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Passively mode locked c.w. dye lasers operating from 490 nm to 800 nm

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Passively mode locked c.w. dye lasers operating from 490 nm to 800 nm

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Résumé. — Les lasers à colorant continu à blocage de mode passif représentent actuellement une importante source d'impulsions optiques femtosecondes accordables dans le spectre visible et proche-infrarouge. Des impulsions aussi courtes que 70 fs ont été obtenues, avec des configurations de cavités à collisions d'impulsions à dispersion optimisée, par l'utilisation de colorants actifs/passifs autres que la combinaison standard Rhodamine 6G et DODCI.

Abstract. — Passively mode locked c.w. dyes lasers now represent an important source of femtosecond optical pulses tunable through the visible and near infra red spectrum. Pulses as short as 70 fs have been obtained from dispersion-optimised CPM cavity configurations using active/passive dyes other than the standard combination of Rhodamine 6G and DODCI.

Passive mode locking is firmly established as a powerful and reliable technique for the generation of femtosecond optical pulses. As well as being proven capable of generating pulses of less than 30 fs duration [1] in dispersion optimised cavities, it has the advantages of high (> 100 MHz) and/or variable repetition rate, low interpulse jitter (< 1 ps) and non-critical adjustment of cavity length. In addition, since it does not require a mode locked pump laser, it is cheaper, easier and potentially more tunable than its synchronously pumped counterparts.

Figure 1 shows the currently reported tuning ranges of passively mode locked c.w. dye lasers. Table I lists the names of the saturable absorbers demonstrated. Apart from the original Rhodamine 6G/DODCI system, all these active/passive dye

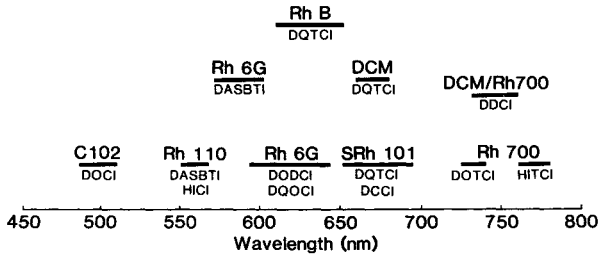

Fig. 1. — Currently available tuning ranges of passively mode locked continuous wave dye lasers. The full names of the saturable absorbers are given in table I.

Table I. — *Saturable absorbers used in c.w. passively mode locked dye lasers.*

DOCI	: 3,3′-diéthyl oxacarbocyanine iodide.
DASBTI	: 2-(p-dimethyl aminostyryl)-benzthiazolylethyl iodide.
HICI	: 1,1′,3,3,3′,3′-hexamethyl indocarbocyanine iodide.
DQOCI	: 1,3′-diethyl 4,2′ quinolyloxacarbocyanine iodide.
DODCI	: 3,3′-diethyl oxadicarbocyanine iodide.
DQTCI	: 1,3′-diethyl-4,2′-quinolylthiacarbocyanine iodide.
DCCI	: 1,1′-diethyl-2,4′-carbocyanine iodide.
DOTCI	: 3,3′-diethyloxatricarbocyanine iodide.
DDCI	: 1,1′-diethyl-2,2′-dicarbocyanine iodide.
HITCI	: 1,1′,3,3′,3′,3′-hexamethylindotricarbo cyanine iodide.

combinations have been developed at Imperial College [2-9]. It should be noted that the Rhodamine 700/HITCI and DOTCI systems were pumped using a krypton ion laser while all the others were argon ion pumped — notably the Rhodamine 6G/Sulphorhodamine 101 and DCM/Rhodamine 700 energy transfer lasers. This technique of using an argon pumped « donor » dye whose fluorescence band overlaps the absorption band of an « acceptor » dye results in a highly efficient dye laser (e.g. [10])

and thus extends the tuning range of argon ion pumped systems into the infra-red. It is interesting to note that the results obtained from the passive mode locking of the DCM/Rhodamine 700 energy transfer dye laser [9] were superior, in terms of minimum achieved pulse duration and amplitude stability, compared to the directly krypton ion pumped Rhodamine 700 laser of reference [2]. The Coumarin 102 laser [8] was pumped by the all lines u.v. output of a Spectra Physics model 171 u.v. enhanced argon ion laser, while the other argon ion pumped systems employed the all-lines visible output of a Spectra Physics model 2020 laser.

