

AN INVESTIGATION OF FEMTOSECOND OPTICAL PULSE GENERATION

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ABSTRACT

Three new femtosecond laser systems operating in the near infrared part of the spectrum have been developed and characterized. The first two of these systems are passively modelocked dye lasers exploiting energy transfer dye mixtures as the gain media. They enable argon-ion pumped operation tunable from 732nm to 761nm and 783nm to 815nm. The latter tuning range is possible using a new energy transfer dye mixture as the laser gain medium.

The third of the infrared femtosecond laser systems is a c.w. pumped Titanium doped sapphire laser. Pulses of 75ps duration, tunable from 725 to 840nm, have been achieved with active modelocking. Applying the relatively recent modelocking technique using feedback from a nonlinear external cavity subpicosecond pulses have been generated directly from this source for the first time.

An experimental investigation into complex pulse evolutions occurring in a dispersion compensated Rhodamine 110 colliding pulse modelocked (CPM) laser has been carried out. This behaviour has been compared with that of a numerical simulation which takes into account the important amplitude and phase varying processes present in the cavity. Good agreement has been found between experiment and simulation, and this has led to an explanation of the experimentally observed instabilities and satellite pulses.

There has also been a detailed experimental investigation into the pulse formation process in the passively modelocked Rhodamine 110 CPM system. The measurements indicate an initial long delay of about 10^4 cavity round trips before pulse formation begins, and another 10^3 round trips to reach the steady state modelocked regime. The results give an indication of the influence of the spectral filter, and its time dependence, on pulse formation in such laser systems.

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1. INTRODUCTION

Ultrashort light pulses are normally taken to have durations ranging from as long as tens of picoseconds down to a few femtoseconds, and for all practical purposes are generated using mode-locked laser systems. As ultrafast pulse techniques become more flexible, better understood and easier to apply they are being used increasingly in more diverse research areas, for example the investigation of molecular dynamics in photochemistry and photobiology, carrier kinetics in semiconductors, laser fusion, and very high speed electronics and communications. A large number of results in the above research areas may be found in the Proceedings of the Ultrafast Phenomena Conferences [1,2,3,4,5,6].

Due to their large gain bandwidth and their availability for tunable laser action over the entire visible and near infrared spectrum, dyes are important laser media used in the generation of ultrashort laser pulses. The main modelocking techniques applicable to dye lasers (which are reviewed in chapter 2 of this thesis) are synchronous modelocking which involves gain modulation, passive modelocking which employs a saturable absorber, and hybrid modelocking which involves both processes. Passive modelocking when applied to the c.w. dye laser, particularly in the colliding pulse ring configuration, has become established as the source of the shortest optical laser pulses (27fs directly [7] and 6fs after compression [8]). Until fairly recently however this technique was largely restricted to one combination of active and passive dyes and thus to a narrow spectral region

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 - 2 Picosecond Phenomena II, Eds. R.M. Hochstrasser, W. Kaiser, and C.V. Shank, *Springer Series in Chemical Physics* 14 (1980)
 - 3 Picosecond Phenomena III, Eds. Eisinger, R.M. Hochstrasser, W. Kaiser, and A. Laubereau, *Springer Series in Chemical Physics* 23 (1982)
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 - 5 Ultrafast Phenomena V, Eds. G.R. Fleming and A.E. Sigman, *Springer Series in Chemical Physics* 46 (1986)
 - 6 Ultrafast Phenomena VI, Eds. T. Yajima, K. Yoshihara, C.B. Harris, S. Shionoya, *Springer Series in Chemical Physics* 48 (1988)
 - 7 C.V. Shank, R.L. Fork, R.Yen, R.H. Stolen, and W.J. Tomlinson, *Appl. Phys. Lett.* 40, 761 (1982)
 - 8 R.L. Fork, C.H. Brito Cruz, P.C. Becker, C.V. Shank, *Optics Letters* 12, 483 (1987)

around 620nm. The development of other active and passive dye combinations at Imperial College [1,2,3] has extended the technique over most of the visible part of the spectrum. In the near infrared (where the band gaps of many semiconductor materials lie) availability was limited to a Krypton-ion pumped Rhodamine 700 laser [4]. The Krypton ion laser is, however, not a very reliable or stable pump source, and argon ion pumped infrared dyes have short upper state lifetimes making them unsuitable for practical passive modelocking. Both synchronous modelocking and hybrid modelocking have been applied over a broader tuning range, which extends well into the near infrared [e.g. 5], than passive modelocking. Neither of these techniques, however, is capable of achieving the stability, or the shortness of duration of passive modelocking.

It was with the goal of developing argon-ion pumped near infrared passively modelocked c.w. dye lasers that energy transfer dye mixtures have been investigated as suitable active media, and the results are present in chapter 3 of this thesis.

New solid state materials such as Alexandrite and Ti:Sapphire show great promise as ultrashort laser media due to their very large gain bandwidths, Ti:Sapphire, in particular has shown promise as a replacement for dyes in the near infrared part of the spectrum. The shortest pulses obtained from Ti:Sapphire so far (6ps- [6]) have been generated by active loss modulation. Synchronous pumping of Ti:Al₂O₃ [7] has proven less effective due to its small gain cross section and its long upper state relaxation time which implies the need for a large flux for saturation. For these same reasons passive modelocking using a slow saturable absorber is unlikely to be as successful with Ti:Sapphire as it is with dye lasers. In chapter 6 the exploitation of a relatively new modelocking technique which uses a non-linear resonant external cavity to provide feedback into a main mode locked laser cavity, and based on the theoretical and

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- 1 P.M.W. French, J.R. Taylor, *Proceedings of Ultrafast Phenomena V*, ed. G.R. Fleming, A.E. Siegman (Springer-Verlag, Berlin 1986), p. 11
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experimental work of Blow et al. on colour-centre lasers [1,2] has been used to modelock this material and thus enabled the generation of subpicosecond pulses from Ti:Al₂O₃.

Modelocked pulses from various laser sources have been compressed in the fibre-grating pulse compressor (first was [3]) which exploits the nonlinear optical process of self phase modulation in a fibre to spectrally broaden and chirp a pulse combined with negative group velocity dispersion to cancel the chirp and recompress the pulse. Pulses as short as 6fs have been achieved employing higher order phase correction with this technique [4]. If the fibre has negative dispersion then the chirp compensation can occur continuously along it, and soliton propagation can occur. Since a fundamental soliton is a pulse which can propagate long distances without dispersive broadening, this mechanism is of much interest for high bit rate communication purposes [5].

Optical nonlinear processes which can give rise to frequency chirp on ultrashort pulses also arise in passively modelocked dye laser cavities [6,7,8], and intracavity dispersion compensation of the chirp has been used to aid ultrashort pulse generation [9,10,11]. It is the optimisation of this process which enabled the generation of 27fs pulses in [12]. The processes and devices involved are reviewed in chapter 2.

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 - 3 C.V. Shank, R.L. Fork, R. Yen, R.H. Stolen, W.J. Tomlinson, *Appl. Phys. Lett.* 40, 761 (1982)
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 - 10 J.A. Valdmanis, R.L. Fork, J.P. Gordon, *Opt. Lett.* 10, 131 (1985)
 - 11 P.M.W. French, J.R. Taylor, *Opt. Commun.* 61, 224 (1987)
 - 12 C.V. Shank, R.L. Fork, R. Yen, R.H. Stolen, and W.J. Tomlinson, *Appl. Phys. Lett.* 40, 761 (1982)

In using dispersion compensated passively modelocked CPM dye lasers several authors [1,2] reported autocorrelation traces with multiple peaked structures which at the time could not be adequately explained. Since the passively modelocked CPM dye laser is now a basic tool it is important that its operation is well understood, and it is the aim of chapters 4 & 5 of this work to improve the understanding of this type of laser system.

In chapter 4 an experimental investigation has been carried out into the existence in a passively modelocked Rhodamine 110 CPM ring dye laser modelocked with HICI and DASBTI of instabilities and satellite pulses which exist over a certain range of intracavity dispersion. These results are compared with those from a numerical model [3] constructed to simulate the laser, with particular emphasis on gaining understanding of the influence of phase effects in such lasers, and thus to explain the observed and previously incompletely explained instabilities etc.

The timescales associated with the pulse formation in passively modelocked dye lasers are important if one wishes to cavity dump such a laser at a high repetition rate [4] or to use a quasi-cw or pulsed pump source [5], however relatively little attention has been paid to the early transient dynamics of pulse evolution in previous studies of c.w. pumped femtosecond passively modelocked dye lasers. The experiments in references [2,3] give only an estimate of the upper limit on the timescale of the pulse evolution. In chapter 5 I have carried out a detailed experimental investigation of the evolution to steady state of femtosecond pulses in the dispersion compensated passively modelocked Rhodamine 110 CPM ring dye laser with the aim of understanding more fully the timescales involved, and the roles of the spectral filter and phase effects.

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- 1 P.M.W. French, J.R. Taylor, *Optics Communications* **61**, 224 (1987)
 - 2 J.A. Valdmanis, R.L. Fork, *IEEE J. Quantum. Electron.* **QE-22**, 112 (1986)
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2. THE GENERATION AND MEASUREMENT OF ULTRASHORT LIGHT PULSES

2.1 The lasing properties of dyes

The importance of dyes as lasing media arises from their large fluorescence bandwidths which permits widely tunable laser action and the support of ultrashort light pulses.

Dyes may be defined to be organic compounds with a high absorption in the visible part of the spectrum. Such compounds normally possess several conjugated double bonds. The thermal and photochemical stability of dyes is important if they are to be used in lasers.

Thermal stability in dyes is related to the long wavelength limit of absorption. A dye which absorbs in the near infrared has a low excited singlet state and a slightly lower metastable triplet state which has two unpaired electrons. Most dye molecules which reach this state by thermal excitation become very reactive and so will chemically decompose [1]. Because of this dye laser operation at room temperature is impractical at wavelengths much beyond 1 μ m.

The short wavelength limit of dye laser operation is given by the absorption of dyes containing only two conjugated double bonds. These have a long wavelength absorption band at about 220nm, so laser operation shorter than about 250nm is not possible with these compounds. If we try to use compounds with only one double bond (e.g. ethylene) which absorb at about 170nm then 200nm is the limit of laser operation. However at this wavelength photochemical decomposition competes effectively with radiative deexcitation since the photon energy is greater than any single bond in the molecule.

The broad spectra of dyes arises because of the large number of atoms involved which give rise to a large number of normal vibrations of the molecular skeleton. Collisional and electrostatic perturbations from the surrounding solvent molecules broaden these vibrational spectra, and every vibronic sublevel has rotationally excited sublevels. These are greatly broadened by

1 *Dye Lasers* ed. F.P. Schafer

collisions resulting in a quasicontinuum of states on every electronic level.

It can also be shown [1] that for every excited singlet state in a dye there exists a triplet state of lower energy. The overall picture of the eigenstates of a typical dye molecule may thus be represented as in figure 2.1.1.

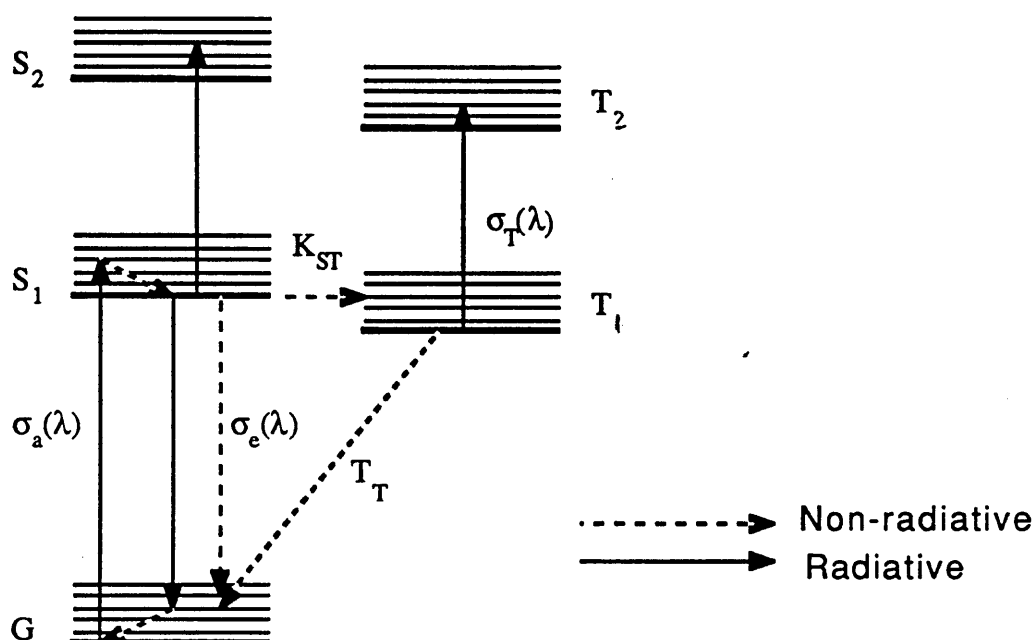


Fig. 2.1.1 Typical dye energy level diagram

Excitation of the dye molecule raises the molecule from the ground state G to the first excited singlet state S_1 . This is characterized by the absorption cross section $\sigma_a(\lambda)$. Often this will leave the molecule in an excited vibronic level of the first excited state from which it will undergo fast ($<ps$) thermalisation to a Boltzmann distribution. Several routes are then possible back to the ground state. It may undergo spontaneous fluorescence (typically after an upper state lifetime of a few nanoseconds) or stimulated emission characterized by cross section $\sigma_e(\lambda)$, or suffer radiationless decay (termed internal conversion) directly to the ground state. Alternatively perturbations (e.g. internally spin-orbit coupling or externally paramagnetic collision partners) may induce a radiationless transition (called intersystem crossing, with rate constant k_{ST}) from the excited singlet state to a triplet state. The transition from the triplet level T_1 back to the ground state (which

corresponds to phosphorescence) is spin-forbidden and has a decay time of a few milliseconds or more. Intersystem crossing can therefore take electrons from the singlet states and inhibit lasing action. Absorption from the first triplet state T_1 to T_2 can overlap the singlet state fluorescence providing a further loss in a laser system. These effects are inhibited in c.w. dye lasers by flowing the dyes fast through the active region or by introducing quenching agents which greatly reduce the time taken to go from T_1 to the ground state.

Other delayed deexcitation routes (via energy transfer to other molecules for instance) can also occur but are usually of lesser significance.

Excitation to higher singlet states (S_2 on up) can occur either directly from the ground state or from the first excited singlet state, however internal conversion back to S_1 is usually very fast ($\sim 10^{-11}$ sec.), and therefore the fluorescence spectra of dyes does not usually depend on pump wavelength.

2.2.The principles of Modelocking

Modelocking is the primary technique used to generate ultrashort light pulses from lasers, and in this section I describe the idea behind it.

A laser consists of a gain medium inside an optical resonator formed by two coaxial mirrors. The range of frequencies over which lasing action can take place in the cavity is determined by the bandwidth over which the optical gain of the medium exceeds the resonator losses. There may be many modes of the resonator which fall inside this lasing bandwidth as represented pictorially in figure 2.2.1 overleaf.

The modes of a resonator formed by two coaxial plane or spherical mirrors has been considered in detail in reference [1]. The principle result necessary in the description of modelocking is that there are a set of longitudinal modes in the resonator with the same energy distribution spatially in the transverse plane but which have different numbers of half-wavelengths along the

1 H. Kogelnik and T. Li, *Appl. Opt.* 4, 1550 (1966)

resonator axis. These longitudinal modes are spaced in frequency by $c/2L$ where c is the velocity of light and L the optical path length of the resonator. The longitudinal mode number is defined to be one more than the number of half wavelengths along the cavity axis.

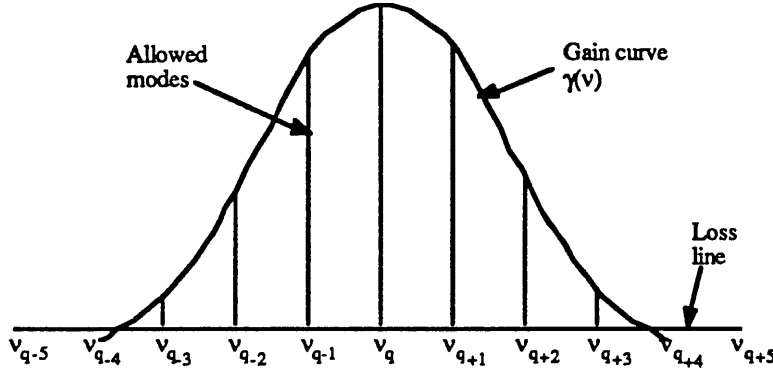


Fig. 2.2.1 Multimode Laser Operation

For each longitudinal mode in the resonator there exists a set of transverse modes which differ from each other by having different energy distributions along the transverse plane. If the resonator has rectangular symmetry this set of energy distributions is given closely by

$$\text{Eqn. 2.2.1 } A(x,y) = A_{m,n} \left(H_m \left(\frac{\sqrt{2} x}{w} \right) H_n \left(\frac{\sqrt{2} y}{w} \right) \right) e^{-\frac{x^2+y^2}{w^2}}$$

where x and y are the transverse coordinates, $A_{m,n}$ is proportional to the field strength of the mode, w is the $1/e$ radius of the fundamental mode ($m=0, n=0$), $H_a(b)$ is the a^{th} order Hermite Polynomial with argument b , and m and n are the transverse mode numbers. As the mode numbers increase the energy is spread out more and more from the axis of the resonator.

For resonators with negligible diffraction losses from the mirrors the resonant frequency ν of a mode (q,m,n) is given [1] by

$$\text{Eqn. 2.2.2 } \nu = \left(\frac{c}{2L} \right) \left((q+1) + \left(\frac{m+n+1}{\pi} \right) \cos^{-1} \sqrt{\left(1 - \frac{L}{R_1} \right) \left(1 - \frac{L}{R_2} \right)} \right)$$

where R_1 and R_2 are the radii of curvature of the two mirrors making up the resonator and other terms are as previously

1 H. Kogelnik and T. Li, *Appl. Opt.* 4, 1550 (1966)

specified.

Normally lasers are operated in the fundamental transverse mode ($m=0$, $n=0$) called TEM_{00} . This may be ensured by inserting an appropriately sized circular aperture into the cavity so that higher order transverse modes suffer appreciably more losses than the fundamental which has the least transverse spatial spread. TEM_{00} operation will be assumed from now on. In this case we see that successive longitudinal modes differ in frequency by $\Delta\nu = c/2L$.

If we consider a laser operating in a number of longitudinal modes which generally will have random amplitude and phase variations (due to mechanical fluctuations in the mirrors, non-linear interactions of modes with the gain medium etc.) then we see that the total field will vary randomly in some uncontrolled way with a characteristic time which is of the order of the reciprocal of the laser bandwidth.

If we somehow fix the relative phases and amplitudes of these modes then the total field will be a well defined function in time. If the n^{th} mode has amplitude E_n , angular frequency ω and phase ϕ then the total field will be given by

$$\text{Eqn. 2.2.3 } E_T = \sum_n E_n e^{i\left(\omega_n\left(t - \frac{z}{c}\right) + \phi_n\right)} + \text{c.c.}$$

If we have equal mode spacing $\omega_n = \omega_0 + n\Delta\omega$ where $\Delta\omega = 2\pi(c/2L)$ and ω_0 is the optical frequency of the laser output then

$$\text{Eqn. 2.2.4 } E_T = e^{i(\omega_0 t - k_0 z)} \sum_n E_n e^{i\left(n\Delta\omega\left(t - \frac{z}{c}\right) + \phi_n\right)} + \text{c.c.}$$

This corresponds to a carrier wave of frequency ω_0 whose envelope depends on the values of E_n and ϕ_n . The envelope travels with the velocity of light and is periodic with period $T = 2\pi/\Delta\omega = 2L/c$. If all ϕ_n and E_n are equal then the result is

$$\text{Eqn. 2.2.5 } E(t) = E_0 e^{i\omega_0 t} \frac{\sin\left(\frac{N\Delta\omega t}{2}\right)}{\sin\left(\frac{\Delta\omega t}{2}\right)}$$

This envelope is a single pulse whose width is roughly the reciprocal of the bandwidth as we would expect from Fourier theory. The time bandwidth product is given by

Eqn. 2.2.6 $\Delta\nu \Delta t > k$

where k is a constant of order unity whose value depends on the exact form of the pulse intensity profile. The parameter $\Delta\nu\Delta t$ is an important measurement of the degree of modelocking of the system. When it equals k then the pulses are (Fourier) bandwidth limited.

In the time domain we may think of modelocking as a pulse bouncing backwards and forwards inside the cavity at the speed of light, and emitting a replica of itself from the cavity at each reflection from the output coupler. Eqn. 2.2.5 is exactly analogous to that pertaining to a diffraction grating except instead of localizing light spatially, modelocking localizes it temporally. Modelocking may be accomplished by several different methods which are discussed in the following sections.

2.3 Active modelocking

(a) Loss and phase modulation

This technique involves an intracavity loss or phase modulator (using acousto-optic or electro-optic effect) driven by an R.F. signal whose period is synchronized to an integral multiple of the cavity round trip time. A good review of the subject is reference [1].

In the frequency domain, due to the loss modulation in the cavity, sidebands to the cavity modes are generated whose frequency coincides with the two neighbouring modes. A result of this interaction between all the modes is that they share the same phase, and thus equation 2.2.3 holds and the cavity is modelocked. In the time domain loss modulation corresponds to a single pulse circulating in the cavity which traverses the synchronized modulator at moments of maximum transmission.

1 A.E. Siegman, D.J. Kuizenga, *Opto-electronics* 6, 43 (1974)

In the case of an electro-optic phase (dielectric constant) modulator light passing through it will be up or down shifted in frequency unless it passes through during the time of the maximum (or minimum) of the dielectric cycle. If the modulator is operated at the synchronous rate this shift will increase in every pass of the cavity until eventually the light frequency falls outside the gain curve of the laser medium. The overall effect of the phase modulator is thus similar to that of the loss modulator except that phase modulation will support two independent pulses in the cavity which will be at slightly different frequencies corresponding to the maximum and minimum of the dielectric cycle. Often bistable operation is observed in such lasers, and the pulses will be slightly chirped. In the frequency domain both these processes correspond to sideband generation from one mode of the laser into the others. In the steady state both AM (loss) and FM (phase) modulators will thus produce the same pulse width, however, the FM case requires a finite laser bandwidth to cause the generation of any pulse at all. This difference is very important in transient modelocking and in attempts to modelock extremely wide linewidth lasers such as CW dye lasers.

Accurate synchronization is essential for successful active modelocking. Modulation frequency tolerances less than 1 KHz out of a modulation frequency of several hundred MHz are typical for CW modelocked Nd:YAG lasers. Wider atomic linewidth systems require proportionately more stringent tolerances. Several schemes have been used to stabilize the modelocker drive frequency, however one of the main problems is drift of the modulators acoustic resonance and the most successful method of stabilizing this is that of reference [1] which controls the RF power driving the modulator (and thus its temperature and resonance frequency) using a feedback signal proportional to the phase difference between the incident and reflected RF power.

If the modulation frequency is the N^{th} harmonic of the axial mode spacing then multiple modelocking occurs, although the pulse repetition rate does not necessarily increase by N . For a fixed modulation depth, an increase in frequency causes stronger compression action (since the modulation extremes are sharper)

1 H. Klann, J. Kuhl, D. Von Der Linde, *Opt. Commun.*, **38**, 390 (1981)

and thus in principle leads to shorter pulses. In multiple modelocking coupling occurs in sets (hypermodes) of axial modes separated by N which may operate separately or together depending on factors such as the positioning of the gain medium in the laser cavity [1].

Electro-optic phase modulators generally use the Pockels effect in LiNbO_3 for the Nd:YAG laser or in GaAs for the CO_2 laser. They are quite easy to construct and operate, requiring only an appropriately cut electro-optic crystal, conducting electrodes and an RF resonant circuit. With several watts of RF power modulation indices of about 0.5 to 1.0 at modulation frequencies of around 1 GHz can be achieved in the visible and near IR. There is normally some optical power loss when a LiNbO_3 crystal or any other phase modulator is put in the cavity. Acousto-optic modulators for AM modelocking employ the resonant light process in materials such as fused quartz for the visible or Ge for the infrared. They require an acoustic transducer bonded to the modulator substrate, with good acoustic impedance matching between the two. The acoustic resonances of the crystal makes frequency tuning difficult. Acoustic losses and heating problems reduce modulation indices to less than 0.5, usually at frequencies of about 200 MHz. However good quality fused quartz is an extremely low loss optical element in the laser cavity.

A laser which is strongly inhomogeneous (e.g. visible gas lasers) is able to oscillate simultaneously on multiple axial modes which burn holes in the spectral gain profile of the laser as shown below.

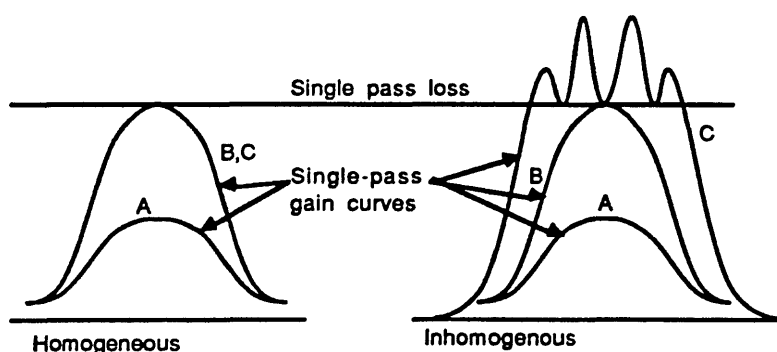


Fig. 2.2.2 Gain curves of laser media (A) below threshold (B) on threshold (C) above threshold

1 M.F. Becker, D.J. Kuizenga, A.E. Siegman, *IEEE J. Quantum Electron*, QE-8, 687 (1972)

Normally these modes have random phase relationships. Mode coupling over the whole spectral width of the laser is achieved by providing just sufficient modulation depth via the active modulator so the weak sidebands from each oscillating mode have energy injected into them from adjoining modes. The expected pulse width is just that of eqn. 2.2.6 where $\Delta\nu$ is the laser oscillation bandwidth. Theoretical studies of active modelocking using inhomogeneous Doppler broadened transitions have used linearised solutions to the problem [1,2,3]. More detailed non-linear calculations using a coupled mode equation approach and assuming that the modes saturate independently have also been tried [4,5].

A homogeneous laser (e.g. CW Nd:YAG laser, low pressure CO₂ laser, dye laser) in the absence of mode coupling oscillates in only the lowest loss mode - at least in principle, however spatial hole burning effects, and various optical nonlinearities, do enable other cavity modes to extract gain. In the homogeneous case modelocking is a competition between the sideband generating active modulator which pushes energy outwards from the gain peak and the limited bandpass laser gain medium which tries to narrow the spectral width and broaden the pulse. The analysis of homogeneously broadened lasers via coupled mode equations is complicated because they do not saturate independently and a large number of coupled axial modes are generated. References [6,7] show an approach which works in the time domain. They assume that a short pulse (temporally Gaussian in shape) exists in the cavity. Such a pulse is followed once around the cavity through the active medium and the modulator and a self consistent solution is found analytically (i.e. no change in the complete round trip). This is similar to the analysis of Cutler's microwave regenerative pulse generator [8]. Loss and phase modulation have been around for quite a few years and so the theories have been examined exhaustively.

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- 1 M. DiDomenico, *J. Appl. Phys.* **35**, 287 (1964)
 - 2 A. Yariv, *J. Appl. Phys.* **36**, 388 (1965)
 - 3 N.H. Crowell, *IEEE J. Quantum Electron.* **QE-1**, 12 (1965)
 - 4 S.E. Harris, O.P. McDuff, *IEEE J. Quantum Electron.* **QE-1**, 245 (1965)
 - 5 O.P. McDuff, S.E. Harris, *IEEE J. Quantum Electron.* **QE-3**, 101 (1967)
 - 6 A.E. Siegman, D.J. Kuizenga, *Appl. Phys. Lett.* **14**, 181 (1969)
 - 7 D.J. Kuizenga, A.E. Siegman, *IEEE J. Quantum Electron.* **QE-6**, 691 & 709 (1970)
 - 8 C.C. Cutler, *Proc. IRE* **43**, 140 (1955)

For appropriate references see New's review [1].

The problem of transient modelocking has been investigated [2], and is important for pulsed laser systems. Essentially the process involves the build up from random noise axial modes across the full laser bandwidth. Spectrally this noise may start very wide but it narrows gradually upon repeated transits as the axial modes near line centre grow most rapidly. At the same time a gradually broadening local coherence is imposed within the spectrum by the sidebands produced by the modulator. When these oppositely acting processes meet steady state modelocking is achieved and the laser output is a fully coherent transform limited pulse. This process has been observed in the repetitively Q-switched Nd:YAG laser [see 3, pp. 54]. Increasing modulation depth or time allowed for build up narrows the modelocked pulses in time, narrows the frequency spectrum and increases the coherence of the pulses.

Dye lasers have the widest bandwidths - typical is Rhodamine 6G which has a linewidth of 10^{13} Hz. They also have low pump power thresholds (.1 - 1W range typically). Thus they are able to support very short pulses (< 1ps). Active modelocking of Rh 6G [4,5] produced pulsewidths of 50-100ps only after overcoming considerable difficulties. The major difficulty in obtaining very short pulses using active modelocking is that the pulse must circulate the cavity many times to reach steady state e.g. for 100 MHz modulation frequency, modulation depth 0.5 it takes about 0.15s to get a 1ps pulse [3]. The circulating pulse and modulator must stay perfectly in phase for this length of time which requires stability to 1 part in 10^8 which is extremely difficult to obtain. Another problem arises because the linewidth is so broad that it allows separate pulses to build up in different parts of the spectrum. Variable group velocity means these pulses arrive at separate times causing satellite pulsing [3].

In He:Ne and Nd:YAG lasers the average power is much the same for the modelocked and non modelocked cases. The argon ion and actively modelocked dye laser show a reduction in average power

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- 1 G.H.C. New, *Rep. Prog. Phys.* **46**, 877 (1983)
 - 2 D.J. Kuizenga, D.W. Phillion, T. Lund, A.E. Siegman, *Opt. Comm.* **9**, 221 (1973)
 - 3 A.E. Siegman, D.J. Kuizenga, *Opto-electronics* **6**, 43 (1974)
 - 4 A. Dienes, E.P. Ippen, C.V. Shank, *Appl. Phys. Lett.* **19**, 258 (1971)
 - 5 D.J. Kuizenga, *Appl. Phys. Lett.* **19**, 260 (1971)

when modelocked. Some causes of this are as follows [see 1]. (1) If the upper state lifetime of the laser transition is less than the round trip time of the cavity then an appreciable fraction of energy will be lost by spontaneous emission between modelocked pulses. This effect is reduced by having the active medium in the centre of the cavity. (2) If the lower state lifetime is longer than the pulsewidth then it will fill up as the pulse passes, reducing the population inversion and thus the energy extraction. This saturable gain effect is in fact essential for pulse formation using saturable absorbers. (3) If the upper and lower states have different degeneracies then the lower level may be bottle-necked as the pulse passes i.e. the pulse can extract energy only until $N_1/g_1 = N_2/g_2$.

(b) Synchronous pumping

This active modelocking technique involves modulating the gain of the laser by pumping with another modelocked source of accurately matched cavity length. Other techniques also making use of a matched master laser are 'synchronous loss modulation' where a loss element in the slave laser is controlled by the master laser (perhaps via an electrical signal in a semiconductor device) and 'double modelocking' where the saturable absorber of the master laser is the gain component of the slave.

In synchronous gain modulation the leading edge of the pulse in the slave laser is suppressed by arranging the timing so that the rising gain in the active medium preferentially amplifies its peak. If this pulse has enough intensity it will deplete the gain and so the trailing edge will also be suppressed, leading to pulse shortening. If the pump pulse is very long then the gain may recover after the passage of the slave laser pulse and amplify other satellite pulses.

Compared with passive modelocking (covered in the next section) synchronous pumping has the advantage that the tunability is not restricted by a saturable absorber bandwidth (almost any laser dye may be synchronously modelocked), energy conversion efficiencies are higher than in the passive case and it is easy to synchronize such a laser to other equipment via the RF signal which drives the mode locker of the pump laser (synchronization is

1 D.J. Kuizenga, *Appl. Phys. Lett.*, **19**, 260 (1971)

more difficult for passive modelocking). However the pulses are not as short as in the passive case and pulse structure critically depends upon accurate matching of slave and master cavity lengths. State of the art femtosecond pulses may be obtained from synchronous modelocking followed by a fibre-grating pulse compressor.

A typical synchronously pumped system is the Ar⁺/Rh 6G combination which has produced subpicosecond pulses. An ultra-stable RF frequency stabilizer to supply the active modulator is essential and as in section 2.2(a) above a large modulation depth is necessary. There are various stabilization systems to control the optical length of the dye laser cavity in relation to the pump laser cavity and thus obtain optimum performance - a common one maximizes the second harmonic of the synchronously modelocked dye laser [e.g. 1]. Many experiments show how pulse characteristics change with cavity mismatch (pump period minus cavity period) (e.g. ref [2,3]). For long cavities the lasers operation is stable but the pulses are like bursts of noise. For dye laser cavities shorter than the pump cavity the power drops rapidly, the pulses broaden and laser operation becomes increasingly unstable. Reference [3] states the experimental result that optimum performance is at the point of exact cavity match plus 2 μ m with an experimental error of 2 μ m.

The theory of synchronous modelocking may be treated using the self reproducing profile approach as used for other active modelocking techniques, however the results are not as good. An alternative stepping model approach allows steady state pulse profiles to be obtained without approximation [4,5,6,7]. The results obtained [6] suggest that even if the practical sources of instability were eliminated the synchronously modelocked laser would still show jitter between it's pulses because the pulse

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- 1 S.R. Rotman, C.B. Roxlo, O. Bebelarr, T.K. Yee, M.M. Salour, *Ultrafast Phenomena II*, 50 (1980)
 - 2 P.G. May, W. Sibbet, J.R. Taylor, *Appl. Phys.* **26**, 179 (1981)
 - 3 P.G. May, W. Sibbet, K. Smith, J.R. Taylor, J.P. Wilson, *Opt. Commun.* **42**, 285 (1982)
 - 4 J.M. Catherall, G.H.C. New, P.M. Radmore, *Optics Letters* **7**, 319 (1982)
 - 5 G.H.C. New, L.A. Zenteno, P.M. Radmore, *Optics Commun.* **48**, 149 (1983)
 - 6 G.H.C. New, J.M. Catherall, *Ultrafast Phenomena V*, Eds. G.R. Fleming and A.E. Sigman, *Springer Series in Chemical Physics* **46** (1986)
 - 7 G.H.C. New, J.M. Catherall, *IEEE J. Quantum Electron.* **QE-22**, 1593 (1986)

evolution never reaches a steady state. This result arises because the authors have taken into account the stochastic nature of the low level spontaneous in the laser cavity, and found that the conditions for eliminating large scale fluctuations and for obtaining short pulses are mutually exclusive.

Normally non transform limited pulses of about a picosecond are produced from synchronously modelocked dye lasers (e.g. 1). It has been found that the pulse duration varies as the square root of the pump pulse duration and as the square root of the bandwidth as determined by an intracavity spectral filter [2]. The pulse duration has also been found to decrease with increasing pump power [3].

2.4 Passive Modelocking

(a) The basic technique

The process of passive modelocking involves placing a saturable absorber near one end mirror of the laser cavity. The saturable absorber is usually an organic dye, however colour centre crystals [4] and MQW semiconductor materials [5] have also been used. The technique is most commonly used in CW dye lasers (first was [6]) but has also been used in solid state systems (e.g. Nd:Silicate, Ruby, Nd:Phosphate glass etc.), flashlamp pumped dye lasers and some gas lasers (e.g. He:Ne). It has the advantage of simplicity and is extremely successful in generating subpicosecond pulses. The technique is reliable in continuous and quasi-continuous lasers but in giant pulse systems the output has stochastic features.

Because of the very short pulses produced by passive modelocking many thousands of cavity modes must be involved and so examination in the time domain is more successful than the frequency domain analysis used to explain active modelocking. In

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- 1 P.G. May, W. Sibbett, J.R. Taylor, *Appl. Phys.* **B26**, 179 (1981)
 - 2 C.P. Ausschnitt, R.K. Jain, *Appl. Phys. Lett.* **42**, 727 (1978)
 - 3 R.K. Jain, C.P. Ausschnitt, *Opt. Lett.* **2**, 117 (1978)
 - 4 K. Smith; PhD Thesis, University of London (1984)
 - 5 Y. Silberberg, P.W. Smith, D.J. Eilenberger, D.A. Miller, A.C. Gossard, W. Wiegmann, *Opt. Lett.* **9**, 507 (1984)
 - 6 E.P. Ippen, C.V. Shank, A. Dienes, *Appl. Phys. Lett.* **21**, 348 (1972)
 - 1 E.M. Garmire, A. Yariv, *IEEE J. Quantum Electron.* **QE-3**, 222 (1967)

early theoretical work [1,2] the pulse duration was found to be limited by the aperture time of the absorbing dye which acts like an intensity triggered optical switch, and this is found experimentally to be true in solid state lasers [3,4].

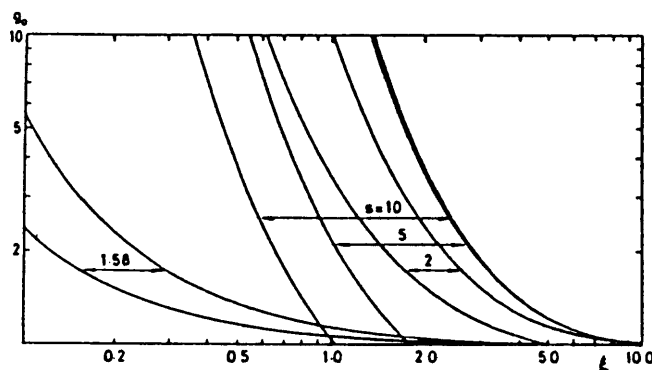


Fig. 2.3.1 Stability diagram for passive modelocking

In homogeneously broadened dye lasers the pulse duration may be much shorter than the recovery time of the saturable absorber, and relatively insensitive to it. The process responsible for this has been analysed by New [5,6] and Haus [7]. The nonlinear processes of both saturable absorption in the passive medium and saturable gain in the active medium are found to play an important role in passive modelocking of dye lasers. The leading edge of a pulse present in the cavity is suppressed by absorption in the saturable absorber before it saturates and the trailing edge is suppressed by gain saturation in the lasing medium (i.e. the peak of the pulse has taken away the gain before it can amplify the trailing edge). According to New [6] the necessary condition for this action is that the absorption saturates before the gain, and the gain does not fully recover between pulses. From New's analysis a stability diagram such as fig. 2.3.1 above can be drawn up. There are three important quantities, the 'S' parameter which is the ratio of

- 1 E.M. Garmire, A. Yariv, *IEEE J. Quantum Electron.* **QE-3**, 222 (1967)
- 2 D.J. Bradley, G.H.C. New, S.J. Caughey, *Opt. Commun.* **2**, 41 (1970)
- 3 E.G. Authurs, D.J. Bradley, A.G. Roddie, *Appl. Phys. Lett.* **23**, 88 (1977)
- 4 E.G. Authurs, D.J. Bradley, P.N. Puntembaker, I.S. Ruddock, T.J. Glynn, *Opt. Commun.* **12**, 360 (1974)
- 5 G.H.C. New, *Opt. Commun.* **6**, 188 (1972)
- 6 G.H.C. New, *IEEE J. Quantum Electron.* **QE-10**, 115 (1974)
- 7 H.A. Haus, *IEEE J. Quantum Electron.* **QE-11**, 736 (1974)

effective absorption cross section to effective gain cross section, the ' ζ ' parameter which is the ratio of cavity round trip time to active medium relaxation time and g_0 the small signal gain of the system (i.e. pump power in practice).

Although New's rate equation theory gives a clear picture of pulse formation from the initial fluctuations in the cavity and predicts the condition under which it will occur it does not set a lower limit on the pulse width (and so no upper on the spectral width). It thus makes no predictions about the shape or duration of steady state pulses formed in a CW dye system. Haus [1] applied a self reproducing profile technique which included a bandwidth limiting term to the problem of a passively modelocked laser with a 'slow' absorber (long recovery time). The pulse shape derived for a parabolic net gain curve $g_T(U)$ (i.e. weak small signal absorption of saturable absorber) was of the form

$$\text{Eqn. 2.3.1 } I(t) = I_0 \operatorname{sech}^2 \left(\frac{t}{\tau_p} \right)$$

where τ_p is the pulse width, inversely proportional to the bandwidth, the pulse energy and the square root of the small signal loss in the saturable absorber. For a stronger 'loading' of saturable absorber the net gain curve $g_T(U)$ becomes sharper than parabolic giving pulses which have sharper peaks than hyperbolic secant but which also have more energy in their tails (for the same full width at half maximum). This distribution of energy in the tails is not always desirable and a suggested way around it was to use several saturable absorbers with an appropriate choice of saturation energies and small signal losses to synthesize a more parabolic dependence of the net gain factor g_T on U . The theoretical approach by Haus does however involve some approximations whose validity has not been proven for dye lasers with 'soft' gain media that saturate easily.

Experimentally the first CW passively modelocked dye laser [2] used Rhodamine 6G pumped by the argon ion laser 514nm line as the active medium and DODCI as the passive medium, both in glass cells. Both active and passive media were put in folded section to increase the energy densities in them during lasing action (this

1 H.A. Haus, *IEEE J. Quantum Electron.* QE-11, 736 (1974)

2 E.P. Ippen, C.V. Shank, A. Dienes, *Appl. Phys. Lett.* 21, 348 (1972)

increases the pump efficiency of the system). The pulses generated were 1.5ps in duration (assuming Gaussian profile). Later a thin saturable absorber cell ($< c\tau_p$ thick) in contact with one of the end mirrors was used to produce pulses of 0.3ps from 598 to 615nm [1] which were transform limited - the bandwidth was limited by an intracavity prism used as a tuning element. This improved performance has been explained as due to an effective tripling of absorber cross section due to the coherent superposition of incident and reflected portions of the pulse (see CPM section for further explanation). Further advances using a linear cavity with the same dye combination reduced the pulse width down to 120fs [2].

(b) Colliding Pulse Modelocking (CPM)

In the colliding pulse regime there are two counterpropagating pulses in a ring cavity which contains dye jets for the active medium and saturable absorber. The two pulses collide in the absorber jet since this involves the least loss. In the absorber medium the pulses coherently interact to create a transient grating in the molecular populations (effectively spatial hole burning) [3,4], which reduces the energy needed to saturate the absorber by about 1.5 [3]. Because there are two pulses involved this increases the effective absorber cross section (and thus the 'S' parameter) by 3, and this should enable the generation of somewhat shorter pulses [5]. There is also an improvement in the modelocking stability brought about by the CPM interaction. The process does however only work well if the absorber jet is narrower than the pulses interacting in it. A thicker jet reduces the effectiveness of the CPM mechanism e.g. an absorber twice as thick as the pulses enhances the effective cross section by only 1.25 [3]. By having the gain medium in the CPM ring cavity a quarter of the way around from the absorber medium the pulses will arrive at the active medium equally spaced in time, and thus both pulses will see the same saturable gain. This is the main advantage of the

-
- 1 I.S. Ruddock, D.J. Bradley, *Appl. Phys. Lett.* **29**, 296 (1976)
 - 2 J.-C. Diels, J. Menders, H. Sallaba, *Picosecond Phenomena II* (see [2])
 - 3 M.S. Stix, E.P. Ippen, *IEEE J. Quantum Electron.* **QE-19**, 520 (1983)
 - 4 D. Kuhlke, W. Rudolph, B. Wilhelmi, *Appl. Phys. Lett.* **42**, 325 (1982)
 - 5 H.A. Haus, *IEEE J. Quantum Electron.* **QE-11**, 736 (1974)

ring cavity over the contacted cell linear cavity [1] which although it makes use of the coherent enhancement of saturable absorption has only one pulse saturating both gain and absorber.

The first experimental pulses of 90fs were produced using the CPM technique [2] with an argon ion pumped Rh 6G/DODCI dye laser combination. The saturable absorber jet stream was about 10 μ m thick. There was no tuning or bandwidth limiting element and the laser ran at 627nm. By changing the dielectric mirrors in the cavity pulses down to 65fs were produced. Further empirical optimizations of critical cavity parameters in similar systems produced pulses of 55fs duration [3].

A variation on the CPM ring cavity is the antiresonant ring [4] which gives similar results but is more critical in alignment.

The basic CPM mechanism has been shown theoretically [5] to produce pulses which are chirped according to the amount of detuning of pulse frequency away from the centre frequency of the saturable absorber. The improvements in pulse duration mentioned above were later shown to be due to fortuitous cancellation of this chirp by balancing the total self phase modulation, saturation chirp and group velocity dispersion in the cavity. The shortest pulses from a laser cavity, of 27fs [6], were obtained by using methods to achieve variable group velocity dispersion in the cavity. See section 2.5 for more detail. In the same paper the authors report pulses of 33fsec from a similarly dispersion compensated linear cavity and conclude that the colliding pulse mechanism is not essential for generating the shortest pulses but does have a useful stabilizing effect.

