Random laser model for Nd$^{3+}$-doped powders and its application to stimulated emission cross-section calculations

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Abstract: A new theoretical approach for random lasing of Nd$^{3+}$-doped powders is presented. The model’s singularity lies in the fact that it proposes a probability distribution of the stimulated photon paths lengths in the sample, as well as a population inversion shared by different photon paths. The model’s predictions satisfactorily compare with the results of laser threshold and absolute input/output energy slope of a real Random Laser. The main novel issue is that the best fit to the experimental results requires considering a high level of shared population inversion among different paths. The model simulation of local and spatially integrated Random Laser emissions, as well as their time evolution, is in accordance with the experimental behavior. These results also provide a new method of measuring the stimulated emission cross-section in this kind of random laser materials.

1. Introduction

Scattering materials have shown their attractiveness as laser sources due to potential applications such as high definition speckle-free imaging, holographic laser displays, optical sensors for medical applications, lithography, miniature spectroscopy, etc [1–4].

Unlike conventional lasers, Random Laser (RL) sources are cavityless; multiple scattering confines the light and the random lasing is emitted in all directions with very low spatial coherence. However, as in conventional lasers, when pumping RLs above a threshold value, the emission pulse becomes suddenly some orders of magnitude more intense and spectrally narrowed.

A very wide range of materials have been tested as RL sources, such as crystal powders, colloidal dye solutions, polymers, biological samples, etc [5–14], showing very different behaviors in emission spectra, energy efficiencies and pulse time-ranges.

Although there have been many previous attempts to develop theoretical models for RLs based on Nd$^{3+}$-doped crystal powders, the current results found in the literature display wide dispersion in their predictions of threshold and output energies and temporal dynamics [13–27]. A clear connection between experiment and theory is still lacking to date.

All models assume that transport phenomena are fast compared to other processes [25]. On the other hand, the laser emission has no spectral structure in this kind of compounds.

In general, there are two approaches to the problem. One is the use of diffusion equations including gain. See [18,24,25,27] for the one dimension approach, based upon which, the authors had extended the numerical calculations to two dimensions [28] showing that diffusion equations model is incomplete, at least for Nd doped powders. On the other hand, in these models the emission area tends to grow indefinitely with growing pumping energies, and on the other hand, related to the above, the output energy grows faster than linearly, even
well above threshold. Neither of both results has an experimental support in Nd compounds [29].

The other approach is the use of rate equations [13,14,16,18,20,26]; in all these cases, a unique length (average) is used to estimate the losses in paths, which in our opinion is an excessive simplification, which fails to simulate the behavior of the system. This average length is provided by the diffusion theory and it turns out to be proportional to the inelastic length (inverse of the absorption coefficient) [19,20,24]. These models are not able to provide results about absolute output energy, and their prediction of temporal behavior does not fit the experimental evidence [30].

In this work, we propose and develop a theoretical model that satisfactorily explains the experimental results about both the energetic and temporal behavior of Nd$^{3+}$-doped crystal powders. The model includes a statistical distribution of path lengths, and a way for the different paths to share the population inversion. The behavior of the threshold, absolute input/output energy, and the yield of RL as a function of the different diffusion and absorption parameters as well as the temporary behavior were tested by the model, and in all cases a good simulation of the experimental results was achieved.

Finally, the empirical application of the results of the model leads to a simple equation which allows to obtain reliable stimulated emission cross-sections by making use of experimental data.

2. Theoretical model

Our model assumes as an approximation that the region pumped by the incident beam is a disk-shaped volume, located in the plane that limits a semi-infinite volume of the powdered material. Its depth is equal to the diffusive absorption length of the incident radiation and its diameter is equal to that of the pumping beam on the surface of the material. If this diameter is much longer than the absorption length, the edge effect in the lateral limits of the pumped region can be neglected. The population inversion is assumed to be uniform in this disk.

According to the conventional diffusion theory, the (diffusive) absorption length, that is, the penetration depth of pump radiation, is given by $l_{abs} = (l_t l_i / 3)^{1/2}$, where $l_t$ is the transport length or mean diffusion length, and $l_i$ the inelastic length, inverse to the absorption coefficient of the bulk material, which is equal to the absorption cross-section of the pump radiation multiplied by the ion concentration per unit volume, corrected by the filling factor of the powder.

