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TUNABLE LASERS FOR WATER VAPOR
MEASUREMENTS AND OTHER LIDAR APPLICATIONS

by

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FINAL REPORT
to the
NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
FOR GRANT
NSG 1156
TUNABLE LASERS FOR WATER VAPOR MEASUREMENTS
AND OTHER LIDAR APPLICATIONS

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ABSTRACT

Under NASA Grant NSG 1156 a tunable dye laser suitable for differential absorption (DIAL) measurements of water vapor in the troposphere was constructed. A multi-pass absorption cell for calibration was also constructed, and both instruments were transported to NASA-LaRC for use in an atmospheric DIAL measurement of water vapor. The electronics, telescope, and photo-detectors for the H₂O lidar were constructed by NASA-LaRC personnel. This final report under NSG 1156 emphasizes the main Maryland effort, namely the tunable dye laser and the calibrating absorption cell.

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I. INTRODUCTION

The purposes of this report are (1) to describe the tunable dye laser constructed for the University of Maryland - NASA:LaRC water vapor lidar, (2) to serve as an operating manual for that laser, and (3) to describe a large aperture, multipass absorption cell for calibrating laser absorption by known amounts of water vapor.

The laser described here is a tunable dye laser operating in the near infrared and pumped by a giant-pulse ruby laser. The output wavelength can be varied from 715 to 740 nm with the present optical elements and dye solution. However, this range can be extended as far as 1000 nm by an appropriate variation of dye, output mirror and etalon. The conversion efficiency of ruby pump light into tunable dye laser output is 5% with an upper limit of 30 mj set by the threshold of grating damage. The spectral width (FWHM) is .01 nm and the beam divergence less than 3 mr. It is assumed that the temporal behavior of the dye laser output follows the ruby pump behavior. The spectral output is a discrete spectrum consisting of lines separated by the mode spacing of the laser cavity which is $\Delta\bar{\nu}_c = 1/2L = 10^{-2} \text{ cm}^{-1}$ corresponding to $5 \times 10^{-4} \text{ nm}$. The layout is shown schematically in Figure 1 and discussed in detail in Section II.

The dye laser cavity itself consists of a dye cell, an output mirror, an echelle grating used as a dispersive rear reflector, and an intra-cavity etalon. Associated with the cavity is a dye circulating system and reservoir, along with beam-handling optics for the ruby laser

pump beam. Components which are in contact with the dye are restricted to stainless steel, glass and polyethylene, with O-ring seals of Teflon or ethylene-propylene. The individual components are discussed in more detail in subsequent sections.

The degree of tuning of each laser pulse to the water vapor absorption lines used in the lidar application can be determined by passing a portion of the laser light through an absorption cell containing a known amount of water vapor. A long cell for this purpose was constructed by Maryland personnel, partly in College Park and partly at LaRC, and installed under the laser table in Bldg. 1271 at LaRC. The cell has been used to measure H_2O line profiles as the laser is tuned, and for extensive direct measurements of "on-line" absorption in the lidar mode. Optical and electronic characteristics of the cell and detectors are described in Section VII.

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II. ASSEMBLY AND ALIGNMENT

A. Aligning the Dye Laser Cavity

The dye laser cavity, diagrammed in Figure 1, consists of a dye cell, a front reflector, an echelle grating, and a fixed-spacing Fabry-Perot etalon. These components are mounted on a dovetail track which establishes the optical axis of the laser. In addition, a circulating pump and dye reservoir are connected to the dye cell, maintaining a continuous flow of dye solution through the cell.

The first step in aligning the dye laser cavity is the definition of a lasing axis. This is conveniently done by setting up a helium-neon laser so that its beam travels along the desired cavity axis. In order to accommodate the heights of the various laser components, this axis must be set at a height 5 1/2" above the breadboard surface.

Once the axis has been defined, the dovetail laser track must be oriented parallel to and roughly centered below the helium-neon beam. A pinhole mounted on a piece of dovetail saddle is helpful for this step. Slide the pinhole up and down the track, repositioning the track until the beam hits the pinhole at both ends of the track. The track is now aligned and should be clamped to the breadboard.

Integration of the dye cell into the system requires that the cell first be filled with clear dye solvent. Since the dye solvent DMSO has an index of refraction comparable to that of glass, the dye-filled cell will translate the laser axis horizontally. The dye cell is wedged by 30° so that the beam is also deflected in angle. Clear DMSO must be used in alignment to view this displacement since the dye solution will not transmit the helium-neon beam. Clamp the filled dye cell to the laser track and, if necessary, translate the cell sideways until the

helium-neon beam passes symmetrically through its center. As shown in Figure 2, the beam should enter and exit the cell about 1 cm from the window edges.

