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# Subpicosecond Pulse Generation in Synchronously Pumped and Hybrid Ring Dye Lasers

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**Abstract.** Subpicosecond pulse generation has been examined in synchronously pumped mode-locked ring dye laser systems. These include hybrid and composite absorber/gain media arrangements as well as a simple synchronous cavity. The shortest pulses recorded were 0.3 ps for the hybrid system, and this has been shown to be critically dependent on the positioning of the absorber jet in the centre of the cavity to better than 50 μm. Stable operation for subpicosecond pulse generation has been achieved in the ring configuration with greater wavelength tunability and higher average power conversion efficiency than with conventional cavity arrangements.

**PACS:** 42.55 Mv, 42.60 Da

Although subpicosecond pulses have been generated with synchronously pumped cw dye lasers using various techniques [1–4], shorter pulses are obtained using the simpler passively mode-locked arrangement [5–7]. However, synchronously mode-locked systems provide a higher average power conversion and also a wider spectral tunability because saturable absorbing dyes are avoided. The recently introduced technique of colliding pulse mode-locking [8], where two counter-propagating pulses interact simultaneously with a saturable absorber in a passively mode-locked ring dye laser, can be directly applied to synchronously mode-locked systems to give rise to highly stable, subpicosecond pulses over a wide tuning range.

In this paper we report on two mode-locked synchronously pumped ring dye laser systems. The first was a straightforward extension of the standard linear cavity, and the second a hybrid system containing a saturable absorber. Some of the factors which give rise to variations in the durations of the generated pulses such as lasing wavelength, pump power, absorber concentration and the timing of the coincidence of the colliding pulses in the absorber jet are given.

## Experimental Systems and Results

Two basic cavity configurations were used, and these are shown in Fig. 1. The cavity elements of the laser have been described in detail previously [9]. An acousto-optically mode-locked argon ion laser was used to provide 80 ps pumping pulses at 514.5 nm at a repetition rate of 140 MHz with typical average powers of $\sim 700$ mW.

Initial measurements were taken with the cavity arrangement shown in Fig. 1a. Mirrors $M_1$ ($r=\infty$), $M_2$ and $M_3$ ($r=10$ cm) were all nominally 100% reflecting in the range 500–700 nm while the plane mirror $M_4$ had a 95% reflectivity in this wavelength range. The rhodamine 6G ($2\times10^{-3}$ M in ethylene glycol) was flowed in a vertical jet stream of width $\sim 100$ μm placed at the common focus of $M_2$ and $M_3$. In this arrangement the dye laser cavity was arranged to be equal in length to that of the argon ion pump laser. Synchroscan streak camera measurements showed that the counterpropagating pulses overlapped at the jet stream to within the limit of the streak camera resolution [10]. Typically, the average power in each

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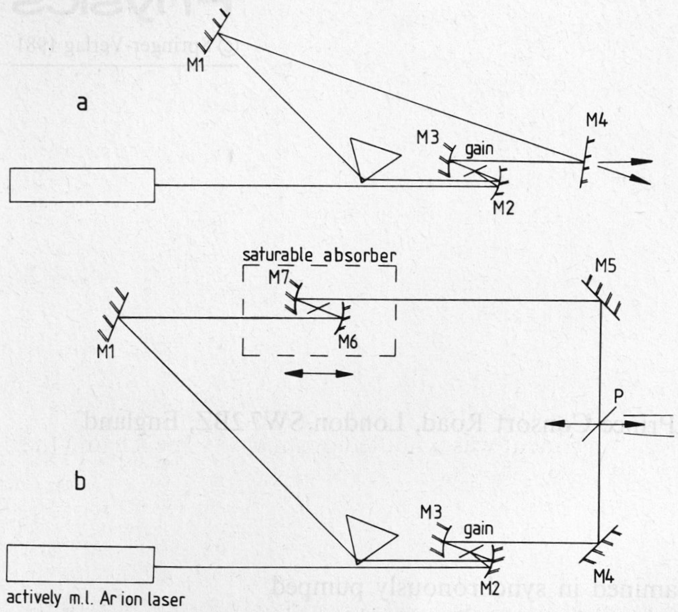


Fig. 1a and b. Schematics of experimental laser arrangements (a) synchronously pumped system (b) hybrid system

