Anomalously delayed stimulated emission in random lasers

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In a random laser based on a mixture of Nd^{3+}:$Ba_5(PO_4)_3$ and Cr^{4+}:$Y_3Al_5O_{12}$ powders, we have observed trains of anomalously delayed stimulated emission pulses, which lasted for several tens of nanoseconds after the end of the pumping pulse. The phenomenon is explained by the color-center formation and reversed-saturable absorption in Cr^{4+}:YAG powder.

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Random lasers, first theoretically predicted by Letokhov in the late 1960s [1] and experimentally demonstrated in the 1980s [2], are the simplest sources of stimulated emission, which do not require any external mirrors or cavity. The feedback in random lasers is provided by highly scattering random laser media. Examples of random lasers include but are not limited to highly scattering powders [2–5] and films [6–9], as well as suspensions of scatterers in liquid dyes [9–14]. The detailed reviews of random lasers can be found in Refs. [14–16].

Relaxation oscillations—oscillations of stimulated emission intensity in response to an abrupt increase of the pumping power—are commonly observed in lasers [17]. Relaxation oscillations in random lasers have been theoretically predicted by Letokhov in 1968 and studied in detail in more recent years [4,18–23]. In neodymium random lasers, relaxation oscillations occur in a highly nonlinear regime and are manifested by narrow stimulated emission pulses (~1 ns) occurring during a much longer (~10 ns) pumping pulse [4]. The dynamics of stimulated emission intensity and population inversion, calculated as described in what follows, is depicted in Fig. 1(a).

As the population inversion exceeds its threshold value [shown in Fig. 1(a) with the arrow], the first stimulated emission pulse appears, depleting the population inversion below its threshold level. The continuing pumping rebuilds the population inversion above the threshold, after which the second emission pulse appears and the process repeats. The process stops at the end of the pumping pulse when no more excitation is available to restore the population inversion.

In this work, contrary to expectations, in a random laser based on a mixture of Nd^{3+}:$Ba_5(PO_4)_3$ (Nd:BFAP) and Cr^{4+}:$Y_3Al_5O_{12}$ (Cr:YAG) powders, we have observed anomalously delayed trains of stimulated emission pulses, which lasted for a long time after the end of the pumping pulse (Fig. 2). Experimentally, the two grounded powders were mixed in the proportion 3/1 (by weight) and compacted to a pellet. The random laser effect, manifested by the narrowing of the emission spectrum and characteristic short laser pulses at $\lambda = 1.06$ μm [4,14], has been observed when the pumping intensity ($t_{pulse} = 5$ ns, $\lambda = 803$ nm) exceeded some critical threshold level. What was absolutely surprising is that the stimulated emission pulses did not stop at the end of the pumping pulse but lasted for another ~80 ns (Fig. 2).

This anomalous behavior could be explained if the pumping energy were stored in some long-living excited state and continued to feed the upper laser level $^4F_{3/2}$ Nd^{3+} after the end of the pumping pulse. However, this is unlikely the case in our system, since the pumping excited the upper laser level directly (rather than one of the upper energy states of Nd^{3+} [24]) and Cr:YAG does not have any emission at ~800 nm.

We infer that the delayed stimulated emission in our random laser is due to a short-lived excited-state absorption (ESA) induced by random laser pulses [Fig. 1(b)]. In this case, stimulated emission causes an abrupt increase of the absorption loss in the system, which stops the lasing process when the population inversion is still *above* its threshold level in the absence of the induced loss. After an appropriate relaxation time, the emission-induced ESA vanishes and the system (still above the stimulated emission threshold) is ready to produce another random laser pulse. Thus, the stimulated emission can be observed long after the end of the pumping pulse—as long as ESA "dispenses" stimulated emission by small portions, stopping the lasing when the population inversion is still above the threshold.

Cr:YAG is known as an efficient saturable absorber used in mode-locked neodymium lasers [25–27]. Two energy-level diagrams of the material, which differ in some details and are commonly discussed in the literature [25,26], are shown in Fig. 3. In a *single-crystal* Cr:YAG, the emission at 1.06 μm depopulates the ground state and increases the sample's transmission [Fig. 4(a)], since its ground-state absorption (GSA) cross section exceeds the ESA cross section.

Grinding laser crystal to powder often creates color centers dominating random laser behavior [28]. By grinding Cr:YAG, we converted the material from a saturable absorber to a *reversed*-saturable absorber, which is evidenced by the reduction of the powder reflectance at high pumping density [Fig. 4(b)].

