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Efficient single-pulse emission with submicrosecond duration from a Cr:LiSAF laser

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Abstract

Using Cr:LiSAF as active medium in a simple, near-hemispherical resonator configuration, laser pulses with energies of 60 mJ and widths of about 180 ns (FWHM) in the TEM_{00} mode at a center wavelength of $\lambda = 830$ nm were obtained without the use of any additional (switching-) components within the cavity. Even longer pulses at the same energy levels are demonstrated using a ring resonator. A self-Q-switching effect in Cr:LiSAF is assumed and discussed.

Keywords: Cr:LiSAF laser; Saturable absorber; Self-Q-switching; Kerr lens Q-switching

1. Introduction

There is a large market for broadly tunable, pulsed solid-state lasers (spectroscopy, lidar, analytical chemistry). In many cases, these laser systems are realized by Q-switching Ti:Sapphire or Cr:LiSAF since both laser materials have a broad emission band in the near infrared (from about 750 to 1000 nm). This band can be extended to a band in the blue range and to a band in the mid-infrared region by second harmonic generation and by sum and difference frequency mixing, respectively, when using nonlinear-optical devices. Ti:sapphire has an emission cross section which is higher than that of Cr:LiSAF but the much lower excited state lifetime (3 μs compared to 67 μs [1,2]) makes it less suitable for flashlamp pumping. Thus, Cr:LiSAF seems to be a good material for a flashlamp-pumped, efficient, broadly tunable, pulsed oscillator-amplifier-system although some loss processes (excited state absorption, upconversion [3]) must not be neglected. The Q-switched Cr:LiSAF laser systems reported up to the present deliver in most cases signals with pulse widths (FWHM) of only a few tens of nanoseconds up to about one hundred nanoseconds [4,5,6]. So, high output energies mean high power densities which may cause damage problems for both the oscillator and amplifier rod [7] and for the other cavity components [4], and, at last, for a frequency transforming nonlinear crystal at the amplifier output. This problem can be diminished by increasing the pulse width, thus reducing the power density. We introduce an efficient Cr:LiSAF oscillator delivering pulses in the TEM_{00} mode with pulse widths in the submicrosecond range. The oscillator consists of a simple near-hemispherical resonator configuration and does not need any supplementary switching components. It is assumed that thereby Cr:LiSAF is working as both, gain medium and passive Q-switcher. This will be discussed in detail.

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2. Experimental set-up

We use a near-hemispherical resonator configuration providing the largest mode size differences (Fig. 1) which is equivalent to the largest power density differences along the resonator axis. The beam diameter reaches a maximum at the curved mirror. So, the utilization of a large active volume and therefore the exploitation of a large part of the input energy can easily be achieved by moving the laser rod versus the HR-mirror ($R_{max,\ curved}$) until the beam diameter corresponds with the rod diameter. Thus, in addition, the sensitive Cr:LiSAF rod is exposed to a minimum of power density yielding maximum protection from destruction. The beam waist occurs at the flat output coupler. This results in a high power density of the output radiation. By making small adjustments in the resonator length the spot size at the curved mirror end can be made as large as desired, whereas the spot size at the flat mirror end becomes correspondingly tiny.

A linear (Fabry-Pérot type) resonator configuration like the near-hemispherical resonator is characterized by standing waves. For this reason spatial hole burning and therefore mode beating occurs, limiting the quality of the radiation. This can be avoided without the use of any intracavity components (e.g. quarter wave plates, etalons etc.) by working with a unidirectional ring resonator as shown in Fig. 2. Here, a traveling wave in the resonator is favored by introducing an additional curved HR-mirror ($R_{max,\ curved}$) outside the ring cavity [8]. Substituting this HR-mirror by an injection seeding component, the linewidth of the oscillator can efficiently be narrowed thereby increasing the spectral brightness.

We use a Cr:LiSAF rod of 3 mm diameter and 55 mm length, 1.5% chromium doped and with flat end surfaces, both AR-coated (750–950 nm).

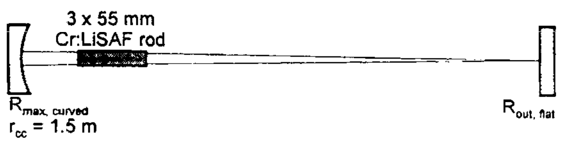

Fig. 1. Experimental set-up: Near-hemispherical resonator configuration, corresponding TEM_{00} mode and location of the Cr:LiSAF rod within the cavity.

