A performance study of Nd-based stoichiometric random lasers

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ABSTRACT

This work presents a comparative study of the random lasing performance of several Nd-based stoichiometric compounds, including threshold and absolute slope efficiencies together with a discussion about the fundamental parameters which control the random laser operation in this kind of compounds.

Keywords: Random laser; Rare-earth stoichiometric materials; Neodymium

1. INTRODUCTION

The luminescence properties of rare earths (RE) in inhomogeneous dielectric structures including highly scattering powders has received much attention because of its potential applications in lighting and displays in the visible spectrum. In particular, RE-activated oxides have been thoroughly investigated [1]. On the other hand, the recent advent of high optical quality RE-doped ceramic materials has increased the research interest on near infrared emitters based on Nd³⁺-activated powder oxides which have been proved to be suitable for solid state lasers [2,3]. In the lasing action of these materials, the effect of the optical cavity is replaced by multiple light scattering, and the lasing properties are determined by the interplay between scattering and gain. Latest results and theories regarding the so-called random lasers can be found in Refs [4-8], where the mechanisms of the random lasing and the nature of the associated laser modes are discussed.

A growing interest has arisen nowadays in the study of lasing properties of activated powders for optoelectronic and medical applications requiring low coherence such as high resolution imaging and/or highly stable optical frequency among others. Among RE-activated powders those based in Nd³⁺ have received much attention due to their low threshold and high efficiency in almost every host [9-12]. The history and the state of the art of these neodymium based random lasers were reviewed by Mikhail Noginov [13].

The first demonstration of laser-like behavior in a powder was performed in 1986 by Markushev *et al.* [14], using Na₅Nd_xLa_{1-x}(MoO₄)₄ at liquid nitrogen temperature. Since then, similar experiments using a variety of Nd-doped powdered materials and highly scattering Nd-doped ceramics have been performed at room temperature. Among them, stoichiometric NdAl₃(BO₃)₄ powder has many desirable properties such as low laser threshold, high Nd³⁺ concentration and high gain, which, together with good physical and chemical properties have made it a very promising material for room temperature solid-state random lasers [15].

In previous papers the transport-mean-free path (l_t) of NdAl₃(BO₃)₄ and Nd_xY_{1-x}Al₃(BO₃)₄ laser crystal powders by using the diffuse reflectance and transmittance of the powders and the absorption coefficient of the crystal materials have been determined by some of the authors [16-17]. It should be remarked that the knowledge of this parameter is very important to understand the operation regime of random lasers. In a recent paper the authors show a reduction of the onset of laser-like emission and an increase of the slope efficiency when increasing the Nd³⁺ content [18]. At a first glance this behaviour seems to be unexpected due to the luminescence quenching at high concentrations. In principle, the main variable that controls the quenching of the luminescence is the nonradiative energy transfer process among RE ions. The high concentration of Nd³⁺ ions gives rise to short-range spatial energy transfer in the metastable level, as well as long-range spatial migration of energy among Nd³⁺ ions, that may affect the threshold and slope efficiency for laser

action. However, account taken of the short build up time of the random laser pulses (a few tens of picoseconds) which can be much less than nonradiative lifetimes, the lifetime shortening of the excited state, as concentration increases, does not affect the random laser process. It is therefore clear that for a random laser material a high effective gain within the build up time scale of the pulse construction is the important issue. Moreover, from a practical point of view, the low coherence and long emission wavelength of a Nd^{3+} random laser make this type of emission source a very promising one for applications in biomedicine, both for medical theranostic and high resolution bioimaging [19-20].

Following this idea, the authors have investigated RL performance of different solid state stoichiometric neodymium crystal laser powder materials: $\text{NdAl}_3(\text{BO}_3)_4$, NdVO_4 , NdPO_4 , and $\text{Nd}_3\text{Ga}_5\text{O}_{12}$. The main characteristic features of these stoichiometric random lasers are exemplified in section 3 with the $\text{NdAl}_3(\text{BO}_3)_4$, NdVO_4 , and NdPO_4 crystal powders whereas section 4 is devoted to the most performant $\text{Nd}_3\text{Ga}_5\text{O}_{12}$ compound.

2. EXPERIMENTAL

The input/output energy measurements have been performed by using a tunable 10 ns time width Ti:sapphire laser as pump source. The pump beam size over the sample was controlled by means of a 40 cm focal lens mounted in a movable holder. The emission was directly collected (with no lens) on a 0.5 mm diameter optic fiber, vertically located at a distance of 25 cm over the sample. Only optical filters were inserted to attenuate or to remove pumping radiation. The fiber drives the optical signal to a detector directly coupled to a digital oscilloscope. Figure 1 shows a scheme of the experimental setup.

