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Analysis of lasing efficiency in neodymium doped laser crystal powders

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ABSTRACT

This work presents a theoretical study of the dependence of the efficiency of laser crystal powders on the sample thickness, on the fraction of volume occupied by the particles, and on the mean particle size. An analytical expression for the lasing efficiency in terms of the sample parameters has been worked out at stationary regime. The expression describes correctly the experimental behaviour of the efficiency of neodymium doped laser crystals powders.

1. Introduction

A random laser is a system formed by randomly distributed scatterers embedded in a host medium, with the particularity that the scatterers or the host medium or both provide optical gain through stimulated emission. In this type of lasers the feedback mechanism is provided by random light scattering caused by the spatial inhomogeneity of the medium [1,2]. This topic has raised a rapidly growing interest due to many reported observations of random lasing in a wide variety of systems, like powders of semiconductor nanoparticles [3–5], organic dyes in strongly scattering media [6–10], organic films or nanofibers [11–13] and ceramics [14]. A detailed discussion about the physics of random lasers can be found in a recent publication by Wiersma [15]. Random lasers based on compacted laser crystal powders (LCP) have no spatial coherence. Besides, they are not stable in phase and the corresponding photon statistics are strongly different from those of a conventional laser. The potential applications of LCP as compact and mirrorless lasers, which are convenient when the coherence is not necessary or the absence of coherence is desirable, motivate the study of their laser properties. As in conventional lasers, the most important properties of a random laser are the lasing threshold and the efficiency. In this work, we present a theoretical study of the efficiency in several laser crystal powders doped with neodymium. An analytical expression for lasing efficiency in terms of the sample parameters has been worked out at stationary regime. Particularly, we have determined the dependence of efficiency on the sample thickness, on the volume fraction occupied by the particles and on the mean particle size. From the analysis, an expression that describes the dependence of the lasing threshold on the sample parameters has been obtained. In addition, it is shown that the performance of the random laser in stationary regime serves to predict its performance in pulsed regime, and vice versa. The theoretical calculations have been made by assuming a diffusive propagation of light in these materials, since the mean-free-path between scatterers in the photon transport in these media is usually much larger than the light wavelength [16,17]. The mean-free-paths have been calculated from the Mie theory for spheres in the independent-scatterer approximation [18].

2. Theoretical assumptions

The propagation of absorbed and emitted photons through a sample of LCP is described by two diffusion equations, each one corresponding to each kind of photons. The volume densities of both kinds of photons are coupled by a rate equation corresponding to the inversion of population of the active ions. For simplicity we have considered a plane wave incident along the z direction upon a slab sample whose dimensions x and y are much larger than the z-dimension. Under this hypothesis the general diffusion equation reduces to the one-dimensional case (the z-direction) and the evolution of the system is described in our model by three differential coupled equations. In pulsed regime, the equations are:

$$\frac{\partial W_p(z,t)}{\partial t} = D_p \frac{\partial^2 W_p(z,t)}{\partial z^2} - \frac{D_p}{l_{\mathrm{abs}}^2} W_p(z,t) + p(z,t) \tag{1}$$

$$\frac{\partial W_e(z,t)}{\partial t} = D_e \frac{\partial^2 W_e(z,t)}{\partial z^2} + f\, v\, \sigma_{\mathrm{em}} N(z,t)\, W_e(z,t) + \beta \frac{N(z,t)}{\tau_s} \tag{2}$$

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$$\frac{\partial N(z,t)}{\partial t} = f\,\mathrm{v}K_{\mathrm{abs}}\,W_p(z,t) - f\,\mathrm{v}\,\sigma_{em}N(z,t)\,W_e(z,t) - \frac{N(z,t)}{\tau_s} \quad (3)$$

where $W_p(z,t)$ and $W_e(z,t)$ are the absorption and emission photon densities, $N(z,t)$ is the density of active ions in the excited state, $\mathrm{v} = c/n_{\mathrm{eff}}$ is the light speed in the medium with effective refractive index $n_{\mathrm{eff}}, \sigma_{\mathrm{em}}$, is the stimulated emission cross section, τ_s is the excited state lifetime, K_{abs} is the absorption coefficient of the bulk material at the pump wavelength and $l_{\mathrm{abs}} = \sqrt{l_t l_i/3}$ is the value of the diffusive absorption length. The definitions of the mean-free-paths involved in the scattering and absorption processes (scattering: l_s, inelastic: l_i, extinction: l^*, transport: l_t) are given elsewhere [16]. $D = \mathrm{v}l_t/3$ is the light diffusion coefficient and β is the fraction of spontaneous emission contributing to the laser process. The fraction of volume occupied by the scatterers, f, has been included in the equations to take into account the effective fraction of photons that penetrates inside the particles. In pulsed regime, the real source of diffuse radiation, $p(z,t)$, is usually a Gaussian pulse entering the sample in the z direction, which is exponentially extinguished (scattered and absorbed) along the scattering sample. The system of equations is solved with the following boundary and initial conditions:

