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Study of lasing threshold and efficiency in laser crystal powders

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Abstract. We have analyzed theoretically the dependence of lasing threshold and efficiency of laser crystals powders on the sample thickness, the volume fraction occupied by the particles and the mean particle size. The optimal range of these parameters that minimize the threshold and maximize the efficiency has been determined.

PACS. 42.55.Zz Random lasers – 42.25.Dd Wave propagation in random media – 61.43.Gt Powders, porous materials

1 Introduction

Light amplification in locally inhomogeneous dielectric materials is a fascinating topic that gathers fundamental aspects of both classical and quantum optics together. In 1967, Letokhov theoretically predicted the possibility of generating laser-like emission starting from scattering particles with negative absorption, the so-called *random* or *powder laser*, in which the feedback mechanism is provided by disorder-induced light scattering due to the spatial inhomogeneity of the medium [1]. The general spectral signatures of random lasing are an overall narrowing of the emission spectrum, and a threshold behavior. A detailed discussion about the physics of random lasers can be found in a recent publication by Wiersma [2]. In spite of the large variety of materials and morfologies studied, random lasers are still of little practical use and lack of a satisfactory theoretical understanding. The reason could be the high degree of complexity in obtaining the required feedback which can be achieved either by the randomly dispersed scatteres or by the medium surrounding them. Since the discovery in 1986 of a laser-like behavior in compacted laser crystal powders (LCP) they have been extensively studied [3,4]. This type of random laser has no spatial coherence, it is not stable in phase and its photon statistics are strongly different from that of a conventional laser. The potential applications of LCP as compact and mirrorless lasers where 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. This work presents a detailed theoretical study of both magnitudes in Neodymium Borate laser crystal powders. Particularly, the dependences of lasing threshold and efficiency on the sample thickness, the volume fraction occupied by the particles, and the mean particle size have been determined. The description of the photon propagation through the sample has been done in terms of the classical diffusion theory since the mean particle sizes of the studied LCP provide transport mean-free-paths in the scattering medium which are much larger than the wavelength and much smaller than the sample thickness [5,6]. By comparing the theoretical predictions with the experimental results, we conclude that the diffusion model is able to describe successfully the laser-like emission features of LCPs. Our findings can be of much interest in the understanding of results obtained from other solid-state random lasers as well as for the development of new systems operating within the diffusive approximation.

2 Theoretical model

The propagation of the pumped and the emitted photons through the sample is described by two diffusion equations: one for every kind of photons. The volume densities of both kind 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 with dimensions x and y 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:

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

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

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

where $W_p(z,t)$ and $W_e(z,t)$ are the pump and emission photon densities, $N(z,t)$ is the density of active ions in the excited state, $\mathtt{v} = \frac{c}{n_{eff}}$ is the light speed in the medium with n_{eff} the effective refractive index, σ_{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 $l_{abs} = \sqrt{\frac{l_t l_i}{3}}$ is 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 [5]. $D = \frac{\mathtt{v} l_t}{3}$ is the light diffusion coefficient. We have distinguished the diffusion coefficients for pump and emitted radiation, D_p and D_e, respectively. β is the fraction of spontaneous emission contributing to the laser process. The volume fraction occupied by the scatterers, f, has been included into the equations to take into the account the effective fraction of the photons which penetrates inside the particles. The source of diffuse radiation, $p(z,t)$, is a Gaussian pulse incoming the sample in the z direction which is extinguished (scattered and absorbed) along the scattering sample:

$$p(z,t) = \frac{J_o \sqrt{\log 2}}{l_s \Delta \sqrt{\pi}} \exp\left(-\frac{z}{l^*}\right) \times \exp\left[-\left(\frac{t - t_{peak} - \frac{z}{\mathtt{v}}}{\Delta}\right)^2 \log 2\right]. \quad (4)$$

Here, J_o is the number of incident photons per unit area, t_{peak} is the time at which the pump pulse is maximum at the sample surface and Δ is the pulse half width at half maximum. We have considered the following boundary conditions:

$$\begin{aligned} W_p(-l_e^0,t) &= W_p(L + l_e^L, t) = 0 \qquad \forall t \\ W_e(-l_e^0,t) &= W_e(L + l_e^L, t) = 0 \qquad \forall t \\ W_p(z,0) &= W_e(z,0) = N(z,0) = 0 \qquad \forall z \end{aligned} \quad (5)$$

where the extrapolation lengths are given by

$$l_e^0 = \frac{2}{3}\left(\frac{1+r_0}{1-r_0}\right) \times l_t \quad \text{and} \quad l_e^L = \frac{2}{3}\left(\frac{1+r_L}{1-r_L}\right) \times l_t,$$

with r_0 and r_L the internal reflectivities at the front $(z=0)$ and rear $(z=L)$ surface.

