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Flashlamp-pumped mode-locked dye lasers

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Recent developments in the production of tunable picosecond pulses by means of flashlamp-pumped mode-locked dye lasers are reviewed. Particular attention is paid to an extension of the passive dye laser mode-locking technique based on the loss modulation of the gain of a secondary oscillator by means of the pulse train of a master oscillator. The resulting two trains of independently tunable picosecond pulses can be used to monitor a transient species by an excite–probe experiment with one shot only.

1. Introduction

Flashlamp-pumped passively mode-locked dye lasers were the first low-priced source of frequency tunable picosecond pulses. Providing pulses of typically 3–10 ps duration with a single pulse energy between 10 and 80 μJ these lasers allowed experiments studying nonlinear interactions in matter as well as the kinetics of primary photophysical and photochemical reactions to be carried out. Frequency tunability has been extended towards shorter wavelength ($\lambda \approx 460$ nm) in recent years, thus opening up new classes of compounds to be investigated. Unfortunately, the beam divergence of flashlamp-pumped mode-locked dye lasers is comparatively high. Extension of the tuning range into other spectral regions by means of nonlinear processes (e.g. parametric oscillators or sum frequency generation) with a sufficiently large conversion efficiency is therefore very difficult.

The ratio between peak power (*ca.* 25 MW) and inter-pulse background, on the other hand, is high (*ca.* 10^4) and pulse quality is good for a considerable number of pulses in the middle and later portion of the train, when the build-up of mode-locking is completed. Because of these properties, mode-locked flashlamp-pumped dye lasers may be able to maintain their position as tunable light sources for time-resolved spectroscopic investigations on a 10 ps – 1 ns time scale.

2. Active mode-locking of flashlamp-pumped dye lasers

Active mode-locking of a flashlamp-pumped dye laser by direct modulation of the resonator loss was first demonstrated by means of an opto-acoustic modulator in the resonator (Ferrar 1969). By using 7-diethylamino-4-methylcoumarin, mode-locked pulses with a width of several hundred picoseconds were generated at $\lambda \approx 460$ nm, the shortest pulses being found at the end of the train of a few microseconds duration. Based upon this experimental result as well as on theoretical considerations (Siegman & Kuizenga 1974) it was assumed that active mode-locking could provide short pulses ($t_p \lesssim 50$ ps) only some 5 μs after the start of laser action.

Just recently, Jasny *et al.* (1978) succeeded in applying a novel type of interferometer (Jasny 1978; see figure 1) to tune and actively mode-lock a dye laser in a very simple and convenient

way. One mirror of the resonator is replaced by this interferometer in which the optical path-lengths in the two arms are changed rapidly by the rotation of a quartz block (figure 2). At a given rotation rate, the condition of maximum reflexion from the interferometer end is fulfilled for a short time and for a certain wavelength only. Therefore, a pulse travelling back and forth within the resonator can develop only at that wavelength for which the fringe count

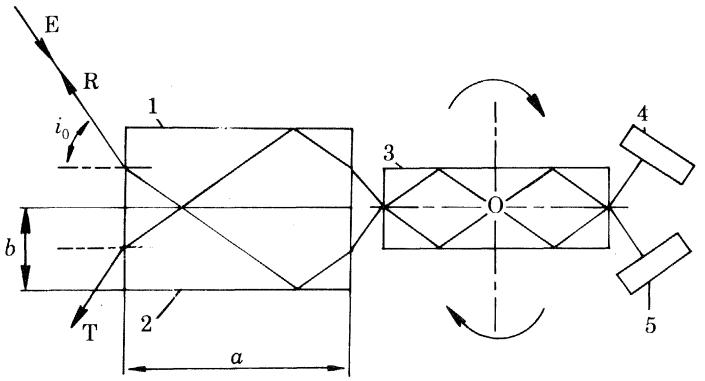

FIGURE 1. The optical system of the interferometer (1, 2, 3, quartz rectangular parallelepipeds; 4, 5, coated mirrors). The blocks 1 and 2 are separated by an air gap of 108 nm thickness; block 3 rotates around the centre O (After Jasny *et al.* (1978).)

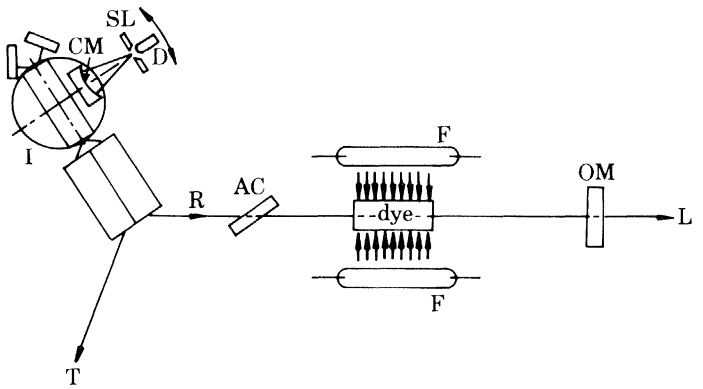

FIGURE 2. Dye laser with interferometer (I, interferometer; CM, concave mirror; SL, slit; D, diode; AC, absorber cell; R, reflected beam; T, transmitted beam). (After Jasny *et al.* (1978).)

period, T_e, is equal to the resonator round-trip time T_r. For small angles of rotation, $\phi < \phi_m$ (with respect to the orientation given in figure 1), the modulation of the reflected and transmitted beam, respectively, is sinusoidal (with a modulation depth of 100% at 580 nm), the modulation period T_e being an almost linear function of the mechanical rotation period T_m:

$$T_e = K(\phi, \lambda)\, T_m, \quad \text{with} \quad \begin{cases} \Delta K/K \leqslant 10^{-5}, & \text{if} \quad \phi \leqslant 0.5^\circ \\ \Delta K/K \leqslant 10^{-3}, & \text{if} \quad \phi \leqslant 5^\circ. \end{cases}$$

Since the factor $K(\phi, \lambda)$ is of the order of 10^{-7}, some 10^4 modulation cycles are completed within the time (*ca.* 50 μs) that is necessary to rotate block 3 in figure 2 by about 3° ($\Delta K/K \approx 10^{-4}$) at 10^4 rev/min. Synchronization between the flashes of the four pumping lamps and the 'zero' position of the quartz block 3 was easily achieved. A photodiode was illuminated by an incandescent lamp situated at the lower part of a stationary slit via a small mirror (CM in figure 2) which is mounted together with the rotating quartz block.

