Random laser thresholds in cw and pulsed regimes

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We analyze the system of rate equations that, according to our previous studies, adequately describes the behavior of neodymium random lasers. We show that random lasers, in which a noticeable fraction of spontaneous emission goes to lasing modes, have different threshold behavior than regular lasers. We also demonstrate that although the dynamics of stimulated emission in random lasers is strongly dissimilar in different pumping regimes, the threshold energy remains constant, equal to the product of the cw threshold power and the spontaneous emission lifetime τ, if the pumping pulse is much shorter than τ. The results of the study suggest that one can compare the performance of random lasers in different operation regimes and use a cw approximation (with some restrictions) in an analysis of thresholds of pulsed random lasers.

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I. INTRODUCTION

Random lasers are simplest miniature sources of stimulated emission. In random lasers, strong optical scattering in the gain medium provides for stimulated emission feedback, thus eliminating the need for an external cavity. Random lasers, first proposed by Letokhov in the late 1960s [1,2], have been intensively studied, both experimentally and theoretically, over the following years. Most of the solid-state lasers operate in a short-pulsed regime with relaxation oscillations. The examples of short-pulsed random lasers include neodymium random lasers [3–5], color center random lasers [6], ZnO random lasers [7,8], random lasers based on scattering polymers [9–11] and many others. Random lasers based on rare-earth doped δ-Al_2O_3 powders pumped with electron beam operate in a cw regime [12,13].

Interesting physics and exciting potential applications of random lasers, including express testing of novel solid-state laser materials [14], inertial confinement fusion [4], information technology [15], and identification [16,17] stimulate increasingly growing research in this field. Thus, in recent years, the dependence of random laser behavior on the diameter of a pumped spot [18–20], the absorption length [19], and the mean particle size [21], have been studied in a number of experimental and theoretical works.

In regular lasers, the properties of stimulated emission in the cw regime are related to those in a pulsed regime. Thus, knowing the behavior of a laser in the cw regime, one can predict its performance in the pulsed regime, and *vice versa*. For example, if the duration of a pumping pulse t_{pump} is much shorter than the spontaneous emission lifetime τ, then the threshold energy E_{th} is independent of t_{pump} and equal to

$$E_{\text{th}} = P_{\text{th}}^{\text{cw}} \tau, \tag{1}$$

where $P_{\text{th}}^{\text{cw}}$ is the threshold pumping power in the cw regime.

In random lasers, the invariance of the threshold pumping energy is not obvious *a priori*. As it is argued in the following sections, in random lasers the contribution of spontaneous emission to laser modes (which is insignificantly small in regular lasers) strongly influences the threshold behavior and cannot be neglected. Thus in cw random lasers [12], the threshold is not very distinct [the inset of Fig. 1(a)]. At the same time, in pulsed neodymium random lasers, a very sharp threshold [the inset of Fig. 2(b)] is manifested by a dramatic shortening of the emission pulse [the inset of Fig. 2(a)], narrowing of the emission spectrum and an increase of the peak emission intensity up to 10^4 times [3]. The question arises as to whether dramatic threshold changes of the emission behavior in a pulsed regime are governed by the same simple balance between gain and loss, which determines a cw threshold.

Despite the majority of random lasers operate in pulsed regime, a number of theoretical studies of random laser emission are carried out in a cw approximation; see, for example, Refs. [19,22,23]. The cw approximation is primarily used because it allows one to derive relatively simple closed-form analytical solutions for many random laser parameters. At the same time, a few studies have been done to justify the use of a cw approximation for the analysis of random lasers operating in a short-pulsed regime.

In this work, we theoretically investigate random laser thresholds at cw and pulsed pumping. We show that although the behavior of stimulated emission is strongly different in the two regimes, the threshold pumping energy of pulsed random lasers can still be described with Eq. (1) if the duration of the pumping pulse t_{pump} is much shorter than the spontaneous lifetime τ. This justifies the use of a cw approximation in the threshold analysis of pulsed random lasers and allows one to compare the behavior of the same (random) laser material in different operation modes.

