Diode-pumped laser characteristics and cross-section determination of high quality Nd:YLF and Nd:GLF grown crystals

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Abstract

The growth and laser performances of Nd:$LiYF_4$ and Nd:$LiGdF_4$, pumped by diode-laser (4W), are described. The laser emission cross-section of the Nd:$LiGdF_4$ was determined to be in the order of 8×10^{-20} cm^2.

Key Words

Laser materials, Infrared and far-infrared lasers, Rare earth and transition metal solid state lasers.

Introduction

To obtain lasers with high c.w. output power, the active medium must have narrow emission line, intense absorption and emission bands and long upper laser level lifetime in order to store energy. The majority of high power solid-state lasers use, as the active medium, neodymium ions incorporated in solid hosts. The rare-earth elements naturally show these favorable characteristics, since they have intra-configurational transitions, with long decay times. The electrons involved in the laser cycle are relatively shielded against the external field, therefore showing low sensitivity to the particular host.

The high optical quality $LiYF_4$ crystal (YLF) is a well-known host material for several rare-earth active-center ions, including neodymium. [1] This material has good thermal and mechanical characteristics, and low photo-elastic coefficients. Moreover, it is birefringent, therefore minimizing the loss of the optical polarization due to induced stress or nonlinearities. The concentration of Nd^{3+} ions, that substitute the Y^{3+} ions, is typically below 1.5% to attain high laser efficiency. Higher concentrations cause an unbearable decrease in the laser lifetime, due to the interaction of pairs of Nd^{3+}, and poorer crystal quality, due to the discrepancy between the ionic radii of the Y^{3+}, 1.015Å, and Nd^{3+}, 1.12Å. An alternative is to use a slightly different host compound: $LiGdF_4$. As the ionic radius of Gd^{3+} is 1.06Å, higher concentrations of Nd^{3+} can be added to the host (~5%), without decreasing the crystal quality. The $LiGdF_4$ crystal (GLF) is isostructural with $LiYF_4$, with almost identical thermal, mechanical and optical characteristics, presenting similar Nd^{3+} absorption and emission spectra. [2] For both crystals, the most intense absorption lines are 792 nm and 797 nm, with cross-sections around $2.5 \times 10^{-20} cm^{-1}$.

The laser action of the Nd:GLF was reported in the past few years. [3, 4] For this crystal, and for Nd:YLF as well, the more efficient laser transition is $^4F_{3/2} \rightarrow {}^4I_{11/2}$, in the π polarization (electrical field parallel to the c-axis), at $\lambda \cong 1.047$ nm. The stimulated emission cross-section, σ_S, is not precisely known for Nd:GLF. The precise measurement of σ_S is a nontrivial work, and it is usually found appreciable differences among the values listed in the literature, for instance, for Nd:YLF. The best methods to perform this task are those realized during laser action, thus leading to an effective value of σ_S, regarding all transitions that contribute to the laser amplification and effects that can modify the spectral parameters (due to the thermal load, for instance).

Recently, high-power diode-lasers, emitting in a wavelength range around 800 nm, have been used to longitudinally pump Nd-doped crystals, with the advantages of lower thermal load and higher optical efficiency, stability and compactness. However, these high-power semiconductor lasers have very asymmetric emission areas, broad in the junction direction and narrow transversally. The beam quality factor in the junction

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direction, M_x^2, is typically very high (≥100), opposed to $M_y^2 \approx 1$, what makes difficult the design, development and modeling of these systems.

Crystal Growth

The yttrium fluoride and the rare-earth fluorides were synthesized from pure oxide powders (Aldrich and/or Johnson Matthey with purity of 99.99%) using the process of high temperature hydrofluorination, in argon gas (purity of 99.995%) and gaseous hydrogen fluoride (HF) (Matheson Products with purity of 99.99%) atmosphere. LiF powder (Aldrich with purity of 99.99%) was zone-refined before it was added to the YF_3 or GdF_3. The YLF was synthesized starting from a mixture of 49.5% YF_3 and 50.5% LiF, which was the most efficient composition to the subsequent purification by the zone-refining process. In the YLF zone refining, a single pass was performed along the boat at a transverse rate of 7 mm h^{-1}, under a stream of purified argon and HF. In general, the stoichiometric YLF phase occupied 90% of the produced ingot. The LiF powder was subjected to four zone-refining passes using a transverse rate of 10 mm h^{-1}. The apparatus utilized is described in reference [5]. The mixture LiF - GdF_3 was synthesized with a composition of 32% GdF_3 and 68% LiF. Since it shows an accentuated incongruent melting behavior, it was not zone refined.

