

# Optimization of a Color-Center Q-Switched Nd:YLF Laser

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## Abstract

An easy and unexpensive optimization method for passive Q-switching of Nd pulsed lasers using LiF:F<sub>2</sub><sup>-</sup> color centers has been proposed and experimentally verified, showing that these centers can be a feasible alternative for an all solid state Q-switched laser. 12 MW peak power in a single pulse of 9 ns FWHM was obtained by proper choice of the output coupling and saturable absorber transmission. Complete mode-locking was also observed indicating a possible stimulated decay of the F<sub>2</sub><sup>-</sup> centers contributing to the field and providing a very fast gate during the intense bursts of the pulse.

## Introduction

Nd:YLF laser crystal has become an alternative to Nd:YAG oscillator lasers due to its optical and thermal properties [1,2,3]. In particular it shows the possibility of lasing in two wavelengths with a much wider emission band allowing for much shorter pulses in C.W. mode-locking operation as well as much higher average output power due to the low thermal lensing effect that can be compensated even under stronger pumping [3]. In the pulsed regime this crystal has two advantages: one of the emission wavelengths coincides with the emission of phosphate Nd:glass lasers, and the lifetime of the metastable <sup>4</sup>F<sub>3/2</sub> level (480μs) [1] is much longer than in the Nd:YAG (230μs). In particular its long decay time makes this laser material very convenient for pulsed Q-switched operation when pumped by conventional Xenon arc lamps. Among the Q-switching techniques, the passive one is the simplest and has been utilized mostly with dye cells [4].

There is, however, a family of color centers in ionic

crystals that absorb efficiently in the emission region of the Nd lasers that provides an alternative for all solid state Q-switched pulsed lasers. There are essentially three well known color centers absorbing in the 1 micron region: F<sub>2</sub><sup>+</sup>:O<sup>-</sup>:NaCl; Tl<sup>0</sup>(1):KCl and F<sub>2</sub><sup>-</sup>:LiF [5,6]. The drawback in the first two is their low temperature operation requirement, what is not the case for the F<sub>2</sub><sup>-</sup> color centers. Besides, these crystals can stand very high peak power at room temperature.

Our aim is to use F<sub>2</sub><sup>-</sup>:LiF color centers as passive Q-switchers in an optimized condition. The measured maximum gain allows for the calculation of the optimum output coupling and non saturated crystal absorption. Theoretical consideration and the experimental results will be described as well as the achievement of mode-locking operation.

## Experimental Conditions

Our laser medium is a 6 mm in diameter by 70 mm of length of Nd:YLiF<sub>4</sub> single crystal doped with 0.9 mol% of Nd oriented with the c axis perpendicular to the axis of the rod, so the laser operates in the π polarization (1,047 nm), grown in our crystal growth facility. The starting materials for the crystal growth were synthesized from ultra pure rare earth oxides utilizing a hydrofluorination procedure. The Nd:YLF thus synthesized was grown by Czochralski's method under Argon atmosphere. The Nd:YLF boule underwent a thermal treatment prior to the rod preparation, to eliminate stress originated from the growth process. Rods were extracted from the boule after the appropriate choice of a region free of scattering defects.

The rod was pumped by a low pressure Xenon lamp, in a cylindrical cavity made of pyrex silver coated. The electrical input energy is 50 J in a 150 μs FWHM pulse duration with a repetition rate up to

10Hz. The cavity mirrors are flat, dielectric coated for high reflection at  $1.05 \mu\text{m}$ . With an output coupler transmission of 53% we obtained a maximum of 300 mJ per pulse. The cavity length is 40 cm in all cases.

Ultra pure (U.P.) LiF crystals were also grown in our laboratory facility by a Czochralski's method from zone refined material treated under HF atmosphere. The concentration of  $\text{OH}^-$  is smaller than  $5 \times 10^{16} \text{ cm}^{-3}$ , controlled by infrared spectroscopy.

The  $\text{F}_2^-$  centers consist of two adjacent anion vacancies shared by three electrons [6]. They are created by radiation damage. We used  $e^-$  beam irradiation and gamma-ray irradiation. In large doses after the usual statistical aggregation process, we observed that in the case of electron irradiation the crystal showed formation of several aggregates ( $\text{F}_2$ ,  $\text{F}_3$ ) in a comparable size with the  $\text{F}_2$  band, showing a distinct brownish color different from the gamma-ray irradiated pure samples. Crystals with significant  $\text{OH}^-$  concentration also show a similar color of the  $e^-$  irradiated samples, due to the fact that  $\text{OH}^-$  doped under irradiation present very efficient electron traps favoring a higher color centers concentration other than  $\text{F}_2$  color centers [7]. A typical absorption spectrum of U.P. LiF crystals irradiated by gamma-ray is shown in Fig. 1. We can clearly see a pronounced band peaking at 960 nm corresponding to the first absorption of these centers. Their emission band is centered at  $1.12 \mu\text{m}$  showing a decay time of 100 ns (Fig. 2) probed by a fast pump (10 ns). According to the literature [6] this decay time is in the range of 65-105 ns depending on the method of creation. The long observed decay time is a strong indication of isolated centers.

