# Color centers as simultaneous active laser media and saturable absorbers ☆

N.D. Vieira Jr., L.S. Assis and S.P. Morato

*Instituto de Pesquisas Energéticas a Nucleares, Comissão Nacional de Energia Nuclear - SP,*
*Caixa Postal 11040, CEP 05499, São Paulo, Brazil*

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There is a family of color centers and metal ion doped crystals that absorb in the emission region of the neodimium lasers. These centers are in general laser active, usually being pumped in a collinear scheme. They can also be used as saturable absorbers for cw passive Q-switching, but this limits the maximum absorption that can be introduced in the laser cavity thus limiting the maximum gain achievable in the saturable absorber/active pumped medium. By using a coupled cavities scheme we were able to optimize the performance of the pumped medium and obtain CW Q-switched operation by an injection mechanism, the saturable absorber being the unique source of loss for the pump. We coupled an astigmatically compensated KCl:$Tl^0(1)$ color center laser cavity with a Nd laser cavity using both properties of the KCl:$Tl^0(1)$ crystal. We obtained complete Q switching of both lasers with a typical pulse width of 1 μs and a pumping efficiency of the color center laser in the order of 40% achieving a maximum output power of 1.3 W for an input pump power of 3 W from the Nd:YAG laser. The Nd:YAG Q-switched operation was also tested in this coupled cavities scheme using LiF:$F_2^-$ color centers. The obtained pulse widths agree well with a simple theoretical model.

## 1. Introduction

Color center lasers (CCL) behave similarly to dye lasers and are their counterpart in the near infrared region [1,2]. They are very efficient mostly due to the possibility of being pumped by another laser in very small areas ($\sim 10^{-5}$ cm$^2$), and achieving high gain in very small volumes ($\sim 10^{-6}$ cm$^3$). In particular there is a class of color centers that can be efficiently pumped by Nd lasers due to their strong absorption bands in the 1 μm region. Some of these centers are $Tl^0(1)$ in KCl [3,4] ($\lambda = 1.040$ μm) and $F_2^-$ in LIF[5,6] ($\lambda = 0.96$ μm) with broad absorption bands that overlap the Nd laser emission. The $Tl^0(1)$:KCl center emits in the 1.5 μm region and must be operated at 77 K. The $F_2^-$ :LiF center emits at 1.12 μm and is stable at room temperature even under strong pumping laser light [7].

Our objective was to use these centers as saturable absorbers and gain media simultaneously. As a saturable absorber medium it controls the temporal behaviour of the Nd:YAG laser depending on its saturation intensities, amount of absorption and decay times. As gain media their decay time depends upon the intracavity intensity of the CCL radiation field.

Particularly, for low absorption laser media in which the efficiency is limited by their small absorption, just a double pass of the pump is enough to improve the laser performance [8]. Ideally, intracavity pumping would be an alternative method to overcome the small absorption problem, but due to the low pumping rate of the pump lasers it would not be feasible. By using a coupled cavities scheme, it is possible to increase the initial absorption of the pumped medium in order to increase the gain of the slave and still keep a low threshold for the cw operation of the Nd:YAG.

## 2. Theoretical

In order to analyse the behavior of the coupled

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cavities scheme we assume that the spatial modes are matched. The main cavity consists of a 100% reflecting mirror ($M_1$) and an output coupler ($M_2$) of reflectivity $R$, as shown in fig. 1. A second branch consists of an absorbing medium (with a transmission $\tau$) and a back mirror with reflectivity $R_0$. By summing up all the electromagnetic contributions reinjected into the main laser the equivalent reflectivity for the set $R$, $R_0$ and $\tau$ is given by [9]

$$R_{\text{eff}} = \frac{[\sqrt{R} - \tau\sqrt{R_0}\exp(\mathrm{i}\phi)]^2}{[1 - \sqrt{RR_0}\,\tau\exp(\mathrm{i}\phi)]^2},$$

where $\phi$ is the phase shift due to the optical length between mirrors $R$ and $R_0$ and due to the absorbing medium and the back mirror. For a constructive interference ($\phi = (2m+1)\pi$, $m$ integer), the reflectivity will be maximum, being in this case

$$R_{\text{eff}} = \frac{[\sqrt{R} + \tau\sqrt{R_0}]^2}{[1 + \tau\sqrt{RR_0}]^2}. \tag{2}$$

Due to the increase in the reflectivity for the constructive modes the pump laser will preferentially oscillate in the selected modes, but if the pump gain laser bandwidth is much greater than the cavity mode spacing there will be allways several modes that will efficiently depopulate the gain medium.