Changing the rotation period T_m between 8.8 ms and 9.3 ms respectively resulted in an output wavelength shift from about 610 to 585 nm, when a 2×10^{-4} M solution of Rhodamine 6G in ethanol was used as lasing material. The spectral bandwidth (time-integrated) was determined to be 0.1 nm without and 0.05 nm with an additional absorber cell AC (10^{-5} M DODCI in ethanol) in the resonator.

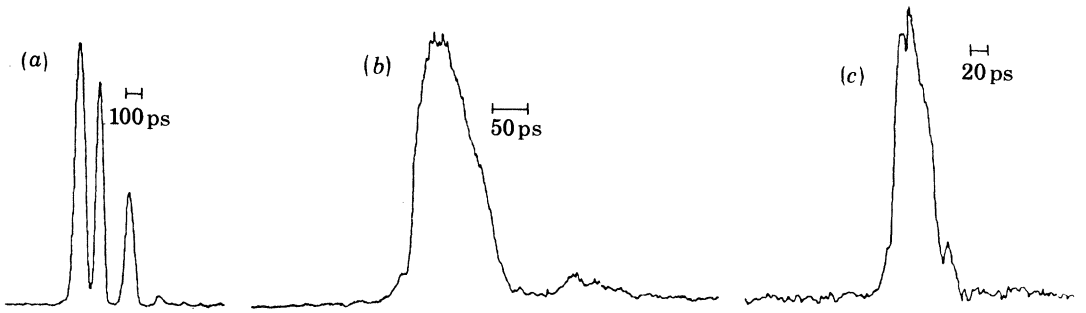

FIGURE 3. Streak camera record of a pulse in the middle of the pulse train (*a*) without absorber cell; (*b*) with absorber cell in short resonator (66.5 cm); (*c*) with absorber cell in long resonator (85.7 cm). (From Jasny *et al.* (1978).)

Without the additional absorber cell AC, short pulses are generated only if the dye is pumped close to threshold, whereas high above threshold the pulses broaden to over 1 ns and show a substructure. With the absorber cell placed in the optical centre of a resonator of increased length (85.7 cm), pulses of typically 50 ps f.w.h.m. duration could be generated (see figure 3).

3. PASSIVE MODE-LOCKING OF A FLASHLAMP-PUMPED DYE LASER

Self-mode-locking of flashlamp-pumped dye lasers was first achieved by Schmidt & Schäfer (1968), using Rhodamine 6G as the laser active medium and the cyanine dye DODCI(3, 3′-diethyloxadicarbocyanine iodide) as the saturable absorber. Bradley & O'Neill (1969) reproduced this result with both Rhodamine 6G and Rhodamine B. By 1972, picosecond pulse generation with interferometric tuning was shown to be possible in the wavelength region $580 < \lambda < 700$ nm by using various combinations of three laser and four absorber dyes (Arthurs *et al.* 1972). At that time it was also established by means of ultra-fast streak camera measurements that in those mode-locked dye lasers that have the saturable absorber cell next to the 100% reflexion mirror (see figure 4), well isolated pulses of less than 6 ps duration were generated. The ratio of peak pulse power to that of the background intensity between the pulses was found to be larger than 10^4 (for a review see Bradley 1977).

By 1972 a large number of laser dyes, e.g. the coumarin and xanthene dyes, were known to cover the spectral range $440 < \lambda < 650$ nm when used in a flashlamp-pumped system (for a review see Drexhage 1973). Nevertheless, it was not until 1977 that mode-locking at wavelengths shorter than 580 nm was reported by Bradley (1977) and Lill *et al.* (1977*a*). According to Bradley, mode-locking of an esculin monohydrate laser was possible in the range $465 < \lambda < 480$ nm with 2-(*p*-dimethylaminostyryl)-pyridylethyl iodide (DASPI) as saturable absorber. With a dye of unknown structure named Fluorol 7 GA, passive mode-locking could be achieved in the spectral range $550 < \lambda < 580$ nm, when an ethanolic solution

of DQOCI (1,3′-diethyl-4,2′-quinolyloxadicarbocyanine iodide) was used as saturable absorber, as well as a Fabry–Perot etalon inserted into the resonator (Lill *et al.* 1977*a*). Unfortunately, the buildup of the pulse train is slow, and short pulses ($t_p \lesssim 5$ ps) can be expected only some 500 ns after the initial laser action.

