PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Synchronously Pumped Continuous Wave Dye Lasers

M. C. Adams, D. J. Bradley, W. Sibbett and J. R. Taylor

Phil. Trans. R. Soc. Lond. A 1980 **298**, 217-223
doi: 10.1098/rsta.1980.0245

Phil. Trans. R. Soc. Lond. A **298**, 217–223 (1980)
Printed in Great Britain

Synchronously pumped continuous wave dye lasers

By M. C. Adams†, D. J. Bradley, F.R.S., W. Sibbett and J. R. Taylor
Optics Section, Blackett Laboratory, Imperial College,
Prince Consort Road, London SW7 2BZ, U.K.

The generation and measurement of picosecond and subpicosecond pulses from synchronously pumped continuous wave (c.w.) dye lasers are reviewed. A streak camera operating at 140 MHz repetition rate in synchronism with the pulses from a synchronously mode-locked Rhodamine 6G dye laser is described, and the results of some dye fluorescence decay studies have also been included to demonstrate the unique capabilities of this synchronous generation and measurement technique.

Laser pulses with durations as short as 0.2–0.3 ps have been generated by passively mode-locked c.w. Rhodamine 6G dye lasers (Ippen & Shank 1975; Ruddock & Bradley 1976; Diels *et al.* 1978), but unfortunately the tuning range over which these subpicosecond pulses can be obtained is limited to *ca.* 15 nm by the absorption characteristics of the saturable absorber (Bradley 1977). Also, since suitable mode-locking dyes for the blue and near infrared spectral regions are not readily available, it is difficult to generate such ultra-short light pulses beyond the wavelength region of 590–615 nm. It is therefore often more convenient and efficient to pump the c.w. dye laser synchronously with an actively mode-locked argon ion or krypton ion laser. In this way picosecond (and at some wavelengths subpicosecond) pulses have been produced over the spectral range extending from 420 to 835 nm (Eckstein *et al.* 1978; Chan & Sari 1974; de Vries *et al.* 1976; Frigo *et al.* 1977; Heritage & Jain 1978; Ryan *et al.* 1978; Kuhl *et al.* 1979; Adams 1979; Fehrenbach *et al.* 1978).

Although the synchronous pumping technique was first applied to pulsed lasers (Bradley & Durrant 1968; Glenn *et al.* 1968; Soffer & Linn 1968) its use in c.w. dye lasers requires more exacting experimental conditions. Particular attention has to be paid to the stability of the pump pulses from the ion laser and to the precise matching of the dye laser cavity length to be equal to, or a multiple or submultiple of, the ion laser cavity length. One type of basic configuration of a synchronously pumped dye laser is shown schematically in figure 1. Alternative configurations are used when the prism tuning element (Chan & Sari 1974; Ryan *et al.* 1978) is replaced by a two or three-plate birefringent filter (Kuhl *et al.* 1979), wedge interference filter (Kuhl *et al.* 1979; Adams 1979) or a Fabry–Perot etalon (Ausschnitt *et al.* 1978).

In all cases it is found that the shortest dye laser pulses are produced when mode-locking of the ion laser is properly optimized. This can be ensured by satisfying three main requirements. A most important factor is the frequency stability of the r.f. oscillator which provides the electrical driving signal to the acousto-optic modulator. The variation in frequency should be less than 1 part in 10^6. The temperature of the tuning prism should also be kept stable to within ± 0.01 °C. Finally it is necessary to accurately align the ion laser beam through the tuning prism of its cavity so that the loss modulation has its maximum value. When sufficient care is taken it

† Present address: E.M.I. Industrial Electronics Ltd, Bury Street, Ruislip, Middlesex HA4 7TA, U.K.

is possible to obtain a reliable mode-locked output from many of the stronger lasing lines of the argon ion and krypton ion lasers.

For the u.v. and blue lines of the Ar^+ laser, pulse widths *ca.* 130–200 ps at average powers *ca.* 10–150 mW have been recorded (Eckstein *et al.* 1978; Adams *et al.* 1978). Shorter pulses of duration *ca.* 90 ps and higher average powers of *ca.* 1 W have also been obtained for the 514.5 nm Ar^+ line (Heritage & Jain 1978; Heising *et al.* 1971; Adams *et al.* 1978). In the Kr^+ laser, similar pulse widths *ca.* 100–150 ps at *ca.* 1 W average powers have been reported for the 647 nm line (Kuhl 1979; Fehrenback *et al.* 1978), and shorter 45–55 ps pulses at 120 mW have been measured for the 407 nm line (Steinmetz *et al.* 1978).