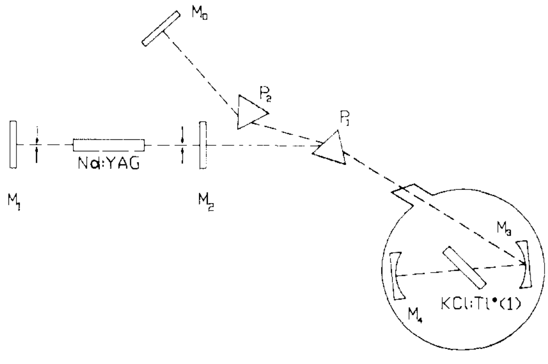

Fig. 1. Scheme of the coupling of the Nd:YAG laser to the Tl$^0$(1):KCl color center laser. The Nd:YAG laser cavity consists of mirrors $M_1$ and $M_2$; mirrors $M_3$, $M_4$ and $M_0$ are the CCL astigmatically compensated cavity. The spatial modes of both lasers overlap at $M_2$. Prism $P_1$, at Brewster angle, allows for the collinearity of both beams at the KCl:Tl$^0$(1) crystal. The apertures inside the Nd:YAG laser are for $TM_{00}$ operation. Mirrors $M_3$, $M_4$ and the crystal are in a vacuum chamber. $M_3$ and $M_4$ are gold coated mirrors. Prism $P_2$ intercepts the CCL beam, directing it to mirror $M_0$.

Assuming now that the back mirror reflectivity is unity ($R_0 = 1$) for the coupled mode, the main cavity threshold is lowered by a quantity $\Delta g = (1-R)\tau/(\tau + \sqrt{R})$, so lasing will occur before achieving the free resonator threshold gain $g_0$.

The transmission of the saturable absorber depends on the incident intensity according to

$$(1 + I/I_S)^{-1}, \quad \tau = \tau_0 \tag{3}$$

where $I_S$ is the saturation intensity.

At the begining of the operation, the main cavity gain is lowered by $\Delta g(\tau_0)$ until the laser action begins. At this point, the intensity grows such that $I \gg I_S$; therefore $\tau \approx 1$ and the new threshold is

$$g_0 - (1-R)/(1+\sqrt{R}),$$

and the system is now with its population inversion much higher than the cavity threshold population, initiating the Q-switched operation.

The photon life time in this coupled scheme depends entirely upon the ratio of energy transfer between both cavities and on the saturable absorber transmission dependence on the time. The dependence of the total intracavity power, normalized by the maximum power, on the transmission of the saturable absorber is given by

$$\frac{P_\tau}{P} = \frac{(1+\sqrt{R})}{2}\left[1 + \left(\frac{t\tau}{1+\tau\sqrt{R}}\right)^2\right], \tag{4}$$

where $t = \sqrt{T}$; and $T = 1 - R$.

The time dependence of $P_\tau$ is given by

$$\frac{1}{P}\frac{\partial P_\tau}{\partial t} = \frac{(1-\sqrt{R})(1+\sqrt{R})^2}{(1+\tau\sqrt{R})^3}\,\tau\frac{\partial\tau}{\partial t}. \tag{5}$$

At the maximum of the saturation, the population of the saturable absorber is fully inverted (for a 4-level scheme). The transmission at this point is given by $\tau = 1 - \sigma nl$ ($\sigma$ is absorption cross section, $n$ is the ground state population and $l$ is the length of the absorbing medium). The recovery of this population is exactly the decay of the total population $n_0$ with an effective decay time $t_d$. Therefore, around $\tau \approx 1$ we have

$$\frac{\partial\tau}{\partial t} = \frac{\ln\tau_0}{t_d}, \tag{6}$$

and consequently

$$\frac{1}{P}\frac{\partial P_\tau}{\partial t}=\frac{1-\sqrt{R}}{1+\sqrt{R}}\frac{\ln \tau_0}{t_d}. \tag{7}$$

At the beginning of the intensity decay the behavior is exponential with a characteristic decay time $t_c$ given by

$$t_c=\frac{1+\sqrt{R}}{1-\sqrt{R}}\frac{t_d}{\ln \tau_0}. \tag{8}$$

When $R=1$, the decay time is infinite (cw operation) corresponding to an uncoupled cavity without output coupling. At $\tau_0$ approaching unity (no absorption), the cavities are coupled but, as there are no time dependent losses, we also obtain cw operation. It is also easily seen that the first factor of the above expression takes into account the coupling between the two cavities (the energy transfer term). The second factor is solely dependent on the saturable absorber physical parameters.