$$W_p(-l_e^0,t) = W_p(L+l_e^L,t) = W_e(-l_e^0,t) = W_e(L+l_e^L,t) = 0\;\forall\, t$$
$$W_p(z,0) = W_e(z,0) = N(z,0) = 0\;\forall\, z \quad (4)$$

where the extrapolation lengths are given by $l_e^0 = (2(1+r_0)l_t)/(3(1-r_0))$ and $l_e^L = (2(1+r_L)l_t)/(3(1-r_L))$, where r_0 and r_L are the internal reflectivities at the front ($z = 0$) and rear ($z = \mathrm{L}$) surfaces

The set of coupled nonlinear partial differential Eqs. (1)–(3), has been numerically solved by the Crank–Nicholson finite-difference method for an incident Gaussian pulse to describe the lasing behaviour in LCP doped with neodymium [19] and in dye-doped silica gel powders [20]. The lasing efficiency has been calculated according to the method shown in [19].

For a continuous pumping $(\partial W_p(z,t)/\partial t = \partial W_e(z,t)/\partial t = \partial N(z,t)/\partial t = 0)$, the equations of the model (1)–(3) can be reduced to the following two equations:

$$0 = D_e \cdot \frac{d^2W_e(z)}{dz^2} + f\,\mathrm{v}\cdot K_{\mathrm{abs}}\cdot W_p(z) \quad (5)$$

$$0 = D_p \cdot \frac{d^2W_p(z)}{dz^2} - \frac{D_p}{l_{\mathrm{abs}}^2}\cdot W_p(z) + p(z) \quad (6)$$

In this case, the source of diffuse radiation would be the light entering the sample in the z direction, which is extinguished (scattered and absorbed) along it according to the expression $(J_0/l_s)\exp(-z/l^*)$. J_0 is the flux of incident photons per unit area. In the same way as in other works [16], we can replace the incoming light beam at the boundary by a source of diffuse radiation located at a certain plane inside the sample. Therefore, it is assumed that the incident photons are left inside the sample at the position $z = l^*$. The source is expressed as $p(z) = J_0\delta(z - l^*)$.

The lasing efficiency (η) has been calculated from the ratio of the emitted intensity to the incident radiation, as follows:

$$\eta = \frac{\lambda_{\mathrm{pump}}}{\lambda_{\mathrm{em}}}\left(\frac{-D_e\frac{dW_e(z)}{dz}\Big|_{z=0}}{J_0}\right) \quad (7)$$

where $(-D_e(dW_e(z)/dz)|_{z=0})$ is the photon flux per unit area corresponding to the emitted photons at the front sample surface ($z = 0$) along the backward direction. The analytical expression obtained for η is the following:

$$\eta(L,f,\bar{\phi}) = \frac{\lambda_{\mathrm{pump}}}{\lambda_{\mathrm{em}}}\left(\frac{L + l_e - l^* - (L+2l_e)\,Cosh\left(\frac{l_e}{l_{\mathrm{abs}}}\right)Csch\left(\frac{L+2l_e}{l_{\mathrm{abs}}}\right)Sinh\left(\frac{L+l_e-l^*}{l_{\mathrm{abs}}}\right)}{(L+2l_e)}\right) \quad (8)$$

The variation of the efficiency with the volume fraction occupied by the particles and with the mean particle size has been obtained by calculating the mean-free-paths from the Mie theory in the independent-scatterer approximation. In this theory the mean-free-paths vary linearly with the particle size and depend inversely on the volume-filling factor ($l \sim \bar{\phi}$ and $l \sim 1/f$). In our case, we have assumed that the dependence of the inelastic mean-free-path with $\bar{\phi}$ is negligible [18].

