On the temporal behavior of Nd³⁺ random lasers

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The temporal dynamics of random laser emission from Nd-doped crystal powders has been investigated by using an improved detection device. The precise conditions to detect high contrast, fast oscillations are described. The measured period is about one order of magnitude shorter than the one given in the literature for relaxation oscillations in these materials. The origin of these differences is discussed. © 2013 Optical Society of America

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The investigation of laser action in disordered material structures such as highly scattering powders, films, colloidal dye solutions, human tissues, etc., is at present a subject of great interest. In this type of cavityless laser, multiple light scattering replaces the standard optical cavity of a conventional laser, and the interplay between gain and scattering determines the lasing properties. Detailed discussions about results and theories concerning the mechanisms responsible for random lasing as well as the history and the state of art can be found in [1–5].

On the other hand, the study of lasing properties of activated powders for optoelectronic and medical applications requiring low coherence such as high resolution speckle-free imaging and/or highly stable optical frequency among others was worth doing [1,2,6,7], in particular, the investigation of the luminescence properties of rare earths in inhomogeneous dielectric structures including highly scattering powders. Among rare-earth-activated powders, those based on Nd³⁺ are especially interesting due to their low threshold and high efficiency in almost every host [8–17].

One of the most relevant points in this field research is the temporal behavior of the laser output in rare-earth-doped random lasers (RLs). These lasing systems show apparent relaxation oscillations of the order of nanoseconds [8,12,13,18], which are typical of this kind of laser where the spontaneous excited state decay time is much longer than the residence time of the emitted photons in the cavity; this behavior is also found in other types of RL [19,20].

In conventional lasing systems the simplest theoretical approach to study their temporal behavior consists in the simulation by means of rate-equations, which has also been widely applied to RL [8,12,13,18,19,21]. However, problems of interpretation of the experimental results still remain, because the period of the observed oscillations is not compatible with the expected value of the photon residence time [8,13,19] and frequently the obtained values are justified assuming a photon occupation volume larger than the pumped volume [8,12,13], or equivalently, two different photon residence times.

A few difficulties dealing with measurements are worth being mentioned. In most cases the obtained

results and inherent conclusions are conditioned by limitations in the temporal resolution of the measuring system along with a not enough good knowledge of the temporal shape of the pumping pulse [12,13]. In general, the obtained results are conditioned by how the measures have been exactly performed, particularly in relation to detection spatial filtering. Due to these limiting factors, it seems quite difficult to derive any definite conclusions from that set of measurements.

The aim of this work is to carry out accurate time-resolved measurements in order to access the temporal dynamics of rare-earth-doped RLs. Although we have investigated laser emission from several Nd-doped powders, for simplicity, the results shown in this work are from the stoichiometric borate NdAl₃(BO₃)₄, due to its high efficiency. For the first time to our knowledge, the measurements have been done with sufficient simultaneous spatial and temporal resolution in order to observe well-resolved quick local oscillations. Moreover, we believe this study will help to clarify the temporal behavior of other types of RLs, even when a more complex spectral structure is present. For example, dye RLs which have spectral widths of a few nanometers, much higher than Nd RLs with a line width less than 1 Angstrom [22].

In a recent work [16] the authors have presented a study of the threshold and energy efficiency in RLs based in Nd³⁺-doped oxysulfide (La₂SO₂:Nd) powders. In that work, the pumping source was a tunable 10 ns time width Ti:sapphire pulsed laser and the pump beam size over the sample surface was controlled by means of a 60 cm focal lens mounted in a movable holder. The output radiation was directly collected through a 500 μm core diameter fiber, with its head located at 120 mm distance from the powder surface. Only attenuation filters or pump removal filters were interposed between the powder surface and the optic fiber. In this configuration, the fiber head collected the radiation from the entire powder surface. For the investigation of the temporal behavior of Nd³⁺ RLs dealt with in the present Letter we have used a similar setup with the same pumping and output radiation collection system but with a faster detection and recording system which consisted in a New Focus 1414 detector from Newport coupled to a digital Tektronix

DP071604 oscilloscope. To collect the RL output at the detector we have tested different optical fibers. The temporal widening introduced by the detection system (oscilloscope + detector + fiber) was measured by using the 30 ps temporal pulse-width of a mode-locked picosecond IR laser as a reference. The temporal width of the reference laser pulse measured by using a 2 m long, 500 μm core diameter fiber was 50 ps, a resolution which became severely degraded for longer fibers with the same core diameter. However, for 1 m long and 60 μm core diameter graded index fibers, the measurement gave 40 ps, and no significant degradation was observed for longer fibers. Therefore, by using this type of fiber we can ensure that the temporal widening introduced by our measurement device is less than 40 ps.