## 3. Experimental

KCl:Tl crystals were grown in our laboratory by the static Bridgman method using reagent grade starting materials sealed under argon in quartz ampoules. Single crystals 5 cm long with diameters of 2 cm were obtained with a Tl concentration of about 0.2 mol% measured by emission spectrography. This concentration is in the ideal range due to the fact that higher quantities of Tl impurities in the crystal generates undesirable Tl aggregates [10]. These samples underwent a thermal treatment of 650°C for 30 minutes followed by a rapid quench to room temperature to eliminate all the undersirable aggregation of Tl centers. This thermal treatment is rapid enough to avoid a considerable loss of substitutional Tl ions through the sample's surface.

Samples of $10\times10\times2$ mm$^3$ cleaved and polished were irradiated on each face at 77 K by an electron beam accelerator with a current of 20 μA min/cm$^2$. The obtained F center concentration was in the order of $10^{18}$ cm$^{-3}$, also within the ideal range to avoid F aggregates. After electron irradiation these samples were optically treated with light in the F center region at −30°C photoconverting F centers into $Tl^0(1)$ centers. Densities up to $10^{17}$ cm$^{-3}$ of the $Tl^0(1)$ were obtained under these conditions with an absorption band peaking at 1.04 μm, corresponding to an initial absorption of 90%.

Crystals of LiF were also obtained in our laboratory by initially zone refining (under HF atmosphere) a reagent grade LiF starting material. The material thus purified was used to grow LiF crystals by Czochralski's pulling method under argon. Single crystal boules 30 mm of diameter and 80 mm of length were obtained and cleaved into plane samples of $10\times10\times2$ mm$^3$. These samples were electron beam irradiated at room temperature during a time enough to produce a statistical concentration of $F_2^-$ centers that caused an optical absorption of 20% at 1.064 μm.

The slave laser cavity is the usual three-mirror astigmatically compensated cavity where the pumped medium is placed in the tightly focusing region ($w_0\approx20$ μm) and is designed to be able to operate at low temperatures. For the CCL operation at 77 K a special cryostat was constructed with a holding $LN_2$ time of approximately 70 hours for a volume of 5 liters.

The pumping of the CCL with the Nd:YAG laser was done using a collinear arrangment as shown in fig. 1. In this figure it is shown the coupling of the two laser cavities: the plano-concave Nd:YAG laser back mirror $M_1$ and the output flat mirror $M_2$ are aligned collinearly with the astigmatically compensated CCL cavity (mirrors $M_3$, $M_4$ and $M_0$). The pump beam mode is matched exactly with the CCL mode by coinciding the beam waists of both lasers at mirror $M_2$, since in the long arm of the CCL the beam waist is almost exactly the waist of the Nd:YAG laser (0.5 mm radius) at the output mirror (see fig. 1). A prism at Brewster angle inserted in front of the CCL vacuum chamber allowed for the collinearity of both laser beams in the highly focused region of the CCL cavity (between mirrors $M_3$ and $M_4$). This equilateral prism, made of dense flint glass produced a dispersion of 1 mm/10 cm for the 1.064 μm laser beam. Another equivalent prism was used to intercept only the 1.5 μm beam deflecting it also in the minimum deviation angle. The latter prism directed the CCL beam into the CCL cavity output mirror. It was used a CCL output mirror with a curvature radius of 3 m thus allowing for the long arm of the CCL to be stretched to an optimum experimental condition ( ≈80 cm) and still keeping its stability range in the 1 mm region (and therefore matching the mode waist at the crystal). The output mirror transmission of this mirror ($M_0$) was 10% at 1.5 μm.

