Relaxation oscillations in long-pulsed random lasers

Karen L. van der Molen,[1,*] Allard P. Mosk,[1] and Ad Lagendijk[2]
[1]*Complex Photonic Systems, MESA+ Institute for Nanotechnology and Department of Science and Technology, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands*
[2]*FOM Institute for Atomic and Molecular Physics (AMOLF), Kruislaan 407, 1098 SJ Amsterdam, The Netherlands*
(Received 12 September 2008; revised manuscript received 30 June 2009; published 19 November 2009)

We have measured the evolution of the intensity emitted by a random laser during a pump pulse that is comparable in duration to the spontaneous emission decay time. The time traces of our random laser, consisting of titanium dioxide particles and sulforhodamine B dye, show clear relaxation oscillations. We compare our experimental results to an analytic, conventional model based on the four-level rate equations for a single-mode laser. The measured frequency of well-resolved fluctuation oscillations suggests an effective mode lifetime of 5 ps.

DOI: 10.1103/PhysRevA.80.055803 PACS number(s): 42.60.Rn, 42.25.Dd, 42.55.Zz

I. INTRODUCTION

Relaxation oscillations of conventional lasers are a well-understood phenomenon [1]. They are especially important for continuous-wave and long-pulsed lasers, for which the duration of the pump pulse is comparable to, or exceeds, the spontaneous emission decay time of the medium. In a random laser, a medium in which gain is combined with multiple scattering of light, relaxation oscillations can also occur, as was predicted by Letokhov in 1968 [2]. A first observation of random lasing in strongly scattering powders was already presented in 1986 [3] and the demonstration of a random laser consisting of microparticles in a laser dye solution by Lawandy *et al.* in 1994 [4] sparked an intense research interest in this field [5–8]. In random lasers, relaxation oscillations and other typical laser phenomena have been observed [9]. Detailed studies on these relaxation oscillations have been performed in neodium random lasers [10]. These random lasers, with a spontaneous emission time in the order of microseconds, were pumped with nanosecond pulses. Soukoulis and co-workers presented measurements of relaxation oscillations in single modes of a picosecond pumped random laser system with a spontaneous emission time of a factor 10 longer than the pump pulse [11]. Different numerical calculations on random lasers also show pronounced oscillatory behavior [12–14]. To our knowledge, no measurements have been performed on relaxation oscillations in random lasers in the interesting regime of long pump pulses, where the duration τ_p of the pump-laser pulse is longer than the spontaneous emission lifetime τ of the emitters.

In this Brief Report, we present measurements of the time evolution of a long-pulsed random laser system just above the laser threshold. We compare our experimental observations to a conventional, simple model based on the four-level rate equations for a single-mode laser.

II. SIMPLE MODEL

The evolution of the number of excited molecules N_1 and the number of photons q in a laser are described by the well-known four-level rate equations [1]

$$\frac{dN_1(t)}{dt} = P(t) - \frac{\beta q(t) N_1(t)}{\tau} - \frac{N_1(t)}{\tau}, \tag{1a}$$

$$\frac{dq(t)}{dt} = -\frac{q(t)}{\tau_c} + \frac{\beta N_1(t)}{\tau}[q(t)+1], \tag{1b}$$

where $P(t)$ is the pump power that is absorbed by the molecules inside the cavity, τ_c the cavity decay time, and β the beta factor of the laser. The beta factor is defined as the fraction of spontaneous emission that contributes to the lasing mode and can also be determined for a random laser [16]. We note that a very similar set of laser equations was used to describe a neodymium random laser [9]. The main difference is that in our case, the spontaneous emission time τ is comparable to the time scale of the laser dynamics, whereas in the neodymium random laser it is much longer. Noginov and co-workers [15] numerically solved the system of equations [(1a) and (1b)] in the long-pulse regime, assuming a smooth Gaussian shape of the pump pulses. In that case, no relaxation oscillations were observed, possibly because there were no frequency components in the pump pulse that could excite them.

Rather than solving the system of laser equations numerically, we make use of the analytic equation for the frequency of the relaxation oscillations of conventional lasers derived by Woerdman and co-workers [17]

$$\omega_{\mathrm{res}} = \sqrt{\left(\frac{M-1}{\tau_c \tau}\right) - \frac{1}{4}\left[\frac{M}{\tau} - \frac{\beta}{\tau_c(M-1)}\right]^2}, \tag{2}$$

where ω_{res} is the frequency of the relaxation oscillations and M is the scaled pump fluence, defined as the ratio of the absorbed pump fluence and the threshold fluence. This equation predicts the frequency of small-amplitude relaxation oscillations when the pump intensity varies slowly compared to $\omega_{\mathrm{res}}^{-1}$. It explicitly includes the effects of spontaneous emission. The derivation of this result assumes the good-cavity regime, i.e., the linewidth of the laser cavity is smaller than that of the laser medium, a condition which is fulfilled as the dye emission linewidth is much larger than the characteristic escape rate of photons from the random laser volume which acts as the cavity.

