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HIGH POWER SOLID STATE LASERS IN THE NEAR INFRARED

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ABSTRACT

Since the early discovery of the laser, Nd:YAG crystals became one of the most used laser medium, in spite of the difficulty of processing the host material and the associated thermal problems during laser operation. As an alternative for the host material we have been developing lasers based on an alphabetic material, $LiYF_4$, where the Nd is one of the possible dopants. Along with the crystal growing developments we have been developing the associated laser technology, including rod preparation, pumping cavities, power supplies, cooling systems, resonator configurations and controlling of the temporal behavior of the laser regime. So far we have developed three prototypes in the pulsed regime and three in the C.W..

1. INTRODUCTION

Since the end of the sixties, a materials science program started at IPEN which included crystals and its defects. During the seventies, a major effort was carried out in the comprehension of the radiation induced defects in either pure or doped crystals. The main concern was with alkali halides and some solution grown crystals. Within this scope, the optical spectroscopic properties of these materials were subject of fundamental investigations, concomitantly with the advent of color centers as useful laser sources[1]. The combined effect of having the crystal growing facilities and the spectroscopic tools led to the first color center laser at our laboratories[2]. In spite of the usefulness of the color center lasers, in most cases they have an intrinsic photothermal instability that limits their practical uses. Ion doped crystals, otherwise, are usually very stable, being mostly limited by the crystal growing processes. Among the most succesful laser materials there is the Nd:YAG ($Y_3Al_5O_{12}$) garnet that nowadays is the most widespread powerful solid state laser.

The main difficulty with the YAG crystal is its very high melting point that imposes thermal gradients in the growing systems affecting the homogeneity of the grown boules. So we looked for a substitute that would present the same properties as a laser material associated to easier crystal growing conditions. An evaluation of the field led to the $LiYF_4$ (YLF) matrix, whose main characteristics are presented bellow. Once the host material was chosen, the main efforts of the laser development group at IPEN have been towards the achievement of Nd:YLF lasers, since Nd was the most promising dopant ion.

The choice of the scheelite-structured YLF matrix[3,4] as the host material for the dopant ions was due to several reasons:

1) The substitutional Nd ion is located in the Yttrium site, with the same valence. This site has no inversion symmetry (group S_4), therefore the proper admixture of even parity terms in the f manifold wave function allows for the optical transition strength needed for the laser transitions. These transitions occurs within the internal f manifold that is shielded by s and p eletrons. Due to the small crystal field strength the laser transition cross section is smaller in YLF than in YAG, but the radiative decay time is conversely longer allowing for a higher energy storage;

2) The YLF is an alphabetic material, concerning the introduction of rare earth ions in the Yttrium site, allowing for higher levels of dopant concentration (in some cases even 100%) and rare earth simultaneous doping;

3) YLF has a melting point much lower than the YAG, therefore, the need for special furnaces and very special crucible materials is slightly less demanding. Nevertheless, platinum crucibles are still needed, but they can be reutilized after the material processing. One drawback of this crystal is the need of hydrofluoric acid atmosphere during the material processing;

4) The thermal conductivity is two times smaller in YLF than in YAG crystal but the index of refraction is less dependent on the crystal temperature. Therefore the thermal lensing effect is less serious than in the Nd:YAG crystal. Besides, its natural birefringence overwhelms the thermally induced one, what makes it a better material for

high power operation;

5) The laser transitions are twofold, one at 1.047 um (π polarized) and another at 1.053 um (σ polarized), and they can be chosen by a proper alignment of the crystal axis with respect to the optical resonator axis. The latter transition matches perfectly with the emission line of the Nd:Glass phosphate amplificator in the inertial confinement fusion experiments;

6) The homogeneous broadening of the laser transition in YLF is greater than in YAG, therefore it supports much shorter pulses in the mode-locked regime.