The coupling was tested using a dummy crystal in the focus of the CCL cavity. The laser threshold for the perfect coupling was essentially the operation threshold of the pumping lamp of the Nd:YAG laser, since this coupled cavity has no output coupling but just minor losses.

The $M_2$ output coupler reflectivity $R$ of the Nd:YAG laser used was 88% for KCl:Tl$^0$(1) and 72% for $F_2^-$ :LiF. In the latter case, mirror $M_1$ has 12% transmission at 1.064 μm. For the $F_2^-$ :LiF crystal a second arrangement was made. As the emission band overlaps the absorption band in the Nd laser emission line, the $F_2^-$ center firstly absorbs, and, after a delay time, emits in the same pumping wavelength; therefore there is no need for a second cavity operating at the peak of the $F_2^-$ :LiF emission band. We then simplified the arrangement by eliminating the prisms.

## 4. Results and discussion

The coupling scheme was tested for both centers described above and the results are

### (A) *Tl$^0$(1):KCl center case*

In this case, the Nd:YAG laser mirror $M_1$ is 100% reflective, $M_2$ has 12% transmission at 1.064 μm. The main cavity gain threshold is ≈6.4% per pass and coupled with the CCL cavity the non saturated gain threshold is reduced to 5.2% per pass. At the saturated regime the threshold gain is just enough to compensate the residual losses. In this case, the net gain modulation for the non saturated and saturated regime is 5.2%, almost 3 times greater than in the $F_2^-$ center case. We observed that in this case the mechanical coupling is more critical and for pump powers above the main cavity threshold a misalignment leads to a mixture of pure cw and Q-switched pulses. By a judicious fine alignment, pure Q-switched operation can be achieved. The lamp operation current at laser threshold was lowered from 24 A to 20 A. The threshold of the CCL was the same as of the Nd:YAG laser. The laser was chopped with a low duty cycle to avoid thermal effects. The typical pulses are 1 μs long, as shown in the insert of fig. 2. The output power of the laser as a function of the lamp current is also shown in fig. 3. The non linearity observed is due to the thermal effect in the laser rod

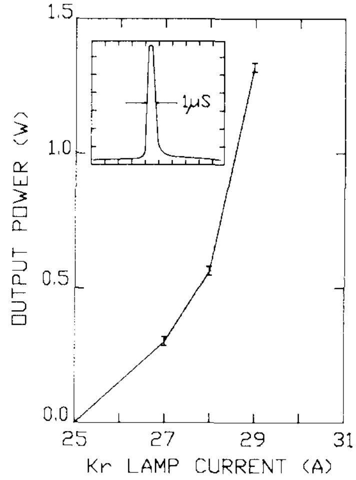

Fig. 2. The figure shows the power output of the Tl$^0$(1) center, in the coupled configuration of fig. 1, as a function of the pumping Kr lamp of the Nd:YAG laser. The observed non linearity is due to thermal effects in the Nd:YAG rod. In the insertion it is shown the pulse width of the Nd:YAG leaking through the back mirror ($T$≈0.2%) at 20 A.

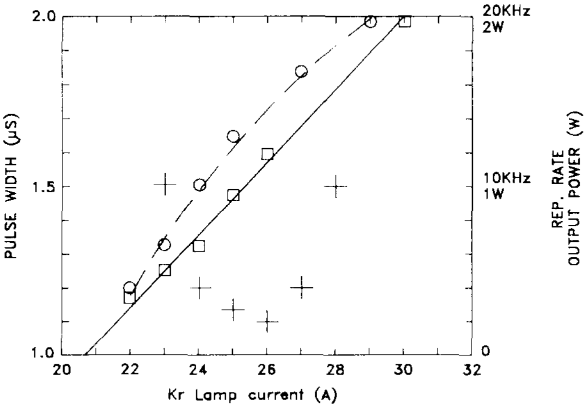

Fig. 3. Operation parameters of the Nd:YAG laser coupled with the CCL cavity of fig. 1 without the prisms, with the $F_2^-$ :LiF crystal. The coupling mirror $M_2$ has 28% transmission at 1.064 μm and the output coupling has 12% transmission (mirror $M_1$). The output power (square dots), the pulse frequency (circular dots) and the pulse width FWHM (crosses) are shown as a function of the Kr lamp pump current.