Figure 2 gives a clear indication of the criticality of the cavity matching for a synchronously pumped dye laser. It can be seen that while lasing continues despite a considerable cavity length

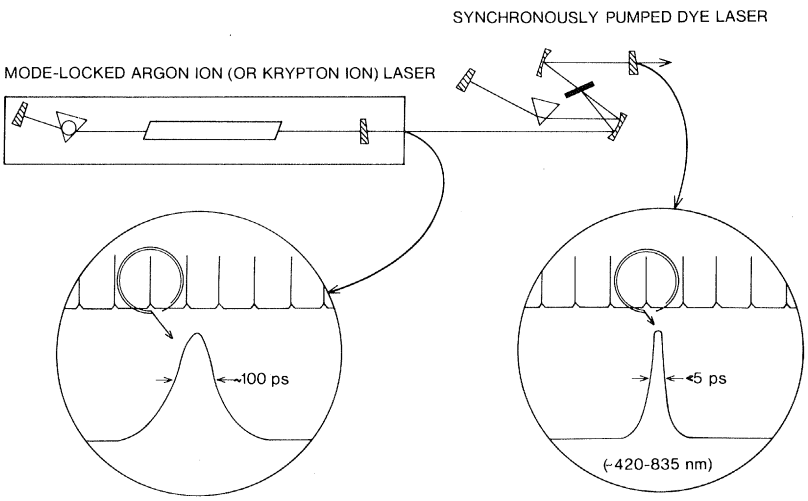


FIGURE 1. Mode of operation of a synchronously pumped c.w. dye laser.

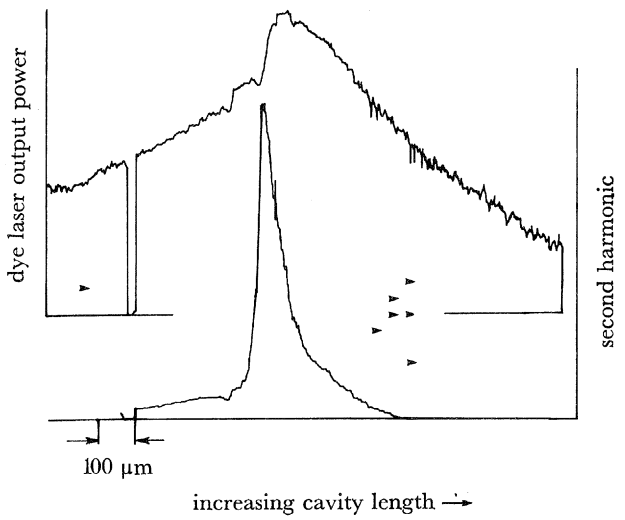


FIGURE 2. Variation of fundamental and second harmonic output power as a function of dye laser cavity length.

mismatch (*ca.* 1 mm), the second harmonic generation is restricted to a mismatching extent of only *ca.* 100 μm. Since the dye pulse duration T_p is related to the fundamental power P_f and second-harmonic power $P_{s.h.}$ by

$$T_p \propto (P_f)^2/P_{s.h.},$$

the experimental result suggests that significant pulse shortening takes place when the cavity lengths are closely matched. Once the maximum second harmonic power has been obtained, the shortest duration pulses are then obtained by fine-tuning the dye cavity length by a few micrometres. Under these optimized conditions, second harmonic generation autocorrelation traces (Bradley 1977) can be recorded for which the peak:background intensity ratio is 3:1. Many such records, which indicate that the dye laser is well mode-locked, have been presented in the literature (Eckstein *et al.* 1978; Chan & Sari 1974; de Vries *et al.* 1976; Frigo *et al.* 1977; Heritage & Jain 1978; Ryan *et al.* 1978; Kuhl *et al.* 1979; Adams 1979; Fehrenbach *et al.* 1978).