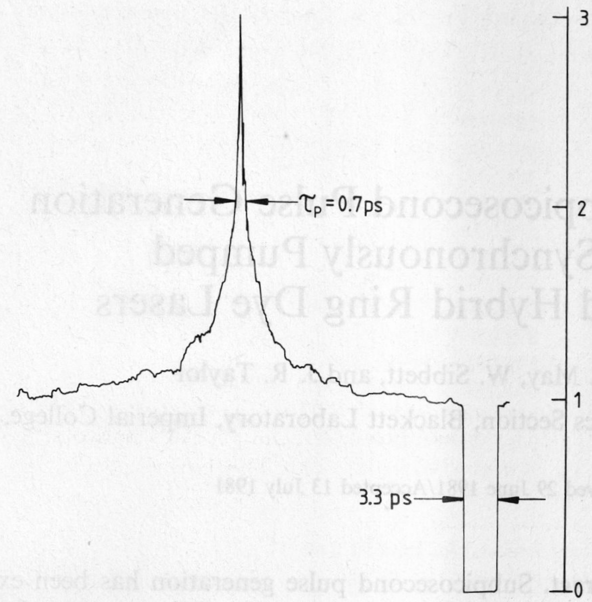


Fig. 2. Intensity autocorrelation trace of a mode-locked pulse train using the cavity arrangement of Fig. 1a ($\lambda = 595$ nm)

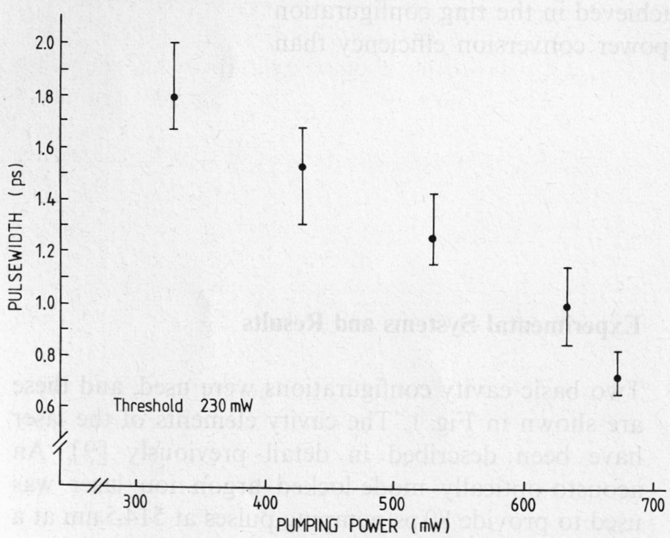


Fig. 3. Variation of average recorded pulsewidth with pumping power for the pure synchronously pumped configuration of Fig. 1b

of the output beams was 30 mW at 585 nm. The pulse widths were measured using a standard second-harmonic generation autocorrelation technique and the durations were calculated from the full width at half maximum assuming a Gaussian pulse shape. (All the measured pulse profiles throughout this work were taken to be Gaussian.) Figure 2 shows a typical autocorrelation trace of the output of the mode-locked dye laser operating at 595 nm, for an argon ion pump power of 640 mW. The measured duration of 0.7 ps was considerably shorter than had been achieved previously with the conventional linear cavity [9]. Simultaneous measurement of the spectral width gave a smooth broad spectrum of 1.5 nm width ($\Delta\nu\Delta t = 0.88$) and indicated that operation was well above the Fourier transform limit. Continuous tuning over the range 562–630 nm was obtained with appreciably shorter pulses and greater stability than with the conventional cavity. This is to be expected in the ring arrangement because pulse shortening is enhanced by the counterpropagating pulses giving rise to a standing wave in the jet which will deplete the gain with greater effect than that of a single unidirectional pulse.

Further measurements were taken using the cavity configuration shown in Fig. 1b. The overall cavity length was increased to be twice that of the argon ion laser and the plane mirrors $M_4$ and $M_5$ were 100% reflecting for radiation $\sim 600$ nm incident at 45°.