2. LASER CRYSTALS

2.1. Crystal Growth[5]

To prepare the Nd:YLF crystals, we first start from ultra pure oxide rare earth powders, Nd_2O_3 and Y_2O_3. These materials are then thermally treated in a dynamic hydrofluoric acid atmosphere in order to obtain the respective NdF, and YF, composites. These fluorides are then mixed with LiF non-stoichiometrically and excess of NdF, is added to achieve the desired final concentration. These materials are then synthesized to obtain the final compound. The next step is to zone refine the compound at the crystal melting point temperature, also under HF atmosphere. The zone refining region is in the order of two centimeters and this hot zone passes through the compound with a constant speed. Impurities and the excess of LiF are left at the end of the ingot. The final ingot is a transparent material, but it is not a single crystal. This procedure guarantees the purity and the stoichiometric composition of the final compound since it melts incongruently. The next step is the single crystal growth by the Czochralski method under Argon atmosphere with a very slow growing rate. The whole process takes about two months. The crystal is then submitted to a strain relief thermal treatment that takes another few days.

2.2. Crystal Characterization

The crystal samples can be characterized in two stages during the preparation procedure: first, just after the zone refining procedure where the ingot composition is analyzed by spectrographic methods with ppm resolution and second, the single crystal is analyzed also in terms of the Nd concentration of the single crystal as well as its spectroscopic characteristics, such as optical absorption, emission, decay time, crystalographic orientation and the presence of domains. Figure 1 shows the energy level

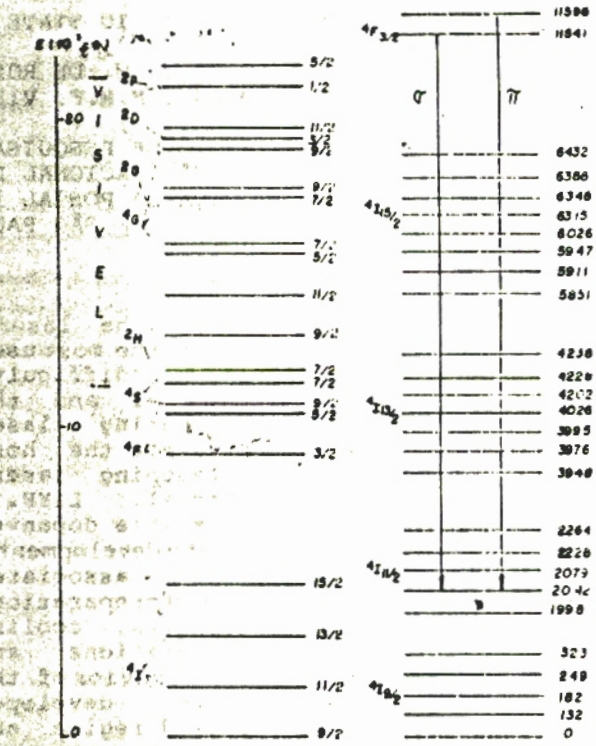

FIGURE 1. Nd^{3+} energy levels in LiYF₄ crystal. The emissions at 1.047 μm (π) and 1.053 μm (σ) are shown.

diagrams of Nd in the $LiYF_4$ crystal. In our lasers, the crystal is oriented with the c axis perpendicular to the rod axis to favor the π polarization, that has the higher emission cross section. These measurements provide a first roughidea of the existence of any structural stress that would lead to some light depolarization which is detrimental to laser action.

In the case of the Nd:YLF crystal its optical properties are well known, such that a comparison with the literature is enough to evaluate the quality of the crystal. Along with the Nd laser development, we are also developing Er:YLF crystals. In this case, the system is much more complex due to the possibility of attaining high Erbium concentrations and therefore there is the possibility of energy exchange between neighbour ions, that affects the total decay time, reducing the laser efficiency. Besides, due to the transitions of low energy and to the possibility of energy migration along the ions, the presence of undesired impurities can lead to a quenching effect of the green luminescence[6]. Table 1 shows the measurements of the luminescence branching ratios in the case of the Er:YLF. We are characterizing this system with conventional spectroscopy techniques and also using photoacoustic spectroscopy which allows for the determination