As we mentioned above, in our first experiments we used a 2 m long, 500 μm core diameter fiber and the distance powder surface-fiber head was 120 mm. Figure 1 shows some different RL single shots registered by using this device, together with their associated pumping pulses. The pumping pulses were registered with a silicon detector (model 818-BB-21 from Newport, with rise and fall time less than 300 ps) [16]. In the first instance, Fig. 1 shows that the pump pulse shapes (blue lines) can be highly variable from shot to shot in a standard Q-switched (flash lamp-pumped) nanosecond laser. The oscillations, due to longitudinal mode beating, are clearly observed in Fig. 1(a) whereas in other pumping pulses, Fig. 1(b), the mode beating effect appears duplicated or even more complicated as shown in Fig. 1(c). These effects can introduce additional confusion in the interpretation of the RL emission because the time period of these oscillations ($\sim$ nanosecond) might be erroneously interpreted as lasing relaxation oscillations. The RL examples shown in Fig. 1 (red lines) clearly demonstrate that when threshold is attained, the RL response faithfully follows the pumping intensity and that no relaxation oscillations are resolved by using the above mentioned detection setup. Nonetheless, what we do observe are small spikes overlapping the laser output, the temporal shape of which shows a period of the order of 100 ps, as can be observed with more detail in the inset of Fig. 1(a). At first glance we thought it might be noise generated by the detection process, so in order to clarify this question, we modified our detection system by using 1 m long, 60 μm core diameter graded-index fiber to collect the laser emission at the same distance from the emitting surface (120 mm). The result was a spectacular increment of the temporal resolution with the appearance of clearly resolved oscillations. The same effect of contrast increment can be obtained by working with the 500 μm core diameter fiber but lengthening the collecting distance to around 1 m, though in this case, noise problems derived from a low signal level may appear. Figure 2 shows an example of a single pulse obtained using a 60 μm core fiber at a distance of 500 mm.

Although we usually detect the radiation emitted from the powder in a perpendicular direction to its plane, we have verified that the observed effect does not depend on the detection angle. Moreover, the temporal shape of simultaneous measures at different angles of the same pulse does not show any short-time correlation. We must also emphasize that if a diffuser glass is introduced

Fig. 1. Time dynamics of three different RL emissions (red lines) collected directly with a 500 μm core diameter optic fiber located at 120 mm from the powder surface, and their corresponding pump pulse profiles (blue lines). In all cases the sample is a powdered stoichiometric borate [11], the pump beam diameter over the sample is 2.0 mm and the pumping energy is approximately four times the threshold level (about 30 mJ). In the inset of the upper picture, it is shown, at a shorter time-scale, the second spike of the laser emission, and there are observed some faster oscillations of period around 100 ps.

between the emitting surface and the detection fiber head, the contrast disappears in all cases and only the pulse envelope remains, as shown in Fig. 1.

In order to explain the oscillations of the collected radiation shown in Fig. 2, account must be taken that the radiation emitted from the powder surface is transversally incoherent [21]. The transverse coherence distance on the emitted wave front is given by

$$d \sim \frac{\lambda L}{\Phi}, \quad (1)$$

where λ is the wavelength, L is the distance between the emission and the detection points and Φ is the diameter of the emitter surface.