For large thicknesses L, Eq. (8) can be simplified to:

$$\eta(f,\bar{\phi}) = \frac{\lambda_{\mathrm{pump}}}{\lambda_{\mathrm{em}}}\left(1 - \frac{e^{-\frac{l^*}{l_{\mathrm{abs}}}} + e^{-\frac{2l_e+l^*}{l_{\mathrm{abs}}}}}{2}\right) \quad (9)$$

By using the expressions of the reflectance (R), the transmittance (T) and the absorptance ($A = 1–R–T$) worked out for the same source of diffuse radiation [18], Eqs. (8) and (9) can be written, respectively, as:

$$\eta = \frac{\lambda_{\mathrm{pump}}}{\lambda_{\mathrm{em}}}\left(A + T - \frac{l^* + l_e}{L + 2l_e}\right) \quad (10)$$

$$\eta = \frac{\lambda_{\mathrm{pump}}}{\lambda_{\mathrm{em}}}(A) \quad (11)$$

The last equation indicates that the lasing efficiency is just proportional to the absorptance of the sample when its thickness is very large.

3. Results and discussion

Figs. 1–3 show the dependence of η on the sample thickness (L), on the volume-filling factor (f) and on the mean particle size ($\bar{\phi}$), respectively. The study has been done in ground powders of $Nd_3Sc_3(BO_3)_4$ and $Nd_{0.5}La_{0.5}Al_3(BO_3)_4$ laser crystals. In the three figures the solid line is the efficiency in stationary regime obtained from Eq. (8) and the triangular points are the efficiencies in pulsed regime obtained from numerical calculation [19]. As can be seen,

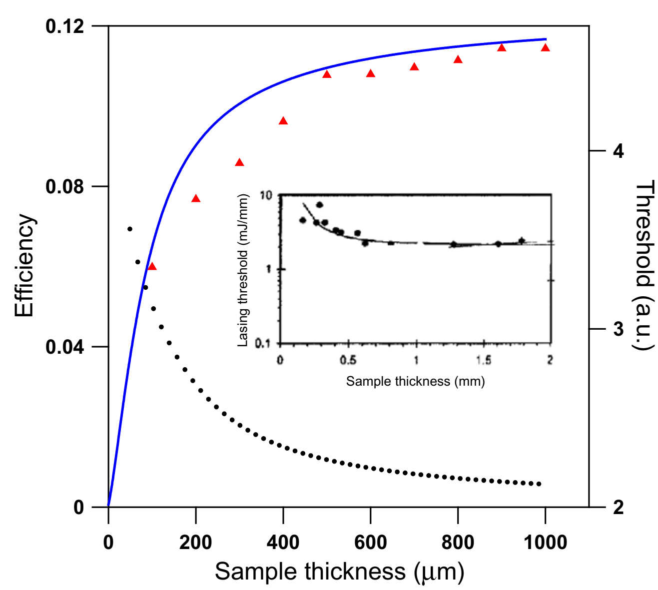

Fig. 1. Efficiency as a function of the sample thickness in $Nd_3Sc_3(BO_3)_4$ powders. The triangles represent the values obtained numerically for an incident Gaussian pulse [19] and the solid line is the efficiency for the stationary regime. The dots represent the lasing threshold. The input values for the calculations are: $\bar{\phi}$ = 4 μm, f = 0.55, σ_{em}(1061 nm) = 2 × 10^{-18} cm^2, τ_s = 24 × 10^3 ns, K_{abs} (532 nm) = 18.2 cm^{-1}, n_{eff} = 1.4, β = 1. The inset corresponds to the experimental results obtained for the same material. It has been reproduced from Fig. 3 of [21].

