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ACTIVE AND PASSIVE MODE LOCKING OF CONTINUOUSLY OPERATING RHODAMINE 6G DYE LASERS

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FOREWORD

This report summarizes the research performed by Dr. André Scavennec during his stay at the National Bureau of Standards, Boulder, Colorado, as a visiting guest scientist from September 1972 to August 1973. Dr. Scavennec's home laboratory is the Centre National D'Etudes des Télécommunications, Issy-les-Moulineaux, France. This report is typical of the high quality research performed in the international scientific exchange programs that NBS participates in and strongly supports. The major discovery of this research, namely the three mirror cavity for a passive mode-locked dye laser, has been reported to the world-wide scientific community. A letter describing this discovery will be published in the January 1974 issue of the IEEE Journal of Quantum Electronics.

The second author, Dr. Norris S. Nahman, was the section chief of the Pulse and Time Domain Section in which Dr. Scavennec performed his research. Dr. Nahman provided scientific counseling and direction of this research effort. He is not longer with NBS. He is presently the chairman of the Electrical Engineering Department of the University of Toledo, Toledo, Ohio.

James R. Andrews

ACTIVE AND PASSIVE MODE LOCKING
OF CONTINUOUSLY OPERATING RHODAMINE
6G Dye Lasers

André Scavennecc* and N.S. Nahman**

ABSTRACT

Using confined and unconfined fluid flow dye cells with suitable mode locking methods continuously operating Rhodamine 6G dye lasers have been built to produce narrow pulses (less than 40 ps) at about a 140 MHz pulse repetition rate. For active (acoustic) amplitude modulation mode locking methods optical pulses were obtained having about a 32 ps pulse width (FWHM). For passive mode locking methods optical pulses of $\leq$ 35 ps pulse width (FWHM) were obtained.

A three mirror cavity was used for the active mode locking studies while three and five mirror cavities were used in the passive studies. The three mirror passive mode locked laser employed a single dye cell containing both the Rhodamine 6G (gain) and DODCI (loss) solutes in a single solution having glycol as the solvent. The active and passive mode locked carrier wavelengths were about 5900 Å and 5800 Å, respectively.

Keywords: DODCI, dye laser; laser; mode-lock; picosecond; Rhodamine 6G.

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1. INTRODUCTION

The mode locking of the continuously operating (CW) rhodamine 6G laser was recently reported with production of extremely short optical pulses: about 50 ps duration (F.W.H.M.) using active mode locking (acoustooptic amplitude modulator) [1], and a few ps (∿ 2) using passive mode locking (with Diethyloxadicarbocyanine Iodide or DODCI as the saturable absorber) [2].*

One of the possible uses of so short optical pulses is in the field of fast electronic metrology. Optical pulses can be used either to drive a gate or to generate electronic pulses. Optical pulses with width in the picosecond range, with a repetition rate high enough to allow averaging or sampling techniques,** would allow a substantial improvement in the field of the ultrafast electronic measurement.

The production of the picosecond pulses is of course dependent on the achievement of very fast optoelectronic transducers, for which one requires a fast response rather than a good sensitivity, which is somehow the opposite of what optical communications require.

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*Other techniques than mode locking may be used to produce short optical pulses: for instance 21 ps duration optical pulses were obtained with a He-Ne laser for which a Fabry-Perot electrooptic modulator was set in place of one of the cavity mirrors [3].

**Short (∿ 7 ps) and quite large (> 100 V) optical pulses were obtained through optical rectification of powerful pulses (.5 GW) emitted by a mode locked Nd doped glass laser [4]. Unfortunately the repetition rate of such a laser is much too low to allow the use of sampling techniques.

This report only deals with the production of narrow optical pulses; we report on the work done on the two techniques used to produce mode locking of the Rh 6G laser. The obtained results are then compared with results from other laboratories.

2. THE CW Rh 6G LASER [5]

2.1 The Active Medium

The active medium is a solution of Rh 6G in a liquid solvent.

The population inversion is obtained through optical pumping of the Rh 6G molecules and can be described by taking into account four electronic vibrationally broadened levels, two singlets and two triplets (fig. 2-1).

The molecules are excited from the lower level S_0 to the excited level S_1. They relax very rapidly to the thermal distribution in S_1 and become available for amplification. The relaxation of molecules to the thermal distribution in S_0 is also a very fast process (10^{-11} sec). As a result, one may consider the dye laser as a four level system though only two levels (S_0 and S_1) are involved. The triplet states act as trapping levels. Actually, the excited molecules in S_1 can decay through spontaneous or stimulated emission down to to S_0, or through non radiative process down to T_0. The CW operation of the Rh 6G laser was made possible only when a quencher was found for the triplet level [5] thus limiting the lifetime of the molecules trapped in T_0. The lifetimes of the different transitions for a CW Rh 6G laser are approximately [6][7]:

$$\tau_S \sim 5.5 \text{ ns}$$

$$\tau_{ST} \sim 35 \text{ ns}$$

$$\tau_T \sim 100 \text{ ns}$$

The loss mechanisms limiting the gain of the laser are the fluorescence loss (spontaneous emission), the triplet trapping, the $T_0 \rightarrow T_1$ absorption and the self absorption resulting from the overlap of the absorption and emission bands. It is worth noting that the last process makes new molecules available for the population inversion.

In order to reach the threshold for gain and lasing, it has been shown that, using the argon ion laser (5145 Å) as the pump, power density greater than 10 kW/cm^2 is required. This can be obtained by focusing the pump laser beam down to a beam waist of approximately 10-30 μm in diameter. On the other hand, losses are reduced by decreasing the length of the active medium [8]. As a result, the active medium is usually confined in a small cylinder (20 μm in diameter, 1 mm or less in length).

To prevent thermal distortion and dye degradation the solution is flowed through the active region; the speed of the flow is between 1 to 20 m/s leading to a residence time of 20 to 1 μsec for the dye molecules in the active region. In order to minimize thermal distortion, a solvent with a low thermal expansion coefficient is required.

2.2 The Laser Cavity

The dye laser and pump beams are usually collinear and their widths are matched. This leads to a maximum gain for the laser. However, for very thin active medium, the collinearity is no longer required and a small angle is allowed.

In order to have a cavity long enough to insert a mode locking device and a beam waist about 20 μm diameter in the active medium, either an internal lens or a third mirror is required. The cavity we used, similar to the one described in [9], is of the three mirror type (fig. 2-2).

In this cavity, the mirror M_2 curvature is chosen to focus the pump beam to the desired size in the active medium. The recollection mirror M_1 curvature is chosen considering the beam waist and ease of mechanical setup considerations. The mirror M_3 is a long radius of curvature -- usually flat -- mirror. The large breadth of the emission band characteristic of the dye lasers allows the broad frequency tuning of the Rh 6G laser. The tuning is done through a dispersive element, the Brewster angle prism P. The prism also allows the pump beam to be injected into the cavity without going through one of the cavity mirrors.

The angle 2θ between the two arms of the cavity is determined to minimize the astigmatism of the cavity [10]; the astigmatism resulting from the Brewster angle dye cell is compensated by the astigmatism from the mirror M_2. For usual solvents and cell thicknesses and for a curvature of M_2 equal to 10 cm, 2θ is of the order of $10°$-$15°$.

2.3 The Dye Cell

The dye is conventionally flowed through a cell, two windows with the desired spacing confining the dye solution, while allowing the pump and the dye laser beams, crossing the cell at the Brewster angle, to interact with the active medium.

Recent development in dye laser technology showed that, by the choice of a viscous solvent, it was possible to use an unconfined film of dye solution as the active medium [1]. The solution is flowed through a nozzle designed so as to give a liquid film of good optical quality.

In the course of our work, we used different cells of the two kinds, i.e., the confined and unconfined.

2.3.1 The Confined Type Cell

This cell is similar to the one designed by D.A. Jennings in the NBS Quantum Electronics Division (fig. 2-3). The only difference was in the choice of inexpensive microscope cover glasses as windows instead of polished quartz. Actually the beam sizes are so small, that even conventional microscope cover glass has sufficient flatness and optical quality to be used inside the laser cavity [12]. However it is worth noting that the poor thermal conduction of glass often results in thermal problems and instabilities [5]. With that type of cell, we used a 4 x 10^{-4} molar solution of Rh 6G in water with 4% Ammonyx LO. The flow speed in the active region was approximately 3 m/s.

2.3.2 The Unconfined Type Cell

The instabilities we encountered resulting from the poor thermal conduction of glass led us to develop cells of unconfined type which are free of such defects.

Moreover using an unconfined type cell avoids window degradation (deposition of burnt particules by the pump beam) which is a persistent problem of confined type cells.

Most of the unconfined type cells we designed used a nozzle made of a copper tubing on which two razor blades were stuck (fig. 2-3), the slit dimensions were approximately 6 x .5 mm. The liquid flowed through that nozzle must have a high enough viscosity (> 10 cp). We used ethylene glycol (viscosity 20 cp at 20°C) and molar concentration of approximately 5 x 10^{-4} with a flow speed of 2 m/s. The free flowing glycol film was estimated to be .2 mm thick in the active region.

Other attempts were made to design a pinched copper tubing as the nozzle [11]. However the poor mechanical quality of the edges resulted in a poor optical quality of the film.

2.3.3 The Circulating Loop

The dye solution is flowed through the cell by means of a magnetically driven pump. A filter is set in line to remove small particules and bubbles.

For the confined type cell using water as the solvent, the filter has pore size of 1.2 μm. With unconfined type cells using high viscosity glycol, it is necessary to have filters with larger pore sizes 7 μm. Since burnt particules are no longer a problem, even larger pore filters might be used (say 15 μm).