Mirrors $M_1$, $M_2$, and $M_3$ were the same as those described in the previous section, and an additional folded section was constituted by mirrors $M_6$ and $M_7$ ($r = 10$ cm) which were totally reflecting over the range 500–700 nm. Wavelength selection was again accomplished using a prism. All the mirrors had reflectivities $\sim 100\%$ and so it was necessary to include a pellicle (P) inside the cavity to couple out some laser radiation as indicated.

Initially, this cavity was operated in the pure synchronously pumped regime, with no dye flowing in the second folded section. Threshold powers were 230 mW in this system compared to 200 mW in the laser previously described. However, pulsewidths and operation were practically identical. The variation of the output pulsewidth with argon ion pumping power is shown in Fig. 3. The points represent the average of about twenty consecutive autocorrelation traces for the pumping powers indicated, and the bars illustrate the full range of recorded pulsewidths at 595 nm. Typically the average output powers per beam for this range of pumping powers varied in an approximately linear manner from about 8 mW for a pump power of 330 mW to 30 mW at 670 mW pump. A clear trend in decreasing output pulsewidth with argon ion pump power can be seen in Fig. 3, where durations $\sim 1.8$ ps for pump powers 100 mW above threshold decreased to $\sim 0.7$ ps for powers of 440 mW above threshold. It is most likely that this decrease in the measured pulsewidth is associated with the increase in the gain bandwidth accompanying the increasing pump power over threshold. The simultaneous spectral measurements again indicated that the operation was above the Fourier transform limit.

Previous measurements have shown [9] that a hydrid mode-locked dye laser system i.e. one which has the addition of a separate saturable absorbing dye stage in a conventional synchronously pumped laser cavity, has given pulse shortening while maintaining comparable peak pulse powers. A brief analysis by Fork et al. [8] for passive mode-locking has indicated that under certain experimental conditions determined principally by the saturation parameters of the absorber, a distinct minimum energy loss occurs when the counterpropagating pulses of the ring laser precisely overlap in the absorber. A similar concept had previously been advanced for the use of thin optically-contacted absorber dye cells in passively mode-locked laser systems [11]. The condition of pulse synchronism is easily achieved in a passively mode-locked ring laser as it arises from the initial pulse formation mechanism in the saturable absorber. However, in a hybrid configuration this is more difficult to achieve, since the pulse formation is determined initially by the active medium and so the saturable absorber must be placed in the cavity such that the path lengths to it from the gain medium for the counterpropagating pulses are equal or differ by a cavity round-trip time of the pumping laser.

We have examined the output pulse width dependence on the position of the saturable absorber for the hybrid ring laser. The scheme used is shown in Fig. 1b as described above. A 100 µm dye jet stream of DODCI (3,3′-diethyloxadicarbocyanine iodide) in ethylene glycol was placed at the common focal points of the totally reflecting mirrors $M_6$ and $M_7$. In order that the jet position could be varied without changing the focus or the overall cavity length, the complete jet/mirror arrangement was mounted on an optical bench and the total system (shown as the dashed area in Fig. 1b) was capable of being positioned with micrometer precision to better than 5 µm. The synchroscan streak camera was again used to determine the approximate overlap position at the saturable absorber of pulses derived in the gain medium. To do this a pellicle was used in place of the saturable absorber jet and the counterpropagating beams reflected off this pellicle were directed into the streak camera. The complete absorber/mirror assembly could then be moved to approximately determine the position of pulse overlap by utilizing the "real time" display of the streak camera. Unfortunately, due to the limitations of streak camera resolution and measurement restrictions on the pellicle position and the re-location of the absorber dye jet the inaccuracy in the timing of the overlap was $\sim 5$ ps.

Further insight into the overlap position was achieved by examining the output pulsewidths and their variations with position of the jet in the ring cavity. A mixed saturable absorber solution of $1 \times 10^{-5}$ M DQOCI (1,3′-diethyl-4,2′-quinolyloxacarbocyamine iodide) and $1 \times 10^{-5}$ M DODCI was used. These dyes have upperstate lifetimes of 16 ps and 1.15 ns, respectively [12]. The system was tuned to operate at 595 nm with a pump power of 650 mW. Variations of the recorded intensity autocorrelations taken in a continuous scan of the position $X$ of the saturable absorber can be seen in Fig. 4. The position of the recorded minimum pulsewidth was specified as $X = 0$ (this lay within that determined from streak camera measurements). Assuming a Gaussian profile, then for the trace shown the inferred pulsewidth is 0.5 ps. The pulsewidth variation with position $X$ showed a fairly symmetrical characteristic. At 50 µm above and below the zero position appreciably broadened pulsewidths $\sim 0.7$–$0.8$ ps were recorded, while at $\pm 100$ µm the pulses were measured to be $\sim 1.3$–$1.4$ ps (Fig. 4). Additional displacement to $\pm 200$ µm yielded no further significant broadening, and many scans through the zero position gave similar variations of recorded pulsewidth with position.