[Er^{3+}]	λ(μm) (Δλ)	Transition	η_T 77 K	η_T 300 K	B_i(%) 77 K	B_i(%) 300 K
1%	0.55 (0.04)	$^4S_{3/2} \longrightarrow {}^4I_{15/2}$	1	0.84	48.6	21.0
	0.67 (0.03)	$^4F_{9/2} \longrightarrow {}^4I_{15/2}$			27.5	8.3
	0.70 (0.03)	$^2H_{9/2} \longrightarrow {}^4I_{13/2}$			4.1	1.3
	0.85 (0.03)	$^4S_{3/2} \longrightarrow {}^4I_{13/2}$			10.5	3.8
	>1.10	mid-IR			9.3	66.6
40%	0.55 (0.03)	$^4S_{3/2} \longrightarrow {}^4I_{15/2}$	0.041	0.006	38.1	1.4
	0.67 (0.03)	$^4F_{9/2} \longrightarrow {}^4I_{15/2}$			13.7	1.6
	0.70 (0.03)	$^2H_{9/2} \longrightarrow {}^4I_{13/2}$			3.5	1.4
	0.85 (0.03)	$^4S_{3/2} \longrightarrow {}^4I_{13/2}$			6.9	0.2
	>1.10	mid-IR			37.8	95.4

TABLE 1. Branching ratio of Er^{3+} in YLF crystal. λ_e is the most intense emission wavelength of the multiplet measured at 77 K, η_T is the total luminescence efficiency, B_i is the branching ratio, and $\Delta\lambda$ is the is the multiplet width.

of the absolute luminescence quantum efficiency. Table 2 shows the typical results obtained for the Nd:YLF crystals using photoacoustic spectroscopy.

Before going into the crystal rod preparation it is also possible to verify the laser action using a collinear pumping scheme where the crystal sample is very small, reducing the need for careful crystal preparation. This scheme was tested with the first Nd:YLF crystals grown at our lab, and once obtained the laser operation parameters we were able to determine the amount of losses introduced by the crystal[7].

λ (μm)	η (1.047 μm)	η (1.054 μm)
0.517	0.12 ± 0.02	0.18 ± 0.02
0.577	0.16 ± 0.02	0.20 ± 0.02
0.742	0.17 ± 0.02	0.13 ± 0.02
0.792	0.29 ± 0.02	0.27 ± 0.02

TABLE 2. Luminescence quantum efficiency of Nd^{3+}:YLF for the absorption bands centered at λ_p (μm) with emission at 1.047 and 1.053 μm.

2.3. Rod Preparation

Single crystals are grown typically with the boule oriented with the c axis perpendicular to the growing direction, with typical useful dimensions of 3 cm of diameter and 8.5 cm long. Rods are then extracted in order to maximize the useful area. Rod diameters can range from 4 mm to 6 mm and they have typically 75 mm of length. The ends are polished in a special jig in order to obtain a surface flatness of λ/10, in the near infrared, with a parallelism of 30" of arc. They are anti reflection coated with a four layer design. The barrel finish is the usual grit 400[8].

2.4. Pumping Cavities

The coupling of the pumping lamp with the laser rod is done by a reflecting enclosure, whose shape depends on the particular application. For pulsed lasers with a low average power, where there is no need for forced cooling, a simple closed coupled cavity consisting of a highly reflecting cylindrical tube is enough for achieving high efficiencies. For C.W. or high power pulsed lasers, where water cooling is necessary, the usual configuration is the elliptical one, with the rod at one of the foci and the pump laser at the other. We have used both configurations, coated with gold, silver or diffuse material.

2.5. Resonator Configurations

For the pulsed mode operation, in order to extract the maximum output power in all possible oscillating modes, the typical mirror configuration is either with flat mirrors or plane-concave configuration, in order to fill the laser rod with the spatial mode. In this case, the rod acts as a dynamic focusing element limiting the useful laser area. In the case of the Nd:YLF this problem is minimized. For the C.W. mode the TEM_{oo} mode is desired so the flat convex resonator is the used one.

2.6. Power Supplies and Cooling Systems

The power supplies for pulsed mode operation were developed to cover the range of 50 to 250 J and pulse durations can range from 100 μs to 310 μs.

For the continuous wave, the electrical output power ranges from 1000 W up to 4000 W. The electrical system has safety interlocks to avoid overheating of the system and also control the deionized water quality. A special triggering circuit is needed for the high pressure arc lamp in the C.W. operation. For high power lasers, the cooling is

firstly done by a primary closed deionized water circuit and then by an external secondary water circuit.