In 1978 complete mode-locking of a flashlamp-pumped laser working with different coumarin dyes was reported by Mialocq & Goujon (1978*a*, *b*). Pulses of typically 10 ps duration were generated at $\lambda \approx 520$ nm with the use of an ethanolic solution of a mixture of 2.5×10^{-4} M coumarin 314 and 10^{-5} M coumarin 6 in combination with a glycerol–water (87% by volume) solution of PIC (1,1′-diethyl-2,2′-quinolyl cyanine iodide) as saturable absorber (Mialocq & Goujon 1978*a*). Pulses shorter than 10 ps in the wavelength region $475 < \lambda < 490$ nm could be generated best by mode-locking coumarin 102 (2.2×10^{-4} M in methanol) with a solution of DOCI (3,3′-dihexyloxacarbocyanine iodide) in an ethylene glycol–methanol (90% by volume) mixture (Mialocq & Goujon 1978*b*). The excited state lifetimes (fluorescence decay times) of the two saturable absorber dyes were found to be 143 ± 20 ps (PIC) and 370 ± 10 ps (DOCI) in the stated solvents. These comparatively long lifetimes do not only confirm the earlier findings that pulse duration in mode-locked dye lasers is independent of the saturable absorber lifetime (see, for example, Bradley & New 1974) but also indicate that larger aperture times may help to start the mode-locking process when dyes with low gain are used as active medium. The drawback of highly viscous saturable absorber solutions, in which long aperture times are observed (see §5), is of course the necessity of fast replacement because of irreversible bleaching effects of the illuminated sample.

Rhodamine 6G and Rhodamine B dye lasers were also mode-locked by using pinacyanol chloride (1,1′-diethyl-2,2′-quinolylcarbocyanine chloride) in various solvents as saturable absorber (Mialocq & Goujon 1977*b*). Tuned mode-locking could be achieved not only with high viscosity but also with low viscosity absorber solutions (methanol, propan-2-ol, DMSO) for which the recovery time is less than 10 ps (Mialocq *et al.* 1977*a*). Untuned mode-locking, however, was achieved in high viscosity solvents only, for which the ground-state recovery time is in the order of 330 ps (glycerol). Similar observations were made in our group when mode-locking of Fluorol 7G A and coumarin 6 was attempted.

4. Mode-locking of a flashlamp-pumped dye laser by loss modulation of the saturable absorber

The nature of the initial build-up phase of mode-locking in a flashlamp-pumped dye laser was elucidated by detailed experimental investigations making use of ultra-fast streak cameras (Arthurs *et al.* 1973; for a review see Bradley 1977). In certain cases, like Rhodamine 6G mode-locked by DODCI at $\lambda = 605$ nm, a very fast build-up occurs. Within a few round trips from the start of the laser action, a fluctuation noise burst of about 100 ps duration exists in the resonator cavity. After some additional 25 round trips the noise burst is substantially compressed, and within another 10 round trips single pulses of *ca.* 2 ps duration are circulating within the resonator. In other cases, e.g. at greater wavelengths or with different combinations of active and passive medium, pulse build-up may take longer, the essential parameter being the ratio

$$\frac{\sigma_p}{\sigma_a}\frac{\tau_a}{\tau_p},$$

with σ and τ denoting the absorption cross sections and relaxation times, respectively, of passive and active media (New & Rea 1976).

A considerable shortening of the build-up phase and/or an extension of the tuning range for a given combination of active and passive media can be accomplished if the saturable absorber of a second laser pumped just below threshold is bleached repetitively by means of a train of picosecond pulses originating from a primary, passively mode-locked laser (see figure 4).

If laser II is operated without a dispersive elements, the spectral distribution of its emission depends on the wavelength of the bleaching pulses, as can be seen from the time-integrated spectra displayed in figure 5. In contrast to solid state lasers, where Opower & Kaiser (1966)

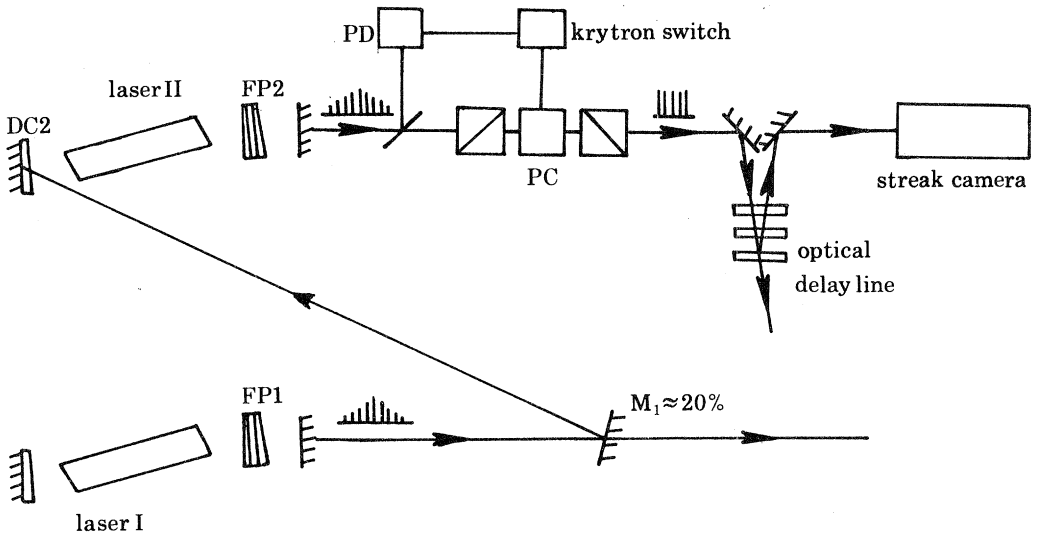

FIGURE 4. Experimental arrangement for the generation of two trains of time-correlated picosecond pulses. The passively mode-locked laser I modulates the loss in the resonator of laser II by repetitive bleaching of the saturable absorber in DC 2. (From Lill *et al.* (1977*c*).)