3 Results and discussion

The study has been done in ground powders of $Nd_{0.5}\,La_{0.5}\,Al_3\,(BO_3)_4$ and $Nd\,Sc_3\,(BO_3)_4$ laser crystal powders. The mean-free-paths in these materials at the required wavelengths have been calculated by using the Mie theory for spheres in the independent-scatterer approximation with a diameter equal to the averaged mean particle size. We have considered $t_{peak} = 60$ ns and $\Delta = 10$ ns as input values for the source. The set of coupled nonlinear partial differential equations (1)–(3) has been numerically solved by the Crank-Nicholson finite difference method ($\Delta z = 0.05$ μm, $\Delta t = 0.01$ ns) and the calculation has been carried out over a time span of 100 ns. As the emitted photons are usually collected along the backward direction of the incident pump beam, the photon flux per unit area corresponding to the emitted photons will be given by $\vec{F}_e(t) = -D_e \frac{\partial W_e(z,t)}{\partial z}\hat{z}$ evaluated at the front sample surface ($z = 0$). The lasing threshold and efficiency have been calculated by plotting the time integrated emitted flux per unit area $\left(J_e = \int_{-\infty}^{\infty} \left|\vec{F}_e(t)\right| dt\right)$ versus J_o. Above threshold, J_e depends linearly on J_o. The slope of this straight line is the efficiency (η) and the lasing threshold $\left(J_o^{th}\right)$ is given by the intersection of the straight line with the abscissa axis. By proceeding in this way, we have studied the dependence of lasing threshold and efficiency on the sample thickness (L), the volume fraction occupied by the particles (f) and the mean particle size (ϕ).

3.1 Thickness dependence

Figure 1a shows the dependence of J_o^{th} and η on the sample thickness in $Nd\,Sc_3\,(BO_3)_4$ powders. As it can be observed, both magnitudes only significantly change for thicknesses smaller than 300 microns. The obtained behaviours agree with those experimentally obtained with this sample [7] and they can be understood in terms of the relation between the thickness and the diffusive absorption length (l_{abs}). This magnitude represents the penetration depth of light in an absorbing and scattering medium [5,8]. In the sample considered in our study $l_{abs} \cong 50$ μm [7]. For a small thickness ($L \lesssim 6 l_{abs}$) the pump beam can not be completely absorbed by the sample and the diffuse transmittance (T) (see Fig. 1b) as well as J_o^{th} will be high. As L is increased the pump beam will be better absorbed by the sample, decreasing T and J_o^{th}. Finally, for L values big enough the pumping beam is completely absorbed, $T = 0$ and J_o^{th} remains constant. With regard to η, it will be proportional to the absorptance (A) of the pump beam by the sample. The thickness dependence of A (Fig. 1b), and therefore of η, can be understood with similar arguments as those given for T and J_o^{th}.

3.2 Volume filling factor dependence

The effect of the volume filling factor on stimulated emission has been studied in $Nd_{0.5}\,La_{0.5}\,Al_3\,(BO_3)_4$ powders.