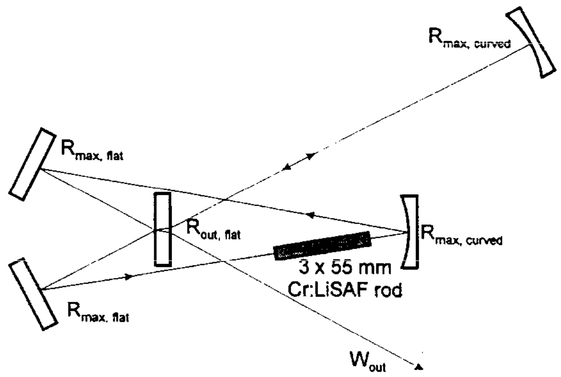

Fig. 2. Experimental set-up: Unidirectional ring resonator.

The pumping head consists of an elliptical gold-plated reflector with a 2″ long flashlamp of 3 mm arc diameter. The coolant is a 1 : 1 mixture of ethylene glycol and water which yields a pH of 7, thus diminishing dissolution of the Cr:LiSAF crystal [9]. The addition of 0.1 g of Rhodamine 6 G per l coolant provides a transfer of the UV and the blue-green emission range of the flashlamp spectrum to the absorption broadband of the Cr:LiSAF in the red. Thus, a better efficiency and a reduction of thermal lensing effects can be achieved. The power supply delivers a nearly rectangular pulse of 60 μs duration with energies of up to 12 J at repetition rates of up to 100 Hz.

In order to generate a single pulse (in both resonator configurations, the near-hemispherical and the ring resonator) no additional switching component is used. The pump energy is increased in such a way that a maximum of energy is deposited in a single spike but that further pulses are still suppressed.

3. Experimental results

In a first set of experiments, we used the near-hemispherical resonator configuration as described in the previous section. The radius of curvature of the HR-mirror was 1.5 m, the distance between HR-mirror and laser rod was 0.11 m. The laser oscillated in the TEM_{00} mode at a center wavelength of $\lambda =$ 830 nm.

In a first experiment, we chose the cavity length to 1.25 m. For different reflectivities of the output coupler we measured the output energies of the single pulse and the corresponding pulse widths (FWHM). The results are shown in Fig. 3.

In a second experiment we measured the output energies and corresponding pulse widths in dependence on the cavity length while choosing the reflectivity of the output coupler to be 50%. These results are shown in Fig. 4.

Reducing the reflectivity of the output coupler to 40% and increasing the cavity length to 1.48 m, a pulse energy of 60 mJ at about 180 ns pulse width was obtained. In all cases, the output pulses were absolutely stable and reproducible concerning the pulse width and the energy.

Using the ring resonator described in the previous section and comparing pulse widths and energies with these of the near-hemispherical resonator with corresponding round-trip paths, we realized that at the same output energies even longer pulse widths can be obtained. This is another advantage of the ring resonator when high oscillator output energies at long pulse widths are desired beside those advantages already mentioned. Increasing the length of the ring resonator to a round-trip path of 2.9 m (which corresponds to a length of the near-hemispherical resonator of 1.45 m), and setting the reflectivity of the output coupler to 80% and the radius of curvature to 1.5 m, an energy of 34 mJ at a pulse width of 500 ns was obtained.

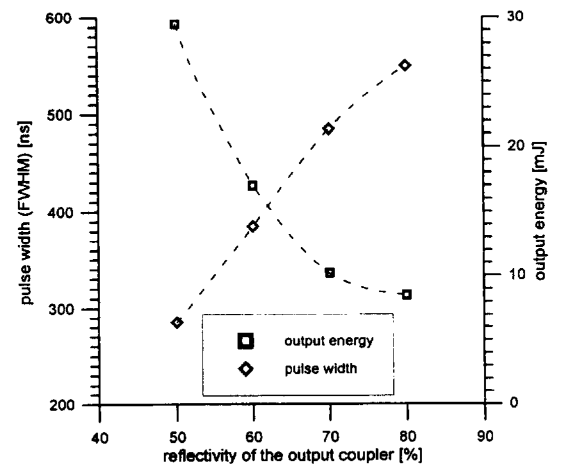

Fig. 3. Pulse width and corresponding output energy versus reflectivity of the output coupler for a cavity length of 1.25 m for Cr:LiSAF.