Clamp the output reflector mount onto the optical track at a distance of about 30 cm from the dye cell, translating sideways if necessary to bring the helium-neon beam through the center of the mirror aperture. Position the mirror in the mount with its reflecting surface facing the cavity and rotate the Allen wrench axis-adjustment screws to return the reflection from the mirrored surface into the helium-neon laser.

Attach the grating mount to the track at a distance of about 10 cm behind the dye cell and clamp the grating into its holder by tightening the two nylon setscrews, making sure grating blaze direction is correct. If necessary, translate the mount sideways to center the helium-neon spot the grating. At this point several reflected diffraction orders of the helium-neon beam should be visible. Adjust the azimuth and pitch micrometers to bring the grating into Littrow with one of the diffracted beams returning to the helium-neon laser. Next manipulate the pitch micrometer to bring an adjacent diffracted beam into Littrow. If this cannot be done by moving the pitch screw alone, the grating clamp must be rotated about the laser axis to bring the grating rulings parallel with the pitch axis. This will be the case when Littrow configurations can be selected for adjacent orders by a single pitch adjustment, assuring that the cavity will remain in alignment as the wavelength is tuned.

Insert the etalon and its mount into the cavity, between the dye cell and output reflector, adjusting the sideways travel if necessary.

Adjust the azimuth and pitch micrometers to bring the etalon normal to the laser axis. As this is done, all the multiple helium-neon return beams will converge and return into the helium-neon laser. Once this alignment has been made, back off one full turn (50 small units) of the azimuth micrometer. This brings the etalon one free spectral range away from the axis normal and prevents lasing between the etalon surfaces and the grating. It is suggested that an iris diaphragm be inserted into the cavity adjacent to the etalon to limit the cross-sectional lasing area to the center centimeter of the etalon plates. At this stage the laser is aligned for a series of wavelengths including 6328 Å. With DTDC and DMSO the laser will lase at approximately 7263 Å. Tuning onto 7243 Å is discussed in Section VI.

F. Alignment of Pump Beam

The dye laser requires a pumping beam from a giant-pulse ruby laser. The pump pulse energy should be adjustable from 0.3 to 0.7 Joule with a beam diameter of one centimeter. It is extremely desirable that the ruby beam have a "clean" mode structure to ensure even pumping of the dye. In this respect it may be advisable to operate the ruby laser at a higher pulse energy region where the lasing is more uniform, and then attenuate the output beam to the proper level. In addition, it is noteworthy that the dye laser output is polarized, and that this polarization is parallel to the grooves of the diffraction grating. Since the dye laser cavity efficiency is optimized for a polarization in the plane of the table, the ruby laser polarization should be fixed in this horizontal plane for this particular dye.

As indicated in Figure 3, the ruby laser beam is directed into the dye cell by two 100% reflectors. In positioning these mirrors, it

is helpful to do a preliminary alignment using a helium-neon laser placed collinear with the ruby laser axis. A more precise alignment can then be made by using the ruby beam to burn exposed Polaroid film placed at various positions along the path to the dye cell.

The point at which the ruby beam enters the dye cell should be adjusted to nearly coincide with the point where the dye laser axis enters the cell. This is accomplished by using the helium-neon alignment laser as an indication of the dye laser axis when a Polaroid burn pattern is taken at the dye cell window. As shown in Figure 4, the burn center should lie slightly (2-3mm) displaced from the helium-neon spot. The displacement is such that the pump beam crosses the dye laser axis within the dye.

To prevent damage to the laser optics, two special considerations should be made in this alignment process. First, care should be taken to center the ruby beam on the 100% reflectors sufficiently well to avoid burning the anodizing of the mirror mounts. Second, whenever Polaroid burns are used as a diagnostic tool, the optics in the vicinity should be blown with dry air to remove the dust and smoke particles which inevitably result. This is especially important since any dirt particles on the optical surfaces can act as absorbers for the laser light and may thus cause damage to the optical coatings.

III. DYE PARAMETERS

The dye used for this application was selected from the family of cyanine dyes which span the near infrared in their capacity as laser dyes. Our selection is Kodak #7663, known as DTDC, short for 3-Ethyl-2-[5-(3-ethyl-2-benzothiazolylidene)-1,3-pentadienyl] benzothiazolium Iodide, (M. W. 518.48) and is useful over a range of 7150-7400 Å. We

have found dimethyl sulfoxide (DMSO) to be the most efficient solvent for this application. In our particular region of interest, 7200-7300 Å, the efficiency curve of this dye in DMSO is nearly flat. We have found it useful to store this dye in a 10^{-3} M stock solution consisting of 260 mg of DTDC dissolved in 500 ml of DMSO. An operating strength solution is prepared by mixing 75 ml of the stock solution with 500 ml of DMSO.