Compared with other techniques for producing ultrashort pulses passive modelocking has the advantages of producing the shortest pulses with high stability, low interpulse jitter and a well defined sech² profile. It is also cheaper than active techniques since no modelocked pump laser is required. Until recently it had the disadvantage of lack of tunability since only the Rh 6G/DODCI dye combination had been successfully used. The technique has now

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- 1 I.S. Ruddock, D.J. Bradley, *Appl. Phys. Lett.* **29**, 296 (1976)
 - 2 R.L. Fork, B.I. Greene, C.V. Shank, *Appl. Phys. Lett.* **38**, 671 (1981)
 - 3 A.M. Weiner, PhD Thesis, MIT Cambridge Mass. (1984)
 - 4 H. Vanherzeele, J.-C. Diels, R. Torti, *Opt. Lett.* **9**, 549 (1984)
 - 5 D. Kuhlke, W. Rudolph, B. Wilhelmi, *Appl. Phys. Lett.* **42**, 325 (1982)
 - 6 J.A. Valdmanis, R.L. Fork, J.P. Gordon, *Opt. Lett.* **10**, 131 (1985)

been extended to other active passive combinations [1] which cover most of the visible and near infrared spectral range.

2.5 Hybrid Mode Locking

This is a combination of both active and passive modelocking. In a typical configuration the dye laser is actively modelocked (almost always via synchronous pumping) but contains a saturable absorber in the cavity which has the effect of reducing the pulse wings and giving pulse durations up to an order of magnitude shorter than with active modelocking alone. It was first demonstrated [2] having both active and passive dyes (Rh 6G/DODCI) flowing in a single jet stream. The pulses produced were of 0.5 to 1ps duration. Other active dyes have been used in similar systems. Cavities with two jets, using CPM [3,4], and with intracavity adjustable group velocity dispersion have also been used for hybrid modelocking and these produce shorter pulses - down to less than 100fs [5].

The mechanism of hybrid modelocking is not well understood. It has been found experimentally that even weak solutions of saturable absorber greatly improve synchronously pumped dye lasers but this has not shown up in theoretical considerations. When passive mode locked systems are synchronously pumped the resulting hybrid system is found to give similar pulse durations but to be less stable. The performance of hybrid systems is found to be more stable with respect to larger cavity mismatches (up to 40 μ m) than for solely synchronously modelocked systems and so the passive modelocking mechanism is probably the dominant effect. However systems which have cavity lengths much longer than the active dye's recovery time, and those which use saturable absorbers with lifetimes much longer than the active mediums lifetime can not be purely passively modelocked but will work in a hybrid system. Simplistically it may be thought that the gain is

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- 1 P.M.W. French, PhD Thesis, University of London (1987)
 - 2 C.V. Shank, E.P. Ippen, *Appl. Phys. Lett.* **25**, 373 (1974)
 - 3 J. Dobler, H.H. Schultz, W. Zinth, *Opt. Commun.* **57**, 407 (1986)
 - 4 P.G. May, W. Sibbett, J.R. Taylor, *Appl. Phys. B* **26**, 179 (1981)
 - 5 J. Dobler, H.H. Schultz, W. Zinth, *Opt. Commun.* **57**, 407 (1986)

'turned off' between pulses in those systems reducing the probability of multiple pulses occurring and thus increasing stability. Theoretically the pulse evolution time of hybrid systems is expected to be longer than that of purely synchronously modelocked systems by about an order of magnitude [1]. This implies increased sensitivity to mechanical instabilities.

The disadvantage of hybrid modelocking is its inferior pulse quality and stability when compared with purely passive systems, however it can be used over a wider range of active and passive dyes and it can easily be synchronized to other systems. Hybrid modelocking is capable of producing pulse durations almost as short as purely passively modelocking (e.g. 29fs [2])

2.6 Group Velocity Dispersion and Self Phase Modulation effects

The often conflicting roles of group velocity dispersion (GVD) and self phase modulation (SPM) have become important for the generation and measurement of hypershort optical pulses. The shortest pulses from a laser cavity (27fs [3]) were generated by balancing intracavity SPM and GVD, saturable absorption and saturable gain. The shortest optical pulses ever (6fs [4]) were generated using controlled GVD and SPM in an optical fibre [5]. GVD may be controlled using diffraction gratings [6], prisms [3,7], interferometers [8] and glass/quartz elements to provide positive dispersion. SPM arises from the non-linear interaction of the light intensity with optical media.

(a) intracavity

After the initial demonstration of the CPM ring laser several similar systems were built, and it was found that to produce the

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- 1 D. Kuhlke, U. Herpers, D. von der Linde, *Appl. Phys. B* **38**, 233 (1985)
 - 2 H. Kubota, K. Kurokawa, M. Nakazawa, *Opt. Lett.* **13**, 749 (1988)
 - 3 J.A. Valdmanis, R.L. Fork, J.P. Gordon, *Opt. Lett.* **10**, 131 (1985)
 - 4 R.L. Fork, C.H. Brito Cruz, P.C. Becker, C.V. Shank, *Optics Letters* **12**, 483 (1987)
 - 5 B. Nikolaus, D. Grischkowsky, *Appl. Phys. Lett.* **43**, 228 (1983)
 - 6 E.B. Treacy, *IEEE. J. Quantum Electron.* **QE-5**, 454 (1969)
 - 7 R.L. Fork, J.P. Gordon, *Opt. Lett.* **9**, 153 (1984)
 - 8 F. Gires, P. Tournois, *C.R. Acad. Sci. Paris* **258**, 6112 (1984)

shortest pulses there should be no bandwidth limiting components in the cavity, and the choice of dielectric mirrors was important. Some experiments revealed that the pulses from CPM lasers were 'down chirped' [1] and the pulse duration could thus be reduced by introducing positive GVD via an adjustable length of glass in the cavity. The net negative chirp in the uncompensated cavity was due [2] to the nonlinear absorption in the saturable absorber jet which gave negative SPM. Some other experiments revealed that there was net positive dispersion in the uncompensated cavity. It was proposed [3] that 'soliton shaping mechanisms' resulting from the interaction of (positive) SPM (due to the fast optical Kerr effect in the dye solvent) and (negative) GVD (from dielectric mirrors etc.) produced the shortest and most stable pulses.

In passively modelocked dye lasers there are two main causes of SPM. There is negative SPM due to off resonant saturation of the absorber which gives a 'down chirp' and positive SPM due to the optical Kerr effect in the solvents which gives an 'up chirp'. The self-phase modulation arising from gain saturation is in general considered to be much less significant than that arising from absorber saturation due to tighter focusing employed in the passive section of the laser.

In the optical Kerr effect the interaction of light produces a change Δn in the refractive index of the medium through which it travels according to

Eqn. 2.6.1 $\Delta n = n_2 I$

where n_2 is a medium dependent coefficient and I the incident intensity. There is thus an intensity induced phase change given by

Eqn. 2.6.2 $\Delta\phi = \frac{2\pi}{\lambda} L \Delta n = \frac{2\pi}{\lambda} L n_2 I$

where L is the length of material. If the intensity varies with time then the phase change will be a function of time and so there will be a frequency chirp $d\omega$ given by

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- 1 W. Dietel, J.J. Fontaine, J.-C. Diels, *Optics Lett.* **8**, 4 (1983)
 - 2 J.-C. Diels, J.J. Fontaine, I.C. McMichael, B. Wilhelmi, W. Dietel, D. Kuhlke, *Sov. J. Quantum Electron.* **13**, 1562 (1983)
 - 3 O.E. Martinez, J.P. Gordon, R.L. Fork, *Ultrafast Phenomena IV* (see [4])

$$\text{Eqn. 2.6.3 } d\omega = \frac{d\phi(t)}{dt} = -\frac{2\pi}{\lambda} L n_2 \frac{dI(t)}{dt}$$

In an optical pulse the frequencies in the leading edge (dI/dt positive) are thus lowered and those in the trailing edge (dI/dt negative) are raised giving an overall up chirp.

The unsaturated gain or loss in the amplifier or absorber gives a chirp which may be obtained [1] by considering a Lorentzian line shape centred at frequency ω_0 . The refractive index at ω is given by

$$\text{Eqn. 2.6.4 } n(\omega) = n_0 + \frac{\pi}{2}(\omega_0 - \omega) \frac{c}{\omega} g(\omega_0 - \omega) \alpha$$

where n_0 is the refractive index in which the transition medium is dissolved, c is the 'in vacuo' speed of light, α is the gain coefficient at the transition peak and $g(\omega_0 - \omega)$ is the Lorentzian line shape -

$$\text{Eqn. 2.6.5 } g(\omega_0 - \omega) = \frac{2}{\pi \Delta\omega_0} \left(1 + \left(\frac{2(\omega_0 - \omega)}{\Delta\omega_0} \right)^2 \right)^{-1}$$

where $\Delta\omega_0$ is the line width (FWHM).

When an intense optical pulse saturates the absorber in a passively modelocked laser the absorption coefficient will have a time dependence which will thus cause a time dependent refractive index via eqn. 2.6.4. This again gives a time dependent phase [2]

$$\text{Eqn. 2.6.6 } \phi(t) = -\omega_0 n(t) \frac{L}{c}$$

which gives a frequency chirp as in eqn. 2.6.3 but opposite in sign to the Kerr effect. As well as this saturation chirp there is a contribution at low light levels arising from phase memory in the absorber [3]. Now that femtosecond pulses are approaching the phase decay time of organic molecules the transition bandwidth has become significant. It has been considered theoretically by introducing an unsaturable linear optical filter into the rate

1 D.G. Parker, *Electron. Lett.* **21**, 778 (1985)

2 S. de Silvestri, P. Laporta, O. Svelto, *IEEE J. Quantum Electron.* **QE-20**, 533 (1984)

3 W. Rudolph, B. Wilhelmi, *Opt. Commun.* **49**, 37 (1984)

1 J.-C. Diels, J.J. Fontaine, I.C. McMichael, B. Wilhelmi, W. Dietel, D. Kuhlke, *Sov. J. Quantum Electron.* **13**, 1562 (1983)

equations [1,2]. These models show the shortest pulses when the CPM lasers are operated off resonance (long wavelength side) with appropriate chirp compensation.

Practically these two non-linear effects are changed in passively modelocked lasers by moving the absorber away from the focus in it's folded section which changes the energy density and thus intensity that the saturable absorber sees.

The dielectric mirrors used in laser cavities can be a major source of GVD. These devices obtain their very high reflectivity by interference between the reflections from multiple dielectric layers of alternating high and low refractive index. As well as a frequency dependent reflectivity (they are designed to have a high reflectivity over a specified wavelength range) they produce a frequency dependent phase shift $\phi(\omega)$ which gives dispersion given by $d^2\phi/d\omega^2$. This can be calculated using the normal matrix formalism [3,4] for the typical single stack dielectric mirror and broad band double stack mirrors.

The results for a single stack dielectric mirror composed of a set of $\lambda/4$ stacks in the form {air H (LH)⁹ glass} with $n_L=1.45$ and $n_H=2.28$ and centred on 563nm are shown in figure 2.6.1. Graphs (a) and (b) show how reflectivity and dispersion vary with wavelength, and show that at normal incidence the narrow band single stack mirror gives no GVD (assuming perfect construction), however if used near the edge of it's high reflectivity zone substantial GVD is introduced - at 4% transmission it's GVD is $300 \times 10^{-30} \text{ s}^2$ which is equivalent to 6mm of quartz at 600nm. At the long wavelength side of the high reflectivity zone the dispersion is positive, while at the short wavelength side it is negative. Graphs (c) and (d) show how the reflectivity and dispersion vary with angle for the central wavelength, and thus it can be seen that using the mirror at an angle can introduce significant positive dispersion. For a summary of the method by which these reflection and dispersion curves were calculated for thin film devices, and for some further

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- 1 J.-C. Diels, J.J. Fontaine, I.C. McMichael, B. Wilhelmi, W. Dietel, D. Kuhlke, *Sov. J. Quantum Electron.* **13**, 1562 (1983)
 - 2 W. Rudolph, B. Wilhelmi, *Opt. Commun.* **49**, 37 (1984)
 - 3 S. de Silvestri, P. Laporta, O. Svelto, *IEEE J. Quantum Electron.* **QE-20**, 533 (1984)
 - 4 W. Dietel, E. Dopel, K. Hehl, W. Rudolph, E. Schmidt, *Opt. Commun.* **50**, 179 (1984)

investigation into their effects on ultrashort pulses see Appendix A.

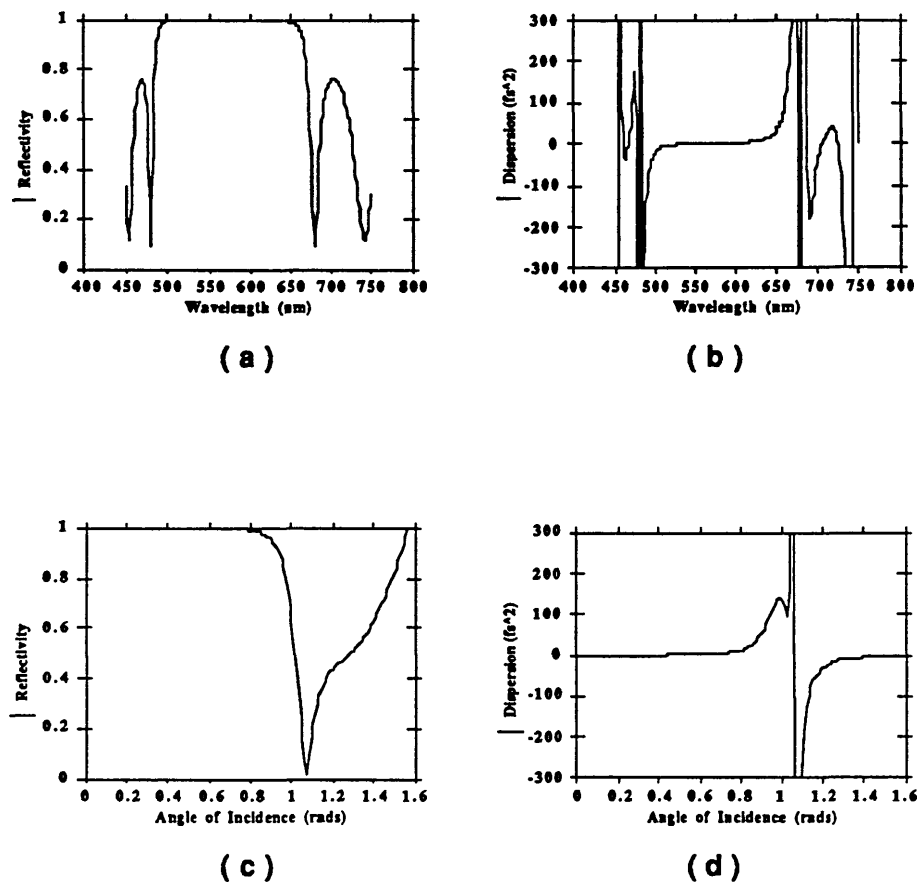


Fig. 2.6.1 Reflectivity and dispersion curves for a single stack dielectric mirror. (a) and (b) show plots vs wavelength at normal incidence. (c) and (d) show plots (for p polarization) vs angle of incidence at central wavelength.

For a broadband double stack mirror type only the outer stack of double stack mirror types tends to affect the phase. Figure 2.6.2 shows how the reflectivity and dispersion vary with wavelength for broadband coatings. In both cases there is a second stack with films 1.25 times those of the first stack which is the same as that mentioned above. In graphs (a) and (b) the short wavelength stack is on top, whilst in graphs (c) and (d) it is at the bottom. If the incident light traverses the first stack to be reflected by the second very large dispersions of either sign of greater than $8000 \times 10^{-30} \text{ s}^2$ may be produced. If the short wavelength stack is at the bottom then it can be seen that there would be very large

phase distortions to pulses being reflected at the short wavelengths of the high reflectivity zone.

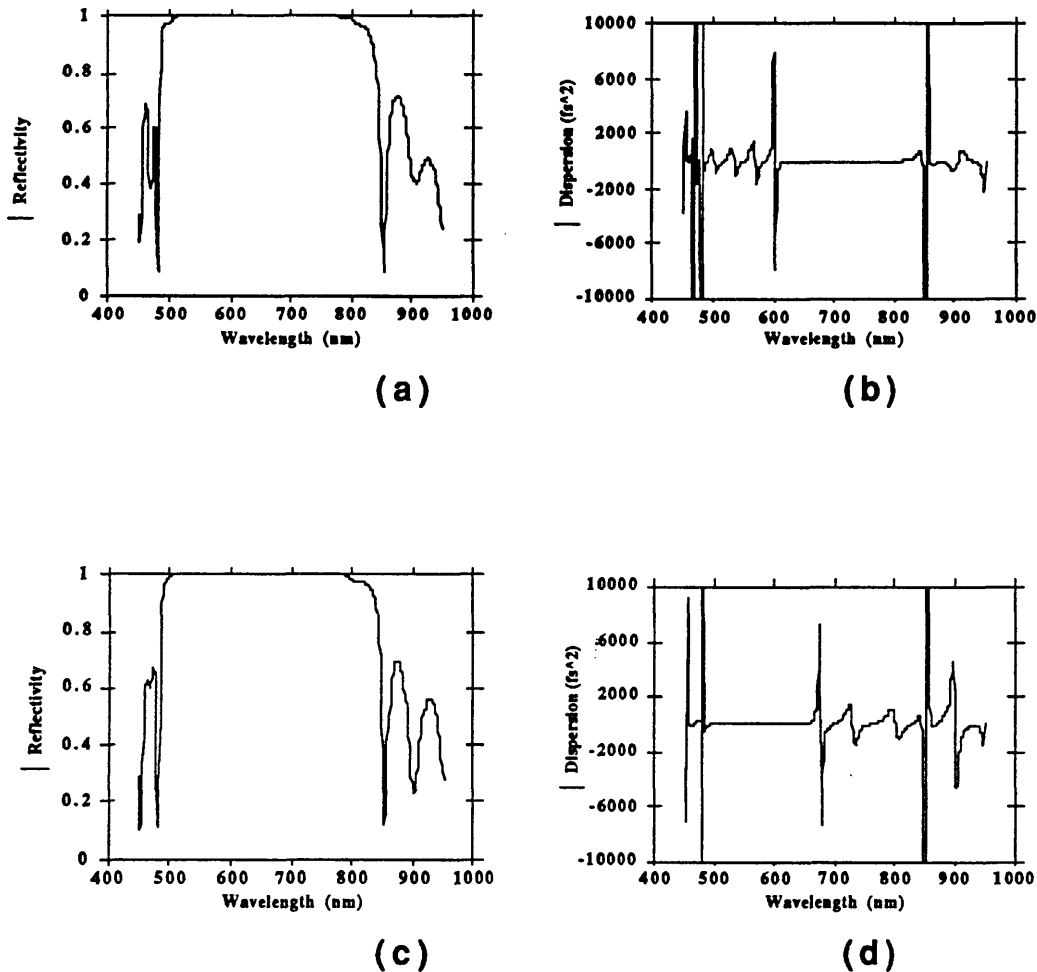


Fig. 2.6.2 Plots of Reflectivity and dispersion verses wavelength for broadband coatings with the red reflecting stack (a & b) and the blue reflecting stack (c & d) on the outside respectively.

Thus we see that dielectric mirrors contribute to GVD if used as high or low frequency cut off filters to limit the laser oscillation wavelength, and early attempts at reducing the pulse width from dye lasers by changing the mirrors were in fact a matter of optimising the intracavity dispersion. This method of intracavity GVD adjustment is obviously clumsy to use in practice and a number of alternative techniques exist.

If the laser is exhibiting negative frequency chirp then positive GVD may easily be provided by introducing normally dispersive glass into the cavity. This may be done controllably by translating

a prism in the cavity, which also serves as a tuning element, along the axis bisecting its deviation angle.

A laser with a positive frequency chirp requires negative GVD which is harder to provide. The usual method is a dispersive delay line employing a prism sequence devised by Fork and Gordon [1,2] and shown in fig. 2.6.1. The prisms are used at minimum deviation and are cut so that the rays enter and leave at Brewster's angle. The first prism introduces an angular dispersion into the beam which means that the lower frequency components will see a longer optical path in the second prism than the higher frequencies. Hence the spectral components emerging from the second prism will be spatially distributed (although all parallel to the incident beam) with the lower frequency components delayed with respect to the higher. The spatial spread may then be undone by passing the rays through another identical prism pair or, in a linear cavity, reflecting them back through the prisms. The four prism sequence is easy to introduce into a ring cavity since there is no net displacement of the incident beam. The net GVD of the sequence is adjustable through zero by translating one of the prisms along an axis perpendicular to its base. The amount of negative GVD which can be introduced is limited by the prism separation L . If the prisms are used away from Brewster's angle the amount of dispersion is greatly increased, at some loss in transmission. Highly dispersive glasses may also be used to increase the amount of dispersion.

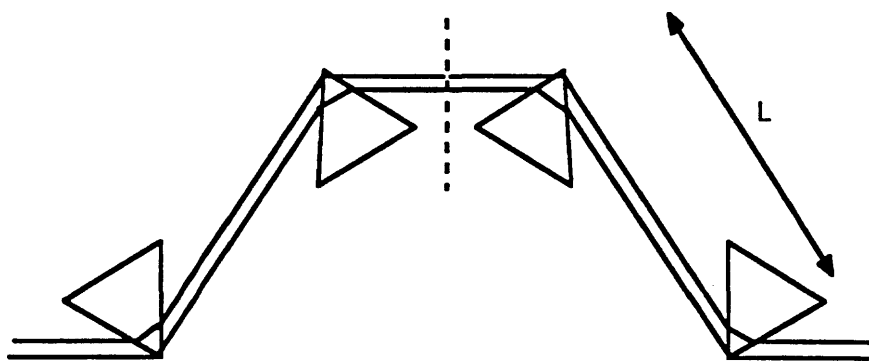


Fig. 2.6.1 Dispersive delay line using prisms

1 R.L. Fork, J.P. Gordon, *Opt. Lett.* **9**, 153 (1984)

2 R.L. Fork, O.E. Martinez, J.P. Gordon, *Opt. Lett.* **9**, 150 (1984)

It has been found that generally lasers with excess negative GVD produce longer pulses but are reasonably stable [e.g. 1] whereas those with excess positive GVD are unstable, giving broad pulses and spiking on a microsecond time scale [e.g. 2].

The shortest pulses do of course require large bandwidths and thus to generate pulses even shorter than those obtainable at present it will be necessary to compensate for terms in the intracavity chirp which are higher than the second order term discussed above. The gain and absorption profiles of the dyes will also need to be modified to enable complete modelocking over a broad spectral region.

(b) extracavity

If a pulse is not bandwidth limited i.e. has a chirp then the application of an appropriate amount of GVD will 'unchirp' and thus narrow the pulse down to its bandwidth limit. The prism sequence mentioned above for intracavity operation can be used extracavity but with loss of effectiveness since it will be traversed only once by the pulse instead of many times as inside a laser cavity. Another device which can produce adjustable negative GVD is a pair of diffraction gratings [3] arranged as shown below in fig. 2.6.3

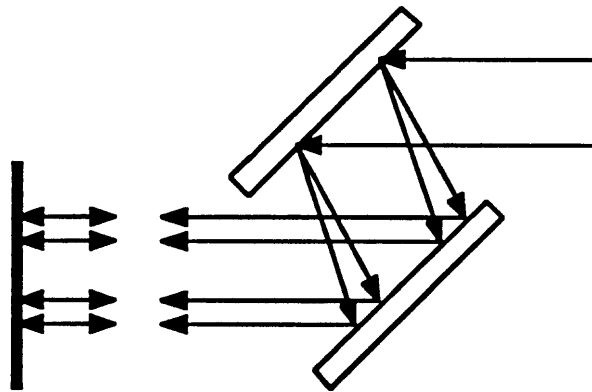


Fig. 2.6.3 Dispersive delay line using a grating pair.

Here again a spatial separation of spectral components is introduced and used to produce a temporal shift i.e. dispersion as in the prism sequence. The emerging beam is reflected back through the gratings (usually not collinearly) so that it emerges parallel to

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- 1 J.A. Valdmanis, R.L. Fork, J.P. Gordon, *Opt. Lett.* **10**, 131 (1985)
 - 2 A.M. Weiner, PhD Thesis, MIT Cambridge Mass. (1984)
 - 3 E.B. Treacy, *IEEE. J. Quantum Electron.* **QE-5**, 454 (1969)

the incident beam, but with a dispersion dependent on the grating separation, the angle of incidence, the frequency of light and the grating constant. The negative dispersion for a typical set-up with a 1m grating separation is about $1.24 \times 10^{-24} \text{ s}^2$ which is larger in a single pass than can be obtained with prisms or dielectric mirrors. A major use for this device has been in the fibre-optic pulse compressor [1] where a pulse is first frequency broadened and linearly chirped by passage through a single mode optical fibre and then compressed by the diffraction grating dispersive delay line. Grating pairs cannot be used inside CW dye lasers because of their relatively high loss.

With both prism and grating dispersive delay lines it is possible to introduce a mask with a spatially varying transmission or phase function in the region where the frequencies are distributed spatially. Thus the frequency spectrum of a pulse may be manipulated to give it tunable wavelengths, durations, amplitude and phase modulations [2,3].

Another device which can give adjustable linear or non-linear GVD is the Gires-Tournois interferometer [4] which is essentially an etalon that has one surface of 100% reflectivity. This has been used in several applications, including intracavity optimisation in a femtosecond laser cavity [5]. For this application the Gires-Tournois has advantages over prism sequences of being potentially more dispersive, more compact, applicable to all polarizations, and being usable over a wide range of frequencies. By cascading several of these devices a range of non-linear group delay functions may be produced to compensate high order dispersion terms in the laser cavity [5].

A combination of a grating sequence and a prism sequence can be used to compensate for both quadratic and cubic phase distortion of the pulse and it is this combination which in a pulse compressor set up has produced the shortest pulses to date (6fs - [6]).

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- 1 B. Nikolaus, D. Grischkowsky, *Appl. Phys. Lett.* **43**, 228 (1983)
 - 2 R.N. Thurston, J.P. Heritage, A.M. Weiner, W.J. Tomlinson, *IEEE J. Quantum Electron.* **QE-22**, 682 (1986)
 - 3 M.D. Dawson, T.F. Boggess, D.W. Garvey, A.L. Smirl, *Opt. Lett.*
 - 4 F. Gires, P. Tournois, *C.R. Acad. Sci. Paris* **258**, 6112 (1984)
 - 5 P.M.W. French, G.F. Chen, W. Sibbett, *Opt. Commun.* **57**, 263 (1986)
 - 6 R.L. Fork, C.H. Brito Cruz, P.C. Becker, C.V. Shank, *Optics Letters* **12**, 483 (1987)

2.7 The Streak Camera

This electro-optic device is the only linear pulse measurement technique capable of subpicosecond resolution [1,2]. The principle of the streak camera was first proposed in 1956 [3] but it was only after 1969 that it became a potential picosecond measurement system [4]. Good reviews on streak cameras are references [5,6]. Here I shall summarize the principles of single-shot and synchroscan operation. The 'circular' streak camera [7] shall be excluded as it is of lesser importance. The figure below shows a schematic of a typical streak camera - the Photochron II [8].

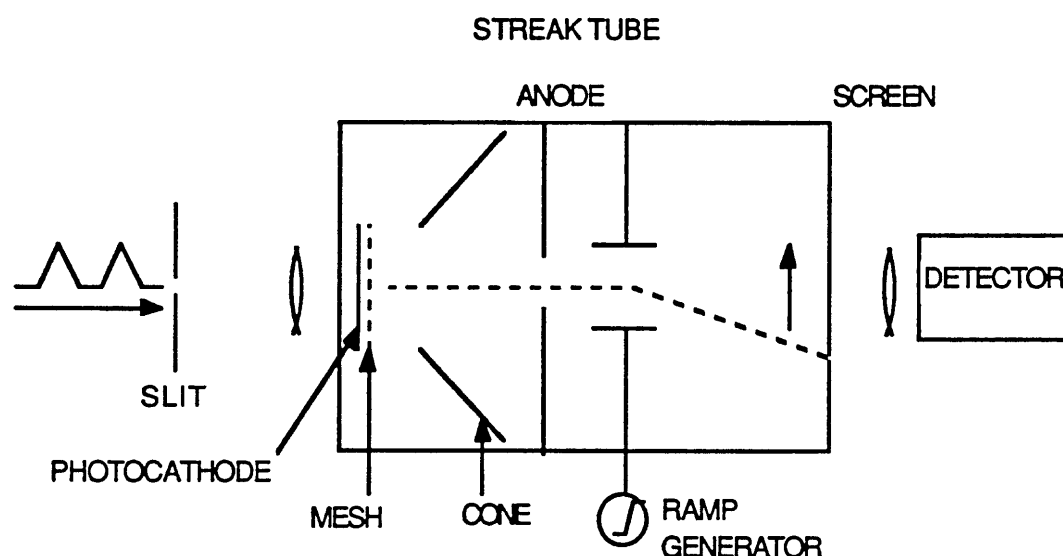


Fig. 2.7.1 The Photochron II streak camera

The optical signal is imaged from a slit onto a photocathode where it liberates electrons. The number of electrons emitted is proportional to the light intensity striking the photocathode. They

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- 1 D.J. Bradley, W. Sibbet, *Appl. Phys. Lett.* **27**, 382 (1975)
 - 2 A. Finch, Y. Liu, H. Niu, W. Sibbett, W.E. Sleat, D.R. Walker, Q.L. Yang, H. Zhang, *Ultrafast Phenomena VI* (Springer Series in Chemical Physics **48**), 159 (1988)
 - 3 E.K. Zavoiski, S.D. Fanchenko, *Sov. Phys. Doklady* **1**, 285 (1956), *Appl. Opt.* **4**, 1155 (1965)
 - 4 D.J. Bradley, U.K. patent spec. 31167 (1970)
 - 5 E.P. Ippen, C.V. Shank, "Techniques for measurements" in *Ultrashort Light Pulses* (Springer 1977) ed. S.L. Shapiro
 - 6 D.J. Bradley, G.H.C. New, *Proc. IEEE* **62**, 313 (1974)
 - 7 W. Sibbett, W.E. Sleat, W. Krause, J.R. Taylor, *SPIE 491, High Speed Photography*, 76 (Strasborg 1984)
 - 8 S.F. Bryant, PhD Thesis, University of London (1976)

are accelerated through various electrodes and deflection plates to be focused onto a phosphor screen. By applying a fast synchronized ramp signal to the deflection plates the signal is streaked across the screen. The time information of the optical signal is thus converted into a temporal distribution of the electron signal which in turn is converted into a spatial intensity distribution on the phosphor screen. This intensity pattern may then be analysed either by taking a density plot of a photograph of it or by direct conversion to an electrical signal using an optical multichannel analyzer (OMA) or similar device, to give the temporal intensity profile of the incident optical signal. Typically a dc bias of about 2.5kV is applied while the ramp has an amplitude of about 5kV. To ensure that the streak across the screen is linear in time only the central 20-30% of the ramp is used and this is enough for a full sweep of the phosphor.

The theoretical time resolution of a photocathode is about 10fs which is insignificant compared to the photoelectron time dispersion t_d and the technical time resolution t_s . The recorded pulse width t_r of a pulse of duration t_p is thus given (assuming Gaussian resolution distributions and pulse shapes) by

$$\text{Eqn. 2.7.1 } t_r = \sqrt{t_p^2 + t_d^2 + t_s^2}$$

The photoelectron time distribution arises from the time dispersion of the energy distribution which the emitted electrons must have due to lattice scattering and pair production in the photocathode [1]. The time distribution is related to the energy spread by [2]

$$\text{Eqn. 2.7.2 } t_d = 2.34 \times 10^{-8} \frac{dE}{E}$$

where dE is measured in electron volts and E , the extraction field in volts per centimetre. The fine mesh near the photocathode in figure 2.7.1 gives the strong fields necessary to minimize this distribution, and is adopted in most streak tubes. For the Photochron II the field strength is about 20kV/cm and dE is about

1 S.F. Bryant, PhD Thesis, University of London (1976)

2 V.V. Korabkin, A.A. Malyutin, M. Ya, Schelev, *J. Photog. Sci.* 17, 179 (1964)

0.4eV at 734nm giving a time dispersion of 0.5ps.

The technical time resolution is the time necessary for the streak tube to sweep through one resolution element of the phosphor screen. This depends on the sweep (or writing) speed across the phosphor, v , and the dynamic spatial resolution, d , according to

$$\text{Eqn. 2.7.3 } t_s = \frac{1}{vd}$$

For the Photochron II the writing speed is $2 \times 10^8 \text{ ms}^{-1}$ and the spatial resolution is 10^4 lines/m giving a technical time resolution of about 0.5ps. Advances in photocathode and deflection plate design has improved the time resolution of streak cameras to about 0.3ps [1].

A calibration of the streak cameras time scale may be achieved by splitting the input pulse in two, and sending one of the resulting pulses along a known optical delay before recombining and sending to the streak tube. Thus two pulses of known temporal separation appear on the streak camera screen.

The trigger for the streak camera deflection plate ramp voltage is often provided by a photoconductive device or photodiode. Although the device has been used in single shot mode, to study CW mode locked lasers [e.g. 2] the repetitive nature of the pulse trains makes synchronous operation attractive.

In the synchroscan mode of operation the voltage ramp is applied to the deflection plates in synchronism with the repetition rate of the laser so that successive streak images superimpose on the phosphor screen. The intensity distribution on the screen is thus an integration of a large number of pulses, giving a large signal to noise ratio and an improved dynamic range since much lower levels of photocurrent can be detected.

The fast ramp required for the deflection plates is in fact derived from the central 1/6 cycle of a sinusoidal signal which is the most linear (to within 5%). At these high frequencies a sine wave is much easier to generate since it does not require the

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- 1 A. Finch, Y. Liu, H. Niu, W. Sibbett, W.E. Sleat, D.R. Walker, Q.L. Yang, H. Zhang, *Ultrafast Phenomena VI* (Springer Series in Chemical Physics 48), 159 (1988)
 - 2 I.S. Ruddock, PhD Thesis, University of London (1978)

support of the higher harmonics necessary for other waveforms (e.g. sawtooth). The sine wave may be derived from the RF signal used to drive the acousto-optic modulator of an actively mode locked laser, or from the optical output of the laser itself. In this later case, which generally provides better synchronization, the laser output is sensed by a photodiode or semiconductor switch whose electrical output is used to trigger a tunnel diode oscillator. The synchronized sinusoidal signal from this is amplified and coupled into the deflection plates via a high Q impedance matching network.

The resolution of synchroscan is very sensitive to interpulse jitter which leads to small time differences between the arrival of the optical pulses and the application of deflection voltage. The input optical signal to streak cameras working at the resolution limits must therefore be gated to minimize this problem. A tunnel diode system is better in this aspect than directly driving from the modelocker signal, and an alternative method using a GaAs photoconductive element [1] has been used to try to get around this problem.

2.8. Second Harmonic Generation Autocorrelation

This measurement technique which is based on the non-linear process of second harmonic generation (SHG) was first proposed by Maier et al. [2], and Weber [3]. It is the most widely used technique for looking at the ultrashort pulses from CW dye lasers and has been used to measure pulses as short as 6fs [4]. SHG autocorrelation was immediately superseded by the two photon fluorescence technique [5] which was more useful for the low repetition rate pulsed lasers of the pre 70's since it could be used to take a single shot, single pulse measurement whereas a stream

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- 1 A.S.L. Gomes, A.S. Gouveia-Neto, W. Margulis, J.R. Taylor, *Rev. Sci. Instrum.* (Oct. 1986)
 - 2 M. Maier, W. Kaiser, J.A. Giordmaine, *Phys. Rev. Lett.* **17**, 1275 (1966)
 - 3 H.P. Weber, *J. Appl. Phys.* **38**, 2231 (1967)
 - 4 R.L. Fork, C.H. Brito Cruz, P.C. Becker, C.V. Shank, *Optics Letters* **12**, 483 (1987)
 - 5 J.A. Giordmaine, P.M. Rentzepis, S.L. Shapiro, K.W. Wecht, *Appl. Phys. Lett.* **11**, 216 (1967)

of pulses are required to build up a SHG type autocorrelation. Both techniques produce the second order correlation function of the pulse profile. Other higher order nonlinear methods have been demonstrated [1,2] but are not commonly used.

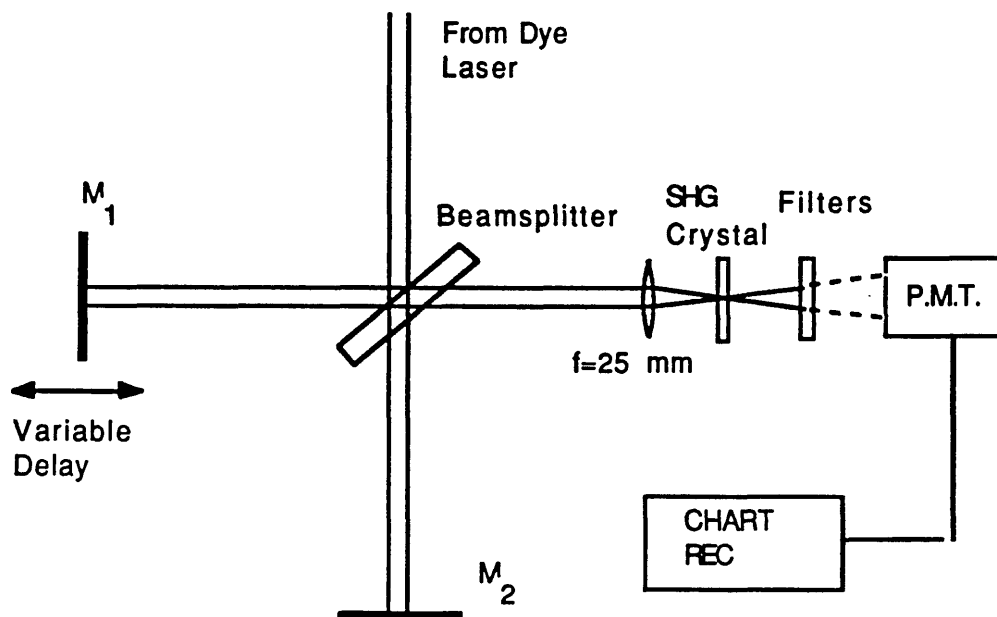


Fig. 2.8.1 Typical configuration for SHG autocorrelation

Optical second harmonic generation [3] is a process whereby two photons of equal energy combine in a nonlinear material to produce a single photon of twice the individual energies. This process can only occur in nonlinear media which are non-centrosymmetric (due to parity conservation) and is only efficient if the fundamental and second harmonic signal are phase matched. The typical set-up for SHG autocorrelation is shown in fig. 2.8.1.

The input stream of optical pulses are divided by amplitude into two beams at the beamsplitter. These two beams then have optical delays imposed upon them by the path lengths to and from the mirrors, and are recombined and focused into the SHG crystal. A recording is made of the average second harmonic power as a function of the relative delay between the two trains of pulses. This is given by

- 1 E.P. Ippen, C.V. Shank, "Techniques for measurements" in *Ultrashort Light Pulses* (Springer 1977) ed. S.L. Shapiro
- 2 D.J. Bradley, G.H.C. New, *Proc. IEEE* **62**, 313 (1974)
- 3 P.A. Franken, A.E. Hill, C.W. Peters, G. Weinreich, *Phys. Rev. Lett.* **7**, 118 (1961)

$$\text{Eqn. 2.8.1 } I_{2\omega} = \int_{-\infty}^{\infty} |E(t)|^2 dt$$

where

$$\text{Eqn. 2.8.2 } E_{\omega} = E(t) + E(t+T)$$

The time dependent field may be written as

$$\text{Eqn. 2.8.3 } E(t) = A(t) e^{i\omega t + \phi}$$

If we substitute 8.3 and 8.2 into 8.1, expand out the squares and average over all interference terms which will not be detected if T is varied fast enough then the result obtained [1] is

$$\text{Eqn. 2.8.4 } I_{2\omega} = c [1 + 2G^{(2)}(T)]$$

where c is a constant and

$$\text{Eqn. 2.8.5 } G^{(2)}(T) = \frac{\int I(t) I(t-T) dt}{\int I^2 dt}$$

Eqn 2.8.4 is true if the two beams entering the crystal are collinear and of the same polarization so type I phase matching is employed. If the two recombining beams have orthogonal polarizations, or they are parallel but not collinear in the SHG crystal then phase matching, and thus an SHG signal is only obtained when both pulses are present. The resulting recorded signal is then

$$\text{Eqn. 2.8.6 } I_{2\omega} = c G^{(2)}(T)$$

When examining the pulses from a modelocked laser the collinear type of autocorrelation is the most useful since the peak to background gives a measure of how well modelocked the laser is. Examples are given in Fig. 2.8.2 Below

1 E.P. Ippen, C.V. Shank, "Techniques for measurements" in *Ultrashort Light Pulses* (Springer 1977) ed. S.L. Shapiro

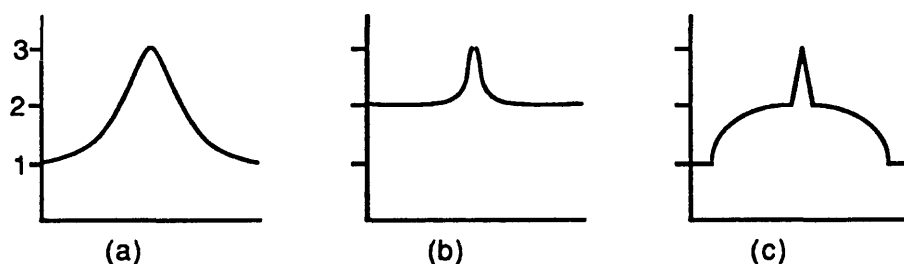


Fig. 2.8.2 Autocorrelation traces resulting from (a) a modelocked laser, (b) a free running laser, (c) a partially mode locked laser.

For perfect modelocking a peak to background ratio of 3:1 is obtained. A free running laser, where the output is random noise, will produce a coherence spike with a contrast ratio of 3:2. The full width half maximum of the spike gives the lasers coherence time. If there is phase locking of only some of the laser modes then (c) is a typical result where there is a noise burst with an autocorrelation contrast ratio of 3:2:1. The interpretation of contrast ratios requires some care since a laser with only 50% of it's modes locked in phase will produce a contrast ratio of 2.9:1 [1].

The second type of autocorrelation (non-collinear) does not give a background and so there is no contrast ratio to determine if modelocking is complete. It's advantage is that the energy and character of the wings of a pulse may be more easily determined without the noise component of the background. It is therefore often used to examine the pulses from fibres, pulse compressors etc. where dispersion of energy is important.

The second order autocorrelation function $G^{(2)}(\tau)$ is symmetric and gives no unique information about pulse shape. When determining the duration of a pulse a particular pulse shape is assumed and this determines the ratio of the FWHM of the autocorrelation function to the pulse duration. This ratio together with the time bandwidth products k for transform limited pulses is given in table 2.8.3 below. A pulse shape may be inferred by matching the measured autocorrelation traces to a set of fingerprinting calculated autocorrelation profiles, and if the pulse is thought to be transform limited by matching the time bandwidth product to those calculated. Care is required however since fluctuating pulse trains always give extended exponential wings

1 R.H. Picard, P. Schweitzer, *Phys. Lett.* **29A** (1964)

independent of pulse profile [1], and if the pulse durations fluctuate the value calculated from the autocorrelation trace will be shorter than the actual average pulse width [2].

$I(t)$	$\frac{\Delta\tau}{\Delta t}$	$k = \Delta t \Delta\nu$
1 ($0 < t < \Delta t$)	1	0.886
$e^{-\frac{(4\ln 2)t^2}{\Delta t^2}}$	$\sqrt{2}$	0.441
$\text{sech}^2\left(\frac{1.76 t}{\Delta t}\right)$	1.55	0.315
$e^{-\frac{(\ln 2)t}{\Delta t}}$ ($t > 0$)	2	0.110

Table 2.8.3 A table of some pulse shapes, the ratio of their widths to their autocorrelation widths, and their time-bandwidth product.

The limits on the resolution of SHG autocorrelation are the effect of GVD of the beams inside the crystal [3], and the finite phase matching bandwidth of the nonlinear crystal [4]. Pulses with a bandwidth greater than this do not generate SHG signals proportional to the square of the fundamental intensity. This can be overcome by ensuring that the beams interact over a very narrow path length in the crystal since phase matching bandwidth is inversely proportional to the interaction length [5].

If an autocorrelation is performed with sufficient resolution the fringe terms obtained from substituting equations 2.8.3 and 2.8.2 into 2.8.1 will be included in the measured function. Note that it is also necessary for the pulses from the laser be coherent and stable for these fringes to be seen. The resulting recorded autocorrelation signal is then given by the complete expression [6]

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- 1 J.M. Catherall, G.H.C. New, To be published
 - 2 E.W. Van Stryland, *Opt. Commun.* **31**, 93 (1979)
 - 3 C.V. Shank, R.L. Fork, R. Yen, R.H. Stolen, W.J. Tomlinson, *Appl. Phys. Lett.* **40**, 761 (1982)
 - 4 I.S. Ruddock, PhD Thesis, University of London (1978)
 - 5 R.C. Miller, *Phys. Lett.* **26A**, 177 (1968)
 - 6 J.-C. Diels, J.J. Fontaine, I.C. McMichael, F. Simoni, *Appl. Opt.* **24**, 415 (1985)

Eqn. 2.8.7

$$I(\tau) = \int_{-\infty}^{\infty} \left[A(t) e^{i(\omega t + \phi(t))} + A(t-\tau) e^{i(\omega(t-\tau) + \phi(t-\tau))} \right]^2 dt$$

The full expansion of this gives interference fringes with a peak to background ratio of 8:1. This type of interferometric autocorrelation is sensitive both to pulse shape and to frequency chirp across the pulse, however sophisticated computer fitting is required for complete analysis. Some of the calculated results for sech^2 pulses, and for Gaussian pulses with varying degrees of chirp are given in ref. [1].

All subpicosecond pulse measurements performed in this thesis were performed using a scanning collinear second harmonic autocorrelator.