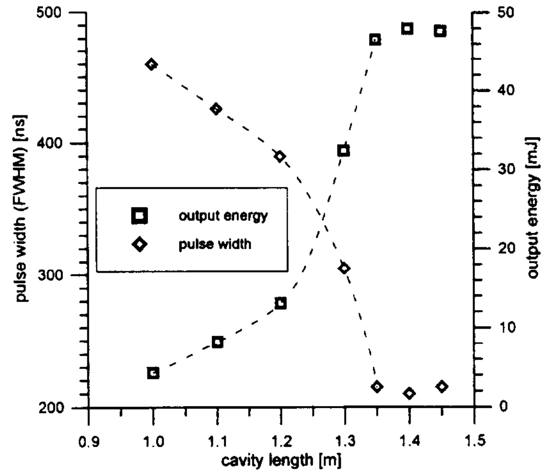

Fig. 4. Pulse width and corresponding output energy versus cavity length for a reflectivity of the output coupler of $R = 50\%$ for Cr:LiSAF.

4. Discussion

When a flashlamp-pumped laser is switched on, a train of distinct, gain-switched subpulses can be observed at the beginning of the laser output. These so-called relaxation oscillations are well known and are also called spiking. The reason for the spiking is that the population inversion under transient conditions exceeds the steady-state inversion since the laser oscillation which regulates the inversion by stimulated emission does not exist at the beginning. So, depositing a maximum of energy in the first spike, i.e. extensively exceeding the steady-state population, inversion can be achieved by strongly damping the buildup of laser oscillation, i.e. strongly damping the increasing of the regulating photon density. This is a process often called gain-switching. Looking at the rate equation system for a four-level laser system [10] and especially at the rate equation for the photon density ϕ

$$\frac{\mathrm{d}\phi}{\mathrm{d}t} = \phi c \frac{1}{L}\left(\sigma_e n l - 0.5\left[\ln(1/R) + \Gamma\right]\right) + k\frac{n}{\tau_f}$$

(c is the speed of light, n is the inversion population density, Γ combines diffraction losses, scattering losses, etc. and k is the quotient of the number of modes at the laser output and the number of resonant modes possible in the laser resonator volume) it becomes obvious that this depositing of maximal energy in the first spike can be achieved by manipulating the rod parameters (short length l of the laser rod, small cross section for stimulated emission σ_e, long fluorescence lifetime τ_f) and/or the cavity parameters (long cavity length L, high transmission of the output coupler, i.e. low reflectivity R). This could be confirmed by a computer simulation based on the rate equations for a four-level system.

Using the near-hemispherical resonator configuration described earlier and the Cr:LiSAF rod as gain material, we have more or less approached the above mentioned requirements for depositing a maximum of energy in the first pulse. Nevertheless, it seems to be unrealistic to attribute the obtained results with the Cr:LiSAF (very high energy in a single pulse) to gain-switching only. Repeating the same experiment with an identical near-hemispherical resonator while using a Nd-doped phosphate glass rod (having a very low stimulated emission cross section – $\sigma_{e,\ glass} = 4.3 \times 10^{-20}$ cm^2 – which is of the same order of magnitude as that one of Cr:LiSAF) of the same geometrical dimensions as the employed Cr:LiSAF crystal, one can see that the energies we measured (Fig. 5) are far below those ones achieved with the Cr:LiSAF, although the fluorescence lifetime of Nd:glass is much higher ($\tau_{f,\ glass} = 330$ μs). So, the suspicion emerges that there exists still another effect causing the observed phenomenon with the Cr:LiSAF crystal.