The film produced by unconfined type cells is very sensitive to pump and other mechanical instabilities. We found that damping the flow pulsations and the pump vibrations was necessary for an acceptable stability of the film. This was achieved through the use of corrugated tubing for vibration isolation and of a damping box inserted between the pump and the filter for the flow pulsations (fig. 2-4).

It was also necessary to use a cooling reservoir to remove the laser heat from the glycol. This was necessary to retain the required high viscosity.

2.4 Characteristics of the Laser

Most of the laser characteristics are related to the confined cell laser.

1. The laser emission bandwidth goes from 5650 Å up to 6200 Å for .5 W of pump power at 5145 Å. No saturation of the output/input power characteristic could be seen up to 1.5 W of pump power, which means that the flow speed is high enough.*
2. The cavity losses (diffraction, scattering ...) were checked and found to be quite small: using three maximum reflectance mirrors, the lasing threshold was only 30 mW (5145 Å pump power). The threshold with the free flowing film laser was a little bit higher (∿ 50 mW).

*This measurement was performed with three maximum reflectance mirrors and the output was the reflected power from one of the prism faces. In usual operation, a 4-5% transmission mirror was used as the output mirror (M_1).

3. At high pump power (> 1 W) a better stability was achieved with the free flowing film laser. However, the use of quartz windows and possibly cooling of the solution [5] would improve greatly the stability of the laser with the confined cell.

4. The laser output stability is a function of the cell and flow stability. It is also dependent on the pump laser stability and on a good optical isolation between the pump laser and the dye laser, in order to prevent feedback and etalon effects.

5. The pump laser noise was < 0.5% peak to peak (10 Hz - 2 MHz) and good isolation was provided by the high absorption of the dye solution. The concentration was adjusted to give a 90-95% absorption per pass, thus limiting the power reflected toward the Ar laser to less than 1%.

6. In the best case, the noise of the dye laser output was observed with a fast detector and a 100 MHz bandwidth to reduce to 8% peak to peak. When the laser is close to the threshold, the output power is more sensitive to the pump laser noise and becomes noisier.

3. MODE LOCKING IN A LASER

We now give a few characteristics of mode locking in a laser. This had been the subject of extensive research, and analysis in both time and frequency domain were published [13].

3.1 General Considerations

Only the laser modes associated with TEM_{ooq} modes of the laser cavity are considered. In the frequency domain, these modes are almost equally spaced. The intermode frequency is c/2L, the fundamental frequency of the laser cavity (∿ 100 MHz). The only modes able to oscillate are the ones for which the laser gain (the gain linewidth is usually 100 MHz $< \Delta\nu_L <$ 100 GHz) is greater than the losses (fig. 2-1).

The modes are oscillating with random phase and amplitudes with a weak intermode coupling. The output signal, characterized by its electric field can be written (fig. 3-1d):

$$E(t) = \sum_n E_n(t)\, e^{j(\omega_n t + \phi_n)}$$

$E_n(t)$: amplitude of the n^{th} mode

ω_n, ϕ_n: frequency and phase of the n^{th} mode

The intermode coupling resulting from the active medium nonlinearities can induce self mode locking (as for the He-Ne laser at 6328 Å). In such a case, the modes are oscillating in phase and with a steady, approximately gaussian amplitude distribution.

$$\omega_n = \omega_{n-1} + \Omega$$

$$\phi_n = \phi_{n-1} = 0$$

$$E_n(t) = E_o(t) \cdot e^{-kn^2}$$ $E_o(t)$ may have small variations with time

The output signal is then:

$$E(t) = E_o\, e^{j\omega_o t} \sum_{n=-p}^{+p} e^{-2n^2 + jn\Omega t}$$

To this signal is associated an optical pulse train with $T = \frac{2L}{c}$ as the repetition period and $\tau \sim \frac{1}{\Delta\nu_L}$ as the pulse-width (fig. 3-1e).

With most lasers, it is possible to produce a similar mode synchronization, by adding nonlinearities inside the laser cavity. One can use either an active modulator varying the phases or amplitudes of modes at a frequency equal to the intermode frequency or a "passive" element such as a saturable absorber.

3.2 Frequency Dispersion of Modes

If the intermode frequencies were equal all over the oscillation width (as are the empty cavity intermode frequencies) the nonlinearities of the laser active medium would be sufficient by themselves to induce mode locking. Several dispersion sources actually prevent the modes from being equally spaced:

1. The nonlinear variation of the active medium refractive index around its resonance results in dispersion of the laser modes resonance frequencies (referring to a linear distribution, the dispersion may be of the order of a few kHz or MHz). This dispersion is quite large for inhomogeneously broadened laser lines (gas lasers); however it is negligible for homogeneously broadened laser lines such as dye lasers [14].
2. Internal optical elements often act as etalons. The well known property of etalons is to modulate the amplitude of the cavity loss as a function of the wavelength. However one must keep in mind that these etalons are also frequency dispersive and can prevent mode locking [15]. It is then necessary to take extra care to get rid of such etalons, either by tilting optical elements or by good quality anti-reflection coatings or, more efficiently, by setting elements at the Brewster angle.

3.3 Rh 6G Laser Mode Locking

The active medium nonlinearities are not large enough to induce self mode locking. However the observation of the dispersion of modes for the laser we built shows that relatively small nonlinearities are required to induce mode locking.

The RF spectrum analyzer observation of beat notes widths, somewhat related to the dispersion [16], gives values around 60-100 kHz. Such values are quite similar to what is observed for the He-Ne 6328 Å laser (40-100 kHz), and much smaller than for the Ar Ion laser (300 kHz - 1.5 MHz).

3.4 Note on the Homogeneous Nature of the Rh 6G Active Medium

The homogeneous line of the Rh 6G dye laser is usually considered to have a width of several Å ($\sim$ 5 Å). This width is larger than the oscillation width of the laser, resulting from the dispersive nature of the cavity (prism). As a result, the Rh 6G laser has essentially a homogeneously broadened active medium. However due to the inhomogeneous spatial saturation of the active medium by modes at different frequencies, there is some inhomogeneity: two modes, spatially offset by $\pi/2$ in the dye cell will saturate the active medium almost independently. That is why a monomode Rh 6G laser has a tendency to oscillate on two modes, spaced c/4d apart, where d is the distance between the cell and the mirror M_1 [17]. The RF spectrum analyzer observation of the output of our laser also shows intense beat notes between 1.2 and 1.7 GHz (c/4d = 1.5 GHz). The modulation frequencies we used in this study were smaller than c/4d, and no problems were expected coming from the inhomogeneous spatial saturation. However with modulation frequencies in the vicinity of c/4d one may expect to observe parasitic phenomena such as simultaneous oscillation of two independent pulse trains.

4. ACTIVE MODE LOCKING OF THE CW Rh 6G LASER

The laser cavity was similar to the one described in the first part. However in order to have more room to set the modulator, a second prism was added to separate the pump and the dye laser beams (fig. 4-1). One of the prisms was a Brewster angle prism, the second was a 60° prism which provided an additional output for triggering purposes. The prisms were set so as to reduce the dispersion of the cavity.

4.1 The Modulator

The results reported in the literature on CW Rh 6G laser active mode locking show that amplitude modulation [1] leads to better results than phase modulation [18]. This is to be expected from the fact that phase modulation is very poor to prevent the oscillations of several intermingled pulse trains corresponding to different etalons transmission peaks. Moreover, for a same modulation index, amplitude modulation gives narrower pulses than phase modulation [19]. As a result, we used an acoustooptic amplitude modulator.* The modulator is made of a silica block (2 mm thick) in which standing acoustic waves are generated by a rectangular X-cut quartz crystal (23.4 MHz fundamental frequency, driven at 70.36 MHz) (fig. 4-2). The acoustic standing waves

*On loan from C.N.E.T.

modulate the refractive index. The optical beam propagating into the silica interacts with the phase grating and sustains losses at twice the ultrasonic frequency [20]. The optical beam transmitted through such a modulator is:

$$I(t) = I_o J_o^2 (\nu \sin \omega^* t)$$

$I(t)$, I_o: Transmitted and incident beams

J_o: Bessel function

ω^*: Ultrasonic frequency

ν: Index of modulation

The modulator we used was built a few years ago for use in an Ar Ion laser mode locking experiment, and slow degradation of the bonding has reduced its efficiency down to approximately 2% $\left(\frac{I_o - \langle I(t) \rangle}{I_o}\right)$ for 40 mW of absorbed electrical power. This efficiency was still sufficient for the Rh 6G laser mode locking.

The modulator is driven by a very stable phase-locked frequency source: in order to reduce the frequency drift resulting from the wide variations of the modulator impedance around resonance. A good electrical isolation is provided between the oscillator and the modulator. Since the modulator is very temperature sensitive it is necessary to monitor the VSWR as a check on the modulator tuning (fig. 4-3).

4.2 The Detection Apparatus

1. Electronic Detection: The output signal was detected with a fast Si photodiode* (transition time 10%-90% $\sim$ 61 ps). A slower (transition time $\sim$ 120 ps) commercial avalanche photodiode is used for triggering. The avalanche photodiode was used in conjuction with a RF spectrum analyzer to align the cavity fundamental frequency and the modulation frequency, and also for the first settings (modulator adjustment for maximum efficiency).

   When good mode locking was evidenced by the RF spectrum, the fast Si photodiode output was directed onto a 28 ps sampler and a display oscilloscope. Careful adjustments of the trigger signal level and trigger stability were necessary to synchronize the oscilloscope to the pulses.