Initially, all the new argon ion pumped active/passive dye combinations reported here were demonstrated in a simple linear cavity in which no attempt was made to optimise the intracavity group velocity dispersion. The spectral region from 550 nm to 760 nm was covered using Rhodamine 110, Rhodamine 6G, Rhodamine B, DCM and an energy transfer mixture of Rhodamine 6G and Sulphurodamine 101 as the active media. A broadband dielectric coating of 100 % reflectivity from 510 nm to 780 nm was used for all the cavity mirrors in the configuration shown in figure 2. A dielectric tuning

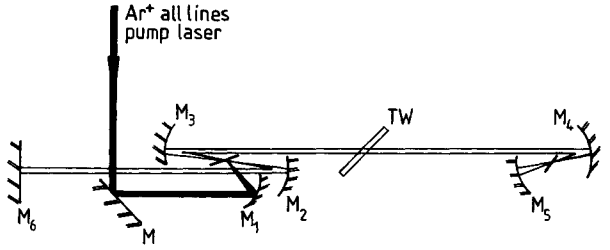

Fig. 2. — Cavity configuration for passively mode locked dye lasers operating from 550 nm to 760 nm.

wedge was used to control the laser wavelength and to provide two output beams. Mirror M_1, of 50 mm radius of curvature, focussed the argon ion pump beam (up to 7.0 W all-lines pump power) into the gain medium, which was a free flowing jet stream of a $\sim 10^{-3}$ M solution of the active dye in ethylene glycol. This concentration was adjusted to yield approximately 90 % absorption of the pump beam. Standard Coherent Radiation Ltd nozzles were used for both the active and passive dye streams. The active folded section comprised two mirrors M_2, M_3, of 100 mm radius of curvature while the passive section consisted of a focussing mirror of 50 mm radius of curvature and a retroreflecting mirror of 25 mm radius of curvature — the cavity parameters being designed in accordance with New's criteria for stable pulse evolution [11]. Pulse durations typically around 250 fs were obtained across the range 550 nm-760 nm, depending on the particular values of the pump power and saturable absorber concentration. Throughout the work presented here, pulse durations have been determined using a standard collinear SHG autocorrelation technique, employing either urea, potassium dihydrogen phosphate or lithium iodate as the doubling crystals and assuming $sech^2$ pulse profiles. The shortest pulses obtained from this cavity, which contained a bandwidth limiting tuning element, dispersive broadband optics and no intentional optimization of the intracavity dispersion, were of ~ 120 fs duration obtained at 667 nm from the Rhodamine 6G/Sulphurhodamine 101 energy transfer laser mode locked with DQTCI [6].

Figure 3a shows the autocorrelation trace of these pulses and figure 3b a typical autocorrelation obtained at 561 nm from Rhodamine 110 mode locked with DASBTI [4]. With all these systems the pulses