Our model assumes that light generated in the powdered material follows a multitude of paths with different possible lengths and gains, which always end up on the emitting surface. Many different paths can cross the pumped volume and parts of each path may go through no pumped regions, since the emitted radiation is not absorbed (four level laser) and can be diffused into the material out of the pumped disk. Thus, we shall only consider the “active length” of these paths, that is, the length along which the photons are amplified, and not the total length, as essential. The “active length” will indicate that the energy emitted from a given path reaching threshold is essentially given by its absorbed energy and not by its “real” length. We do not care about the microscopic random walk picture of the individual photons, and just consider a collection of photon beams which propagate independently through the sample and follow discrete paths in which the photon densities and shared population inversion may change as a function of time, due to stimulated and/or spontaneous emissions and losses. Moreover, as we consider a large enough amount of photons and inverted population in each path, the time evolution of the system is followed by using their average densities. The wavelength of the incident radiation that fills non radiatively the $^{4}I_{3/2}$ metastable laser level is about 800 nm, whereas the wavelength of the emitted laser radiation is always close to 1064 nm; the pumped energy per unit area is about 10 mJ/mm$^2$. Transport lengths are around ten microns, much longer than wavelengths involved, and the pumping
beam diameter on the powder surface is much longer than the absorption length, typically of a few tens of microns.

Assuming a path of active length $x$, $N(x,t)$ denotes the mean population inversion in particles per unit volume along this path, and $\phi(x,t)$ the mean photon density in the same path, both as a function of time $t$. Under these assumptions, the evolution of these variables is given by:

$$\frac{dN(x,t)}{dt} = W - AN - c\sigma\left[pN\phi + \frac{(1-p)}{S_{\text{abs}}} \int_{0}^{\infty} n(x')V(x')\phi dx'\right]$$

(1)

and

$$\frac{d\phi(x,t)}{dt} = \beta AN + c\sigma N\phi - \frac{c}{x}\phi$$

(2)

where $c$ is the mean speed of light in the material, $n(x)$ is the number of paths of active length $x$ per unit length, $V(x) = sx$ is the volume associated to a path of active length $x$, being $s$ its cross section, $S$ is the area of the pumping beam on the surface of the powder, $\sigma$ is the stimulated emission cross-section, $W$ is the spatially averaged absorption rate (in number of particles per unit volume and time), $A$ is the spontaneous decay probability per time unity, and $\beta$ is the ratio of spontaneous emission that is added to the stimulated one. The parameter $p$, that defines the contribution of the population inversion to the stimulated emission in a concrete path, deserves some clarification. Our model assumes that, when an excited ion decays in a path by the emission of a stimulated photon, this photon may have been emitted in the same path or in another different one. This effect is introduced by means of this parameter $p$, with $p = 0$ standing for the limiting case in which the emission in any path is shared by all paths, and $p = 1$ the opposite case, in which the emission in a path causes a decrease in the population inversion only in that path.

The relationship between the average absorption rate $W$ and the absorbed power $P_{\text{abs}}$ or energy $E_{\text{abs}}$ is given by

$$W(t) = \frac{P_{\text{abs}}(t)}{hvS_{\text{abs}}} = \frac{E_{\text{abs}}}{hvS_{\text{abs}}} f(t)$$

(3)

where $h$ is the Planck constant, $v_p$ is the frequency of pumping radiation, and $f(t)$ is a normalized function that defines the temporal shape of the pumping.

The output energy is given by:

$$E_{\text{out}} = \int P_{\text{out}}(x) dx, \quad \text{with} \quad P_{\text{out}} = hv\int_{0}^{\infty} n(x)V(x)\frac{c}{x}\phi(x) dx$$

(4)

where $v_e$ is the frequency of emitted radiation. We must emphasize that the calculation (as well as the measurements) of the output energy is made by only recording while the laser emission lasts and not indefinitely, which would include the residual spontaneous emission. Therefore, it can be said that only the stimulated emission is present, both in calculations and measurements.

Following the hint for the transport path distribution in [31], an exponential distribution for the path distribution $n(x)$ is assumed.

$$n = \frac{M}{x_0} \exp\left(-\frac{x}{x_0}\right)$$

(5)

where $M (>>1)$ is the total number of paths considered.

The total volume of the paths, which should be equal to the volume where energy is absorbed, can be calculated by using expression (5):
\[
\int_0^n n(x)W(x)\,dx = V_{\text{total}} = SI_{\text{abs}} = Msx_0
\]  

(6)

and from (4), (5) and (6) we obtain the output power in photons per unit time and unit pumped area by:

\[
P_{\text{out}}(t) = \frac{cL_{\text{abs}}}{x_0^2} \int_0^\infty \exp \left( -\frac{x}{x_0} \right) \phi(x,t)\,dx
\]

(7)

and the population inversion rate, by:

\[
\frac{dN(x,t)}{dt} = W - AN - c\sigma \left[ pN\phi + \frac{(1-p)}{x_0^2} \int_0^\infty x'\exp \left( -x'/x_0 \right) \phi N\,dx' \right]
\]

(8)

where magnitudes \( s \) and \( M \) have disappeared, which is essential for the consistency of the model.