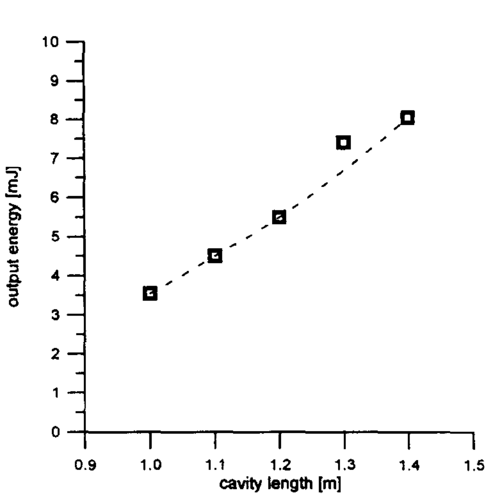

Fig. 5. Output energy versus cavity length for a reflectivity of the output coupler of $R = 50\%$ for Nd:glass.

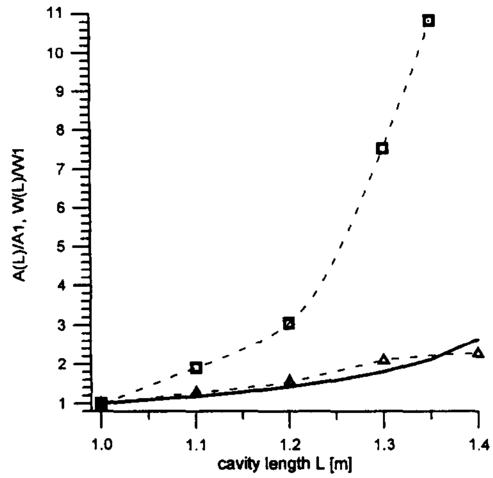

Fig. 6. Output energies $W(L)$ normalized to $W_1 = W(L = 1$ m) for Cr:LiSAF (squares) and Nd:glass (triangles) and cross sections of the laser rod transmitted by the beam $A(L)$ normalized to $A_1 = A(L = 1$ m) (solid line) versus cavity length.

This suspicion is corroborated when investigating the strong increasing of the output energy W with increasing resonator length L (see Fig. 4) which cannot only be explained by the growing transmitted cross section of the active volume; Fig. 6 shows the growth of the pulse energies with increasing resonator length L. Thereby the values of the energy $W(L)$ are normalized to the value $W_1 = W(L = 1$ m). For comparison, the corresponding beam cross sections at the laser rod $A(L)$ (which are nearly constant along the laser rod and which are therefore proportional to the transmitted active volume) are drawn in the same figure, also normalized to the value $A_1 = A(L = 1$ m). So, the strong differences of the gradients of these two graphs imply the assumption that there is still another effect (than the growing transmitted active volume) causing those high output energies and their extremely strong growth when the cavity length approaches the curvature of the HR-mirror. In addition, Fig. 6 contains the normalized values of the energies of Nd:glass which are in good accordance with the curve of the normalized beam cross sections.

The energies we measured (Fig. 3 and Fig. 4) in a single pulse with the Cr:LiSAF rod resemble very

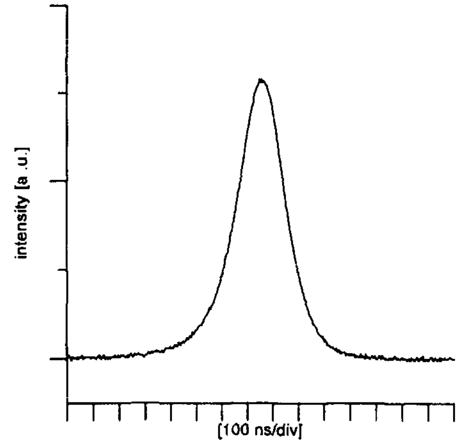

Fig. 7. Typical temporal profile of the Cr:LiSAF laser output pulse.