2. Optical Detection: Since the width of the optical pulses is approximately equal to the photodiode transition time, a large uncertainty on the actual pulse width results from the electronic measurement. In order to have an accurate measurement of the optical pulse width, an optical autocorrelation measurement, similar to the one reported in [1] was performed, using Second Harmonic Generation (SHG)

*On loan from C.N.E.T.

in KDP (fig. 4-4). Such a measurement is similar to the well known Two Photon Fluorescence (TPF) measurement performed to measure pulses from flash-lamp pumped lasers. The measurements were done around 5900 Å, which is the wavelength for which the SHG crystal was cut, (no pulse width dependence with frequency is expected). Considerable care was given to the alignment of the reflecting prisms in order to avoid false results which might result from non-collinear recombined beams.

4.3 Results of Active Mode Locking Experiments

Most of the experiments with active mode locking were conducted with the free flowing film type of laser and with a 4% transmission mirror as M_1. Insertion of the modulator inside the cavity raised the threshold up to 500 mW.* In usual operation, the pump power was set to 1.0-1.2 W depending on the nozzle and solution used. With the modulator operating, the output goes down to 10-12 mW average power approximately, from 50 mW without modulation. The use of a smaller modulator very close to M_3 would probably improve that figure.

---

*One may note that increasing the losses of the cavity increases the population in the excited singlet state and thus increases the active medium loss due to the triplet states.

Single transverse mode operation was obtained by introduction of a small ($\sim$ 2 mm diameter) iris into the cavity. Good mode locking resulted from careful alignment of the cavity and modulation frequencies, and a good stability of the output pulses was obtained through dye cell adjustments. The different waveforms we observed as we detuned the cavity frequency were quite similar to what is observed with an Ar Ion laser [16]. The short fluorescence lifetime characteristic of these two lasers might be the reason of those observations [21]. Only for the optimum frequency and for a high enough modulation index did we observe pulsing with a single pulse per modulation period; detuning in either direction resulted in multiple pulsing, usually with broader pulses.

With a 28 ps transition time sampler, the pulses we observed with the sampling oscilloscope had a width around 70 ps (F.W.H.M.), and a risetime (10%-90%) of 50 ps, which are consistent with a gaussian pulse (fig. 4-5).

The optically detected pulse widths (F.W.H.M.) were measured to be 32±2 ps, assuming a gaussian optical pulse.

On the figure 4-6 is a recording of the signal detected by the photomultiplier (pulse autocorrelation function). Because the translation length for the recording system was limited to 10 mm, we scanned only part of the curve. However we first checked by manual translation for symmetry of the curve. The contrast ratios don't exactly agree with theoretical predictions (3:2:1 for the central spike, the broader spike, and the flat background): this is attributed to slightly different beam sizes coming from the two interferometer arms. The sharp central spike is related to the coherence of the laser (or the width of the oscillating spectrum) the broader spike is related to the actual pulse width and the background to the sum of the SHG signals resulting from each interferometer arm signal.

From the two measurements, we were able to determine a transition time (10%-90%) of 61 ps for the fast photodiode.

Larger pulse widths were reported by Dienes et al. [1], with not so large a pulse width dependence on the modulation frequency as predicted for homogeneously broadened lasers [19]. Their measured values would give, for the modulation frequency we used ($\sim$ 140 MHz), a pulse width of 62 ps. It is not clear to us why we obtained shorter pulses than theirs, though the expected value of the mode locked Rh 6G laser pulse width according to the theory developed in [19] is still much shorter (a few picoseconds).

The optical measurement of the pulse widths was performed when the laser was operating with a poor optical quality dye film and other experiments with better dye films (thinner ?) might give even better results.

However, one should note that the theory developed in [19] applies to the case of an homogeneously broadened active medium with a fluorescence lifetime much larger than $2L/_c$, which is not at all the situation for the Rh 6G laser. Non-linear amplification, not included in theory [19], is thought to be of major importance for the dye laser and is probably responsible for the larger pulse width.

5. PASSIVE MODE LOCKING OF THE CW Rh 6G LASER

A few years ago, passive mode locking of the flashlamp pumped Rh 6G dye laser [22] gave pulses with a few picoseconds widths. The same absorber, diethyloxadicarbocyanine Iodide (DODCI) was used to produce mode locking of the CW laser [2][23].

In order to drive the absorber into its nonlinear region, focusing of the CW Rh 6G laser beam in the absorber is required to obtain a high enough power density. The cavity of a mode locked CW Rh 6G laser has then to be designed so as to produce small beam diameters in both the amplifying and the absorbing medium.

This was done by Ippen, et al. [2] through the use of a five mirror cavity which is an extension of the three mirror cavity [9]. O'Neill [23] used an inline type of cavity with lenses to focus the beam. The five mirror cavity seems to give better results, so we decided to try that type of configuration.

5.1 The Five Mirror Cavity Passively Mode Locked Laser

The confined type cell was used for the amplifying medium (Rh 6G in water + Ammonyx LO) and the absorber (DODCI in glycol) was flowed through a free flowing type cell. The mirrors had the same curvature and reflectivity as the ones used in [2], leading to beam diameter of 25 μm in the Rh 6G solution and 15 μm in the DODCI solution. The cavity length (1.60 m) was a bit shorter than in [2] (fig. 5-1a).

We first had the three mirror cavity Rh 6G laser lasing. The mirror M_3 was the tilted so as to direct the flurescence pattern onto the mirror M_4, and the five mirror cavity was adjusted for lasing with the glycol film in the beam. DODCI was then carefully added to the glycol. Most of the time, extinction of the lasing action resulted and it was necessary to readjust the laser cavity to obtain lasing again. This probably results from the increase in absorption of the glycol film and associated change in thermal focusing. It is our belief though that a slow change in DODCI absorption may occur after mixing with glycol (conversion to an isomeric configuration) which would explain that we always had to wait a little bit after adding DODCI before the laser would oscillate again. Degradation of DODCI (bleaching) actually occurred and approximately twice a week, it was necessary to add some DODCI to the absorber solution.

Adjustment of the cavity parameters and of the absorber concentration resulted in mode locking. The photodiode detected pulse widths were 72 ps (F.W.H.M.), as for the pulses produced by active mode locking. However, due to large instabilities of the cells, the observed signal was quite noisy.

Only with smoothing and persistence on the display oscilloscope were we able to observe the detected pulse.* The average output power had to be restricted to a few mW for clean pulsing. Increasing the laser power -- by increasing the pump power -- resulted in multiple pulsing (several pulses superimposed to a DC level per period). Detuning of the absorber cell from its best position led to an increase of the pulse width.

The increase in pulse width seemed to be gradual with detuning and we did not observe any sudden jump to a regime with 300 ps wide pulses as reported in [2]. During this experiment we had not the opportunity to carry on an optical pulse width measurement.

In order to reduce the noise due to thermal instabilities of the confined cell, we decided to try using the free flowing type cell for the amplifying medium (Rh 6G in glycol) and the confined cell for the absorber (DODCI in methanol). The confined cell, no longer exposed to the powerful pump laser, was supposed to be more stable. However we were unable to obtain good mode locking with that arrangement. This may have resulted from the wavelength shifts of Rh 6G and DODCI resonances as a function of the solvent.

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*This was the first experiment we conducted and the stability has since then improved. The free flowing type cell was redesigned and the thermal instabilities of the confined cell reduced by increasing the flow speed. Moreover a better optical isolation was provided for the Ar Ion laser, thus reducing its noise.

5.2 The Three Mirror Cavity Passively Mode Locked Laser

The five mirror arrangement described in the preceding section is very sensitive to mechanical noise and adjustment of the numerous settings is not easy.

In order to simplify alignment of the cavity we decided to fold that five mirror arrangement and to have a single cell through which both the amplifying and the absorbing media are flowed (fig. 5-16).

Both Rh 6G and DODCI may be used with glycol as the solvent, so we chose a free flowing type of cell. The expected problems were:

- -- bleaching and saturation of the DODCI absorption by the Ar Ion pump laser;
- -- reaction between Rh 6G and DODCI modifying the properties of either one.

Actually no noticeable reduction of the mode locking properties of DODCI resulting from saturation of the absorption by the 5145 Å pumping radiation could be seen, nor did we observe a faster bleaching of DODCI than when used in a separate cell with the five mirror cavity configuration.* The properties of either Rh 6G or DODCI didn't seem to be impaired by the presence of each other in the same solution.

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*It seems however that DODCI is bleaching faster in glycol than in methanol. According A. Dienes, it is necessary to change the methanol solution of DODCI only once a month in the arrangement of [2]. In our case, it's necessary to add some DODCI to the solution approximately twice a week.

Good mode locking was obtained with the new arrangement. The alignment of the cavity was as easy as for the simple three mirror laser and this resulted in a much easier mode locked source to work with.

We used molar concentrations of approximately 6×10^{-4} of Rh 6G and 5×10^{-5} of DODCI, though no attempt has been made to determine the best concentrations. Moreover, due to the bleaching of DODCI, we added some from time to time, so the absorber concentration is not really meaningful.

The observed mode locked pulses, detection limited, had a width of 72 ps (F.W.H.M.) for the best settings (fig. 5-2). Detuning the cell resulted in broader, more stable pulses; the decrease of the stability with decreasing pulse width is attributed to the broader oscillating bandwidth which makes slight mechanical or pumping variations more critical. The stability was much better than with the five mirror arrangement. However we still needed smoothing and persistence on the display section. We later found that part of the problem can be attributed to a poor triggering of the oscilloscope. From the photodiode and sampler response times, we can infer an upper limit to the optical pulse width to be around 35 ps. We didn't perform any optical measurement of the pulse width, which we believe to be quite small (< 10 ps).

The average output power was 6-10 mW and mode locking was obtained around 5800 Å with a 100 Å tuning range. In order to avoid multiple pulsing, the cavity length was chosen so as to have a round trip time (∿ 6 ns) of the order of the recovery time of the laser gain. For the same average power, an increase in the cavity length led to the presence of a second pulse or more in the cavity; it was then necessary to reduce the pump power to obtain single pulsing again.