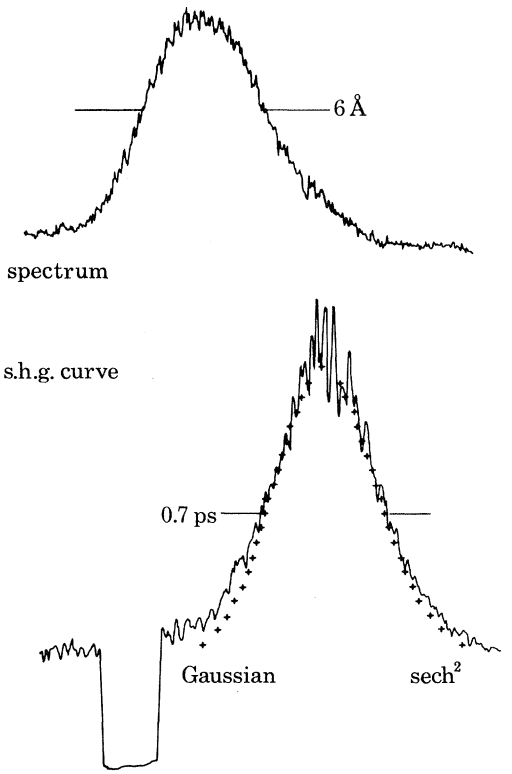


FIGURE 3. Spectrum and second harmonic generation (s.h.g.) autocorrelation profile for the subpicosecond pulses generated by a hybrid c.w. dye laser. ($1\,\text{Å} = 10^{10}\,\text{m} = 10^{-1}\,\text{nm}$.)

More direct and unambiguous observations of the pulses have been carried out by using a single-shot picosecond streak camera (Ryan *et al.* 1978). The pulse widths deduced from the streak images were often in close agreement with those determined from the autocorrelation traces. However, there were instances when some of the streak records obtained immediately after the recording of an acceptable autocorrelation trace showed the presence of substantially longer pulses in which substructure could frequently be seen. In other streak photographs there was clear evidence of the presence of satellite pulses. These variations in pulse character were usually short-term and unpredictable. Consequently, it is likely that during the recording of some auto-correlation traces, short-term instabilities occur which serve to complicate the results. This

could explain why the published work contains a variety of suggested or inferred pulse shapes which in many cases are shown to be imperfect fits to the autocorrelation profiles (e.g. fig. 3 in Ryan *et al.* 1978). Perhaps small departures from exact matching of the two laser cavity lengths are partly responsible for such pulse degradation because similar behaviour has been seen when cavity mismatches were introduced deliberately (Kuhl *et al.* 1979).

The first demonstration of subpicosecond pulse generation by a synchronously pumped c.w. dye laser was made by Heritage & Jain (1978). They used a synchronously mode-locked Rhodamine 6G dye laser to tandem-pump a Rhodamine B laser. With 55 ps pump pulses a 570 nm they showed that pulse durations *ca.* 0.7 ps could be generated within the wavelength range of 620–640 nm. Tandem pumping of an oxazine 750 dye laser by the 7 ps pulses from a synchronously pumped oxazine perchlorate laser has also been demonstrated as a method for generating subpicosecond pulses in the 770–785 nm spectral region (Kuhl *et al.* 1979). Subpicosecond pulses have been produced more directly by using a low finesse Fabry–Perot etalon (Ausschnitt *et al.* 1978) or wedge-shaped interference filter (Kuhl *et al.* 1979; Adams 1979), both of which permit lasing over a relatively broad bandwidth. In the extreme situation when no bandwidth limiting tuning element is included in the laser cavity, pulses as short as 0.5 ps have been obtained (Adams 1979).

Hybrid mode-locked c.w. dye lasers, in which gain modulation by the pump laser is supplemented by the loss modulation of a saturable absorber, are capable of producing near transform-limited subpicosecond pulses. Sech^2 or Gaussian pulse profiles with durations *ca.* 0.7 ps have been obtained (Ryan *et al.* 1978) when the mode-locking dye is in contact with the 100% reflector of a Rhodamine 6G dye laser. Furthermore, the time duration–bandwidth product for the pulses was 0.4, which agrees well with the expected values for sech^2 (0.32) or Gaussian (0.44) pulse shapes. When Fehrenback *et al.* (1978) added the saturable absorber to the oxazine 750 lasing dye, they found that subpicosecond pulses were only produced when the dye cavity length was equal to half that of the Kr^+ pump laser. Pulse durations of *ca.* 0.8–4.0 ps were obtained over the wavelength range 750–835 nm with the shortest pulses being generated around 770 nm, where the duration–bandwidth product was approximately unity. It would appear that the hybrid arrangements give better pulse shapes than their purely synchronously pumped counterparts. However, the severe restriction on tuning range imposed by the saturable absorber characteristics returns. Thus the particular output wavelength desired will largely dictate the nature of the laser system that can be used and ultimately the quality of the ultra-short pulses generated.