1 J.-C. Diels, J.J. Fontaine, I.C. McMichael, F. Simoni, *Appl. Opt.* **24**, 415 (1985)

3. PASSIVE MODE LOCKING OF C.W. ENERGY TRANSFER DYE LASERS IN THE NEAR INFRARED

3.1 Introduction

Until 1985 the passive mode locking of dye lasers was largely restricted to the Rhodamine 6G/DODCI system initially demonstrated in 1972 [1]. Since then active/passive dye combinations have been developed to extend the spectral coverage of this technique into most of the visible spectrum [2,3]. In 1985 a Krypton-ion-pumped Rhodamine 700 system was passively mode locked in the near infrared [4], however the Krypton-ion laser is not a reliable pump source and the argon-ion is preferred for pumping c.w. dye lasers. Unfortunately the active dyes which lase in the near infrared and have absorption spectra overlapping with the argon-ion lasing lines (e.g. Styryl 9) tend to have short upper state lifetimes making them unsuitable for passive mode locking. In order to overcome this difficulty and thus enable argon-ion pumped passively mode locked c.w. dye lasers in the near infrared two energy-transfer dye mixtures have been investigated as possible suitable active media for passive modelocking.

The energy transfer process dyes occurs when one molecular species (the donor or sensitizer) which absorbs at the pump wavelengths has its fluorescence band at the longer wavelength absorption peak of the second (acceptor) species which is consequently pumped and can thus fluoresce at longer wavelengths than the first species alone could. The main mechanisms by which an energy transfer process can take place between two molecular species are: (1) radiative transfer where the donor emits a photon which is reabsorbed by the acceptor, (2) short range collisional transfer, (3) the formation of a complex between sensitizer and acceptor molecules and (4) a non-radiative resonant long range dipole-dipole interaction. This fourth mechanism is found to be the most important for the dyes as used in c.w. dye lasers and is

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- 1 E.P. Ippen, C.V. Shank, and Dienes, *Appl. Phys. Lett.* **21**, 348 (1972)
 - 2 P.M.W. French and J.R. Taylor, *Appl. Phys. Lett.* **50**, 1708 (1987)
 - 3 P.M.W. French, J.A.R. Williams, and J.R. Taylor, *Revue de Physique Appliquee* **22**, 1651 (1987)
 - 4 K. Smith, N. Langford, W. Sibbett, and J.R. Taylor, *Opt. Lett.* **10**, 559 (1985)

discussed by Forster [1] and Muto et al. [2]. The basic mechanism of this interaction for molecular species is illustrated by figure 3.1.1 below.

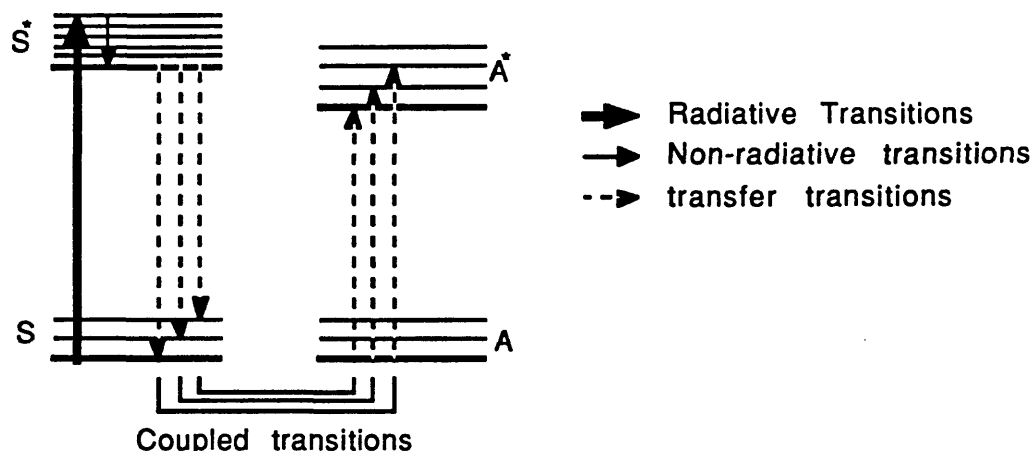


Fig. 3.1.1 The simplified energy level diagram for energy transfer between two molecular species.

Absorption of a pump photon leaves the donor molecule in a high vibrational level of its first excited electronic excitation state. Fast thermalisation with the surrounding medium brings it down to a lower vibrational level of the same excited state where it remains for its upper state lifetime. If the energy difference for a possible deexcitation of the donor molecule corresponds exactly to that for a possible absorption transition of a nearby acceptor molecule then with sufficient coupling both processes may occur simultaneously. Due to the broad spectra of large dye molecules in solution there is always sufficient coincidence of donor and acceptor transitions if there is some overlap between their absorption spectra, with this being favourable if the acceptor has a slight red shift in its spectra.

Quantitative analysis of this energy transfer mechanism leads to an expression for the rate constant as follows [3]

$$\text{Eqn. 3.1.1 } n_{S \rightarrow A^*} = \frac{9000 \ln 10 \kappa^2}{128 \pi^2 n^4 N \tau_s R^6} \int_0^{\infty} f_s(\nu) \epsilon_A(\nu) \frac{d\nu}{\nu^4}$$

1 T. Forster, Discuss. Faraday Soc. 27, 7 (1959)

2 S. Muto, C. Ito, and H. Inaba, *Electron. Commun. Japan* 66-C, 120 (1983)

3 T. Forster, Discuss. Faraday Soc. 27, 7 (1959)

where ν is the wave number, $\epsilon_A(\nu)$ the molar decadic extinction coefficient, $f_s(\nu)$ the spectral distribution of fluorescence (measured in quanta and normalized to unity on a wave number scale), N Avogadro's number and τ_s the mean lifetime of the excited sensitizer. n is the refractive index of the solvent, R the mutual distance between both molecules and κ an orientation factor whose average value for a random directional distribution is $2/3$.

The important features of this equation are that the probability of energy transfer decreases as the sixth power of distance between the molecules, and is inversely proportional to the mean sensitizer lifetime τ_s . Provided that the sensitizer upper state lifetime is not reduced to be comparable to its thermalisation time, then, for this mechanism, the sensitizer fluorescence spectrum remains the same as transfer efficiency increases.

This dipole-dipole interaction can be distinguished experimentally from the more trivial possible interactions. The observation that increasing the concentration of both components (but keeping a constant ratio) increases transfer efficiency rules out pump absorption by the acceptor species. Measurements indicating a reduction in the sensitizer lifetime with increasing acceptor concentration rules out radiative transfer, and if the process was collisional in nature increasing solvent viscosity would reduce the transfer efficiency.

3.2 The passive mode locking of the DCM/Rhodamine 700 dye laser

The DCM/Rhodamine 700 energy transfer dye combination had previous to this work [1] been demonstrated in an argon-ion pumped c.w. dye laser to have an efficiency as high as 13% at it's peak wavelength of 730nm, and to be capable of lasing up to 805nm.

Figure 3.2.1 below shows the tuning range we obtained with this medium in a simple 3 mirror laser cavity employing high reflectivity single stack dielectric mirrors centred on 750nm and tuned using a Spectra-Physics tuning wedge. Both mirrors in the folded section were of 100mm radius of curvature.

1 E.G. Marason, *Opt. Commun.* 40, 212 (1982)

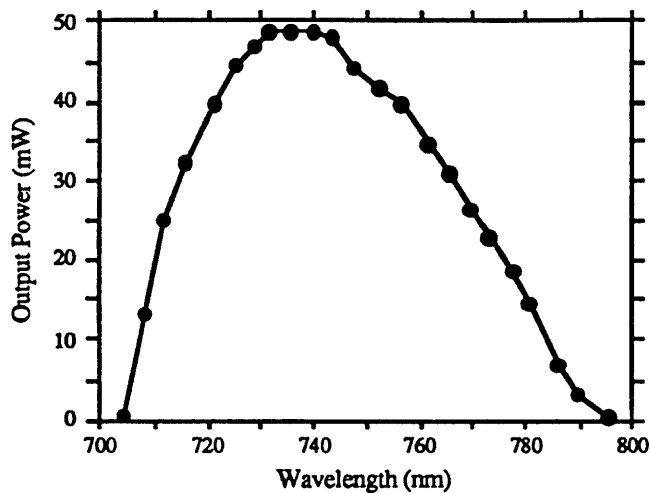


Fig. 3.2.1 Tuning curve for DCM/Rh700 dye mixture

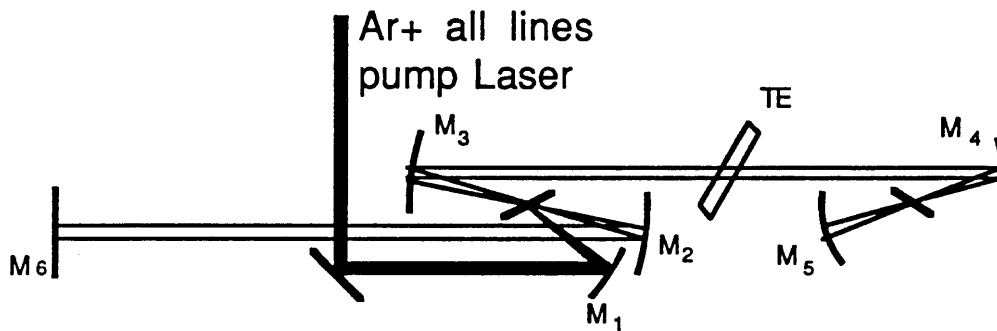


Fig. 3.2.2 Simple linear dye laser cavity

Figure 3.2.2 above shows the initial experimental configuration used to passively modelock this system. A five mirror cavity was employed which was configured to meet New's criteria [1] for stable single pulse evolution. The argon-ion pump beam (up to 7.0W 'all lines' visible output from a Spectra Physics model 2020) was coupled into the amplifying dye jet (2×10^{-3} M DCM and 2×10^{-3} M Rhodamine 700 in a 1:4 propylene carbonate:ethylene glycol mixture) by mirror M_1 of 50mm radius of curvature. Two mirrors (M_2, M_3) of 100mm radius of curvature completed the active folded section and the passive section consisted of a focusing mirror of 50mm radius of curvature and a retroreflecting mirror of 25mm radius of curvature about the absorber jet stream. Both jets were vertical and of about $100\mu\text{m}$ thickness (nozzles from Coherent

1 G.H.C. New, *IEEE J. Quantum Electron.* QE-10, 115 (1974)

Radiation Ltd). The cavity round trip time was 8ns. All the cavity mirrors supported a single stack dielectric coating of 100% reflectivity for normal incidence centred on 750nm and a Spectra Physics dielectric coated tuning wedge was used to control the laser wavelength, and to provide two output beams. The bandwidth limit imposed by this wedge, and its dispersion properties probably limited the pulse duration to being greater than 120fs. The lasing thresholds in the complete linear cavity (without any saturable absorber added to the second jet) were typically about 1.7W. There was a slight problem with the DCM occasionally precipitating out of solution, this was thought to be due to the presence of water as a contaminant in the glycol.

One of the output beams from the laser was directed onto a fast photodiode (BPW28) while the other permitted pulse width measurements via a standard collinear second harmonic generation autocorrelation technique employing Lithium Iodate as the frequency doubling crystal. The autocorrelation function could be observed in real time using a fast scanning autocorrelator, and this combined with the pulse train information from the diode facilitated optimisation of cavity parameters, absorber concentrations etc and made it possible to check for complete stable mode locking.

Two saturable absorbers were demonstrated in this linear cavity: DDI (1,1'-Diethyl-2,2'-dicarbocyanine iodide) and Cryptocyanine (1,1'-Diethyl-4,4'-carbocyanine iodide) for which the peak extinction coefficients of $19.3 \times 10^4 \text{ l mol}^{-1} \text{ cm}^{-1}$ and $23.0 \times 10^4 \text{ l mol}^{-1} \text{ cm}^{-1}$ were measured respectively for 1:9 ethanol:ethylene glycol solutions. Figure 3.2.3 shows the structural formulae and absorption profiles of these dyes.

With DDI pulses of less than 400fs were generated over the spectral range of 742nm to 761nm. Figure 3.2.4 shows typical autocorrelations of pulses recorded at (a) 747nm and (b) 761nm. Absorber concentrations which varied between 10^{-5} and $3 \times 10^{-5} \text{ M}$ resulted in lasing thresholds of the order of 3.5W. The best results were obtained for pump powers 1W above threshold for which total output powers of about 20mW were measured. As the absorber concentration was increased the lasing wavelength increased (since the gain peak is on the long wavelength side of the absorber peak). The tuning wedge was used to provide a cut off at long

wavelengths and thus force laser operation back within the absorption profile of the saturable absorber (otherwise the wavelength would keep on increasing and mode locking would not take place). No attempt was made to optimise the intracavity group velocity dispersion and a spectral investigation of the laser output revealed considerable excess bandwidth - the measured bandwidth (e.g. Figure 3.2.5) of 4.8nm being able to support pulses of 120fs duration.

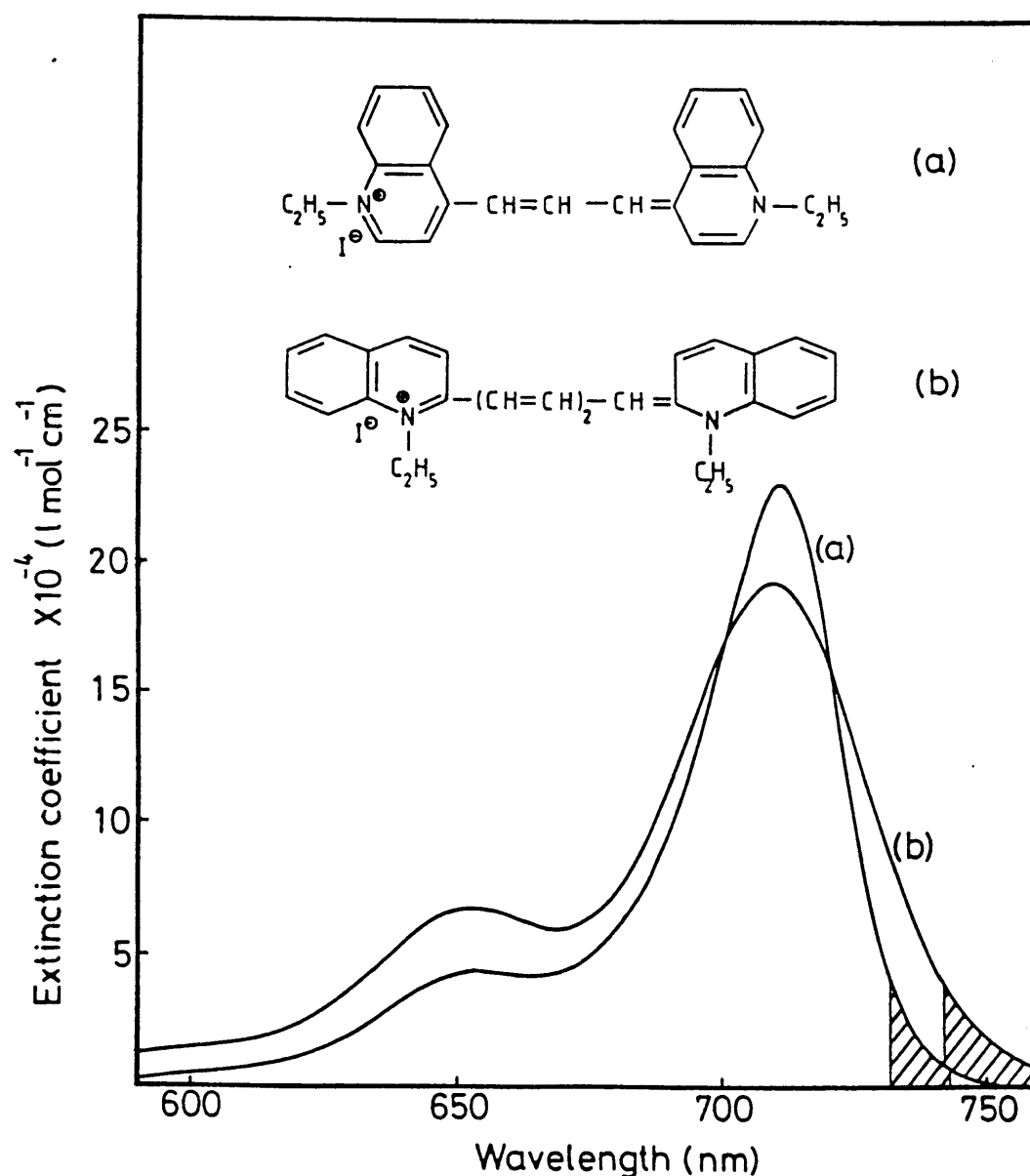


Fig. 3.2.3 Structural formulas and absorption profiles of (a) Cryptocyanine and (b) DDI. The hatched regions represent the tuning range in which sub-400fs pulses were generated.

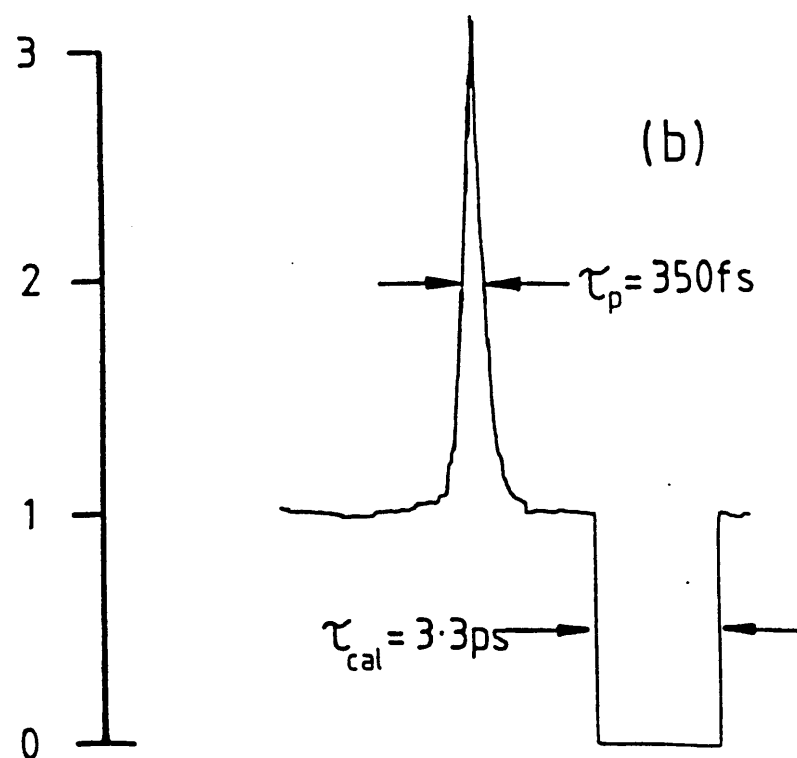
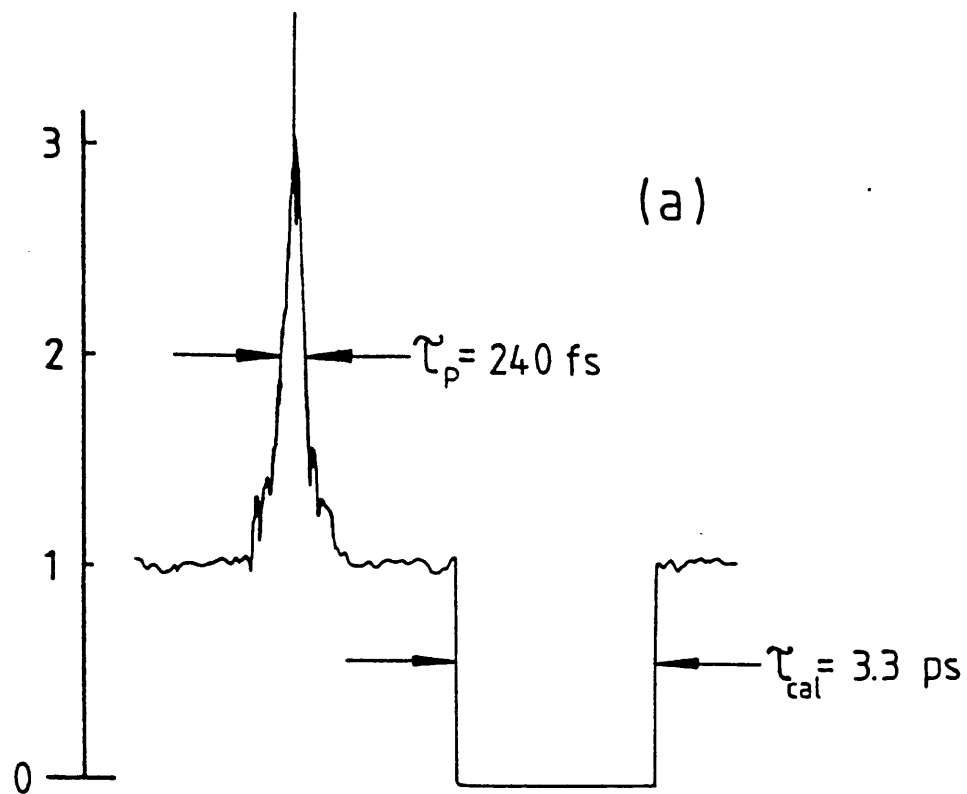


Fig. 3.2.4 Autocorrelation traces obtained with DDI as the saturable absorber for wavelengths centred at (a) 742 nm and (b) 761 nm.

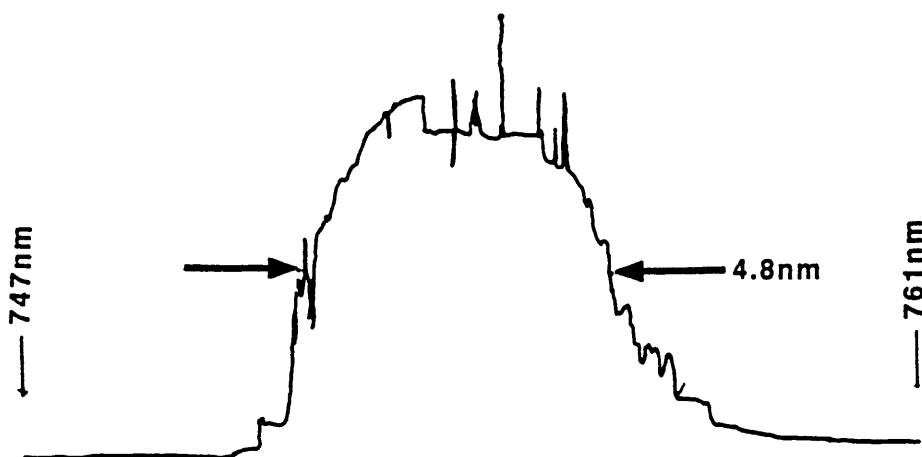


Fig. 3.2.5 Typical spectrum with DDI as the saturable absorber.

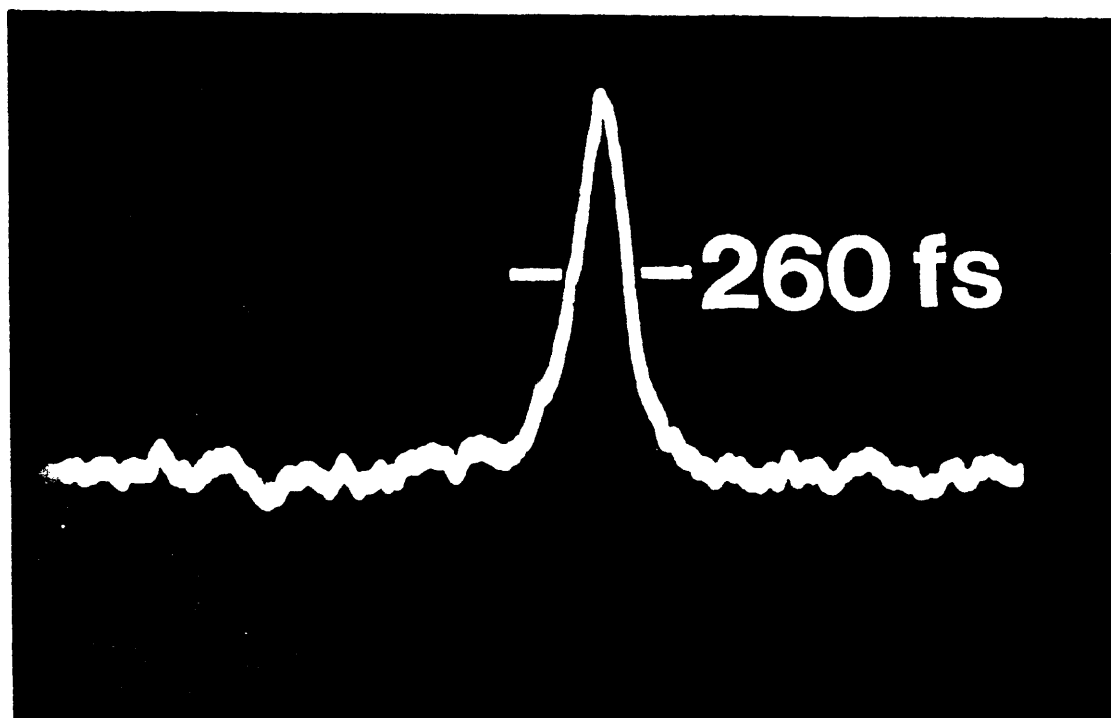


Fig. 3.2.6 Autocorrelation trace of a 260fsec pulse generated with Cryptocyanine as the saturable absorber.

When cryptocyanine was used as the saturable absorber pulses of less than 400fs duration were generated from 732nm to 743nm for absorber concentrations between 2×10^{-5} and 9×10^{-5} M. Lasing thresholds of 3.5W were again observed and pump levels around 5W

produced total output powers of 6mW, with the pulses again displaying considerable excess bandwidth. Figure 3.2.6 shows a photograph of an autocorrelation trace taken from the scanning autocorrelator which corresponds to a pulse duration of 260fs. In general the laser performance was not as stable with this saturable absorber and, if applicable, DDI is a more attractive alternative.

In order to try to generate shorter, transform limited pulses, the CPM ring cavity shown in figure 3.2.7 was constructed. The folded sections were as in the linear configuration except that the passive section mirrors were now both of 50mm radius curvature. Two extra plane mirrors were included in the cavity to permit a colliding pulse ring laser geometry. All the mirrors supported the same single stack coating already described and so mirrors M_6, M_7 , and M_8 being used away from normal incidence, acted as long wavelength cut-off filters and sources of adjustable group velocity dispersion [see e.g. 1]. The reduced reflectivities at these mirrors provided adequate output coupling, and by varying the absorber concentration and the angles α, β, γ the laser wavelength could be controlled thus eliminating the need for the tuning wedge. DDI was used as the saturable absorber and the total cavity round trip time was again 8ns. Typical lasing thresholds without any saturable absorber added were about 1.5W.

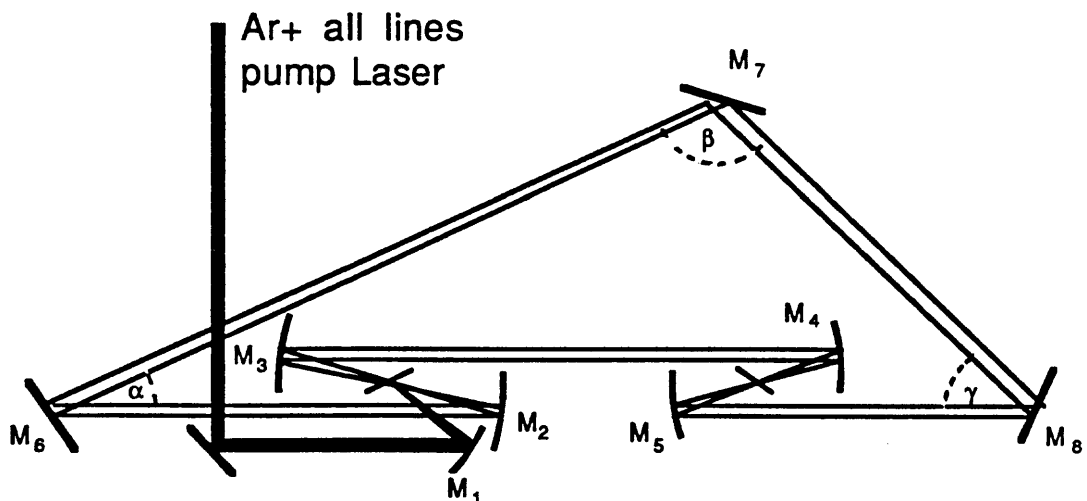


Fig. 3.2.7 Schematic of ring cavity

1 P.M.W. French, A.S.L. Gomes, A.S. Gouveia-Neto, and J.R. Taylor, *Opt. Commun.* **60**, 389 (1986)

By varying the DDI concentration between 10^{-5}M and $2 \times 10^{-5}\text{M}$ and the angles α, β, γ between $29^\circ, 55^\circ, 96^\circ$ and $17^\circ, 27^\circ, 136^\circ$, pulses of less than 120fs duration were obtained from 743nm to 754nm which were within 10% of their transform limited duration. Figure 3.8 shows an autocorrelation trace and figure 3.2.9 the corresponding spectrum of pulses of 110fs at 754nm. Under these conditions the lasing pump threshold varied between 4W and 6W and the laser was pumped 1W above threshold yielding output powers of up to 20mW per beam from mirror M7.

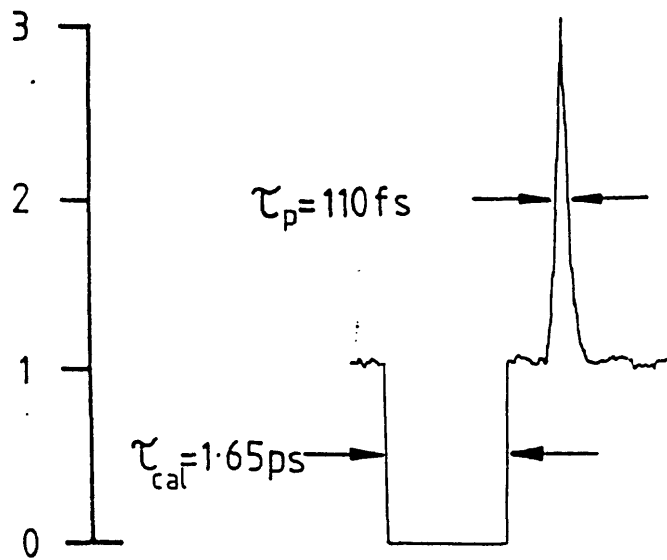


Fig. 3.2.8 Autocorrelation trace of a 110-fsec pulse obtained at 754nm in a ring configuration.

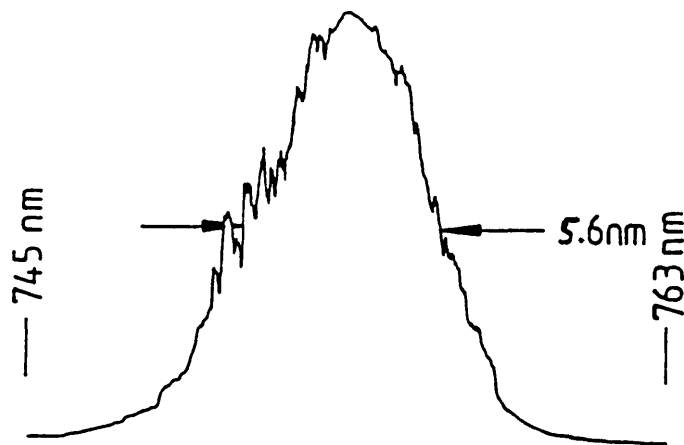


Fig. 3.2.9 Spectrum corresponding to the above 110-fsec pulse

3.3 The passive mode locking of the Pyridin 1 / Rhodamine 800 dye laser.

The lasing band of Rhodamine 800 extends further into the infra-red (to about 835nm) than that of Rhodamine 700 and thus use of it in an energy transfer should extend the spectral region covered by argon-ion pumped passively mode locked dye laser systems to about 820nm. Rhodamine 800 has a structural form similar to that of Rhodamine 700, with the exception of a CN grouping attached to the 9 position of the Xanthene ring instead of the CF₃ group [1]. With absorption coefficients, lifetime and lasing efficiencies similar to Rhodamine 700, at the first instance, an energy transfer system employing Rhodamine 800 as the acceptor dye would be expected to give similar operational characteristics as the previously demonstrated Rhodamine 700 system. Pyridin 1 has the appropriate absorption and fluorescence bands to absorb all-lines argon ion pump radiation and act as a donor to Rhodamine 800. It also has a short upper state lifetime allowing efficient dipole-dipole non-radiative energy transfer (Eqn. 3.3.1). Figure 3.3.1 below shows the absorbance and fluorescence spectra of both Pyridin 1 and Rhodamine 800 separately.

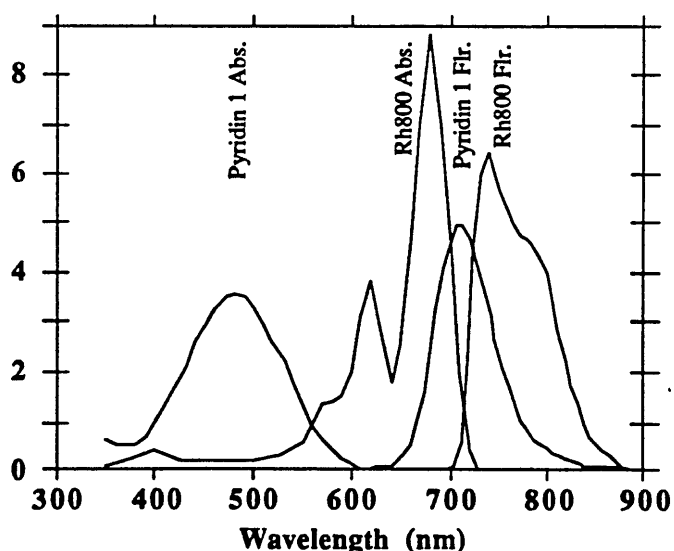


Fig. 3.3.1 The absorbance (Abs.) and fluorescence (Flr.) spectra of Pyridin 1 and Rhodamine 800 separately.

1 R. Raue, H. Harnisch, and K.H. Drexhage, *Heterocycles* 21, 167 (1984)

An active medium comprising a mixture of the two dyes Pyridin 1 and Rhodamine 800 was investigated in a simple three mirror cavity as a possible candidate to exhibit an efficient energy transfer mechanism. The argon-ion pump beam (up to 7.5W all-lines output from a Spectra-Physics Model 2020 laser) was coupled into a 100 μ m vertical free flowing active dye jet (2.6×10^{-3} M Pyridin 1 with 2.3×10^{-3} M Rhodamine 800 in a 1:9 propylene carbonate:ethylene glycol mixture) using a mirror of 50mm radius of curvature. About 84% of the pump power was absorbed by this active medium. A mirror of 100mm radius of curvature acting as a retroreflector and another, also of 100mm radius of curvature, formed the active folded section. The cavity was completed by a plane retroreflector. All the cavity mirrors had a single stack dielectric coating of 100% reflectivity at normal incidence centred around 760nm. Two output beams were provided by the reflections from the surfaces of an intracavity dielectric tuning wedge (Spectra Physics) which was used for wavelength control. Some output was also obtained from the plane mirror.

Figure 3.3.2 is a plot of total output power vs pump power with the cavity described above tuned to 797nm. A lasing threshold of 0.83W and a slope efficiency of 0.76% for the energy transfer mixture in this cavity is thus demonstrated. Figure 3.3.3 shows a plot of the output power from one of the output beams versus wavelength for a pump power of 7.2W. A maximum of 18.5mW average output power at 782nm was obtained and laser operation occurred from 742nm to 823nm. Such a generated tuning range, together with the observation that lasing efficiency increased with increasing Pyridin 1 concentration, is evidence of the energy transfer process contributing to the laser action.

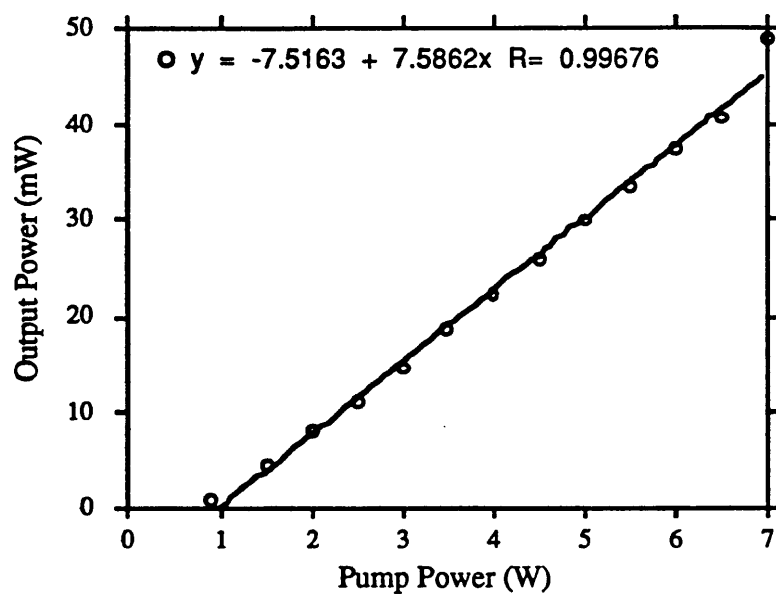


Fig. 3.3.2 Output power vs pump power for the c.w. Pyridin 1/Rhodamine 800 energy transfer dye laser.

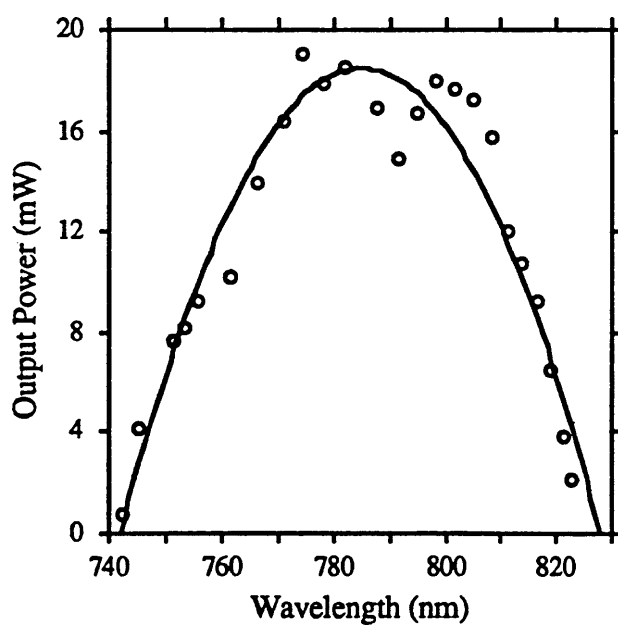


Fig. 3.3.3 Tuning range of the c.w. Pyridin 1/Rhodamine 800 energy transfer dye laser.

This new active medium was then passively mode locked in a five mirror cavity (see figure 3.2.2). The pump, mirror coatings, active dye mixture, tuning wedge and jet thicknesses were as above in the three mirror c.w. cavity. The active folded section however employed tighter focusing to achieve higher efficiencies from the gain medium. Mirror M_1 was of 45mm radius of curvature and mirrors M_2 and M_3 were of 50mm radius of curvature. Mirrors M_4 and M_5 which formed the second folded section around the absorber dye jet were each of 25mm radius of curvature. The cavity round trip time was 7.5nsec. The output beams were employed as for the Rhodamine 700 system. The lasing threshold in this cavity with no absorber in the second jet was 2.1W.

Two saturable absorbers were investigated in this laser system. One of these, 3,3'-Diethylthiatricarbocyanine Iodide (DTTCl), with a peak extinction coefficient of $22 \times 10^4 \text{ L mol}^{-1} \text{ cm}^{-1}$, produced a train of pulses observable on the diode (Fig. 3.3.4) when used at low concentration ($2.5 \times 10^{-5} \text{ M}$). The pump powers needed to be right on threshold (3.9W), and for this reason only very low output powers (about 0.1mW) could be obtained. These output powers were insufficient to enable measurement of the second harmonic so autocorrelation traces could not be taken. If higher pump powers were attempted the laser output became very unstable. This indicated that the absorber had a long recovery time (comparable to the cavity round trip time) thus enabling more than one pulse/noise spike per round trip.

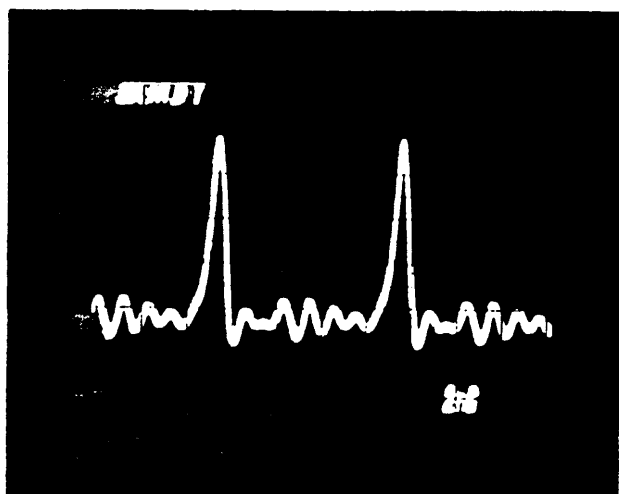


Fig. 3.3.4 Pulse train with DTTCl as the saturable absorber.

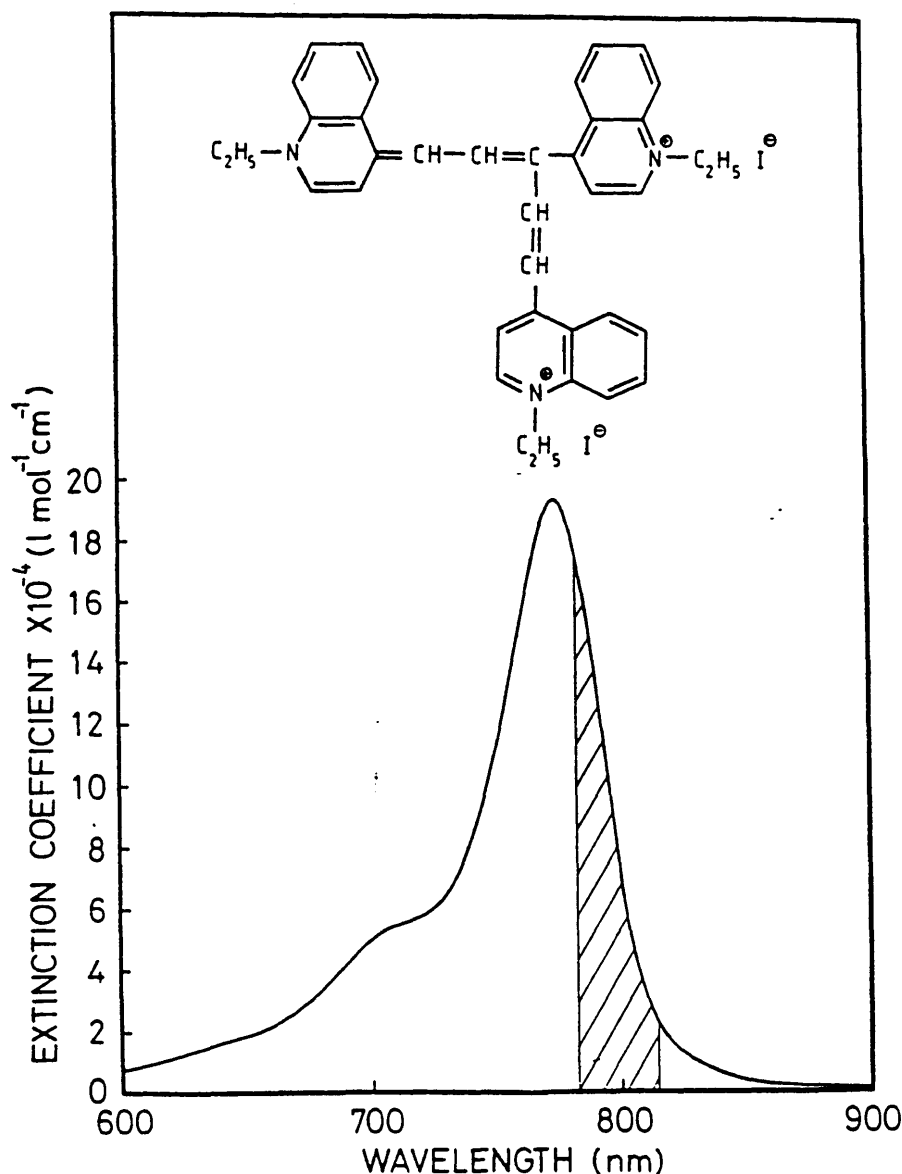


Fig. 3.3.5 Structural formula and absorption profile of Neocyanine. The hatched region represents the tuning range in which 500 fsec pulses were generated.

The other saturable absorber investigated in this laser system was Bis-4-(1-ethylquinoline) (γ -4'-(1'-ethylquinoline)) pentamethine cyanine diiodide. This is commonly known as Neocyanine and had a measured peak extinction coefficient of $19.2 \times 10^4 \text{ L mol}^{-1} \text{ cm}^{-1}$ in a 1:9 ethanol: ethylene glycol solution. Figure 3.3.5 shows the structural formula and recorded absorption profile of this dye. Stable mode locking was observed when absorber concentrations in the laser were varied between $2 \times 10^{-5} \text{ M}$ and $2 \times 10^{-4} \text{ M}$ (giving lasing thresholds from 2.8W to 6W) and pump

powers of between 0.1W and 0.3W above threshold were used (giving average output powers of about 1mW per beam). The shortest pulses obtained with the system were 260fsec at 814nm, and figure 3.3.6 shows an autocorrelation trace of such a pulse. Sub 500fsec pulses were generated from 783nm to 815nm and representative autocorrelation traces at the shortest and longest wavelengths are given in figure 3.3.7. In this cavity no intentional intracavity group-velocity dispersion compensation was present, and measurements of the generated spectra showed some excess bandwidth. Figure 3.3.8 is a typical bandwidth of 2.0nm at 783nm which in the transform limit would be able to support Sech² pulses of 320fsec duration. The corresponding actual measured pulse width (see Figure 3.3.7) was 400 fsec.