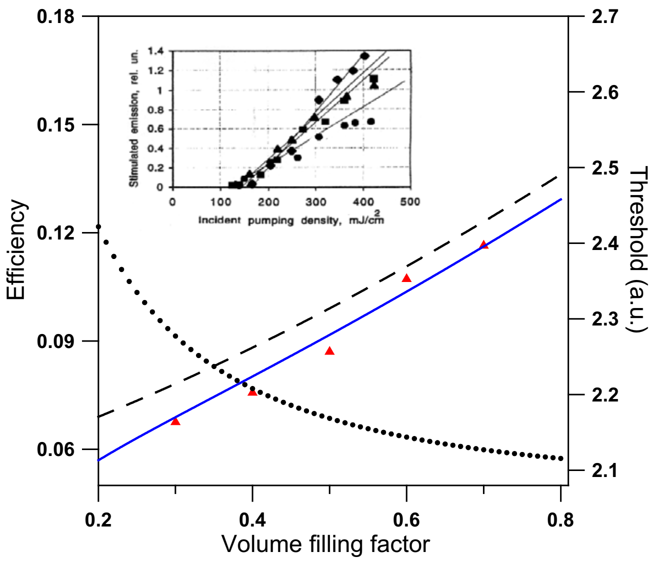

Fig. 2. Efficiency as a function of the volume-filling factor in $Nd_{0.5}La_{0.5}(BO_3)_4$ powders for $L = 1000$ µm. The triangles represent the values obtained numerically for an incident Gaussian pulse [19], the solid line is the efficiency for the stationary regime and the dashed line is the limit of the efficiency for large thicknesses. The dots represent the lasing threshold. The input values for the calculations are: $\bar{\phi} = 4$ µm, $L = 1000$ µm, $\sigma_{em}(1061\ \text{nm}) = 1.5 \times 10^{-19}\ \text{cm}^2$, $\tau_s = 20 \times 10^3$ ns, K_{abs} (532 nm) = 12.5 cm^{-1}, $\beta = 1$. The inset corresponds to the experimental results obtained for the same material ($f = 0.48$ (circles), $f = 0.54$ (triangles), $f = 0.59$ (squares) and $f = 0.63$ (diamonds)). It has been reproduced from figure 2.25 of [22].

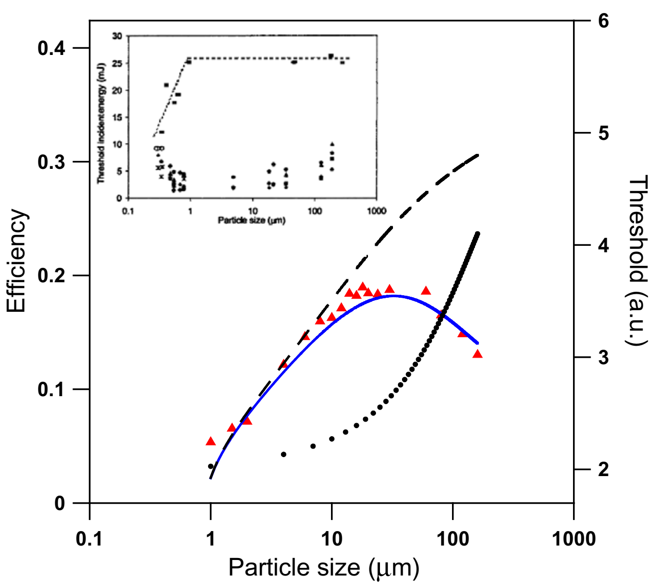

Fig. 3. Efficiency as a function of the particle size in $Nd_3Sc_3(BO_3)_4$ powders. The symbols represent the values obtained numerically for an incident Gaussian pulse [19], the solid line is the efficiency for the stationary regime for $L = 1000$ µm and the dashed line is the limit of the efficiency for large thicknesses. The dots represent the lasing threshold. The input values for the calculations are the same as in Fig. 1 with $L = 1000$ µm. The inset corresponds to the experimental results obtained for the same material. (The dashed line represents the efficiency in log–log scale. Characters below correspond to the threshold at different pumped spot diameters). It has been reproduced from figures 2.28 and 2.30 of [22].

the agreement between corresponding values is excellent in all cases and matches with that experimentally obtained [21,22]. These results suggest that it is possible to analyze the efficiencies of random lasers in different operation regimes by using the result obtained in the continuous-wave (cw) approximation. This fact is not surprising because many properties of pulsed lasers are closely

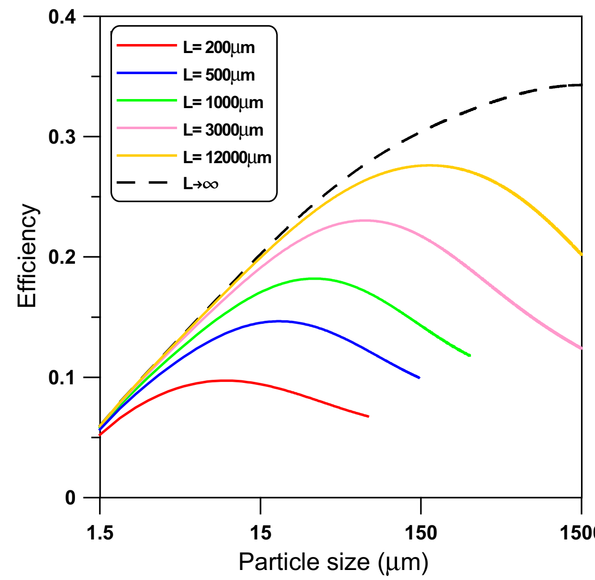

Fig. 4. Efficiency as a function of the particle size in $Nd_3Sc_3(BO_3)_4$ powders in the stationary regime for different thicknesses. The input values for the calculations are the same as in Fig. 1.

related to the corresponding properties of cw lasers. Indeed, values for pulsed and cw thresholds observed in random lasers are linearly proportional to each other [23].