### 3. Results from the numerical model in the energy domain

The values of the parameters introduced by default in the model equations are \( A = 10^5 \text{ s}^{-1}, c = 2\times10^8 \text{ ms}^{-1}, \beta = 0.01, l_i = 10 \text{ \mu m}, l_f = 40 \text{ \mu m}, \sigma = 10^{-22} \text{ m}^2, \) and \( f(t) \) is a normalized Gaussian function of 5 ns width, centered at 5 ns.

These values are reasonable for the problem we are dealing with. Regarding parameter \( \beta \), its effects have already been studied both in conventional and RL works [27], where there are no geometrical restrictions. We have estimated its value taking into account the emission quenching, the spectral narrowing of the stimulated emission, and the spontaneous emission to levels other than the lower laser level. In any case, it must be born in mind that the effect of great changes in the value of \( \beta \) is irrelevant for the results of the model, which is related to the pulsed character of the laser and with the limited time window of recording output energy.

First we analyze the effect of parameter \( p \), shown in Fig. 1, where the output energy (in photons per unit area) is plotted against absorbed energy (also in photons per unit area). When \( p = 0 \) the result in output/input energy is a line of slope unity from the origin, which is totally unreal. We must bear in mind that in this case (see Section 2), we are assuming that if a single path (as long as we want) begins to lase, it detracts population inversion from all the other paths, at the same rate as from itself, and then threshold would be zero, which does not make any sense, given the experimental evidence. However, when \( p \) is not too close to zero, the results of the model can be realistic, pointing to the evident fact that at least part of the population inversion must be detracted from the path itself because population inversion is only partially shared. Therefore, to start with, we can only say that \( p \) must be greater than zero, and noticeably smaller than one, because in this case the curvature of the energy output/input function is better simulated, as will be discussed below.

As the RL experiments show [12,29,32,33], far enough above threshold, the output/input energy function asymptotically tends to a line of slope one. As can be seen in Fig. 1, in our model, there is a noticeable dependence on parameter \( p \); the higher slopes correspond to the smaller values of parameter \( p \). On the opposite side, if we focus our attention on threshold, Fig. 1 also shows the very weak dependence on \( p \) of the apparent threshold energy, except for too low \( p \) values.
Fig. 1. Output energy in photons per unit area as function of absorbed energy also in photons per unit area, for different values of $p$ parameter and assuming $x_0 = l_{abs}$.

On the other hand, Fig. 2 shows the results for different values of $l_i$ assuming $x_0 = l_i$, the inelastic length. This approach, based on one-dimensional RL models [19,20,24], would imply that the average path length $x_0$ be proportional to the inelastic length $l_i$. It is easy to verify that under this assumption the threshold energy is actually proportional to the square root of the ratio between transport and inelastic lengths. However, these results are against the experimental evidence, according to which, for a given sample, the threshold energy per unit area only depends on the sample absorbance at the pump beam wavelength [29,31].

In our opinion, the clue is that one-dimensional models consider path lengths for passive diffusion, while in our model the key length is what we have called “active length”, the one in which amplification takes place. The distribution of “passive path lengths” usually given by the diffusion theory which leads to an average length proportional to the inelastic length (assuming a similar transport length at pumping and emission wavelengths), does not represent the real amplification path distribution involved in RL emission.

If we perform our calculations taking $x_0$ equal to the absorption length $l_{abs}$, the energy results are not altered in any way by changes in transport or inelastic lengths.

Likewise, it can be shown that the threshold absorbed energy per unit area is inversely proportional to the stimulated emission cross-section, as is shown in Fig. 3, for two different values of $p$. Therefore, the product of the apparent threshold absorbed energy per unit area multiplied by the stimulated emission cross-section is a constant, approximately 0.2.
Fig. 2. Output energy in photons per unit area for two different values of $p$ parameter and three different inelastic lengths, assuming $x_0 = l_i$.

Fig. 3. Output energy in photons per unit area as function of absorbed energy in photons per unit area for two different values of $p$ parameter and two emission cross-sections, and assuming $x_0 = l_{abs}$.