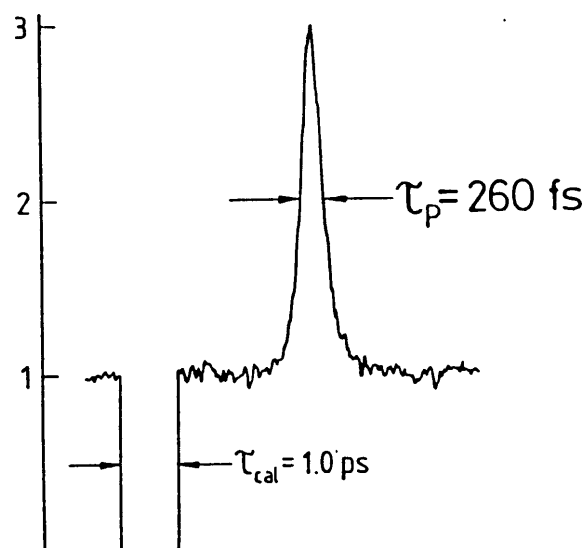


Fig. 3.3.6 Autocorrelation trace of 260fsec pulses obtained at 814nm.

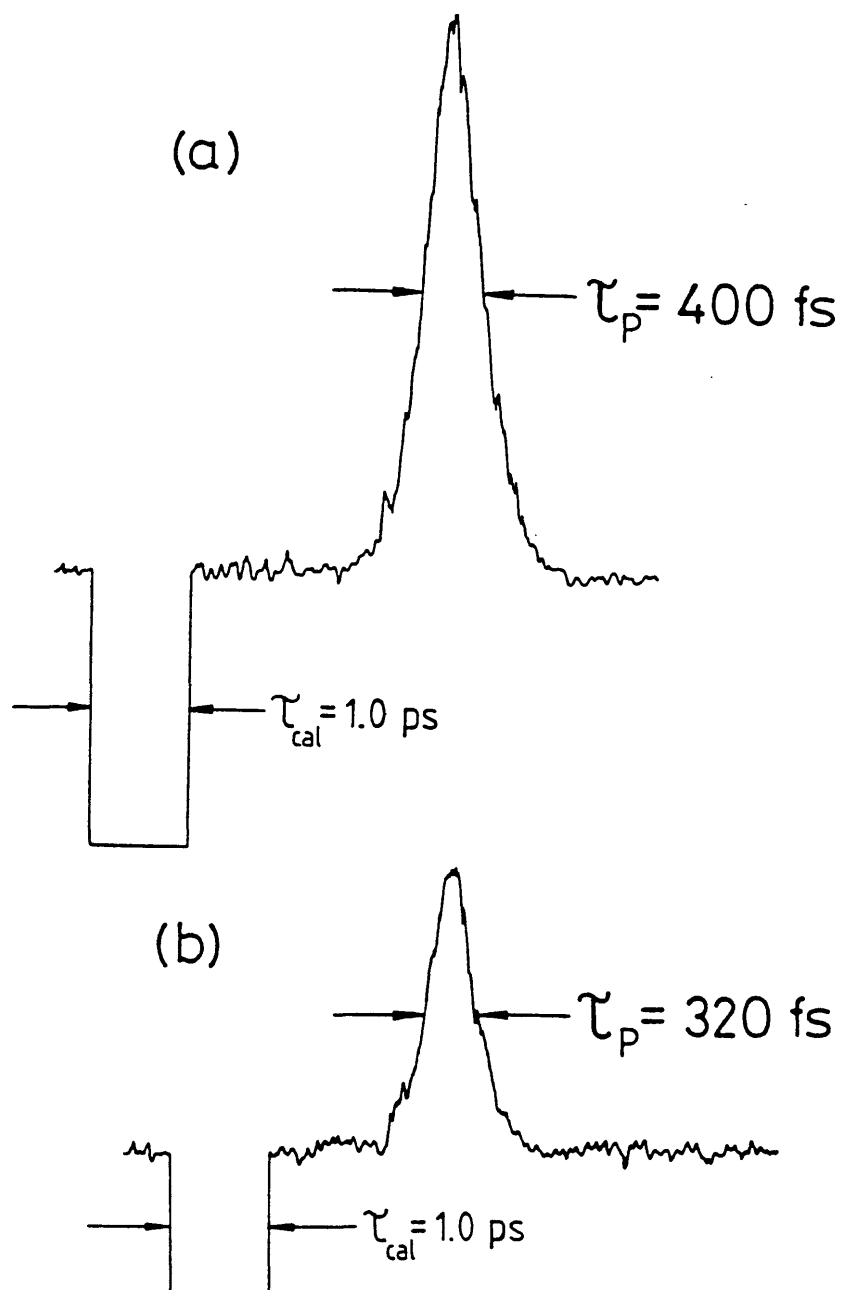


Fig. 3.3.7 Autocorrelation profiles obtained at wavelengths centred at (a) 783nm and (b) 815nm, the extremes of the passive mode locked tuning range.

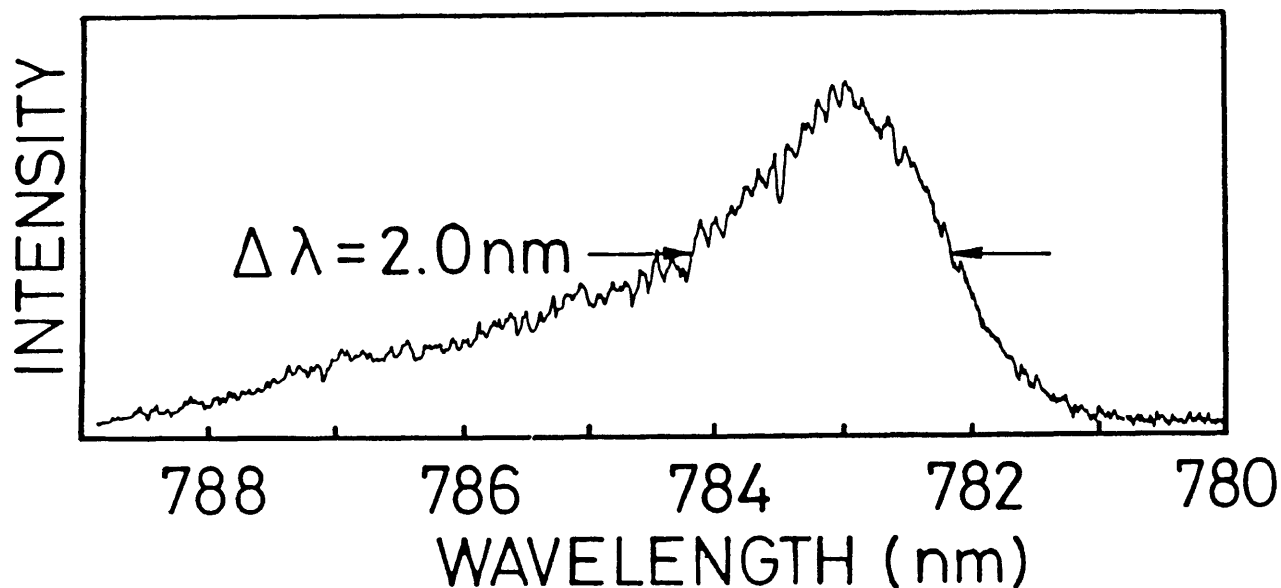


Fig. 3.3.8 A typical pulse spectrum centred on 783nm.

3.4 Conclusions

In conclusion, the spectral coverage of c.w. argon-ion pumped passively mode locked dye lasers has been extended into two new regions in the near infra-red by employing energy transfer dye mixtures as the active media.

The first of these systems used the DCM/Rhodamine 700 energy transfer as the active medium. In a simple linear cavity with no optimisation of the intracavity dispersion this has yielded pulses of less than 400fs duration continuously tunable from 732nm to 761nm. Transform limited pulses as short as 110fs have been obtained from a simple CPM ring laser whose wavelength is tunable through 747nm - of particular interest since frequency tripling would yield sub 100fs pulses at 249nm suitable for amplifying in KrF excimer amplifiers [e.g. 1]. Improved laser performance could perhaps be obtained by using a thinner absorber dye jet to enhance

1 I.A. McIntyre, A.P. Schwarzenbach, T.S. Luk, A. McPhearson, K. Boyer, and C.K. Rhodes in *Digest of Conference of lasers and Electro-Optics* (Optical Society of America, Washington D.C., 1986), paper TUC3

the CPM mechanism and by incorporating a prism sequence [1] to facilitate more precise control of the intracavity GVD.

The second system used a new active lasing medium consisting of an energy-transfer dye mixture of Pyridin 1 and Rhodamine 800 which has been demonstrated to have a c.w. tuning range of 742nm to 823nm, excited using the all-lines pump from an argon ion laser. This has been passively mode locked using Neocyanine as the saturable absorber to produce pulses of less than 500fsec duration tunable from 783nm to 815nm, with pulses as short as 260nm within this region. Since the pulses have been shown to have excess bandwidth, shorter bandwidth limited pulses could perhaps be obtained using a prism sequence [2] to control intracavity dispersion. Because of the inefficiency of the active medium a ring configuration may be unobtainable for this system.

Recent improvements in the performance and reliability of Krypton ion lasers means that in future this pump source could be used to provide more efficient pumping and therefore improved passive modelocking of the Rh700 and Rh800 systems.

1 R.L. Fork, O.E. Martinez, and J.P. Gordon, *Opt. Lett.* **9**, 151 (1984)

4. COMPLEX PULSE EVOLUTIONS IN THE PASSIVELY MODE LOCKED RHODAMINE 110 SYSTEM

4.1 Introduction

The roles of intracavity group velocity dispersion (GVD) and nonlinear frequency chirp have been extensively investigated in passively mode locked CW dye lasers. It has been shown experimentally that shorter pulses can be generated if intracavity GVD is optimised [1,2,3] and that the linear components of GVD can be controlled by use of a prism sequence [4]. Frequency chirp in passively mode locked dye lasers arises primarily from self-phase modulation (SPM) by the Kerr effect (positive chirp) in the solvents and by time dependent saturation of the absorption (mainly negative chirp) of the absorber dye [5,6,7]. The chirp arising in the active section from gain saturation and SPM is less important than that in the passive jet due to the tighter focusing employed in the passive section [7]. Theoretical investigations [8,9,10,11] have shown that if SPM and GVD are of opposite sign then 'soliton-like' pulse shaping can, to a certain extent, give rise to shorter pulses and increased stability compared to if there is no intracavity frequency chirp. This effect is strongest when the contribution from either type of SPM can be made negligible compared to the other, or when both types can be contrived to have the same sign [12,13]. Valdmanis

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- 1 J.C. Diels, J.J. Fontaine, I.C. McMichael, B. Wilhelmi, W. Dietel, D. Kuhlke, *Sov. J. Quantum Electron.* **13**, 1562 (1983)
 - 2 J.A. Valdmanis, R.L. Fork, J.P. Gordon, *Opt. Lett.* **10**, 131 (1985)
 - 3 P.M.W. French, J.R. Taylor, *Opt. Commun.* **61**, 224 (1987)
 - 4 R.L. Fork, O.E. Martinez, J.P. Gordon, *Opt. Lett.* **9**, 150 (1984)
 - 5 J.J. Fontaine, W. Dietel, J.C. Diels, *IEEE J. Quantum Electron.* **QE-19**, 1467 (1985)
 - 6 O.E. Martinez, J.P. Gordon, R.L. Fork, *Ultrafast Phenomena IV*, Springer-Verlag Series in Chemical Physics **38**, 7 (1984)
 - 7 S. de Silvestri, P. Laporta, O. Svelto, *IEEE J. Quantum Electron.* **QE-20**, 533 (1984)
 - 8 O.E. Martinez, R.L. Fork, J.P. Gordon, *Opt. Lett.* **9**, 156 (1984)
 - 9 J.-C. Diels, W. Dietel, J.J. Fontaine, W. Rudolph, B. Wilhelmi, *J. Opt. Soc. Amer. B* **2**, 680 (1985)
 - 10 O.E. Martinez, R.L. Fork, J.P. Gordon, *J. Opt. Soc. Amer. B* **2**, 753 (1985)
 - 11 H.A. Haus, Y. Silberberg, *IEEE J. Quantum Electron.* **QE-22**, 325 (1986)
 - 12 O.E. Martinez, R.L. Fork, J.P. Gordon, *J. Opt. Soc. Amer. B* **2**, 753 (1985)
 - 13 J.A. Valdmanis, R.L. Fork, *IEEE J. Quantum Electron.* **QE-22**, 112 (1986)

et al. [1,2] have successfully exploited intracavity chirp in the laser system Rhodamine 6G modelocked with DODCI and its photoisomer by arranging the pulse energy density to be large compared to the saturation value of the photoisomer but small compared to that of the ground state. Thus one component was deeply saturated by the pulse early on while the other was virtually unsaturated by the pulse. This greatly reduced the time dependent absorber saturation (which occurred very early on in the pulse) and allowed the positive SPM from the Kerr effect to predominate - this result was apparent in the very asymmetric dependence of pulse width on intracavity GVD.

French and Taylor [2,3] in investigating a CPM dye laser employing Rhodamine 110 as the gain medium with a HICI/DASBTI saturable absorber observed unexpected behaviour in the way the autocorrelation traces of the laser output varied with intracavity glass path. Similar behaviour was reported by Wise et al. [4] and Valdmanis et al. [2] in the Rhodamine 6G/DODCI system. What was called 'Soliton-like' behaviour was observed in a standard Rh 6G/DODCI CPM laser by Salin et al. [5], where a periodic modulation of the laser output similar to an N=3 soliton was observed together with a strong modulation of the laser output at about 70kHz which they considered to be twice the soliton period. Recently results very similar to those mentioned here were observed in systems using Rhodamine 6G and Sulphur Rhodamine 101 mode-locked with DQTCI [6]. Theoretically Diels et al [7] have employed a soliton description in investigating the cycling of the pulse in the cavity.

In a fibre the soliton pulse evolutions are governed by the nonlinear Schrodinger equation, however in a CPM laser the GVD and SPM components are discretely distributed in the cavity (as compared to continuously distributed as in a fibre), and the effects of gain and absorber saturation also occur (again in discrete elements). It would therefore be expected that solutions of the

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- 1 J.A. Valdmanis, R.L. Fork, J.P. Gordon, *Opt. Lett.* **10**, 131 (1985)
 - 2 P.M.W. French, J.R. Taylor, *Opt. Lett.* **11**, 297 (1986)
 - 3 P.M.W. French, J.R. Taylor, *Opt. Commun.* **61**, 224 (1987)
 - 4 F.W. Wise, I.A. Walmsley, C.L. Tang, *Opt. Lett.* **13**, 129 (1988)
 - 5 F. Salin, P. Grangier, G. Roger, A. Brun, *Phys. Rev. Lett.* **56**, 1132 (1986)
 - 6 P. Georges, F. Salin, G. Le Saux, G. Roger, A. Brun, *Opt. Commun.* **69**, 281 (1989)
 - 7 J.-C. Diels, W. Dietel, J.J. Fontaine, W. Rudolph, B. Wilhelmi, *J. Opt. Soc. Am. B* **2**, 680 (1985)

nonlinear Schrodinger equation would not be a totally accurate description of CPM pulse evolutions.

The work in this chapter is an attempt to achieve a more complete description of CPM behaviour including the observed instabilities. I shall start by describing the characterization of the passively mode locked Rhodamine 110 CPM dye laser cavity, and the details of an experimental investigation into the roles of GVD and SPM in this cavity. Subsequently the experimental results will be compared with those from a detailed numerical model of H. Avramopoulos [1], and the conclusions will be drawn.

4.2 The linear cavity

In order to investigate the properties in passively modelocked dye lasers of dielectric mirrors used away from normal incidence, the dispersion compensated passively mode locked linear cavities shown schematically below (fig. 4.2.1) were constructed.

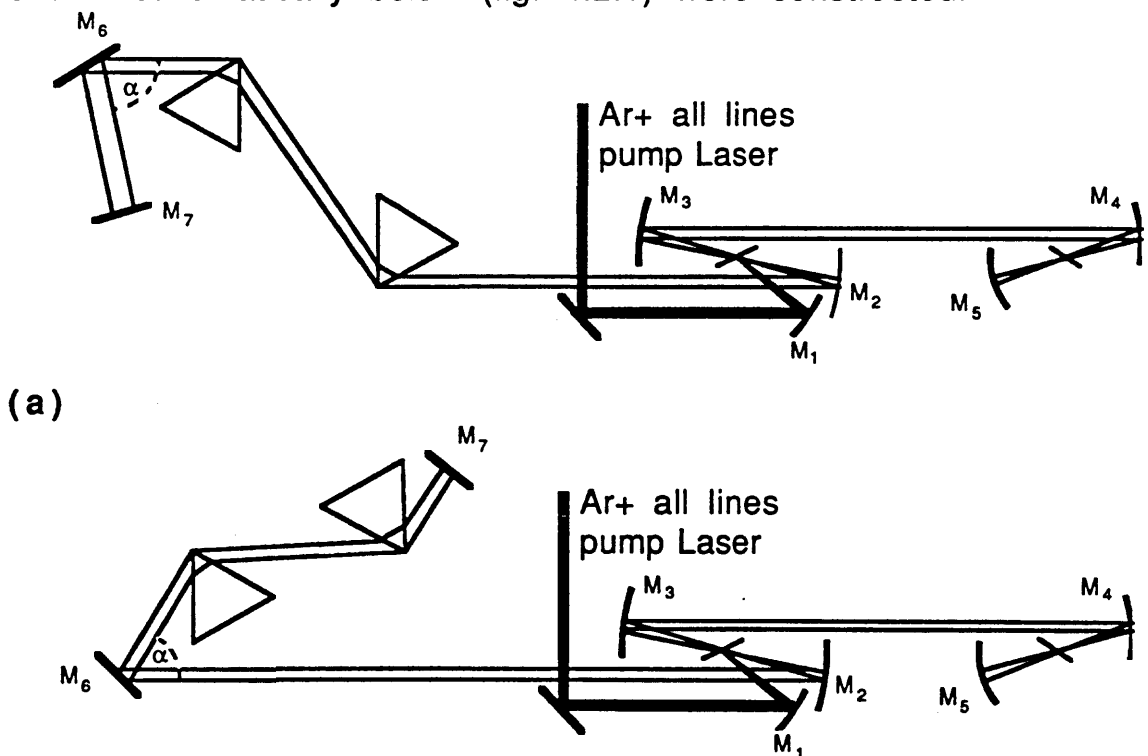


Fig. 4.2.1 Dispersion compensated linear cavities

1 H. Avramopolous PhD Thesis, London University (1989)

All the mirrors M_1 - M_7 supported single stack dielectric coatings of 100% reflectivity centred on 560nm. The active folded section consisted of two mirrors (M_2 and M_3) of 100mm radius of curvature about a vertical dye jet. The passive section had focusing mirror M_4 of 50mm radius of curvature, and retroreflecting mirror M_5 of 25mm radius of curvature. The cavity round trip time was 11ns in cavity (a) and 10ns in cavity (b), and the prism separation was 350mm in each case. The prisms were cut for minimum deviation operation at Brewsters angle and were made from fused silica. These dye laser configurations as well as the subsequent ring cavities were pumped by the all-lines visible output of a small frame Spectra-Physics argon ion laser (model 2020) which gave up to 7W cw output power. The pump radiation was coupled into the gain medium (2×10^{-3} M Rhodamine 110 dissolved in a 2:10 ethanol:ethylene glycol solvent mix) via mirror M_1 of 50mm radius of curvature. Tuning was accomplished by using mirror M_6 away from normal incidence to act as a long wavelength cut-off filter and to give two output beams, and dispersion control was accomplished by moving one of the prisms along the axis perpendicular to it's base. One of the laser outputs was focused onto a fast photodiode (BPW 28) enabling it to be monitored on a fast oscilloscope, while the other enabled pulse width measurements by real time collinear second harmonic autocorrelation using 2mm thick Urea as the nonlinear crystal. Sech² pulses were assumed in interpreting the autocorrelations near the position of optimum glass path.. The saturable absorber used was 1,1,3,3,3',3'- hexamethylindocarbocyanine iodide (HICI). It's absorption profile for a 3:1 ethylene glycol:ethanol solution is given in fig. 4.2.2 opposite.

In cavity (a) for a fixed concentration of 3.7×10^{-4} M HICI the pump threshold power and the output wavelength and pulse duration for the optimum intracavity glass path were monitored as angle α was varied. The results are given in table 4.2.3.

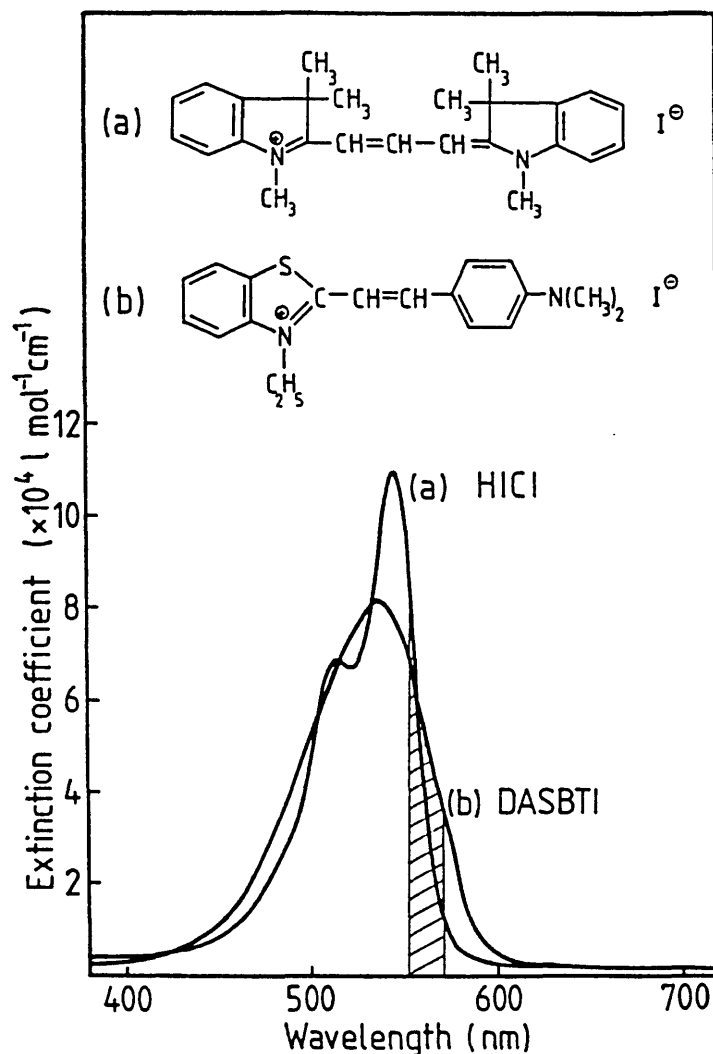


Fig. 4.2.2 Structural formulas and absorption profiles of (a) HICI and (b) DASBTI. The shaded areas indicate the tuning range in which sub-500fs pulses were obtained.

Angle α Degrees	Wavelength (nm)	Pulse Duration (fs)	Threshold Pump Power (W)	Optimum Glass Path (mm)
61	581	-	2.8	8
68	578	250	2.4	8
75	578	260	3.2	9.5
80	579	240	3.3	10.5
85	575	220	4.2	11
89	570	150	5.4	11.5

Fig. 4.2.3 Table of variation of pump threshold power, output wavelength and pulse duration at the optimum intracavity glass path vs angle.

From these results it can be seen that with increasing angle the wavelength of the laser becomes shorter as the long wavelength cut off of the mirror moves to shorter wavelengths. As the wavelength moves towards the peak absorption of the saturable absorber, so mode locking improves, producing shorter pulses. The negative dispersion arising from the mirror also increases, and this must be compensated for with increasing glass path.

In order to ensure that the spatial dispersion of frequencies which occurs on the output beams of cavity (a) due to the prism does not affect the autocorrelation measurement cavity (b) was used for further measurements. In this cavity it was impractical to change the angle α often since this involves moving the prisms, so it was set to 77° , about the mid range of the angles investigated for cavity (a), and the absorber concentration was varied. Passive mode locking was observed for HICI concentrations from $2 \times 10^{-4} \text{M}$ to $1.4 \times 10^{-3} \text{M}$ (where there was insufficient pump power to continue increasing the concentration). The graph below (Fig. 4.2.4) shows how pulse duration and wavelength varied with increasing saturable absorber concentration. The optimum glass path remained at around 8.5mm for all these results. A typical autocorrelation trace is given in figure 4.2.5 - 130fs at 579nm with $7 \times 10^{-4} \text{M}$ HICI and 8mm of intracavity glass with a prism separation of 347mm, and angle $\alpha = 77^\circ$. The optimised laser was pumped about 1W above lasing threshold to give typical output powers of about 7mW. From these results we can see how adding the absorber forces the laser to longer wavelengths until the long wavelength cut off of the dielectric mirror takes effect, at which point the threshold of the laser starts increasing as the loss in the cavity goes up. As more saturable absorber is added the pulses get shorter until the limit is reached. At high absorber concentrations the threshold is so high that there is insufficient pump power to maintain stable mode locking action.

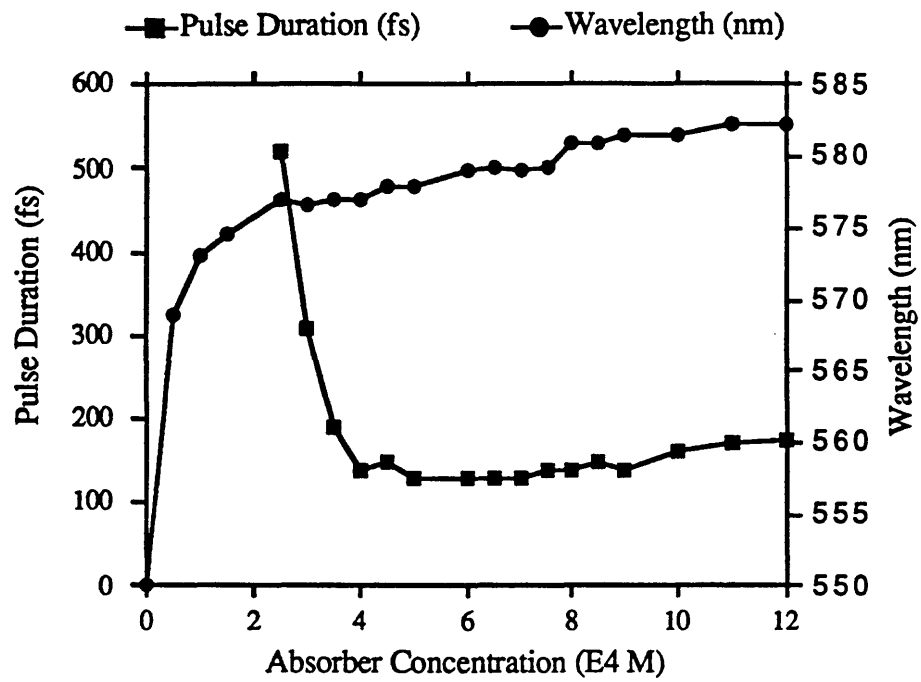


Fig. 4.2.4 Graph showing how wavelength and pulse duration vary with increasing absorber concentration in a dispersion compensated linear cavity.

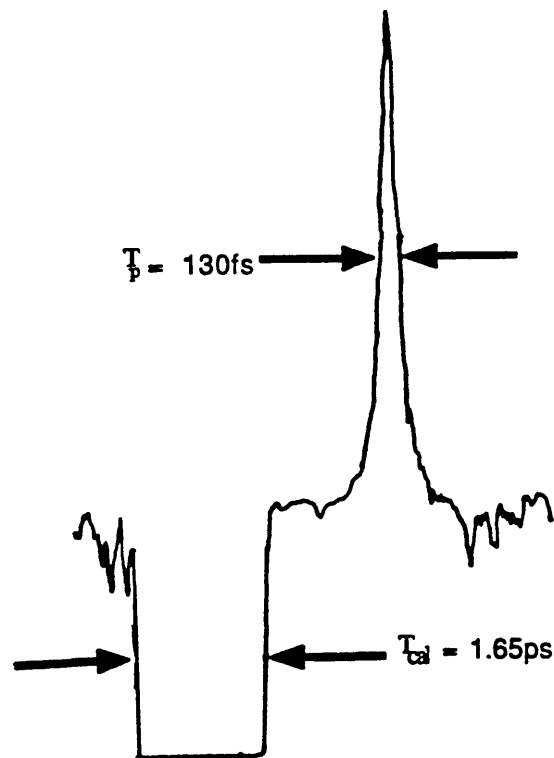


Fig. 4.2.5 130fs Autocorrelation from optimised dispersion compensated linear cavity.

4.3 The CPM ring cavities

The CPM ring cavity shown below without dispersion compensation was initially set up. All the mirrors had a single stack dielectric coating of 100% reflectivity. Mirrors M_2 to M_6 , and M_8 were coated for a central wavelength of 550nm, and mirror M_7 was coated for 600nm. The active and passive folded sections used mirrors of 100mm and 50mm radius of curvature respectively. The mirrors M_5 - M_7 were used away from normal incidence to act as long wavelength cut off filters, become dispersive elements, and to provide output coupling. Mirror M_7 had a coating for a longer wavelength (at normal incidence) than the others so that when used at a large angle it would not present an excessive loss, or introduce a large amount of dispersion. The cavity round trip time was 8ns. Optimisation of the cavity angles and the absorber concentration yielded pulses of 110fs duration at 574nm (Fig. 4.3.2). For this result the angles α, β, γ were $20^\circ, 90^\circ, 70^\circ$ respectively and the concentration of the saturable absorber, HICI, was $1.5 \times 10^{-4} \text{M}$.

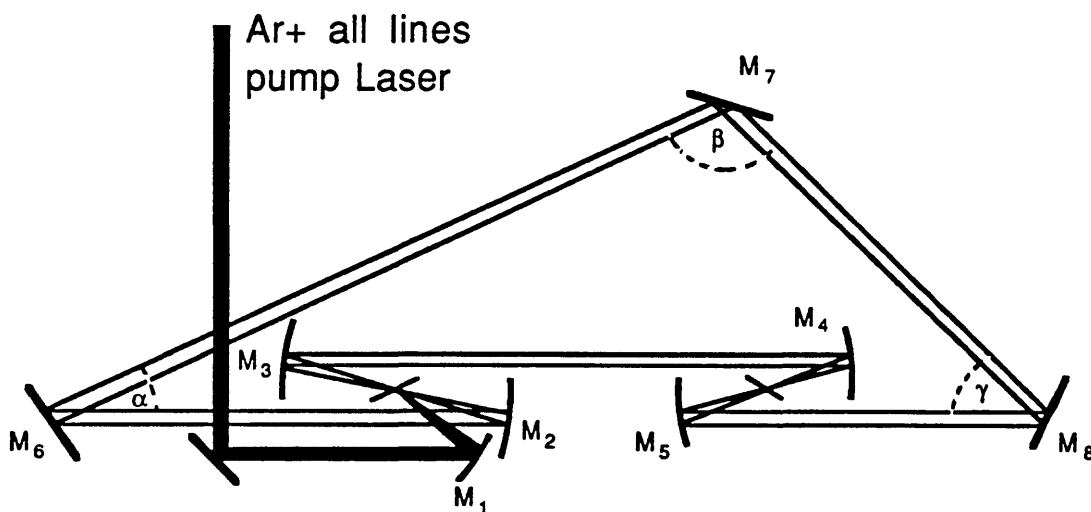


Fig. 4.3.1 Simple CPM ring cavity

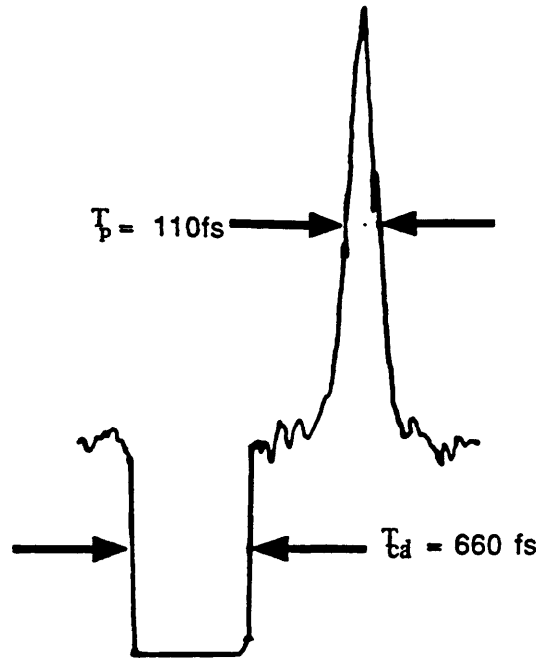


Fig. 4.3.2 Autocorrelation trace of 110fs pulse from a simple CPM ring cavity

Having ascertained the optimum angles for the mirrors using the above cavity, a prism sequence was inserted to give dispersion control. The resulting cavity is as shown below (figure 4.3.3). The addition of Brewster angled fused silica prisms (with separation $L = 26\text{cm}$) increased the cavity round trip time to 8.5ns

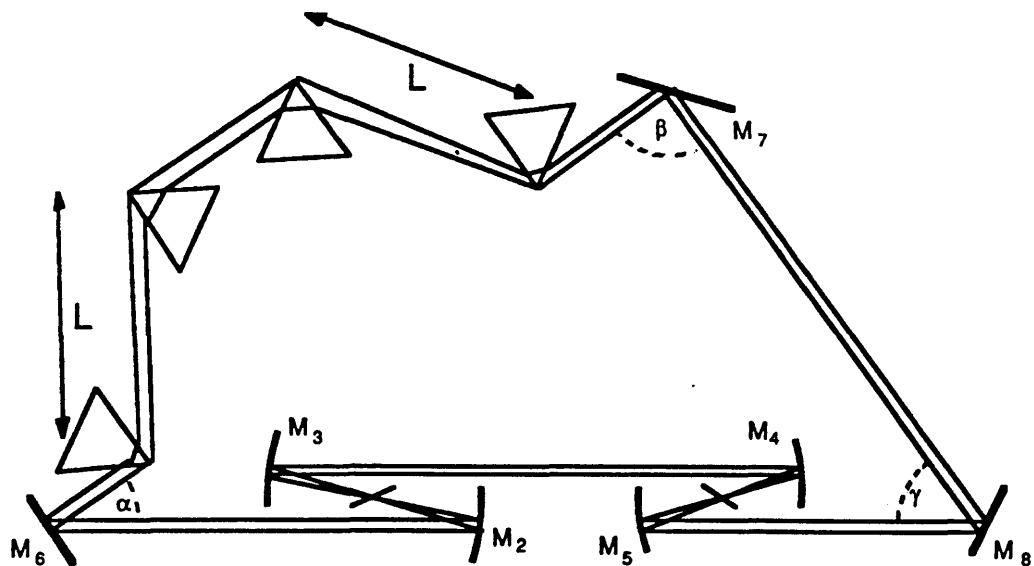


Fig. 4.3.3 Dispersion Compensated CPM ring cavity

The gain medium, cavity mirrors and pump source were kept the same as for the previous cavity. Optimisation of this cavity lead to

the generation of pulses as short as 70fs at a wavelength of 573nm as shown in the autocorrelation trace 4.3.4 and it's bandwidth 4.3.4 below, for an absorber concentration of $0.24 \times 10^{-3} \text{M}$ HICI. The pump power was 5.5W for these pulses - 1W above threshold and there was 13.75mm of intracavity glass introduced by the prisms.

For pulses this short it is important that in the autocorrelator there is no phase matching restriction on the bandwidth of second harmonic generation arising from group velocity mismatch between fundamental and second harmonic [1]. Thus for these and subsequent pulse duration measurements a thin (100 μm thick) Lithium Iodate crystal was used to minimize this.

Next an investigation was carried out to determine if either the autocorrelation traces or the spectral traces of the pulses varied at outputs from different parts of the cavity. This was done by taking simultaneous measurements of the outputs from the angled mirrors M_6 - M_8 . No obvious differences were noticed, and after statistical comparison over several measured parameters of the traces no significant differences could be found.

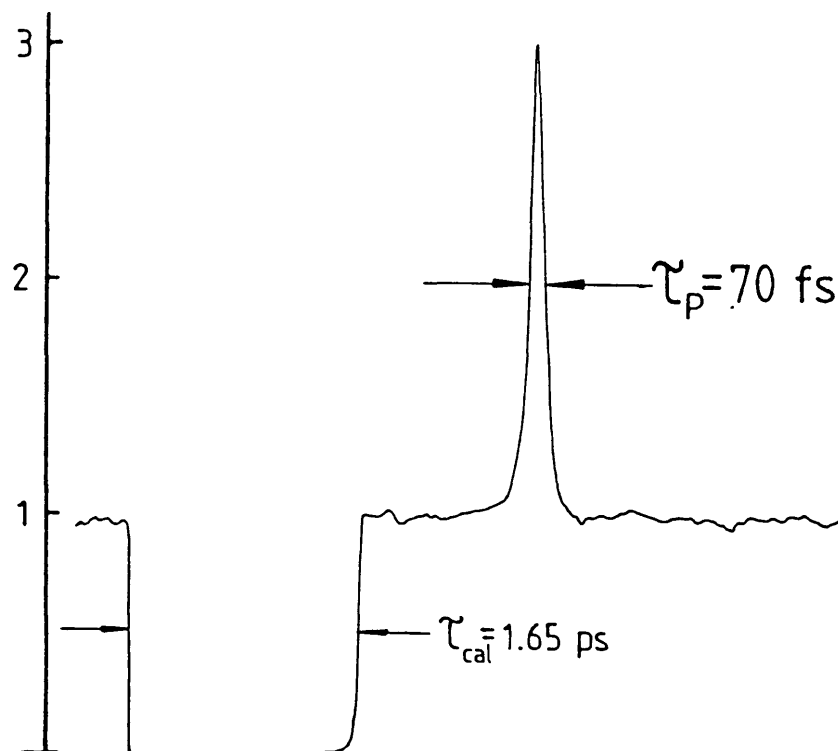


Fig. 4.3.4 Autocorrelation of a 70fs pulse from the dispersion compensated CPM cavity.

1 R.C. Miller, *Physics Letters* **26A**, 177 (1968)

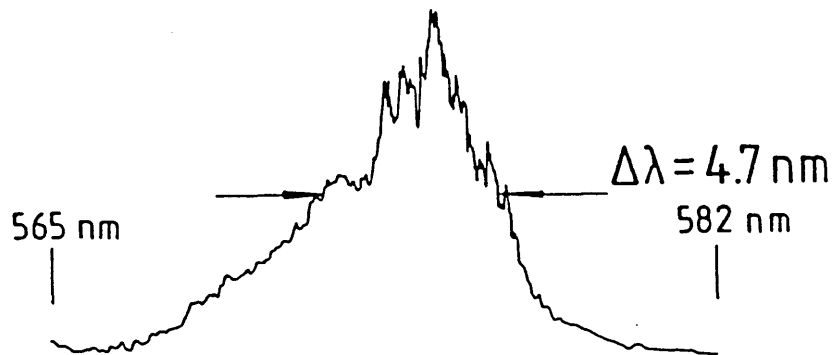


Fig. 4.3.5 Bandwidth corresponding to autocorrelation trace 4.3.4 above.

Subsequently, an investigation into the frequency chirp of the pulses arising at different values of glass path was carried out. This was done by comparing the autocorrelation traces of the pulses coming directly from the laser with those which had traveled through a 13.3cm long dispersive glass rod. The results are presented in the graph 4.3.6 below, and it is apparent that for lower glass paths in the cavity the pulses have a negative chirp which can enable compression of them in the extracavity glass. For increasing intracavity glass the chirp of the pulses decreases so that the difference between the cases of with and without extracavity glass decreases. For the optimum intracavity glass (shortest pulses) the extracavity glass leads to considerable dispersion broadening, however as excess intracavity glass is introduced the difference becomes smaller again. This is because the longer laser pulses entering the glass rod have a narrower bandwidth, and thus suffer less dispersive broadening (although of course the pulses are always broadened by the extracavity glass).

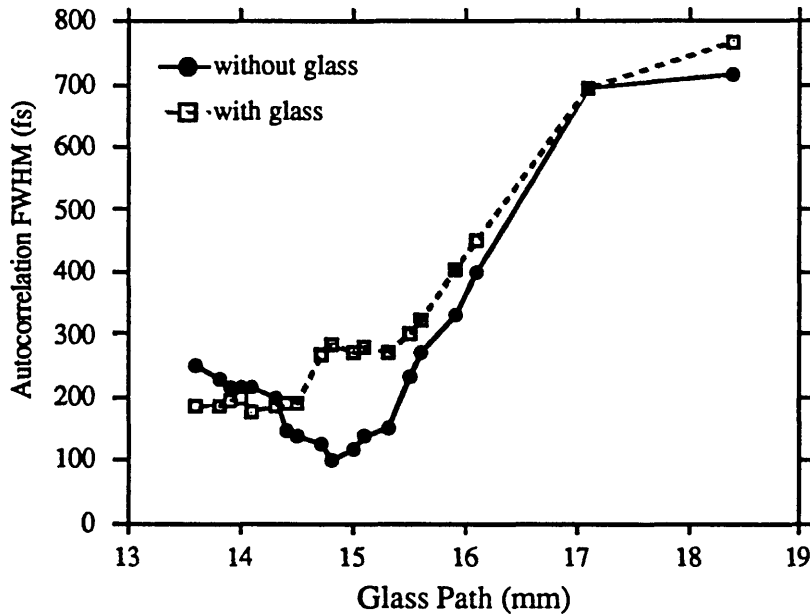


Fig. 4.3.6 Plot of Autocorrelation FWHM vs Intracavity Glass Path for the cases of with and without extracavity glass.

It has been found experimentally [1] that the stability of this CPM system can be improved if a composite absorber of HICI and DASBTI (2-(p-dimethyl aminostyryl)-benzthiacarbocyanine iodide) is used rather than just HICI alone, and so the composite absorber was employed for most of the subsequent experiments described here. The absorption profile of DASBTI is given in figure 4.2.1.

A characterization was carried out of how the pulse duration at optimum glass path, and the glass path itself, varied with the position of the passive jet in it's folded section. These results are presented below, and due to the symmetry of this curve it is suspected that the minimum pulse durations are obtained when the absorber jet is at the centre of it's focus where the optical energy density is at a maximum. If this were not the case two minima would be expected - one either side of the focus. The amount of intracavity glass required is at a maximum here (i.e. the amount of negative dispersion required is at a minimum). By comparison with a pulse compressor [2] where the required compressor factor (i.e. dispersion - either sign) is inversely proportional to the spectral

1 P.M.W. French, J.R. Taylor, *Opt. Lett.* **11**, 297 (1986)

2 W.J. Tomlinson, R.H. Stolen, C.V. Shank, *J. Opt. Soc. Am. B* **1**, 139 (1984)

broadening of the pulse (i.e. amount of frequency chirp picked up in the fibre) and where the compression is proportional to the spectral broadening we can deduce that the pulse chirping in the jet is mainly due to the positive Kerr effect rather than the negative time dependent saturation of absorption. Thus with the absorber jet at the centre of the focus, with the maximum energy density focused onto it, the induced nonlinear chirp is at a maximum, the required compensating negative dispersion is at a minimum and the pulse duration is at a minimum.

The absorber concentrations used for this experiment were $2 \times 10^{-3} \text{M}$ HCl with $7 \times 10^{-5} \text{M}$ DASBTI.

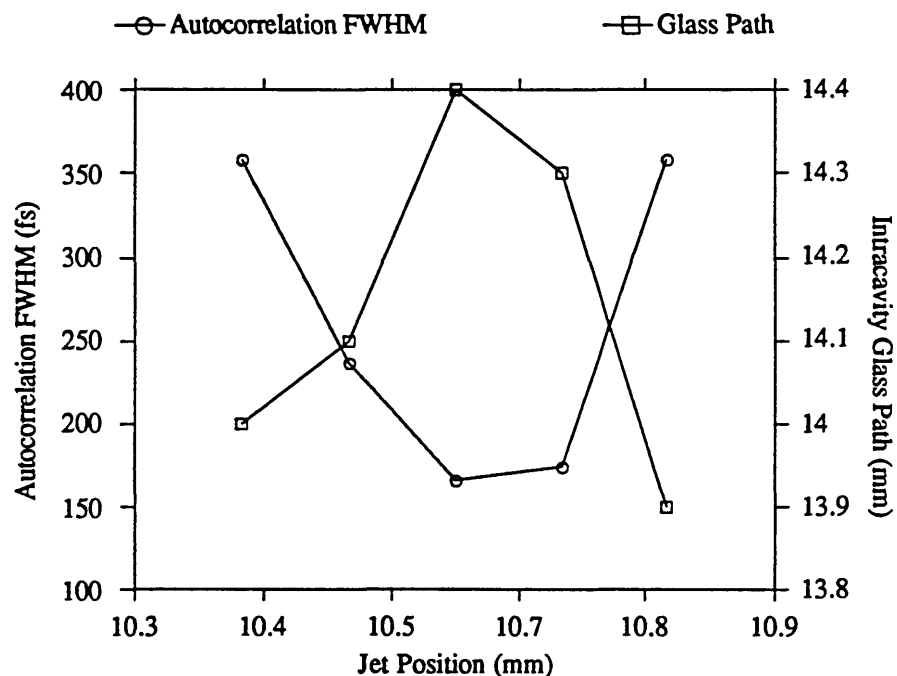


Fig. 4.3.7 A plot of autocorrelation FWHM, and optimum glass path vs Absorber Jet Position.