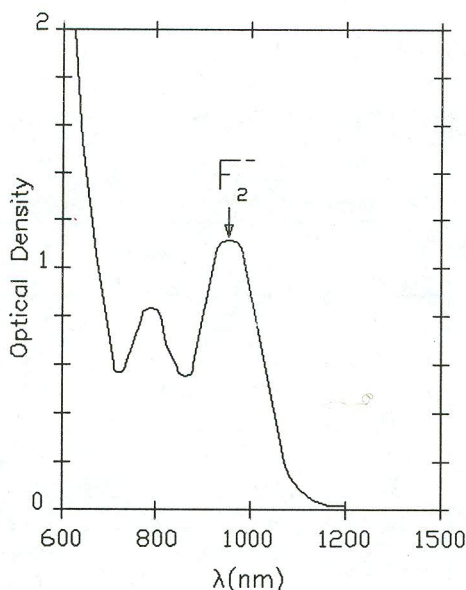


Figure 1 - Absorption spectrum of a 4.4cm U.P. LiF crystal irradiated with 63 Mrad of  $^{60}\text{Co}$   $\gamma$ -rays

The dependence of the peak absorption coefficient of the  $\text{F}_2^-$  as a function of the gamma dosage is shown in Fig. 3. We can see that above 60 Mrad there is a saturation effect, so longer exposures do not increase the  $\text{F}_2^-$  center concentration significantly. The absorption cross section at 1047 nm is  $1.5 \cdot 2 \times 10^{-17} \text{ cm}^2$  [6].

## Theoretical Analysis

There are two main concepts in passive Q-switching. First, the capacity of the saturable absorber to block the initial laser operation by the losses due the light absorption during the pumping, in order to achieve the maximum inversion population possible. Second, the rapidly decreasing absorption of the Q-switcher due to the intra cavity intensity increase in the crystal, during the growth of the laser field.

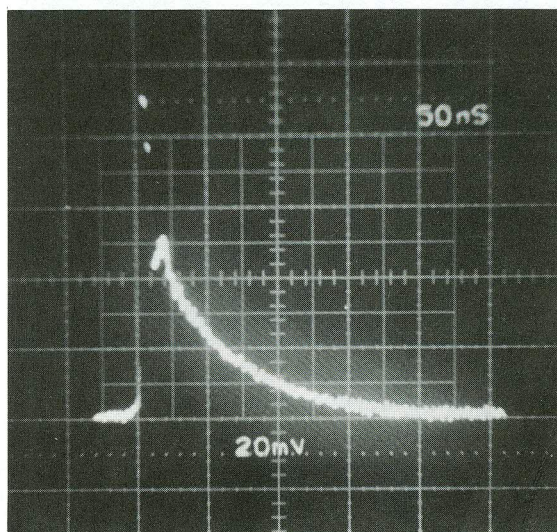


Figure 2 - Emission decayment of the  $\text{F}_2^-$  in LiF at 1080 nm ( $\Delta t$  pumping  $< 10\text{ns}$ )

The transmission of the saturable absorber as a function of the intensity is given by:

$$\ln T = \left( \frac{1}{1 + I/I_s} \right) \cdot \ln T_0 \quad (1)$$

where  $I_s$  is the saturation intensity. For a 4 level system  $I_s$  is given by:

$$I_s = 1 / (\sigma_a \cdot \tau) \text{ cm}^{-2} \text{ s}^{-1} \quad (2)$$

where  $\sigma_a$  is the absorption cross section and  $\tau$  the total decay time. The Q-switched operation starting sequence is the following: the laser medium is pumped in a time scale preferably shorter than the decay time producing an inverted population that follows the integral of the pump pulse; during this process

spontaneous emission in the laser channel builds up. As the losses due to the saturable absorber are still high, the gain is still below threshold preventing lasing [8]. As the intensity grows, the absorber starts the saturation process decreasing the losses until the gain reaches the losses and laser action starts. The intensity grows several orders of magnitude in few cavity round trips bringing down the Q-switcher absorption to negligible amounts according to Eq.(1). It can be shown that the output peak power, in these conditions, is given by [9]:

$$P_p = (Vh\nu / 2 t_r) \ln \frac{1}{R} \left[ N_i - N_t \left( 1 + \ln \frac{N_i}{N_t} \right) \right] \quad (3)$$

where V is the laser mode volume;  
 $t_r$  is the cavity round trip time;  
 R is the output mirror reflectivity;  
 $N_i$  is the initial population inversion at the laser pulse formation;  
 $N_t$  is the cavity laser threshold population without the absorber.