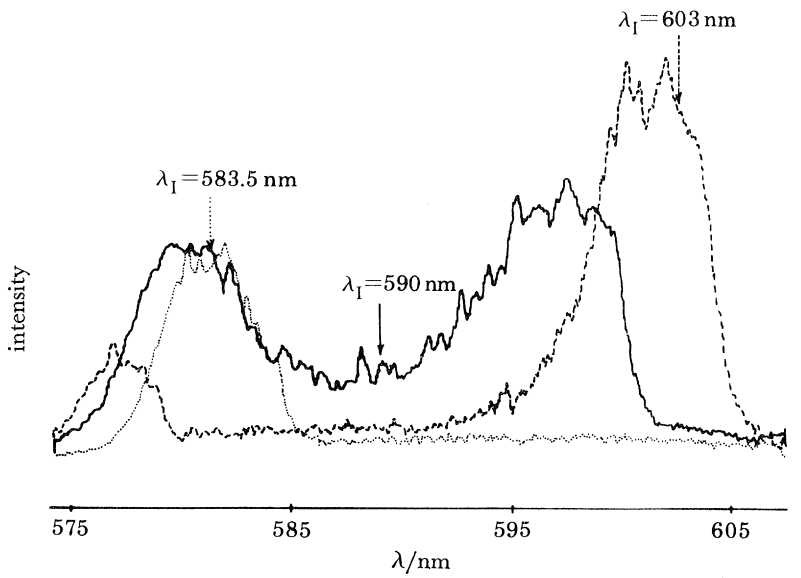

FIGURE 5. Spectral distribution of the output of laser II (Rhodamine 6G/DODCI) as a function of the wavelength of the bleaching pulses. Pump energy of laser II is 0.5 % under threshold. (From Lill *et al.* (1977*b*).)

found that the slave oscillators became frequency-locked to the master oscillator, the situation is rather more complex here because of the large gain–bandwidth product that the dye solution can provide. The laser will start to oscillate with its frequency distribution centred at the bleaching wavelength, then slowly broaden the distribution according to the net gain of the system (see figure 6), which depends both on the losses (saturable absorber) as well as on the strength of pumping of the active medium, as can be seen by inspection of figure 7.

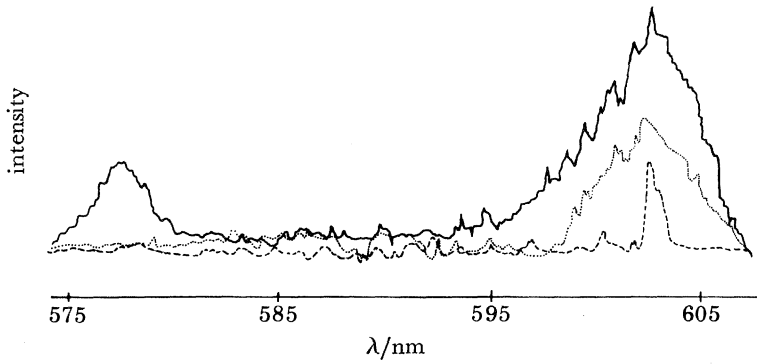

FIGURE 6. Evolution of the spectral distribution of the output of laser II (Rhodamine 6G/DODCI), when pumped 0.5 % below threshold. Spectra are taken 40 ns (– – –), 80 ns (· · ·) and 120 ns (—)after the start of oscillation. (From Lill (1978).)

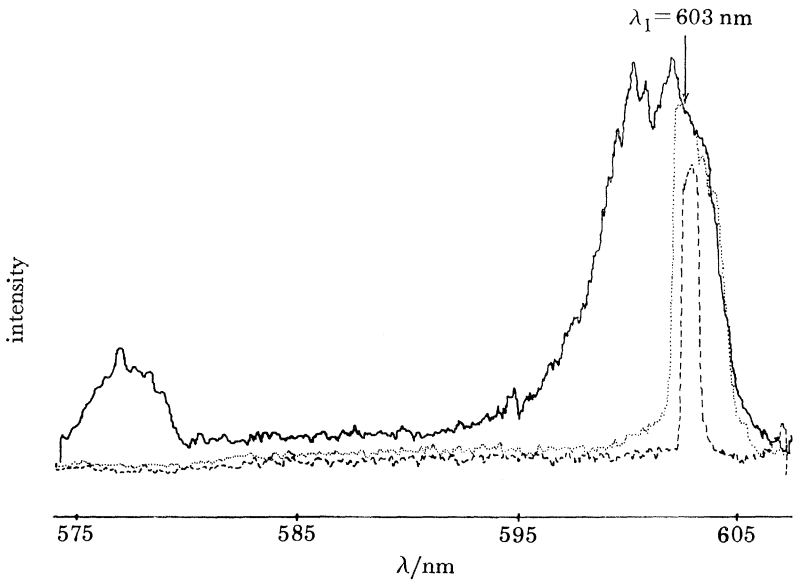

FIGURE 7. Influence of pump energy of laser II (Rhodamine 6G/DODCI) on the spectral distribution of the output (—, 0.2 %; · · ·, 0.4 %; – – –, 0.6 % below threshold). Bleaching wavelength $\lambda_I = 603$ nm. (From Lill *et al.* (1977*c*).)

Insertion of the Fabry–Perot etalon FP2 into the resonator of laser II results in the production of picosecond pulses tunable independently from λ_I over a considerable wavelength region. Table 1 lists the experimental data for various combinations of active and passive media in the two oscillators. The wavelength range given for the passively mode-locked system L_I indicates within which limits the wavelength of the bleaching pulses can be tuned if the slave oscillator L_{II} should remain independently tunable in the wavelength region indicated. The tuning

range of the latter is shifted towards shorter wavelengths compared with that of the equivalent, passively mode-locked system when DODCI or DQOCI are used as saturable absorber dyes. The explanation for this shift is based on the observation that photoisomers are created which absorb at a longer wavelength (see §5).