These frequency-tunable picosecond and subpicosecond light pulses can be applied widely to time-resolved spectroscopy. It is obviously advantageous to have a sensitive diagnostic technique that combines good dynamic range, high temporal and spatial resolution with a linear response over a wide spectral range. All of these characteristics are provided by a streak camera system that operates repetitively in synchronism with the pulse train from a mode-locked c.w. laser.

The operating principle and performance of the 'synchroscan' camera have already been discussed elsewhere (Adams *et al.* 1978; Bradley 1978) and only a few details need be included here.

A 140 MHz sinusoidal voltage applied to the deflexion plates of the image tube results in the streaked images of the dye laser pulses (or e.g. dye fluorescence resulting from laser pulse excitation) to be successively superimposed on the tube phosphor. In this way the camera becomes much more sensitive than when used as a single-shot streak camera, while retaining all the temporal and spatial resolution capability. When used in conjunction with a passively

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

mode-locked c.w. dye laser, the time resolution of synchroscan has been demonstrated to be *ca.* 3 ps (Adams 1979). Used with the synchronously pumped laser, however, the best temporal resolution so far demonstrated is *ca.* 10 ps (Ryan *et al.* 1978) which is adequate for most of the fluorescence studies being carried out at this stage.

The experimental configuration normally employed is shown schematically in figure 4. The fluorescence decay profile is displayed on the storage oscilloscope and a permanent record is made on a chart recorder. In the example shown in figure 5 for DSCI (3, 3′-diethylselenacarbocyanine iodide) in methanol, the excitation laser wavelength was 585 nm and the fluorescence signal at 628 nm was measured. It was thus possible to study how the fluorescence

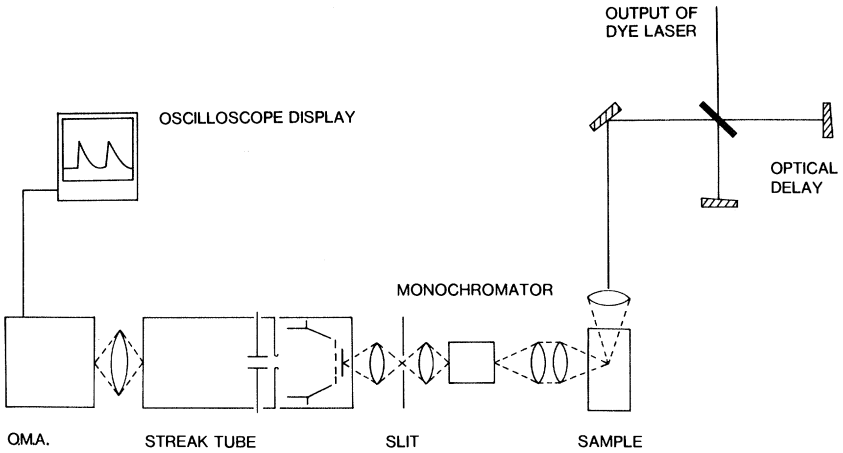


FIGURE 4. Schematic of the experimental arrangement used for fluorescence decay measurements with the synchroscan streak camera. (O.M.A., Optical Multichannel Analyser.)

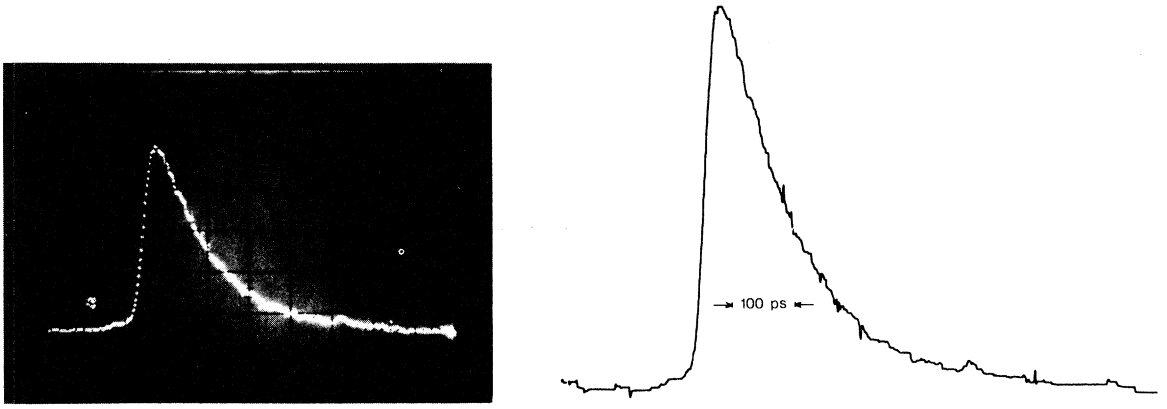


FIGURE 5. Storage oscilloscope display and chart recorder trace for the fluorescence decay of DSCI in methanol.