Single crystals were grown by the Czochralski technique, under a purified argon atmosphere, using a conventional stainless steel furnace with graphite resistance heating. The zone-refined YLF with addition of 3 mol % excess of LiF, or the synthesized LiF - GdF_3, and the neodymium fluoride, were melted in a platinum crucible after a heat treatment under vacuum. The crystal pulling rate was 1 mm h^{-1} for [100] oriented boules, and rotation rates were around 30 rpm. Finally, the crystals suffered a thermal treatment, under argon atmosphere, to eliminate internal stress.

Characterization and active element preparation

Due to the high incongruent melting behavior of the GLF, good quality crystals were obtained only by maintaining the diameter of the grown boule shorter than 10 mm and the length around 10 cm. Efforts to enhance the diameter have resulted in crystals with bubble lines disposed at 45° to the pulling axis. For the Nd:YLF, the described procedure has allowed us to obtain boules with diameter of $\phi \approx 30$mm and length $\ell \approx 100$mm, with high homogeneity and suitable for the extraction of laser rods. Samples from various positions of the crystals were chemically analyzed, for the determination of the neodymium concentration and the occurrence of eventual impurities. The Nd concentrations were 0.6(1) mol% (0.85×10^{20} cm^{-3}) for the Nd:YLF crystal and 2.0(1) mol% (2.85×10^{20} cm^{-3}) for the Nd:GLF crystal. The regions of minimum optical scattering were identified by passing through each crystal a laser beam and evaluating the scattered light. The polarization preservation of a transmitted optical beam was also verified, what allowed the identification of stressed or multi-domain regions. The samples used in this work were extracted along the growth direction.

The optimum active medium length, for a longitudinally pumped laser at $\lambda_P = 797$ nm, and considering diffraction limited Gaussian beams, [6] is around 1.5 cm for a Nd:YLF crystal with Nd concentration of 0.6 mol%, and around 4 mm for the Nd:GLF crystal with 2 mol%. The dimensions of the Nd:YLF sample were 9 mm^2 x 13 mm and were 50 mm^2 x 4 mm for the Nd:GLF. The optical faces were prepared at Brewster angle, in the π polarization, and polished to a flatness of $\lambda/4$. The transmissions of both samples at $\lambda = 1.06\,\mu$m were 0.995(5).

Diode-pumped laser performance

The diode laser used to pump both crystals was a *SDL-2382-P1* (*SDL, Inc.*, CA, USA). This is a broad-area (1 µm x 500 µm), 4 W c.w., GaAlAs laser, tunable in the range 790-800 nm by temperature control. The diode laser beam has a high asymmetry of size and quality, with $M_y^2 \approx 1$ and $M_x^2 \approx 100$. It was collimated by a diffraction-limited, N.A.=0.5, f=8 mm objective, and transformed by a 3x anamorphic prism pair (both from *Melles Griot*), that expands the beam in the *x* direction. The simplest condition to deliver this pumping beam to the active medium was found to be just by focusing it, with a 5 cm focal length lens, into the laser crystal. Close to the focus, and for a longitudinal range of 2 mm, the beam had a rectangular profile, with transverse dimensions of 60 µm x 300 µm.

The general setup of the laser cavity, used for both the Nd:YLF and the Nd:GLF laser, is shown in Fig. 1. The resonator has 3 mirrors, with plane end mirrors M_1 and M_2, and a concave folding mirror M_3. The tilting angle of M_3 was chosen in order to compensate for the astigmatism of the intracavity elements, placed at Brewster angle. The laser crystal is positioned close to the mirror M_1, that has a high transmission for $\lambda_P = 797$ nm, and is highly reflector for the laser emission wavelength ($\lambda = 1047$ nm).