TABLE 1. EXPERIMENTAL DATA FOR VARIOUS COMBINATIONS OF 'ACTIVELY' AND PASSIVELY MODE-LOCKED SYSTEMS

active medium		saturable absorber (concentration/M)		lasing wavelength/nm		electron energy/J		max. pulse energy/μJ	
L_I	L_{II}	L_I	L_{II}	L_I	L_{II}	L_I	L_{II}	L_I	L_{II}
Fl 7 GA	Rh 6 G	DQOCI (10^{-4})	DQOCI (5×10^{-5})	550–580	570–603	290	360	60	70
—	—	—	DODCI (5×10^{-5})	—	580–618	290	360	60	80
Rh 6 G	Fl 7 GA	DODCI (10^{-4})	DQOCI (10^{-4})	585–620	550–580	170	650	60	30
—	Rh 6 G	—	DODCI (5×10^{-5})	—	570–618	170	360	60	70
—	Rh B	—	DQTCI (8×10^{-5})	—	610–643	170	500	60	15

Pulse generation in the slave oscillator was studied with a fast streak camera (Lill *et al.* 1977*c*). To this end, pulses were selected from various parts of the train of laser II by using a Pockels cell switch (see figure 4). After passing through a calibrated optical delay line, single pulses were directed on to the S-20 photocathode of the Photochron II streak tube (for details see Bradley 1978). The instrumental time resolution of the camera was 1.5 ps in the wavelength region used.

Unlike dye lasers synchronously pumped by second harmonic pulses of a mode-locked Nd:YAG laser (Goldberg & Moore 1975) or injection locking by pulses from a mode-locked c.w. laser (Moses *et al.* 1976), mismatch in cavity length up to ±40 mm did not effect the width of the pulses of laser II after mode-locking was complete. In figure 8, microdensitometer traces of streak camera records are displayed characterizing pulses selected *ca.* 300 ns after the start of the pulse train of laser II. Both lasers were operated with Rhodamine 6G and DODCI as active and passive mediums respectively. Operating wavelengths were $\lambda_I = 604$ nm and $\lambda_{II} = 595$ nm, at which wavelength mode-locked pulse formation occurred in a time of *ca.* 100 ns, when the system is used as a passively mode-locked laser (pulse duration is essentially the same in both modes of operation). To study the initial region of the pulse train of the secondary oscillator, laser II was tuned to 585 nm, where under passive mode-locking conditions the build-up time of single pulses is relatively long (150–200 ns). The pulses examined were selected *ca.* 80 ns from the beginning; typical microdensitometer traces are displayed in figure 9. In general, single pulse development was not yet complete, although in most cases the pulses recorded showed the evolution of a single pulse with a duration of less than 5 ps. Single pulse generation after this type of profile usually takes place within a few more cavity round trips (see also Arthurs *et al.* 1973; New & Rea 1976). Therefore, as a source of ultrashort pulses, the initial region of the actively mode-locked train should be avoided. Furthermore, it is recommended that the secondary oscillator be operated at a slightly shorter resonator

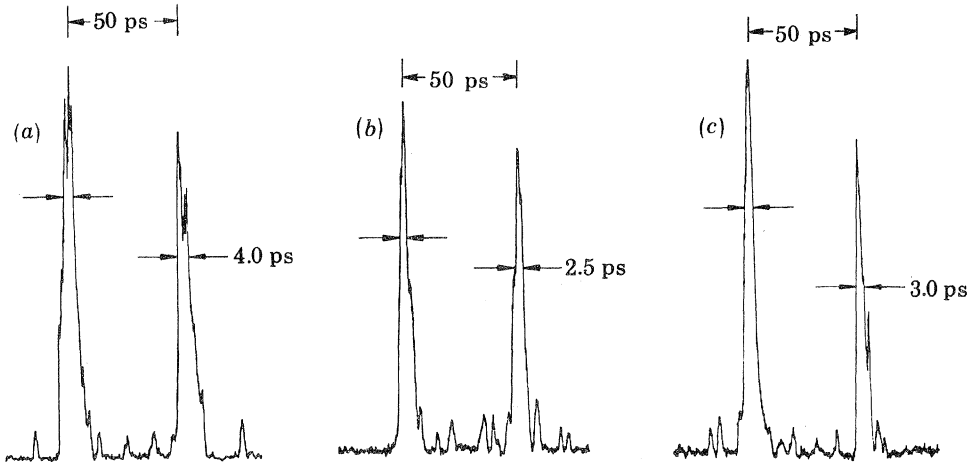

FIGURE 8. Microdensitometer traces of pulses selected *ca.* 300 ns from the beginning of the pulse train of laser II (cavity length 580 nm) for various cavity lengths of the master oscillator: (*a*) 620 mm, (*b*) 560 mm and (*c*) 520 mm. For more details see text. (From Lill *et al.* (1977*c*).)

