Optical and Quantum Electronics **21** *(1989) 55–61*

Pulse shortening of 5 × 10^3 by the combined pulse forming of dye oscillators, saturated amplifiers and gated saturable absorbers

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Received 14 April; accepted 18 May 1988

A simple excimer-laser-pumped cascade dye laser-amplifier system has been presented which due to the combined pulse-forming effects of cascade dye lasers, saturated amplifiers and gated saturable absorbers, is capable of generating single 3 ps pulses.

There is increasing interest in excimer-laser-pumped short-pulse dye lasers. These lasers can have broad tunability, and they are in automatic synchronization with the following amplifier chain [1, 2]. However, it was shown [1] that the shortest pulse duration achievable with these lasers is roughly an order of magnitude longer than that of mode-locked continuous wave (c.w.) lasers [3]. Still, there are numerous applications [4, 5] where the use of excimer-laser-pumped short-pulse dye lasers may be preferred.

In [6] a cascade dye laser set-up was reported which consists of a quenched dye laser (QDL), a short-cavity dye laser (SCDL) and subsequent amplifier and gated saturable absorber (GSA) stages (for details, see [6] and references therein). In [1, 2, 4, 5] this laser is part of a system designed to provide single, picosecond, ultraviolet pump pulses for pumping a subpicosecond distributed-feedback dye laser (DFDL).

In this paper we present a simplified version of that set-up while preserving the original output characteristics. It is also shown that not all of the processes which were considered in [6] are essential for pulse shortening. This realization led to further simplification of the set-up.

The experimental arrangement is shown in Fig. 1. One channel of a Lambda Physik EMG 150 excimer laser is used for pumping, delivering 80-mJ, 15-ns pulses at 308 nm, when filled with a partial pressure of 4.5 mbar HCl, 70 mbar Xe, and He to a total pressure of 2.5 bar. A quartz plate acts as a beamsplitter, which couples out about 8 mJ of pump energy. One-third of the out-coupled beam is used to pump a dye laser, where the resonator is formed by the two planparallel surfaces of a 5-mm thick Hellma dye laser cell. When this laser is used as a quenched laser, the second quenching resonator is formed by inserting an additional slightly tilted mirror – indicated by broken line in Fig. 1 – about 2 cm behind the dye cell, as in [7]. In this case, by proper focusing of the pump beam, the laser is set to

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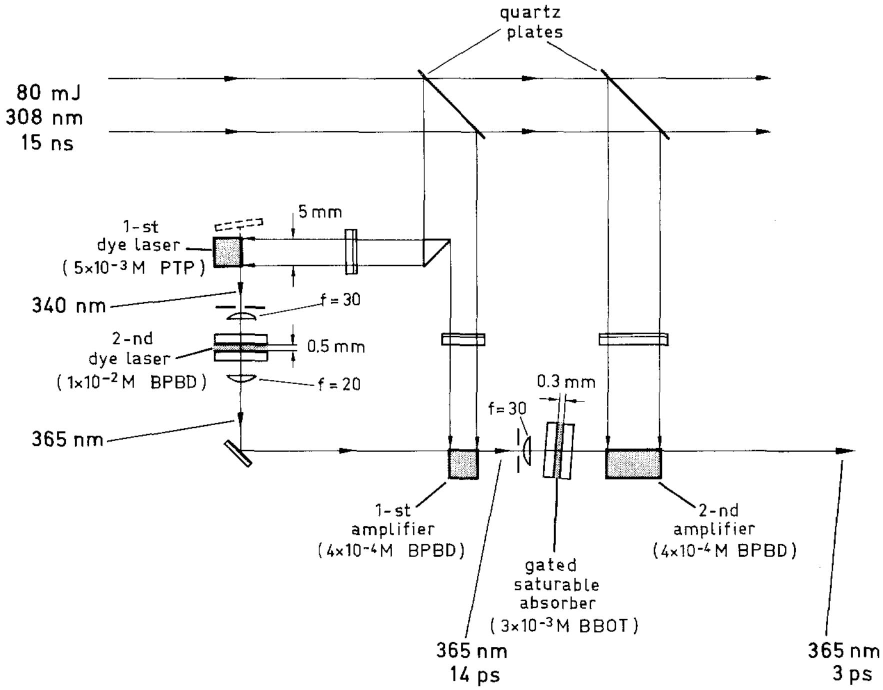


Figure 1 Experimental arrangement.

operate approximately three to four times above its threshold. Paraterphenyl (5×10^{-3} M) in cyclohexane is used as an active medium yielding a 1-nm broad emission centred at 340 nm. The rise time of the pulse at this point is about 100 ps, with a trailing component of several nanoseconds. When the laser is quenched, the trailing component is partially suppressed (see below).