*k.l.vandermolen@alumnus.utwente.nl

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In Eq. (2), the first term in the square root describes the free oscillation frequency, while the second term is a correction due to damping. In Ref. [10], the same equation was obtained for a random laser system, however, the damping correction, which is negligible far above threshold, was omitted. We note that the argument of the square root becomes negative due to the damping correction when the oscillations are strongly overdamped. The damping constant is given by

$$\gamma_{\text{res}} = \frac{1}{2}\left(\frac{M}{\tau} + \frac{\beta}{\tau_c(M-1)}\right). \tag{3}$$

The frequency spectrum of oscillations in the laser output will be the product of the noise spectrum driving the oscillations and the spectral response function of the laser. This response spectrum has a peak at ω_{res} with a characteristic width γ_{res}. Well above threshold, $\gamma_{\text{res}} \ll \omega_{\text{res}}$, and the relaxation oscillations have a well-defined frequency [1,17]. Fluctuations of the pump intensity at off-peak frequencies will be strongly damped and therefore not be visible in the output. Close to threshold, on the other hand, $\gamma_{\text{res}} \geq \omega_{\text{res}}$ and relaxation oscillations are not well resolved. The laser acts as a driven oscillator with a very broad, damped resonance and the fluctuation spectrum of the random laser output will resemble the fluctuation spectrum of the pump pulse.

We apply the analytic Eq. (2) to the multimode pulsed random laser by assuming that oscillation predominantly occurs in modes with a cavity decay time τ_c and that the fraction of spontaneous emission that enters the laser mode is given by the beta factor of the random laser [16]. For our random laser, these assumptions accurately model the nonlinear relation between pump fluence and output power around threshold [18].

III. EXPERIMENTAL APPARATUS

The random laser used in our experiments is essentially the same as used in earlier work [18]. It consists of a suspension of TiO_2 particles (mean diameter of 180 nm, volume fraction of 10%) in a solution of sulforhodamine B in methanol (1 mmol/liter; pump absorption length, 104 μm; minimal gain length, 83 μm [19]). The spontaneous emission time of the dye is 3.2 ns [19]. The suspension is contained in a fused silica capillary tube, with internal dimensions $100 \times 2 \times 0.2$ mm^3. The mean-free path of light in the sample in the absence of pumping was measured in a separate apparatus using enhanced-backscatter cone measurements [21] and escape function measurements [22]. We found a transport mean-free path of 0.46 ± 0.1 μm at 633 nm (effective refractive index 1.48 ± 0.04).

In our random laser experiments, the samples were excited by a pump pulse at 532 nm, provided by an optical parametric oscillator (OPO) pumped by a Q-switched Nd:yttrium aluminum garnet (YAG) laser (Coherent Infinity 40-100/XPO). The pump pulse had a duration of 3 ns and a repetition rate of 50 Hz. The pump light was focused with a microscope objective [water-immersed, numerical aperture (NA) 1.2] onto the sample (focus area, 12 ± 6 μm^2), reaching a fluence in the order of 1 mJ/mm^2. The pump fluence

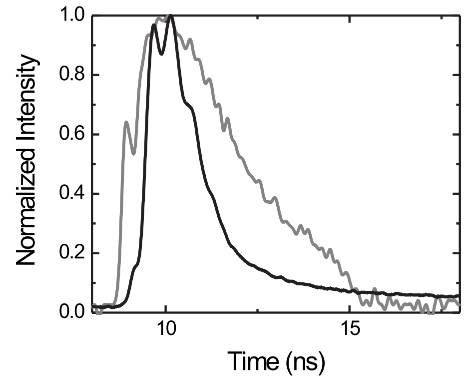

FIG. 1. Measured time traces of the pump pulse (gray) and emission output above threshold (black, input fluence $=0.47$ mJ/mm^2) of a titania random laser normalized to their maximum value. The pump pulse duration is much longer than the duration of the emitted light of the random laser above threshold. Relaxation oscillations in the emitted light are clearly visible near the peak intensity. The decay time of the emitted light is first dominated by stimulated emission. In the second part of the decay curve, spontaneous emission dominates.