lifetime of DSCI and DQOCI (1,3′-diethyl-4,2′-quinolyloxcarbocyanine iodide) vary as a function of solvent viscosity. Thus the variation of solvent viscosity provides a convenient method for adjusting the ratio of the recovery times of the amplifying and absorbing media in passively mode-locked dye lasers (Bradley 1977).

It has been observed that the addition of malachite green (Ippen & Shank 1975; Bradley 1978) or DQOCI (Ruddock 1976) to the DODCI (3,3′-diethyloxadicarbocyanine iodide) saturable absorber has improved the performance of passively mode-locked c.w. dye lasers. The effect of these additive dyes in the shortening of the DODCI fluorescence lifetime has been the

subject of a recent study with the use of the synchronously pumped dye laser and a synchroscan camera (Adams *et al.* 1979*a*). With this direct method it was possible to examine singlet–singlet non-radiative resonance energy transfer from DODCI to malachite green and to DQOCI at sufficiently low laser excitation power densities so that complicating nonlinear effects are avoided. The results showed that the 1.2 ns fluorescence lifetime of a 10^{-4} M ethanolic solution of DODCI was reduced to *ca.* 960 ps and 750 ps respectively when 10^{-4} M and 10^{-3} M solutions of malachite green were added. For the addition of 10^{-4} M and 10^{-3} M solutions of DQOCI the lifetime of DODCI was more markedly reduced to 814 ps and 555 ps respectively.