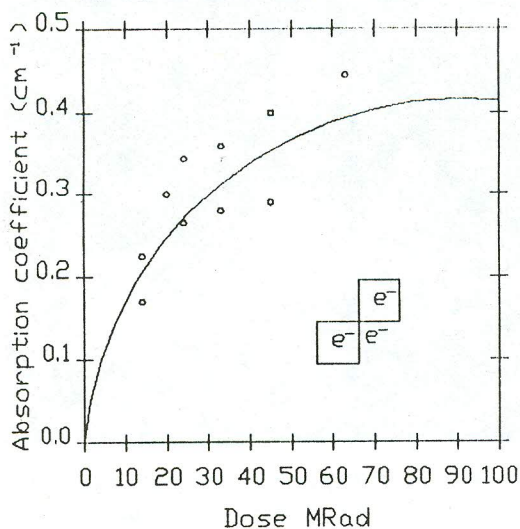


Figure 3 - Absorption Coefficient of the  $F_2^-$  in LiF (960 nm) as a function of  $\gamma$ -ray dose. The insert shows the  $F_2^-$  Color Center Scheme.

In order to achieve the maximum output peak power it is necessary to maximize the inverted population before laser action starts. The threshold population is dependent upon the mirror reflectivity that determines the cavity photon lifetime and therefore the pulse duration.

The limiting parameter is the maximum gain, what is a characteristic of the laser medium, the pump system and the optical cavity. We make the assumption that the maximum gain is achieved by a proper choice of output coupling and initial Q-switcher absorption. The laser action is prevented until the maximum possible population is achieved providing a maximum

gain given by  $g_m = \sigma_e N_i L$  which is a measurable quantity. We can then express the output power given by Eq.(3) as a function of the threshold inverted population through the usual laser threshold condition:

$$1 = R \exp [ 2 g_t - \gamma ] \quad (4)$$

where  $g_t = \sigma_e N_t L$  is the threshold gain, L is the length of the gain medium and  $\gamma$  the internal loss per double pass. Using Eq.(4) in Eq.(3) and maximizing the output peak power as a function of the mirror reflectivity by imposing  $\delta P_p / \delta R = 0$  we obtained:

$$\frac{g_m - g_t}{g_t} = \ln \left( \frac{g_m}{g_t} \right)^2 \quad (5)$$

where R is determined by the value of  $g_t$  using Eq.(4). In order to achieve the maximum population inversion the non saturated transmission  $T_o$  then is given by:

$$T_o = \exp [ g_t - g_m ] \quad (6)$$

The method for determining the maximum gain [10] is achieved by introducing slightly tilted microscope slides until lasing is prevented. The maximum gain is:

$$g_m = \gamma / 2 - \frac{1}{2} \ln R - n \ln t \quad (7)$$

where t is the microscope slide transmission (0.92) and, n is the number of slides. Assuming 5% internal losses per pass, we obtain a maximum gain of 1.25.

## Laser Operation

Inserting the LiF: $F_2^-$  crystal in the resonator close to the back mirror and slightly tilted to avoid spurious reflections we measured the number of pulses, the

TABLE 1  
Parameter variation With  $F_2^-$  initial Absorption

Number of pulses	P.Width (ns)	Out. Energ.(mJ)	Output Coupling	Initial Absorp.
> 10	200	300	0.47	0.017
> 10	100	240	0.47	0.038
> 10	100	290	0.47	0.053
> 10	100	300	0.47	0.064
6-7	28	180	0.42	0.22
4	25	180	0.42	0.33
2-3	15	160	0.42	0.36
2-3	28	80	0.04	0.22
2	25	80	0.04	0.33
1	12	100	0.04	0.36
1	9	110	0.42	0.48

pulse width and the total energy as a function of the transmission of the front mirror and the initial absorption of the saturable absorber at 1.047 nm. These results are shown in TABLE 1.

According to the theoretical considerations and taking the measured value of  $g_m = 1.25$  we obtained the optimum gain threshold value by Eq.(5), obtaining  $g_t = 0.36$ . Including the assumed internal loss of 0.05 per pass, the optimum reflectivity is  $\approx 54\%$ . Therefore the optimal initial transmission of the saturable absorber can be obtained from Eq.(6) and is 41%. It is clearly seen that as we increase the absorption of LiF:F<sub>2</sub> we decrease the number of pulses due to repumping of the gain medium and the peak power increases accordingly. As we get closer to the theoretical optimum value, the number of pulses tends to one and the pulse width shortens to a minimum value. The best result is obtained with an output coupling transmission of 42% and an initial transmission of the saturable absorber of 52%. In this case the pulse width is 9ns measured with a scope with 3 ns of rise time as shown in Fig. 4. This is comparable to electro-optic switcher devices.