After the initial cavity characterization a detailed investigation into the complex pulse evolutions which have previously been reported in this [1,2] and other [3] CPM systems was carried out. For a fixed composite absorber (10^{-3}M HCl and $2 \times 10^{-4} \text{M}$ DASBTI) the variation of various parameters of the laser output with changing intracavity glass path was recorded (other cavity parameters were

- 1 P.M.W. French, J.R. Taylor, *Opt. Commun.* **61**, 224 (1987)
- 2 P.M.W. French, J.R. Taylor, *Opt. Lett.* **11**, 297 (1986)
- 3 F. Salin, P. Grangier, G. Roger, A. Brun, *Phys. Rev. Lett.* **56**, 1132 (1986)

held fixed). The series of autocorrelation profiles are given in figure 4.3.11, with a corresponding plot of the FWHM autocorrelation width against intracavity glass path is given in figure 4.3.8 below. It is noticeable that this plot is fairly symmetrical contrasting to the results from the Rh6G/DODCI system of Valdmanis et al. [1,2]. This suggests that neither positive nor negative SPM is strongly dominant, and perhaps explains why the minimum pulse widths are longer than those of [1]. The very slight asymmetry which does exist indicates that overall the Kerr effect is a little stronger than the time dependent absorption saturation effect. For values of intracavity glass path between 14.9 and 15.5mm (traces d,e,f in figure 4.3.11) satellite peaks occur on the autocorrelation profiles indicating the presence of a secondary pulse. As the glass path is increased this secondary pulse moves in towards the central peak and eventually merges with it. This occurs at a value of approximately 15.5mm for the intracavity glass, and gives rise to the step like feature in figure 4.3.8. It is sometimes possible to see a very weak tertiary pulse further out in autocorrelation trace as this occurs. Also at this point of merger the output power of the laser exhibits a strong periodic modulation at a frequency of about 100kHz. This modulation can also be seen on the spectrum of the pulses. Having a composite HICI/DASBTI absorber rather than just HICI alone enhances these effects considerably. A plot of how typically the modulation frequency varied with glass path is given in figure 4.3.9. At the point of autocorrelation trace (d) the modulation was periodic as shown on oscillograph (a) in figure 4.3.10, while at point (f) the laser output was irregular as shown in oscillograph (b).

1 J.A. Valdmanis, R.L. Fork, J.P. Gordon, *Opt. Lett.* **10**, 131 (1985)

2 J.A. Valdmanis, R.L. Fork, *IEEE J. Quantum Electron.* **QE-22**, 112 (1986)

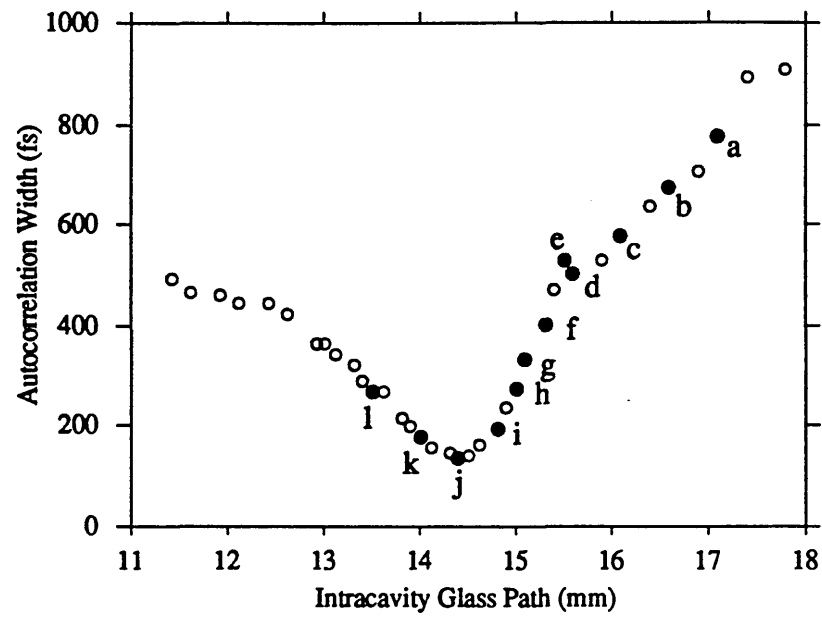


Fig. 4.3.8 Variation of the autocorrelation FWHM of the laser pulses with intracavity glass path. The points labeled (a) to (l) correspond to the photographs in figure 4.3.11.

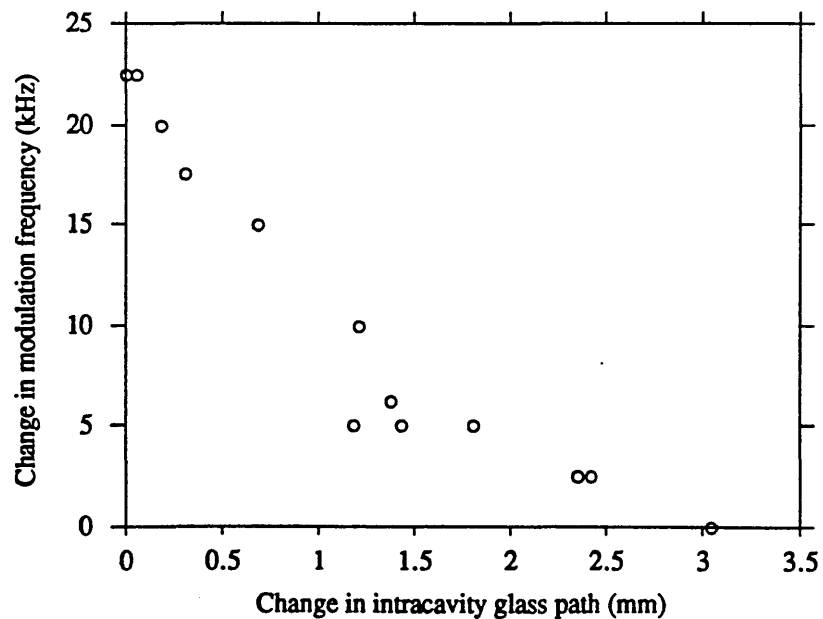


Fig. 4.3.9 Typical variation of the modulation frequency of the laser output with intracavity glass path

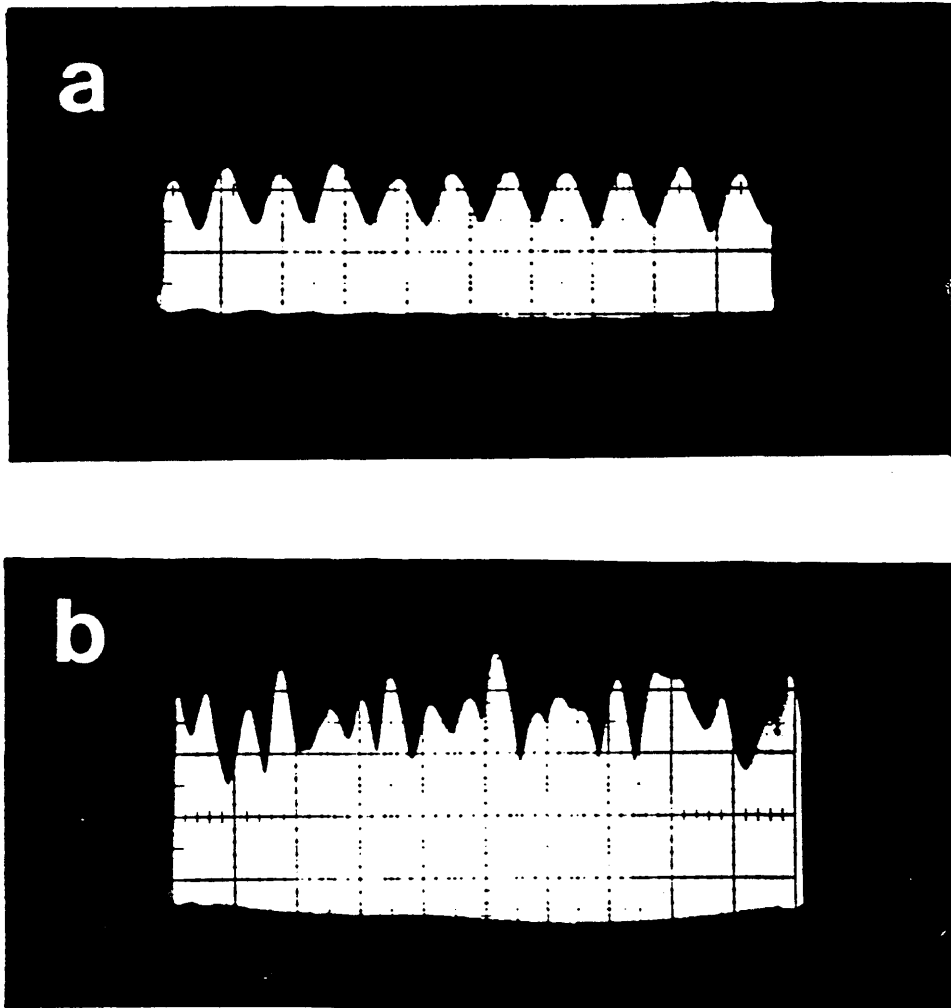


Fig. 4.3.10 (a) Oscillographs of laser output (with 10ms/div) for the values of intracavity glass path corresponding to (a) trace d and (b) trace f

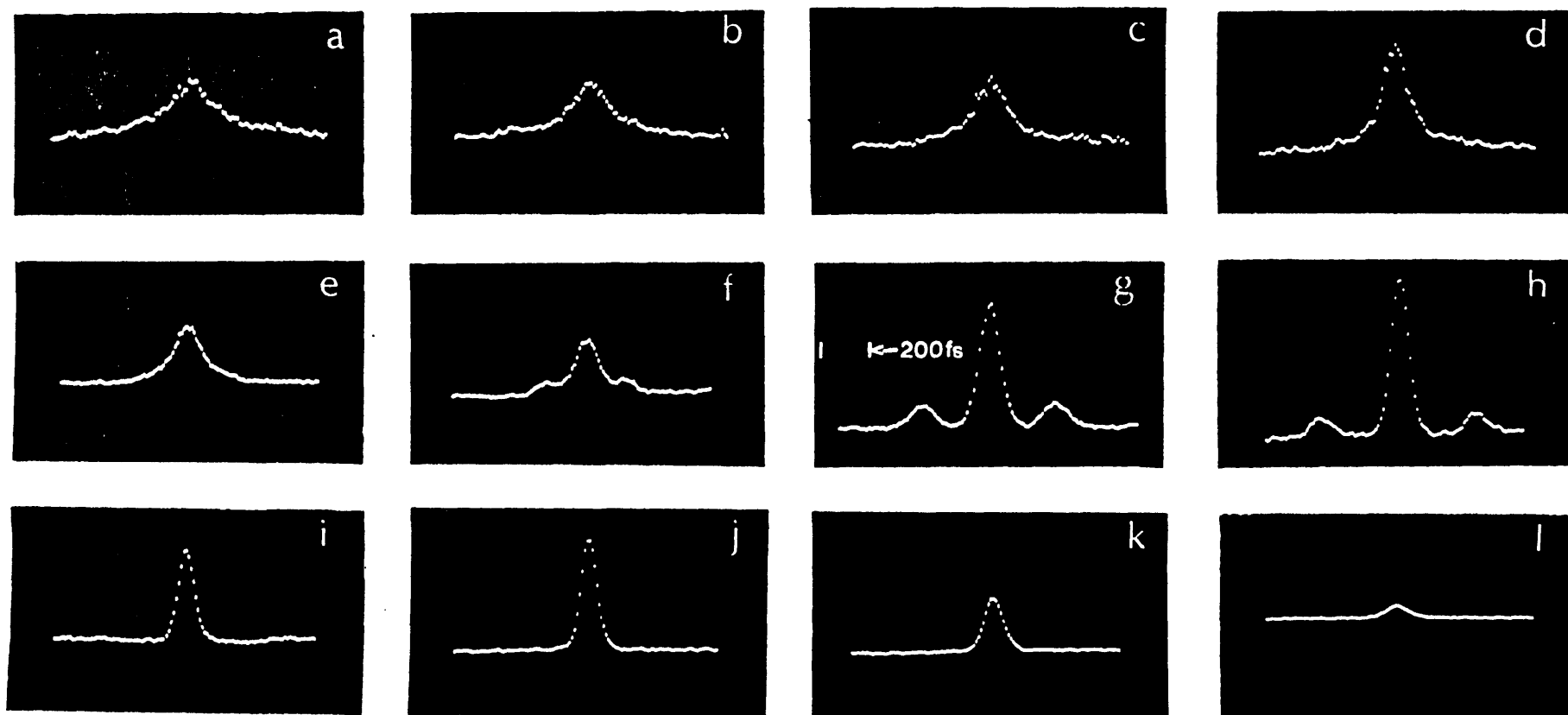


Fig. 4.3.11 Autocorrelation traces (a) to (l) recorded as the intracavity glass path was decreased

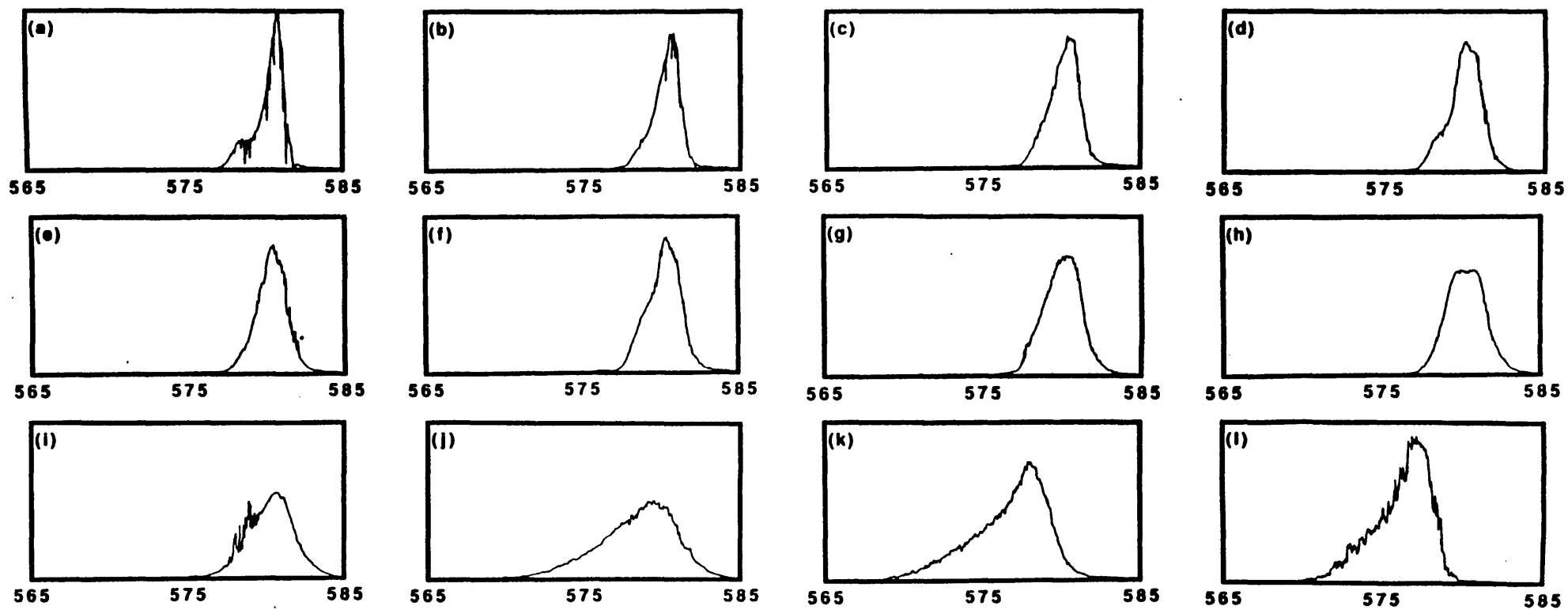


Fig. 4.3.12 The measured power spectra (a) to (l) recorded as the intracavity glass path is decreased, corresponding to the autocorrelation traces in figure 4.3.11

The observation of secondary peaks resembles the results of Salin et al. [1] for the standard Rhodamine 6G/DODCI laser. These results were interpreted as higher order solitons, and a modulation on the laser output was observed which was used to provide a trigger signal for a gating experiment using a boxcar averager. The results from this showed a sequence of traces similar to those that would occur for a $N=3$ soliton, with an inferred soliton width of 500fs. In the case presented here, however, no significant modulation of the laser output on a kHz time scale was observed with intracavity glass paths corresponding to autocorrelation traces (g) and (h). At those points where modulation on the laser output was observed (traces (d) to (f)) a gating experiment similar to that of Salin et al. was attempted in order to try and observe any temporal periodic modulation of the autocorrelation profile which may exist. The modulation signal from the diode monitoring the laser output was filtered with an LC filter, and triggered a voltage comparator. The resulting logic signal was divided down to drive a boxcar averager which then gated the signal from the autocorrelator. The case of the most periodic modulation which had the largest modulation depth and which could therefore provide the best triggering corresponds to autocorrelation trace (d) (see fig. 4.3.10 (i)) where it is clear that any satellite pulse would be of low intensity compared to the main peak. The situation corresponding to trace (f) where the satellite peaks are clear unfortunately corresponds to a much less periodic weaker modulation (see fig. 4.3.10 (ii)) on the laser output. The best results that could be obtained from this experiment, corresponding to autocorrelation trace (e) are shown in figure 4.3.13 below. The boxcar was triggered at 14kHz and the autocorrelation traces (a) and (b) were recorded at the peak and trough of the modulated laser output.

The spectra recorded simultaneously with the autocorrelation profiles of figure 4.3.11 are shown in figure 4.3.12. A plot of the bandwidths measured from the spectra, against intracavity glass path is given in figure 4.3.14, and a plot showing how the central wavelength of the laser output varied with intracavity glass path is given in figure 4.3.15. This wavelength was measured at the

1 F. Salin, P. Grangier, G. Roger, A. Brun, *Phys. Rev. Lett.* **56**, 1132 (1986)

centre of the half intensity mark of the spectra rather than at the peak wavelength to take into account the possible effect of asymmetry in the traces.

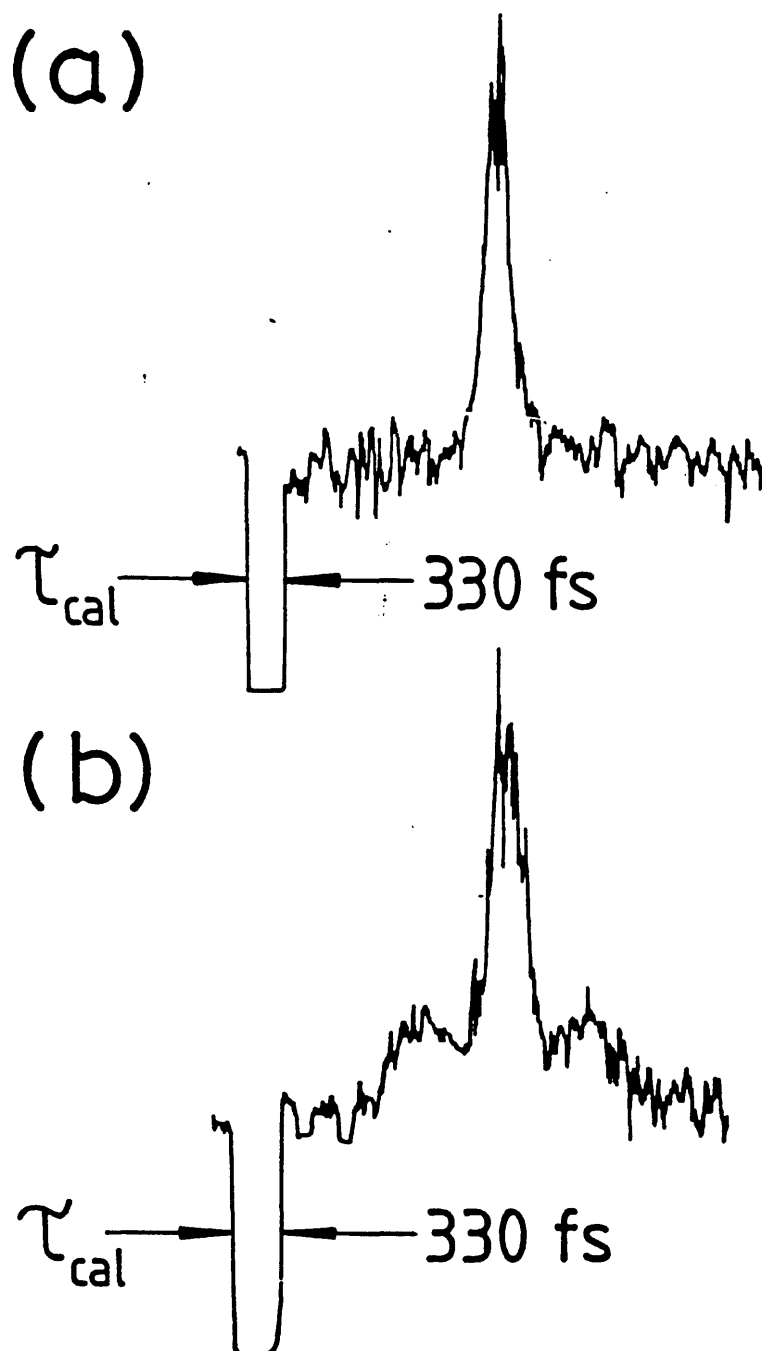


Fig. 4.3.13 Autocorrelation profiles recorded using a boxcar averager gated at 14kHz for the value of intracavity glass path corresponding to autocorrelation trace (e). (a) was recorded at the peak of the 100kHz modulation, and (b) at the trough.

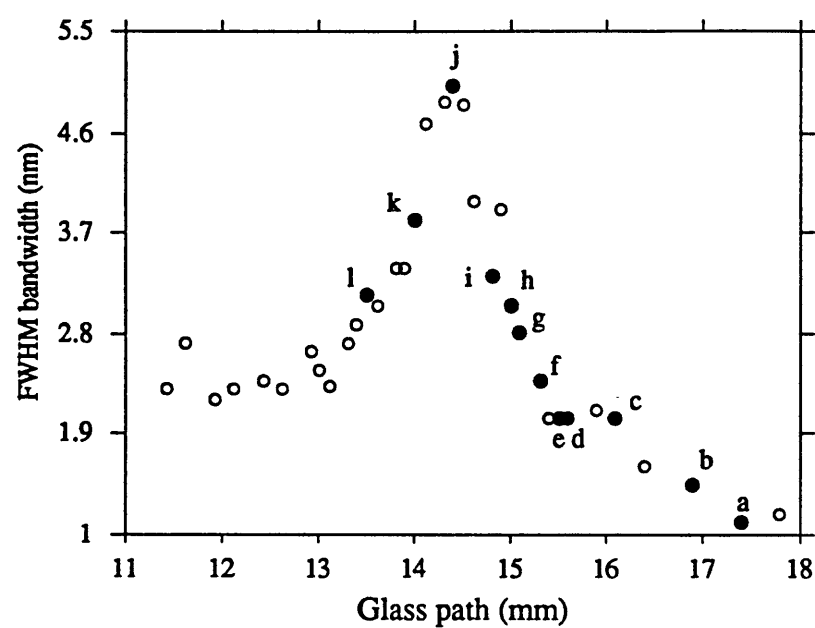


Fig. 4.3.14 The variation of bandwidth with intracavity glass.

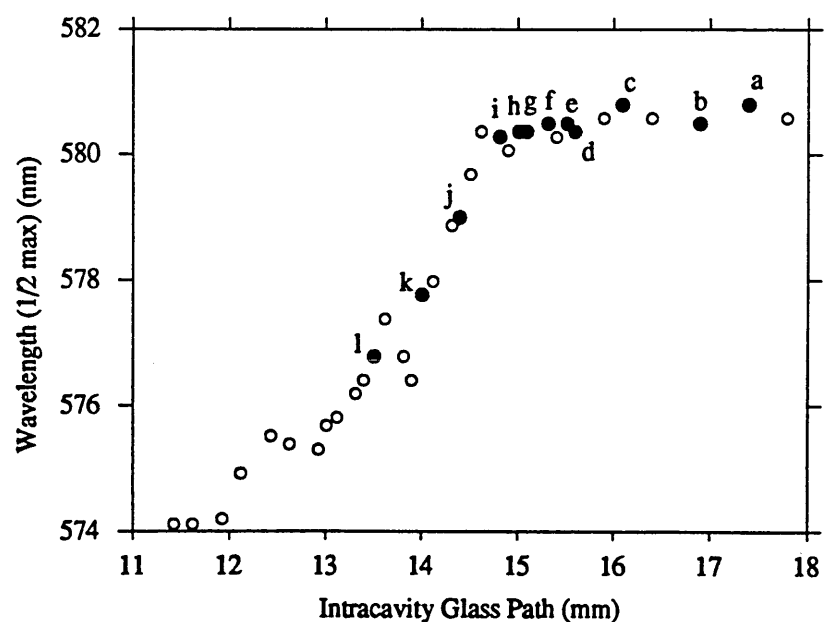


Fig. 4.3.15 The variation of 'centre' wavelength with intracavity glass.

4.4 Introduction to the numerical model

In order to interpret the behaviour of C.P.M. dye lasers in general, and the Rhodamine 110 system mentioned in this chapter in particular H. Avramopoulos [1] constructed an evolutionary numerical model which included the elements shown schematically in figure 4.4.1 overleaf. Due to the discrete nature of the model the separate processes can be described by a set of algebraic equations (as compared to a set of coupled non-linear partial differential equations required for a continuous medium exhibiting the processes). This enables the effect of each process on the pulse evolution to be isolated and studied easily, and direct identification of the processes and effects with elements in typical passively modelocked dye lasers.

The model followed the pulse in one direction around the laser cavity and therefore did not take into account the colliding pulse mechanism. However previous theoretical work [2,3] indicated that effectively the colliding pulse mechanism is the same as increasing the s-parameter by a factor of 2-3. In the model the parameters were set to give good modelocking so that the omission of the CPM mechanism is not thought to be important. Furthermore the pulse evolutions being studied here are directly due to phase effects which do not require the CPM mechanism.

The relative strengths of the two phase modulation effects were inferred by observing that the experimental autocorrelation FWHM vs GVD curve (Figure 4.3.8) is only slightly steeper on the excess glass path side than on the negative dispersion side. Thus the two non-linear phase effects must be opposing each other and have similar magnitude, with the self-phase modulation slightly exceeding the absorber saturation modulation (ASM) in strength [4,5].

The choice of other parameters in the model was then made to give a good fit of the experimental data, however in order to reduce computational time the magnitude of the parameters in the model

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- 1 H. Avramopolous PhD Thesis, London University (1989)
 - 2 M.S. Stix, and E.P. Ippen, *IEEE J Quantum Electron.* QE-19, 520 (1983)
 - 3 D. Kuhlke, W. Rudolph, and B. Wilhelmi, *IEEE J Quantum Electron.* QE-19, 526 (1983)
 - 4 J.A. Valdmanis, R.L. Fork, J.P. Gordon, *Opt. Lett.* 10, 131 (1985)
 - 5 J.A. Valdmanis, R.L. Fork, *IEEE J. Quantum Electron.* QE-22, 112 (1986)

were scaled to increase the per round trip pulse modification and thus reduce the evolutionary period . A comparison between the parameters of the Rhodamine 110 laser and the simulation are given in table 4.4.2 below. The values for the experimental system are very approximate. For the real system the pulse energy (J_{∞}) was estimated from the 15mW average output power and a 10% linear loss. The small signal gain coefficients in the amplifier are given in the second row. The comparison of percentage change in pulse spectrum and temporal widths due to SPM and GVD per transit are calculated for a $10\mu\text{m}^2$ spot size area, $50\mu\text{m}$ jet thickness and 2mm of intracavity glass for a 100fs pulse. Thus we see there is a considerable difference between experiment and simulation, however the differences are reduced when cavity lifetimes are accounted for. This difference does, however, mean that quantitative comparisons between simulation and experiment were not possible.

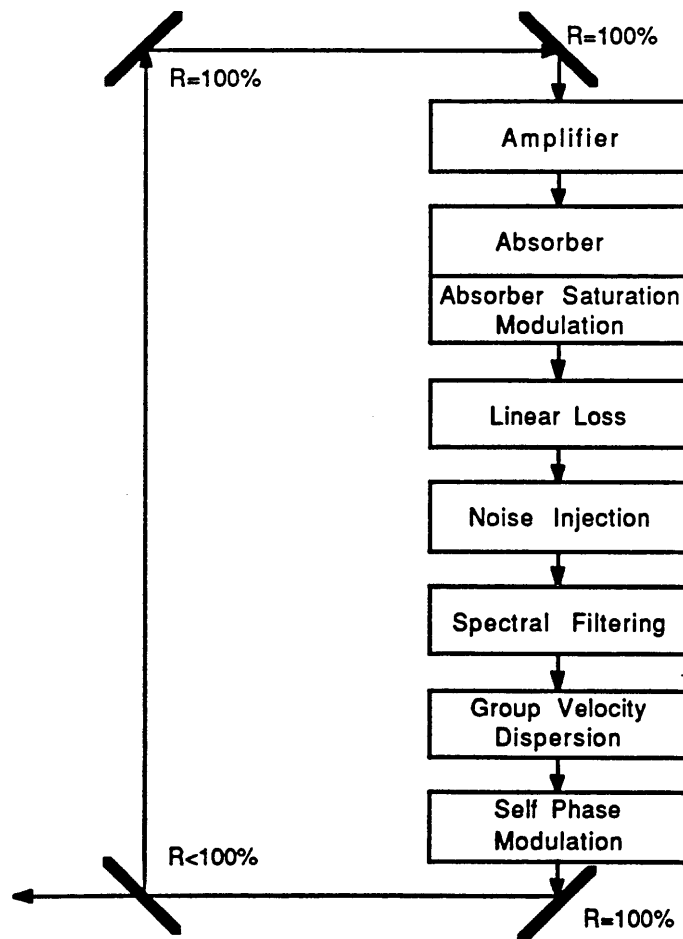


Fig. 4.4.1 A Schematic of the Numerical Model

	Rhodamine 110 Laser	Simulation
J_{∞} -value	~2.0	1.4
Small Signal Gain, A_s	~0.05-0.5	2
$\left[\frac{\Delta\omega^{\text{out}} - \Delta\omega^{\text{in}}}{\Delta\omega^{\text{in}}} \right]$ per transit	~0.2%	16%
$\left[\frac{\Delta T^{\text{out}} - \Delta T^{\text{in}}}{\Delta T^{\text{in}}} \right]$ per transit	~0.01%	0.7%
Cavity Lifetime in Transits	~10	2
$\left[\frac{\Delta\omega^{\text{out}} - \Delta\omega^{\text{in}}}{\Delta\omega^{\text{in}}} \right]$ per Cavity Lifetime	~14%	50%
$\left[\frac{\Delta T^{\text{out}} - \Delta T^{\text{in}}}{\Delta T^{\text{in}}} \right]$ per Cavity Lifetime	~1%	3%
Period of Evolutions in Transits	60-1000	30-120

Table 4.4.2 A comparison between parameters for the Rhodamine 110 Laser and the simulation.

4.5 The Numerical Results

Figure 4.5.1 below shows the variation of autocorrelation width with varying intracavity GVD obtained from the numerical model. These are average FWHM values in the cases where no steady state is reached and so are directly comparable with the measured time averaged autocorrelation traces obtained from the experimental system. The relative symmetry of the curve was obtained by the choice of the strengths of the phase modulation effects. Near the point of 0 GVD there is a step in the curve, and a strong modulation in the output. Between points labeled (b) to (d) there are satellites in the autocorrelation traces. This agrees well qualitatively with the experimental results in figure 4.3.8.

Figure 4.5.2 shows the series of average autocorrelation profiles corresponding to the labeled points in figure 4.5.1. Profiles (h) to (d) show satellite pulses that move towards the central peak as negative GVD is increased, and the step like feature is again due to the merging of the satellites with the main peak. Between traces (g) and (h) the movement of the satellite away from the peak

is very rapid. Strong modulation on the output was observed for profiles (d) to (f), and figure 4.5.3 shows how the frequency of output varies as the dispersion is changed. These results are very similar to the experimental results, although there is not an exact one to one correspondence.

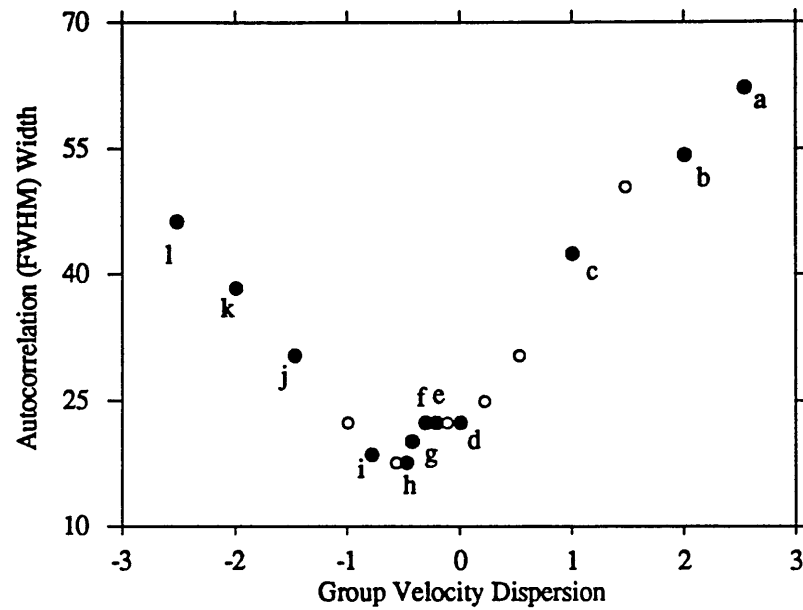


Fig. 4.5.1 Variation of the average FWHM of the autocorrelation function from the numerical model with intracavity GVD. The points labeled (a) to (l) correspond to the profiles in figure 4.5.2.

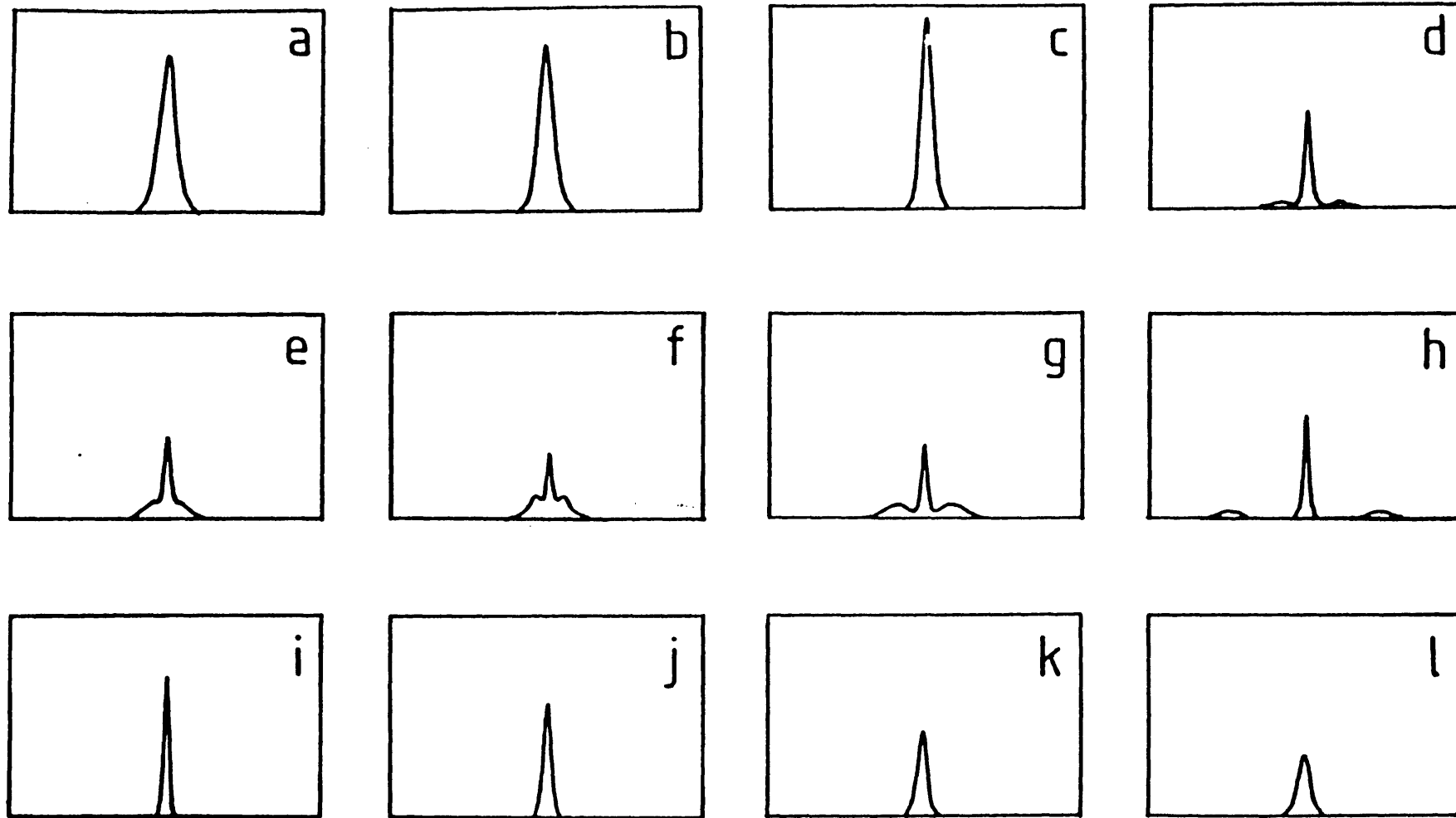


Fig. 4.5.2. Average autocorrelation profiles (a) to (l) obtained from the numerical model as GVD is varied from positive to negative values. (d) corresponds to 0 dispersion.

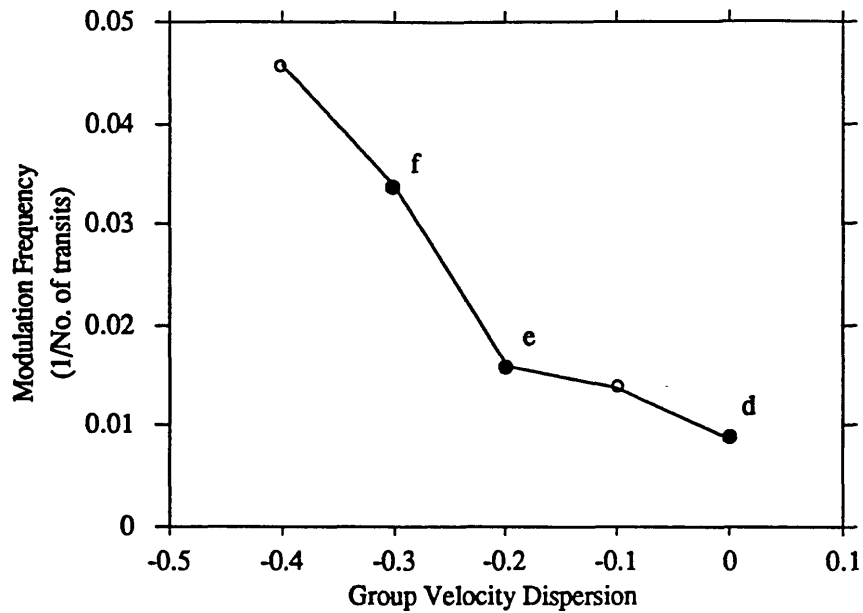


Fig. 4.5.3 Variation of modulation frequency versus intracavity GVD for the numerical model.

Figure 4.5.4 shows the evolution of the value of J_{∞} (the total pulse energy) of the pulse corresponding to the average autocorrelation profiles shown in figure 4.5.2. The strongest modulation occurs at the 0 GVD point - profile (d), while profiles (d) and (e) show the periodic and irregular modulation as shown in the experimental oscillographs in figure 4.3.10.

Figure 4.5.5 shows how the spectrum shifts with varying GVD. The centre of the half maximum is used to give a better indication of the asymmetries that occur. This agrees well with the experimental result - figure 4.3.15.

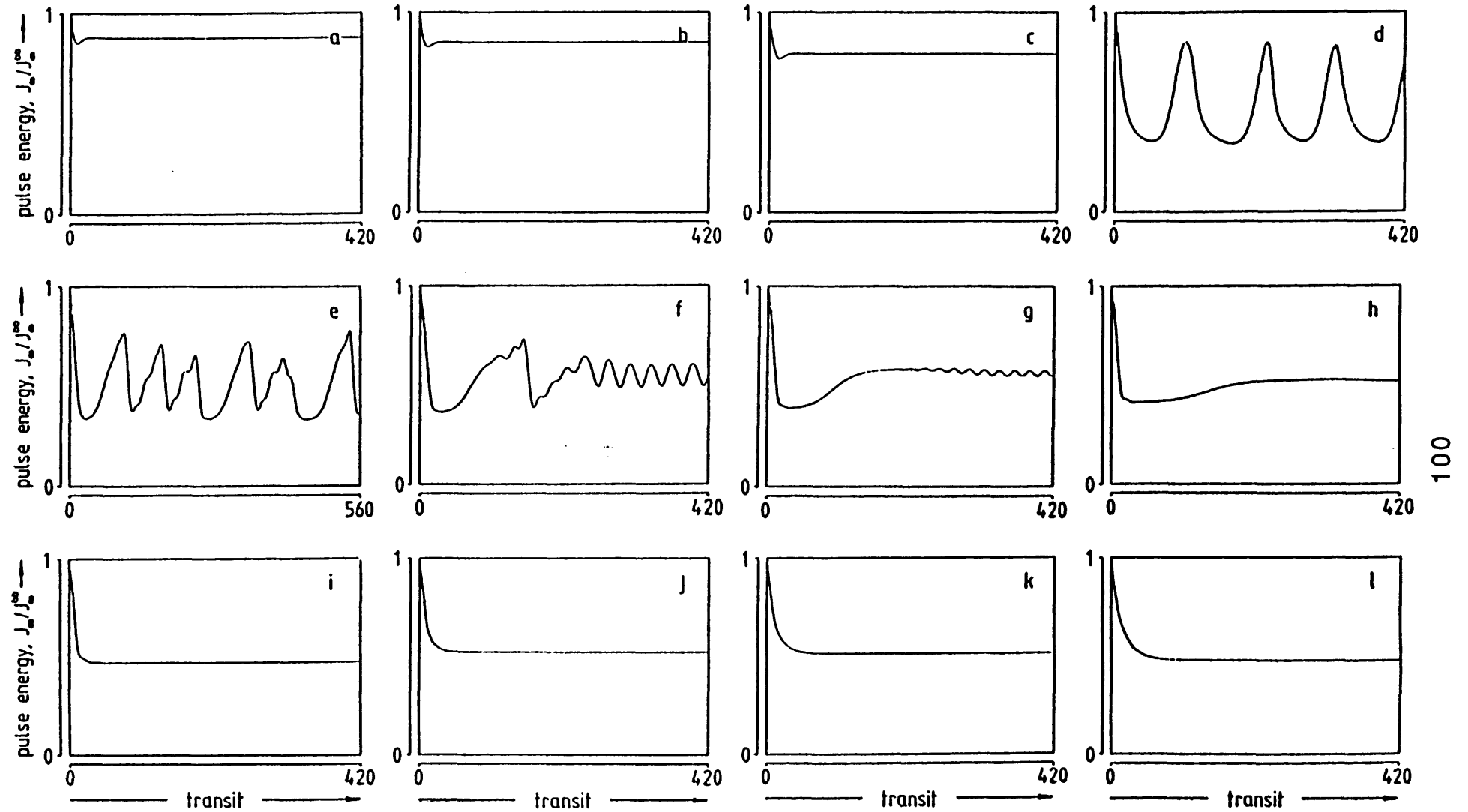


Fig. 4.5.4 Evolution of the pulse J_∞ -value as GVD is varied from positive to negative values. (d) corresponds to 0 dispersion.

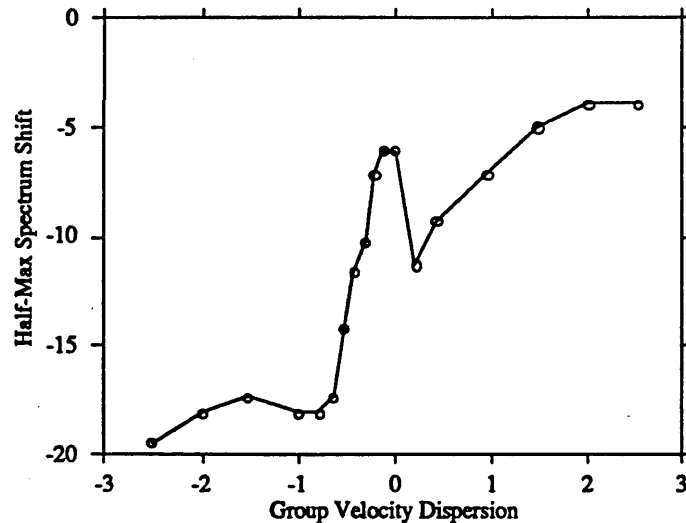


Fig. 4.5.5 Shift of the centre of the pulse spectrum as group velocity dispersion is varied.