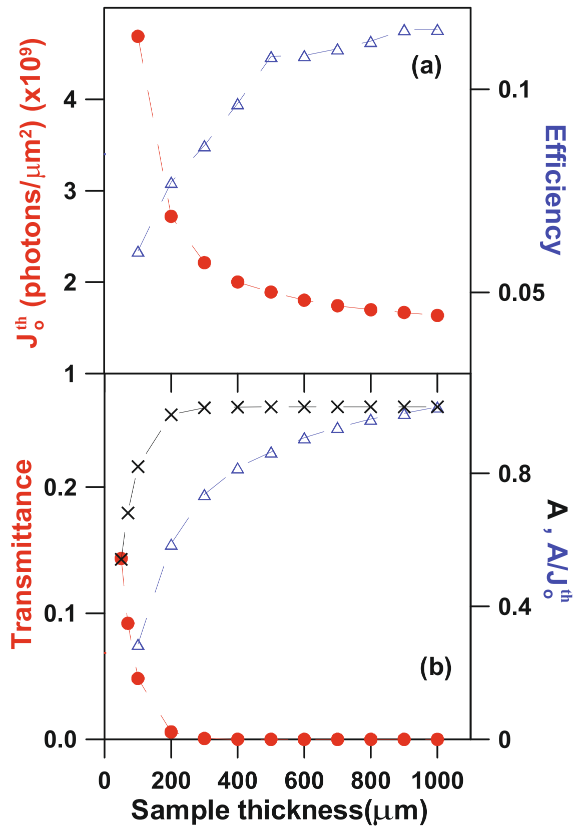

Fig. 1. (Color online) (a) Lasing threshold J_o^{th} (circles) and efficiency (triangles) as a function of the sample thickness in $Nd\,Sc_3\,(BO_3)_4$ powders. (b) Transmittance (circles), absorptance A (crosses) and A/J_o^{th} (triangles) of the pump beam for the same sample. Dashed lines are guides for eye. For clarity, the data corresponding to A and A/J_o^{th} have been normalized to unity. The input values for the calculations are: $\phi = 4\mu m$, $f = 0.55$, $\sigma_{em} = 2 \times 10^{-18}$ cm^2, $\tau_s = 24 \times 10^3$ ns, $K_{abs}(532$ nm$) = 18.2$ cm^{-1}, $n_{eff} = 1.4$, $\beta = 1$.

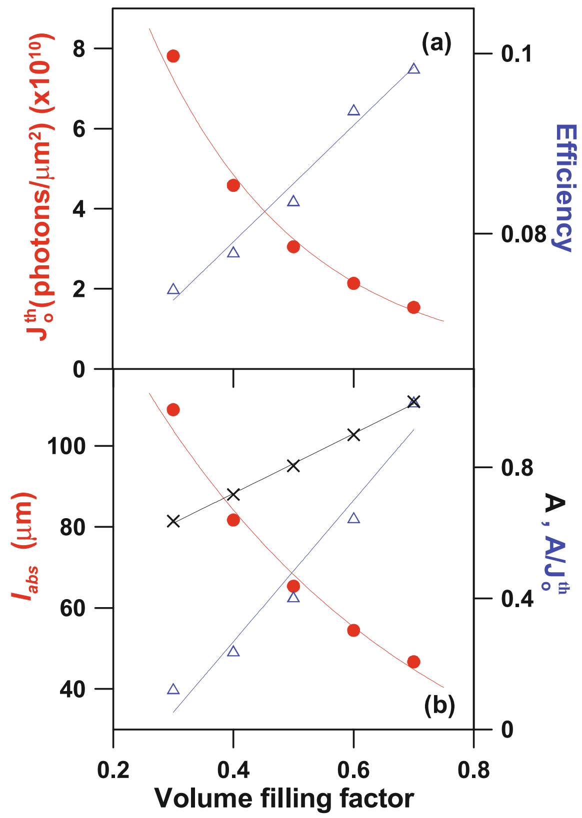

Fig. 2. (Color online) (a) Lasing threshold J_o^{th} (circles) and efficiency (triangles) as a function of the volume filling factor in $Nd_{0.5}\,La_{0.5}\,Al_3\,(BO_3)_4$ powders. (b) Diffusive absorption length (circles), absorptance A (crosses) and A/J_o^{th} (triangles) of the pump beam for the same sample. For clarity, the data corresponding to A and A/J_o^{th} have been normalized to unity. The input values for the calculations are: $\phi = 4\mu$m, $L = 1000$ μm, $\sigma_{em} = 1.5 \times 10^{-19}$ cm^2, $\tau_s = 20 \times 10^3$ ns, $K_{abs}(532$ nm$) = 12.5$ cm^{-1}, $\beta = 1$.

As follows from Figure 2a high values of f favour the laser action, given rise to smaller values of laser threshold and higher efficiencies. The f-dependence of J_o^{th} is mainly determined by l_{abs} and this length varies with f according to a law $1/f$ [8] (see Fig. 2b). Therefore, as f is increased, the absorption of the pump beam takes place in a more confined region of the sample. The inversion of population in this region will be higher and can more easily exceed the critical value to produce the laser emission, provoking a decreasing of J_o^{th}. Besides, for increasing values of f, the effective density of active ions in the sample increases causing an increasing of the absorptance (see Fig. 2b) and, in consequence, an improvement of the laser emission efficiency. The result obtained for the efficiency agrees with the experimentally detected behaviour in the same sample [4].