The output of the first dye laser, after spatial filtering and focusing by an $f = 30$ mm lens, is used for longitudinal pumping of a second dye laser having a cavity length of 0.5 mm. (The focus of the pump beam is set to be just behind the cavity.) This cavity is formed by an output mirror of 80% reflectivity and a dichroic back mirror, which has 100% reflectivity at the maximum of lasing (365 nm), and 60% transmission for the pump wavelength. In order to avoid parasitic oscillations from these surfaces, the cavity is slightly tilted with respect to the pump beam. The cavity contains a solution of 1×10^{-2} M butyl-PBD 2-(4-biphenylyl)-5-(4-t-butylphenyl)-1,3,4-oxadiazol in methanol. The second dye laser is pumped approximately eight times above threshold by proper adjustment of the $f = 30$ mm focusing lens. Its output is collimated by an $f = 20$ mm lens to produce a parallel beam of diameter about 1.5 mm. It is then amplified in an amplifier which is pumped by the rest of the beam out-coupled by the quartz plate. The amplifier uses a 4×10^{-4} M solution of butyl-PBD in cyclohexane. The amplified pulse has an energy of 12 μJ and a pulsewidth of 14 ps. The pulse duration measurements were done with a Hamamatsu C1587 streak-camera, having a temporal resolution of about 2 ps. The typical pulse form of the amplified pulse is shown in Fig. 2a, exhibiting a pronounced initial overshoot with a sharp rise time, and some weak trailing component. (For the evaluation of the pulse width of these pulses, only the width of the initial overshoot is taken into account and the trailing component is

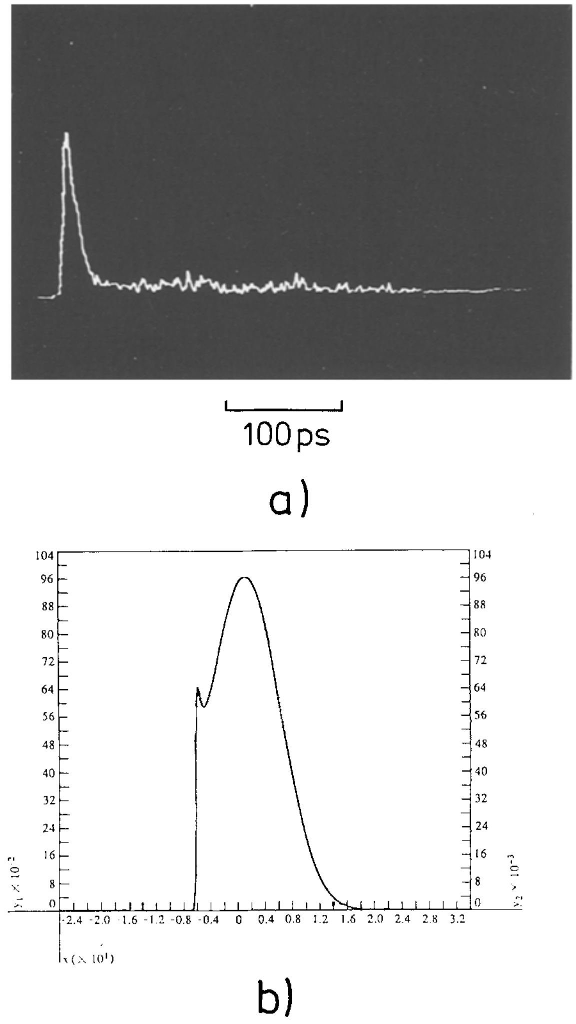


Figure 2 (a) Typical pulse form of the amplified pulse of the cascade dye laser. (b) Computed temporal evolution of a dye laser output when pumped high above threshold (from [8]). $\alpha = 250$, $\beta = 3.0$, $\gamma = 6.0$.

neglected.) The large amplitude of the initial overshoot compared with that of the trailing component is remarkable, which seems to contradict the theoretically predicted temporal behaviour of dye lasers pumped well above threshold [8]. According to the theory, the response of a dye laser on a strong pump pulse is a pulse consisting of an initial overshoot with a sharp rise time and a trailing component which roughly follows the pump pulse and has an amplitude comparable with that of the initial overshoot. This behaviour is clearly seen in Fig. 2b (taken from [8]). The reason for the difference between our experimental pulse form in Fig. 2a and the computed one in Fig. 2b is the pulse-forming effect of our saturated amplifier, which amplifies more the initial overshoot and less the tail. In our experimental configuration (Fig. 1) pumping of the first dye laser, and the pulse generated by the cascade dye laser, is delayed so that the initial overshoot to be amplified sees maximum gain at the amplifier. The input energy density is chosen to be sufficiently high for the initial overshoot to saturate the amplifier, resulting in much less gain for the residual part of the pulse.