The optimum TEM_{00} beamwaist at the active medium, to the maximum laser output power, was experimentally found to be around 230 µm, for both crystals. This can be obtained using $R_3 = 1$ m, $L_1 = 70$ cm and $L_2 = 130$ cm. Both samples absorb almost 100% of the pump power, without laser action. Thus, pumping saturation effects are not present.

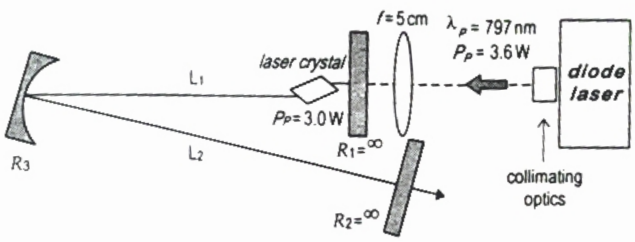

Figure 1. General scheme for the optical cavity used in the laser experiments.

The optimum transmissions of the output mirror, M_2, were 25% for the Nd:YLF and 8% for the Nd:GLF. The output versus input power, for both crystals, is shown in figure 2. The Nd:YLF laser shows a higher slope efficiency and lower pump power threshold.

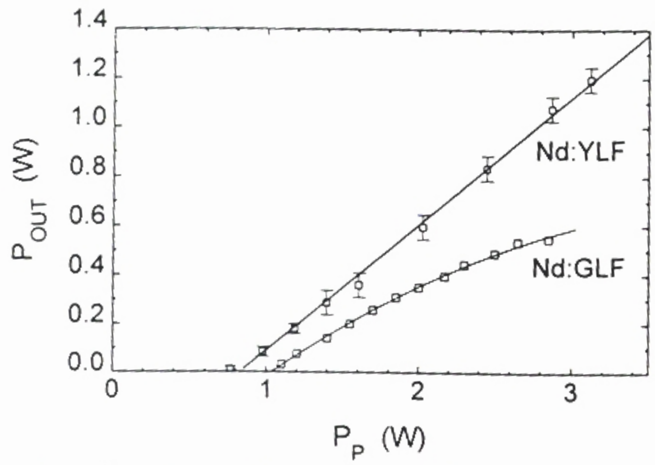

Figure 2. Output versus pump power. The output mirror transmission was 25% for Nd:YLF and 8% for Nd:GLF.

Fundamental laser parameters

For a better understanding of the material differences, measurements of the output power as a function of the output reflectivity were performed, as shown in figure 3. By fitting the following expression from the laser oscillator model: [7]

$$P_{OUT} = AI_S \frac{(1-R)}{2}\left[\frac{\Gamma^0}{L-\ln(R)} - 1\right],$$

it was determined the equivalent non-saturated gain, Γ^o, the cavity losses, L, and the saturation power, equal to the beam area at the active medium, A, times the material saturation intensity, I_S. It was found, for the Nd:YLF: $\mathbf{\Gamma^o = 1.2(4)}$, $\mathbf{L = 0.2(1)}$, and $\mathbf{A.I_S/2 = 3.5(4)\,W}$; for the Nd:GLF: $\mathbf{\Gamma^o = 0.4(1)}$, $\mathbf{L = 0.1(1)}$, and $\mathbf{A.I_S/2 = 8(2)\,W}$. The higher losses in the Nd:YLF system were due to the fact that the resonator had other optical elements inserted (a Brewster-angled glass and an acousto-optical modulator). Assuming an average mode area at the active medium: $A \cong 1.7\times10^{-3}\,\text{cm}^2$, the Nd:YLF saturation intensity can be estimated as: $\mathbf{I_S \approx 4\,kW/cm^2}$. A more precise determination of this parameter, realized in a previous work, [8] was $I_S \approx 1.2(1)\,\text{kW/cm}^2$. The Nd:GLF saturation intensity could be estimated from the measurements as: $\mathbf{I_S \approx 9\,kW/cm^2}$.