There is a marked deterioration of the dye solutions when they are exposed to fluorescent room lights for an extended period of time. To prevent this photodegradation the dye solutions should be stored in a dark place in amber glass bottles. In addition, the dye lines should be shielded from room light.

The biological properties of DTDC are not well studied but in sufficient dosage it may well be toxic¹. The toxicity problem in handling dye solutions is complicated by the use of DMSO as a solvent, since DMSO readily diffuses through the skin, carrying solute molecules with it. It is therefore important that any contact with dye solutions be avoided, and that gloves be worn as a minimum precaution whenever dye solutions are handled.

IV. DYE CIRCULATING SYSTEM

The dye circulating system maintains a continuous flow of solution through the dye cell by means of a circulating pump, reservoir, and connecting tubing as shown in Figure 5. This constant recirculation of dye prevents the formation of temperature gradients in the cell, ensuring lasing of the dye.

1. Kues, H. A., Latty, G. A., Laser Focus, 11 No. 5, 59 (May, 1975).

Given the chemically reactive nature of DMSO, the circulating system is constructed to place only substances inert to DMSO in contact with the dye solution. All metal parts are constructed of stainless steel, with seals made of either Teflon or ethylene-propylene. The tubing used for dye lines is made of polyethylene.

The pump is a Micro-Pump model 12-50-316-761 gear pump with an induction motor. It incorporates a 316 stainless steel body and shaft with Teflon gears. The dye reservoir, also of stainless steel, has a one-liter capacity and can be sealed airtight to prevent the escape of DMSO vapor. The dye lines are $\frac{3}{8}$ " Eastman Poly-Flo tubing, and stainless steel fittings are used throughout.

V. CAUTIONS INVOLVED IN OPERATION OF DYE LASER

The most singularly delicate component of the dye laser is the sapphire grating, in that it is not only easily damaged by careless handling, but it is subject to burn damage from excessive laser cavity energy. In this latter regard we have found that the damage threshold corresponds to a dye laser output energy of approximately 30 millijoules. When one considers that the shot-to-shot energy variation can be as much as 30% (and long-term drifts even greater), it appears advisable that a "safe" upper limit for output energy be set below 30 millijoules. We have found that a value of 15 millijoules has resulted in no damage over several hours of operating time. The energy can be measured directly with the Gen Tac joulemeter and an oscilloscope.

In order to restrict the dye laser pulse energy to safe limits, the ruby pulse energy should be held to less than 0.7 Joule. We have found that the beams from ruby lasers often exhibit an undesirable mode structure at these low energies, and in order to clean up the beam it is

necessary to operate the ruby at higher energy settings. This problem is easily resolved by insertion of a 0.5 glass neutral density filter in the ruby beam, thus allowing operation of the ruby in a "clean" energy region while limiting the energy seen by the dye cell. Another approach is to use the full laser energy with a more dilute dye solution.

As noted in the sections covering laser alignment, the presence of dust or dirt on the laser optics can result in burns on the optics when the laser is being operated. Therefore these optics should be blown with dry air before each operating session and after making any Polaroid burn patterns. As an additional precaution the optics should be covered when not in use.

A further precaution involving both grating safety and output beam quality involves the angular setting of the etalon. In the process of wavelength tuning, care must be taken to avoid positioning the etalon normal to the dye laser axis, since lasing would then occur between the etalon surfaces and the grating. A good rule of thumb is to maintain the etalon at least one free spectral range (50 small units, or one turn of the micrometer) away from normal. This still allows a useful working region of three or four free spectral ranges.

In regard to the dye circulating system, a few cautions are in order. As mentioned earlier, the circulating pump must be running when the dye laser is being operated. It is also advisable to switch on the pump a few minutes before initially firing the laser, since this will give the solution in the lines a chance to reach thermal equilibrium with the dye in the reservoir, reducing thermal gradients in the dye which could give rise to laser instability.

VI. FINE ALIGNMENT AND WAVELENGTH-TUNING

In the process of optimizing both the magnitude and mode structure of the dye laser output, it is a useful exercise to observe the output beam as fine adjustments are made in the grating azimuth angle. There are two convenient methods for making this adjustment. A rough alignment is facilitated by measuring the output with a joulemeter as the azimuth angle is stepped about its original setting. It is then useful to photograph the laser spot on a target. By photographing the beam at several settings around the peak-energy point, the optimum setting can be selected as the one which produces the narrowest spot.

The process of wavelength-tuning is accomplished by manipulation of both the grating and the etalon. The dye molecules have a characteristic lasing curve about 150 \AA wide. The grating presents the cavity a pass band approximately 1 \AA wide, and the etalon in turn offers a series of pass bands of width 0.1 \AA and separated by about 1.2 \AA . A spectrum of the dye laser output will therefore consist of one or more etalon lines which appear in a region determined by the grating pitch angle.