The streak-camera record in Fig. 2a was made when the quenching mirror at the first dye laser was in place. By removing this mirror, both the characteristics of the initial overshoot and the relative amplitude of the trailing component remained practically the same, but the duration of the latter increased from about 200 ps to several nanoseconds.

Since the trailing component can be completely removed as is shown below, the parameters of the final pulse are not influenced by quenching of the first dye laser. This means that only the rise time of the initial overshoot of each laser is important for the temporal behaviour of the output.

It is known from earlier investigations [8] that the rise time of the output of a dye laser pumped high above threshold is determined mainly by the rise time of the pump pulse and by the length and the Q of the resonator, and is only slightly dependent on the pump level. Even with an optimized resonator, the rise time of the initial overshoot cannot be more than about 100 times shorter than that of the pump pulse. Under our experimental conditions the use of two dye lasers in cascade was necessary to decrease the risetime of about 4 ns of the excimer pump laser to the 10-ps level.

The output pulse of the first amplifier is then used as input pulse for the following gated absorber–amplifier stage. After spatial filtering the beam is focused by an $f = 30$ mm lens just beyond the gated saturable absorber. The transmitted beam is then amplified in the following amplifier, resulting in a divergent output beam. (This divergent beam is tailored to the pump requirements of our new DFDL to be pumped by this output can be collimated by an appropriate lens.)

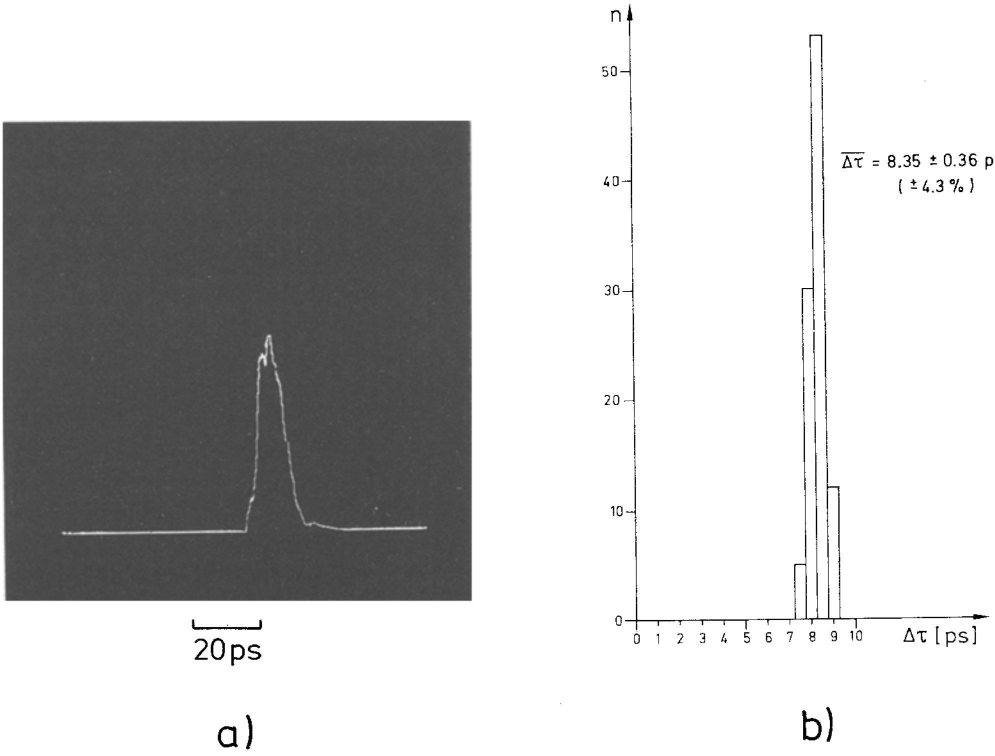


Figure 3 (a) Typical pulse form and (b) pulsewidth distribution of the output pulses for 1 mm mirror separation in the GSA. $\overline{\Delta\tau} = 8.35 \pm 0.36$ ps (±4.3%).