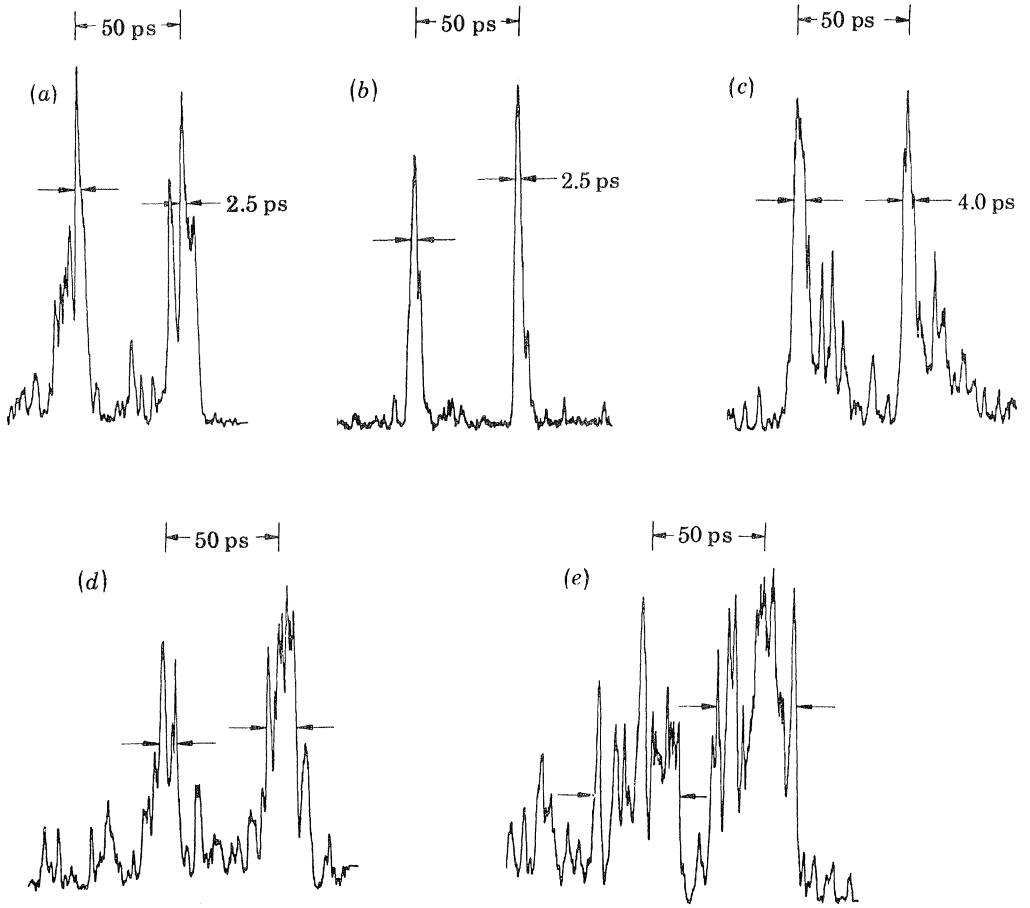

FIGURE 9. Microdensitometer traces of pulses selected *ca.* 80 ns from the beginning of the pulse train of laser II (cavity length 580 mm) for various lengths of the master oscillator: (*a*) 620 mm; (*b*) 600 mm (*c*) 580 mm; (*d*) 560 mm and (*e*) 540 mm. For more details see text. (From Lill *et al.* (1977*c*).)

length. Pulses circulating in the secondary oscillator will then, after the initial starting help, 'walk fast' through the gate open time of the saturable absorber with the effect that the consecutive pulse development will be comparable with that occurring in passively mode-locked systems.

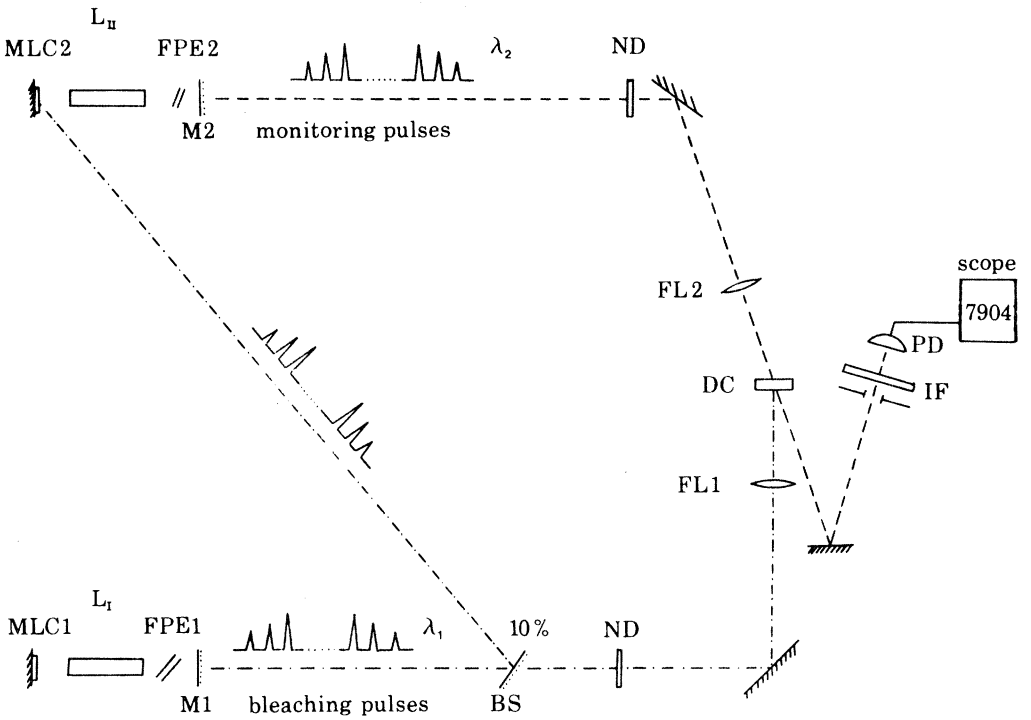

FIGURE 10. Set-up for excite–probe experiment with the use of two independently tunable trains of picosecond pulses (FPE, Fabry–Perot etalon; BS, beam splitter; FL, focusing lens ($f = 100$ mm); ND, neutral density filter; DC, dye cell; IF, interference filter; PD, photodiode).

5. RAPID OPTICAL SAMPLING OF RELAXATION PHENOMENA EMPLOYING TWO TRAINS OF TIME–CORRELATED PICOSECOND PULSES

In the past, the decay curve of a transient, photoinduced transmission of, for example, a dye solution was measured point by point by splitting one laser pulse train and mechanically varying (between pulse firings) the delay between the arrival of excitation and probing pulses. By making use of two optically coupled mode-locked dye lasers in the arrangement displayed in figure 10, it is possible to monitor the decay curve by one firing only of the two lasers (Lill *et al.* 1977*d*). Furthermore, the restriction of the previous technique, that pump and probe pulse have the same frequency, is abolished because both lasers can be frequency tuned independently.