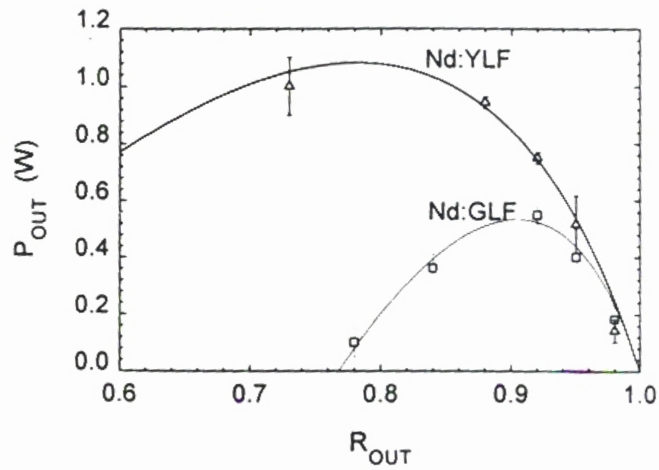

Figure 3. Output power as a function of the output reflectivity for both crystals.

The theoretical model used to calculate I_S [6] imposes constant area beams, what is not obeyed here. The pumping beam is nearly constant only in 2 mm and the Nd:YLF gain medium is 1 cm long (the 0.6 mol% sample absorption length). For the 2 mol% Nd:GLF, the absorption length is around 3 mm. However, even for this case, the obtained results must be viewed only as estimates. Besides, the local heating of the laser crystal, caused by the 3 W laser beam, leads to a broadened emission line that corresponds to a higher I_S.

Because of the absence of any previous determination of I_S for the Nd:GLF, it is worth to realize a second analysis. Considering the obtained equivalent non-saturated gain, $\Gamma^o = 0.4(1)$, and the following expression from the model of longitudinal pumping: [6]

$$\Gamma^o = P_{AB}\cdot\frac{4.\eta_P.(\lambda_P/\lambda).\beta}{\pi.(w_L^2 + w_P^2).I_S}$$

where $P_{AB} \cong 3.0$ W is the absorbed power, $\beta \cong 0.5$ is the Boltzmann factor for the upper laser level population. $\eta_P \cong 1$ is the pumping quantum efficiency, $(w_L)^2 \cong (230\,\mu\text{m})^2$ and $(w_P)^2 \cong (60\,\mu\text{m} \times 300\,\mu\text{m})$, it is obtained: $\mathbf{I_S = 6(2)\,kW/cm^2}$.

Discussion

During the experiments, intense visible fluorescences (around 425, 525, 590 and 655 nm) were observed along the pumping path. This could be characterized as up-conversion processes that occur in the Nd^{3+} ions in both hosts. These effects have been studied for Nd:YLF, [9] and it is known that they degrade the performance of the diode-pumped laser. The degradation increases with the population density of the upper laser state, thus with the pump beam intensity, the total Nd^{3+} concentration and the absorption cross section.

A fundamental difference between the spectroscopic parameters of Nd:YLF and Nd:GLF is responsible for the observed lower efficiency of the second. If the Nd:GLF upper laser level lifetime, for the sample with 2 mol% of Nd, can be assumed as: [4] $\tau \cong 430\,\mu s$, the Nd:GLF stimulated emission cross-section in the π polarization can be estimated as $\sigma_S \approx \mathbf{6x10^{-20}\ cm^2}$, for the first determination of I_S, and $\sigma_S \approx \mathbf{8(3)x10^{-20}\ cm^2}$, for the second. It must be stressed that this value of σ_S was obtained in an *in situ* measurement, therefore affected by the system characteristics, mainly by the pump beam intensity, geometry, and the absence of any cooling of the active medium.

Acknowledgments

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Note added in proof: further measurements using a beam-shaped diode-laser pump beam have led to an estimate of the emission cross-section as $\sigma_S \approx \mathbf{1.5x10^{-19}\ cm^2}$, at the same pump power.

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