II. MODEL

As it has been shown [5], the dynamics of neodymium random lasers can be described both qualitatively and quantitatively using a system of rate equations for the population inversion n and the volume density of emitted photons E. A similar system of rate equations is used in the present study [24],

$$\frac{dn}{dt} = \frac{P(t)}{h\nu_p S l_p} - \frac{n}{\tau} - \frac{E}{h\nu_e} c\sigma_e n, \tag{2a}$$

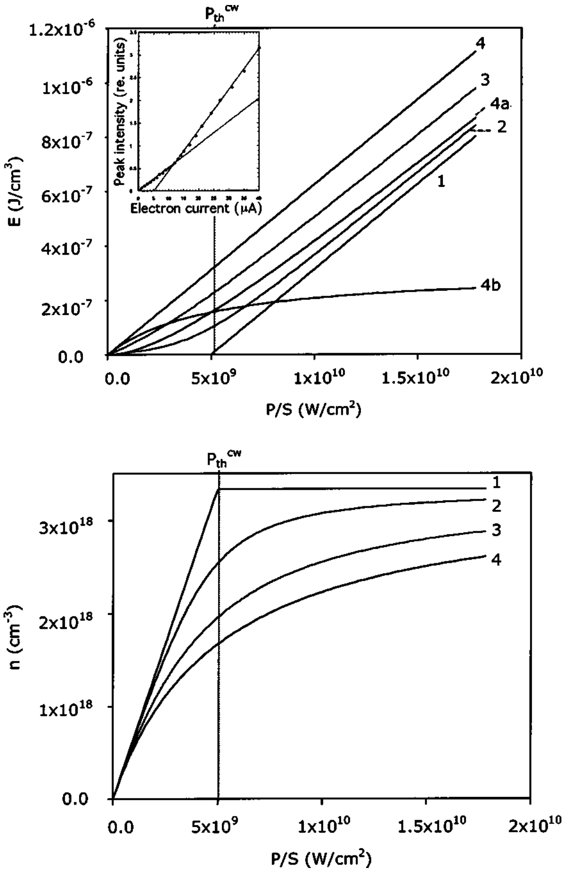

FIG. 1. (a) Calculated input-output curves of random laser emission. Curve 1, calculated at $\zeta = 10^{-6}$, is typical to regular lasers. Curves 2, 3, and 4 are calculated at $\zeta = 0.1$, 0.5, and 1, correspondingly. Curves 4a and 4b, calculated at $\zeta = 1$, correspond to the stimulated emission contribution and spontaneous emission contribution to the total emission. (b) Functions $n(P/S)$ corresponding to the emission curves in (a). Inset of (a): peak emission intensity (at $\lambda_{em} = 362$ nm) of a Ce:δ-Al_2O_3 random laser pumped with a dc electron beam as a function of current. After [12].

$$\frac{dE}{dt} = -\frac{E}{\tau_{res}} + \frac{\zeta n}{\tau} h\nu_e + Ec\sigma_e n. \tag{2b}$$

Here $P(t)/S$ is the pumping power density; l_p is the depth of the pumped layer (Sl_p is the pumped volume); σ_e is the emission cross section, $h\nu_p$ ($h\nu_e$) is the photon energy at the pumping (emission) wavelength; τ is the spontaneous decay time determined by the spontaneous emission, multiphonon relaxation, and cross-relaxation; $\zeta n/\tau$ is the rate with which spontaneously emitted photons populate lasing mode(s); τ_{res} is the photon residence time in a scattering medium; and c is the speed of light.

III. RANDOM LASER THRESHOLD IN A cw REGIME

In the cw approximation, the system of equations (2a) and (2b) has a solution:

$$n = \frac{E}{\tau_{res}\left(\frac{\zeta h\nu_e}{\tau} + c\sigma_e E\right)}, \tag{3}$$