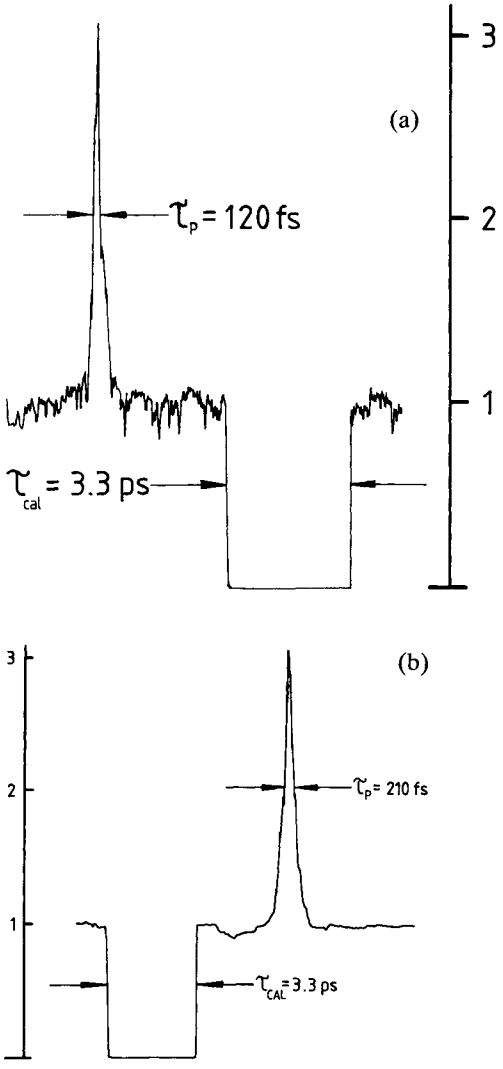

Fig. 3. — a) Autocorrelation trace of pulses of 120 fs duration obtained at 667 nm using the Rhodamine 6G/Sulphurhodamine 101 laser mode locked with DQTCI. b) Typical autocorrelation trace of pulses of 210 fs duration obtained at 561 nm using the Rhodamine 110 laser mode locked with HICI.

were not generally transform limited but exhibited sufficient bandwidth to support pulses of ~ 120 fs duration. Typical average output powers for these systems varied between ~ 20 mW and 140 mW depending on the dye combination and level of pump power. All of these systems exhibited good amplitude stability, as monitored *via* a photodiode (BPW28) and oscilloscope, which is characteristic of passively mode locked systems.

Passive mode locking in the blue-green spectral region was achieved using Coumarin 102 pumped by a u.v. enhanced argon ion laser [8]. A linear four mirror cavity incorporating two retroreflecting folded sections and the same dielectric tuning wedge was adopted. Mode locking was obtained from 487 nm to 508 nm using DOCI as the saturable absorber, the shortest pulses being of 580 fs duration at 498 nm with 8 mW total average output power. Figure 4 shows a typical autocorrelation trace obtained. No attempt was made to optimise the intracavity group velocity dispersion and the pump power was limited to 2.6 W. At this pump level the pulses exhibited sufficient bandwidth to support a pulse of 160 fs duration. With higher pump powers and appropriate dispersion-compensation, this laser should readily generate pulses of the order of 100 fs duration.

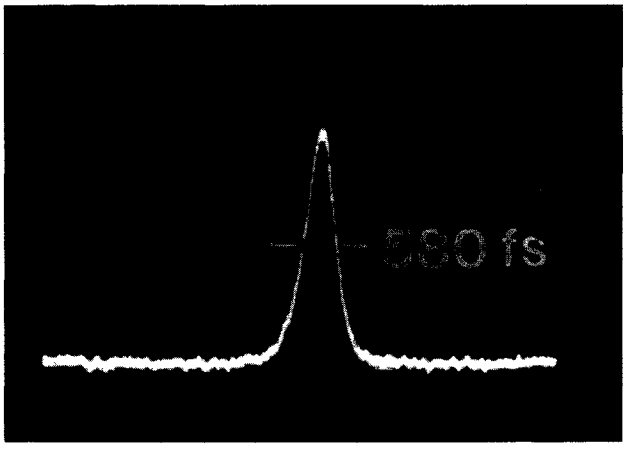

Fig. 4. — Autocorrelation trace of pulses of 580 fs duration obtained at 498 nm using the Coumarin 102 laser mode locked with DOCI.