Therefore, for a given wavelength, a longer emitter-collector distance implies a higher transverse coherence distance on the wave front which allows for a better coherence sensing. Moreover, the use of a small fiber diameter (detection area) also improves the resolution of the transverse coherence measurement along the emitted

Fig. 2. Time dynamics of RL emission collected with a 1 m, 60 μm core diameter graded-index optic fiber located at 500 mm from the crystal powder surface. The sample and the pumping conditions are the same as those of Fig. 1.

wave front. In our experiments, the wavelength is about 1 μm , the diameter of the pumped zone around 1 mm, and the collecting distances 120/500 mm so, for a coherent source, relation (1) should give a transverse coherence size d of about 120/500 μm which explains the differences in temporal resolution between the pulses shown in Fig. 1, collected with the 500 μm fiber at a distance of 120 mm, and those in Fig. 2, collected with the 60 μm fiber at 500 mm. In the last case we can consider that the detection area, where all the waves emitted from each local incoherent emitter interfere, is nearly punctual (contrast ratio $\sim 1/10$), whereas in the first case, the detection area (500 μm fiber core) is larger than the transverse coherence length (about 120 μm) and as a consequence the full overlapping of the waves emitted from the sample surface by incoherent emitters produces a contrast degradation of the registered oscillations.

In order to better understand the relation between temporal and spatial properties of the RL emission from Nd^{3+} -doped powders, we have modified our experimental setup to be able to collect the laser output only emitted from a predetermined size area. The device simply consisted of a movable 5 cm focal lens, which projects the image of the powder surface over a plane where the fiber head was located. Controlling both powder-lens and lens-fiber head distances we can obtain a well-focused image of the emitting surface in a known plane with the desired lateral magnification. Since the fiber head we use is always the same (60 μm core diameter), we control the size of the area whose emitted radiation is collected, by changing the lateral magnification. So, lateral magnifications from 0.3 up to 3 allow selecting emissions from areas between 200 and 20 μm .

The results of these measurements are summarized in Fig. 3, where we can observe a contrast decrease as the integrated emitting area is increased. The disappearance of the contrast as the emission area is enlarged implies that the emissions from different sample zones are not correlated, though the enveloping curve remains similar

Fig. 3. Time dynamics of three similar RL emission pulses obtained imaging with different lateral magnifications the surface of the powder over a plane where is located the head of a 60 μm core diameter graded-index optic fiber. The sample and the pumping conditions are the same as those of Fig. 1.

for all the local emissions following the pumping pulse. Due to the limitation of spatial resolution of our measurement device, it is difficult to precisely estimate the minimal size of correlated emitters; nevertheless it seems logical to presume it could be of the order of the transport length, which in our samples is of the order of 10 μm [23].

Referring to the oscillations period, it must be emphasized that the relatively fast evolution of the pumping intensity, among other factors, makes it difficult to give an interpretation of the measurements, because the period value can significantly vary within the pumping pulse due to strong changes of the pumping intensity in time intervals comparable to the oscillations period. Despite these difficulties, the average time period can be estimated to be about 100 ps when pumping is not too close to threshold and in general, it shows a slight tendency to decrease as the pump level is increased.

A similar behavior in the period and contrast in oscillations has been observed in different kinds of Nd^{3+} -doped powdered materials tested in our laboratory, like borates, vanadates, phosphates, or oxysulfides. Probably, the photons residence time varies not much in all cases we have worked out, because their grain size

and compactness are not very different. Nor were significant changes observed by using different pumping diameters in the 0.5–2.5 mm range.

It should also be mentioned that though the temporal resolution of our measurement system is about 40 ps this limitation should not have a big influence on the time delay measurements between oscillation peaks, but might be important for peak-width values, possibly shorter than measured.

As we mentioned above, in earlier works the rate-equation model applied to Nd-doped RL [8,12,13] gave a too short time period for the observed oscillations if compared to the known experimental results. As an example, if we apply these rate-equations to the stoichiometric borate and the calculation is done without any consideration about larger photon occupation volumes or residence times so that only active pumped volume is used, the period of the oscillations is in the order of 100 ps [13] which coincides with the one we have obtained in our present experimental results by using an improved time resolution setup. Therefore, it seems reasonable to conclude that the observed quick RL oscillations are relaxation oscillations, probably affected by internal or external interferences between multiple waves, depending on the precise experimental conditions.

As we have demonstrated, the former oscillations observed in the nanosecond time domain can hardly be related to RL relaxation oscillations, but instead its origin should be attributed to pumping fluctuations.