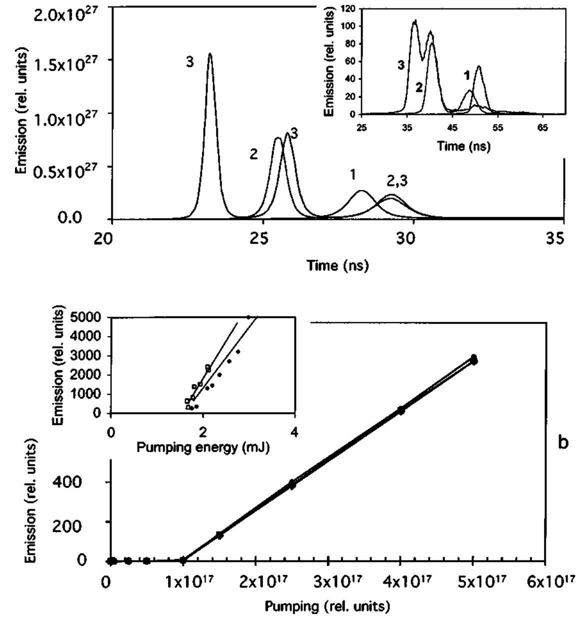

FIG. 2. (a) Calculated kinetics of stimulated emission in a neodymium random laser. The ratio between energies corresponding to traces 1, 2, and 3 was $E_3:E_2:E_1 \Leftrightarrow 136:114:100$. Inset: Stimulated emission kinetics experimentally measured in a $NdSc_3(BO_3)_4$ random laser at 532 nm pumping ($t_{pump} \approx 10$ ns). The ratio between energies corresponding to traces 1, 2, and 3 was $E_3:E_2:E_1 \Leftrightarrow 139:126:100$. (b) Three overlapped input-output curves calculated in a neodymium random laser at t_{pump} equal to 2.5, 5, and 10 ns. Inset: Input-output curves recorded in a $NdSc_3(BO_3)_4$ random laser, at the pumping pulse duration equal to 7 ns (open squares) and 17.5 ns (closed circles). The two thresholds and the two slopes are the same within the experimental accuracy. ($\lambda_{pump} = 532$ nm, diameter of the pumped spot ≈ 0.8 mm.)

$$E = \frac{\tau_{res}\nu_e}{2Sl_p\nu_p}\left[(P - P^*) \pm \sqrt{(P - P^*)^2 + 4\zeta 4PP^*}\right], \tag{4}$$

where

$$P^* = \frac{h\nu_p Sl_p}{\tau\tau_{res}\sigma_e c}. \tag{5}$$

[In Eq. (4), only the "+" sign has a physical meaning.] At $P \to \infty$, the asymptotic behavior of E is

$$E = \frac{\tau_{res}\nu_e}{Sl_p\nu_p}[P - P^*(1 - \zeta)], \tag{6}$$

while at $P \to 0$,

$$E = \frac{\tau_{res}\nu_e}{Sl_p\nu_p}P\zeta. \tag{7}$$

According to Eq. (6), the slope efficiency above the threshold is independent of ζ and equal to $\eta \equiv E/P = \tau_{res}\nu_e/Sl_p\nu_p$.

At $\zeta \to 0$, the threshold pumping power is equal to $P_{th}^{cw} = P^*$ [Eq. (6)] and the slope efficiency below the threshold approaches zero [Eq. (7)]. This is the case of regular lasers, in which $\zeta \to 0$.

In random lasers, especially in random lasers with incoherent feedback, where the emission is omnidirectional and the density of spectrally overlapped modes is very high, the value of ζ is determined by the quantum yield of luminescence (in a spectral band of laser emission) and cannot be neglected. The nonzero value of ζ lowers the value of the threshold to $P^*(1-\zeta)$ [Eq. (6)] and provides for nonzero slope below the threshold [Eq. (7)].

The input-output curves and the functions $n(P/S)$ calculated at different values of ζ and the spectroscopic parameters, which are typical to high-cross-section neodymium random lasers [3] ($\sigma_e = 1\times10^{-18}$ cm^2, $\tau = 2\times10^{-4}$ s, $\tau_{res} = 1\times10^{-11}$ s, $l_p = 0.8$ mm, and $\lambda_p = c/\nu_p = 532$ nm), are depicted in Fig. 1. The calculated value of P_{th}^{cw}/S is equal to 5×10^2 W/cm^2, expectantly, much lower than those experimentally determined in *short-pulsed* neodymium random lasers, 2×10^5–10^9 W/cm^2 [25]. The curves calculated at $\zeta\rightarrow0$, which exhibit sharp thresholds and locking of the population inversion above the threshold, are typical to regular lasers. As ζ increases, the input-output curves become smoother and the value of population inversion determined at the threshold differs from that as $P\rightarrow\infty$. The predicted smoothness of a cw threshold is in agreement with that observed experimentally [12]; the inset of Fig. 1.

At $\zeta=1$, the input-output curves does not have a threshold at all. This result is easy to understand since the laser process cannot improve the quantum yield of emission (which is already equal to unity), and the output is linearly proportional to the input. The pumping-dependent contributions from the spontaneous emission and the stimulated emission to the total emission are also shown in Fig. 1. Note that at $P\rightarrow\infty$, the asymptotic behavior of *stimulated* emission is given by Eq. (6) at $\zeta=0$. (The same asymptotic behavior of stimulated emission contribution holds for all values of ζ.)

Note that diffusion, which governs photon motion in scattering random laser media, is not abandoned in the developed model. The diffusion theory is used to calculate the thickness of the pumped layer l_p and the residence time τ_{res} entering Eqs. (2)–(7) [21].

IV. RANDOM LASER THRESHOLD IN A PULSED REGIME

We started the study of a random laser dynamics with examining the case of infinitely long step-like pumping pulse turned on at $t=0$. The kinetics of emission $E(t)$ and population inversion $n(t)$ have been calculated numerically at different pumping intensities.