In order to demonstrate that the new active/passive dye combinations compare favourably with the standard of Rhodamine 6G with DODCI — thus showing that passive mode locking is an attractive source of femtosecond pulses over the visible and near infra-red spectrum (perhaps the most attractive) — some of the new combinations were demonstrated in dispersion-optimised cavities.

Rhodamine 110 mode locked with HICI and DASBTI was examined in both linear and CPM ring configurations [12] which incorporated narrow band single-stack dielectric mirror coatings, prism pairs and no standard bandwidth-limiting tuning elements. Optimisation of the intracavity group velocity dispersion was facilitated by the prism pairs (after Fork *et al.* [13]) and by adjusting the angle of incidence to the narrow band dielectric mirrors (see e.g. [14, 15]). This latter adjustment also permitted some control of the laser wavelength since the mirrors acted as « long wavelength cut-off filters ». Figure 5 shows two of the cavity configurations used.

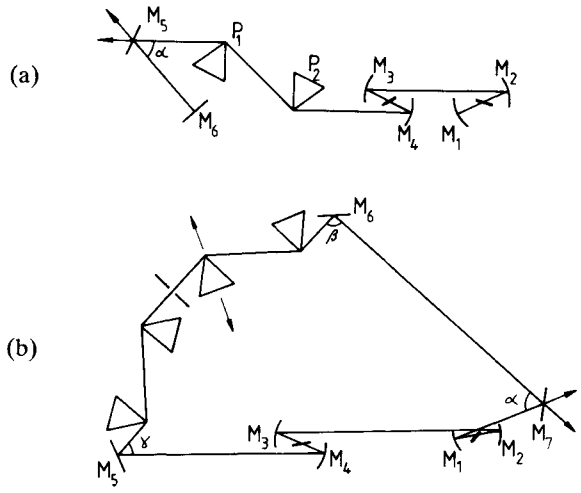

Fig. 5. — a) Schematic of a linear cavity with adjustable intracavity dispersion. b) Schematic of a CPM ring cavity with adjustable intracavity dispersion.

From the linear cavity of figure 5a, transform limited pulses of 120 fs duration were obtained for an angle α of 76° and an HICI concentration of 10^{-3} M. In this instance it was only mirror M_5 which restricted the laser wavelength or contributed to the intracavity dispersion — the other mirrors all being used near normal incidence. It was found that, unless there was some spectral control, the laser wavelength steadily increased with increasing absorber concentration and pulses of duration below ~ 300 fs could not be generated.

The shortest pulses were obtained from the configuration shown in figure 5b. Here the mirror M_5, M_6 and M_7 contributed to both wavelength restriction and intracavity dispersion. Transform limited pulsed of 70 fs duration were obtained at 573 nm with up to 60 mW total average output power for 6.5 W pump power with a 9 ns cavity round trip time and angles α, β, $\gamma = 70°$, 90°, 27° respectively. Figure 6 shows an autocorrelation trace of these pulses and figure 7 shows how the pulse duration varied as the intracavity glass path was adjusted *via* the translation of one of the prisms. The lack of a strong asymmetry in the latter figure is in contrast to the results of Valdmanis *et al.* [1] as is the obser-

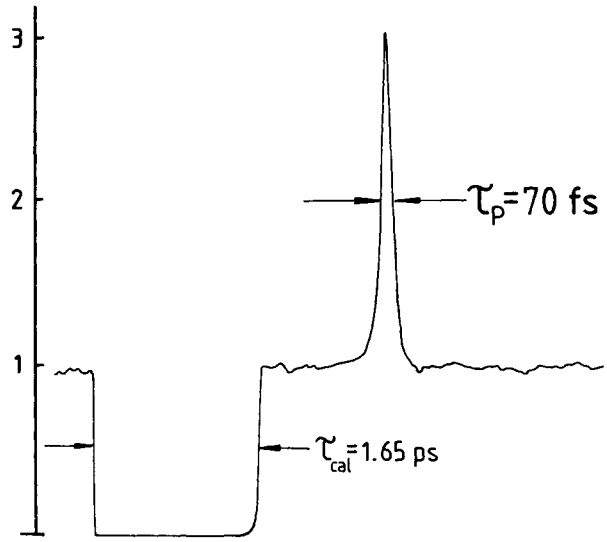

Fig. 6. — Autocorrelation trace of pulses of 70 fs duration obtained at 573 nm from the cavity of figure 5b using Rhodamine 110 mode locked with HICI.