The principle of GSAs was introduced by Yasa [9] and discussed by Christov and Tomov [10]. This makes use of a saturable laser dye, surrounded by a tilted dichroic resonator, where non-linear absorption and stimulated emission from the dye solution determine a time window where the arrangement is transparent. It is also shown in [6] that the time window, which is mainly determined by the separation of the dichroic mirrors of the GSA, must be tailored to the input pulse; it has an optimum at half to one-third of the input pulse duration (for details, see [6, 9, 10]).

Our later investigations showed that it is not the pulse duration, but rather the rise time of the input pulse, which counts in determining the optimum time window for the GSA. This means that pulse-shortening of GSAs is limited for symmetrical pulses and can be more pronounced for asymmetric pulses having short rise time, as in our case (see Fig. 2a).

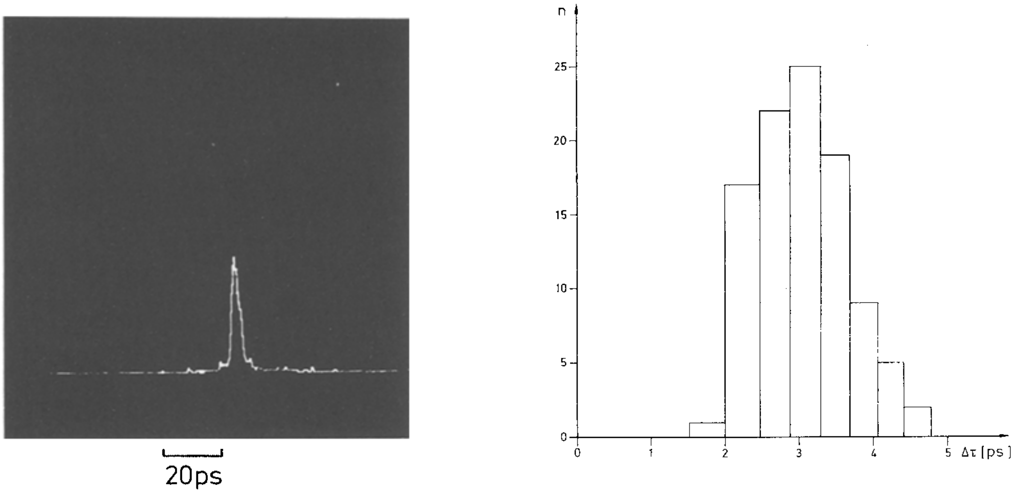
In this case the duration of the transmitted pulse can be roughly one-third of the rise time of the input, which means even higher pulse-shortening referring to the input pulse duration. The trailing component is completely suppressed.

In our experiment the gated saturable absorber consists of a solution of BBOT 2,5-di-(5-tert-butyl-2-benzoxazolyl)-thiophene of proper concentration, and a tilted cavity formed by two identical dichroic mirrors having approximately 10% reflectivity at 365 nm and 100% reflectivity at 430 nm, acting at the same time as dye-cell windows. The small-signal transmission of the absorber is adjusted to be approximately 10^{-5}. Two configurations were tested experimentally; when 1×10^{-3} M BBOT in a cavity 1 mm long and when 3×10^{-3} M of the same dye in a cavity 0.3 mm long was used as a GSA. In each case the GSA was followed by a 4-cm long dye amplifier, containing a 4×10^{-4} M solution of butyl-PBD in cyclohexane. The amplifier was pumped by a pump pulse of about 7 mJ out-coupled by a quartz plate. The temporal behaviour and the pulsewidth distribution of 100 successive output pulses (after amplification) is shown in Figs 3 and 4, for cavity lengths of 1 and 0.3 mm, respectively. It is seen that using 1 mm separation the duration of the output pulse is relatively long (about 8 ps), but the pulsewidth stability is excellent (about 4.5%). With decreased cavity length the pulse duration can be shortened to about 3 ps, but at the expense of the increase of the pulsewidth fluctuation (19%).

This figure agrees with our earlier observation concerning the optimum time window of GSAs. If the time window is too long, then no significant shortening can be achieved; if it is too small compared with the input pulse duration, then the transmitted energy and the stability of the transmitted pulse decrease [6]. In [6] 8.2-ps (3.0-ps) pulse duration with 5% (13%) pulsewidth fluctuation is achieved, when a cavity length of 1 mm (0.3 mm) is used in the GSA. Compare these results with Figs 3 and 4. Although the corresponding pulse durations are practically the same (they seem to be determined only by the separation of the cavity of the GSA) the pulsewidth fluctuations are different, in agreement with the different pulse-shortening factors of the GSAs in the two experiments.