The pumping energy was calibrated by comparing the measurement given by an optical head detector, Ophir PE25BF-V2 ROHS, located at the exact position where powder samples were pumped, and the one provided by a silicon detector which measures the light dispersed by the folder mirror used to address the pump beam. Thus it is possible to measure pumping and output pulse energies simultaneously. The stimulated emission energy was calibrated by using a low energy calibrated head (Ophir PE-10) over the sample, assuming a Lambertian emission [21]. The energy calibration allows the use of absolute scales in the input/output energy representations and therefore, obtaining the absolute value of the slope efficiency.

Fig. 1. Scheme of the experimental setup.

3. MAIN FEATURES OF $\text{NdAl}_3(\text{BO}_3)_4$, NdVO_4 , and NdPO_4 RANDOM LASERS

3.1 Laser slopes

In a previous work, the authors have proved [22] that the slope efficiency of the stimulated emission energy versus incident pumping energy in Nd-doped crystal powders can be approximated (only to powder materials with spontaneous decay times much longer than pumping time and with not too small pump beam sizes) by the following expression:

$$slope = \eta(\lambda_p) \frac{\lambda_p}{\lambda_{EM}} \quad (1)$$

where η is the absorbance of the powder at the pump wavelength and λ_p and λ_{EM} , the wavelengths of the absorbed and emitted radiation respectively. We have tested this expression for all the stoichiometric crystal powders.

Figure 2 shows, as an example, the absorbance results obtained by using a Cary spectrometer with integrating sphere of $\text{NdAl}_3(\text{BO}_3)_4$, NdVO_4 , and NdPO_4 . Note that the maximum absorbances are about 50%, quite similar for the three compounds.

Fig.2. Spectral diffuse reflectance for $\text{NdAl}_3(\text{BO}_3)_4$, NdVO_4 , and NdPO_4 .

Laser output energies as a function of incident pumping energy are displayed in Fig. 3. The corresponding experimental slope efficiencies are 0.33 for $\text{NdAl}_3(\text{BO}_3)_4$, 0.30 for NdVO_4 and 0.36 for NdPO_4 , with an estimated error of ± 0.05 , whereas the slopes obtained from the measured absorbances and expression (1) are 0.37, 0.43 and 0.40 respectively showing a reasonably good accordance with the experimental laser slope efficiencies.

Fig. 3. Laser output energy as a function of incident pumping energy for $\text{NdAl}_3(\text{BO}_3)_4$ [$\lambda_p=810$ nm, $\lambda_{em}=1063$ nm], NdVO_4 [$\lambda_p = 808$ nm, $\lambda_{em} = 1061$ nm], and NdPO_4 [$\lambda_p = 795$ nm, $\lambda_{em} = 1059$ nm]. Pump area is 0.4 mm^2 .

It is worth noticing that these slopes remain unchanged in the three powders for different pumping beam diameters above a given value (about 0.5 mm). Below this pumping size the reproducibility of our results is not good. We have often observed that the surface is affected by pumping, small subsidence (no burning) appears, and the slope and even directionality of the emission are affected. Sometimes we have observed that the slope tends to increase, surely due to an increment of absorbance.

Figure 4 shows, as an example, the wavelength dependence of the experimental slope with absorbance, obtained from diffuse reflectance for the NdPO_4 sample. In accordance with expression (1) it can be seen that almost all absorbed pumping photons above threshold are reemitted as stimulated emission.

Fig. 4. Laser slope efficiency versus pumping wavelength for NdPO_4 with a pump area of 0.4 mm^2 .

3.2 Laser threshold properties

The measured values of the pump threshold energy as a function of the pumping area showed that the threshold energy per unit area remains constant for pump beam diameters above 0.5 mm, and it starts to rise below this value. The energy thresholds per unit area obtained from Fig. 3 were about 8.1 mJ mm^{-2} for NdPO_4 and 3.8 mJ mm^{-2} for $\text{NdAl}_3(\text{BO}_3)_4$ and NdVO_4 samples.

The random laser models predict [22] that the incident pumping threshold energy is given by:

$$E_{th} = \frac{Ah\nu_P}{\eta(\lambda_P)\sigma_{EM}} \sqrt{\frac{l_t}{l_i}} \quad \text{where} \quad l_i = \frac{1}{c\rho\sigma_{ABS}(\lambda_P)} \quad (2)$$

where A is the pumping area, $h\nu_P$ is the energy of pump photons, σ_{EM} is the stimulated emission cross-section, l_t is the transport length and l_i is the inelastic length, being σ_{ABS} the absorption cross-section, ρ the concentration, and c the volume filling factor of the powder. This expression is considered to be valid for not too small pumping areas, because it is obtained from a one-dimensional model.

From expression (2) it is not easy to predict relationships between threshold pumping energies in different materials, because differences in the transport length l_t and/or the inelastic length l_i , also imply differences in absorbance η . For example, the emission cross-section and the absorbance are higher in NdVO_4 than in $\text{NdAl}_3(\text{BO}_3)_4$, but nevertheless, both have a very similar laser threshold. This is justified by a shorter inelastic length in NdVO_4 powder.