The detailed spectroscopic study of ESA in pulverized Cr:YAG is beyond the scope of this work. The calculations have shown that several plausible scenarios involving strong *grinding-induced* ESA can lead to delayed trains of stimulated emission pulses [Fig. 1(b)], similar to those observed experimentally (Fig. 2).

The population inversion of Nd^{3+} ions (the population of the upper laser level $^4F_{3/2}$) n, the emission energy density E, and the excited-state populations of Cr^{4+} ions m_1 and m_2 are described with the system of rate equations following the

energy-level diagram in Fig. 3 (left),

$$\begin{aligned}
&\frac{dn}{dt} = \frac{P}{h\nu_{\text{pump}} l_p S} - \frac{n}{\tau_0} - \frac{E}{h\nu_{\text{em}}} c\sigma_{\text{em}} n, \\
&\frac{dE}{dt} = Ec\sigma_{\text{em}} n + \frac{n}{\tau_0} h\nu_{\text{em}} - \frac{E}{\tau_{\text{res}}} - Ec\sigma_{\text{gs}}(M - m_1 - m_2) - Ec\sigma_{\text{es}}^1 m_1 - Ec\sigma_{\text{es}}^2 m_2, \\
&\frac{dm_1}{dt} = \frac{E}{h\nu_{\text{em}}} c\sigma_{\text{gs}}(M - m_1 - m_2) - \frac{m_1}{\tau_1} + \frac{P}{h\nu_{\text{pump}} S}(M - m_1 - m_2)\sigma_{\text{abs}} - \frac{E}{h\nu_{\text{em}}} c\sigma_{\text{es}}^1 m_1 + \frac{m_2}{\tau_2}, \\
&\frac{dm_2}{dt} = \frac{E}{h\nu_{\text{em}}} c\sigma_{\text{es}}^1 m_1 - \frac{m_2}{\tau_2}.
\end{aligned} \tag{1}$$

Here P is the pumping power; S is the area of the pumped spot; l_p is the thickness of the pumped layer; $h\nu_{\text{pump}}$ ($h\nu_{\text{em}}$) is the pumping (emission) photon energy; τ_0 is the spontaneous emission lifetime of Nd^{3+} ions; σ_{em} is the Nd^{3+} emission cross section; c is the speed of light; τ_{res} is the residence time of photons in the random laser (determined by the scattering strength and the volume); M is the total concentration of Cr^{4+} ions; τ_1 and τ_2 are the life-times of Cr^{4+} excited states $|1\rangle$ and $|2\rangle$; σ_{gs}, σ_{es}^1, and σ_{es}^2 are GSA and ESA cross sections of Cr^{4+} at the transitions $|0\rangle \to |1\rangle, |1\rangle \to |2\rangle$, and $|2\rangle \to |3\rangle$, respectively; and σ_{abs} is the Cr^{4+} absorption cross section at the pumping wavelength. In calculations, neodymium random laser parameters were adopted from Refs. [4,29] and plausible parameters of Cr^{4+}:YAG powder (following Refs. [25–27]) were as follows: $M = 8.0 \times 10^{17}\ \text{cm}^{-3}$, $\tau_1 = 4\ \mu\text{s}$, $\tau_2 = 10\ \text{ns}$, $\sigma_{\text{gs}} = 1.4 \times 10^{-18}\ \text{cm}^2$, $\sigma_{\text{es}}^1 = 5.0 \times 10^{-19}\ \text{cm}^2$, $\sigma_{\text{es}}^2 = 1.5 \times 10^{-17}\ \text{cm}^2$, and $\sigma_{\text{abs}} = 2.7 \times 10^{-18}\ \text{cm}^2$.

The part of Eq. (1) enclosed within the dashed rectangle describes the behavior of the random laser without Cr:YAG [Fig. 1(a)]. As discussed earlier, in a "pure" neodymium random laser, stimulated emission pulses drive the population inversion below its threshold level and the lasing inevitably stops after the end of the pumping pulse [Fig. 1(a)]. Full Eq. (1) describes the stimulated emission in a random laser mixed with a reversed-saturable absorber. Short-lived ESA in the system