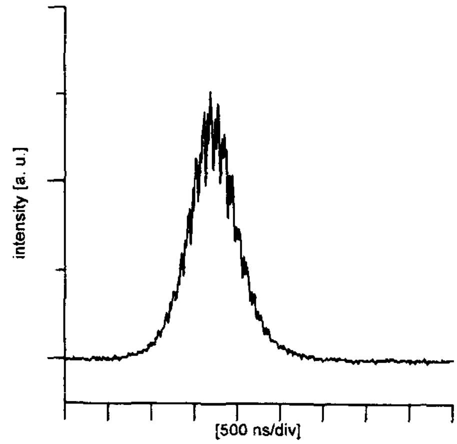

Fig. 8. Typical temporal profile of the Nd:glass laser output pulse.

much those ones which can be obtained when Q-switching a solid-state laser of comparable geometrical dimensions; an example provides Ref. [4]. Verifying the temporal development and the form of the output pulse (Fig. 7), similarities with a passively Q-switched laser can be stated: there is a relatively long rising edge with a small gradient at the beginning of the pulse which results when passively Q-switching a laser from a non-abrupt rising of the cavity quality factor Q as a consequence of an interaction between the photon density and the cavity quality factor. In addition, an increase of the ratio rise time/fall time with increasing resonator length (increasing output energy) could be observed while the pulse width is decreasing. This also stands in contrast to the results obtained with the Nd:glass rod when inreasing the resonator length: rise time, fall time and pulse width remained unchanged and the fall time is slightly longer than the rise time as this should be expected when only an increase of the transmitted cross section of the active volume (change in geometry) and not a supplementary switching effect is responsible for an increase of the pulse energy.

Comparing the temporal development of the output pulse of the Cr:LiSAF laser (Fig. 7) with that one of the Nd:glass laser (Fig. 8) it can be stated that the Nd:glass output is full of ripples due to beating between adjacent modes, while the Cr:LiSAF delivers a smooth output which is a hint for (if not one single so at least) one dominant axial mode [11] (we used a fast detector and a fast oscilloscope). This considerably enhanced mode selection is typical for a passive Q-switched laser [8] and therefore another hint for the self-Q-switching behavior of Cr:LiSAF.

Thinking about the mode of working of this observed switching mechanism, there are not too many possibilities. One of these possibilities is the assumption of particles in the crystal which work as saturable absorber. Therefore, we investigated Cr:LiSAF for absorption by determining the transmission, using the set-up as shown in Fig. 9. A diode-pumped cw $Nd:YVO_4$ laser provided a stable radiation of 150 mW at 1.064 μm. Comparing this output power with the power detected after passing the Cr:LiSAF rod twice provided a transmission of $T = 88.5\%$ which corresponds to a transmission of about $T = 94\%$ per transit. Remembering that the AR-coating covers the range between 750 nm and 950 nm, the losses probably arise mainly from mismatching of the AR-coating for 1.064 μm; the scattering losses of the crystal are lower than 0.3% per transit (product

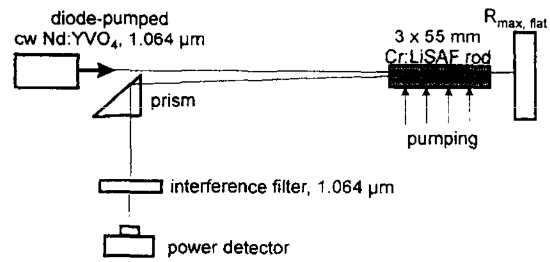

Fig. 9. Set-up for the determination of the transmission of the pumped and the unpumped Cr:LiSAF crystal.

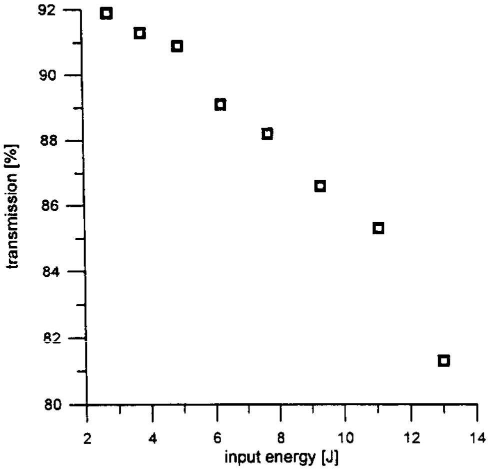

Fig. 10. Transmission of the pumped Cr:LiSAF versus the pump energy.

information of the manufacturer) and therefore negligible. This assumption of no absorption could be confirmed when an unpumped Cr:LiSAF rod was inserted in the hemispherical cavity described earlier; thereby, the pumped Cr:LiSAF rod was installed pretty close to the flat output coupler in order to generate only a small output pulse, while the unpumped rod found its place close to the HR-mirror, so that the transmitted area was large (in order to provide an interaction of the intracavity beam with a high number of the supposed absorbing particles), but still small enough that diffraction losses at the unpumped rod were still negligible: the energy of the laser output pulse was not affected by the supplementary unpumped Cr:LiSAF rod in this set-up which means that there does not exist any absorption from the ground-state.