To tune the cavity to a specific wavelength, first adjust the grating pitch such that the center of its pass band is located approximately at the wavelength of interest. The angular dispersion $\frac{\Delta\lambda}{\Delta\theta}$ for the pitch controls is roughly 50 \AA per turn of the rough pitch adjustment screw and 4.87 \AA/mm of the pitch micrometer. This done, adjust the etalon azimuth micrometer until one of the etalon pass bands is located exactly on the desired wavelength. The free spectral range of the etalon is 2.5 cm^{-1} (1.2 \AA); its angular dispersion is 3.3 \AA/mm .

on the azimuth micrometer. Finally, adjust the grating pitch control to maximize the lasing energy in the selected etalon mode and simultaneously eliminate lasing in all other modes. This amounts to repositioning the grating pass curve so that its central peak exactly coincides with the wavelength of the selected etalon mode. This step is accomplished by observing the intensity of the modes with a spectrograph.

VII. MULTIPASS ABSORPTION CELL

The use of an auxiliary cell to calibrate concentration measurements to be carried out with tunable lidar systems was first suggested to one of the authors in a private communication from M. L. Wright of SRI in 1971. When G. B. Northam of LaRC again suggested it for the water vapor experiment, we considered several possible versions and finally adopted the one described here partly on the basis of optics available from University of Maryland laboratories.

It is a long path cell (~ 300 M) employing ~ 75 successive reflections from large spherical mirrors separated by about 4 meters. The gas in the cell is room air^{*}, and is enclosed by a light-tight wooden box which has been blackened inside with flat black and "black velvet" paint.

In most of the work since carried out with the Maryland absorption cell, the 300 M pathlength in room air has proved sufficient (optical depth ~ unity) for calibrating lidar operation on the H₂O lines near 724.3 nm. These lines are among the strongest in that band system, according to a separate Maryland study^{**} with the NASA-Ames Research Center, while a few lines have roughly 1.5 times the strength of λ 724.371; therefore a variety of lines can be used in that spectral region, depending on humidity variations and pathlength adjustments. In any case, the

* Other designs involving shorter paths through denser H₂O vapor were given lower priority because they involved heated cells and the attendant problems of temperature non-uniformity, H₂O condensation and the optical perturbation by strong air currents.

** T. Wilkerson, private communication

primary humidity standard is a psychrometer measurement of water vapor in the room air. On some occasions, humidity in the absorption cell has been observed to remain constant for days.

The use of such a cell is based on detection of both the input and output beams, preferably under circumstances where the two signals are comparable in magnitude and can be well resolved in time for measurements of pulse amplitude or the integrated energy in a pulse. We accomplished this by using two identical fast photomultipliers which, together with their electronics, gave response times ~ 3 ns. Because of the 300 M pathlength through the cell, there is a 1 μ sec delay between the input and output signals - which themselves are only 25 ns in duration. This enables one to use gated pulse measurements to great advantage, and to ascertain that no light is being detected from scattering off the optics partway through the long path system.

The PM tubes were RCA-4832, whose GaAs photocathodes give reasonably high quantum yields (7-10%) around 720 nm; response times of the tubes are 1.5-2 ns. We used AC-coupled, non inverting buffer amplifiers to couple the PM outputs to coaxial cables leading to the lidar data acquisition system; this was done because we anticipated the possibility of an electrically noisy environment, under circumstances where very long signal cables might be needed. The current gain of the buffer amplifiers was about 10, and the overall system bandwidth about 250 MHz. The detector systems were tested at Maryland with fast, yellow light pulses from a dye laser pumped with the 6 ns output of a pulsed N_2 laser.* In the LaRC lidar system, the data

*University of Maryland Coordinated Laser Facility

from the absorption cell PM tubes were collected in LeCroy gated A/D pulse integrators having 8 + 1 bit digital outputs. Gated detection was very important because of the noise signals from the flash lamps of the main ruby pump laser. In retrospect it was clear that the good time separation of the two absorption cell signals would have permitted the use of one rather than two PM detectors.

This section closes with a description of the optical arrangement of the cell. Figure 6 shows a trial configuration of the main mirrors facing one another in vertical tripod mounts. These are 12.5" (31.75) cm diameter Tinsley mirrors having focal lengths of 2.5 M. They were recoated* with gold, and overcoated with SiO₂, to obtain 95% reflectivity at 730 nm; the signal loss at 44 reflections for example is about a factor of ten.

The available space in Bldg. 1271 at LaRC made it more convenient to run this cell with a 4 meter mirror separation, than with the 5 meter separation that one might expect from the focal length. Moreover, we found in extensive tests at Maryland that a re-entrant or "perturbing mirror" system** could be used to obtain from the 4 meter configuration of this cell a sufficient number of passes for 300 meter total pathlength. This was a conservative design based on a minimum size requirement for the insertion and perturbing mirrors, to allow for size and position variations of the dye laser beam spot.