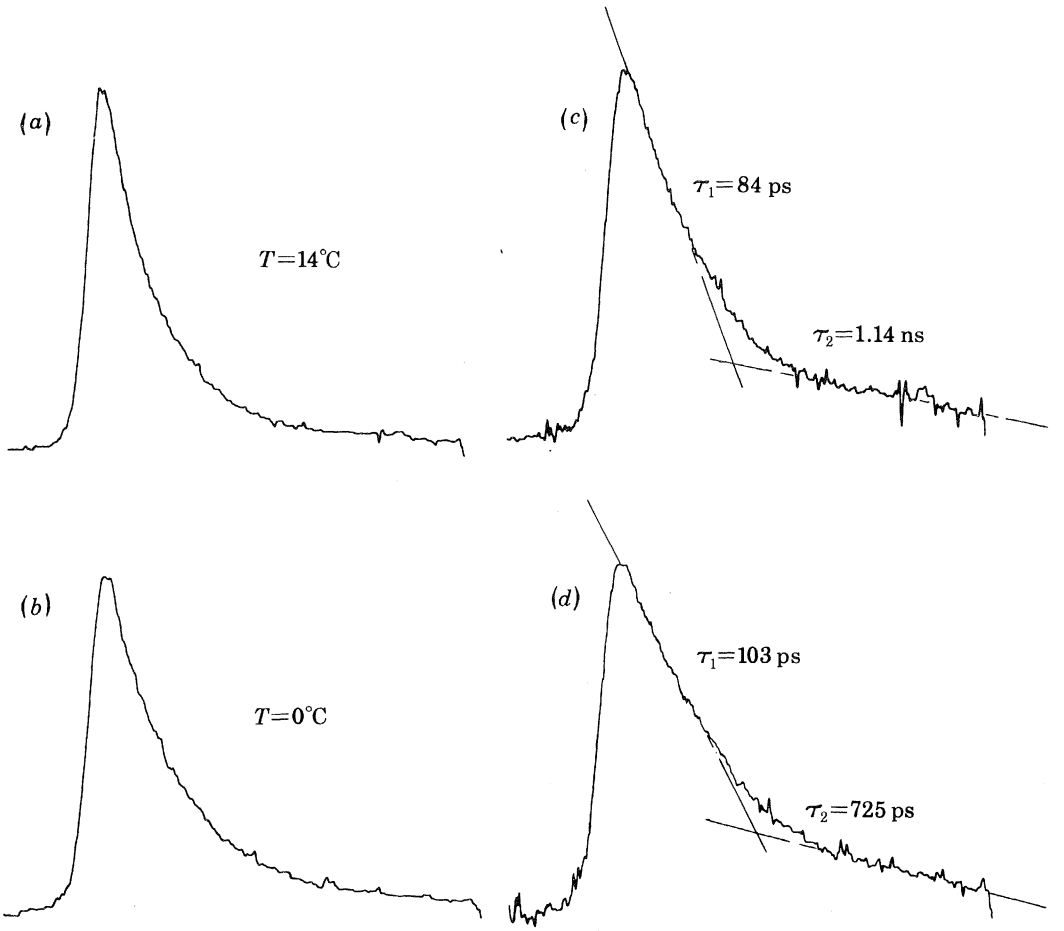


FIGURE 6. Linear scale (*a*, *b*) and logarithmic scale (*c*, *d*) fluorescence decay profiles for *trans*-stilbene.

The excellent sensitivity and high dynamic range of the synchroscan camera were also clearly demonstrated during the investigation of the fluorescence decay characteristics of *trans*-stilbene (Adams *et al.* 1979*b*). The *trans*-stilbene was excited by 300 nm laser pulses which were produced by intra-cavity second harmonic generation. A bi-exponential fluorescence decay profile is expected to result from the twisting mechanism of the excited *trans*-stilbene molecules (Birch & Birks 1976). Although this is rather difficult to see from figure 6*a* and *b*, corresponding to temperatures of 14 and 0 °C, the semi-logarithmic displays of figure 6*c* and *d* confirm that two rate constants are present. In the context of this paper it is worth emphasizing that the fluorescence signal was measured for an excitation power density of only *ca.* 10 kW cm^{-2}, produced

by focusing pulses of energy 0.7 pJ. The excellent dynamic range of the camera enabled the long-lived but much lower intensity component (*ca.* 50 times less intense than that of the short-lifetime component) of the fluorescence emission to be clearly distinguished while only the fast decay component was observed previously (Heisel *et al.* 1979) when a single-shot streak camera was used.

