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Nd:YLF Lasers

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ABSTRACT

Since the discovery of the laser, Nd:YAG crystals became one of the most used laser medium. 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..

LASER PROGRAM AT IPEN

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 in our laboratories [2]. At that time we realized that Nd doped crystals would be an interesting field of either basic research and technology. Since then, the main efforts of the laser development group at IPEN has been towards the achievement of Nd:YLF lasers.

LASER CRYSTALS [3,4]

The choice of the scheelite-structured $LiYF_4$ (YLF) matrix as the host material for Nd^{3+} ions is due to several reasons:

1) The substitutional Nd ion is located at 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. 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 crystals 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 mm (π polarized) and another at 1.053 mm (σ polarized), and 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.

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_3 and YF_3 composites. These fluorides are then mixed with LiF non-stoichiometrically and excess of NdF_3 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 of the order of two centimeters and this hot zone passes through the compound with a constant speed. Impurities and the excesses of YF_3 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 Czochraslki 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.

CRYSTAL CHARACTERIZATION

The crystal samples can be characterized in two stages of the preparation procedure: first, just after the zone refining procedure where the ingot composition is analyzed by spectrographyc 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, crystalographyc orientation and the presence of domains. Figure 1 shows the energy level 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 shows the higher emission cross section. These measurements provide a first rough idea of the existence of 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 neighbor ions , that affect 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 also characterizing this system either with conventional spectroscopy techniques and also with the photoacoustic methods which allow for the determination of the absolute luminescence quantum efficiency. Table 2 shows the typical results obtained for the Nd:YLF crystals .

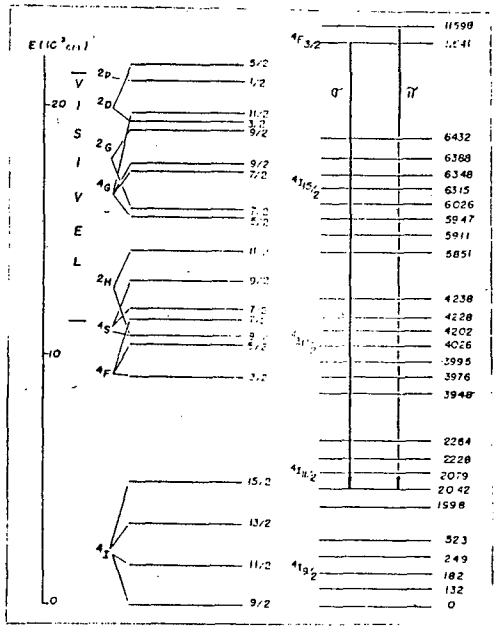

Figure 1: Nd^{3+} energy levels in $LiYF_4$ crystal. The emissions at 1.047 $\mu m(\pi)$ and 1.053 $\mu m(\sigma)$ are shown.

Table 2: Luminescence quantum efficiency for the absorption bands centered at λ (μm) with emission at 1.047 and a 1.053 μm.

λ (μ m)	η (1.047 μ m)	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

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 in our lab, and with the laser operation parameters we were

able to determine the amount of losses introduced by the crystal [7].

Table 1: Branching ratio of Er3+ in YLF crystal; λ is the most intense emission wavelength of the multiplet measured at 77 K, η_T is the total luminescence efficiency, B_1 is branching ratio, and the $\Delta\lambda$ is the multiplet width.

$[Er^{3+}]$	$\lambda(\mu m)$ ($\Delta\lambda$)	Transition	η_T 77 K	η_T 300 K	B_1 (%) 77 K	B_1 (%) 300 K
1%	0.55 (0.04)	$^4S_{3/2} \longrightarrow {}^4I_{15/2}$	1	0.54	48.6	21.0
	0.67 (0.03)	$^4F_{9/2} \longrightarrow {}^4I_{15/2}$			27.5	8.3
	0.70 (0.03)	$^2H_{9/2} \longrightarrow {}^4I_{11/2}$			4.1	1.3
	0.85 (0.03)	$^4S_{3/2} \longrightarrow {}^4I_{13/2}$			10.5	2.8
	>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)	$^2H_{9/2} \longrightarrow {}^4I_{11/2}$			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

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 then polished in a special jig in order to obtain a surface flatness of 1/10, 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].

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.

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 convex-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_{00} mode is desired so the flat concave resonator is the used one.

POWER SUPPLIES AND COOLING SYSTEMS

The power supplies for pulsed mode operation were developed to cover the range of

Figure 2: Pulse of the Q-switched Nd:YLF laser using $LiF:F_2^-$ crystals as the passive Q-switcher.

50 to 250 J and pulse durations can range from 100 ms to 310 ms. For the continuous wave, the electrical output power ranges from 1000 W up to 3000 W. The electrical system has safety interlocks to avoid overheating of the system and also to 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.

LASER PROTOTYPES

a) Pulsed models

Due to the simplicity of construction, the first lasers developed were the pulsed ones, and 3 models were already constructed [8]. One is a hand held, rugged model, with a parallel flat mirror configuration, with 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 multi mode free running energy is 500mJ.

Using a saturable absorber consisting of LiF:F_2 crystal we were able to optimize the Q-switching operation [9] of this laser, obtaining pulses as short as 5 ns, what is comparable to that 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 in our lab. It was found that the best LiF crystals are the ultra pure ones, that can be obtained in our 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 the second harmonic.

The third model is an industrial prototype 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 ms, tailored to milling of stainless steel.

b) CW Lasers

For the c.w. operation mode, we have developed three laser models. The first laser 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 preceding one, using two lamps for pumping. In this case, the maximum output power is 1 W. 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 thermal lens can be easily compensated and therefore the resonator configuration is the standard plane concave. The maximum electrical input power is 3 KW and in this condition we obtained 15 W in multimode operation and 3 W in the TEM_{00}, in a concave-convex resonator.

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