was adjusted by attenuating the pump light using two polarizers, making sure that the polarization of the pump light on the sample was not changed. The central wavelength of the emitted light was 594 nm and the narrowing factor (defined as the spectral width of the emitted light far below threshold divided by the spectral width far above threshold) is 8. The spontaneous emission factor β was 0.07 ± 0.03 at a threshold fluence of 0.10 ± 0.02 mJ/mm^2, as determined from the curve of the output energy versus pump fluence [18]. The light emitted by the random laser was collected by the same microscope objective, detected by a fast photodiode (New Focus 1404, bandwidth 25 GHz), and read out by an oscilloscope (Tektronix TDS 7404, analog bandwidth, 4 GHz). The position of the detector was chosen to collect as much emission light as possible; no angular or spatial selection was performed. The resulting time resolution was 100 ps. To obtain a good signal-to-noise ratio, the data shown are an average over 100 laser pulses. The pump light was filtered out of the detection path by use of a colored glass filter with an optical density of more than 4 at the wavelength of the pump laser.

IV. MEASURED RELAXATION OSCILLATIONS

The normalized time traces of the pump pulse and of the total emitted light from the random laser far above threshold are shown in Fig. 1. The pump pulse is not smooth. It contains broadband fluctuations with frequency components between 1 and 10 GHz. Overall, the duration of the pump-laser pulse is longer than the duration of the pulse of light the random laser emits. The pulse emitted by the random laser first exhibits a fast decay, followed by a slower exponential decay. The fast decay is due to stimulated emission in the random laser. In the second part of the decay, the population inversion is no longer present and spontaneous emission causes a slower decay of intensity. These observations are in

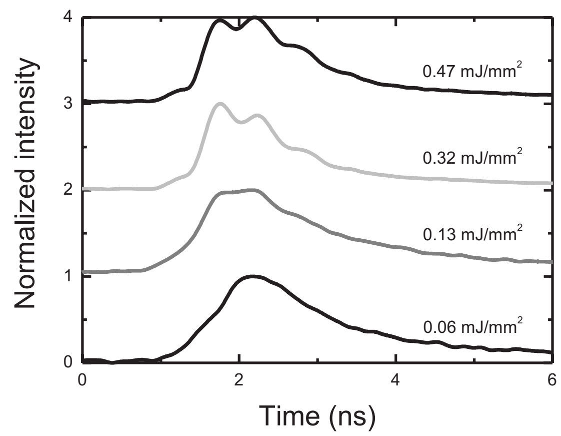

FIG. 2. Measured time traces of the emission intensities of a titania random laser for four different pump fluences. The traces are vertically shifted with respect to each other for clarity. Relaxation oscillations become more pronounced at higher pump fluences.

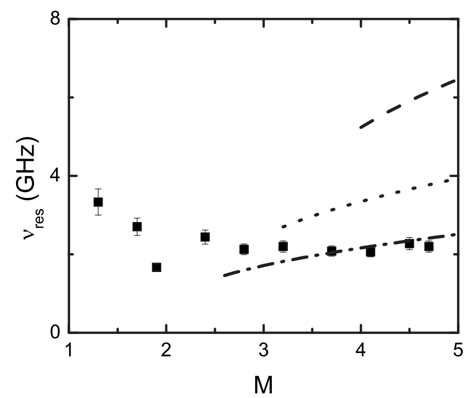

FIG. 3. Measured relaxation oscillations frequency as a function of the normalized pump fluence M, defined as the ratio of the absorbed fluence P and the threshold fluence P_{th} (squares). The simple model [Eq. (2)] is plotted for different cavity decay times 0.7 ps (dashed line), 2 ps (dotted line), and 5 ps (dash-dotted line). The curves are shown for values of M where relaxation oscillations are well resolved. At low pump fluence, $M<3$, relaxation oscillations are not well-resolved. In this regime, oscillations in the pump intensity determine the frequency of the oscillations in the random laser output.

agreement with other random laser experiments [11]. In Fig. 1, relaxation oscillations in the emitted light are clearly visible near the peak intensity.

In Fig. 2, the normalized output intensity is plotted versus time for four different pump fluences [25]. The time traces are shifted vertically with respect to each other for clarity. The time trace at a pump fluence of 0.06 mJ/mm^2 is below threshold, while the other time traces are above threshold. We observe that relaxation oscillations occur above threshold and become more pronounced when the pump fluence increases. We measured at pump fluences up to a factor 5 above threshold. At higher pump fluence, our samples were damaged.

The frequency of the relaxation oscillations is computed from the time traces as follows. The difference of two consecutive local maxima Δt is interpreted as the period, so that the frequency of the relaxation oscillation $\omega_{\rm rel}$ is obtained as $2\pi/(\Delta t)$.