Tuning the laser from the orange toward the yellow resulted in strong CW oscillation, and we did not observe any bistability as observed by Ippen, et al. [2]. However, when the laser was tuned to the red, CW oscillation accompanied by such a bistability was seen. When the lasing action was interrupted, it was possible to initiate lasing again either by tuning back to the orange and then continuously to the red, or by increasing the 5145 Å pump intensity. Such a behavior is rather similar to the hysteresis observed by Lisitsyn and Chebotaev [24] with a He-Ne laser and an intracavity Ne absorbing cell. Additional information on saturation parameters of Rh 6G and DODCI should be helpful in interpreting the bistability phenomena.

We finally found that this three mirror arrangement gave better results (mainly in stability and reproducibility) than the five mirror arrangement tried before and further work on it is expected to result in a very attractive source for the continuous production of picosecond optical pulses.

6. COMPARISON OF THE TWO TECHNIQUES

In this last section we plan to compare the results we obtained with the two mode locking techniques.

A simple way to describe the results is to say that active mode locking leads to a widely tunable (∿ 800 Å or more) source of short optical pulses while passive mode locking gives ultrashort optical pulses on a smaller bandwidth (∿ 100 Å). In table 6-1 are summarized the values of the pulse widths, average output power, and peak power of the pulses. Also shown are the results published by other laboratories.

For active mode locking it is worth noting the small width (32 ps F.W.H.M.) we obtained as compared to the results of reference [1]. Increasing the efficiency of the modulator and decreasing its size would allow operation at higher average power. Since operating at higher frequency leads to shorter pulse widths, we believe that 25 ps F.W.H.M. optical pulses with peak power greater than 10 W may easily be obtained on a broad bandwidth with our laser. Moreover we don't know of any reason why the pulse width could not be reduced to 10 ps or less, and more work in this direction could lead to a large improvement in the pulse widths obtained with active mode locking. However it is worth noting that in active mode locking, the achievement of very narrow pulses and the stability of the pulse train depends on the good matching of the modulation frequency and cavity frequency. The shorter the pulse width, the more critical is this matching. A mismatch of 1 kHz (or cavity length variation of 10 μm) actually results with our laser in a broadening of the pulse. This effect would be much more severe with 10 ps pulses.

For that reason, one may prefer the passive mode locking technique for which no frequency matching is required. The other advantage of this second technique is in the ultra-short pulse widths observed in [2] and [23]. In our case we had not the opportunity to make an accurate measurement of the pulse width, but there is no reason why the pulses should be larger than 10 ps. In the table 6-1, one can see that the pulse peak power obtained with passive mode locking is much higher than with active mode locking, which is another reason to prefer the former technique. The latter however is more likely to allow higher energy per pulse since passive mode locking average output power seems to be limited to 10 W. The bleaching of DODCI also requires changing the absorber solution from time to time, which is not really too much of a problem.

7. CONCLUSION

The mode locking of the CW Rh 6G dye laser has been presented. We have compared the two usual mode locking techniques, active mode locking (with an acoustooptic modulator) and passive mode locking (with DODCI).

We would like to outline two points which give some originality to our work in regard to previous work:

-- The observed pulse width using active mode locking (35 ps F.W.H.M.) shows a real improvement of the pulse width measured by Dienes, et al. [1], though the modulation indices were almost the same. This result shows that it is probably still possible to improve the pulse width with active mode locking.

-- The realization of the laser using a single active medium, solution of Rh 6G and DODCI in glycol, allows the simultaneous production of gain and non-linear absorption. Such a laser, corresponding to a substantial simplification of the laser developed by Ippen, et al. [2], has the great advantage to be very easy to align.*

Some more work should lead to an improvement of the mode locked lasers characteristics:

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*A letter on that point is to be published in the Correspondence section of the IEEE Journal of Quantum Electronics, January 1974.

-- The design of more stable cells should suppress part of the laser noise.

-- A Perot-Fabry optical spectrum analyzer would be helpful to assist in the search for shorter pulses.

-- A smaller modulator (with a deposited mirror on the back surface) working at a higher frequency would increase the capacities (shorter pulses, higher peak power) of the active mode locked laser.

The mode locked Rh 6G laser used in conjunction with ultrafast optoelectronic transducers should make possible the achievement of recurrent electric pulse generations and gates with faster response than presently available devices.

The study of photodetectors is presently carried on at NBS. Among the different types are:

-- Photodiode detectors: The classical vacuum or semiconductor photodiodes are limited by their transit time and junction capacitance. Metallic point contact photodiodes have a better potential for ultrafast detection. These photodiodes are now working in the visible [25]. The main defect seems to be their short lifetime.

-- Optical rectification: The phenomena is very fast. However high optical power densities are required, and optical rectification probably can't be used in conjunction with a CW Rh 6G mode locked laser.

-- Photoconductivity: Photoconductors have not the transit time and junction capacitance limitations of classical photodiodes. The only limitation is with the carrier lifetime, which may be quite small (a few ps for GaAs). Such photoconductors have already been realized [26]. However they are not very sensitive. An improvement of the sensitivity seems necessary (for instance through the addition of Al in order to move the absorption toward the red).

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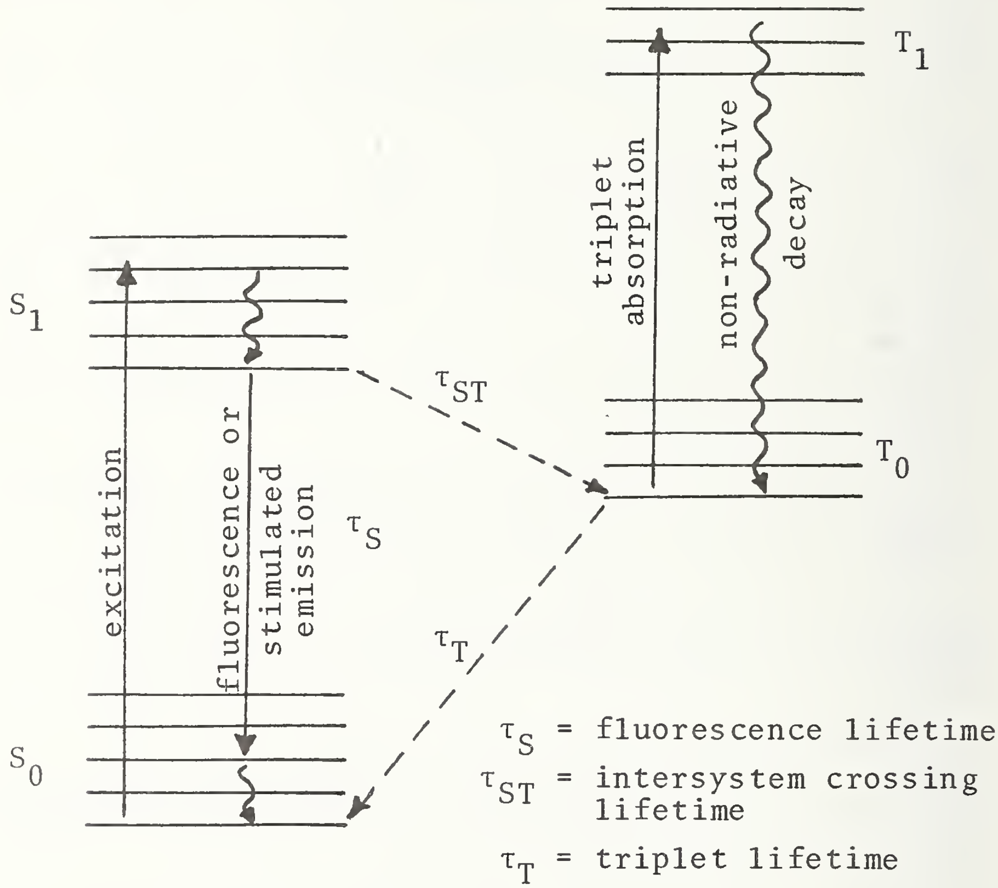


Figure 2-1. Energy level diagram of Rh 6G.