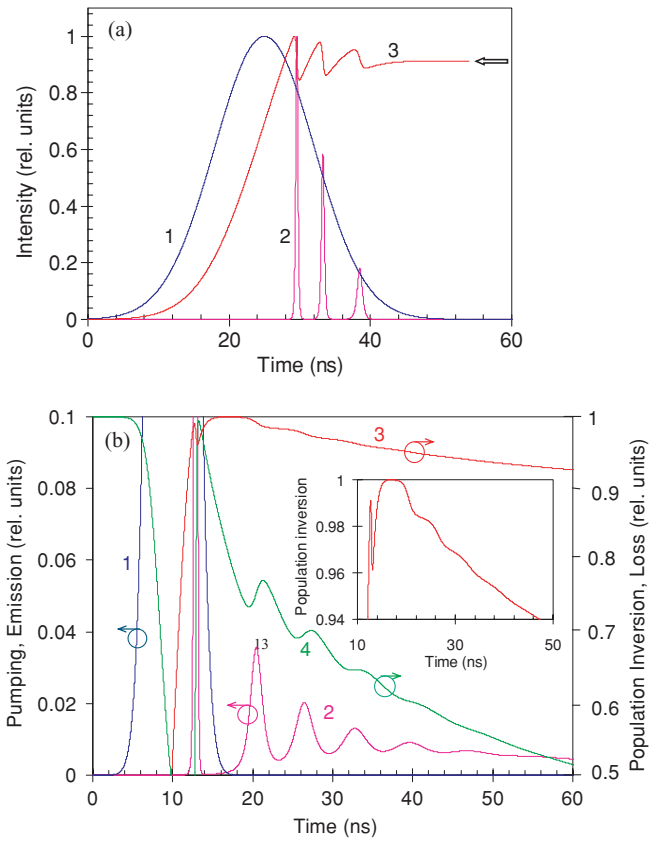

FIG. 1. (Color online) Gaussian pumping pulse (trace 1), stimulated emission pulses (trace 2), population inversion (trace 3), and absorption loss [trace 4 in panel (b)] calculated for $\tau_0 = 400\ \mu\text{s}$, $\tau_{\text{res}} = 10\ \text{ps}$, $\sigma_{\text{em}} = 5.0 \times 10^{-19}\ \text{cm}^2$, $l_p = 0.8\ \text{mm}$, $\lambda_{\text{pump}} = 803\ \text{nm}$, and $\lambda_{\text{em}} = 1055\ \text{nm}$. Panel (a) is calculated using the part of Eq. (1) enclosed with the dashed rectangle, which models stimulated emission in pure neodymium-doped powder. Panel (b) is calculated using full Eq. (1), which models the random laser effect in the mixture of Nd^{3+}:BFAP and Cr^{4+}:YAG powders, assuming the energy level diagram shown in Fig. 3 (left). All traces are normalized to unity. (Inset) A fragment of the population-inversion kinetics showing a characteristic "staircase"-line decay.

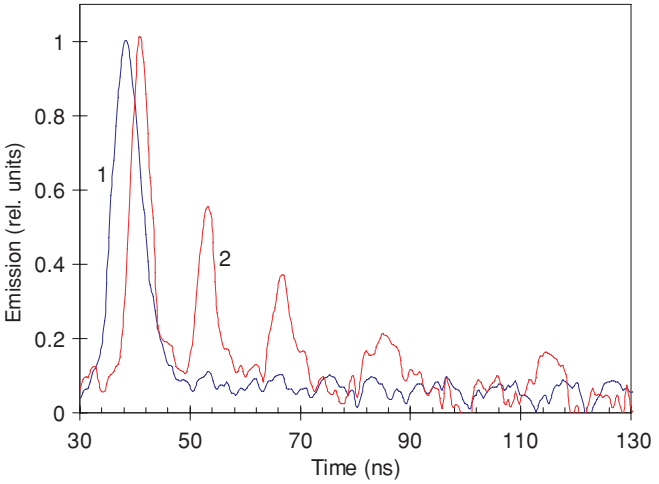

FIG. 2. (Color online) The pumping pulse (trace 1) and the train of stimulated emission pulses (trace 2) in the $(\text{Nd:BFAP})_{0.75}$:$(\text{Cr:YAG})_{0.25}$ random laser. The diameter of the pumping beam is ~0.42 mm and the pumping energy is ~4.6 mJ (two times larger than the threshold value).

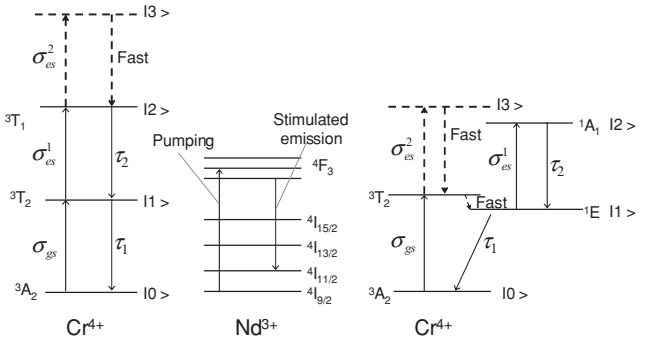

FIG. 3. The energy-level diagram of Nd^{3+} ions (center) and two energy-level diagrams of Cr:YAG commonly discussed in the literature (left [25]; right [26]). Possible strong ESA transitions making the material a *reversed*-saturable absorber are shown with bold dashed lines. The cross sections of these transitions exceed all other absorption cross sections in the system.

stops the stimulated emission when the population inversion is still above the threshold in the absence of ESA. Reversed-saturable absorption "dispenses" the population inversion by small portions, allowing the train of random laser pulses to continue long after the end of pumping [Fig. 1(b)].