Repeating the investigations of the transmission with the 1.064 μm radiation, thereby pumping the Cr:LiSAF rod, delivers the results of Fig. 10 where the transmission per rod transit versus the pump energy (electrical energy reaching the flashlamp) is shown. To avoid detection of the fluorescence light of the Cr:LiSAF rod, we inserted an interference filter between prism and power meter (Fig. 9).

The results show that an increasing of the pump energy of the Cr:LiSAF results in a growth of absorption by the crystal. Since the absorption occurs only when the crystal is pumped, this must be excited-state absorption (ESA). ESA of the laser radiation between the $^4T_2$-level and the $^4T_1$a-level has been reported and values determined [3,12]. But this transition cannot operate as saturable absorber: its saturation intensity is far above the intensity which is reached in our experiments as will be shown in the following.

The saturation intensity I_s for a saturable absorber is given by [10]

$$I_s = h\nu/\sigma\tau.$$

Here, $h\nu$ is the energy of a photon of the wavelength $\lambda = 830$ nm, σ is the cross section for the considered transition which is $\sigma_{ESA}(\lambda = 830\text{ nm}) = 2 \times 10^{-20}$ cm^2 [12], and τ is the effective lifetime of the upper excited level which is here the $^4T_1$a-level. This lifetime is assumed to be not longer than 1 ns [13] (Ref. [14] reports of rapid relaxations). This leads to a saturation intensity of at least 1.2×10^{14} W m^{-2}. Taking the lower value of $\sigma_{ESA}(\lambda = 830\text{ nm}) = 1.3 \times 10^{-20}$ cm^2 reported in Ref. [3] an even higher saturation intensity is obtained.

Assuming an output pulse with a nearly gaussian profile (concerning the temporal behavior, Fig. 6), the maximum output power we obtained can be valued at 3.3×10^5 W ($W_{out} = 60$ mJ, $\Delta t_{FWHM} = 180$ ns). Thus, the maximum intensity in the laser rod can be calculated to about 2×10^{11} W m^{-2}, assuming a reflectivity of the output coupler of 40%, and a relation of the output power P_{out} to the power within the cavity P_{in} which is given by [10]

$$P_{out} = \frac{1-R}{\sqrt{R}} P_{in},$$

and an intensity distribution which has a gaussian profile over the laser rod cross section which is completely utilized.

Comparing the calculated saturation intensity for the transition between the $^4T_2$-level and the $^4T_1$a-level with the maximum intensity obtained in our experiments, it becomes obvious that this transition cannot operate as saturable absorber and can therefore not be made responsible for the assumed Q-switching behavior.

So, one can think of another transition operating as saturable absorber, for example a transition in Cr^{4+}. LiSAF is doped with Cr^{3+} but it can not be excluded that Cr^{4+} is generated when growing the crystal [15]. Charge transfers from an excited state could deliver the high cross section necessary for a saturation effect at intensities which are observed in our experiments. On the other hand, it can also not be excluded that oxygen is included in the growing process of the Cr:LiSAF crystal [16] responsible for the generation of color centers [17] which could work as saturable absorber. But, at last, there is no proved existence of such absorbing particles which could be saturated by the measured intensities.

So, at the moment, we favor another effect probably causing the switching mechanism in Cr:LiSAF. A Ker nonlinearity was reported in Cr:LiSAF [18]. This nonlinearity has already been used for mode locking [18,19]. We believe that the Kerr effect gives rise to a passive Q-switching behavior in our resonator configuration.