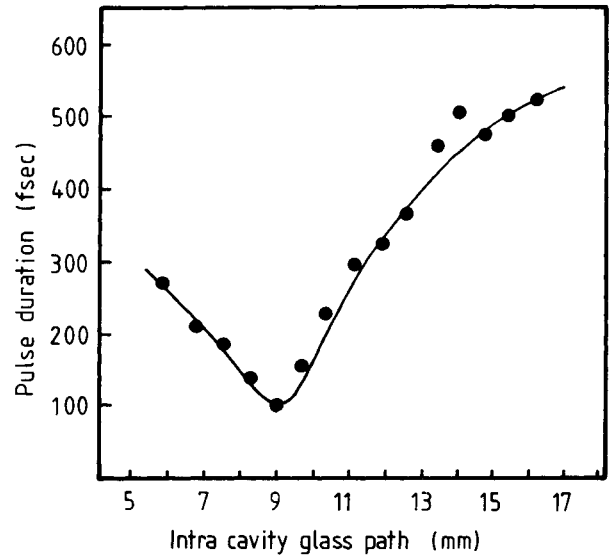

Fig. 7. — Variation of pulse duration with intracavity glass path for the laser described in figure 5b.

vation that the shortest pulses appear to be obtained with the absorber at the focus of the passive folded section. It appears that the strong « soliton shaping » of Martinez *et al.* [16] is not dominant in this laser. With more intracavity glass path than that which yields the shortest pulses, a triple peaked autocorrelation trace appears which is reminiscent of the observations of « high order soliton evolution » by Salin *et al.* [17], though no strong periodic modulation was observed at kHz frequencies as in reference [17]. Recent theoretical work by Avramopoulos and New [18] suggests that our observations can be simulated by a numerical model incorporating saturable absorption and gain, group velocity dispersion, the optical Kerr effect and the chirp arising from time dependant saturation of the absorber. A detailed account of these experimental and theoretical results is under preparation. Using the same active/passive dyes, pulses of 80 fs duration were obtained from a similar cavity in which the prism sequence was omitted — thus only the dielectric mirrors were used to optimise the intracavity dispersion. With the DCM/Rhodamine 700 energy transfer dye laser mode locked using DDCI, a CPM ring cavity was constructed which yielded transform limited pulses as short as 110 fs duration from 742 nm to 754 nm [9]. Again the prism sequence was omitted and so both the laser wavelength and intracavity dispersion were determined by the angles of incidence at the cavity mirrors.

In conclusion, passive mode locking has been demonstrated as an effective technique for generating femtosecond pulses over the visible and near infra red spectrum. It has been shown that pulses of ~ 100 fs duration are obtainable from these new active/passive dye combinations when used in lasers with appropriate optimisation of the intracavity group velocity dispersion. Most, if not all, of these new dye combinations should yield such short pulses and it is highly likely that the preliminary results presented here will be considerably improved upon. All of these dyes will also work in synchronously pumped, hybridly mode locked lasers. Various differences have been noted in comparison with lasers using the standard combination of Rhodamine 6G and DODCI. In particular, the CPM configuration appears to yield shorter pulses than the dispersion-optimised linear cavities ; the lasers need to be constrained to work at the wavelengths which give the shortest pulses and the dependance of the laser performance on the intracavity frequency chirping appears to differ for the case of the dispersion-optimised CPM Rhodamine 110 dye laser. Further investigations of new active/passive dye lasers should afford greater insight into the mechanisms of passively mode locked lasers generally.

Acknowledgments.

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