On the other hand, the agreement between the theoretical results and the experimental data shows that the light diffusion model is able to successfully describe the laser-like emission features of laser crystal powders. According to the results obtained, the diffusive absorption length (l_{abs}) and the absorptance (A) of the incident beam play a fundamental role in the main features of the laser action [19]. It must be noted that the diffusive absorption length represents the penetration depth of light in an absorbing and scattering medium. The L-dependence of η (see Fig. 1) is mainly explained by the L-dependence of A. When the thickness is small the incident beam can not be completely absorbed by the sample. As L is increased, the pump beam will be better absorbed by the sample and A will increase too. Finally, when L is large enough the incident beam is completely absorbed and A remains constant. With regard to the dependence of η with the volume-filling factor, Fig. 2 shows a nearly linear dependence of the efficiency with the compacticity of the sample. As f is increased, the absorptance of the incident beam takes place in a more confined region of the sample, increasing A and, in consequence, improving the laser emission efficiency. The dependence of η on the mean particle size, depicted in Fig. 3, shows that there is a maximum of efficiency for a certain value of the particle size. Besides, this maximum shifts to larger sizes as the sample thickness is increased (see Fig. 4). These behaviours can be explained assuming that η is proportional to the quotient A/J_0^{th} where J_0^{th} is the lasing threshold [19]. The value of $\bar{\phi}$ that maximizes η corresponds to a compromise solution that maximizes, as far as possible, the value of A while it minimizes, as much as possible, the lasing threshold. When the thickness is small the shift is nearly constant, but when L is large enough the shift tends to diminish, due to the weak dependence of A on the thickness and on the lasing threshold.

To obtain the dependence of the threshold with the sample parameters ($L, f, \bar{\phi}$) we can use the fact that the quotient between A and the threshold describes all the behaviors of the lasing efficiency [19]. By using the expression of A worked out for the same source of diffuse radiation [18], the next expression for the lasing threshold has been obtained:

$$J_0^{\mathrm{th}}(L,f,\bar{\phi}) \propto \frac{(L+2l_e)\left(-1+Cosh\left(\frac{l_e}{l_{\mathrm{abs}}}\right)Sech\left(\frac{L+2l_e}{2l_{\mathrm{abs}}}\right)Cosh\left(\frac{L-2l^*}{2l_{\mathrm{abs}}}\right)\right)}{\left((L+2l_e)Cosh\left(\frac{l_e}{l_{\mathrm{abs}}}\right)Cosch\left(\frac{L+2l_e}{2l_{\mathrm{abs}}}\right)Sinh\left(\frac{L+l_e-l^*}{l_{\mathrm{abs}}}\right)-L-l_e+l^*\right)} \quad (12)$$

Although this expression does not give the value for the lasing threshold, it can be verified that it describes correctly the behaviour of the threshold when there are variations in the sample thickness, the volume-filling factor, and the mean particle size (see Figs. 1–3). In the literature there are several theoretical expressions for the threshold of random lasers obtained from other physical approaches [23,24], but they do not describe the dependence of the threshold on the three parameters at once (L, f, $\bar{\phi}$). With respect to the efficiency, this is the first time, as far as we know, that a simple expression for η as function of sample parameters has been worked out.

4. Summary

We have theoretically analyzed the dependence of the lasing efficiency of laser crystal powders (LCPs) on the sample thickness (L), on the fraction of volume occupied by the particles (f) and on the mean particle size ($\bar{\phi}$). Analytical expressions of the random laser efficiency have been worked out for stationary regime. The theoretical behaviour of the efficiency agrees with the experimental measurements and with the computational results obtained for an incident Gaussian pulse. The assumption that η is proportional to the ratio of the absorptance and the lasing threshold, has been used to obtain an analytical expression for the lasing threshold. This expression for J_0^{th} also describes correctly the behaviour of the threshold of the laser crystal powders as a function of the sample parameters.

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