* Denton Vacuum, Cherry Hill, N. J.

** D. R. Herriott and H. J. Schulte, Appl. Optics 4, 883 (1965).

Figure 7 illustrates the absorption cell configuration as finally installed under the main laser table at LaRC. A fraction of the dye laser output was brought down to the cell via a pellicle (see Fig. 3) and turning mirrors, and first attenuated (as needed) by a choice of ND filter at the entrance to the box containing the multipass mirror geometry. The "input" channel for the cell is provided by a glass plate which picks off ~ 4% of the input, reflecting it into the detector PM(1), and allows most of the light to continue into the long path system. The insertion mirror position and angle determine an elliptical configuration of spots on each mirror where the beam hits. The perturbing mirror is placed at one of these spots, so as to return the beam to the mirror system at a different angle relative to the optic axis. This generates yet another set of beam spots on the mirrors, a set which is adjustable in locus and spacing independently from the first set. The extraction mirror is placed at one of these spots, and reflects the beam into the detector PM(2). A choice of ND filter in front of PM(1) makes the input and output signals for the cell as nearly comparable as is necessary to ensure that both PMs are operating at the same signal level, and the LeCroy integrators are covering the same portion of the dynamic range of signal.

The small mirrors throughout the system are standard 1/8 wave, aluminized elliptical mirrors epoxied to 1" x 1" NRC adjustable mounts. In the final configuration adopted for the absorption cell, 75 passes were used; these were established by means of an alignment laser and adjusting the system for 37 beam spots at each of the large mirrors. The 75th pass takes place in the beam passage from the extraction mirror to PM(2).

As well as being used routinely for lidar calibration, the cell has been used for quantitative scans of H_2O line shapes near 724.3 nm. This work was reported* by E. Browell at the Eighth Lidar Conference, and has been described in numerous internal NASA presentations.

* E. V. Browell, M. L. Brumfield, J. M. Siviter, G. B. Northam, T. D. Wilkerson and T. J. McIlrath, "Development and Evaluation of a Near-IR H_2O Vapor DIAL System", Proc. Eighth International Conference on Laser Radar, Drexel Univ., Philadelphia, PA. June 6-9, 1977