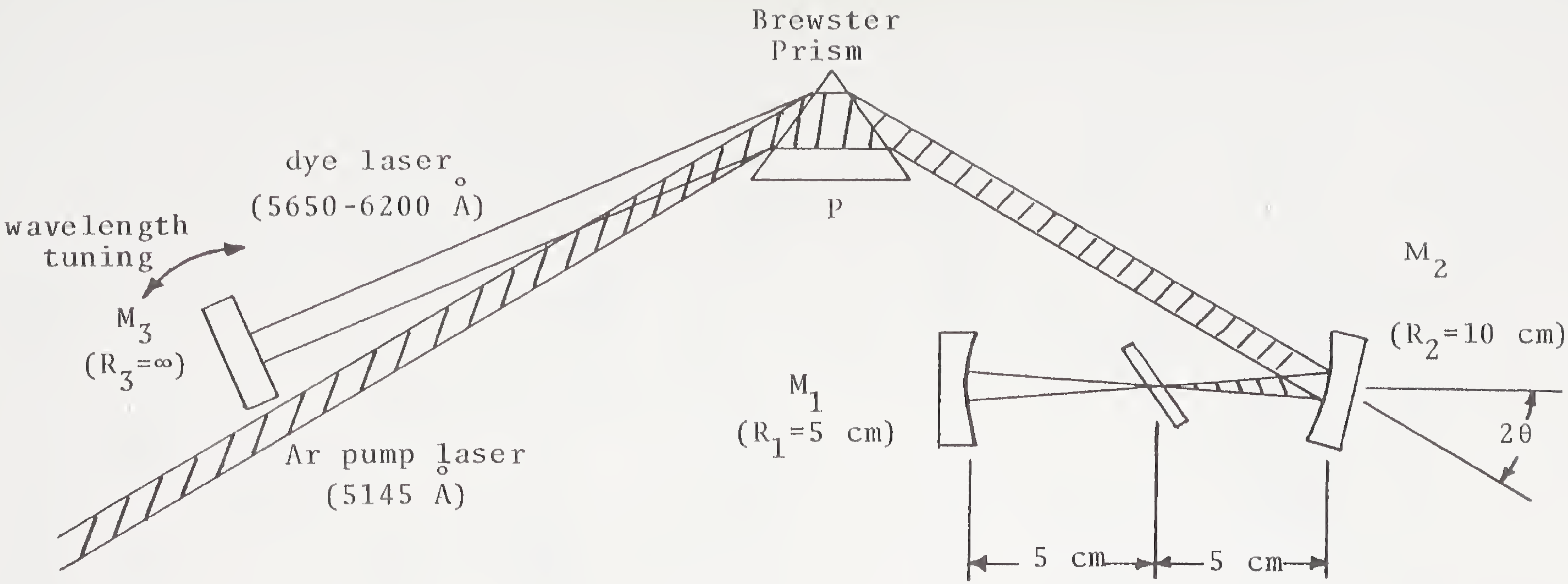
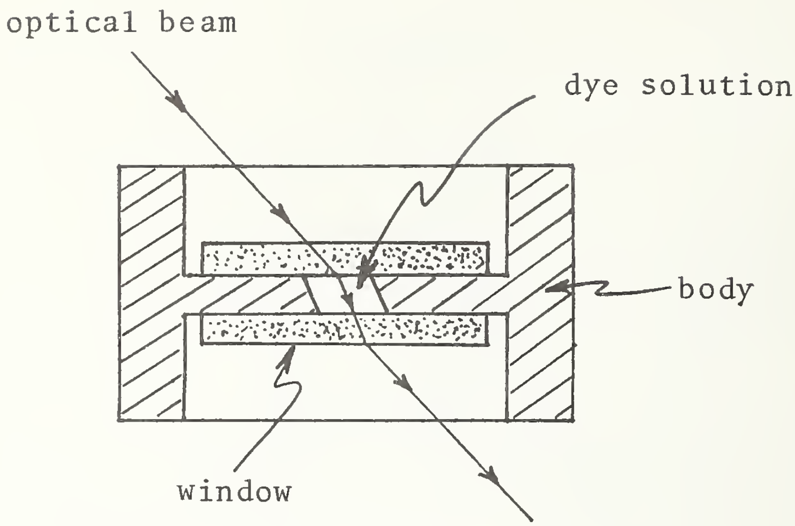
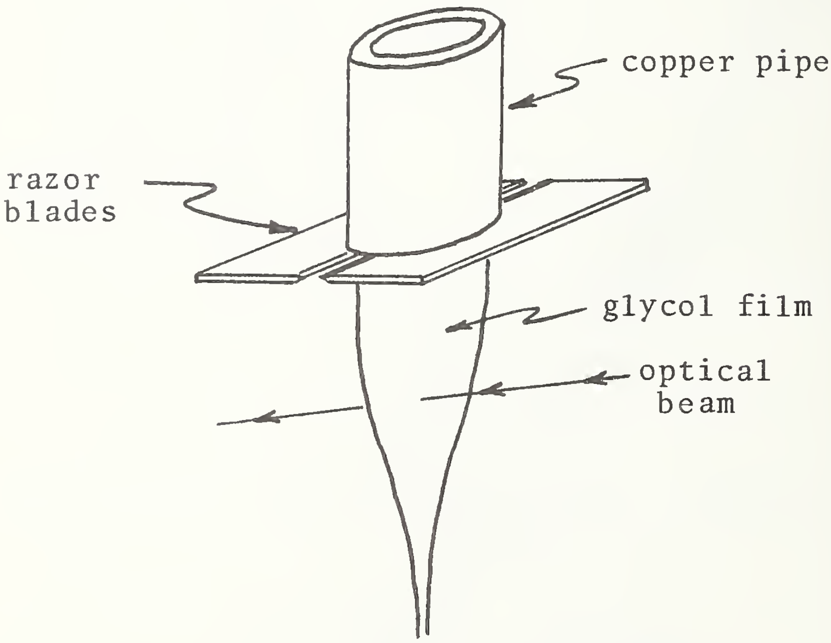


Figure 2-2. CW dye laser.



(a) Schematic view of a confined cell.



(b) Free flowing dye cell.

Figure 2-3. Dye cells.

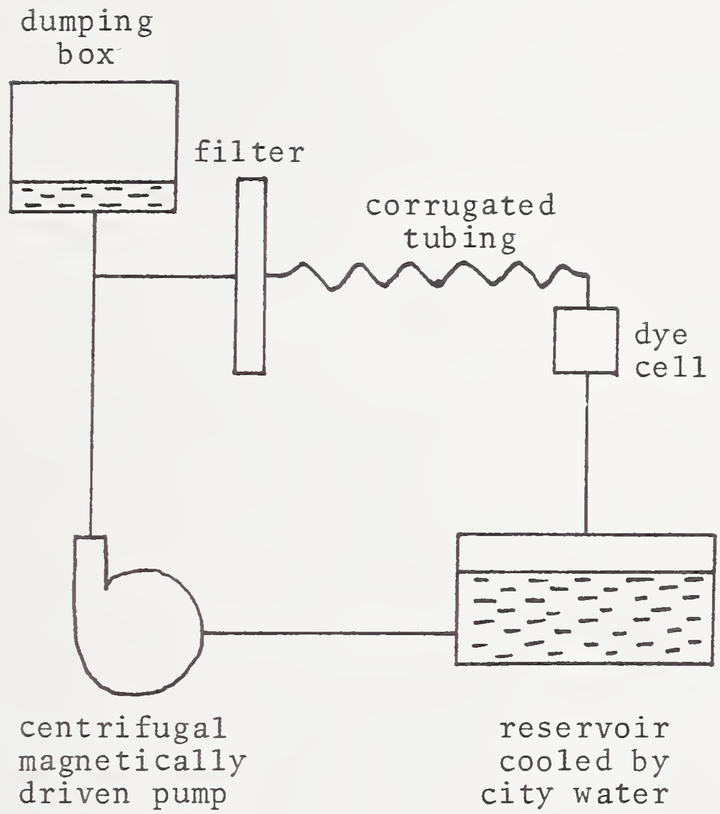


Figure 2-4. Dye circulating system.

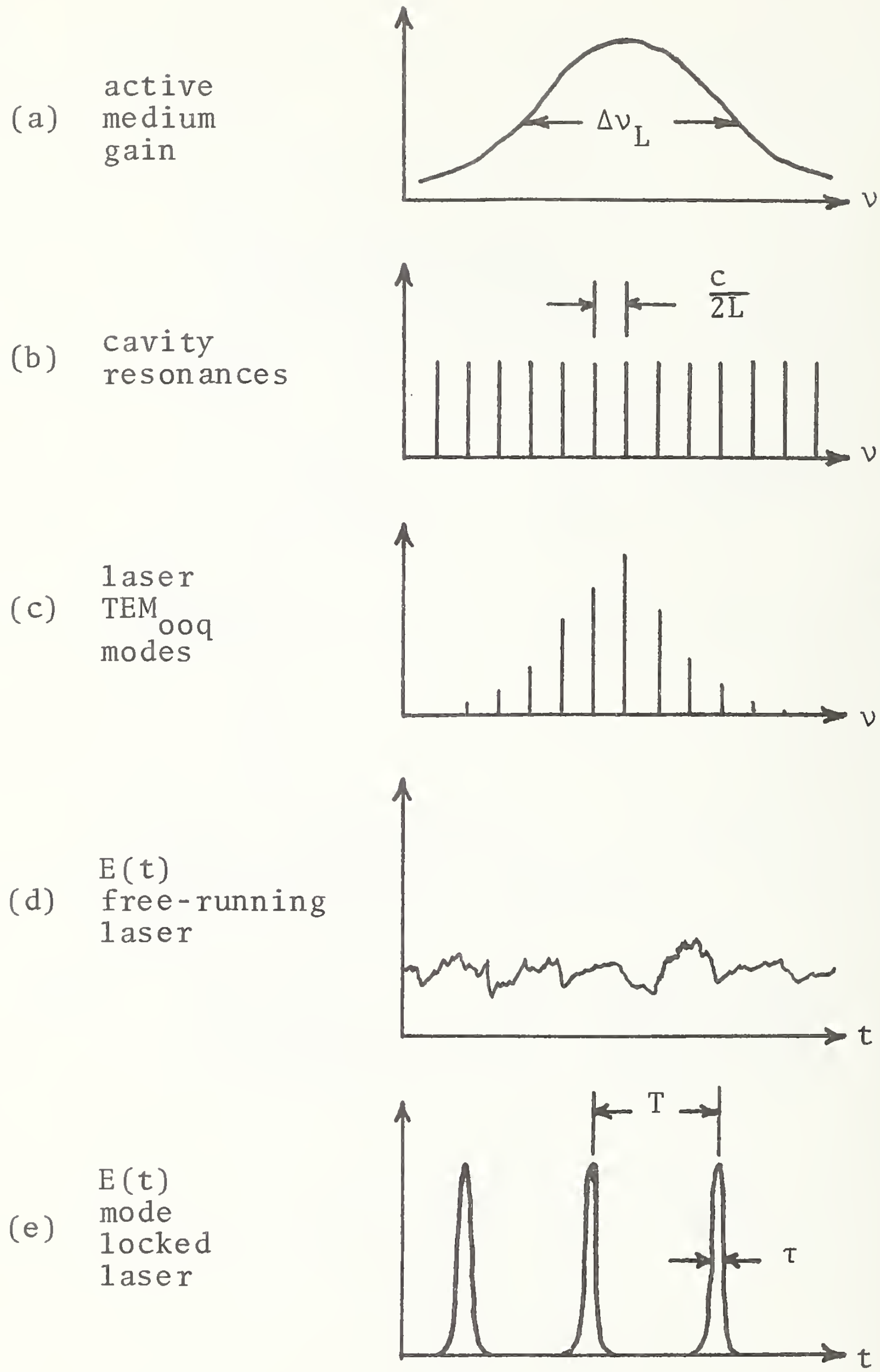


Figure 3-1. Laser modes and mode locked output.