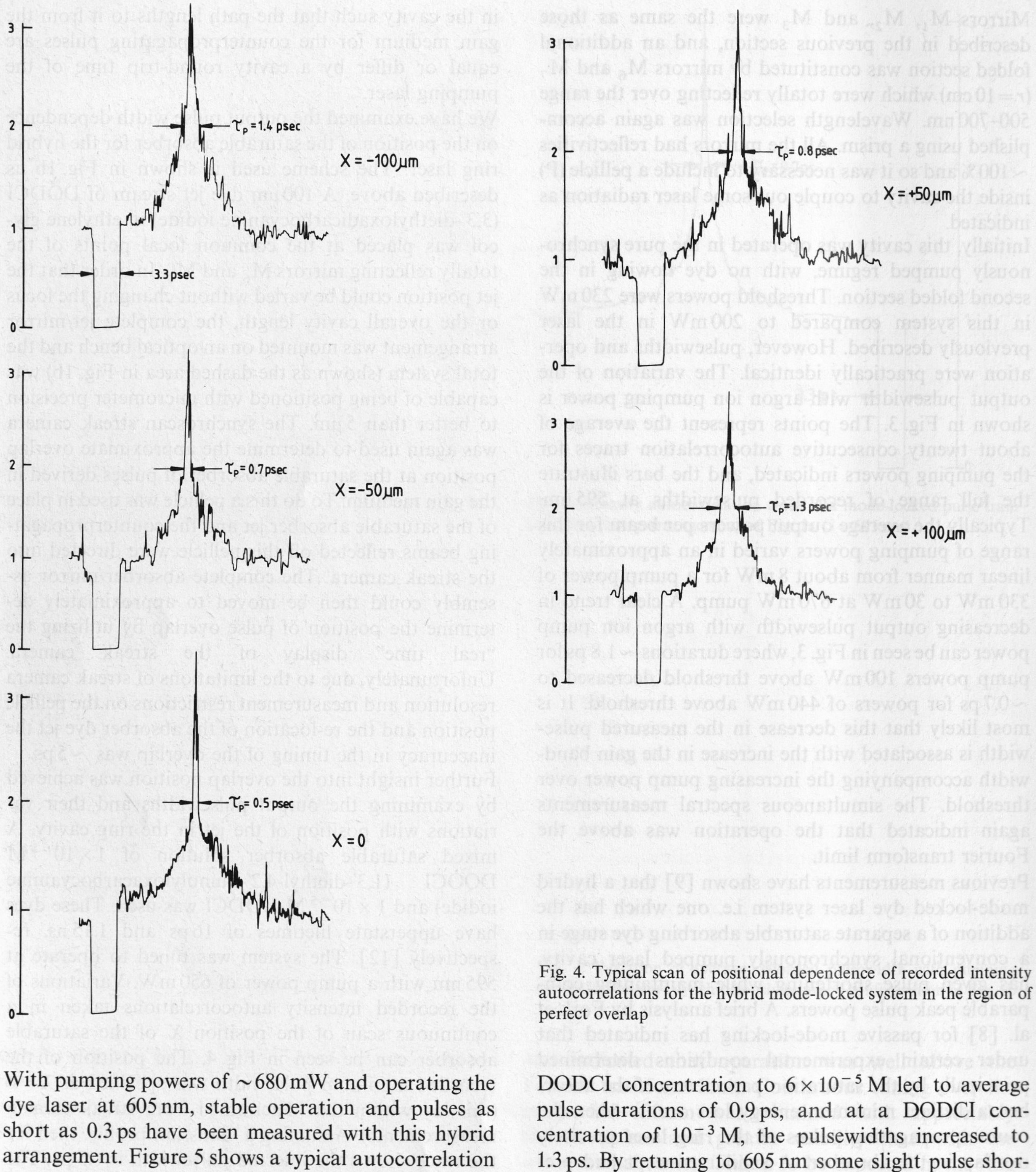


Fig. 4. Typical scan of positional dependence of recorded intensity autocorrelations for the hybrid mode-locked system in the region of perfect overlap