We must point out that the experimental observation of the laser threshold is somehow uncertain, because it depends on factors such as the sensitivity of the detectors, the range of working energies, and even the human factor, since the apparent input threshold energy is defined when a minimal appreciable signal is detected above the one coming from spontaneous emission, and this minimal signal is relative and can be subjective.

On the other hand, we must keep in mind that though transport and inelastic lengths do not have an explicit influence, since we work with absorbed energy (threshold as a function of the incident energy) they do so implicitly, since they affect the diffusive absorbance of the powdered material.
Let us compare the results of the model in terms of threshold and shape of the efficiency curves, with our previous experimental results. In Fig. 4 we show the experimental energy output/input of a Neodymium Gallate [29,32,33] together with the results of our simulation using different values of \( p \) and \( \sigma \). The optimal fit is obtained for values of \( p \) slightly lower than 0.1 and the stimulated emission cross-section is not very sensitive to changes in \( p \).

Figure 5 shows the theoretical fitting for a Neodymium Borate [29,32,34]. The best \( p \) value is also close to 0.08 as in the former case. On the other hand, the mean slope efficiency of the obtained curves is slightly below the unit, close to experimental slopes [29]. We first thought that this could be due to spurious losses of the stimulated emission, but these results lead us to believe that it is intrinsic to the laser process in these diffusive materials, because slopes are very close to one only well above threshold.

The non-linearity observed in output/input energy close to the threshold is observed also in some recent works [35]. Obviously, the instability is much greater close to the threshold than far above, in line with other works that deal with fluctuations in energy [36,37].
4. Measurement of the stimulated emission cross-sections

Although the values of the fitted cross-sections in Figs. 4 and 5 are not absolute, because there are scale factors to be determined ($x_0$, $l_{abs}$ and others), they may allow to be compared with each other. In both cases, the product between the apparent threshold energy (in number of photons per unit area) and the cross section is approximately 0.2. Therefore, it is reasonable to conclude that the ratio between both emission cross-sections should be a meaningful number.

Although the constancy of the product between threshold energy and emission cross-section cannot be given for granted, the fact that the different samples studied show similar values for $p$, with similar absorbance, suggests that in samples with not very different absorbance, the $p$ values would scarcely change, and as a consequence, the observed results could be generalized.

In summary, for powders that meet the conditions expressed in sections 2 and 3 and excited at similar wavelengths and pumping areas, we can state that the following expression is fulfilled:

$$\eta E_{th}^6 \sigma = \text{constant}$$  \hspace{1cm} (9)

where the constant is the same for different powders. In this expression, $E_{th}^6$ is the incident pump threshold energy and $\eta$ is the diffuse absorbance of the powder. This last magnitude is usually measured by means of an integrating sphere, and therefore $E_{abs} = \eta E$, being $E$ the incident pump energy. By using expression (9) it is possible, to compare stimulated emission cross-sections of different materials. If we have a standard powder sample with a well-known emission cross-section, by measuring the threshold incident energy and the absorbance in this powder we can obtain the value of the emission cross-section of any other material.

If we take the product of threshold energy per unit area by cross-section equal to 0.2, assuming $x_0 = l_{abs}$, expression (9) takes the simple form:

$$\eta \sigma E_{th} / h\nu \rho S \equiv 0.2$$  \hspace{1cm} (10)
It is worth noticing that the results obtained from (10) give a good absolute value for one of the best known stimulated emission cross sections, the one of Nd:YAG [38]. Thus, taking this advantage, Table 1 displays the values of the stimulated emission cross-sections of several compounds, for the 1064 nm transition, by using the RL experimental thresholds [29] in expression (10).

Table 1. Stimulated emission cross-sections for \(^{4}F_{3/2} \rightarrow ^{4}I_{11/2}\) transition of Nd\(^{3+}\) ion in different materials obtained from (10).

| MATERIAL | \( \lambda \) pump \( \text{nm} \) | Diffuse absorbance \( \eta \) at \( \lambda \) pump | Threshold (mJ/mm\(^2\)) | Stimulated emission cross-section \( \times 10^{23} (\text{m}^2) \) |
|----------|-------------------|------------------|--------------------|-------------------|
| YAG: 1% Nd | 808 | 0.26 | 8.0 | 3.0 |
| NdAl\(_3\)(BO\(_3\))\(_4\) | 810 | 0.47 | 4.0 | 3.3 |
| NdPO\(_4\) | 797 | 0.52 | 8.0 | 1.5 |
| La\(_2\)O\(_2\)S: 2% Nd | 819 | 0.13 | 3.0 | 16 |
| La\(_2\)O\(_2\)S: 9% Nd | 819 | 0.23 | 2.0 | 14 |
| Nd\(_3\)Ga\(_5\)O\(_12\) | 807 | 0.56 | 6.5 | 1.7 |
| LiNdP\(_4\)O\(_12\) | 800 | 0.48 | 13.0 | 1.0 |
| NdVO\(_4\) | 808 | 0.54 | 4.0 | 2.9 |

5. Results from the numerical model in the temporal domain

The temporal behavior of the output intensity is another fundamental aspect of how a RL operates. However, to check this model issue, we must first specify which kind of output is considered and how it has been measured.