Two experimental characteristics of a random laser, which can be easily compared to calculations, are the threshold energy density E_{th}/S and the time interval between relaxation oscillation pulses (e.g., between the first and the second pulses, Δt_{12}). The experimentally measured threshold in the $(Nd:BFAP)_{0.75}$:$(Cr:YAG)_{0.25}$ random laser is 1.6 J/cm^2, while the theoretical threshold is >0.9 J/cm^2. (The latter value is calculated taking into account the reflection of pumping light by the sample [30]. However, the fraction of energy absorbed by Nd:BFAP (as opposed to Cr:YAG) in a mixed scattering sample is difficult to evaluate. That is why the theoretical value is known only approximately.) Similarly, the experimental time interval $\Delta t_{12} = 12$ ns is fairly close to its calculated value 8 ns. Thus, the proposed model describes the behavior of the random laser studied both qualitatively and quantitatively.

One of the parameters of the system, which is known with lower accuracy, is the photon residence time in the system, τ_{res}. It affects both E_{th}/S and Δt_{12}. The fact that both experimental values are in a good agreement with the theoretically predicted ones suggests that the estimated value $\tau_{res} = 10$ ps, which is in agreement with the one determined in a neodymium random laser in Ref. [4], is very reasonable. In this work, we do not differentiate between the photon residence time in a pumped volume and the one in an effective scattering cavity (which includes both pumped and not-pumped parts of the sample [31]). This helps us to keep the model simple and reduce the number of not-very-well known parameters.

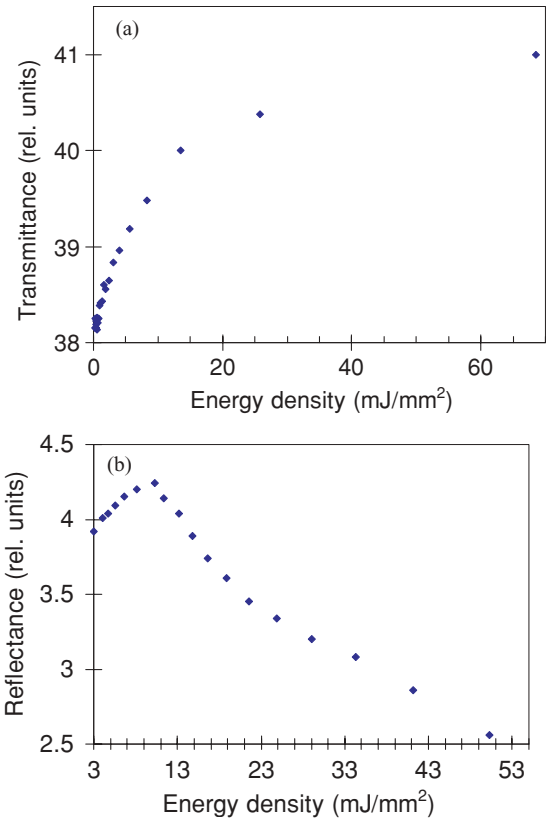

FIG. 4. (Color online) Transmittance of a 2.5-mm Cr^{4+}:YAG slab (a) and reflectance of Cr^{4+}:YAG powder (b) as a function of the pumping-energy density ($\lambda = 1064$ nm, $t_{pulse} \sim 10$ ns).

Note that the effect of a much smaller magnitude—a single, very weak stimulated emission pulse occurring after the end of the pumping pulse—can occasionally be observed in neodymium random lasers without Cr^{4+}, implying an existence of color centers in powders of neodymium-doped laser media.

To summarize, in the random laser based on the mixture of the Nd^{3+}-doped and Cr^{4+}-doped powders we have observed trains of anomalously delayed stimulated emission pulses, which lasted for several tens of nanoseconds after the end of the pumping pulse. The phenomenon is explained by the formation of color centers and the effect of reversed-saturable absorption in Cr^{4+}-doped powder, which damps laser pulses and does not allow them to deplete the population inversion in one shot.

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