Studies of the spectral and temporal evolutions obtained from the simulation corresponding to the autocorrelation traces labeled (a) to (i) reveal that for the 0 GVD case - (d) the laser output is at its most deeply modulated. The stability increases as the amount of negative GVD is increased. At point (f) the satellite is found to become permanent, and it changes the modulation characteristics of the output causing the kink in graph 4.5.3. With a further increase in negative GVD the satellite lags further behind and reduces in energy, with a large change occurring for a small change in dispersion between points (g) and (h). Similar behaviour has been reported by Wise et al. while introducing extracavity dispersion [1] into the output (which gave a triple peaked autocorrelation trace) from their laser, and such behaviour cannot be entirely explained by dispersion alone. At point (i) the satellite disappears leaving the single shortest pulse which has exponential wings a pulse width-bandwidth product of 0.32 which is close to that of a sech^2 profile, agreeing with experiment.

1 F.W. Wise, I.A. Walmsley, C.L. Tang, *Opt. Lett.* **13**, 129 (1988)

4.6 Interpretation

As we have already seen the experimental and theoretical results have a good qualitative agreement - quantitative agreement being ruled out by the need to exaggerate the model parameters, and by imprecise knowledge of the experimental system. Thus we believe that the model is a good description of the laser. Since the interpretation of all operations interacting at once is difficult, the cavity operations may be separated into operations which predominantly affect the amplitude and operations which predominantly affect the phase variation.

The operations which predominantly affect the amplitude are the saturable gain, the saturable absorption, linear loss, spectral filtering and noise injection. In the numerical model noise injection is important for the long term stability of the pulse rather than it's formation. The other four parameters can lead to stable single pulse formation, if the pulse compression (saturable gain, saturable absorption and linear loss) balances pulse broadening from the spectral broadening, and if pulse gain (amplification) is balanced by loss (absorption, linear and spectral loss). In this stable region there is absorption loss in the front of the pulse until the absorber saturates. Thus the leading edge is steepened, and there is gain at the peak of the pulse until the gain saturates giving a loss to the trailing edge of the pulse, and steepening there too. This process is balanced by the spectral filter which gives successively heavier losses to frequencies away from resonance. The energy balancing occurs, because if the pulse energy were to decrease from it's steady state value then there would be excess gain available on the trailing edge of the pulse on this transit. It is important to note that the trailing edge receives the extra energy rather than the leading edge since this explains why a satellite pulse is formed behind the main pulse rather than in front.

The processes which predominantly affect the phase variation of the pulse are phase modulation due to the time dependant absorber saturation (ASM), phase modulation due to the intensity dependant Kerr effect (SPM), and group velocity dispersion (GVD). The first two effects couple energy from the oscillating modes into new modes and thus extend the spectrum. For SPM the frequency

expansion is symmetrical in time (for a symmetrical pulse) but it is generally asymmetric for ASM since this doesn't occur until the absorber saturates. As the pulse energy decreases the ASM becomes more important in relation to SPM because the frequency change due to SPM is linearly related to energy while the ASM depends on the depth of saturation of the absorber which is almost complete and thus weakly dependant on energy. Figure 4.6.1 shows the effect on the carrier wave frequency from the two phase modulation effects for a more energetic (b) ($J_{\infty} = 1.4 J_{\infty}^{bb}$) and less energetic ($J_{\infty} = 0.4 J_{\infty}^{bb}$) (c) pulse of hyperbolic secant form (a). (J_{∞}^{bb} is the pulse energy that would occur from the 'bouncing ball' rate equation approximation model of New [1] for the small signal parameters used in this model. J_{∞}^{bb} was used as the seed pulse energy for this model).

GVD delays different frequencies by different time and always broadens unchirped pulses. Chirped pulses can be broadened or compressed depending on whether the GVD has the same sign as the chirp, or opposite.

1 G.H.C New, *IEEE J. Quantum Electron*, QE-10, 115 (1974)

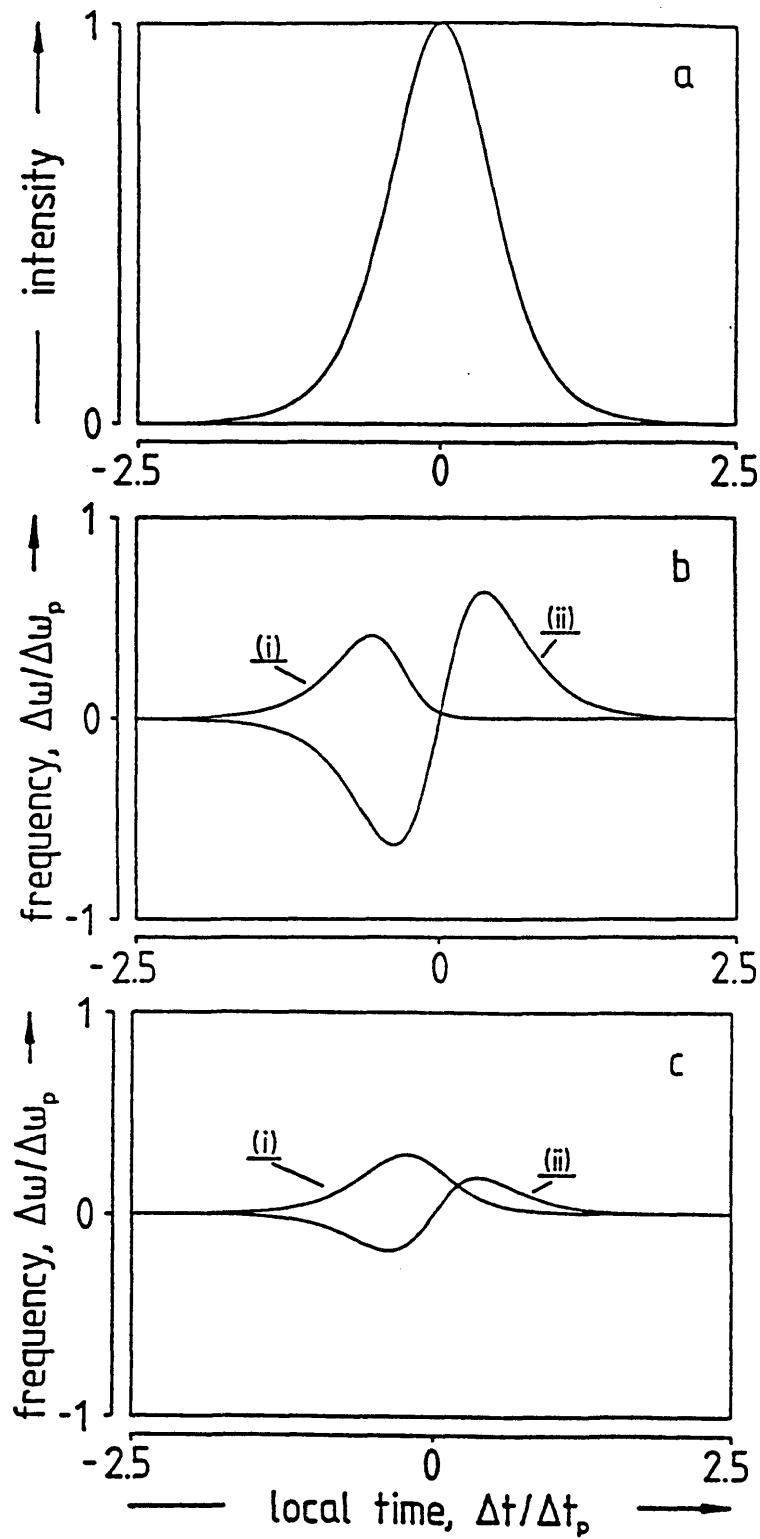


Fig. 4.6.1 Change of carrier wave frequency across a sech^2 profile (a) due to (i) absorber saturation modulation and (ii) optical Kerr effect, for pulse energies of (b) $J_\infty = 1.4 J_\infty^{\text{bb}}$ and (c) $J_\infty = 0.4 J_\infty^{\text{bb}}$. [1]

In the following discussion the description of the pulse formation in the model cavity which arises from detailed consideration of the numerical results will be given. The dispersion parameter range will be considered in three regions - (a) 0 GVD to just before the shortest pulse, (b) shortest pulse to excess negative GVD and (c) positive GVD.

In the region with 0 GVD (trace (d)) the new frequency modes coupled by phase modulation do not move within the pulse profile. Spectral expansion from both ASM and SPM generates both high and low frequencies, and this increases the loss in the filter, reducing the pulse energy and delaying the net gain function towards the high frequency components. Also the loss of pulse energy favours ASM to SPM which adds to the shift of the spectrum to high frequencies. The pulse energy is continually reduced by the spectral shift until it is insufficient to saturate the gain completely and thus there is gain left at the trailing edge of the main pulse which enables a second pulse to form. The first pulse continues to shift to high frequencies due to ASM and the second pulse receives increasing gain. As the energy of the second pulse increases it receives SPM but not ASM since the first pulse saturates the absorber. Eventually the first pulse disappears and the second pulse takes over to continue the cycle.

For the early parts of the above cycle SPM dominates, and the pulse is not bandwidth limited. The introduction of negative GVD in the cavity causes a phase change in the pulse profile which is opposite in sign to that induced by SPM. It can therefore partly compensate for it and cause pulse compression. The introduction of negative GVD can also reduce the amount of spectral expansion of the pulse by moving the newly generated frequencies away from the pulse edges, thus reducing the accumulative effect of the SPM. The reduction in spectral expansion reduces the losses of the pulse to the filter and thus has a stabilizing effect. The main pulse therefore increases in energy causing it to saturate the gain more deeply, therefore leaving less gain for the satellite pulse which reduces in energy and trails further behind (trace (e)). As negative GVD is further increased eventually the first pulse no longer disappears (trace (f)), and a periodic double pulsed evolution arises. The first pulse always contains most of the high frequencies of the spectrum because it saturates the absorber and

receives ASM. The second pulse has less phase modulation and primarily suffers SPM, so its spectrum lies near the centre of the filter. The evolution at this point works as follows. If the energy of the first pulse increases, its high frequency shift arising from ASM increases, leading to an increase in the high frequency part of the spectrum. This energy will then be shed to the filter reducing the pulse energy and leaving gain which enables the energy of the second pulse to increase. The second pulse will therefore then develop further SPM characteristics. This reveals itself in the power spectrum where a low frequency component occurs, and the peak at the centre of the filter increases. The second pulse will then lose its energy due to its spectral expansion and this will cause the leading edge gain coefficient in the next pass to increase and provide more energy for the first pulse again. Increasing negative GVD further increases compensation of SPM and reduces the losses to the first pulse from spectral expansion so that the gain for the second pulse decreases reducing its energy and causing it to lag further behind (It takes longer for the lower gain to establish the second pulse). Thus we see a small change in dispersion (traces (g) to (h)) causes a large time lag in the satellite as observed experimentally.

In the region from shortest pulse to excess negative GVD (traces (i) to (l)) the pulse is stable. For the shortest pulse (trace (i)) the GVD compensates for SPM and the effect of ASM causes the bias of the spectrum to high frequencies. The pulse profile is symmetric, and has the characteristics of a sech^2 intensity profile. More negative GVD (trace (j) to (l)) further delays the low frequency components broadening the temporal profile. However this is stable because the pulse energy is not significantly changed although the low frequency components of the spectrum do move further away from the gain peak causing them to be reduced in amplitude.

In the region of 0 to excessive positive GVD (traces (a) to (c)) the pulses are stable, long and asymmetric. The positive GVD enhances the positive chirp of the SPM and broadens the pulse. This reduces the spectral width with respect to the 0 GVD case (where there is a cumulative change of frequency) and decreases the losses suffered by the pulse at the filter. As the pulse energy is increased the net gain moves to the front of the pulse where the

frequencies are lower. The spectrum thus moves towards lower frequencies.

The strength of the spectral shifting and filtering process mentioned above would increase if the strength of the ASM were to increase. This was indeed found in the cavity where a mixture of DASBTI and HICI when used as a saturable absorber increased the strength of the effects over just having HICI as the saturable absorber. Over most of the wavelength range of operation of the laser DASBTI has about twice the absorbance of HICI and so will be more deeply saturated by the pulse.

The results obtained from the numerical model have shown that at 0 GVD there is a strong modulation in both the pulse profile and the pulse energy. This is contrary to soliton formation by the NLS equation which requires dispersion, and for which pulse energy is conserved. The instabilities have instead been explained as arising from phase modulation effects causing the spectral walk-off of the pulse inside the filter. The results shown below in table 4.6.2 illustrate how the ratio of pulse spectral width to filter bandwidth $\Delta\nu_p/\Delta\nu_f$ varies for different amounts of GVD.

Intracavity GVD	Corresponding Trace	$\Delta\nu_p/\Delta\nu_f$
2.0	(b)	0.21
1.0	(c)	0.32
0.5	(c)-(d)	0.41
0.0	(d)	0.18-0.45
-0.5	(h)	0.42
-1.0	(i)-(j)	0.30
-2.0	(k)	0.22
No phase effects		0.21

Table 4.6.2 Variation of the ratio of pulse spectral width to filter bandwidth ($\Delta\nu_p/\Delta\nu_f$) with GVD.

For the real Rhodamine 110 laser this can be estimated by considering the mirror reflectivities and absorption profiles which give a filter width of about 30nm. This is about 6x the bandwidth required for a 70fs sech² pulse. N=2 and N=3 solitons have a 4x to 8x expansion of their bandwidth during their evolution, and from the results presented in table 4.6.2 it is obvious that there is insufficient bandwidth for the spectral expansion of solitons of the required duration.

Although the shortest pulse both experimentally and theoretically has the characteristics of a fundamental soliton (i.e. it is symmetric, has exponential wings and has a time-bandwidth product within 5% of a sech^2) it cannot in general be described by the NLS since with stronger ASM in the cavity the spectrum would be shifted to higher frequencies and the system would be unstable where GVD compensates for SPM (i.e. where the shortest pulse would be expected from the NLS).

The experimental results of Salin et al. [1] do however indicate that higher order solitons (in particular $N=3$ solitons) can occur in passively modelocked dye lasers. Salins results suggested a fundamental soliton width of 470fs - they were unable to obtain the higher order solitons with their laser (Rhodamine 6G/DODCI) optimised to produce 70fs pulses. Thus the spectral width occupied by their solitons was a small part of the available bandwidth. This would occur if the modelocking mechanism were weak, as was indeed the case since the pump power was lower than optimum when the solitons were observed. Solitons have also been obtained by the numerical model set to give weak mode locking operations. However both experimentally and theoretically it has been found that the pulse energy decreases slightly where the evolving soliton was at it's point of maximum compression. This arises from loss to the spectral filter due to the large spectral expansion at this point. For a pure NLS evolution there would be no modulation in the pulse energy. Increasing the strength of the mode locking processes in the model caused the available relative bandwidth for pulse spectral expansion to decrease, increasing losses to the filter and the pulse energy modulation preventing the soliton evolution.

4.7 Conclusions

In this chapter the role of intracavity GVD in a Rhodamine 110 CPM laser passively modelocked with HICI and DASBTI has been investigated. An evolution of the autocorrelation profile with intracavity glass path has been described, and it contained regions where there were multi peaked autocorrelation traces indicating

1 F. Salin, P. Grangier, G. Roger, A. Brun, *Phys. Rev. Lett.* 56, 1132 (1986)

the presence of satellite pulses. For particular values of intracavity GVD periodic modulation was observed in the laser output.

The results from a numerical model which was constructed to incorporate the processes of SPM, GVD, saturable absorption and saturable gain have also been presented. This model was used to simulate the real laser, and good qualitative agreement was observed by fitting the strengths of the two phase modulation effects. The numerical results show that for zero and small negative values of GVD instabilities occur due to spectral shift and expansion arising from the phase modulation processes in the cavity which thus give rise to losses in the spectral filter and allow the formation of a satellite pulse. Large values of negative GVD, and positive GVD gives a stable result since the changes in the pulse spectrum are reduced. The numerical analysis suggests that the pulse evolution cannot be due to high order solitons since there is insufficient bandwidth to support such a pulse throughout its evolution. Only at the point where the shortest pulse is observed does it have the bandwidth limited sech^2 profile characteristic similar to a fundamental soliton. The model has however shown soliton evolutions in agreement with the experimental work of other workers if the modelocking mechanism is weak. In this case the pulses are long and require only a small amount of the total available bandwidth.

The observation from the analysis of the numerical results that the strongest modulation in the laser output occurs at the point of zero GVD should allow the determination of absolute intracavity GVD in real passively modelocked systems.

5. AN INVESTIGATION INTO FEMTOSECOND PULSE FORMATION IN A CONTINUOUSLY PUMPED PASSIVELY MODELOCKED CPM RING DYE LASER

5.1 Introduction

Most previous studies on c.w. passively modelocked dye lasers consider steady state or periodic pulse evolutions and relatively little attention has been paid to the early transient dynamics of the pulse evolution. The timescales associated with the pulse formation in c.w. passively modelocked dye lasers are important if one wishes to cavity dump such a laser at a high repetition rate [1] or to use a quasi-cw or pulsed pump source [2]. Experiments such as those in references [1,2] can only provide an estimate of the pulse formation time for the appropriate pumping levels.

Pulse formation has been observed directly in a flash lamp pumped passively modelocked dye laser using a single shot streak camera [3]. It was found that single pulses of a few picosecond duration evolved from initial ~ 100 ps noise bursts after ~ 45 cavity round trips. This observation was in good agreement with the theoretical model of New [2]. For typical c.w. lasers, however, the pump powers and therefore pulse energies are generally lower and so pulse formation takes rather longer. Using a powerful numerical model which did not require the rate equation approximation, Petrov et al. [4] calculated that the pulse evolution took several hundred to more than a thousand round trips to reach a steady state. The work presented in the chapter is to the best of my knowledge the first experimental investigation of the evolution to a steady state of femtosecond pulses in a passively modelocked dye laser.

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- 1 M.C. Downer, R.L. Fork, M. Islam, *Ultrafast Phenomena IV*, Springer-Verlag Series in Chemical Physics **38**, pp. 27-29 (1984)
 - 2 H. Ansari, A. Dienes, J.R. Whinnery, *Optics Letters* **10**, pp. 19-21 (1985)
 - 3 E.G. Arthurs, D.J. Bradley, A.G. Roddie, *Appl. Phys. Lett.* **23**, pp. 88-89 (1973)
 - 4 V. Petrov, W. Rudolph, B. Wilhelmi, *Optical and Quantum Electronics* **19**, pp. 377-384 (1987)

5.2 Experimental Results

Streak cameras do not have the necessary sensitivity or temporal resolution to observe the pulse formation in a c.w. passively modelocked dye laser. Achievement of both high temporal resolution and sensitivity in measuring the pulse evolution is, however, possible by periodically modulating the pump power and using a standard second harmonic generation autocorrelator in conjunction with a gating boxcar averager so that the temporal information at different delays can be obtained over many thousands of cycles.

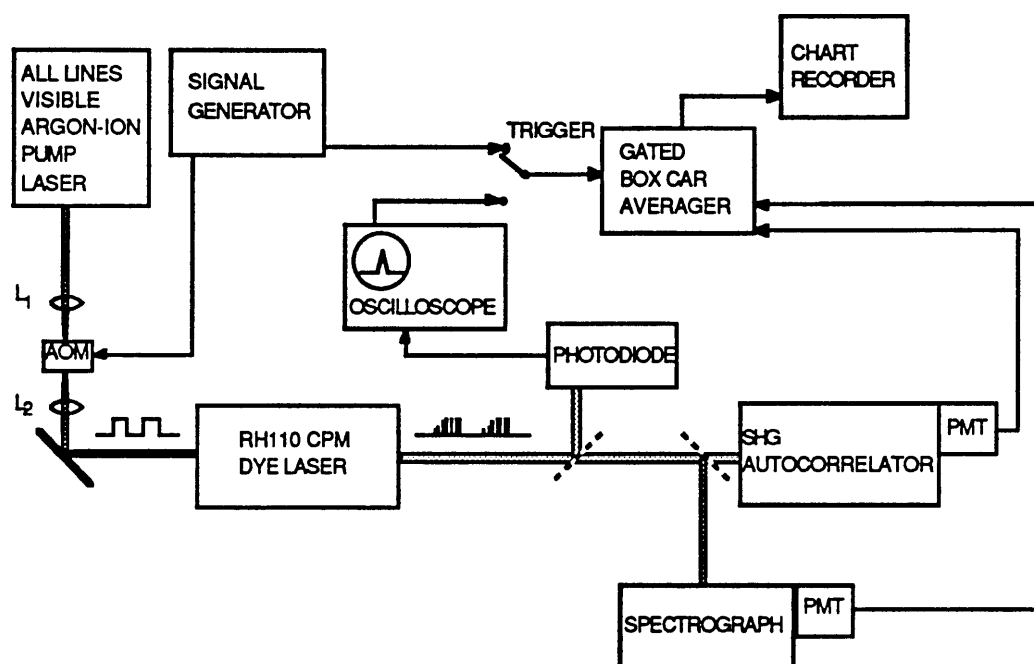


Fig. 5.2.1 Schematic of experimental set up used to study pulse formation in a c.w. passively modelocked dye laser.

The experimental configuration employed is shown in fig. 5.2.1 above. Lens L_1 and L_2 , both of 110mm focal length, focussed the pump beam to a spot size of $\sim 50\mu\text{m}$ diameter in the Tellurium dioxide acousto-optic modulator (AOM). The rise time of the induced modulation in the beam traveling through the modulator is limited by the transit time of the acoustic wave across the beam waist, and therefore tighter focusing which gives a smaller beam waist gives a faster rise time. However tight focusing also gives a more divergent beam which reduces the efficiency of the Bragg reflection (which is angle dependant) and thus reduces the

modulation depth. The afore mentioned focusing arrangement gives a reasonable compromise with a rise time of 20 ns (about two cavity round trips) and a modulation depth of about 60% which is easily sufficient to switch the CPM laser from good modelocked operation to well below lasing threshold.

The Rhodamine 110 CPM ring dye laser, passively modelocked with HICI as the saturable absorber, has been described in detail, and its performance as a function of intracavity GVD recorded and explained in the previous chapter. For the experiment described here the maximum available pump power for the CPM dye laser was limited to ~2.7W due to thermal lensing in the modulator which caused the pump beam to become decoupled from the CPM laser as the power was increased beyond this amount. The absorber concentration ($8 \times 10^{-4} \text{M}$) was therefore lower than optimum and consequently the minimum pulse widths achievable were ~120fs rather than <100fs as in the previous chapter. The cavity round trip time here was 9ns, and the interprism separation was 41cm. The behaviour of the laser as a function of intracavity GVD etc. was found to be qualitatively the same as in the previous chapter.

The output from the CPM laser was analysed temporally using the standard collinear second harmonic generation technique with a $1\mu\text{m}$ thick BBO crystal, and spectrally with a 1m grating spectrometer. The optical signal from both devices was measured using photomultipliers with <30ns response time, and the resulting electrical signals were gated in a boxcar averager. An additional output from the laser enabled monitoring of the pulse energy using a BPW28 photodiode and an oscilloscope.

Initially the pump radiation was modulated with a square waveform at a frequency of 700Hz, and the modulating signal was used to trigger both the oscilloscope and the boxcar averager. Figure 5.2.2 below shows an oscillograph of the evolution of the pulse energy from the CPM laser for one cycle. This is taken for the optimum value of intracavity glass path (i.e. that value which produced the shortest pulses). From this figure we see that the pulses start to build up about 100 μs after the laser is switched on - this corresponds to 11000 rounds trips in the cavity. There was ~20 μs jitter associated with this rise time which was probably due to small changes in the intracavity loss arising from fluctuations of the dye jets and the pump power. It was found that

increasing the pump power reduced both this initial start up time period, and the amount of jitter. Quantitative measurements of this variation, however, were impractical due to the inability to increase the pump power without thermal lensing in the acousto-optic modulator.



Fig. 5.2.2 Oscillograph of one cycle of the modulated pulse train from the laser. (DETECTOR WAS A.C. COUPLED)

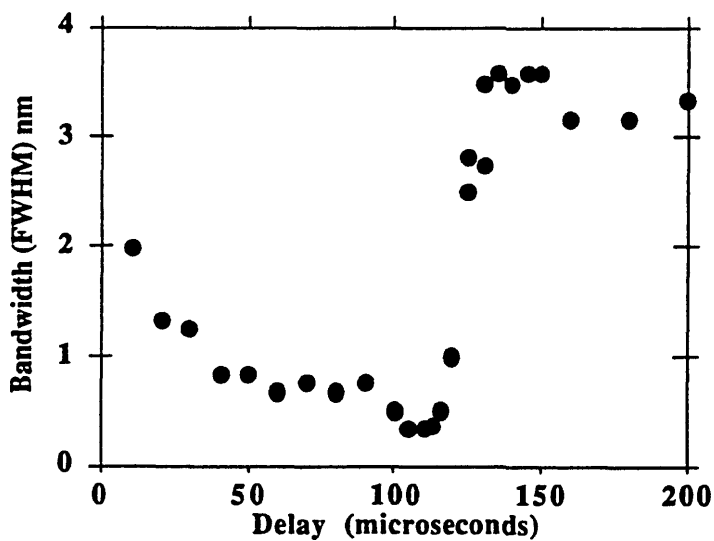


Fig. 5.2.3 A plot of the bandwidth FWHM versus delay from switch on, for optimum intracavity glass path.

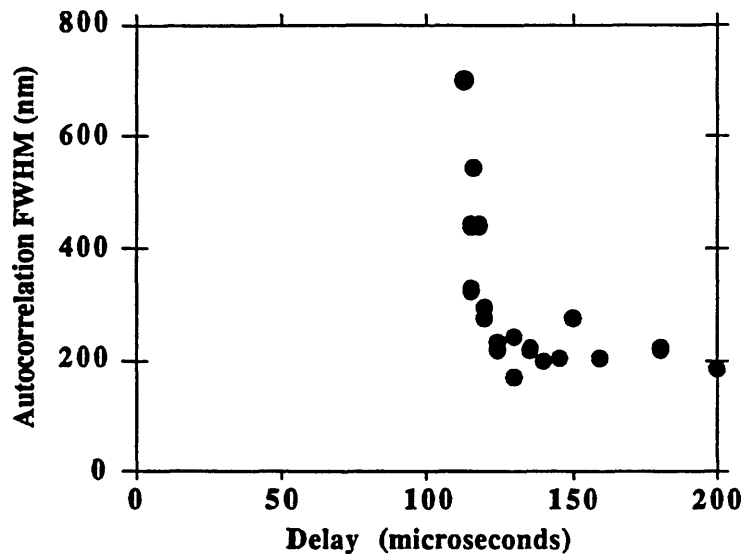


Fig. 5.2.4 A plot of the Autocorrelation FWHM versus delay from switch on, for optimum intracavity glass path.

Employing the gating boxcar averager with a gate width of 30ns, and using a 30 sample average an examination was carried out of how the autocorrelation and spectral profile of the laser output varied with delay from the initial switch on. Due to the temporal jitter in the point where pulse formation starts relative to the switch on time of the laser it was found necessary, for measurements on the pulse formation process, to trigger the boxcar averager from the gate output of the oscilloscope which was in turn triggered from the output of the photodiode. This trigger method was used for the measurements given after 100 μ s in figures 5.2.3 & 5.2.4, and measurements of delay in all subsequent figures are quoted relative to this point. Figures 5.2.3 & 5.2.4 show how the FWHM bandwidth and autocorrelation width of the laser output varied with delay from switch on for the optimum value of intracavity glass path. Initially we see the laser starting with a broad weak noise spectrum which in time grows in energy and coherence. The energy in the laser cavity continues building up until modelocking begins after about 100 μ S. At this point figures 5.2.3 & 5.2.4 show the bandwidth expanding and the autocorrelation FWHM narrowing rapidly as both absorber and gain saturation take their effect. The autocorrelation FWHM contracts from 700fs to 200 fs almost linearly in 12.5 μ s gives a pulse shortening velocity

in this cavity of $40\text{fs}/\mu\text{s}$. Figure 5.2.5 shows a set of the spectral and autocorrelation profiles taken for various delays corresponding to some of the points on these graphs. Eventually in figures 5.2.3-5.2.5 we see that a steady state is reached in the autocorrelation and spectral profiles.

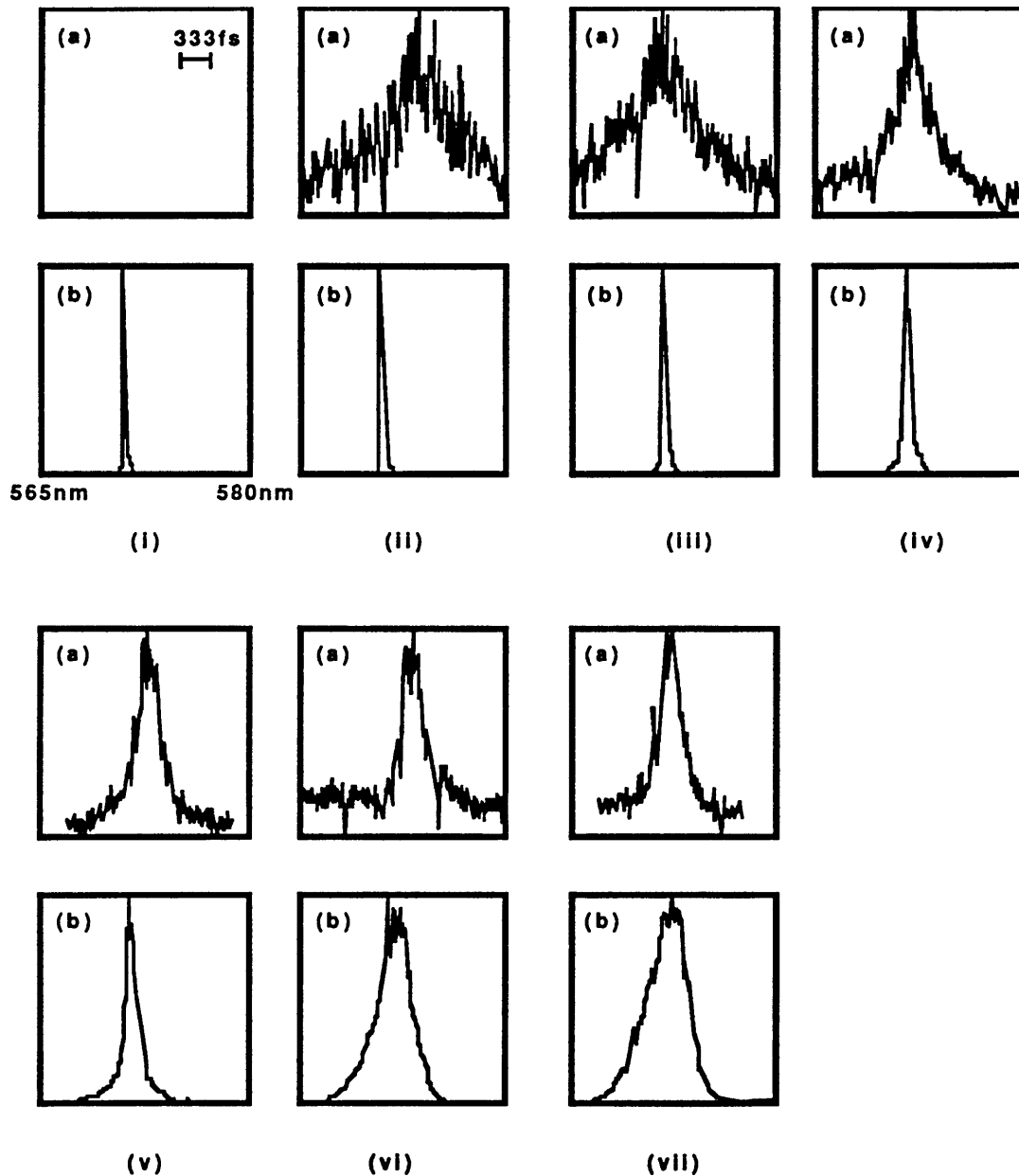


Fig. 5.2.5 The sequence of (a) autocorrelation and (b) spectral traces taken for the optimum intracavity glass path at delays of (i) $0\mu\text{S}$, (ii) $12.5\mu\text{S}$, (iii) $15\mu\text{S}$, (iv) $17.5\mu\text{S}$, (v) $20\mu\text{S}$, (vi) $25\mu\text{S}$ and (vii) $50\mu\text{S}$ from the oscilloscope trigger point, all with the same horizontal scales.

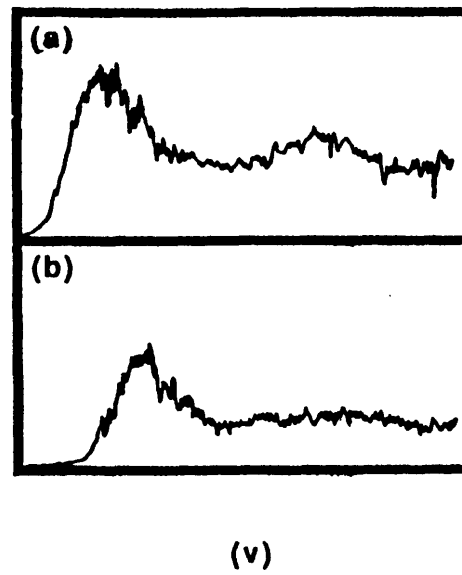
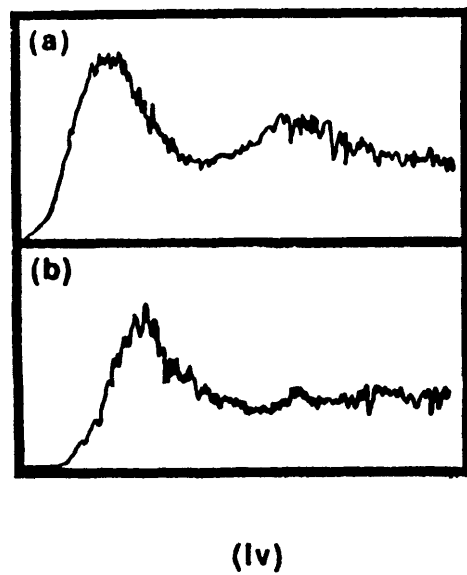
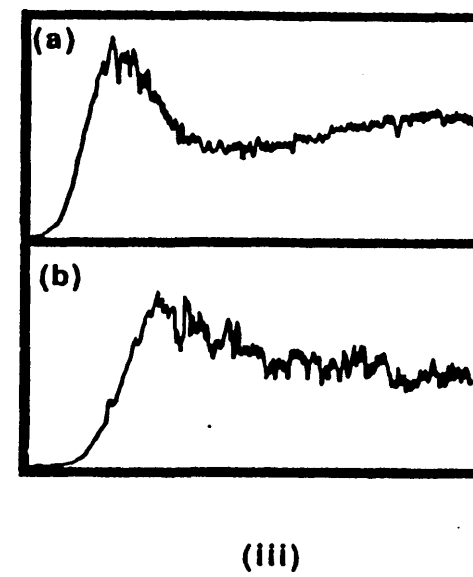
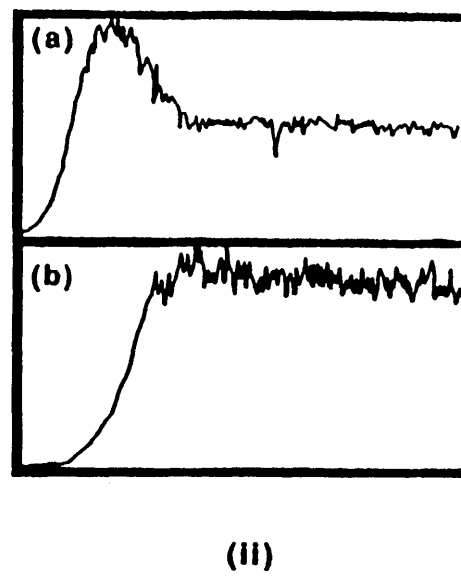
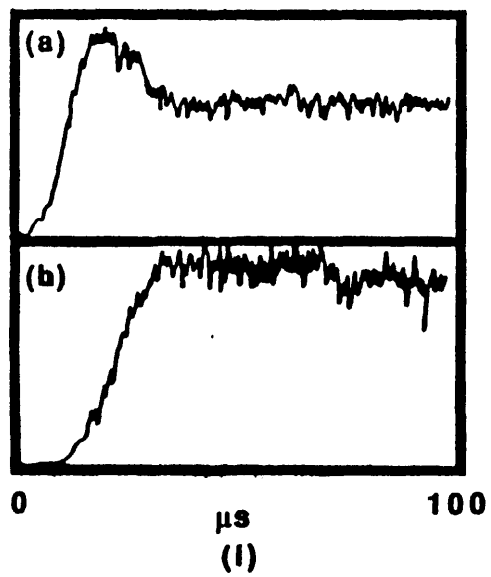
A study was made of how the power of the laser output (monitored using a PMT) and the second harmonic of the laser

output, varied with delay from the start of pulse build up for various intracavity glass paths. The glass paths are quoted relative to the point of 0 GVD, as ascertained in the previous chapter. These experimental traces are presented in figure 5.2.6, and data taken from these traces was used to construct figures 5.2.7 and 5.2.8 which have respectively the power traces and second harmonic traces superimposed thus showing more clearly the effect of varying glass path. From these results we observe that the slopes of the rising edges of the power and second harmonic signal curves are the same for the differing glass paths. The earlier cw lasing part of the evolution not shown in figures 5.2.6-5.2.8 was also invariant with change in intracavity glass path. It should be noticed that the power of the laser output (fig. 5.2.6(a)) overshoots its steady state value for all intracavity glass paths whereas the second harmonic only overshoots for glass paths that are larger than optimum. Also the peak in the second harmonic signal always occurs after the peak in the power output. For equal or less intracavity glass path than optimum (fig. 5.2.6 (ii) & (i)), both the second harmonic and power signal reach a steady state whereas for a greater amount of glass (figs. 5.2.6 (iii)-(v)) the power is periodically modulated. The second harmonic signal also shows some modulation for these values of intracavity glass path, but the modulation depth is reduced. The magnitude of the second harmonic signal decreases for increasing amounts of intracavity glass.

Sequences of autocorrelation and spectral traces with delay were taken for various values of intracavity glass path other than the optimum. It was observed that for values of glass path that were less than optimum, the features of the pulse evolution were similar to that of optimum but the asymptotic bandwidth and autocorrelation FWHM reached were broader. For values of glass path beyond optimum increasingly complicated behaviour was observed in the structure of the spectra. It was not possible to observe the corresponding temporal behaviour in the autocorrelation traces due to excessive noise (although use of a more sensitive but slower PMT showed that weak satellites were present on the autocorrelation profile). Figure 5.2.9 shows a sequence of spectra taken for different delays for an intracavity glass path corresponding to approximately zero GVD. This shows that an initially broad modelocked bandwidth (iii) is seen to evolve

out of the narrow cw spike (i). Subsequently however, the evolution continued with reemergence of the spike ((iv)-(vi)), and a periodic evolution ensued, with alternating double peaked and single peaked spectral profiles.

Fig. 5.2.6 The evolution of power (a) and second harmonic signal (b) from the laser output for intracavity GVD corresponding to (i) - 5.8mm, (ii) -3.5mm (optimum), (iii) -2.2mm, (iv) 0mm and (v) 3.4mm of glass. They start at the oscilloscope trigger point and end 100 μ s later.



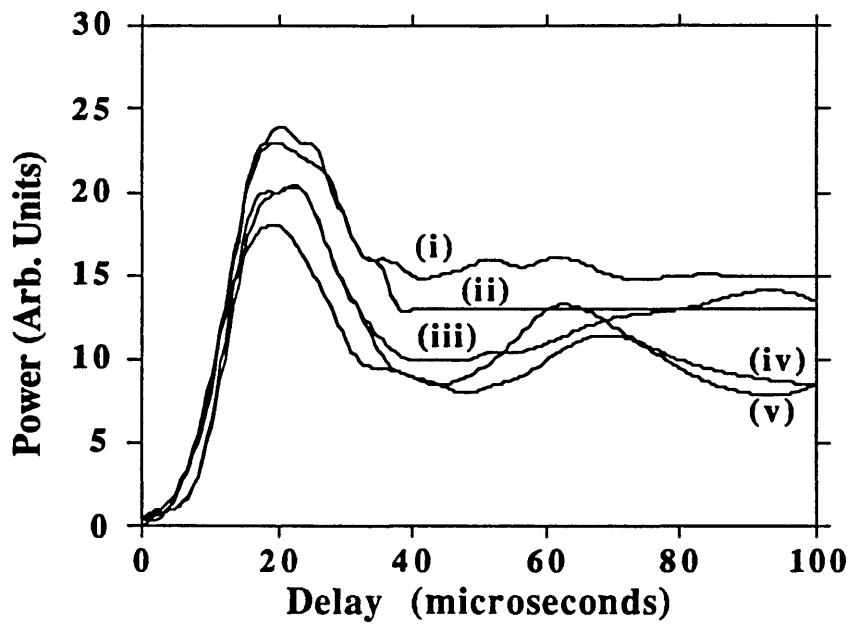


Fig. 5.2.7 Superimposed traces showing the evolution of average fundamental power with delay for varying intracavity GVD values, labeling as in figure 5.2.6.

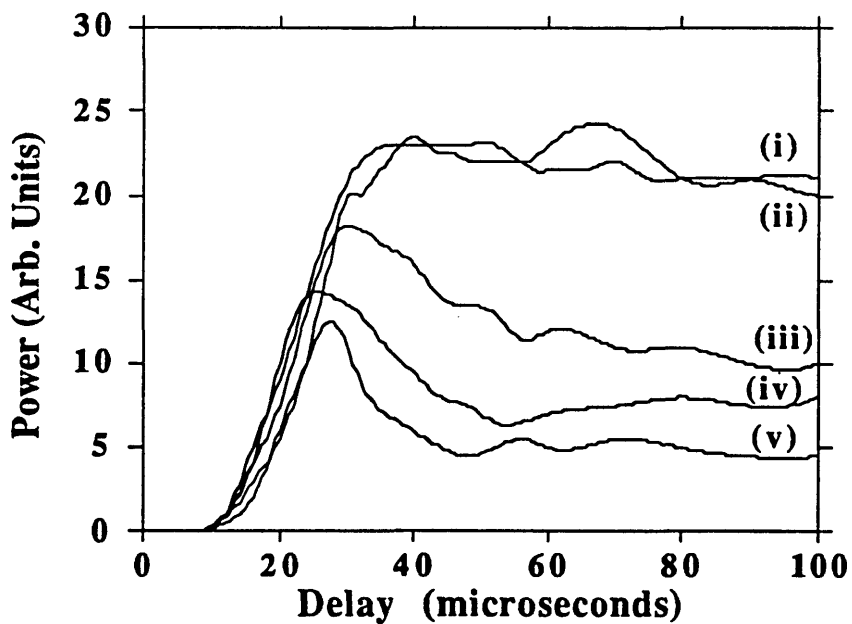


Fig. 5.2.8 Superimposed traces showing the evolution of average second harmonic power with delay for varying intracavity GVD values, labeling as in figure 5.2.6.

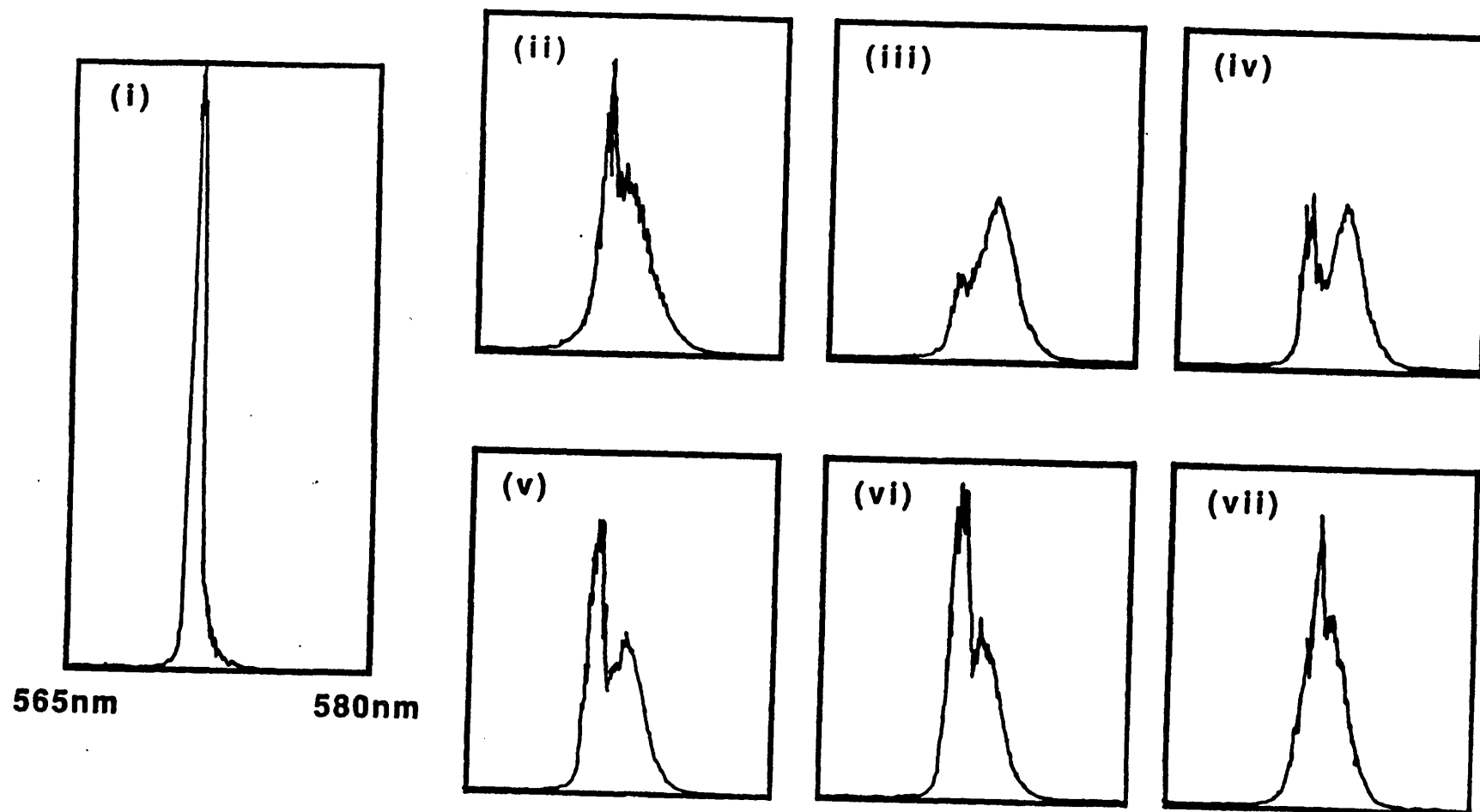


Fig. 5.2.9 Sequence of spectral traces taken for intracavity GVD corresponding to 0mm of glass at delays of (i) 25 μ S, (ii) 35 μ S, (iii) 45 μ S, (iv) 55 μ S, (v) 65 μ S, (vi) 75 μ S and (vii) 95 μ S from the oscilloscope trigger point, all with the same horizontal scale.