At low pumping power, $P<P_{th}^{cw}$, the emission is predominantly spontaneous. In this regime, the characteristic time constant of the build-up of $E(t)$ and $n(t)$ $[\propto(1-e^{-t/\tau})]$ is close to the lifetime τ. With an increase of the pumping power to P_{th}^{cw}, gain balances loss and the system reaches the lasing threshold. However, at $\zeta=1$ (which is approximately the case of random lasers), this does not cause any qualitative change of the stimulated emission dynamics, Fig. 3(a). Likewise, the character of stimulated emission remains to be the same in some range of powers exceeding P_{th}^{cw}. In the latter case, the only predicted change is the shortening of the characteristic buildup times for $E(t)$ and $n(t)$.

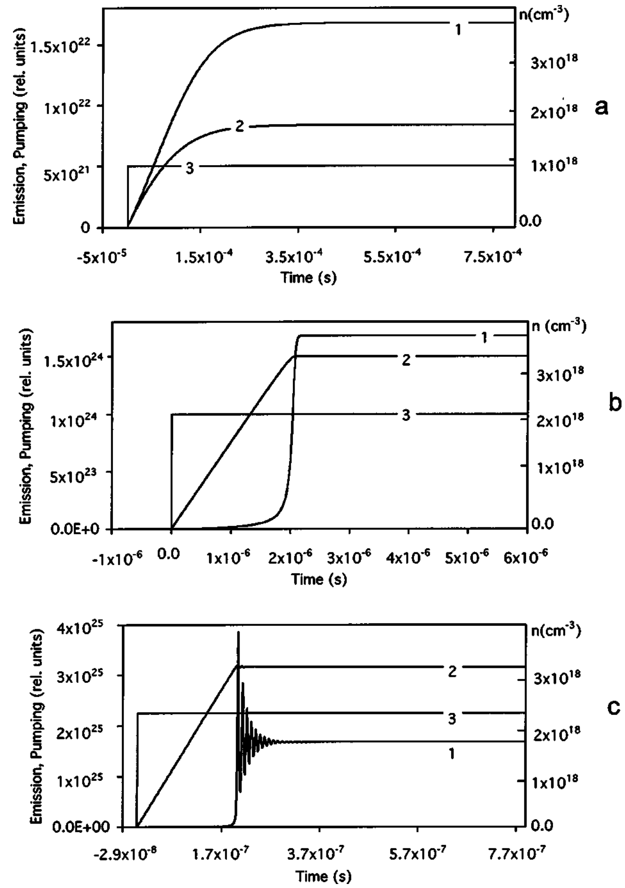

FIG. 3. Kinetics of emission energy density $E(t)$ (trace 1) and population inversion $n(t)$ (trace 2) calculated at $\xi\rightarrow1$ at step-like pumping (trace 3). (a) $P/S=5\times10^2$ W/cm^2$=P_{th}^{cw}/S$; (b) $P/S=5\times10^4$ W/cm^2$=100P_{th}^{cw}/S$; and (c) $P/S=5\times10^5$ W/cm^2 $=1000P_{th}^{cw}/S$.

At much higher pumping power, the character of stimulated emission changes significantly: after certain duration of the pumping pulse, the emission intensity increases in an almost step-like manner, Fig. 3(b). A sharp increase of the emission density can be associated with the threshold of the random laser. At even stronger pumping power, characteristic relaxation oscillations are predicted in the stimulated emission kinetics, Fig. 3(c).

Note that at $\zeta\rightarrow0$ (the case of regular lasers), the shape of the stimulated emission kinetic calculated at $P=P_{th}^{cw}$ approximately resembles the one in Fig. 3(b), but not that in Fig. 3(a). At the same time, kinetics computed at much higher pumping intensities do not strongly depend on the value of ζ.

We then investigated the dynamics of stimulated emission under pumping of the system with short rectangular and Gaussian pulses, which duration ranged between 5 and 500 ns. In this regime, which closely resembles the one realized in many random laser experiments, the threshold is associated with the appearance of the first short pulse in the stimulated emission kinetics. In an agreement with experiment, the number of calculated emission pulses increases with the increase of the pumping energy, Fig. 2(a). When the

TABLE I. Invaiance of pumping energy in random laser excited with pulses of different duration.