V. COMPARISON OF THE MEASUREMENTS TO THE MODEL

In Fig. 3, the measured frequencies of the relaxation oscillations are plotted versus the scaled pump fluence M. The measured frequency of the oscillations decreases when the scaled pump fluence increases from 1 to 2. A further increase of the pump fluence does not significantly change the frequency of the oscillations.

The result of Eq. (2) is plotted for different cavity decay times. This cavity decay time is the only parameter that was not fixed by measurements. We have limited the curves to the parameter space where the relaxation oscillations are well resolved, $\omega_{\rm res}>2\gamma_{\rm res}$.

The model predicts well-resolved relaxation oscillations for $M>3$. The frequency of the relaxation oscillations in the model increases slightly with pump fluence. We find a reasonable correspondence with the measured data assuming a cavity decay time of 5 ps. We note that while model frequencies slightly increase with pumping, the measured frequencies do not significantly change for $M>3$. Neither the pump pulse shape nor its duration enters the model Eq. (2) as parameter, as the model assumes a constant pump intensity after turn on. This situation is not too different from the experimental one, as the relaxation oscillations occur simultaneously with the peak of the pump pulse, at which time the pump intensity varies only little over one oscillation period.

The mean cavity decay time $\bar{\tau}_c$, which is the time scale on which energy escapes the passive system, is approximated by the mean of the phase delay-time distribution for light traveling from a source to the outside of the medium. We calculate this mean under the assumption that the source is in the center of the gain volume and find [18]

$$\bar{\tau}_c = \frac{1}{8}\frac{L^2}{D}, \tag{4}$$

with L the linear size of the gain volume and, for nonresonant scattering, the diffusion constant $D=c_0\ell/(3n')$, where c_0 is the speed of light in vacuum, ℓ the transport mean-free path, and n' is the real part of the effective refractive index of the medium. When the pump beam does not saturate the gain medium, L is approximately $L_a=\sqrt{\ell\ell_a/3}$, where ℓ_a is the pump absorption length. Although above threshold the gain volume may be larger, numerical calculations [20] show $L<3L_a$ in the regime $3<M<5$. For these parameters, we estimate the mean decay time in the gain volume $\bar{\tau}_c$ to be 0.6 ps. The effective decay time of the cavity may be longer since in a nonabsorbing medium, the effective cavity may extend significantly beyond the pumped volume, as the light can be scattered back into the gain volume by the scatterers outside.

The comparison to theory in Fig. 3 suggests an effective cavity lifetime of the random laser modes of 5 ps, much longer than the estimated 0.6 ps. This longer effective lifetime is not unexpected as the modes may extend beyond the pump volume, increasing their lifetime, and the lasing process is expected to occur selectively in the modes with the longest dwell times [23]. Our experiment corroborates the expectation that modes which have long dwell times contribute to the emitted laser light.

At low pump fluence, ($M<3$) the relaxation oscillations are not well-resolved [17]. In this regime, fluctuations of the output intensity occur, but their frequency strongly depends on the spectrum of the pump oscillations that drive them. Close to threshold, we observe a frequency of 3.3 GHz, which coincides with a strong component of oscillations in the pump pulse (see Fig. 3). The interpretation is that oscillations of the pump intensity drive the relaxation oscillator off-resonance at low pumping. At high pumping, the relaxation oscillation peak frequency is well-defined and the only frequency component observed is the relaxation oscillation frequency ω_{res}.

At much higher pump fluence, a clear increase of the relaxation oscillation frequency has been found in random laser experiments and accompanying calculations by Noginov and co-workers [10]. Their experiments, which were conducted far above threshold, qualitatively follow the behavior of the simplified model [Eq. (2)].

The field of relaxation oscillations in random lasers, and especially the quantitative determination of τ_c, is open to a more detailed theoretical analysis. A promising approach is the concept of self-consistent modes [24].

VI. CONCLUSIONS

We have seen relaxation oscillations in our random laser while looking at the time evolution of the total emitted light for different realizations of the sample. Multiple modes contributed to these time traces and we averaged the time traces over several realizations of disorder of our random laser sample. The resulting time trace still showed relaxation oscillations: a weighted average of the oscillations of all the underlying modes.

The measured relaxation oscillations were compared to a simple analytic model based on a single-mode continuous-wave laser. The cavity decay time at which the simple model matches the data is about a factor 10 higher than the mean cavity decay time. Our experiment supports the idea that modes with cavity decay times far above the average play a crucial role in random laser physics. Closer to threshold, the measured frequencies of the intensity oscillations show a remarkable decrease with pump fluence. This decrease is due to a crossover from strongly damped forced oscillation to well-resolved relaxation oscillations.

ACKNOWLEDGMENTS

This work is part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie (FOM), which is financially supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO). We thank Boris Bret for the work that lead to the estimation of the transport mean-free path.

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