3.3 Mean particle size dependence

Figure 3a shows the dependence of J_o^{th} and η on the mean particle size in $Nd\,Sc_3\,(BO_3)_4$ powders. The results obtained for J_o^{th} are in good agreement with those experimentally determined with the same sample [9]. Contrary to the dependences previously considered, now there is no value of the variable (ϕ) that optimizes at the same time all laser action features: while J_o^{th} is minimum for $\phi \cong 1$ μm, the maximum of η is reached for $\phi \cong 24$ μm. Again the ϕ-dependence of J_o^{th} will be determined by l_{abs} (see Fig. 3b): when l_{abs} is minimum the region where the pumping beam is absorbed is the smallest and, therefore, J_o^{th} is minimum too. On the other hand, the absorptance increases for increasing values of ϕ (see Fig. 3b). The maximization of the absorptance requires, therefore, a more extended spatial distribution of the pump beam. Consequently, the value of ϕ that maximizes η will correspond to a compromise solution that maximizes, as far as possible, the absorptance and, at the same time minimizes, as much as possible, the lasing threshold. In fact, it can be verified that the ϕ-dependence of the quotient A/J_o^{th} is similar to the one of η, both reaching a maximum for approximately the same value of ϕ (see Fig. 3b). As it can

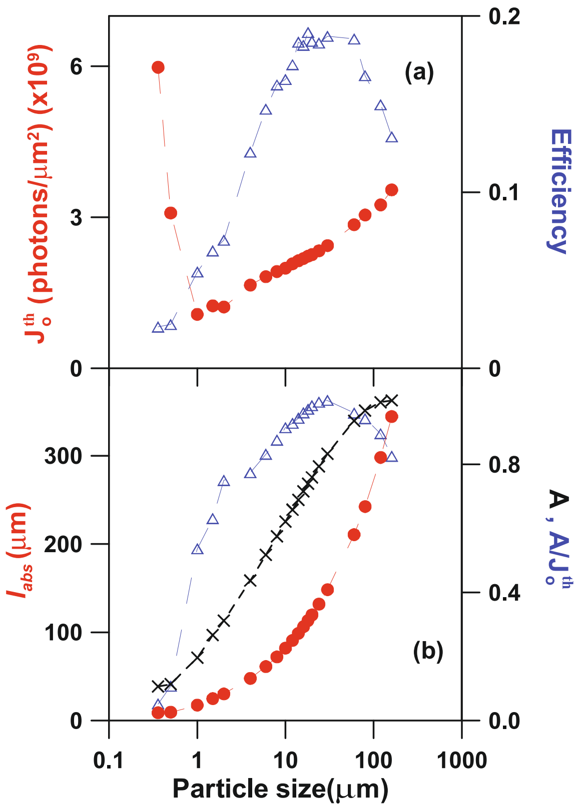

Fig. 3. (Color online) (a) Lasing threshold J_o^{th} (circles) and efficiency (triangles) as a function of the particle size in $\mathrm{Nd\,Sc_3\,(BO_3)_4}$ powders. (b) Diffusive absorption length (circles), absorptance A (crosses) and A/J_o^{th} (triangles) for the same sample. Dashed lines are guides for eye. For clarity, the data corresponding to A and A/J_o^{th} have been normalized to unity. The input values for the calculations are the same as in Figure 1 with $L = 1000$ μm.

be seen in Figures 1b and 2b, this quotient also determines the dependence of the efficiency on the thickness and the volume filling factor.

4 Conclusions

We have theoretically analyzed the dependence of lasing threshold $\left(J_o^{th}\right)$ and the efficiency (η) of LCP on the sample thickness (L), the volume fraction occupied by the particles (f) and the mean particle size (ϕ). The results obtained are in agreement with the experimental data showing that the light diffusion model is able to successfully describe the laser-like emission features of LCP. According to the obtained theoretical results the diffusive absorption length (l_{abs}) an the absorptance (A) of the pump beam play a fundamental role in the main features of the laser action: J_o^{th} will be minimum when the region where the pumping beam is absorbed is the smallest (l_{abs} minimum), while η will be maximum when the quotient $\left(A/J_o^{th}\right)$ is maximum. In the case of the L- or f-dependence of J_o^{th} and η both extremal conditions can be satisfied at the same time, and the optimum values of the parameters are: $L \gtrsim 6 - 8l_{abs}$ and f as high as possible. Nevertheless, there is no value of the particle size that fulfils both conditions at the same time. The optimum particle size will be in the interval $\left[\phi\left(J_o^{th} minimum\right), \phi\left(\eta\ maximum\right)\right]$ and its particular value will depend on the characteristics required for each particular random laser considered. The obtained results are general and they establish the restrictions on the sample parameters that have to be taken into account in the design of optimized LCP random lasers operating within the diffusion approximation.

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