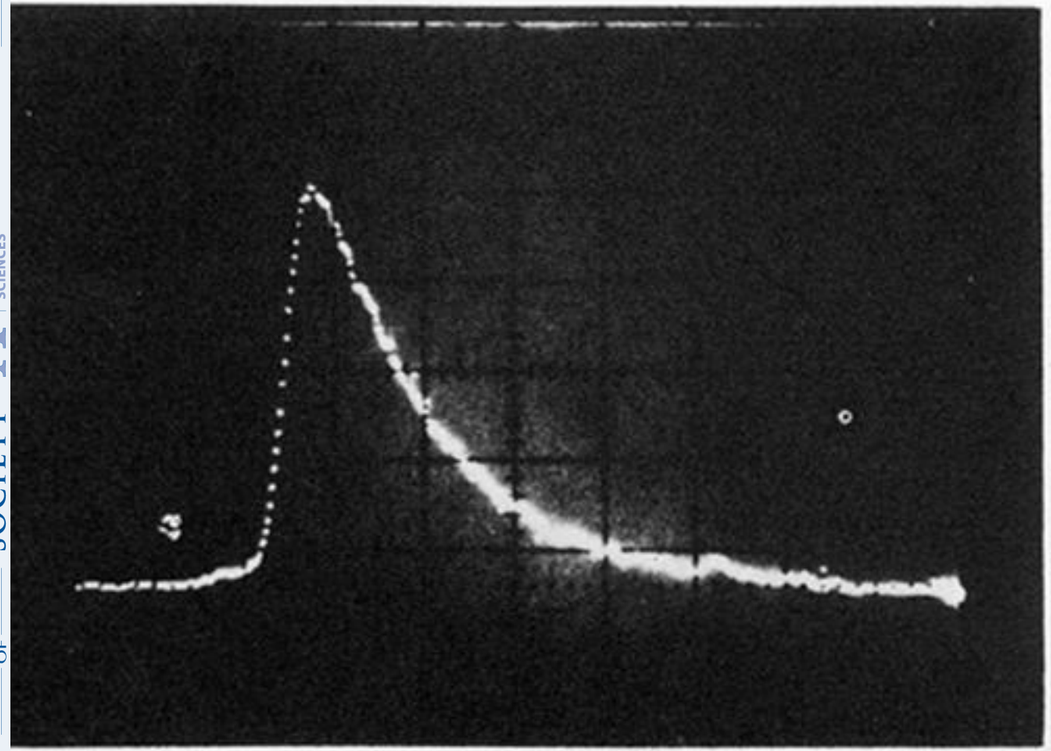
Conclusion

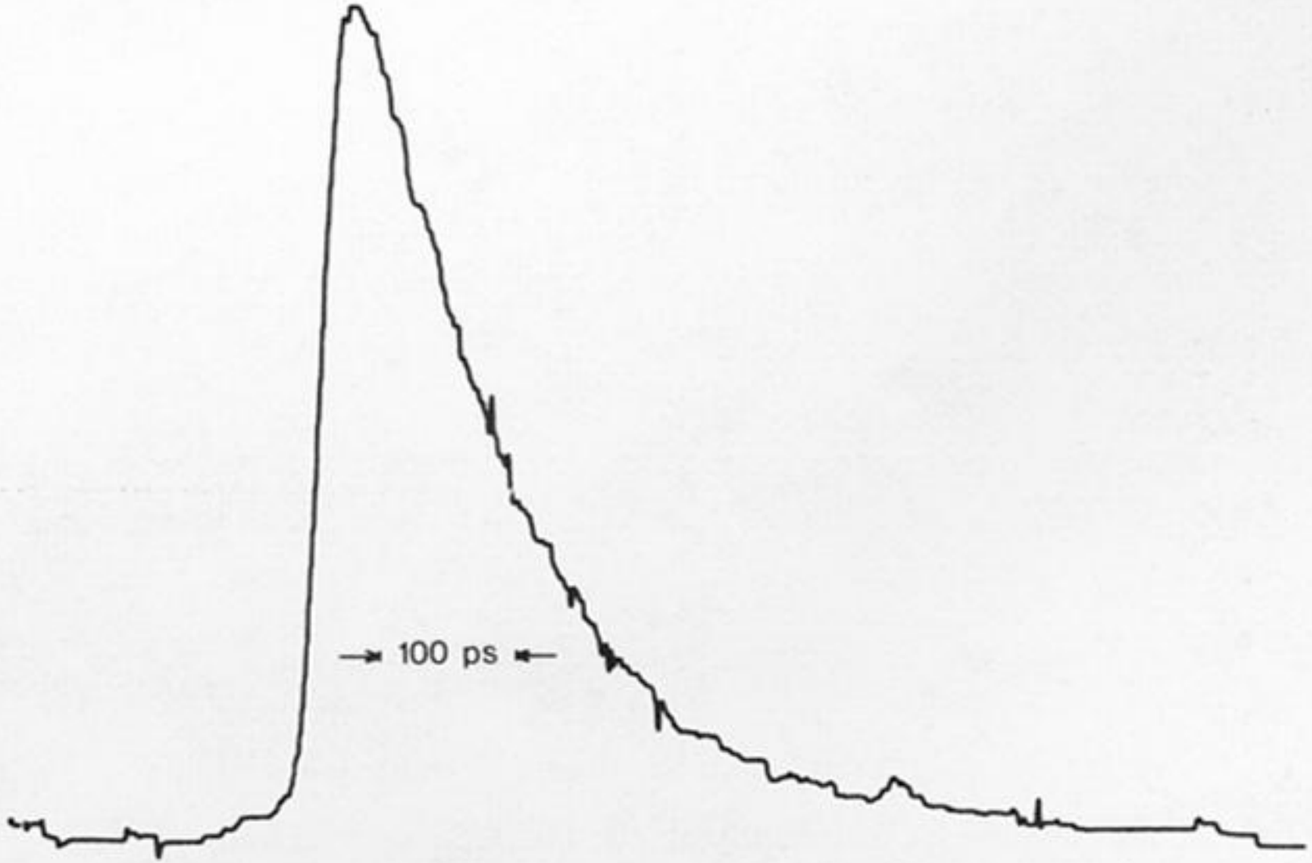
From the results presented in this review, it should be clear that synchronously pumped c.w. dye lasers are convenient sources of picosecond and subpicosecond light pulses over the spectral range of *ca.* 420–835 nm. The shortest duration pulses are produced by increasing the lasing bandwidth or by including a saturable absorber to form a hybrid arrangement. The resulting restriction imposed on tunability is the price paid for obtaining minimum pulse durations. The novel features of the synchroscan camera and mode-locked c.w. dye laser combination are likely to lead to wide application in time-domain spectroscopy. The use of the two-dimensional read-out capability of the Optical Multichannel Analyser to provide the direct display of spatial/spectral information on a picosecond timescale, should further enhance the attractiveness of this technique.