FIGURE 1 : DYE LASER CAVITY

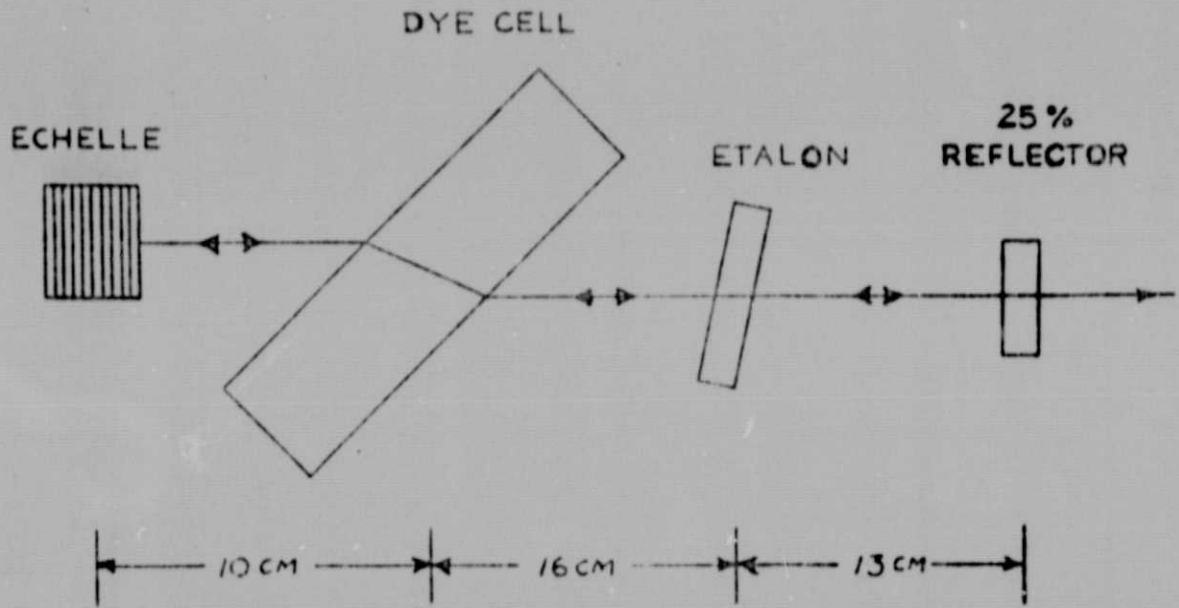


FIGURE 2 : BEAM PATH IN DYE CELL

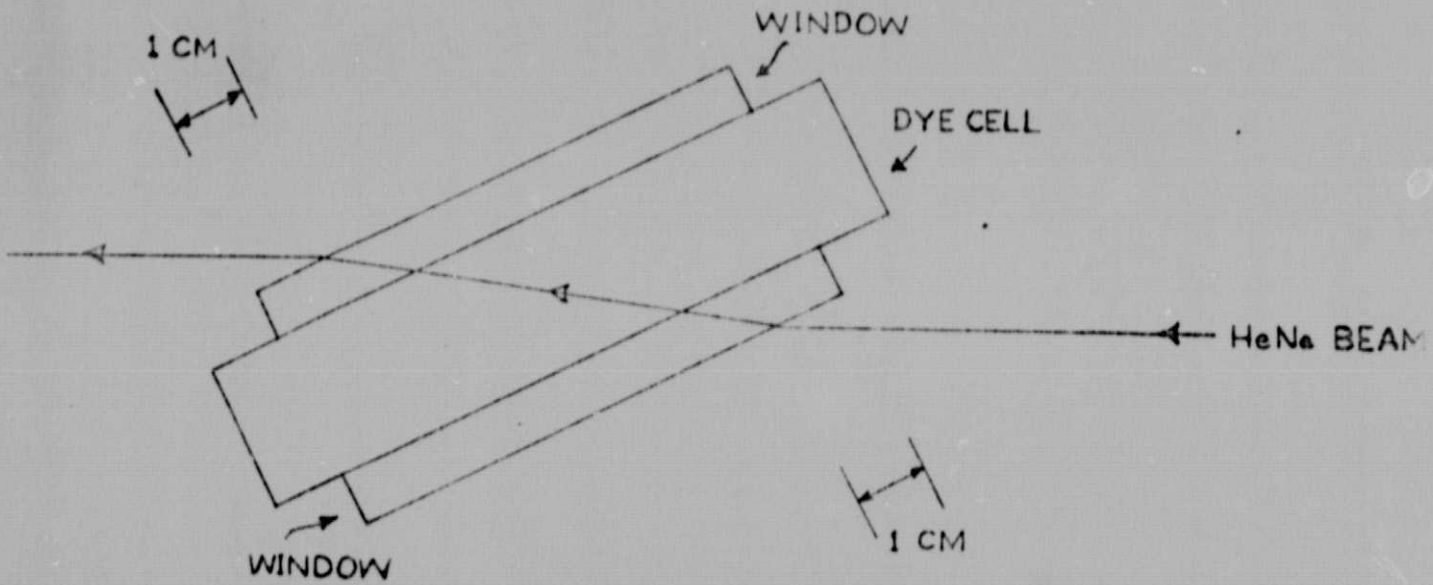


FIGURE 3 : DYE LASER SYSTEM

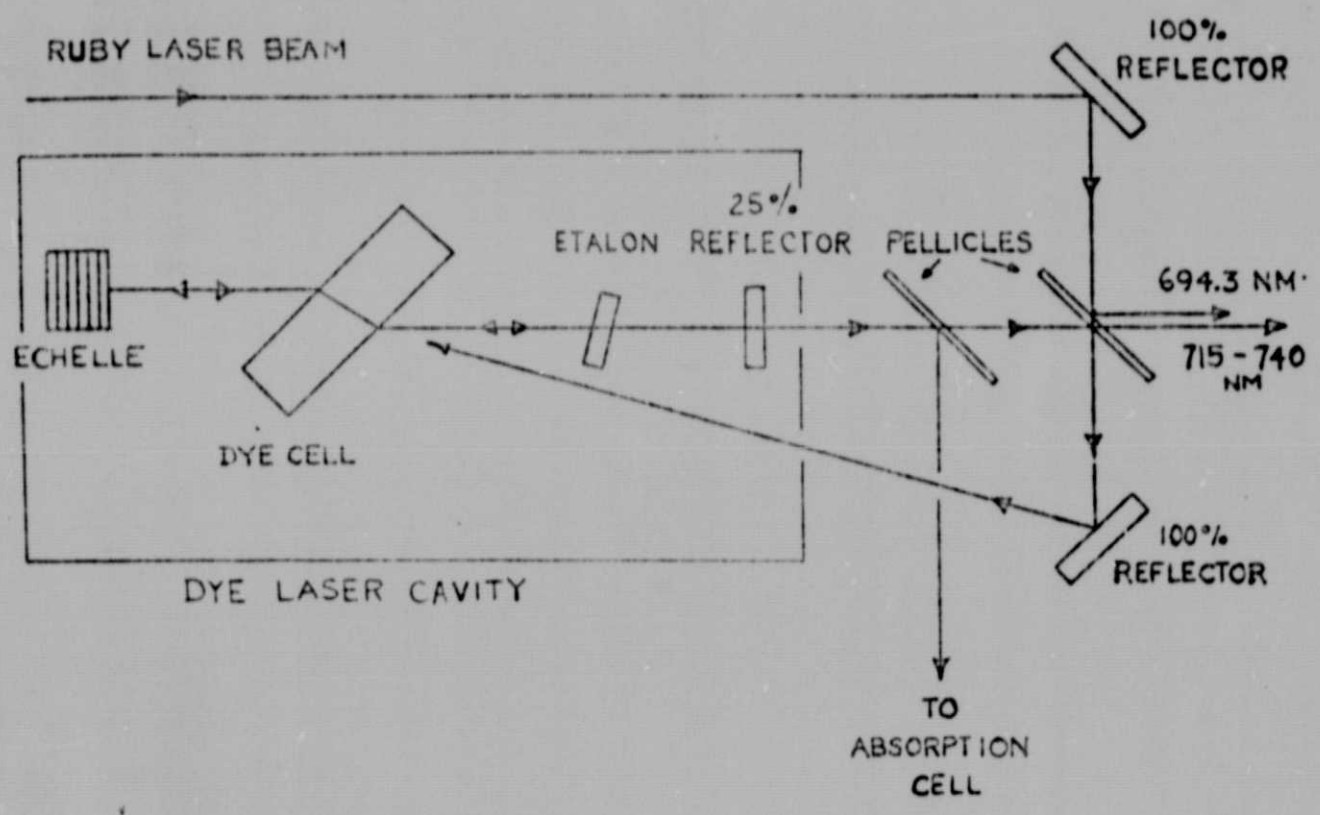


FIGURE 4 : ALIGNMENT BURN PATTERN
AT DYE CELL WINDOW

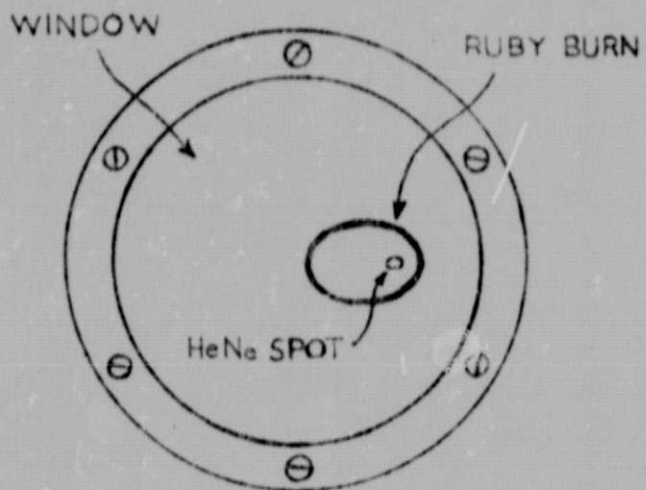