3. LASER PROTOTYPES

3.1. Pulsed Models

Due to the simplicity of construction, the first lasers developed were the pulsed ones, and 3 models were already constructed. One is a hand held, rugged model, with a parallel flat mirror configuration, a compact power supply, low repetition rate (0.3 Hz), that delivers 70 mJ in a 0.3 ms pulse duration in multimode operation. The pump lamp is a low pressure Xe flash lamp.

The second pulsed laser developed uses the same pump lamp but the electrical power source can deliver up to 50 J. The resonator configuration is also flat and parallel with a diffuse pumping cavity. The multimode free running energy is 500 mJ.

Using a saturable absorber consisting of a LiF:F_2^- crystal we were able to optimize the Q-switching operation of this laser, obtaining pulses as short as 5 ns, what is comparable to what is obtained using active Q-switchers. Figure 2 shows the pulse output obtained in this case. These centers are long lived at room temperature even under working peak powers of tens of megawatts. The mechanism of formation, its spectroscopic properties and stability were subject of investigation at our lab. It was found, in our case, that the best LiF crystals are the ultra pure ones, that can be obtained at our crystal growth facilities. By increasing the length of the resonator and decreasing the output coupling, mode locking regime with 100% modulation, was observed in this system. An estimate of the pulse duration with a background free second harmonic generation autocorrelator showed pulses with, at most, 80 ps of duration. With this high peak powers we then used a home grown KDP to generate second harmonic.

The third model is an industrial phototype made of a Nd:Glass rod, pumped by two lamps in a double ellipse configuration, with an electrical input power of 250 J in a plane concave configuration, using a long radius mirror. The output energy of this laser is 6 J, with a pulse duration of 100 µs, tailored to the dynamic balancing of stainless steel rotors.

3.2. CW Lasers

For the C.W. operation mode, we have developed three laser models. The first one is pumped by a tungsten halogen lamp in an elliptical pumping cavity, gold plated, cooled by forced air. The laser rod is cooled by water flow. The resonator configuration is plano concave, multimode, and the useful output power achieved was 400 mW. The second model is an upgrading of the proceding one, using two lamps for pumping. In this case, the maximum output power is 1 W.

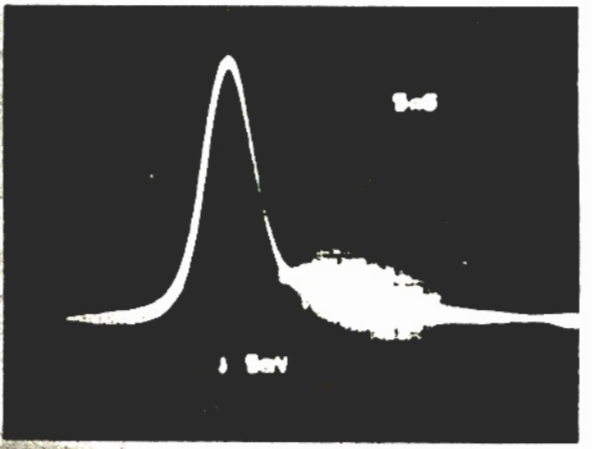

FIGURE 2. Pulse of the Q-Switched Nd:YLF laser using LiF:F_2^- crystals as the passive Q-switcher (⟷ = 5ns).

The third model uses a high pressure Kr arc lamp and in this case the rod and the flash lamp are water cooled in a flooded pumping cavity. In this case, the resonator configuration is the standard plane concave. The maximum electrical input power is 4 KW and in this condition we obtained 38 W in multimode operation. It was obtained 6 W in the TEM_{oo} mode, in a concave-convex resonator, where the thermal lens effect can be easily compensated.

4. CONCLUSIONS

It was presented the evolution of the Laser Development Group at IPEN. It started with the alkali halides single crystals growth and the conventional color center spectroscopy, that associated led to the development of a color center laser. This capability and the difficulties of the color center laser led us to look for another kind of laser that could be developed at our laboratories. The YLF matrix was choosen and its growth is well established at our laboratories, as well as the spectroscopies for its characterization. At the same time it was developed the solid state laser engineering that led to the obtention of the six prototypes described in the text.

5. ACKNOWLEDGMENTS

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