As a conclusion, for the first time to our knowledge, measurements of laser emission from Nd³⁺-doped crystal powders have been done with sufficient temporal and spatial resolution to observe high contrast, quick local oscillations. The way to resolve them consists in selecting a small enough emitting surface area of the powder sample (Fig. 3, 20 μm), as well as in using a small enough area detection device (Fig. 2). The contrast of the oscillations decreases either when the emission or the detection areas are increased (Fig. 3, 200 μm , and Fig. 1, respectively). The decrease of contrast when the detection area increases can be explained in terms of correlation distances on the wave front by using expression (1). The emissions from different sample zones separated by more than a given coherence length ($\sim$ transport length) are not correlated, though the instantaneous enveloping curve is the same for all local emissions. This conclusion is sustained by the disappearance of contrast as the selected emission area is enlarged.

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References

1. D. S. Wiersma, *Nat. Phys.* **4**, 359 (2008).
2. M. A. Noginov, *Solid-State Random Lasers* (Springer, 2005).
3. H. Cao, *Waves Random Media* **13**, R1 (2003).
4. D. S. Wiersma and M. A. Noginov, *J. Opt.* **12**, 020201 (2010).
5. Q. Baudouin, N. Mercadier, V. Guarrera, W. Guerin, and R. Kaiser, *Nat. Phys.* **9**, 357 (2013).
6. B. Redding, M. A. Choma, and H. Cao, *Nat. Photonics* **6**, 355 (2012).
7. A. N. Paddock, *Photon. Spectra S* (Sept. 2012).
8. M. Bahoura, K. J. Morris, G. Zhu, and M. A. Noginov, *IEEE J. Quantum Electron.* **41**, 677 (2005).
9. V. M. Markushev, V. F. Zolin, and C. M. Briskinia, *Sov. J. Quantum Electron.* **16**, 281 (1986).
10. M. Bahoura and M. A. Noginov, *J. Opt. Soc. Am. B* **20**, 2389 (2003).
11. S. García-Revilla, I. Iparraguirre, C. Cascales, J. Azkargorta, R. Balda, M. A. Illarramendi, M. Al-Saleh, and J. Fernández, *Opt. Mater.* **34**, 461 (2011).
12. M. A. Noginov, N. E. Noginova, H. J. Caulfield, P. Venkateswarlu, T. Thompson, M. Mahdi, and V. Ostroumov, *J. Opt. Soc. Am. B* **13**, 2024 (1996).
13. J. Azkargorta, M. Bettinelli, I. Iparraguirre, S. García-Revilla, R. Balda, and J. Fernández, *Opt. Express* **19**, 19591 (2011).
14. G. A. Kumar, J. Lu, A. A. Kaminskii, K. I. Ueda, H. Yagi, and T. Yanagitani, *IEEE J. Quantum Electron.* **42**, 643 (2006).
15. Yu. V. Orlovskii, T. T. Basiev, K. K. Pukhov, M. V. Polyachencova, P. P. Fedorov, O. K. Alimov, E. I. Gorokhova, V. A. Demidenko, O. A. Khristich, and R. M. Zakalyukin, *J. Lumin.* **125**, 201 (2007).
16. I. Iparraguirre, J. Azkargorta, O. Merdignac-Conanec, M. Al-Saleh, C. Chlique, X. Zhang, R. Balda, and J. Fernández, *Opt. Express* **20**, 23690 (2012).
17. C. Gouedard, D. Husson, C. Sauteret, F. Auzel, and A. Migus, *J. Opt. Soc. Am. B* **10**, 2358 (1993).
18. M. A. Noginov, J. Novak, and S. Williams, *Phys. Rev. A* **70**, 063810 (2004).
19. K. L. van der Molen, A. P. Mosk, and A. Lagendijk, *Phys. Rev. A* **80**, 055803 (2009).
20. C. M. Soukoulis, X. Jiang, J. Y. Xu, and H. Cao, *Phys. Rev. B* **65**, 041103 (2002).
21. D. Wiersma and A. Lagendijk, *Phys. Rev. E* **54**, 4256 (1996).
22. M. A. Noginov, S. U. Egarievwe, N. Noginova, H. J. Caulfield, and J. C. Wang, *Opt. Mater.* **12**, 127 (1999).
23. M. A. Illarramendi, I. Aramburu, J. Fernández, R. Balda, S. N. Williams, J. A. Adegoke, and M. A. Noginov, *J. Opt. Soc. Am. B* **24**, 43 (2007).