5.3 Discussion

The basic picture of the passively mode locked c.w. dye laser (e.g. [1]) demands a close balance between small signal gain and loss in the cavity such that CW lasing operation is very close to lasing threshold. This is illustrated by the 10^4 or so round trips which we observe for the radiation in the cavity to build up to the point where modelocking can take place. For the same reason the initial build up of radiation in the cavity is very sensitive to small changes in loss, and this results in the jitter observed in the time taken to begin modelocked operation. The growth and narrowing of the spectrum in time is due to preferential amplification by the gain at the wavelength of least loss and is as expected for a homogeneous gain medium [2]. It is only when the circulating energy in the cavity is sufficient to saturate both the absorber and the gain that mode locked operation can begin, and once this occurs, the overall gain in the temporal region of the pulse increases due to absorber saturation [1] and so the circulating energy can increase rapidly as shown in figures 5.2.2 & 5.2.6. Our measurement of about 1300 round trips for the pulse to shorten to the steady state value (see figures 5.2.2 and 5.2.6) is in reasonable agreement with the results from the numerical model of Petrov et al. [3] when phase modulation effects were included. This observation may appear to conflict with conclusions drawn from [4] where a passively modelocked dye laser was cavity dumped at 3MHz. In the latter case, however, the pulse energy was modulated rather than the pump power, and was thus maintained at a level where it could saturate the absorber.

Since the amount of dispersion in the cavity will only have an effect on a short pulse with a significant bandwidth, it is expected that the initial rate of pulse shortening is independent of the amount of intracavity glass, and this is observed (fig. 5.2.6-5.2.8). The overshoot in pulse energy which occurs, and which has also

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- 1 G.H.C. New, *IEEE Journal of Quantum Electronics* QE-10, pp 115-124 (1974)
 - 2 A.E. Siegman, *An Introduction to Lasers and Masers*, Pub. McGraw-Hill 1971
 - 3 V. Petrov, W. Rudolph, B. Wilhelmi, *Optical and Quantum Electronics* 19, pp. 377-384 (1987)
 - 4 M.C. Downer, R.L. Fork, M. Islam, *Ultrafast Phenomena IV*, Springer-Verlag Series in Chemical Physics 38, pp. 27-29 (1984)

been shown in the numerical work of Petrov *et al.* [see reference 1 figure 2] may be explained as being due to the effect of the limited spectral support in the cavity. Initially a broad pulse with a narrow spectrum suffers little loss in the spectral filter, and so its energy can build up fast. As the pulse narrows temporally it broadens spectrally increasing the loss arising from spectral filter. This causes a reduction in the the pulse energy as the pulse continues to shorten. In figure 5.3.1 the traces of figure 5.2.7&5.2.8 (ii) for the evolutions of average laser output power (a) and the average second harmonic signal (b) have been taken, and from these there has been derived (using the relationship that the second harmonic signal will be proportional to the pulse energy squared over the pulse duration for a fixed pulse profile) a trace of the pulse duration evolution (c). In this figure we see clearly the pulse duration reaching its steady state value at the point in time where the pulse energy has decreased back to its steady state value. The pulse duration scale on this graph was set to correspond to the measured autocorrelation widths obtained once steady state had been reached.

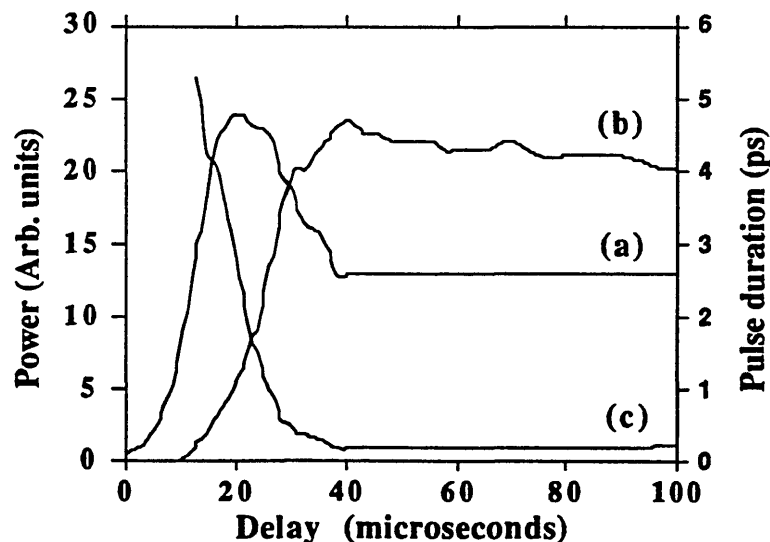


Fig. 5.3.1 The evolution of power (a) and second harmonic signal (b) for the case of optimum intracavity GVD (trace (ii) in figure 6) plotted with the pulse duration trend calculated from these traces.

1 V. Petrov, W. Rudolph, B. Wilhelmi, *Optical and Quantum Electronics* 19, pp. 377-384 (1987)

Once a short pulse has been formed, the effects of intracavity dispersion and phase modulation arising from time dependent absorber saturation and the optical Kerr effect are expected to become important in the pulse evolution. These can give rise to complicated behaviour in the laser output which has been described in the previous chapter where it was shown that, for this particular laser system, the total amount of group velocity dispersion is negative at the point where the shortest pulse is obtained. As in the previous chapter, it is convenient therefore, in considering the results, to split the dispersion parameter range into three regions: (a) shortest pulse to excess negative GVD, (b) 0 GVD to just before the shortest pulse, and (c) positive GVD.

For glass paths less than optimum (Region (a) - corresponds to figure 5.2.6(i)) the features of the modelocking process observed are the same as those at optimum (figure 5.2.6(ii)), however the steady state pulse duration reached is broader, and the pulse energy is larger. This is exactly as expected from the previous chapter.

In figure 5.2.6(iii) (region (b)) a modulation is observed in the laser output. The temporal development of the spectrum for this dispersion range which is shown in figure 5.2.9 shows the same initial spectral broadening and temporal narrowing as before (figure 5.2.5). Instead of settling down to a steady broad spectrum, however, a narrow spike grows back out of this spectrum (fig. 5.2.9 (iv) to (vi)), and a periodic evolution ensues. It is thought that this spectral spike corresponds to the satellite pulse which appears in the autocorrelation trace of the laser output, while the broad spectral component corresponds to the main pulse. The absorber saturation modulation in this laser causes an overall shift of the main pulse to higher frequencies. For values of intracavity group velocity dispersion close to zero, this frequency shift will accumulate on each round trip until the pulse loses energy to the filter. It is this spectral walk-off which is the cause of the observed periodic pulse evolutions arising in CPM dye lasers in the strongly mode locked regime as described in the previous chapter. Since the absorber is deeply saturated immediately after the main pulse the satellite pulse does not experience any absorber saturation modulation. It does, however, see a different spectral

filter to the main pulse. In our laser the filter is formed by the dielectric mirror cut off on the long wavelength side of operation, and by the absorption profile of the saturable absorber on the short wavelength side of laser operation. The filter thus changes as the absorber saturates, and the second pulse sees reduced loss at shorter wavelengths than the main pulse. Thus the spectral peak corresponding to the satellite occurs at a shorter wavelength than the main pulse. This is illustrated in figure 5.2.9.

5.4 Conclusions

An investigation of the formation of femtosecond pulses in a c.w. passively modelocked laser has been undertaken for the first time. The timescale of this evolution has been measured, using an electronic gating technique. It is found to be very much slower than for pulsed dye lasers [1] due to the much lower gain available in c.w. systems. Initially, we measure a delay of $\sim 100\mu\text{s}$ for the circulating energy to build up to the level at which the gain and absorption can begin to saturate. From this point the pulse evolution reaches a steady state - with or without a periodic evolution - after ~ 1300 cavity round trips. This is in reasonable agreement with the estimates of Petrov et al. [2]. These observations should be important when considering the stability of passively modelocked c.w. dye lasers in the presence of 'dropouts' and when cavity-dumping or pumping with quasi-c.w. sources.

Observations of overshoot in the pulse energy during pulse formation are evidence that the intracavity spectral filter is important in determining pulse formation in that it causes shorter pulses with broader bandwidths to lose energy. This may therefore be the limiting factor in ultrashort pulse formation in passively modelocked dye lasers.

Measurements into the effect of intracavity GVD on pulse formation have been carried out, and these show that this only becomes important when the pulse duration becomes quite short.

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- 1 E.G. Arthurs, D.J. Bradley, A.G. Roddie, *Appl. Phys. Lett.* **23**, pp. 88-89 (7/1973)
 - 2 V. Petrov, W. Rudolph, B. Wilhelmi, *Optical and Quantum Electronics* **19**, pp. 377-384 (1987)

For values of GVD for which satellite pulses were reported in the previous chapter, spectral measurements show that the satellite pulse lies at shorter wavelengths than the main pulse, and this is evidence for the transient behaviour of the intracavity spectral filter arising from absorber saturation. These measurements have also shown the relative insensitivity of autocorrelation techniques to changes in the spectral content of the pulses.

Similar gating experiments may be used to study pulse formation dynamics in other laser systems - of particular interest would be a comparative study on synchronously and hybridly modelocked dye lasers.

6. FEMTOSECOND PULSE GENERATION FROM A TITANIUM-DOPED SAPPHIRE LASER

6.1 Introduction

Titanium^{that is} doped in Al_2O_3 replaces an Al^{3+} ion, and may occur as either trivalent Ti^{3+} or quadrivalent Ti^{4+} . The Ti^{4+} ion has the closed shell configuration of Argon while the Ti^{3+} ion has a single outer d-electron whose energy levels are degenerate in the isolated ion, but which are split (to first order) in the presence of a crystal field into a ${}^2T_2$ triply degenerate lower level and a doubly degenerate ${}^2E_g$ upper level. This enables 4-level laser operation, with no upper state absorption to higher d electron levels, and with a fast phonon assisted relaxation from the metastable excited state [1]. The relevant configuration diagram is shown below.

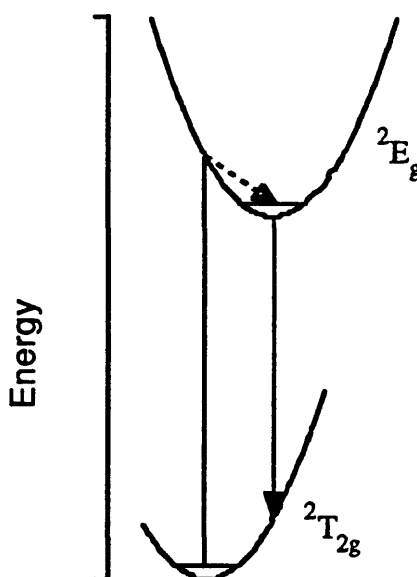


Fig. 6.1.1 Configuration diagram for Ti^{3+} in sapphire. The dashed line indicates a phonon assisted relaxation.

The fluorescence spectra of $\text{Ti}:\text{Al}_2\text{O}_3$ extends from about 600nm to $>1.1\mu\text{m}$, while the absorbance extends from 400nm to about 600nm [2] making it suitable for argon-ion pumping.

Until relatively recently poor crystal quality has prevented development of $\text{Ti}:\text{Al}_2\text{O}_3$ as a laser source. With the use of growth

-
- 1 L.G. DeShazer, J.M. Eggleston, K.W. Kangas, *Opt. Lett.* **13**, 363 (1988)
 - 2 P.F. Moulton, *JOSA* **B3**, 125 (1986)

methods employing heat exchange techniques, and adjustment of crystal growth parameters to minimize the precipitation of Ti^{2+} ions as scattering centres [1] good optical quality has been achieved. The residual absorption in the infrared in this material which is thought mainly to arise from the formation of Ti^{3+} - Ti^{4+} pairs has been minimized by annealing in a reducing atmosphere [2]. $Ti:Al_2O_3$ is now a good solid state alternative to argon ion pumped dyes in the near infrared, and therefore potentially a useful tool for spectroscopy of semiconductor materials.

Once continuous laser operation using this material at room temperature was demonstrated [3] the way to c.w. mode locked operation was opened. If the entire gain bandwidth could be exploited then in principle pulses of 4fs duration could be amplified in $Ti:Al_2O_3$. Amplification, in a $Ti:Al_2O_3$ regenerative amplifier, of 300fs pulses from a femtosecond dye laser has been realized by Schulz et al. [4]. The shortest pulses directly generated previous to this work (6ps- [5]) used active loss modulation employing a high Q, high modulation depth acousto-optic mode locker. Synchronous pumping of $Ti:Al_2O_3$ [6] has proven less effective due to its low gain cross section ($\sim 3 \times 10^{-19} \text{ cm}^2$) and its long upper state relaxation time (3.2 μ s) which implies the need for a large flux for saturation, and for the same reasons passive modelocking using a slow saturable absorber is unlikely to be very successful [7].

Most of the early experiments referenced above employed research grade pieces of the material which are selected to have a high figure of merit (defined as the ratio of pump absorption at 500nm to residual absorption at 800nm). In this chapter I shall describe the CW and CW mode locked operation of a commercial

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- 1 F. Schmid, and C.P. Khattak, *Tunable Solid State Lasers* (Springer Optical Sciences series vol. 47), 122
 - 2 R.L. Aggarwal, A. Sanchez, M.M. Stuppi, R.E. Fahey, A.J. Strauss, W.R. Rapoport, and C.P. Khattak, *IEEE Journal of Quantum Electron.* **24**, 1003 (1988)
 - 3 P. Albers, E. Stark, and G. Huber, *JOSA* **B3**, 134 (1986)
 - 4 P.A. Schulz, M.J. LaGosse, R.W. Schoentein, and J.G. Fujimoto, OSA Annual Meeting, 1988 Technical Digest Series **11**, Paper MEE2.
 - 5 J.D. Kafka, A.J. Alfrey, and T. Baer, *Proc. Ultrafast Phenomena VI, Springer series in Chem. Phys.* **48**, 64 (1988)
 - 6 G.B. Al'tshuler, V.B. Karasev, N.V. Kondratyuk, G.S. Kruglik, A.V. Okishev, G.A. Skripko, V.S. Urbanovich, and A.P. Shkadarevich, *Sov. Tech. Phys. Lett.* **13**, 324 (1987)
 - 7 G.H.C. New, *EEE J. Quantum Electron.* **QE-10**, 115 (1974)

grade crystal (from Union Carbide) with a figure of merit guaranteed only to be greater than 100. This will be followed by a description of a non-linear resonant external cavity, based on the theoretical and experimental work of Blow et al. on colour-centre lasers [1,2], which has enabled the generation of subpicosecond pulses from this material.

6.2 CW operation

Initially a simple three mirror astigmatically compensated cavity as shown below was constructed.

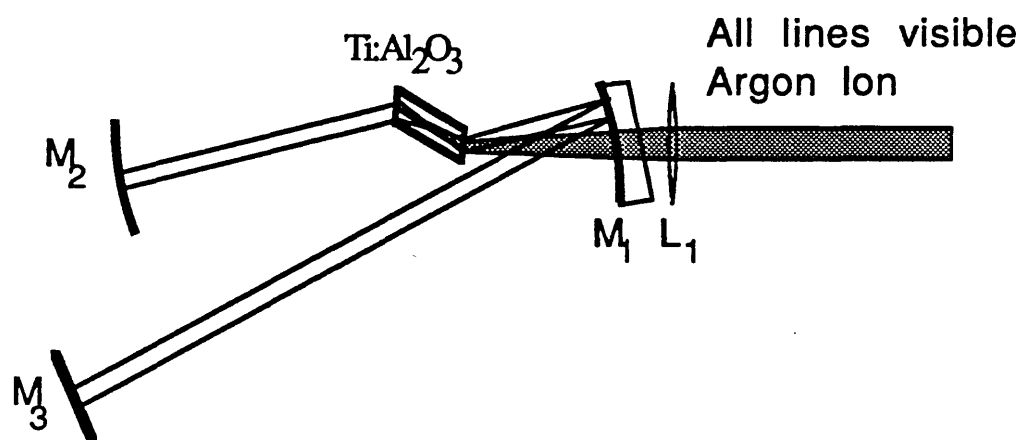


Fig. 6.2.1 Cavity configuration for c.w. Ti:Al₂O₃ laser

Mirrors M₁ and M₂ were of 15cm radius of curvature and mirror M₃ was plane. All the laser mirrors supported broadband dielectric coatings with a high reflectivity zone from 650nm to 950nm. The Brewster-angled rod of Ti:Al₂O₃ was cut perpendicular to the c axis, had about 0.1% doping of Ti ions and was 17mm in length. It was mounted on a water cooled copper block which was kept at 17°C. Lens L₁ (of 10cm focal length) focussed the pump radiation (all lines visible from Spectra-Physics model 2020 or 2035 Argon ion lasers) onto the laser rod which absorbed about 70% of the pump power. With 100% reflectivity laser mirrors all round the lasing threshold was 0.9W of absorbed pump power. With a 4% output coupler for mirror M₃ slope efficiencies of 16% were achieved as shown in figure 6.2.2 below.

1 K.J. Blow and D. Wood, *JOSA B*5, 629 (1988)

2 K.J. Blow and B.P. Nelson, *Opt. Lett.* 13, 1026 (1988)

Introduction of a Spectra-Physics tuning wedge into this cavity enabled continuous tuning from 724nm to 800nm with 8W total pump power (Fig. 6.2.3a). With 15W total pump power the tuning range extended from 720nm to 845nm with a large gap in the tuning range around 815nm (Fig. 6.2.3b) which may be due to some residual absorption in the Ti:Al₂O₃ crystal [1] or perhaps to some as yet unidentified spectral filter in our resonator.

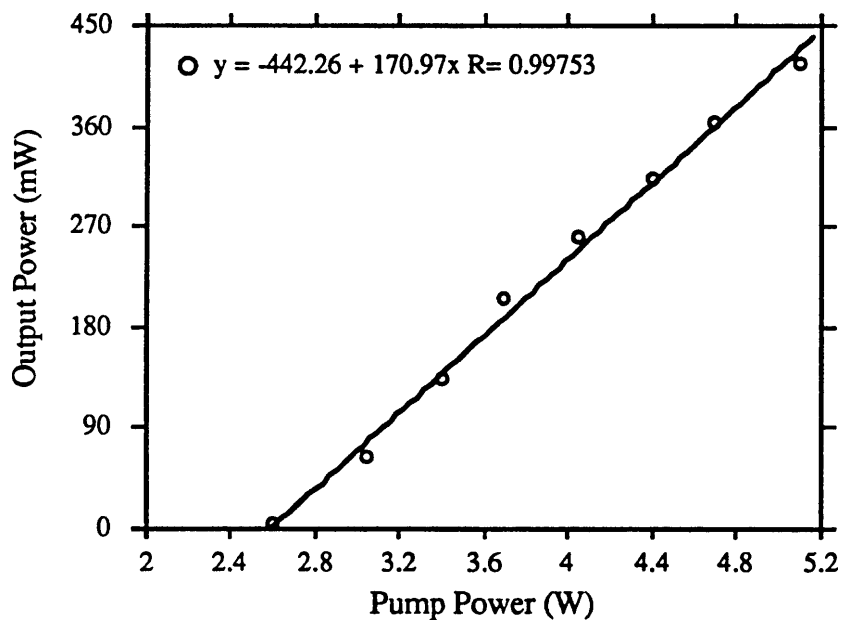
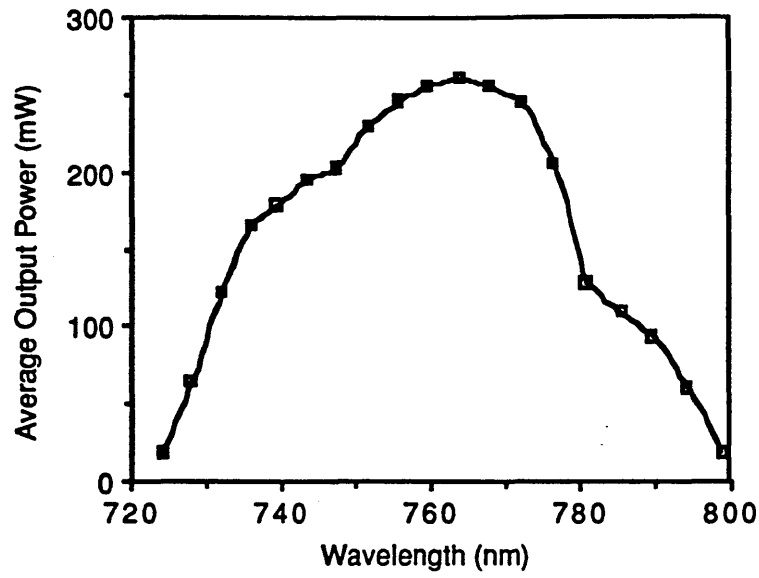
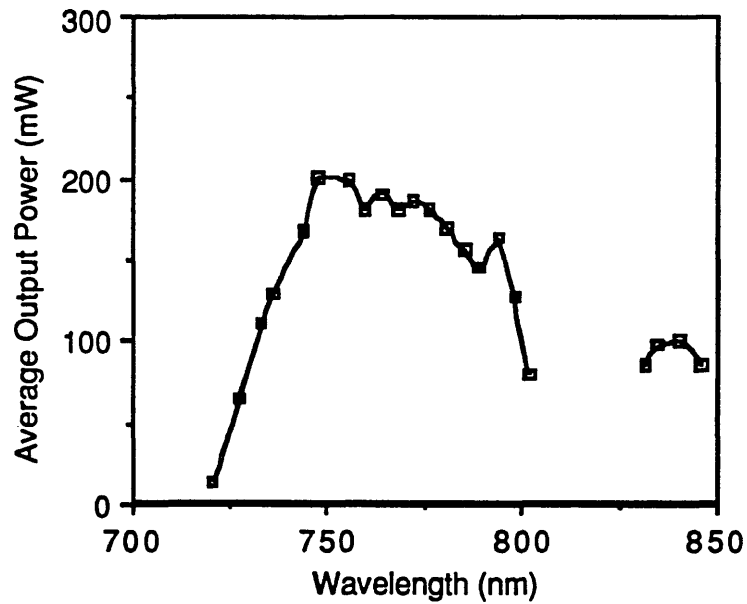


Fig. 6.2.2 Slope efficiency curve for cw Ti:Al₂O₃ laser.[2]

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- 1 R.L. Aggarwal, A. Sanchez, M.M. Stuppi, R.E. Fahey, A.J. Strauss, W.R. Rapoport, and C.P. Khattak, *IEEE Journal of Quantum Electron.* **24**, 1003 (1988)
 - 2 Result obtained by P.M.W. French



(a)



(b)

Fig. 6.2.3 Tuning curves for c.w. Ti:Al₂O₃ laser for (a) 6W pump power and a 4% output coupler and (b) 15W pump power and output coupling from the tuning wedge.

6.3 Mode locked operation

The cavity shown in figure 6.3.1 below was constructed to mode lock the $\text{Ti:Al}_2\text{O}_3$ using active loss modulation at 138MHz from a standard Spectra-Physics acousto-optic modulator (see section 2.3). The output coupler M_3 had 15% transmittance.

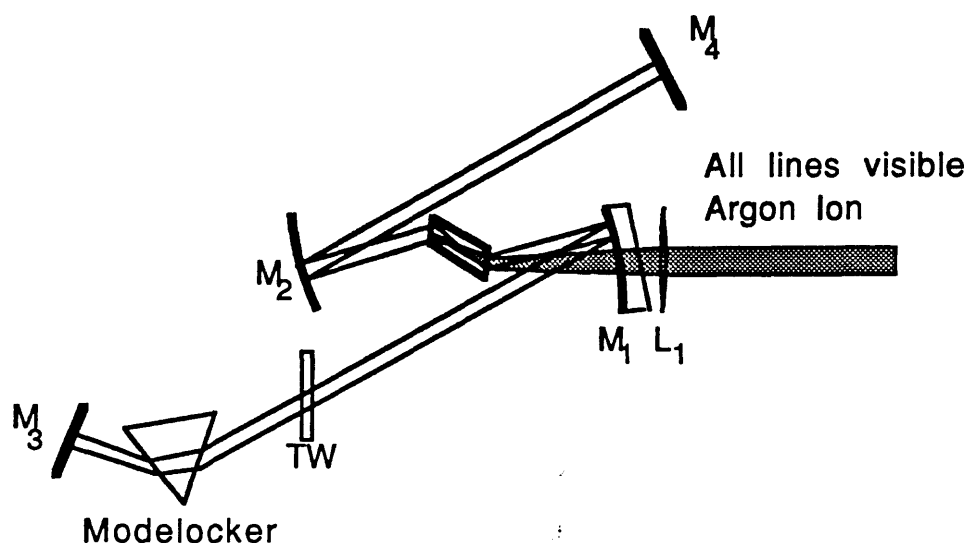


Fig. 6.3.1 Cavity configuration for actively mode locked $\text{Ti:Al}_2\text{O}_3$ laser

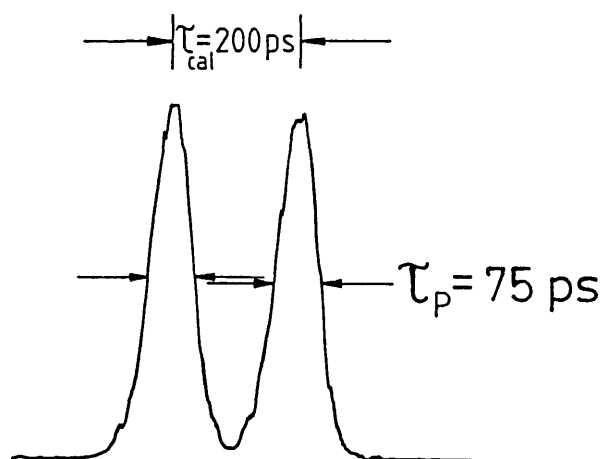


Fig. 6.3.2 Traces from synchroscan streak camera of 75ps pulses from actively modelocked $\text{Ti:Al}_2\text{O}_3$ laser.

The output from this laser was monitored using a synchroscan streak camera driven from the mode locker frequency source. Typically pulses of 75ps duration were obtained as shown in figure 6.3.2.

The modelocked slope efficiency was about 10% as shown in figure 6.3.3 below.

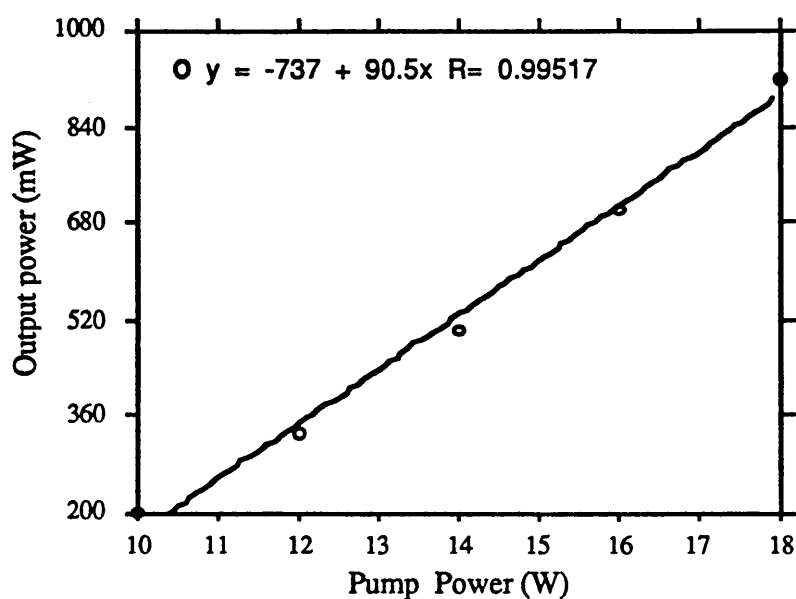


Fig. 6.3.3 Slope efficiency curve for modelocked Ti:Al₂O₃ laser with 15% output coupler.

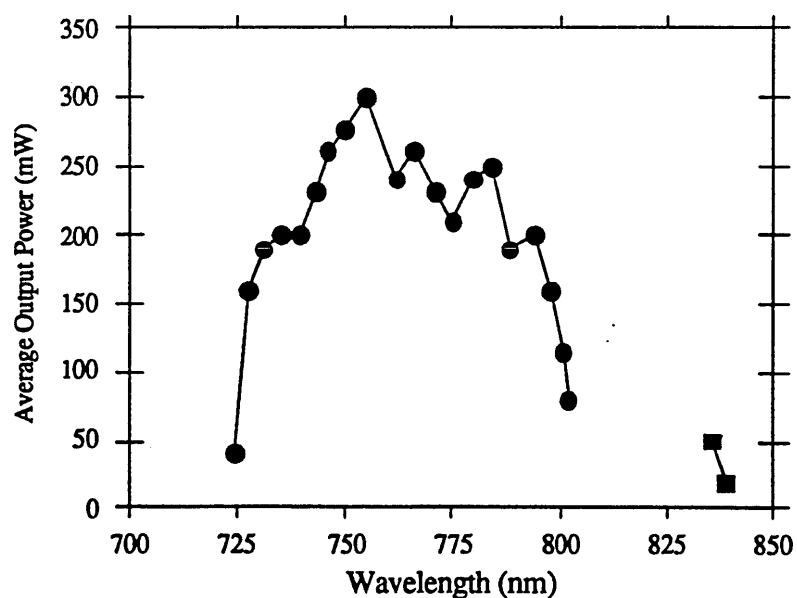


Fig. 6.3.4 Tuning curve for modelocked Ti:Al₂O₃ laser with a 5% output coupler and 15W pump power.

With a 5% output coupler and 15W of pump power the laser tuned from 720nm to 840nm with a gap between 800nm and 830nm as for the c.w. case. Output powers were about 270mW. With a 20% output coupler a maximum average output power of 920mW was obtained for 18W pump. The lasing threshold in this case was 8.2W pump power. Pulse durations were below 100ps throughout. These pulses are longer than those of [1] because a standard prism acousto-optic modulator produces a lower modulation depth and so is not as efficient at coupling the cavity modes.

6.4 Nonlinear resonant feedback operation

(a) Background

The feedback from an empty resonant external cavity into a modelocked ring laser was used experimentally to obtain stable unidirectional mode-locked operation in 1979 [2]. The first use of feedback from a non-linear resonant external cavity to enhance the generation of ultrashort pulses was the soliton laser of Mollenauer and Stolen [3]. With the two cavities matched such that the pulse returning from the external cavity was synchronous with that in the main cavity 60fs pulses were produced from a synchronously pumped colour centre (Ti:KCl) laser which otherwise produced 8ps pulses. This result was explained using the properties of N=2 soliton propagation in the fibre in the external cavity which led to the pulse being fed back being compressed. Subsequently Belanger [4] obtained an integral solution for this action considering the system as a laser injection locked by the compressed pulse being fed back.

Blow and Wood [5] used a numerical simulation which showed that negative dispersion (and thus pulse shortening) in the external cavity was not essential for improving the modelocking. In their work both a fast saturable gain (which can broaden the pulse) and a

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- 1 J.D. Kafka, A.J. Alfrey, and T. Baer, *Proc. Ultrafast Phenomena VI, Springer series in Chem. Phys.* **48**, 64 (1988)
 - 2 V.K. Batovrin, Y.D. Golyaev, A.I. Ladanov, S.V. Lantratov, V.F. Papulovskii, *Sov. Phys. Tech. Phys.* **49**, 1395 (1979)
 - 3 L.F. Mollenauer, R.H. Stolen, *Opt. Lett.* **9**, 13 (1984)
 - 4 P.A. Belanger, *J. Opt. Soc. Am.* **B5**, 793 (1988)
 - 5 K.J. Blow and D. Wood, *JOSA B* **5**, 629 (1988)

fast saturable absorption (which tends to compress the pulse) were considered separately as nonlinear elements in the external cavity, and the relevant equations were iterated to obtain quasi-steady state solutions. It was found, in their simulation, that both nonlinearities could give rise to greatly improved modelocking. The mechanism by which this improvement was achieved involved the nonlinear external cavity inducing stronger coupling between the longitudinal modes of the main cavity and thus enabling increased communication of phase information to the edge of the laser gain bandwidth. This mechanism does not require that the external cavity directly inject energy into the wings of the gain spectrum, and thus pulse compression can still occur even if the pulse in the external cavity is spectrally narrowed (as in the case of the fast saturable amplifier). Stability in this simulation was found to be critically dependant upon the interferometric phase mismatch between the two cavities and thus it was concluded that cavity stabilization would be required in a real system. More detailed numerical work into the dynamics of a laser coupled to an external cavity has been carried out by Kelly [1], and this found that either active, passive or hybrid modelocking behaviour could be obtained depending on the interferometric mismatch between the two cavities. This compression mechanism does not allow a steady state (for a single cavity) to be reached since the external cavity must continually inject new phase information into the mode structure of the laser.

Experimentally Blow and Nelson [2] demonstrated the improved modelocking of a synchronously pumped colour centre laser using an optical fibre operating in the normally dispersive regime (where the pulse is broadened) as the nonlinear element in the external cavity. This experiment found many operating points which enabled improved modelocking, and the results were found to obey the scaling relations of the nonlinear Schrodinger Equation even though the fibre did not support solitons. Similar results have now been obtained by other workers e.g. [3,4] using differing synchronously

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- 1 S.M.J. Kelly, Ph.D. Thesis, London 1989
 - 2 K.J. Blow, B.P. Nelson, *Opt. Lett.* **13**, 1026 (1988)
 - 3 P.N. Kean, X. Zhu, D.W. Crust, R.S. Grant, N. Langford, W. Sibbett, *Opt. Lett.* **14**, 39 (1989)
 - 4 C.P. Yakymyshyn, J.F. Pinto, C.R. Pollock, *Opt. Lett.* **14**, 621 (1989)

modelocked colour centre lasers, and in [1] it was claimed that a nonlinear gain element had been used in the external cavity.

A second treatment of the improved modelocking which a nonlinear external cavity can bring about is the "Additive Pulse modelocking" picture of Ippen et al. [2]. This approach considers the interferometric adding of the pulse in the main cavity with the pulse returning from the external cavity. For the Kerr effect the induced nonlinear phase shift in the external cavity is zero at the peak of the pulse profile, but increases further out in the wings (see Fig. 4.6.1). The pulse coming back from the external cavity interferes with the pulse in the main cavity, and for particular values of cavity mismatch the peaks of the two pulses may be in phase and thus constructively interfere while the wings may be out of phase and destructively interfere. This can lead to a pulse shortening in a single round trip of the cavities. Experimental results have been given in the above reference to support the predictions of the model.

Since this technique has been shown, both theoretically and experimentally, to work in the normally dispersive regime it was believed that it may be applicable to the Titanium sapphire laser for which, because it lases at too short a wavelength, there is no soliton supporting (anomalously dispersive) fibre available. It was also important to show experimentally that the technique was applicable to laser systems other than those based on colour centres.

(b) Experiment

The experimental configuration used for the $\text{Ti:Al}_2\text{O}_3$ laser with nonlinear feedback from an external cavity containing an optical fibre is shown in figure 6.4.1 below.

Initially Mirror M_4 was used as the output coupler (20% transmittance), from which the power was coupled into the nonlinear external cavity. The nonlinear element in the external cavity was 7.3m of birefringent single mode fibre (YORK HB750). With 14W of pump power there was 600mW of output power of which (due to the small core size) only 200mW could be coupled

1 P.N. Kean, X. Zhu, D.W. Crust, R.S. Grant, N. Langford, W. Sibbett, *Opt. Lett.* **14**, 39 (1989)

2 E.P. Ippen, H.A. Haus, L.Y. Liu, *J. Opt. Soc. Am.* **B6**, 1736 (1989)

into the fibre. Autocorrelation and spectral measurements of the pulses entering and leaving the fibre did not resolve any temporal or spectral broadening. This indicates that the dispersion and SPM of pulses of this duration and power in a fibre of this length are very small.

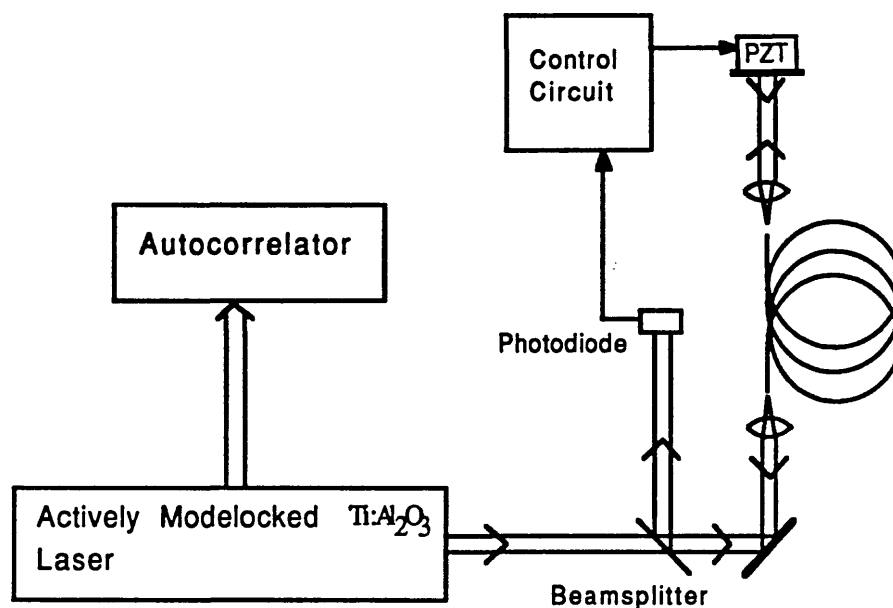


Fig. 6.4.1 Schematic of femtosecond $\text{Ti:Al}_2\text{O}_3$ laser with feedback from a nonlinear external cavity.

The resonance condition of the external cavity was determined by looking at the output from the tuning wedge on a fast photodiode. Although the fed back pulse could not be seen on the photodiode, near resonance the main pulse from the cavity was enhanced. On the streak camera the pulse was broadened and pulled either forward or backward in time (relative to the streak camera drive and thus modulator signal) depending on the relative mismatch of the feedback. Within $\sim 100\mu\text{m}$ of resonance the signal on the streak camera was not displaced in time but did not appear to be shortened. At this condition a rather noisy enhancement could be seen in the second harmonic signal of the autocorrelator, and some spectral broadening (bandwidth up to 0.5nm) was measured.

A servo stabilization scheme based on the design of Mitschke and Mollenauer [1] was employed to interferometrically match the main and external cavity lengths. This works by assuming that the power in the control cavity varies sinusoidally with ϕ the round

1 F.M. Mitschke and L.F. Mollenauer, *IEEE J. Quantum Electron.* QE-22, 2242 (1986)

trip optical phase shift. This power is monitored using a slow detector (a silicon diode) and is amplified to drive a piezo translator coupled to the feedback mirror. Servo adjustments include gain, integration time and the level and sign with which the monitored signal is compared (effectively setting ϕ). The system employed is described in Appendix C. Varying these parameters did not produce a more stable second harmonic signal, nor could a short pulse be observed on the autocorrelation profile, and it was found that out of phase optical feedback from the fibre ends was affecting the system.

To prevent the unwanted reflections from the fibres feeding back and disturbing the system two steps were taken. First the fibre was cut to have an optical path equal to a multiple of the main cavity length so that the feedback from both ends were in phase. Then the output was taken from mirror M_3 next to the modelocker prism, and the fibre was placed $0.25 \times$ the cavity length from this. Thus the unwanted out of synchronization feedback was rejected by the modelocker before it reached the gain in the main cavity. With this new scheme the enhancement in second harmonic signal by having the external cavity was found to be greater than before. However the signal was still very noisy and autocorrelation traces could not be taken.

To optimise the system the maximum enhancement of second harmonic signal achieved with feedback over that occurring without was plotted against the power measured at the output of the fibre for several different lengths of fibre. The results are presented below from which it can be seen that the shorter the fibre and the higher the power the larger the enhancement. Since the larger the second harmonic enhancement the larger the pulse compression this observation agrees with that of Blow et al. [1] for an F-centre laser using a nonsoliton external cavity and thus supports the idea that the pulse shortening scales according to the nonlinear Schrodinger equation. The two curves for the 63cm fibre case occur because for low powers the measured enhancement was unstable between these two values. This is not at present understood. In taking these measurements the interferometric servo control was not turned on and the signal was very noisy.

1 K.J. Blow and B.P. Nelson, *Opt. Lett.* **13**, 1026 (1988)

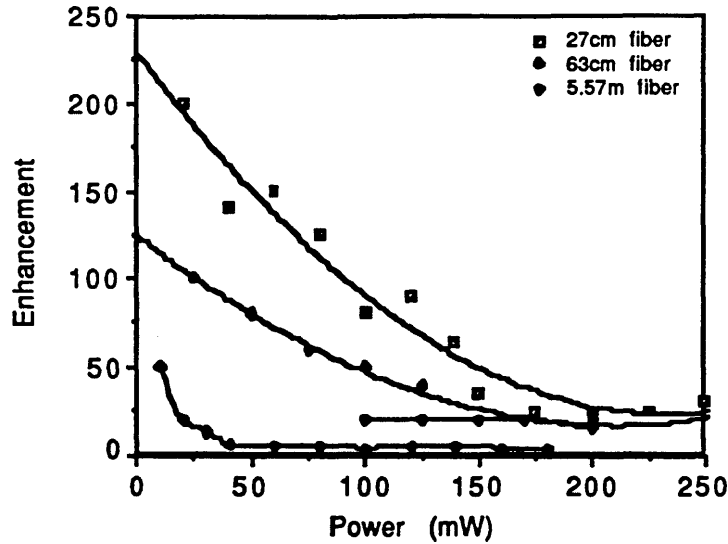


Fig. 6.4.2 Peak Second harmonic enhancement with the nonlinear external cavity versus power in the fibre for varying fibre lengths

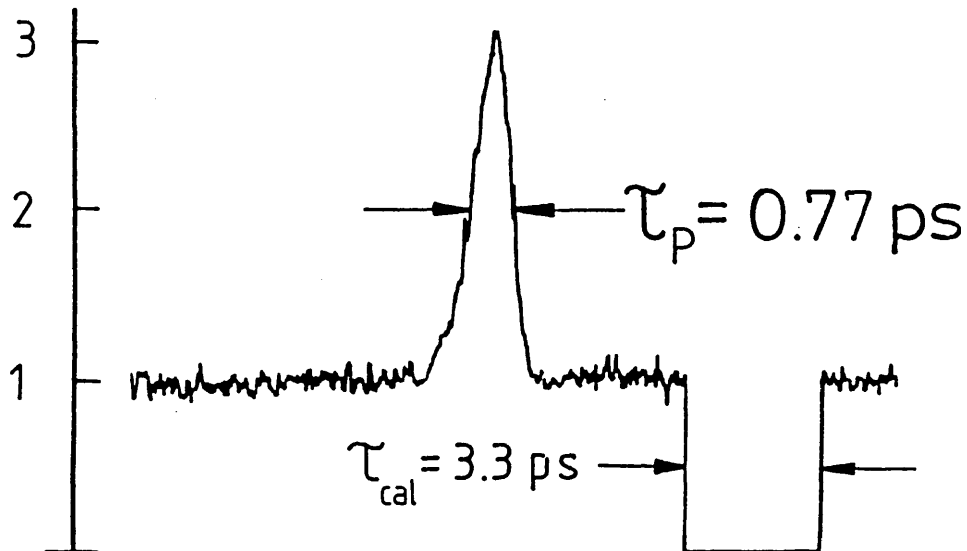


Fig. 6.4.3 Autocorrelation trace of pulses of 770fs duration.

With the shortest fibre length (27cm) in the external cavity short pulses were observed on the autocorrelator. The output from the system was stabilized by optimising the adjustments on the servo amplifier. With 200mW average power coupled into the fibre, pulses of 770fs duration (within 10% of their transform limit assuming sech^2 pulse profiles) were generated using the standard collinear SHG autocorrelation technique with Lithium Iodate employed as the doubling crystal. Figure 6.4.3 shows the autocorrelation trace of these pulses recorded at 788nm.

6.5 Conclusions

I have shown here successful c.w. and actively modelocked operation of a $\text{Ti:Al}_2\text{O}_3$ laser using a commercial grade crystal. The modelocked system generated sub-100ps pulses over a tuning range extending from 725nm to 840nm, with typical average output powers of 1W. The generation of sub-picosecond pulses was achieved for the first time directly from a $\text{Ti:Al}_2\text{O}_3$ laser by employing nonlinear feedback from a resonant external cavity. This result again confirms that feedback from a nonlinear external cavity into a modelocked laser cavity can lead to improved modelocking without directly injection seeding the laser bandwidth. This is the first time that this modelocking technique has been successfully employed with systems other than colour centre lasers.