[12]. At the maximum lamp current used (29 A), the time average laser output was 1.3 W, corresponding to an absolute efficiency of 40%. The output pulses of the CCL were also modulated as it could be seen by a slow detector (Ge detector frequency response ≈ 100 kHz), but the actual pulse width was not measured.

Due to the fast stimulated decay rate in the color center laser, we expect that the CCL Q-switched pulses have approximately the same temporal pulse width (1 μs). Therefore, at the maximum pump lamp current (29 A), with a pulse rate of 25 kHz and an average power of 1.3 W, the energy per pulse is 52 μJ and the output peak power is 52 W. The repetition rate of the cw, Q-switched pulses was the same of the Nd laser as one would expect, and was 25 kHz at maximum lamp input power. The pulse frequency increases with the pump power.

In order to calculate the pulse width we have to consider that the total decay time is due to spontaneous and stimulated emission and is given, for high intensities [13], by $t_d = t_{d,0}\, I/I_S$. The saturation intensity for the $Tl^0(1)$ center is $4.8\times10^{22}$ cm$^{-2}$ s$^{-1}$ and, considering the average power as 1.3 W, for an area of $1.2\times10^{-5}$ cm$^{-2}$, we obtain an average decay time of 94 ns. The calculated characteristic decay time $t_c$ in this case is 1.3 μs, very close to the 1 μs measured.

The remarkable fact in this operating scheme is that, as the color center is the unique effective source of loss for the Nd:YAG laser, the pumping efficiency is shown to be very high. The fact that the laser is operating in the Q-switched regime justifies this increase in efficiency since the population is inverted in a time that is a fraction of the decay time. Then, losses due to the excited state population fluorescence are minimized.

### (B) $F_2^-$:*LiF center case*

The $F_2^-$ centers are formed by electron beam irradiation in concentrations orders of magnitude smaller than other primary color centers, therefore the maximum center concentration is limited to small values. For a thickness of 1.74 mm, the absorption at 1.064 μm is 20%. Using mirror $M_2$ with 12% transmission, the coupling was very weak, producing long Q-switched pulses (several microseconds). In order to increase the coupling factor, the transmission of mirror $M_2$ was increased to 28%. The main cavity threshold gain per pass in this case is 23% plus residual losses. By coupling the CCL cavity, the non saturated single pass gain threshold is reduced to 14.6% and the saturated one is reduced by an extra factor of 1.8% (this net gain modulation is very small). The uncoupled threshold gain is higher than the maximum possible laser gain so only the coupled laser cavity allows for laser action. The results are shown in fig. 2. The increase of the pump power reduces the gain build up time; consequently, the pulse frequency increases proportionally, increasing the average output power. By increasing the pump power, the pulse width initially decreases, achieving a minimum of ~1 μs. As the thermal lens effect in the Nd:YAG rod takes over, the cavity mismatch diminishes the role of the saturable absorber and produces longer pulses.

Considering that the $F_2^-$ saturation intensity is $4\times10^{23}$ cm$^{-2}$ s$^{-1}$, and using expression (3), the effective decay time is reduced to 35 ns, thus the characteristic photon decay time is 2 μs, much longer than the experimental value of ~1 μs. The reasons for this discrepancy are: firstly, the single pass gain variation during the saturation of the absorber is very small (1.8%), what produces a slow build up of the pulses; secondly the 12% coupling of mirror $M_1$ presents an extra loss per pass that should account for the further reduction in the cavity photon life time [14].

The output power obtained is ~65% of nominal cw mode due to the high saturation intensity of the $F_2^-$ color centers. These losses are due to the energy required to bleach the saturable absorber. Part of this power is lost by luminescence (the decay time is ten times faster than the pulse width).

## 5. Conclusions

It was shown that by coupling two cavities, a cw Q-switching of the Nd:YAG laser was obtained due to the injection mechanism of the beam after passing through the color center medium. Secondly, the unique loss mechanism of the Nd laser is the absorption of the $Tl^0(1)$:KCl centers implying, therefore, a very efficient pumping scheme. In the $F_2^-$:LiF case, the stimulated emission of the $F_2^-$ center occurs at the pumping wavelength due to its small Stokes shift. Therefore, in this case, a simple coupled cavities arrangement was enough to produce Q-switched pulses of essentially the same pulse width (1 μs) and with constant amplitude.

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