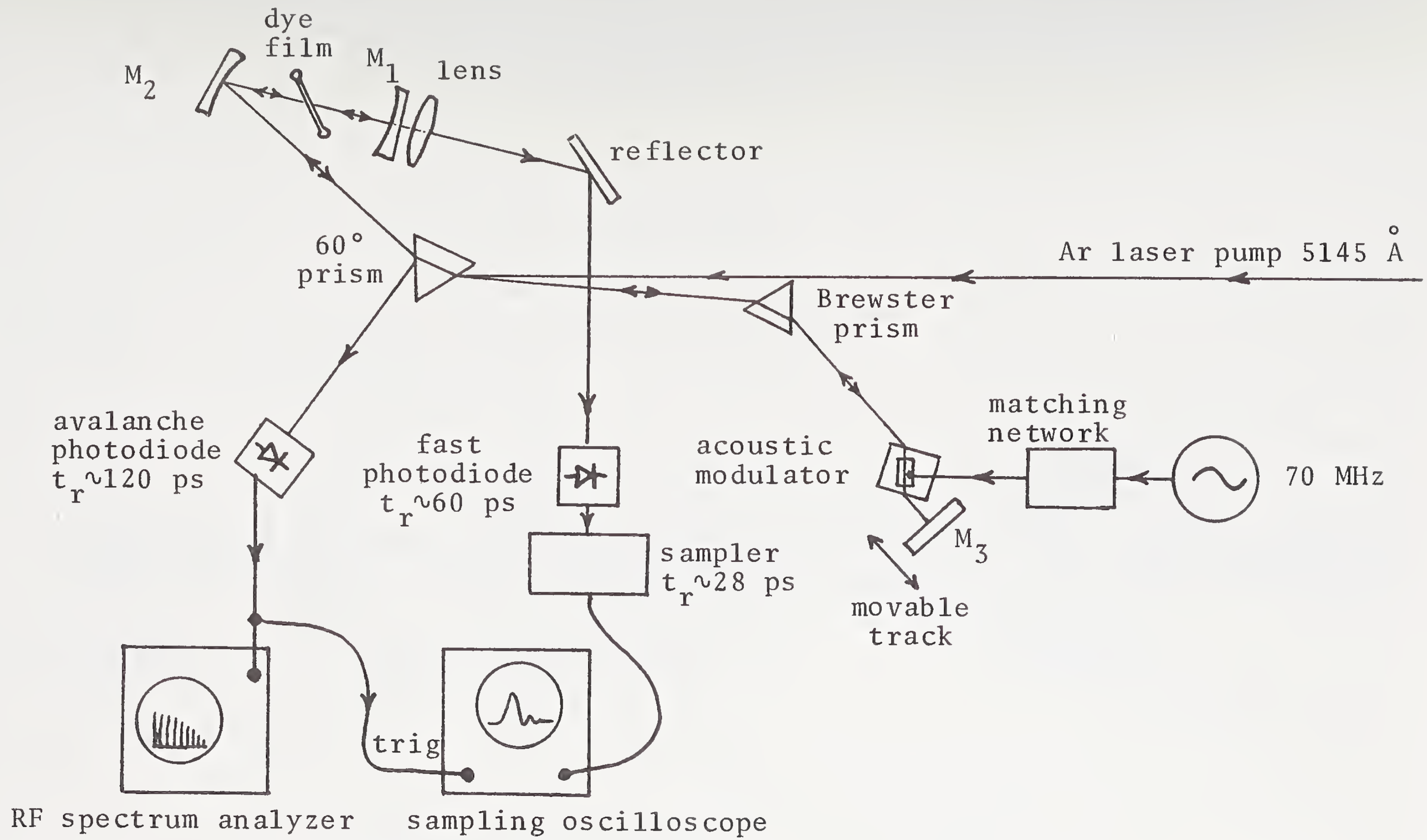


Figure 4-1. Active mode locking, experimental setup.

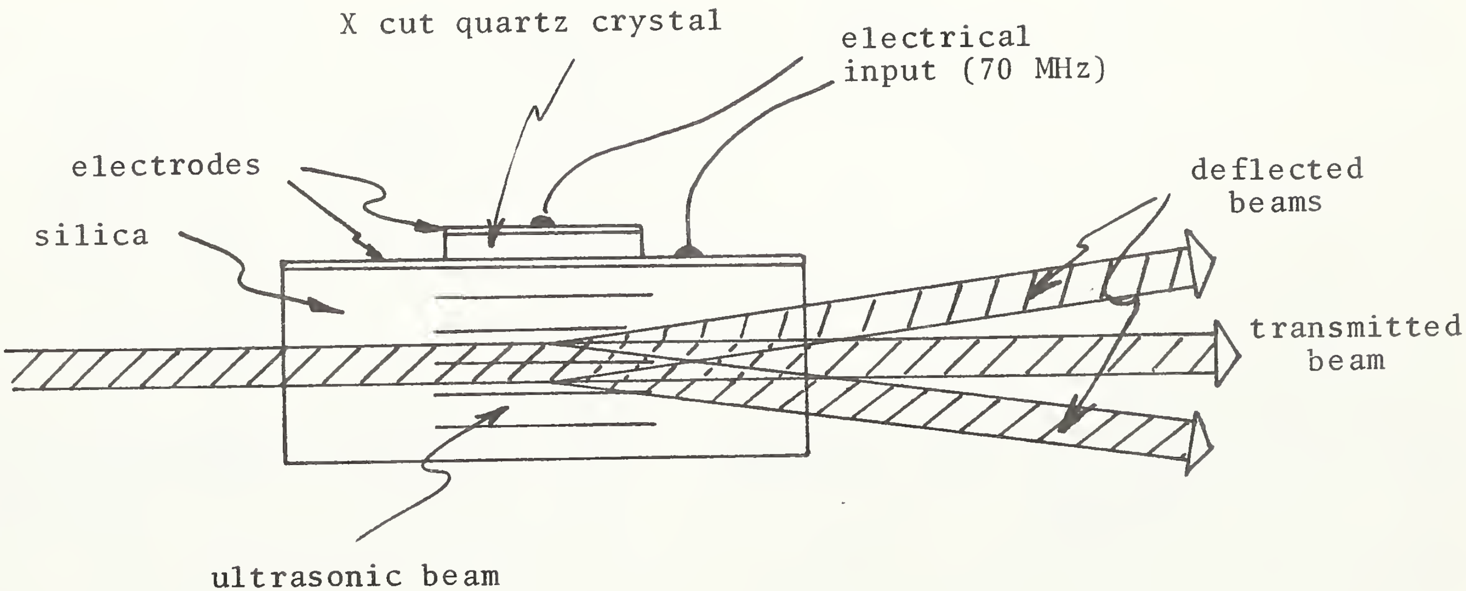


Figure 4-2. Acoustooptic modulator.

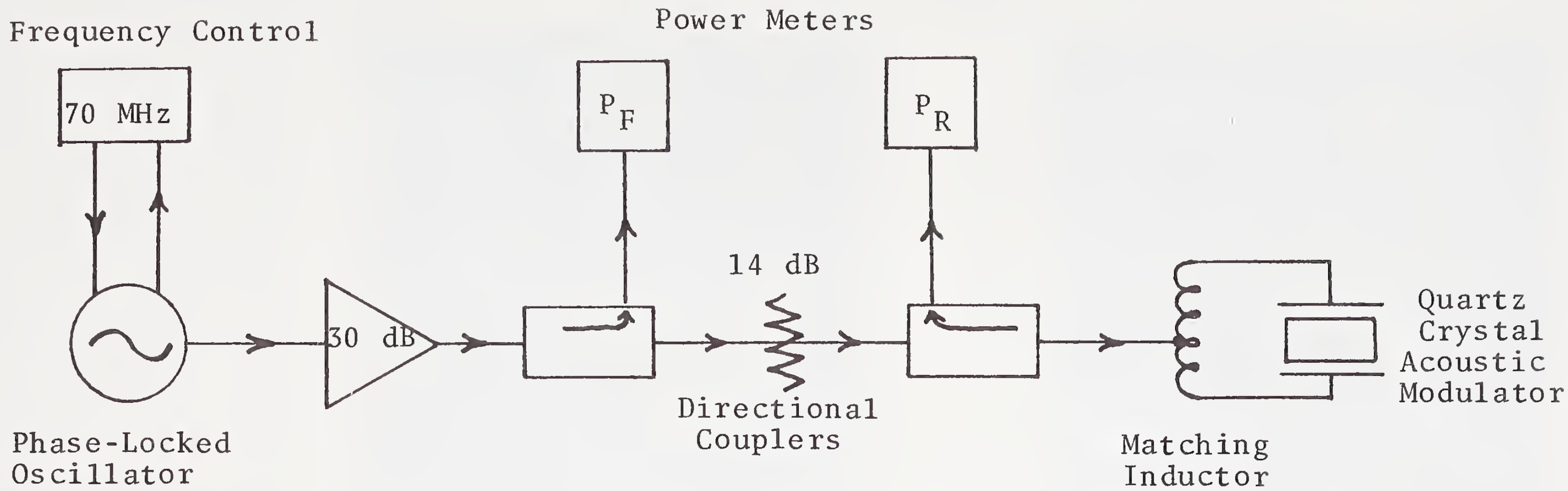


Figure 4-3. Modulator driving circuit.