Two facts limited the improvement of the laser operation parameters. The mirror with 47% transmission which value is closer to the predicted, was destroyed during the tests; and the absorption of the crystal achieved the saturation, as shown in Fig. 3. In order to increase the absorption it would be necessary to increase the length of the crystal to avoid this saturation problem [7]. The actual crystal length in our experiments is 44 mm. The peak power obtained was 12 MW below the calculated 22 MW for an effective mode corresponding to a diameter of 6 mm, that would correspond to a complete utilization of the rod volume. Due to some imperfections in the border, thermal effects and filamentary Q-switching in the saturable absorber a smaller rod area is being utilized

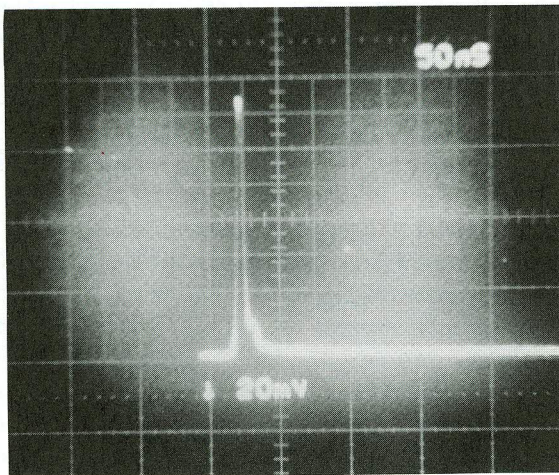


Figure 4 - Single Q-Switched Pulse (50 ns/div Reduced Scale).

In fact, just a reduction of 25% in the rod diameter would match exactly the obtained experimental value of the power output. Besides there are reflection losses in the LiF crystal that were not considered here.

During the experiments, we also increased the cavity length observing a change in the pulse profile with a longer envelope in the range of tenths of nanoseconds with a mode-locking structure with 100% modulation, as shown in Fig. 5. The pulse period corresponds to a cavity length of 1 m. The parameters range for this mode-locking operation is quite large. We used several sets of mirror transmissions and saturable absorber transmissions, always observing the

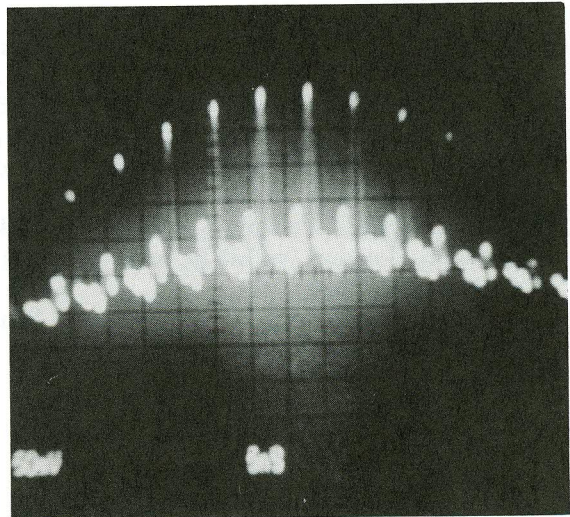


Figure 5 - Full mode-locked train. The cavity length is 100 cm (5 ns/div reduced scale)

mode locking operation. We believe that this is due to a pulse shaping mechanism occurring in the saturable absorber.

These centers have a small Stokes shift presenting a strong overlap, at room temperature, of the emission and absorption bands [7]. This overlapping occurs at the Nd lasers emission such the pump light that is absorbed by these centers can also induce stimulated emission of the inverted population. As a matter of fact the emission cross section at the peak of the band (1.12nm) is just 1.4 times greater than at the 1047 nm (Nd:YLF  $\pi$  polarized). We suppose that, at first, the leading edge of the pump pulse is absorbed producing an inverted population, with some short delay, due to non radiative relaxations in the sub picosecond or in the picosecond time scale [11]. This will provide a net gain to the more intense part of the pulse that, in turn, depopulates the excited state bringing back the saturable absorber to the absorbing state again. Therefore, in spite of having a long decay time (100 ns) the F<sub>2</sub> center, actually behaves as a very fast gate, producing entirely the pulse shaping mechanism for the mode-locking regime. This idea is also

corroborated by the results of C.W. operation of the  $F_2^-:LiF$  using Nd:YAG lasers [12] whereas a negligible efficiency was obtained due to this temporal mechanism that will greatly reduce the pumping efficiency. Time resolved spectroscopy studies are under way to exploit this temporal behavior.

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