Generally spoken, on the one hand, the Kerr effect causes an induced lens (dependent on the intensity of the transmitting beam) which results from wavefront distortion inflicted on the beam by itself while traversing a nonlinear medium [20], for example Cr:LiSAF. On the other hand, Q-switching means an increasing of the cavity quality factor Q being identical with a lowering of the cavity losses after some energy has been stored in the gain medium. In our resonator configuration, the switching mechanism becomes conspicuous (the energies considerably exceed those obtained with the Nd:glass rod for example) when the resonator length approaches the radius of curvature of the HR-mirror, i.e. the resonator approaches instability which is identical to an increase of diffraction losses. The necessarily higher pump energies cause an increase of the negative thermal lens of the Cr:LiSAF crystal [9] still intensifying the losses. But as soon as the laser oscillation starts, the growing radiation intensity induces a (positive) Kerr lens thereby lowering the diffraction losses (by more or less compensating the negative thermal lens). This means an increase of the cavity quality factor Q leading to a further intensity growing, intensifying the Kerr lens effect etc.

An experiment which might confirm this idea will be given in the following. We used the set-up shown in Fig. 1 choosing the cavity length to $L = 1.1$ m. Inserting a diaphragm inbetween the Cr:LiSAF rod and the HR-mirror we determined the output energy of a single pulse and its width for different diameters of the diaphragm. The results are shown in Fig. 11.

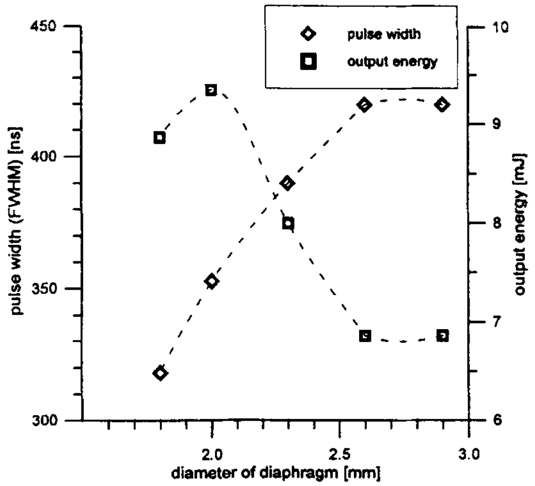

Fig. 11. Pulse width and corresponding output energy of the Cr:LiSAF laser versus the diameter of the inserted diaphragm.

For the present, it might be amazing that the output energies are growing when the diameter of the diaphragm and therefore the cross section of the transmitted area of the Cr:LiSAF crystal is reduced. But considering the Kerr lens effect as described above this phenomenon becomes clear: By inserting a diaphragm with small diameter, the diffraction losses and therefore also the negative thermal lens (induced by the pump energy) are increased. A higher amount of pump energy can be stored in the gain medium. With beginning oscillation and growing intensity the positive Kerr lens will be induced compensating to a certain amount the negative thermal lens and the diffraction losses, thereby increasing the cavity quality factor Q. A shorter getting pulse width with reduced aperture is another hint for a stronger switching mechanism. Generally, the meaning of the Kerr lens in the Cr:LiSAF crystal can be shown when working again with the cavity as described in Fig. 1 and with a cavity length of about 1.35 m (high radiation intensity). Inserting an additional unpumped Cr:LiSAF rod (4×55 mm) pretty close to the output coupler means a completely different output pulse concerning the temporal width and the measured energy which can be obtained in a single pulse although the unpumped rod does not show any absorption losses or diffraction losses (the latter could be confirmed by a diaphragm of slightly smaller diameter than the unpumped rod which substituted in another experiment the rod). Using this configuration, the high intensity at the output coupler induces a strong Kerr lens in the unpumped rod changing the stability behavior of the resonator. On the other hand, in the experiment for measuring the absorption (at $\lambda = 830$ nm) of the unpumped crystal as described earlier, the unpumped rod did not manipulate the output pulse: there, we had low output energies and the intensity at the unpumped rod was very small since it was installed next to the output coupler where the beam diameter has its maximum. So, a Kerr effect was not detectable.

In summary, it can be said that the observed self-Q-switching seems to be caused by a Kerr lens Q-switching. So to our knowledge it would be for the first time that the Kerr nonlinearity is utilized for passive Q-switching. We are still doing detailed research of this problem.

5. Summary

We reported of a Cr:LiSAF oscillator delivering pulses with several tens of millijoules of output energy and pulse widths of several hundreds of nanoseconds without any form of external Q-switching. We studied the possibility of a self-Q-switching effect of the crystal and provided hints confirming this assumption.

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