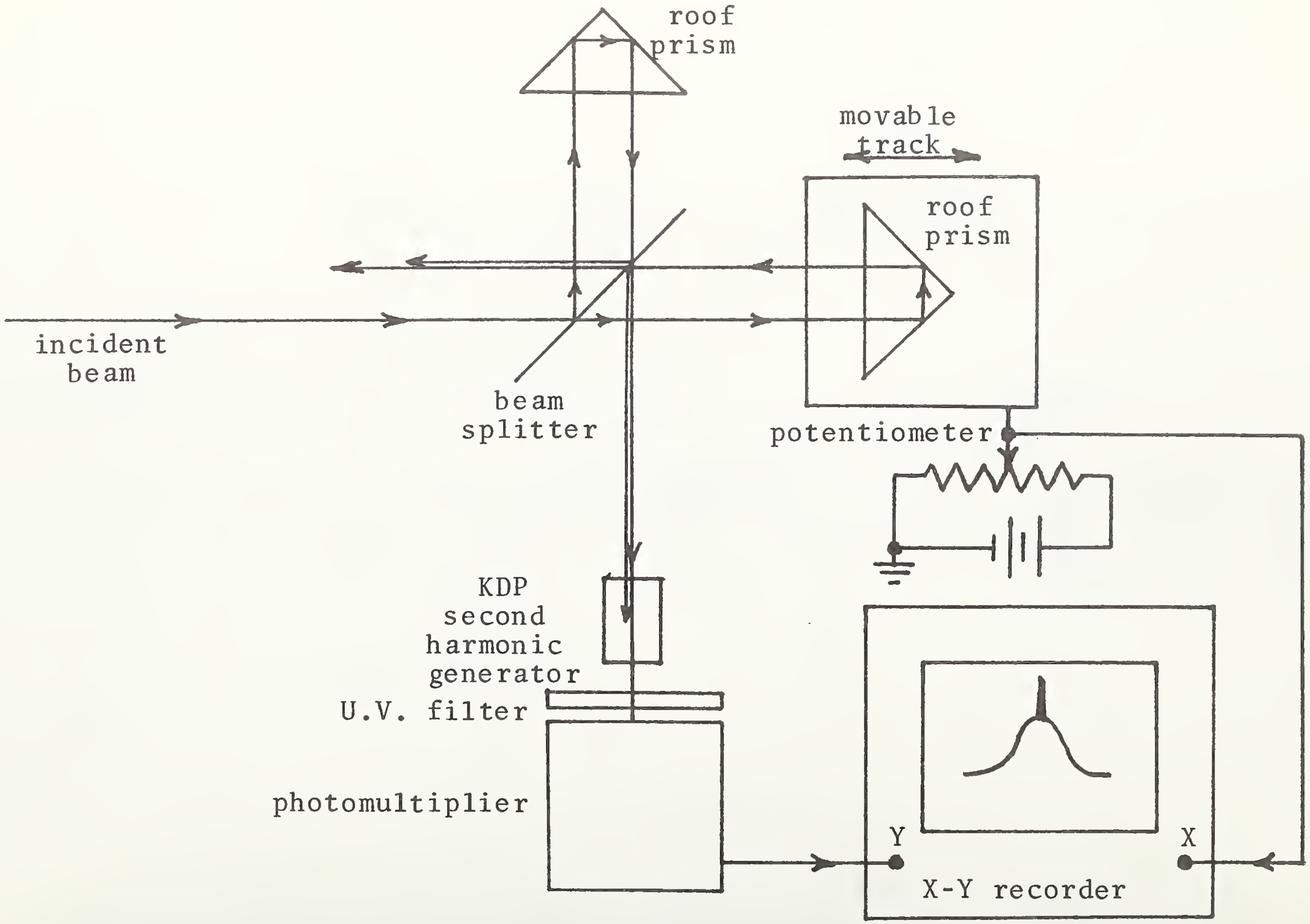
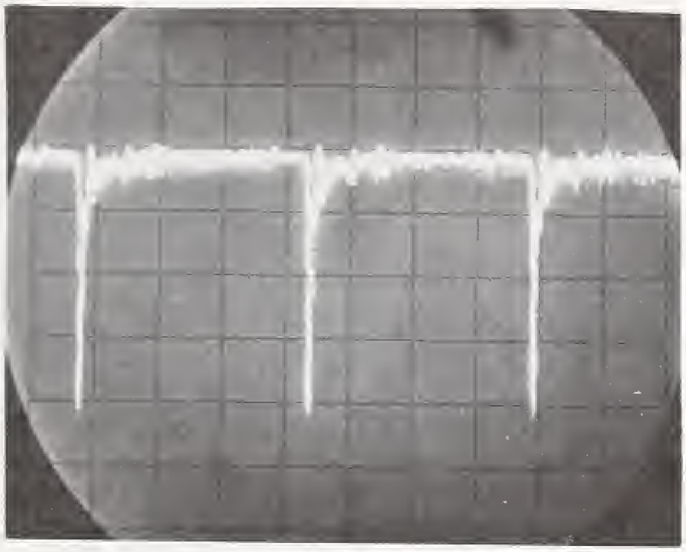
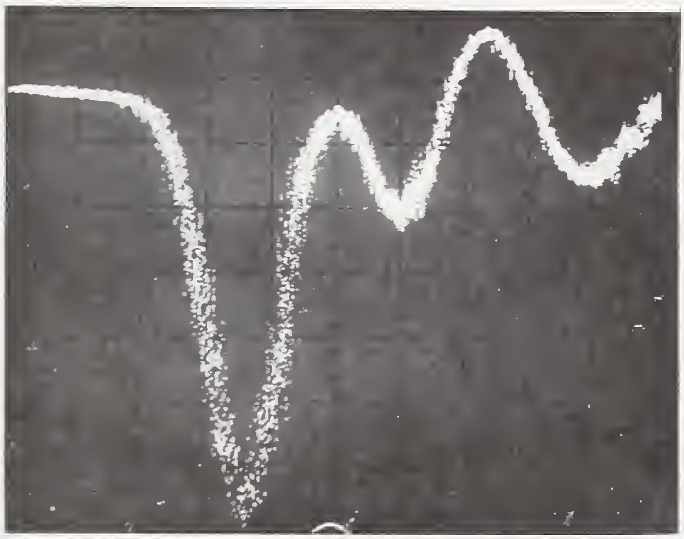


Figure 4-4. Optical autocorrelation pulse width measurement setup.



(a) 2 ns/div., 50 mV/div.



(b) 50 ps/div., 20 mV/div. The secondary pulses come from the detector. The sampler has a 20 ps transition time.

Figure 4-5. Active mode locking, detected pulse waveforms.