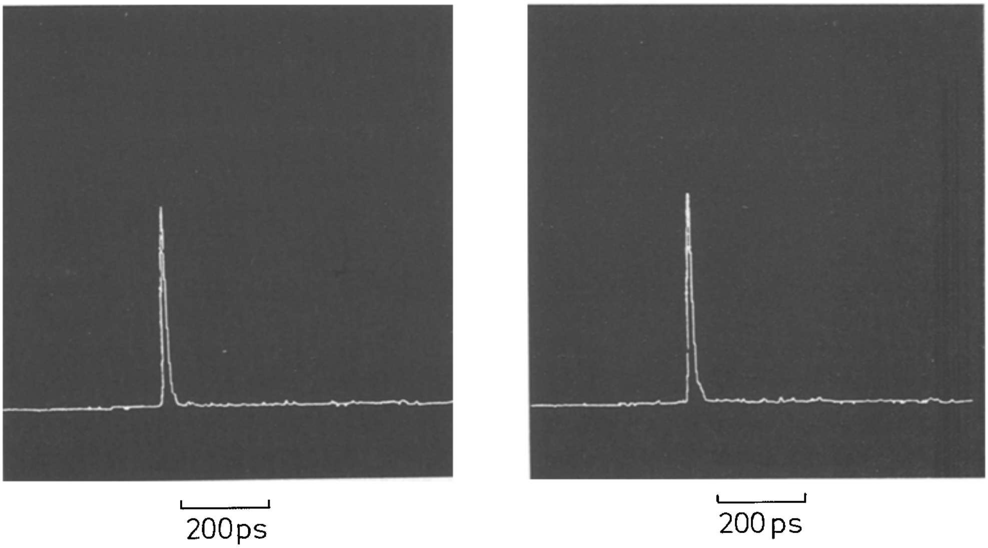
We also found that using this technique no significantly shorter pulses can be generated than these presented here. On an even shorter timescale not only did the geometrical delay of the resonator play a role in the build-up time of lasing, but also the excited-state relaxation time, making the switching process slower and less pronounced.

Finally, we checked the eventual effect of quenching of the first dye laser on the temporal behaviour of the output. We found no difference between when the quenching mirror was in place (Fig. 5a) or removed (Fig. 5b). In all cases the background level was < 1%. In later experiments, when this set-up was used as a part of a hybrid excimer–dye laser system [11, 12], the quenching mirror was generally used, its purpose being not to form the pulse



a) b)

Figure 4 (a) Typical pulse form and (b) pulsewidth distribution for 0.3 mm mirror separation. $\overline{\Delta\tau} = 3.06 \pm 0.59$ ps ($\pm 19\%$).



a) b)

Figure 5 Temporal behaviour of the output (a) with and (b) without quenching the first dye laser.

shape, as shown here, but to remove the unwanted, several-nanosecond-long tail of the pulse, already at the first stage, and then to avoid eventual damage of the following stages related to the high energy density. We would emphasize that for this purpose no exact positioning of the quenching resonator is necessary, the quenching mirror can be placed several centimetres from the dye laser, not affecting the first sub-nanosecond operation, but cutting the following, several-nanosecond-long, trailing component.

This means that, by proper choice of the cavity length of the dye lasers and the GSA, and by driving the amplifiers into saturation, the desired single, picosecond output can be reliably achieved even without having to make any temporal measurement at the various stages for proper adjustment and timing.

It is noteworthy that $< 20\%$ of the total pump energy is used for the above arrangement; the remainder may be utilized for further amplification [12].

In conclusion, a simple excimer-laser-pumped cascade dye laser–amplfier system has been presented which, due to the combined pulse-forming effects of cascade dye lasers, saturated amplifiers and gated saturable absorbers, is capable of generating single 3-ps pulses.

It is shown that the output pulse duration, within certain limits, is determined only by geometrical parameters, which makes the construction and operation of this laser easy, even without the help of temporal measurements.

Acknowledgements

The author wishes to thank Professor F. P. Schäfer and Dr N. Ernsting for stimulating discussions and critical reading of the manuscript, and Dr G. Szabó for his help in making the streak-camera measurements. This work was supported by the Bundesministerium für Forschung und Technologie and by the OTKA Foundation of the Hungarian Academy of Sciences.

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