3.3 Spatial properties of stimulated emission

A better understanding of the interaction between amplification and diffusion in this kind of lasers, can be experimentally obtained by imaging the emission surface of the powder onto a CCD analyzer (Newport LBP-3) with lateral magnification one.

The obtained results are very similar for all stoichiometric samples and are summarized in Fig. 5 where sizes of pumped and stimulated emission areas can be compared for different pumping areas in NdPO_4 powder.

From these results, a strong competition between diffusion and amplification processes can be deduced. For large pumped areas, the amplification prevails, and the stimulated emission is concentrated in a zone smaller than the pumped region (Figs. 5a and 5b), whereas for small pumped areas, the diffusion is dominant, and the emission is expanded over a region larger than the pumped area (Figs. 5e and 5f).

Fig. 5. Images of the emitting surfaces of NdPO_4 (a-f) powders. The left hand images (a,c,e) are images of the pumped area, since the stimulated emission has been removed by filtering. Those to the right (b, d, f) are images of the stimulated emission area, since the reflected or re-emitted pumping has been removed by filtering. All images have been obtained at input energies approximately twice the threshold. Width of beams at 37% of maximal intensity are: a) 1.49x1.56 mm, b) 1.14x1.24 mm, c) 0.75x0.85 mm, d) 0.77x0.81 mm, e) 0.20x0.20 mm, and f) 0.35x0.35 mm.

4. A HIGH PERFORMANT $\text{Nd}_3\text{Ga}_5\text{O}_{12}$ POWDER CRYSTAL RANDOM LASER

In order to perform RL experiments, the $\text{Nd}_3\text{Ga}_5\text{O}_{12}$ single crystal was powdered in a mechanical mill to average grain size of the order of tens of microns. The diffuse reflectance of the powder is shown in Fig. 6. The maximal absorbance observed in the $^4\text{I}_{9/2} \rightarrow ^2\text{H}_{9/2}, ^4\text{F}_{5/2}$ band is given at 807 nm wavelength, and its value is 56%. This wavelength was selected as pump wavelength in RL experiments, since it gives the minimal input threshold energy and maximal laser slope efficiency.

Fig. 6. Absorbance of the powdered $\text{Nd}_3\text{Ga}_5\text{O}_{12}$ crystal.

Figure 7 shows a pumping pulse and the corresponding stimulated emission pulse at a pumping energy approximately three times the threshold energy. It is worth noticing that, after the threshold is reached, the random laser intensity closely follows the pump intensity. The temporal resolution of the measurement system was about 300 ps. From Fig. 7 it follows that the photon residence time of the laser system, which is of the order of the response time, is very short compared to the spontaneous decay time (300 ns). Even if as in this case, the quenching effects are very strong, and therefore the main decay channels are non-radiative, the response of the stimulated emission is faster.

Fig. 7. Pump pulse intensity (below) and the corresponding output laser intensity (above) as a function of time. The oscillations of the pump intensity are due to the longitudinal mode beating. The pumping energy is approximately three times the threshold.

The spectrum of the laser emission, measured driving the emission through the optical fiber to a spectrometer, simply consisted on a single and intense narrow peak at 1061.8 nm, just at the maximum of the spontaneous emission band. The

wavelength of this emission line is independent of the pump wavelength, and it has a spectral width of less than 0.3 nm (the resolving power of the detection system).

The laser output energy versus pumping energy is shown in Fig. 8. For a diameter of pump beam of 0.66 mm and a pumping wavelength of 807 nm, a slope efficiency of 0.42 ± 0.03 and a threshold pumping energy close to 2.2 mJ was obtained (6.5 mJ/mm^2). Both the laser slope efficiency and the pumping threshold energy per unit area are not dependent on the pump spot size.

Fig. 8. Output laser energy versus pumping energy for powdered $\text{Nd}_3\text{Ga}_5\text{O}_{12}$ crystal. The experimentally obtained laser slope efficiency is 0.42 and threshold input energy is 2.2 mJ using a pump beam diameter of 0.66 mm.

5. CONCLUSIONS

Absolute slope efficiencies of four stoichiometric Nd-based random lasers have been measured. The higher slope corresponds to the $\text{Nd}_3\text{Ga}_5\text{O}_{12}$ garnet crystal powder with 42% followed by NdPO_4 , $\text{NdAl}_3(\text{BO}_3)_4$, and NdVO_4 with 36%, 33%, and 30% respectively. The lowest threshold, 3.8 mJ mm^{-2} , corresponds however to NdVO_4 and $\text{NdAl}_3(\text{BO}_3)_4$ compounds followed by the $\text{Nd}_3\text{Ga}_5\text{O}_{12}$ garnet, 6.5 mJ mm^{-2} , and NdPO_4 , 8.1 mJ mm^{-2} . It is worth noticing that the higher slope efficiency of the garnet compound is related to its higher absorbance because, above threshold, the absorbed emission is reemitted as stimulated emission.

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