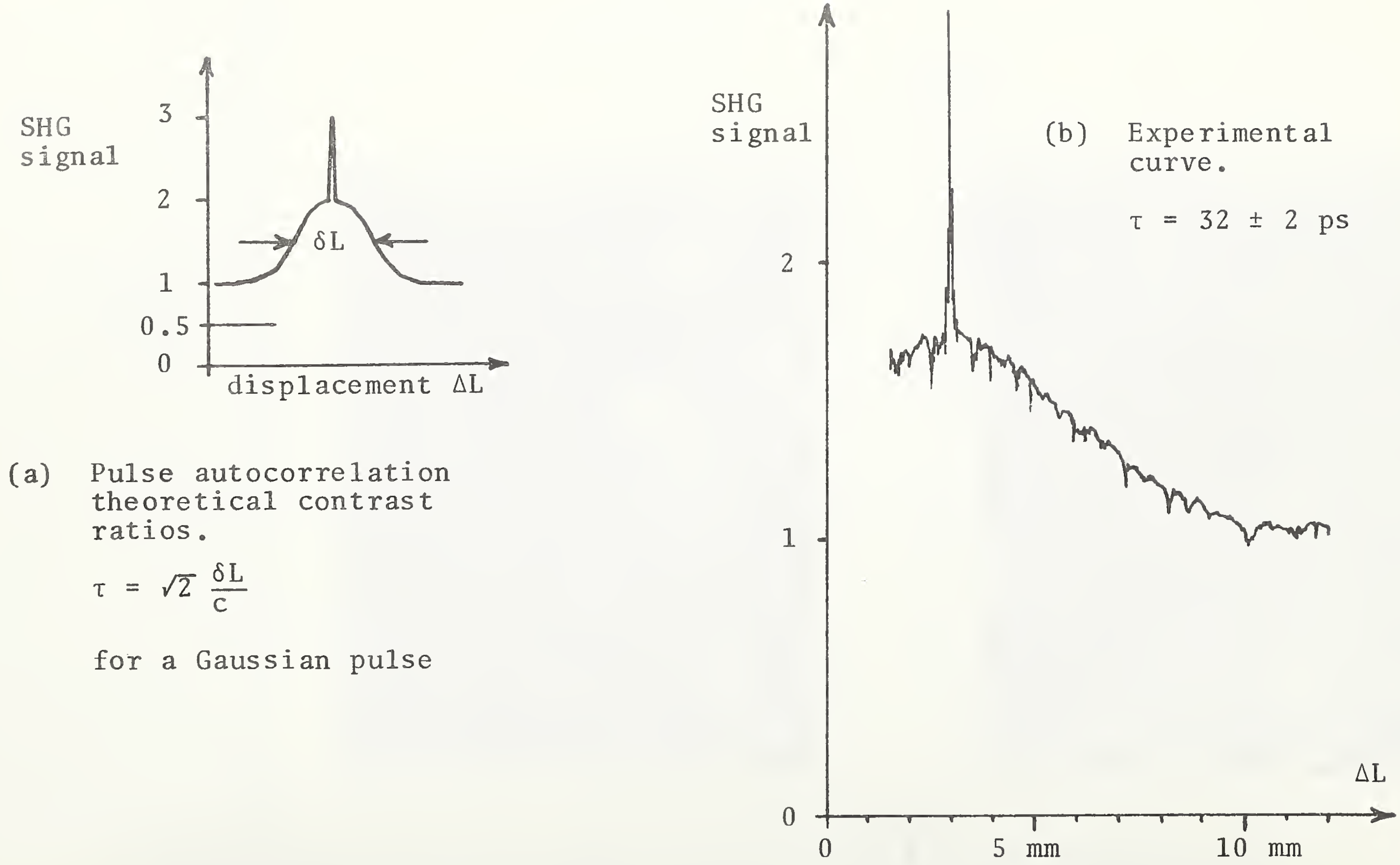
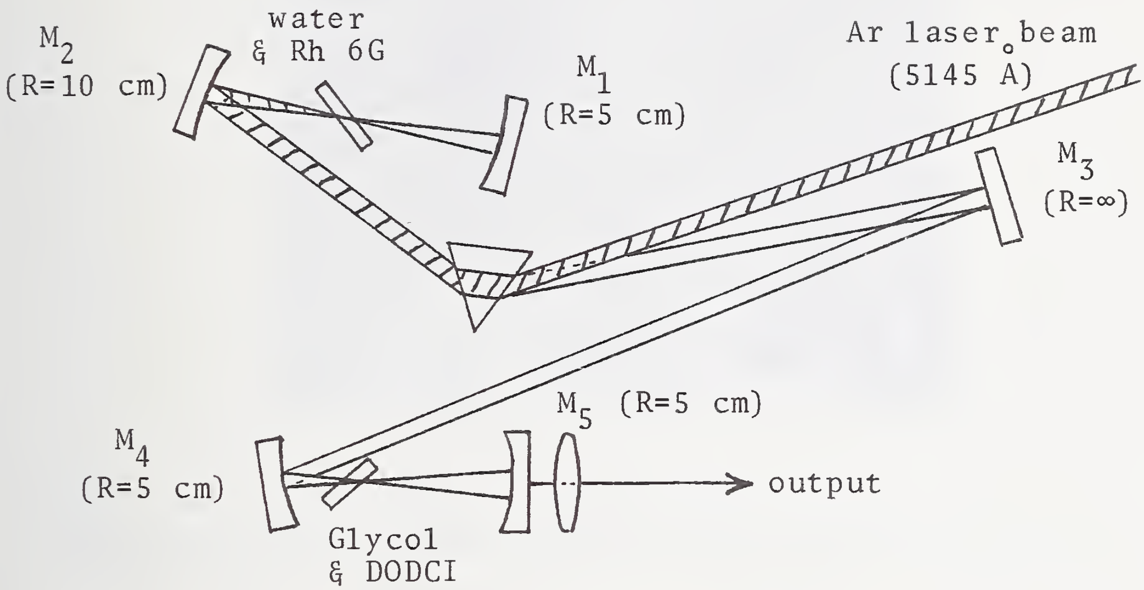
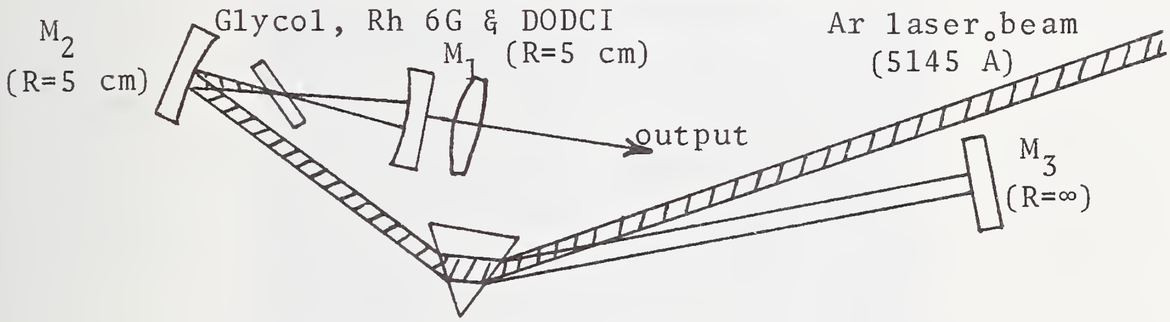


Figure 4-6. Optical Autocorrelation measurement result.

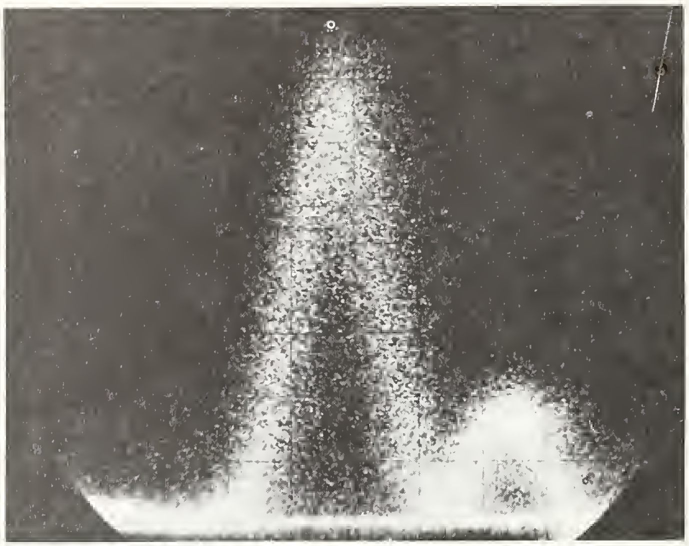


(a) Five mirror arrangement.

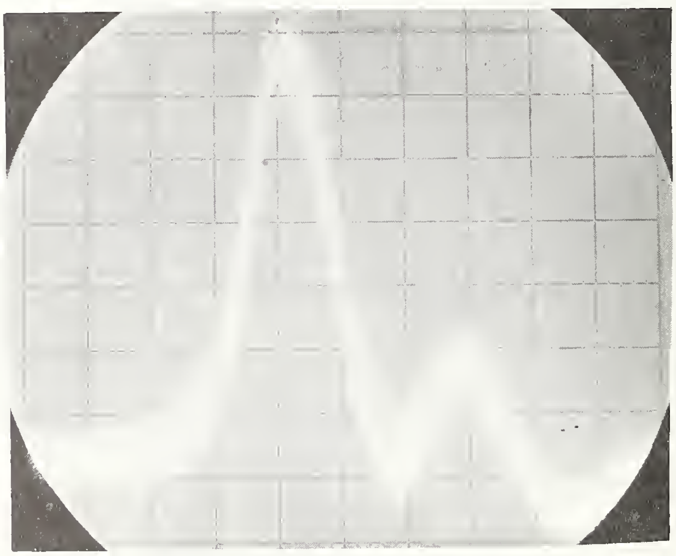


(b) Three mirror arrangement.

Figure 5-1. Passive mode-locking, experimental setup.



(a) Five mirror cavity. The signal is very noisy. The pulse width is estimated to be 80 ps (FWHM) on this photograph. 20 mV/div., 50 ps/div.



(b) Three mirror cavity. The pulse width is 74 ps (FWHM) here. 5 mV/div., 50 ps/div.

Figure 5-2. Passive mode locking, detected pulse waveforms.

Table 6-1. Comparison of the results obtained with the two mode locking techniques.

	Other Laboratories			Our Work			
	τ (FWHM) (ps)	$P_{av.}$ (mW)	P_{peak} (W)	τ (FWHM) (ps)	$P_{av.}$ (mW)	P_{peak} (W)	λ_o (Å)
Active Mode locking							
Acoustooptic [1]	55 (225 MHz)	50	4				
[1]	70 (75 MHz)	--	--	32 (140 MHz)	10	2.2	5900
Electrooptic [18]	<200 (∿400 MHz)	--	--				
Passive Mode locking							
DODCI							
[2]	1.5 (83 MHz)	10	50-100	<35* (135 MHz)	10	≥ 2	5800
[23]	4.3 (140 MHz)	1	1.7				

*Probably detection-limited.

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Report of research performed by Dr. André Scavennec as an NBS guest scientist from Centre National D'Études des Télécommunications, Issy-les-Moulineaux, France

16. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here.)

Using confined and unconfined fluid flow dye cells with suitable mode locking methods continuously operating Rhodamine 6G dye lasers have been built to produce narrow pulses (less than 40 ps) at about a 140 MHz pulse repetition rate. For active (acoustic) amplitude modulation mode locking methods optical pulses were obtained having about a 32 ps pulse width (FWHM). For passive mode locking methods optical pulses of ≤35 ps pulse width (FWHM) were obtained.

A three mirror cavity was used for the active mode locking studies while three and five mirror cavities were used in the passive studies. The three mirror passive mode locked laser employed a single dye cell containing both the Rhodamine 6G (gain) and DODCI (loss) solutes in a single solution having glycol as the solvent. The active and passive mode locked carrier wavelengths were about 5900 Å and 5800 Å, respectively.

17. KEY WORDS (Alphabetical order, separated by semicolons)

DODCI; Dye Laser; Laser; Mode-Lock; Picosecond; Rhodamine 6G

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