The principle of operation is shown in figure 11. Since laser II starts to oscillate in response to the loss modulation of the saturable absorber in its resonator, it is guaranteed that one pair of pulses of the middle of the train of each laser arrives at essentially the same time at the sample cell DC. An inequality in optical length (round-trip time) of the two resonators then causes the difference in arrival time of each consecutive pair of pulses to increase automatically by the amount

$$\Delta t = t_2 - t_1 = 2\Delta L/nc,$$

[31]

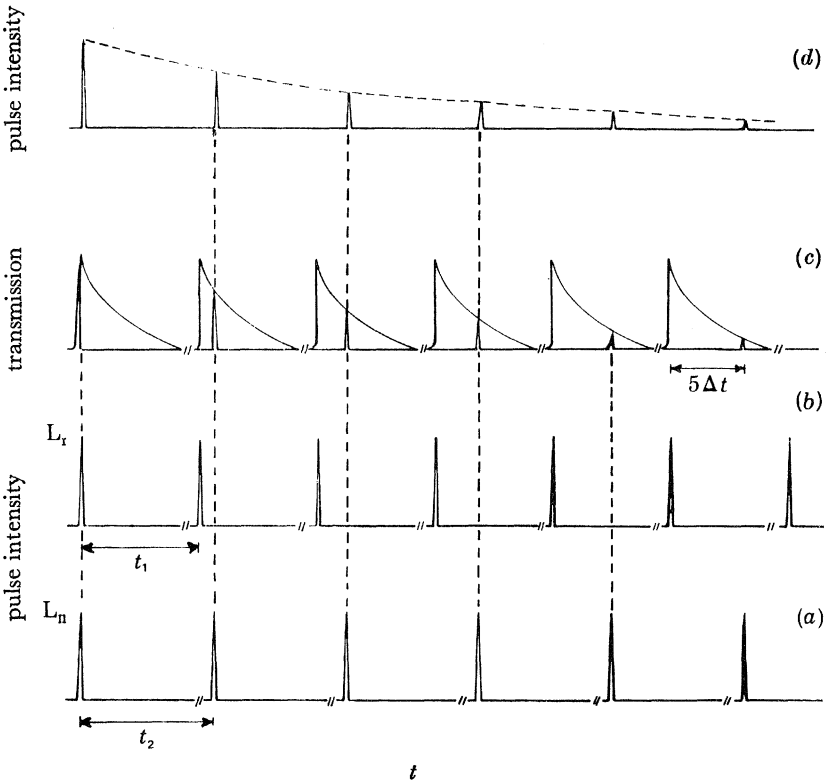

FIGURE 11. Optical sampling of the induced transmission decay: (*a*) series of probing pulses originating from laser II; (*b*) series of exciting pulses originating from laser I; (*c*) transient transmission induced by each bleaching pulse (laser I) and monitoring by the corresponding probing pulse of laser II; (*d*) intensity distribution of the transmitted probing pulses as seen on the oscilloscope.

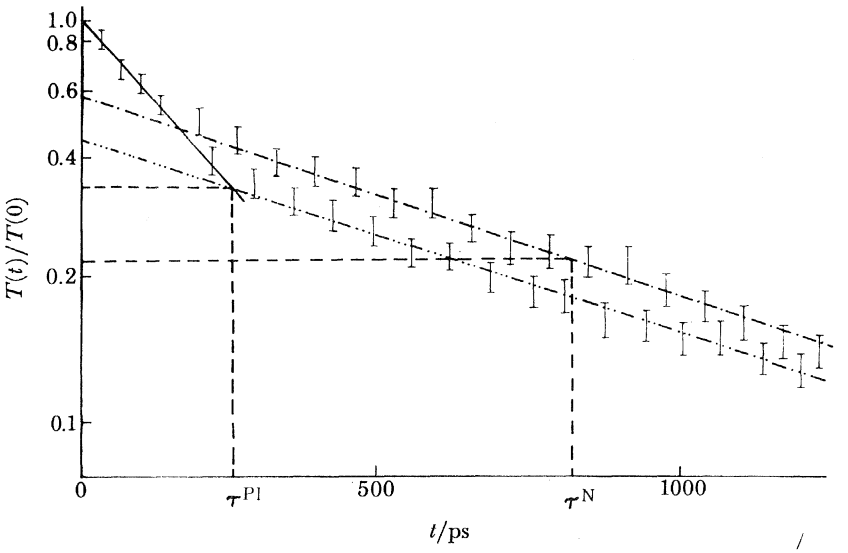

FIGURE 12. Normalized transmission of consecutive probing pulses. Sample is an ethanolic solution of DODCI. Excitation wavelength $\lambda_I = 580$ nm, probing wavelength $\lambda_{II} = 580$ nm (–·–·–) and 610 nm (–···–). (After Lill (1978).)

which can be adjusted to the appropriate value by a simple micrometer-controlled mounting of the output mirror of one of the two lasers (figure 11*a*, *b*). The transmission decay of the repetitively bleached sample is thereby transformed into the intensity distribution of the transmitted pulse sequence (see figure 11*c*, *d*; inter-pulse distance *ca.* 5 ns).