In Fig. 6 we show the output intensity as a function of time corresponding to individual paths of a given length as provided by our model, where (a, b, c) correspond to three paths of different lengths. Chosen lengths are one below threshold (Fig. 6(a)), another one not very far above threshold (Fig. 6(b)) and a third one far above threshold (Fig. 6(c)). In all cases, relaxation oscillations are observed above threshold with periods that change according to specific conditions such as path length or pumping energy, but always lie in the 100-200 ps range. Nevertheless, we should make clear that these intensities are unmeasurable, since it is not possible to separate or distinguish different paths.

On the other hand, the result provided by the model for the total output intensity, that is, the sum of intensities emitted by all paths is shown in Fig. 7(a). In this case, the output intensity essentially follows in time the pumping intensity (after build-up of the laser pulse), as observed in experiments [30], and in Fig. 7(b) [39]. This kind of measurements can be performed, for example, by collecting emissions directly from the entire emitting surface with a multimode optical fiber.

Although separate paths cannot be measured, we can measure the emission from small areas, of a size of the order of the distance of minimum transversal coherence, the transport length, which is about 10 microns. This can be achieved by using a lens that projects the image of the emitting surface on a plane where a pin-hole of the appropriate size [30] is placed. Under these conditions, no periodical fluctuations with a time range of tens of ps are detected. From the point of view of the model, this measuring way is equivalent to adding the amplitudes coming from many different paths, all emitted from the same small area. The phases of these amplitudes are random, and they change in a time of the order of about 5 ps, which is the coherence time of the stimulated emission photons in the material [40]. The results provided by the model, shown in Fig. 7(c) yield a good simulation of the
experimentally observed fluctuations in these conditions (see references [29,30] and Fig. 7(d)). In this simulation a broadening of 20 ps by convolution has been included in order to take into account the widening effect of our measuring system.

In summary, the observed intensity is the result of the internal overlapping of the amplitudes from many paths. It is possible to observe the same effect with external overlapping of amplitudes from all paths, by directly using (with no lens) a detector (or fiber) meeting the condition that the emission wavelength multiplied by the sample-detector distance and divided by the square of the detector size be less than unity [32]. In other case, the fluctuations tend to disappear in the measured intensity, and the result is the one obtained in Fig. 7(b).

Fig. 6. Output intensities in relative units as function of time. The absorbed pump is 7x10^{21} photons/m^2. In all three Figs., black line shows the pump pulse time shape, and red lines as follows: a) Output from a below threshold path (x = 10 μm). b) from an above threshold path (x = 25 μm) c) from a far above threshold path (x = 120 μm).
6. Summary and conclusions

A rate-equations model based on photon paths in an amplifying medium has been developed for RL in Nd$^{3+}$-doped powders. The novelty of the model lies in the fact that a probability distribution is used for the lengths of the paths in the rate equations, as well as their possibility of sharing population inversion to a certain degree.

The analysis of the model prediction and its comparison with previous experimental results indicate that the average “active length” (length in which amplification is produced) is proportional to the diffusive absorption length and not to the inelastic length. On the other hand, the inverse proportionality between threshold energy per unit area and stimulated emission cross-section is shown. Likewise, the time evolution predicted by our model accords well with local and integral experimental measurements performed in these powdered compounds.

The experimental slope efficiency, measured in photons per unit area, is well reproduced by the model, the slope tending to one as pump energy grows. However, to achieve a good adjustment the introduction of a high level of path overlapping (that is, paths sharing population inversion) is required. Even though the issue of the emission area is not treated by the model, our results suggest an important emission relocation, in the sense that a photon emitted into a path causes a decrease in population inversion in other paths. This
consequence of the model is supported by the experimental evidence that shows that the emission area on the powder surface hardly grows once the threshold has been reached.

Our results support a new method for comparative, and even absolute measurements of the stimulated emission cross-section in this kind of compounds. Besides, it is noteworthy that this method may avoid the difficulty bound to other indirect methods, such as the use of Judd-Ofelt calculations.

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