The authors wish to acknowledge the financial support of the Science Research Council and the Paul Instrument Fund.

References (Adams *et al.*)

Adams, M. C. 1979 Ph.D. thesis, University of London.
Adams, M. C., Bradley D. J. & Sibbett W. 1978 In Springer series in chemical physics, vol. 4 (*Picosecond phenomena*) (ed. C. V. Shank, E. P. Ippen & S. L. Shapiro), p. 108. Heidelberg: Springer-Verlag.
Adams, M. C., Bradley, D. J., Sibbett, W. & Taylor, J. R. 1979*a* *Chem. Phys. Lett.* **66**, 428.
Adams, M. C., Sibbett, W. & Bradley, D. J. 1978 *Opt. Commun.* **26**, 273.
Adams, M. C., Sibbett, W. & Taylor, J. R. 1979*b* *Appl. Phys. Lett.* (In the press.)
Ausschnitt, C. P., Jain, R. K. & Heritage, J. P. 1978 Paper J9, 10th Int. Quant. Electron. Conf., Atlanta, May 1978.
Birch, D. J. S. & Birks, J. B. 1976 *Chem. Phys. Lett.* **38**, 432.
Bradley, D. J. 1977 In *Topics in applied physics*, vol. 18 (*Ultrashort light pulses*) (ed. S. L. Shapiro), p. 17. Heidelberg: Springer-Verlag.
Bradley, D. J. 1978 *J. phys. Chem.* **82**, 2259.
Bradley, D. J. & Durrant, A. J. F. 1968 *Phys. Lett.* A**27**, 73.
Bradley, D. J. & Sibbett, W. 1975 *Appl. Phys. Lett.* **27**, 382.
Chan, C. K. & Sari, S. O. 1974 *Appl. Phys. Lett.* **25**, 403.
de Vries, J., Bebelaar, D. & Langelaar, J. 1976 *Opt. Commun.* **18**, 24.
Diels, J. C., van Stryland, E. & Benedict, G. 1978 Paper J8, 10th Int. Quant. Electron. Conf., Atlanta, May 1978.
Eckstein, J. N., Ferguson, A. J., Hansch, T. W., Minard, C. A. & Chan, C. K. 1978 *Opt. Commun.* **27**, 466.
Fehrenbach, G. W., Gruntz, K. J. & Ulbrich, R. G. 1978 *Appl. Phys. Lett.* **33**, 159.
Frigo, N. J., Daly, T. & Mahr, H. 1977 *IEEE J. Quantum Electron.* **QE-13**, 101.
Glenn, W. H., Brienza, M. J. & de Maria, A. J. 1968 *Appl. Phys. Lett.* **12**, 54.
Heisel, F., Miehe, J. A. & Sipp, B. 1979 *Chem. Phys. Lett.* **61**, 115.
Heising, S. J., Jarrett, S. M. & Kuizenga, D. J. 1971 *Appl. Phys. Lett.* **18**, 516.
Heritage, J. P. & Jain, R. K. 1978 *Appl. Phys. Lett.* **32**, 101.
Ippen, E. P. & Shank, C. V. 1975 *Appl. Phys. Lett.* **27**, 488.
Kuhl, J., Klingenberg, H. & von der Linde, D. 1979 *Appl. Phys. Lett.* **25**, 514.
Ruddock, I. S. 1976 Ph.D. thesis, University of London.
Ruddock, I. S. & Bradley, D. J. 1976 *Appl. Phys. Lett.* **29**, 296.
Ryan, J. P., Goldberg, L. W. & Bradley, D. J. 1978 *Opt. Commun.* **27**, 127.
Soffer, B. H. & Linn, J. W. 1968 *J. appl. Phys.* **39**, 5859.
Steinmetz, L. L., Richardson, J. H. & Wallin, B. W. 1978 *Appl. Phys. Lett.* **33**, 163.





IGURE 5. Storage oscilloscope display and chart recorder trace for the fluorescence decay of DSCI in methanol.