With pumping powers of ~680 mW and operating the dye laser at 605 nm, stable operation and pulses as short as 0.3 ps have been measured with this hybrid arrangement. Figure 5 shows a typical autocorrelation trace of a 0.35 ps pulse recorded at 605 nm with average powers per beam of ~15 mW. It is highly likely that the jet stream thickness was limiting the pulsewidths in this case and as pointed out by Fork et al. [8] reduction in jet thickness should further reduce the pulse durations.

When operating at 595 nm, a slight increase in the DODCI concentration did not affect the average minimum pulsewidth recorded. However, increasing the DODCI concentration to $6 \times 10^{-5}$ M led to average pulse durations of 0.9 ps, and at a DODCI concentration of $10^{-3}$ M, the pulsewidths increased to 1.3 ps. By retuning to 605 nm some slight pulse shortening was detected which was probably due to lowering of the absorption cross section at this wavelength.

The precise timing problem of pulse overlap can be simplified in hybrid systems by using a composite gain-absorber dye solution, as has been demonstrated for conventional passively mode-locked [13] and synchronously mode-locked lasers [14]. The cavity arrangement of Fig. 1b was used except that the saturable

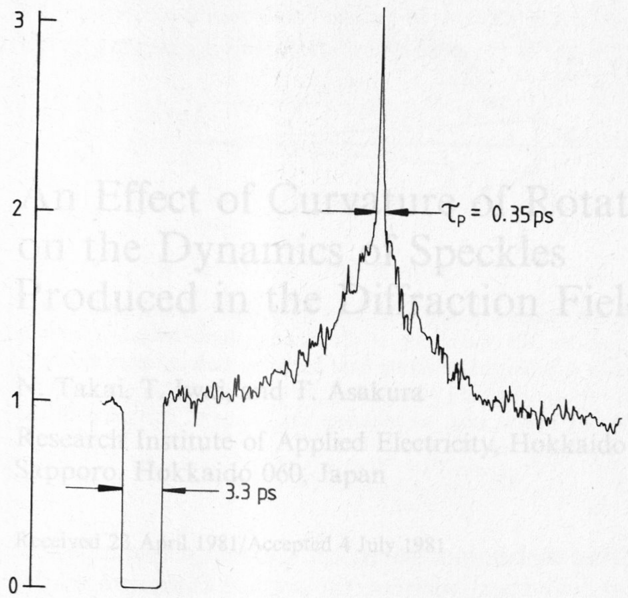


Fig. 5. Autocorrelation trace of a 0.35 ps pulse obtained with the hybrid laser

absorber was not circulated in its separate system, but added to the gain medium. For a $2 \times 10^{-5}$ M DQOCI absorber concentration and lasing at 595 nm for a pump power of 640 mW, a minimum pulse duration of 0.8 ps was obtained. Again small amounts of DODCI ($\sim 10^{-5}$ M) added to the system made little significant difference to the pulses generated. As in the case of the hybrid system, increasing the concentration of the DODCI to $\sim 10^{-4}$ M increased the average measured pulsewidth to $\sim 1.4$ ps and further increases to $\sim 5 \times 10^{-4}$ M gave rise to pulses of $\sim 1.7$ ps. Retuning to 605 nm decreased the pulse durations for the latter concentration to 1.3 ps. This observation would suggest that the saturable absorber in a composite medium may be less effective due to direct heating effects from the pumping Ar ion laser.

## Conclusion

It has been shown here that the method of colliding pulse mode-locking in a ring cavity when applied to a synchronously pumped laser does give rise to greater stability, a wider tuning range, higher average power conversion and shorter pulses compared to a conventional cavity. However, the hybrid system is not entirely suitable as an experimental laser as the positioning of the second jet tends to be rather critical to ensure the generation of the shortest pulses. On the other hand, the simple synchronously pumped ring system gives high stability and subpicosecond pulses which are well suited for applications in time-resolved spectroscopy.

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