In figure 12, the normalized transmission of consecutive pulses is displayed (logarithmic scale) for an ethanolic solution of DODCI. The wavelength λ_I of the exciting pulses was chosen to coincide with the absorption maximum of DODCI in its 'normal' form (see figure 13). The wavelength of the probing pulses, on the other hand, was tuned to 580 and 610 nm, respectively. In both cases the biexponential nature of the decay is apparent. Since probing at 610 nm monitors relatively more of the fast decaying transmission change, we conclude that the shorter aperture time is correlated to the photoisomer whose absorption spectrum was determined by Dempster *et al.* (1972).

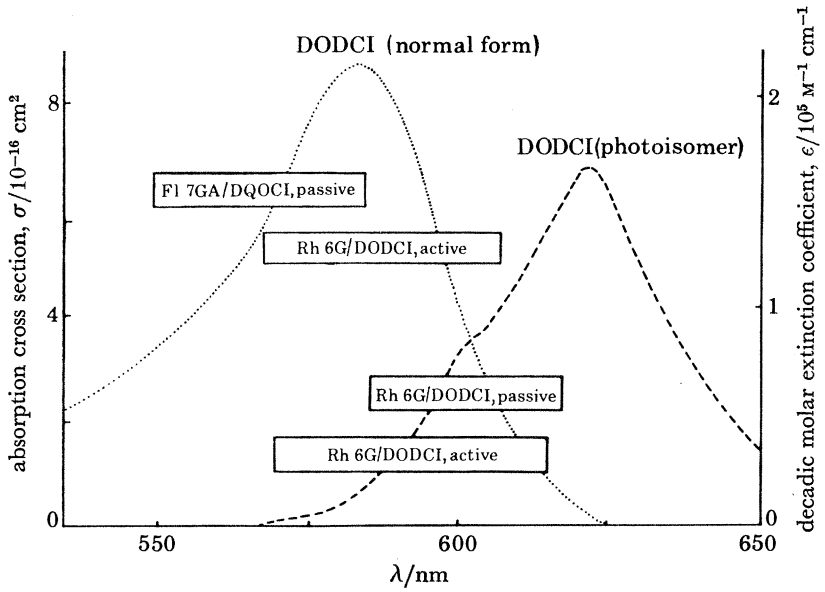

FIGURE 13. Absorption spectra of DODCI 'normal' form and photoisomer. The tuning range of the 'actively' mode-locked laser, when operated together with a Fluorol 7GA and Rhodamine 6G master oscillator, is displayed.

As in any experiment of this kind, the excited state lifetime of the dye under study must be deduced from the aperture time by means of a model. Applying, as usual, a simple two-state model, lifetimes of 1.3 ± 0.2 ns and 430 ± 40 ps are found for the two forms of DODCI. These numbers are in fair agreement with the results of other groups (see, for example, Jaraudias *et al.* 1977). Formation of the DODCI photoisomer is, in our opinion, also the explanation for the shift in tuning range of the 'actively' mode-locked laser (see figure 13) and its modified behaviour during the build-up phase of mode-locking, when this dye is used as saturable absorber.

The technique just described has also been used to study the viscosity dependence of the excited state lifetime of a merocyanine dye (*N*,*N*-dimethyl-amino-7-dicyanomethylene-heptamethyne). In glycerol–water mixtures, an $\eta^{\frac{2}{3}}$ dependence was found (see figure 14) as in triphenylmethane dyes (see, for example, Yu *et al.* 1977). One may expect to observe a similar

viscosity dependence for other dyes too. The result of Mialocq *et al.* (1977*a*) confirms this for pinacyanol (see §2). Therefore, more attention is recommended in choosing the proper solvent for making up saturable absorber solutions. With a recovery time in the 100 ps region, mode-locking may become possible for many more laser dyes than has so far been achieved.

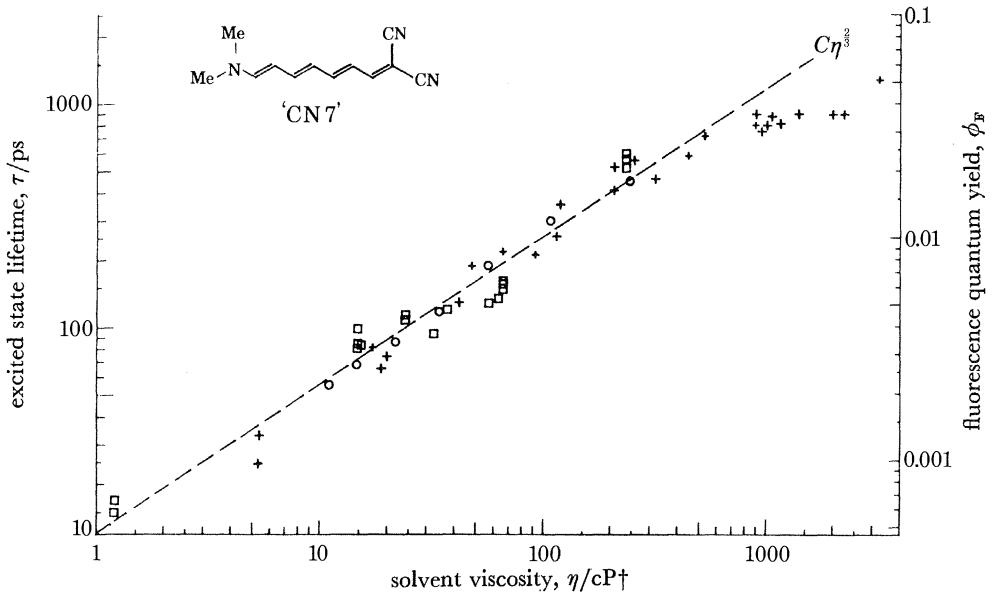

FIGURE 14. Influence of solvent viscosity on excited state lifetime and fluorescence quantum yield (+, fluorescence decay (from Yu 1978); □, ground state recovery; ○, quantum yield). †1 cP = 1 mPa s.

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