Type of pumping	P/S (W/cm^2)	t_{pump} (ns)	E/S (J/cm^2)
cw	5×10^2		
Infinitely long rectangular step	5×10^4	2000[a]	0.1
Infinitely long rectangular step	5×10^5	200[a]	0.1
Rectangular pulse	2×10^5	500	0.1
Rectangular pulse	2×10^6	50	0.1
Rectangular pulse	2×10^7	5	0.1
Gaussian pulse		50	0.1
Gaussian pulse		10	0.1
Gaussian pulse		2.5	0.1

[a]Time of the step-like increase of the emission intensity

energy of the emission pulse(s) is plotted versus the pumping energy, it results in typical for lasers input-output dependence with a very sharp threshold, Fig. 2(b). In accord with the discussions above, a very sharp threshold in the short-pulse regime is explained by a strong predominance of the stimulated emission over the spontaneous emission.

In several pumping regimes studied above, the behavior of stimulated emission was strongly different. However, as follows from Table I, summarizing pumping powers and pulse durations at different excitation conditions, the threshold energy is an invariant value given by Eq. (1) if $t_{pump} \ll \tau$. Thus, knowing the threshold pumping power of a random laser at one pulse duration, it is possible to predict its value at a different pulse duration or cw excitation. Correspondingly, it is possible to study the dependence of a random laser threshold on some of the system parameters (transport mean-free path, diameter of the pumped spot, etc.) in a cw regime and expect to have the same dependence in a pulsed regime.

V. DISCUSSION AND SUMMARY

We analyzed the system of rate equations, which describes the behavior of neodymium random lasers [5] in cw and pulsed regimes. We have found that the cw threshold behavior of random lasers, characterized by strong feeding of spontaneous emission into laser mode(s), can be strongly different from that in regular lasers: the cw thresholds in random lasers are predicted to be much less distinct than those in regular lasers. When the quantum yield of spontaneous emission is equal to unity and *all* spontaneous emission is coupled to lasing modes ($\zeta=1$), then at cw pumping or pumping with long rectangular pulses, no qualitative changes occur to the emission signal at the threshold. This surprising and important result can be commented on as follows.

The developed model does not account for any changes of the emission spectrum. In the first approximation, this corresponds to an experiment in which emission is detected in a spectral band exceeding (or equal to) the bandwidth of spontaneous emission. When the condition *GAIN*=*LOSS* is satisfied, stimulated emission becomes self-supporting. However, at $\zeta=1$, the quantum yield of emission is already equal to 100%. Stimulated emission cannot further improve this value. That is why no changes of the emission intensity *integrated over a broad spectral band* can be seen at the threshold.

In real random lasers (for example, neodymium random lasers), the emission line narrows above the threshold. This narrowing can be theoretically predicted if the model is upgraded to include large number of spectral channels, Ref. [26]. If the emission signal is detected at the maximum of a narrow laser line, then, even at $\zeta=1$, the input-output curve will resemble trace 1 in Fig. 1(a) and the shape of the kinetics at the threshold will be close to that in Fig. 3(b). In other words, the behavior of emission *detected in a narrow spectral band* is qualitatively similar to that in a random laser with $\zeta=0$. This formal analogy has a physical meaning and corresponds to the fact that the fraction of spontaneous emission going to a spectrally narrow channel is very small, even if for the emission integrated over a relatively broad spontaneous emission band, ζ is equal to 1.

In the short-pulsed regime, the emission kinetics are significantly different from those in the long-pulsed regime, and the input-output curve has a distinct threshold even at $\zeta=1$. The latter statement seems to be nontrivial, since at $\zeta=1$, one expects 100% quantum yield of emission. So, one can ask the following: "Where does the energy go below the threshold?" At $\zeta=1$, the value of the emission power integrated over long ($\gg\tau$) period of time, is exactly equal to the pumping energy. If such time-integrated emission is plotted against pumping energy, the input-output curve will resemble trace 4 in Fig. 1(a). Emission of *pulsed* neodymium random lasers manifests itself in form of short high-intensity spikes, which last during the pumping pulse. The intensity of residual spontaneous emission following short spikes of stimulated emission (after the end of the pumping pulse) is very small. Correspondingly, the residual spontaneous emission is never taken into account in experimental measurements or calculations.

This is the reason of the distinct threshold behavior theoretically predicted (and experimentally observed) in short-pulsed random lasers even at $\zeta\to1$. In fact, below the threshold, the intensity of emission, integrated over the duration of the pumping pulse, is negligibly small; however, above the threshold, when the short emission spikes appear, its value is increased significantly.

We have proven that in spite of strongly different dynamics of random laser emission in different regimes, the threshold pumping energy remains constant, determined by the product of the cw threshold pumping power and the spontaneous lifetime if $t_{pump} \ll \tau$. This allows one to study the dependence of the random laser threshold on various system parameters in a cw regime and then extrapolate the results to the pulsed regime. This also allows one to predict and compare the behavior of random lasers operating in different pumping regimes.

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