FIGURE 5 : DYE CIRCULATING SYSTEM

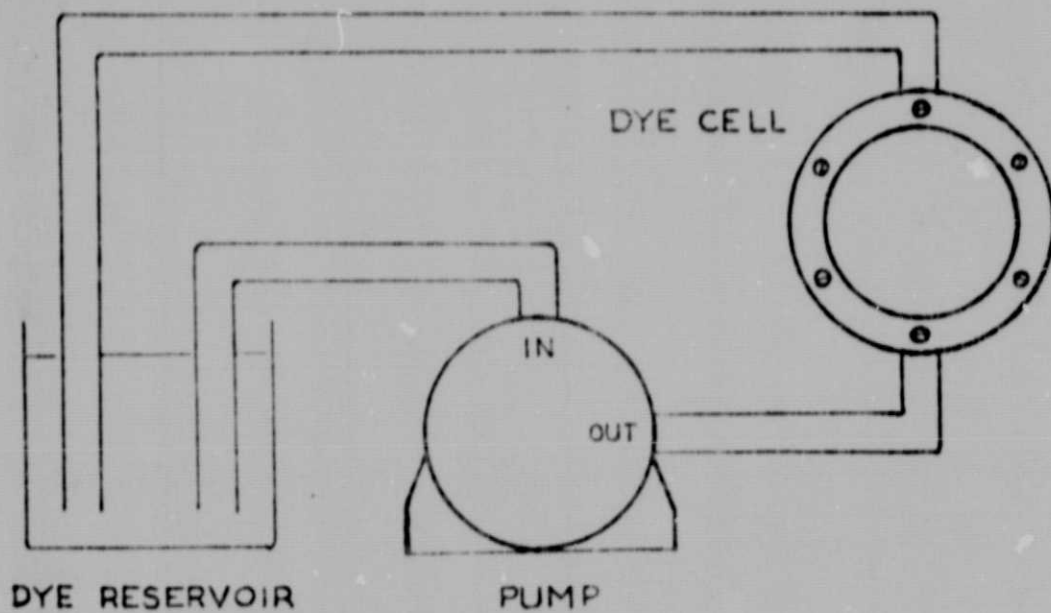
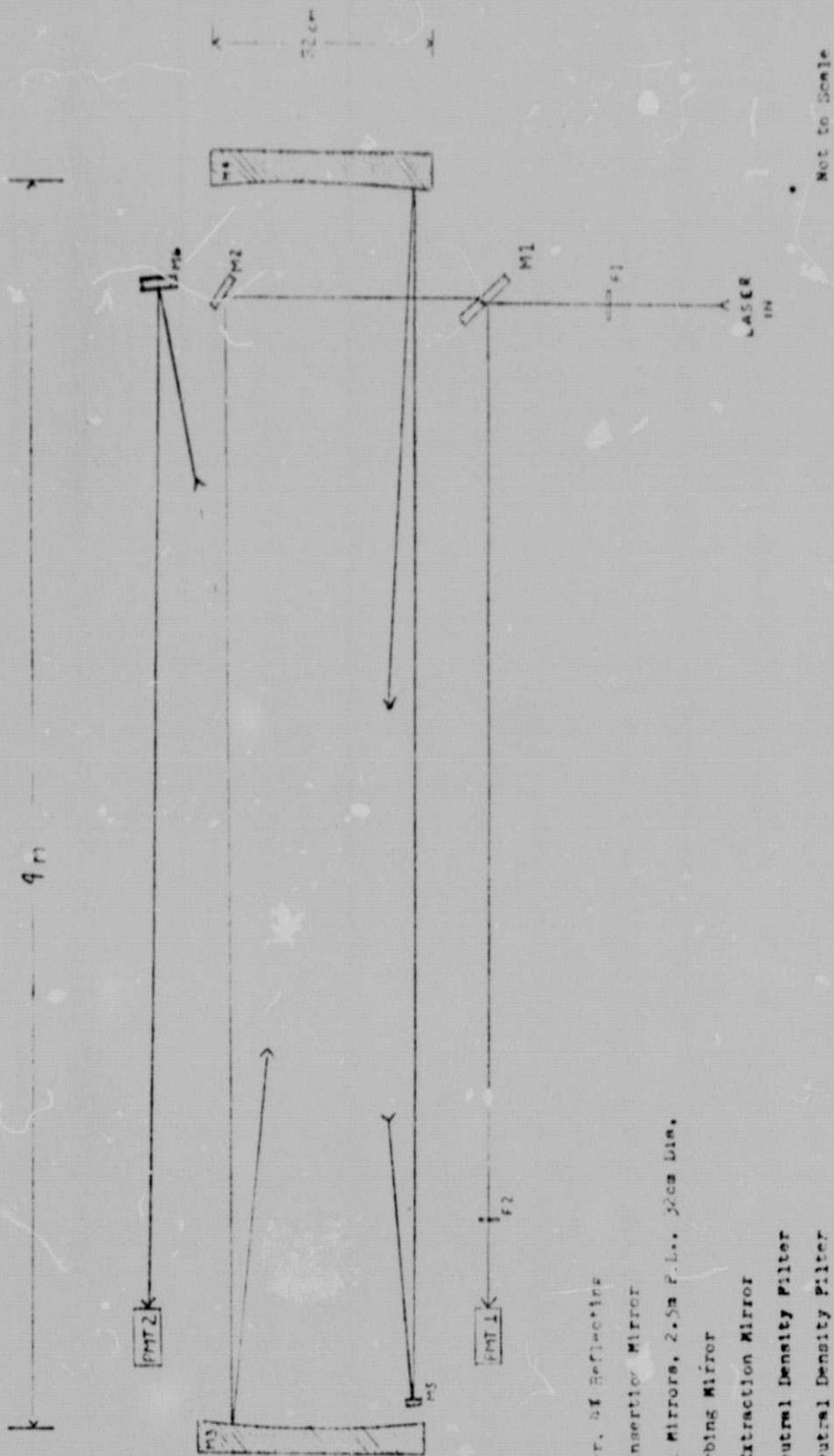




Fig. 6 Test Setup for Absorption Cell using
32 cm dia. Mirrors

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- M1 Beam Splitter, 4K Reflective
- M2 Plane Beam Insertion Mirror
- M3, M4 Spherical Mirrors, 2.5m f.l., 52cm Dia.
- M5 Plane Perturbing Mirror
- M6 Plane Beam Extraction Mirror
- F1 ND 3 to 5 Neutral Density Filter
- P2 ND 0 to 2 Neutral Density Filter
- PMT1, PMT2 RCA 4832 Photomultiplier Tubes

FIG. 7 - ABSORPTION CELL OPTICAL SCHEMATIC