The minimum pulse durations reported here are somewhat longer than those of the soliton laser and this is thought to be due the birefringence of the Brewster-angled $\text{Ti:Al}_2\text{O}_3$ rod acting as a tuning element and limiting the available bandwidth. This proposition is backed up by the observation that the output wavelength jumps in 4nm steps which we believe arises from the birefringence of the rod.

Subsequent to the work presented here the group at MIT have obtained 1.6ps pulses directly from a $\text{Ti:Al}_2\text{O}_3$ laser employing nonlinear external cavity feedback [1], and which was not otherwise modelocked - showing that the pulse formation process can start from noise (i.e. no seed pulse was required).

Furthermore, very recent work [2] has shown that a resonant linear (i.e. empty) external cavity which has its return mirror dithered slowly can lead to modelocked operation of the $\text{Ti:Al}_2\text{O}_3$ laser, generating 40psec pulses without a modulator, and further characterization and numerical modelling is required to understand this rather surprising result.

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- 1 J. Goodberlet, J. Wang, J.G. Fujimoto, Proceedings of CLEO 1989, Paper FQ4.
 - 2 P.M.W. French, J.A.R. Williams, and J.R. Taylor, Presented at QE9, Oxford (1989)

7. GENERAL CONCLUSIONS

The understanding of femtosecond optical sources, and their subsequent development is important for time resolved spectroscopy, which requires not just shorter pulses, but also reliability and tunability. It is hoped that this thesis has contributed to the understanding, and development of femtosecond pulse laser sources.

In chapter 2 the importance of dyes as the active media for ultrashort pulse lasers has been discussed. The various modelocking techniques have been reviewed, and their merits and demerits discussed. Passive modelocking of c.w. dye lasers is found to be not only the source of the shortest, but also the most stable pulses, however it has in the past suffered from limited tunability. Argon-ion pumped systems using new active/passive dye combinations have been demonstrated over the visible part of the spectrum by French and Taylor [1].

Previous to the work presented here, there was however, only one passively modelocked dye laser demonstrated in the near infrared part of the spectrum [2], and this system required Krypton-ion pumping and therefore was not particularly stable. In chapter 3 two passively modelocked dye laser systems are described which extend the tuning range further into the near infrared part of the spectrum, and because these systems employ energy transfer dye mixtures as the gain media, they can be pumped by the more conventional and stable argon-ion laser.

The development of intracavity dispersion compensated passively modelocked dye lasers arose from the understanding that phase modulation effects, and group velocity dispersion are important in ultrashort pulse generation [3,4,5]. The understanding of these effects in CPM dye lasers has traditionally been based on the Nonlinear Schrodinger equation, which successfully describes soliton propagation in optical fibres [6]. Analytical treatments of

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- 1 P.M.W. French and J.R. Taylor, *Appl. Phys. Lett.* **50**, 1708 (1987)
 - 2 K. Smith, N. Langford, W. Sibbett, and J.R. Taylor, *Opt. Lett.* **10**, 559 (1985)
 - 3 J.C. Diels, J.J. Fontaine, I.C. McMichael, B. Wilhelmi, W. Dietel, D. Kuhlke, *Sov. J. Quantum Electron.* **13**, 1562 (1983)
 - 4 J.A. Valdmanis, R.L. Fork, J.P. Gordon, *Opt. Lett.* **10**, 131 (1985)
 - 5 P.M.W. French, J.R. Taylor, *Opt. Commun.* **61**, 224 (1987)
 - 6 J.-C. Diels, W. Dietel, J.J. Fontaine, W. Rudolph, B. Wilhelmi, *J. Opt. Soc. Amer. B* **2**, 680 (1985)

the passive modelocking mechanism, however, have employed the self reproducing profile principle and thus required that a steady state be reached [e.g. 1,2,3]. Recent experimental results [e.g. 4,5], have shown that these premises may not always be true in CPM dye lasers. In chapter 4 an experimental investigation was carried out into the existence in a passively modelocked Rhodamine 110 CPM ring dye laser modelocked with HICI and DASBTI of instabilities and satellite pulses which exist over a certain range of intracavity dispersion. The agreement between these experimental results, and those from a simulating model constructed by H. Avramopolous [6] to investigate the pulse evolution was found to be qualitatively excellent [7]. In the model the satellite pulses and pulse energy modulation behaviour occurred for zero and negative group velocity dispersion values, and arose not from high order soliton formation but from an accumulative spectral walk-off of the pulse arising from asymmetric phase modulation. This walk-off caused loss of energy to the filter and thus enabled formation of a satellite pulse. Subsequent to this work several authors have reported similar instabilities [8,9,10], and it is believed that the proposed mechanism affects the pulse formation in femtosecond passively modelocked dye lasers in general. By comparison between model and experiment we believe that the position of zero intracavity group velocity dispersion in CPM dye lasers can be determined for the first time.

Relatively little attention has been paid to the early transient dynamics of pulse evolution in previous studies of c.w. pumped femtosecond passively modelocked dye lasers. The timescales associated with the pulse formation are important if one wishes to cavity dump such a laser at a high repetition rate [11] or to use a

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- 1 O.E. Martinez, R.L. Fork, J.P. Gordon, *Opt. Lett.* **9**, 156 (1984)
 - 2 O.E. Martinez, R.L. Fork, J.P. Gordon, *J. Opt. Soc. Amer. B* **2**, 753 (1985)
 - 3 H.A. Haus, Y. Silberberg, *IEEE J. Quantum Electron.* **QE-22**, 325 (1986)
 - 4 P.M.W. French, J.R. Taylor, *Opt. Commun.* **61**, 224 (1987)
 - 5 J.A. Valdmanis, R.L. Fork, *IEEE J. Quantum. Electron.* **QE-22**, 112 (1986)
 - 6 H. Avramopolous PhD Thesis, London University (1989)
 - 7 H. Avramopoulos, P.M.W. French, J.A.R. Williams, G.H.C. New and J.R. Taylor, *IEEE J. of Quantum Electron.* **QE-24** (1988) p.1884
 - 8 F.W. Wise, I.A. Walmsley, C.L. Tang, *Opt. Lett.* **13**, 129 (1988)
 - 9 F. Salin, P. Grangier, G. Roger, A. Brun, *Phys. Rev. Lett.* **56**, 1132 (1986)
 - 10 W.L. Nighan, T. Gong, P.M. Fauchet, *Opt. Lett.*, **14**, 447 (1989)
 - 11 M.C. Downer, R.L. Fork, M. Islam, *Ultrafast Phenomena IV*, Springer-Verlag Series in Chemical Physics vol. 38, pp. 27-29 (1984)

quasi-cw or pulsed pump source [1]. In chapter 5 I have carried out an experimental investigation of the evolution to a steady state of femtosecond pulses in the passively modelocked Rhodamine 110 CPM ring dye laser. The results indicate that initially there is a delay of $\sim 10^4$ cavity round trips for the intracavity circulating energy to build up to the level at which gain and absorption saturation become significant, and the modelocking process begins. From this point the pulse evolution reaches a steady state - with or without a periodic evolution - after $\sim 10^3$ cavity round trips. This result is in good agreement with the numerical estimates of Petrov et al. [2] which include the off-resonant interaction the saturable absorber. It was also found that the pulse energy overshoot it's steady value during the pulse evolution, and this we believe is evidence of the limited spectral support in the cavity - the pulse energy may overshoot because an initially broad pulse has a narrow spectrum and suffers less loss to the filter than the steady state short pulse. In addition measurements were made on the effect of glass path on pulse build up, and it was found that intracavity dispersion only became important once short pulses had been formed. Spectral measurements showed the effects of the time dependant absorption saturation on the intracavity spectral filter, with a narrow spectral spike, corresponding to the satellite pulse occurring for negative near zero dispersion values, occurring at shorter wavelengths than the broad spectral component corresponding to the main pulse. This occurred because the satellite pulse occurs after the main pulse where the saturable absorber is still fully saturated. Thus the satellite pulse has less loss than the main pulse at shorter wavelengths.

These results show the importance of the intracavity spectral filter in passively modelocked dye lasers, and further theoretical work into pulse evolutions in CPM dye lasers will be required to include the time dependence of this filter arising from absorber saturation, as well as the phase effects.

In chapter 6 a possible alternative for the modelocked dye laser as a source of ultrashort pulses in the near infrared has been

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- 1 H. Ansari, A. Dienes, J.R. Whinnery, *Optics Letters*, vol. 10, pp. 19-21 (1985)
 - 2 V. Petrov, W. Rudolph, B. Wilhelmi, *Optical and Quantum Electronics*, vol. 19, pp. 377-384 (1987)

described. This is the Titanium doped sapphire laser, which with its very broad tuning range in one system provide the same spectral coverage as many different dye laser systems. Characterization of c.w. and modelocked operation of such a laser has shown the possibilities and limitations of employing a commercial rather than research grade $\text{Ti:Al}_2\text{O}_3$ crystal. Active modelocking enabled the generation of 80ps pulses, and up to 1W average output power while pumping with a large frame argon-ion laser. Subpicosecond pulse generation from this source was obtained for the first time using the relatively recent technique exploiting feedback from a nonlinear external cavity. This result again confirms that pulse compression in the external cavity is not required to give an overall pulse compression, and therefore gives further evidence for the mode-coupling mechanism [1] of operation of such lasers. This is the first time that such a technique has been successfully employed with systems other than colour centre lasers.

Other workers [2] have now shown that the modelocking action can start without a modelocker in the main cavity and therefore that the process can start from noise. Very recently it has been found that modelocking can occur in this laser with a linear (empty) external cavity. This result still requires explaining. Future work in this area would probably require the use of higher quality $\text{Ti:Al}_2\text{O}_3$ crystal to overcome the tuning problems associated with the material used for this work. Further characterization is also necessary in these systems to improve stability, however if the problems are overcome there will be a robust, high power femtosecond laser source broadly tunable in the near infrared.

1 K.J. Blow and D. Wood, *JOSA B*5, 629 (1988)

2 J. Goodberlet, J. Wang, J.G. Fujimoto, Proceedings of CLEO 1989, Paper FQ4.

APPENDICES

A - The phase distortion arising from dielectric mirrors.

Dielectric mirrors, which because of their very high reflectivities are usually used in laser cavities, normally consist of a number of layers of dielectric material of alternating high and low refractive indices. Normally the dielectric layers have a thickness of $\lambda/4$, where λ is the required central wavelength of the high reflectivity zone of the mirror. Extended high reflectivity zones may be obtained by placing two stacks of differing layer thicknesses, one on top of the other. The standard matrix formalism for multilayer high reflectance coatings enables, for a given stack, a given wavelength, and a given angle of incidence, the calculation of complex amplitude reflectance and transmission coefficients for each of the two polarizations (s and p). Applying this formalism over a range of wavelengths enables the generation of the curves such as figures 2.5.1 and 2.5.2. Although plots of group velocity dispersion have also been calculated, the phase response of the mirrors is by no means linear and higher order terms are important when considering the effects of such mirrors on pulses. In order to calculate the effect of these mirrors on ultrashort pulses at differing wavelengths I constructed an algorithm which uses the discrete Fast Fourier Transform to determine the frequency components of a pulse of a particular duration, and a particular carrier frequency. These complex frequency components are then multiplied by the corresponding calculated mirror amplitude reflectivities, and transformed back to the time domain. The results for a sech^2 pulse profile reflected from differing mirror types are presented in figure A below.

Fig. A Graphs showing variation of the reflected pulse intensity profile (i), and FWHM of this profile (ii) versus wavelength for 100fs (a) and 30fs (b) incident sech^2 pulse profiles. A.1 is for a single stack coating (i.e. $\lambda/4$ stacks in the form {air H (LH)⁹ glass} with $n_L=1.45$ and $n_H=2.28$ and centred on 563nm). A.2 is for a double stack, with the second stack, layer thicknesses 1.25 times those of the first stack which is the same as that in A.1, on the outside. A.3 has the thicker stack on the inside. The mirror intensity reflectivity coefficient is shown alongside each graph.

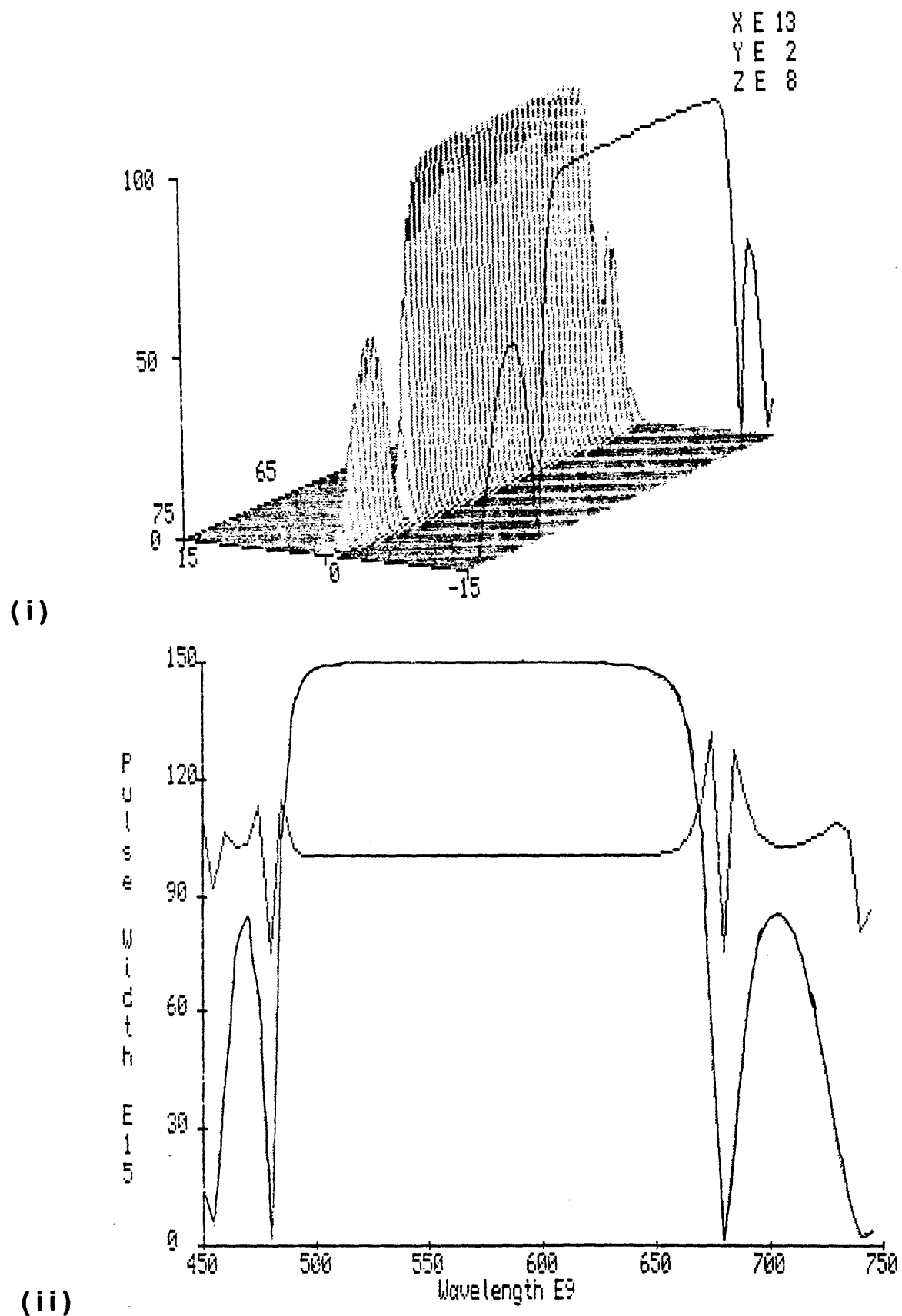


Fig. A.1 (a)

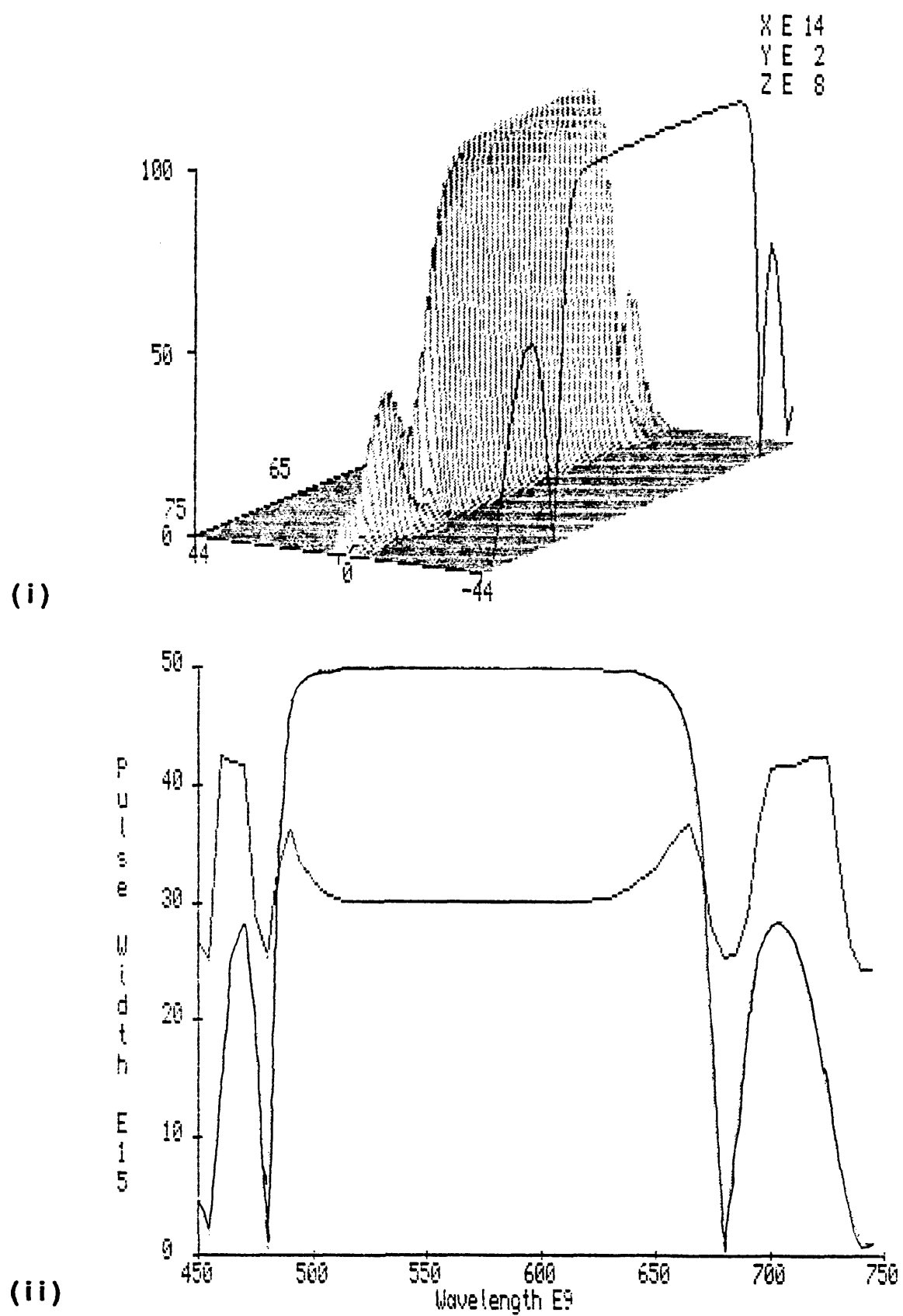


Fig. A.1 (b)

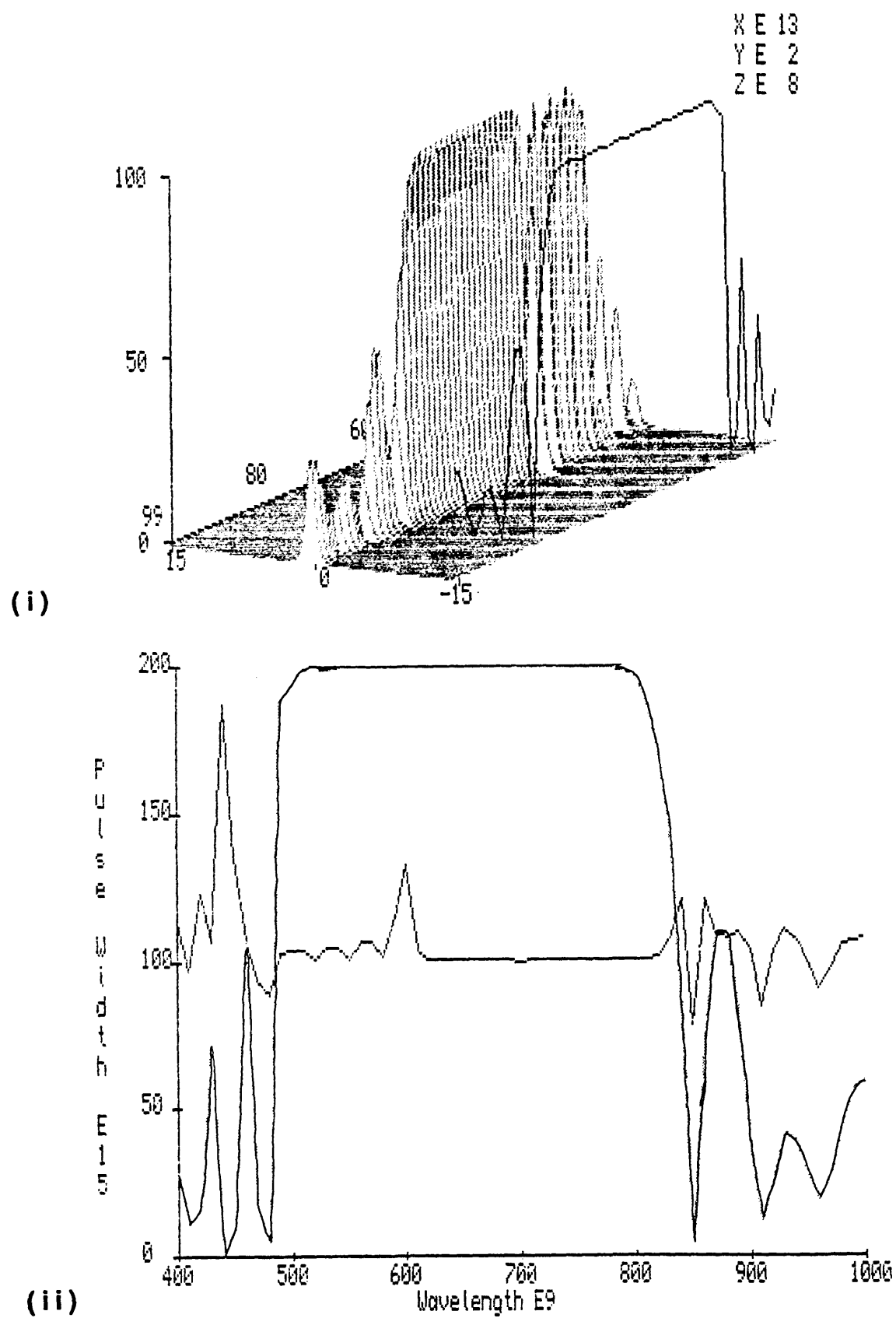


Fig. A.2 (a)

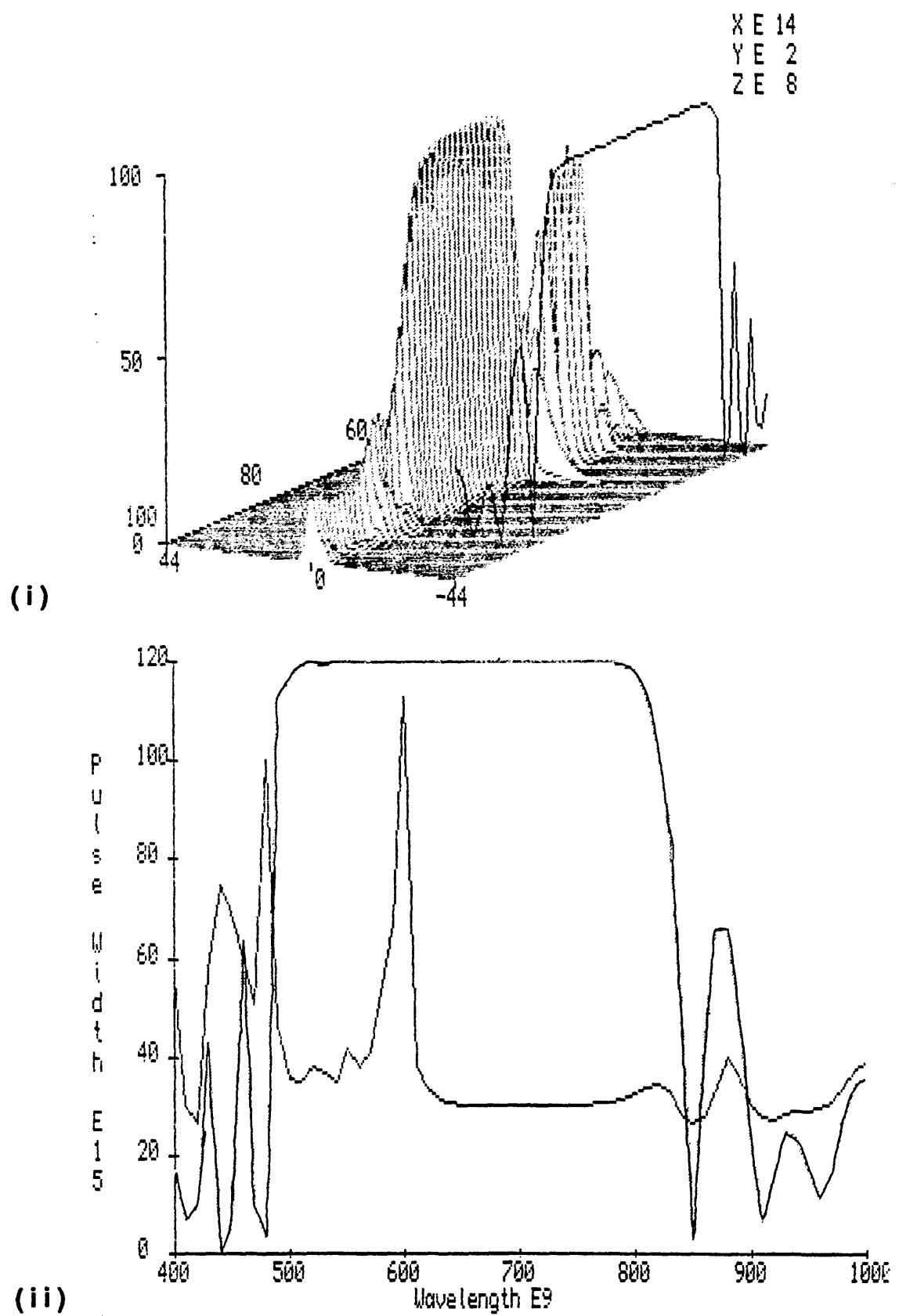


Fig. A.2 (b)

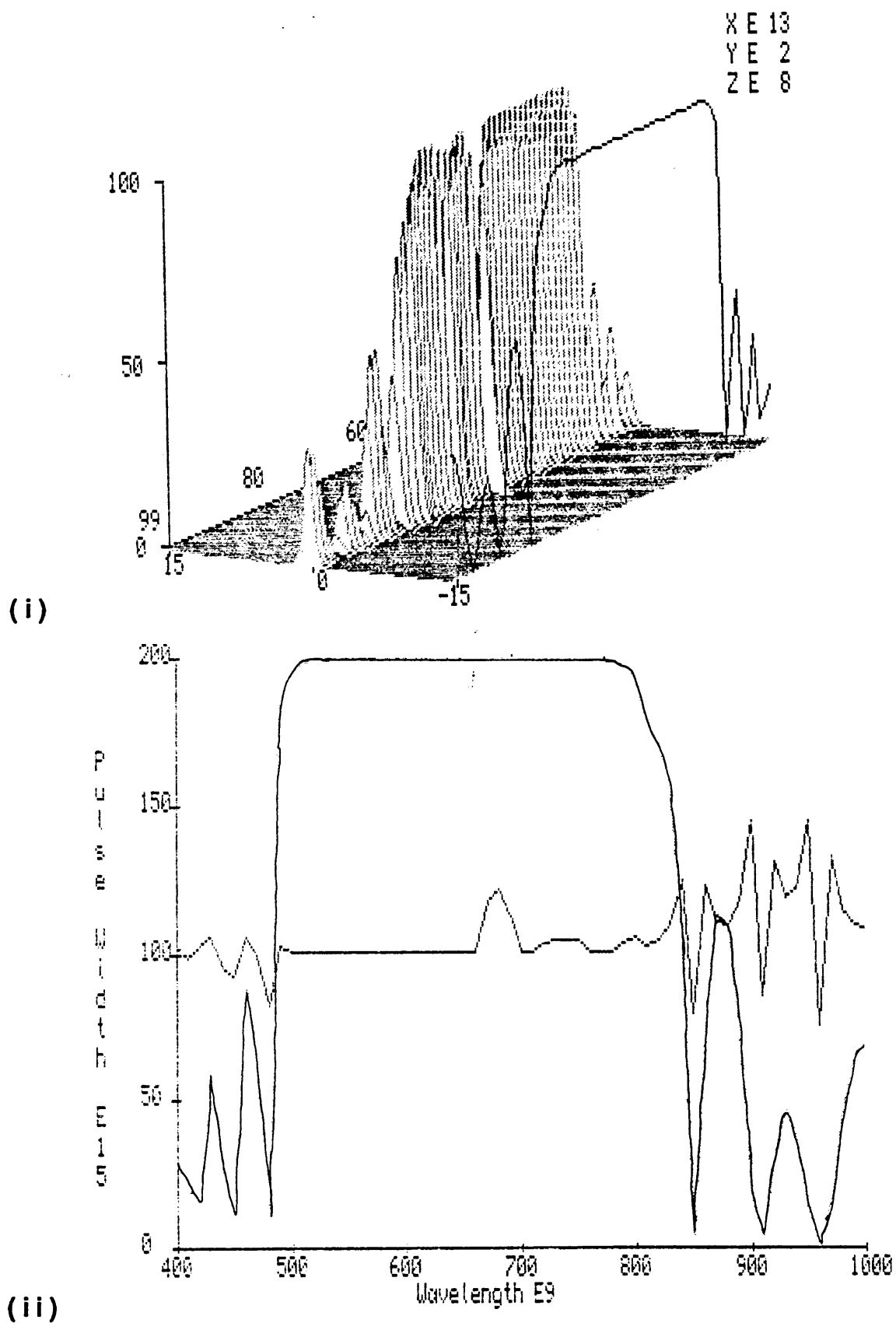


Fig. A.3 (a)

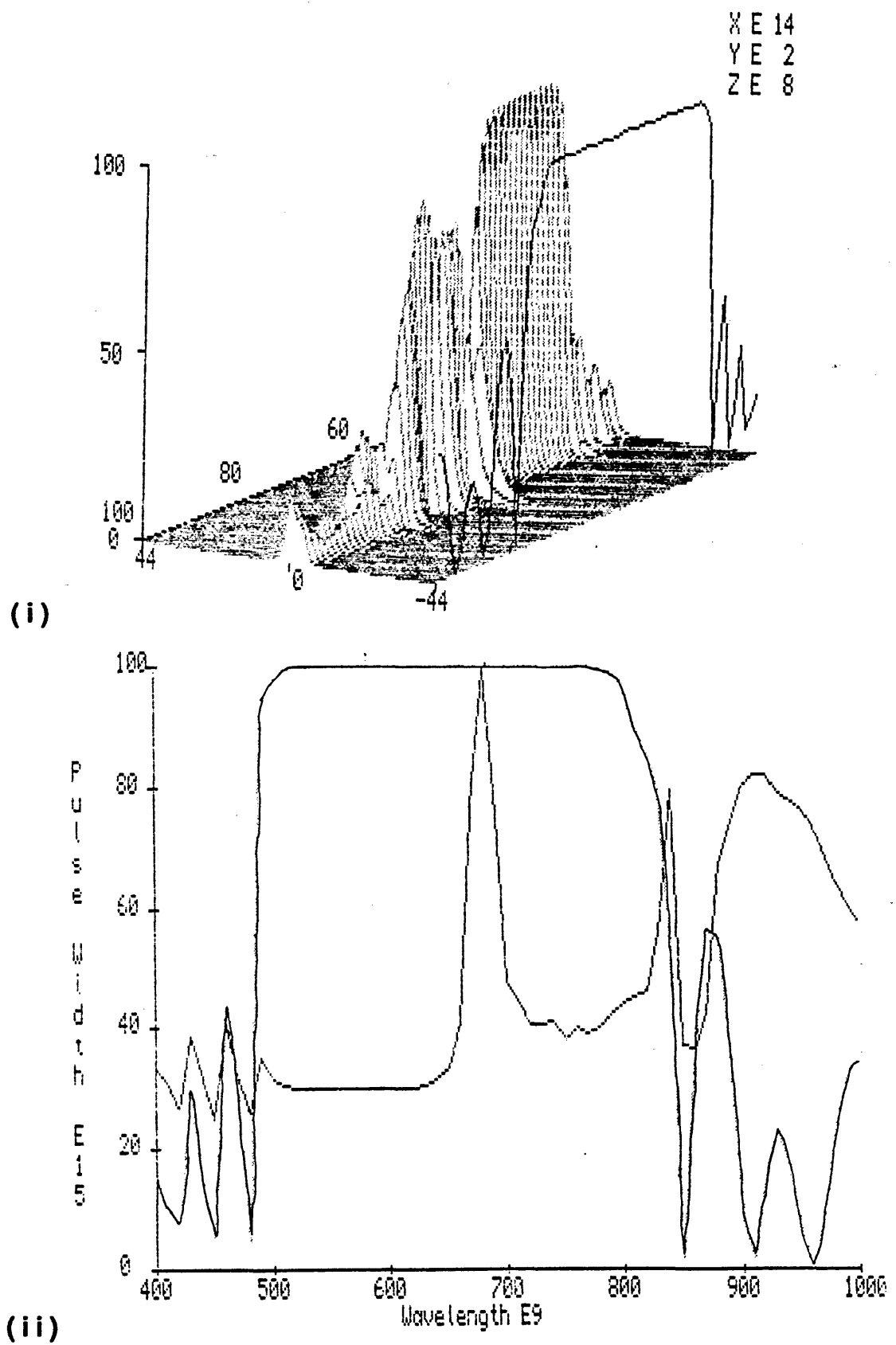


Fig. A.3 (b)

Looking first at the single stack results A.1 we find that the pulse is only distorted if some of its bandwidth lies outside the high reflectance zone of the mirror. In particular the 100fs pulse is broadened by about 10% at a reflectivity at its carrier wavelength of ~96%, and the 30fs pulse is broadened by about 18% at the same wavelength. A reduction in pulse width may also occur when the spectrum of the pulse straddles a low reflectivity resonance of the mirror. This preferentially reduces the energy at the centre of the bandwidth, increasing relatively the contribution from the wings. The reflected pulse is therefore much less energetic, but shorter, and there may also be a satellite pulse produced.

In the case of the two types of broadband stacks A.2 & A.3 very large amounts of pulse broadening occur inside the high reflectance zone if the pulse has to travel through the outer stack to be reflected by the inner. In particular near the centre of the high reflectance zone a 30fs pulse (A.2.(b) & A.3. (b)) may be broadened up to as much as 100fs on a single reflection. This is obviously a very major change indeed. However even a 100fs (A.2.(a) & A.3.(a)) pulse is noticeably broadened, and in a laser cavity where many reflections occur on each mirror such distortion would be sufficient to prevent ultrashort pulse generation. Broadband dielectric stacks are therefore unsuitable for use inside femtosecond laser resonators, and even on a single reflection they can lead to severe distortion of an ultrashort pulse.

B - The scanning autocorrelator

The scanning autocorrelator used for this work is of the standard design. The shaker used (Bruel & Kjaer type 4810 Mini-Shaker) on one arm of the interferometer can scan over a displacement of about 6mm when driven with a sinusoidal waveform of up to 7W r.m.s. power. The generation of this sine wave is carried out using the circuit whose schematic is shown overleaf. There are three main units in this drive unit - an oscillator, a delay line and a power amplifier.

The oscillator circuit employs an 8038 waveform generator chip as the main semiconductor, however such devices when used in the standard circuit are unstable at low frequencies (which may be useful when performing interferometric autocorrelations), and for this reason in this circuit there is an extra level of frequency control feedback. The oscillator has an external frequency adjustment between about 5 and 150Hz. There are three outputs from the oscillator, a sine wave and two trigger lines (one for driving low impedances, the other for driving high impedances) which are in phase with the sine wave.

The sine wave from the oscillator goes through a delay line using a TDA 1022 semiconductor. The delay circuit is of standard design taken from R.S. data sheet 5077. The amount of delay imposed is controllable externally between 0.5 and 50ms.

Finally the delayed sinewave goes through a level control and is amplified by a high power amplifier circuit using a 165 op. amp. (as of R.S. data sheet 5386) before driving the shaker.

The signal from the autocorrelator photomultiplier is monitored on an oscilloscope with the timebase triggered by the circuit. About 1/6 of the sine wave is linear (to >95%) and can be used for autocorrelation purposes, and by varying the delay it is possible to make sure that this linear portion with the autocorrelation trace is in the centre of the oscilloscope screen. Normally a frequency of about 20Hz is used to give a real time trace without excessive vibration, although lower frequencies enable interferometric resolution if required.

C - The stabilization system for the nonlinear external cavity

A servo stabilization scheme based on the design of Mitschke and Mollenauer [1] was employed to interferometrically match the main and external cavity lengths. This works by assuming that the power in the control cavity varies with ϕ the round trip optical phase shift. A mirror on one of the cavities is mounted on a piezoelectric transducer (Burleigh) which with a 150V signal can be moved 30 μm . The circuit to drive this transducer is in two parts, a large d.c. power supply and the servo amplifier itself.

The d.c. power supply (figure C.1) can provide 0-120 V D.C. controllable via a 10 turn pot. This enables accurate manual control of the piezoelectric transducer. The output from this power supply forms the signal ground of the servo amplifier (figure C.2).

The signal for the servo amplifier is provided by a silicon photodiode operating in photovoltaic mode. This is operated upon by a set of four op. amps which provide gain, damping, and control of the sign of the output. A final amplification stage of a darlington transistor pair provides protected driving of the piezo. The signal output from this amplifier has a $\pm 15\text{V}$ range about the input from the power supply which corresponds to $\pm 3\mu\text{m}$ translation about the preset position. This should normally be sufficient.

1 F.M. Mitschke and L.F. Mollenauer, *IEEE J. Quantum Electron.* QE-22, 2242 (1986)

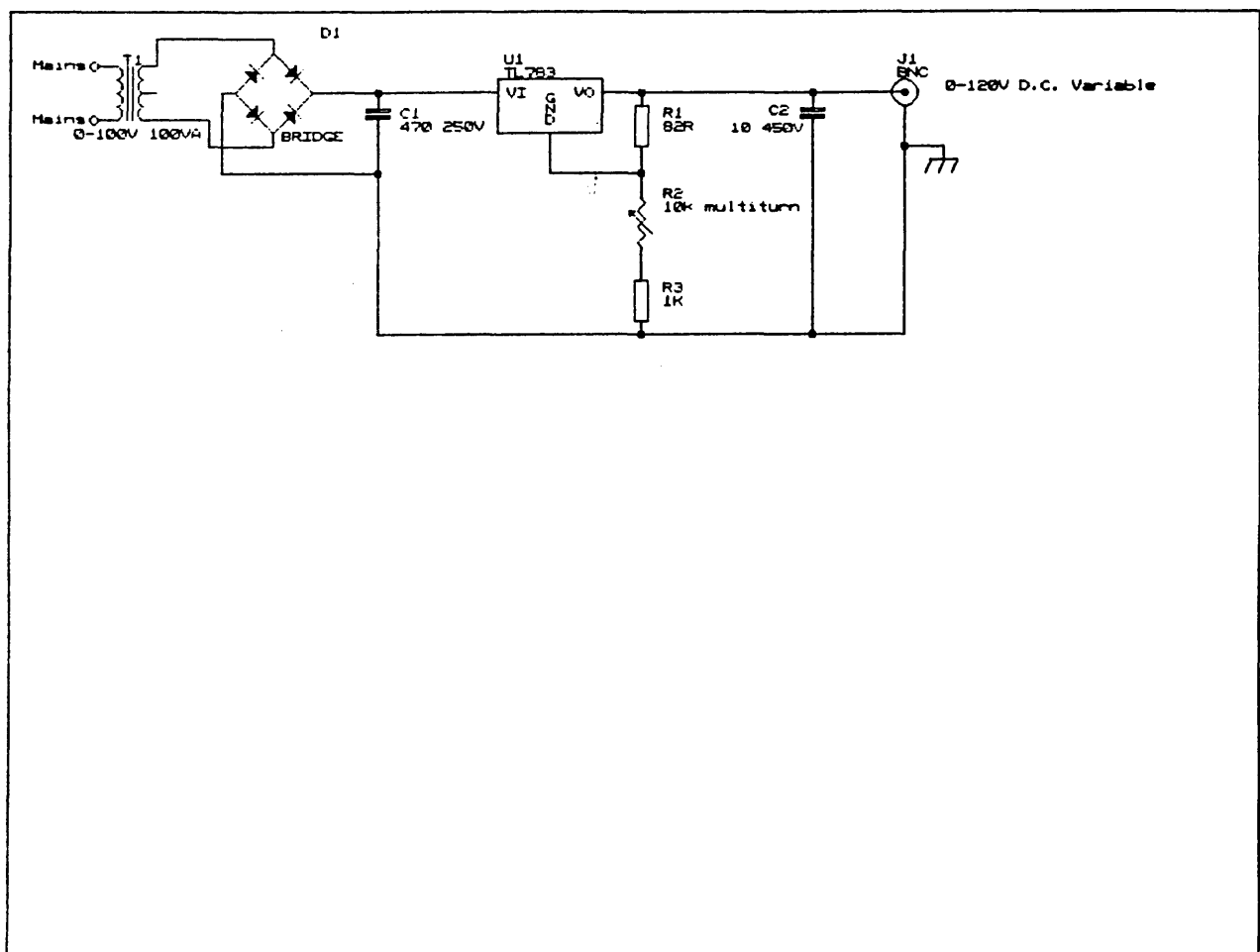


Fig. C.1 The manual variable 0-120 V d.c. piezo power supply circuit

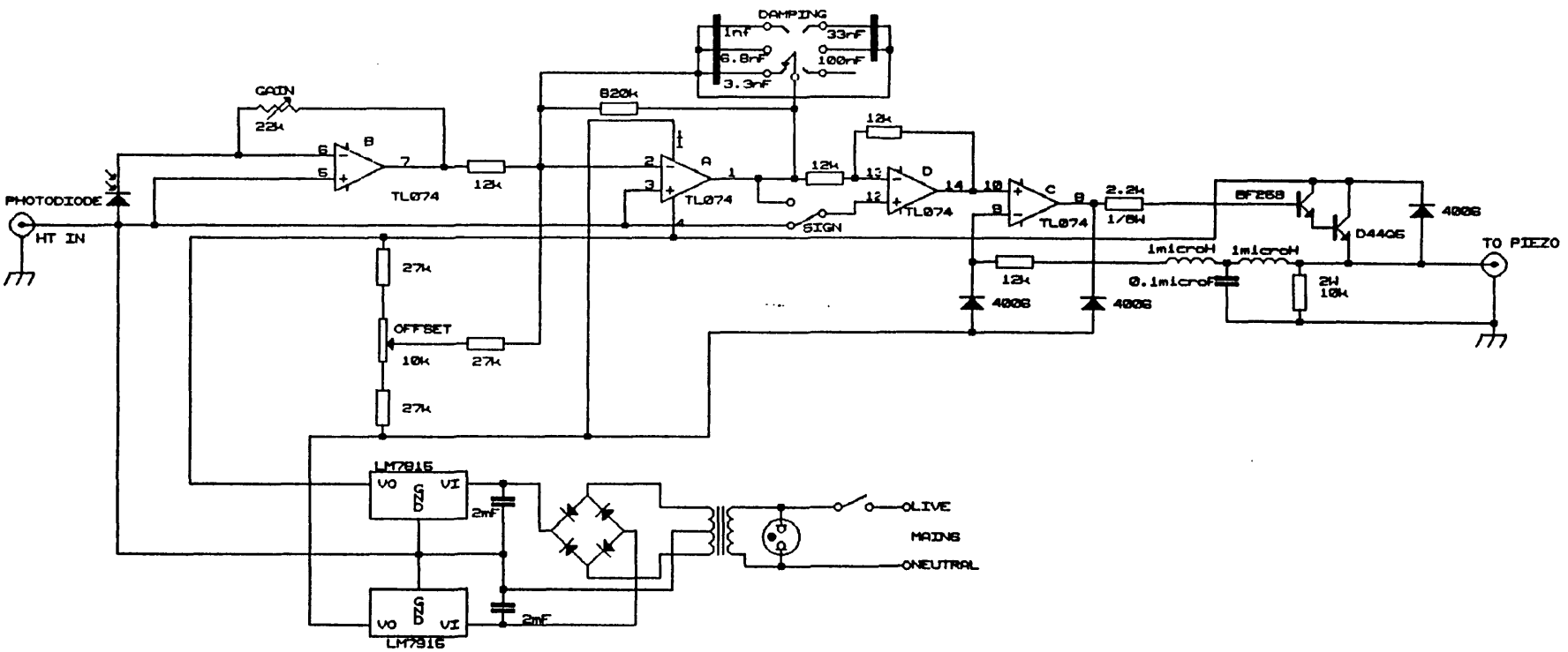


Fig. C.2. The servo amplifier circuit.

ACKNOWLEDGEMENTS

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