

## A HIGH-RESOLUTION CW DYE LASER SPECTROMETER<sup>☆</sup>

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The construction and utilization of a high-resolution cw dye laser spectrometer is described. The laser incorporates pressure tuned intra-cavity etalons enabling the single mode output laser frequency to be tuned more than one nm in a continuous scan. In the scan the laser jumps from one cavity mode to the next in  $\sim 50$  MHz steps. In comparison to molecular Doppler widths this mode jumping is negligible. As an example of the use of the instrument, a high-resolution absorption spectrum of molecular iodine between the sodium D lines is presented. A unique frequency counter provides the calibration of the absorption spectrum.

### 1. Introduction

The cw dye laser spectrometer consists of a narrow linewidth (about  $10^{-4}$  nm) source of coherent radiation and a frequency marker by which the output frequency can be accurately offset from a known calibration point such as an atomic line to any desired value within the scanning range. In being continuously tunable over several nm, the instrument is particularly useful in experiments involving optical pumping of a particular molecular absorption line because the proper absorption may be identified by the line pattern of the molecular spectrum. With the wavelength location problem thereby solved, the laser may be used for a variety of optical experiments such as double resonance, resonant scattering, level crossing, or isotope separation.

### 2. Principle of operation

The narrow output linewidth of the dye laser is achieved by forcing the laser to oscillate in a single longitudinal mode. This is accomplished using a com-

bination of intracavity dispersive prisms and Fabry-Pérot etalons. Two intracavity high refractive index Brewster angle prisms narrow the linewidth from several hundred GHz to about 20 GHz. Two intracavity (20% reflection) coated etalons of FSR 150 GHz<sup>†</sup> and 7.9 GHz limit the oscillation to a single longitudinal mode. The linewidth is then limited, in practice, by mechanical instabilities in the laser cavity to typically 100 MHz.

A most important observation is that over an interval of somewhat less than the bandwidth prior to the insertion of the etalons (about 20 GHz) the frequency of oscillation is determined by the etalons. This frequency is that of the longitudinal cavity mode for which best overlap of the etalon transmission maxima with the laser gain profile occurs. The tuning mirror, when tilted through a small angle determines solely the gain at the frequency of oscillation. Quasi-continuous<sup>‡</sup> tuning of the dye laser frequency, therefore, amounts to synchronous tuning of the etalons. The

<sup>†</sup> An etalon should be used which has a FSR larger than the spectral bandwidth over which the laser has sufficient gain to lase (in this case about 100 GHz) rather than the 20 GHz bandwidth over which it actually lases.

<sup>‡</sup> The frequency is discontinuous in hopping from that of one longitudinal cavity mode to that of the next as the laser is tuned. In the dye laser described below, the cavity was made sufficiently long that the longitudinal mode spacing was less than the longterm linewidth of the laser. For practical purposes we may, therefore, regard the tuning as continuous.

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tuning mirror need not be adjusted for frequency scans of less than 10 GHz, and for larger frequency scans the tuning mirror need only be periodically adjusted to maximize the output laser power.

Etalons are used as intracavity mode-selecting elements in cw dye lasers because of their low insertion loss. The etalons must be slightly misaligned from the cavity mirrors so that reflections from the etalon plates are not coupled back into the cavity. Were there any reflective coupling, then the cavity mode frequencies would become extremely sensitive to mechanical vibrations which introduce a relative motion between the cavity mirrors and the etalon plates.

An intracavity etalon introduces a selective loss into the cavity, suppressing most strongly those longitudinal modes which do not lie near any of its transmission maxima. The success of the etalon technique relies on homogeneous broadening of the gain medium, to permit only a single longitudinal mode near a transmission maxima to lase. For an etalon of plate separation  $d$ , inclined at a small angle of incidence  $\theta$  to the collimated intracavity laser beam, the condition that a transmission maxima exists at a frequency  $\nu_d$  of the dye laser is given by\*

$$2nd \cos \theta = mc/\nu_d, \quad (1)$$

where  $n$  is the refractive index of the medium separating the etalon plates,  $c$  is the velocity of light, and  $m$  is an integer. For a given value of  $m$ , the frequency  $\nu_d$  can be varied by changing  $n$ ,  $d$  or  $\theta$ . These changes correspond respectively to pressure tuning, temperature tuning and tilt tuning.

For tuning single mode over frequency ranges of less than 10 GHz the tilt tuning technique is most commonly used, and is the most convenient. For larger frequency tuning, the tilt tuning technique has two serious disadvantages, however. Firstly, it can be seen from eq. (1) that  $\nu_d$  is quadratically related to  $\theta$ , which makes linear frequency scans difficult. Secondly, and most importantly, as  $\theta$  increases walk-off losses in the etalon increase, and the effective finesse of the etalon decreases because of an increasingly poor overlap of successive reflections within the etalon. Thus, it becomes increasingly difficult to maintain a single

mode when tilting  $\theta$  more than a few mrad, and the tilt tuning technique would thus seem to be unsuitable for large range frequency tuning.

Both the pressure and temperature tuning techniques are suitable for tuning over large frequency intervals. The temperature tuning technique offers the advantage that problems in etalon plate alignment can be overcome by using commercially available solid etalons. The pressure scanning technique, employing air-spaced etalons, offers the advantage of rapid frequency tuning while overcoming the problem of thermal inertia. The temperature scanning technique is limited by the maximum temperature to which the (coated) etalons can safely be raised (at 600 nm a solid quartz etalon will scan at the rate of  $4 \text{ GHz}^\circ\text{C}^{-1}$  or  $200^\circ\text{C nm}^{-1}$ ). Setting a limit of several atmospheres for pressure scanning still permits tuning over a much greater range.

When considering pressure tuning it is appropriate to write eq. (1) in the approximate form

$$d\nu_d/dp \approx \alpha \nu_d (1 - \alpha p), \quad (2)$$

where  $p$  is the gas pressure and  $\alpha$  is a constant (of order  $10^{-3}$ – $10^{-4} \text{ atm}^{-1}$ ). In deriving eq. (2) we have assumed that  $\theta^2$  is small in comparison to unity, and that the refractive index,  $n$ , is related to  $p$  by the equation

$$n = 1 + \alpha p. \quad (3)$$

From eq. (2) the rate of etalon frequency tuning is seen to be independent of the etalon plate separation and is to a good approximation also independent of the gas pressure. Thus, several intracavity etalons are synchronously tuned as the pressure is varied.

### 3. Construction of the spectrometer

A schematic diagram of the dye laser spectrometer is shown in fig. 1. It is essentially the 3 mirror configuration of Dienes et al. [1]. Several other laser cavities have been examined for stable single mode operation, in particular a variation of the 3 mirror configuration in which mirror  $M_1$  in fig. 1 is replaced by a lens-plane-mirror combination, and the "crossed" 4 mirror configuration of Johnston and Runge [2]. Both of

\* Inclusion of the effect of the small wedge angle between the front and back surfaces of each etalon plate adds a constant to the value of  $\theta$  in eq. (1).

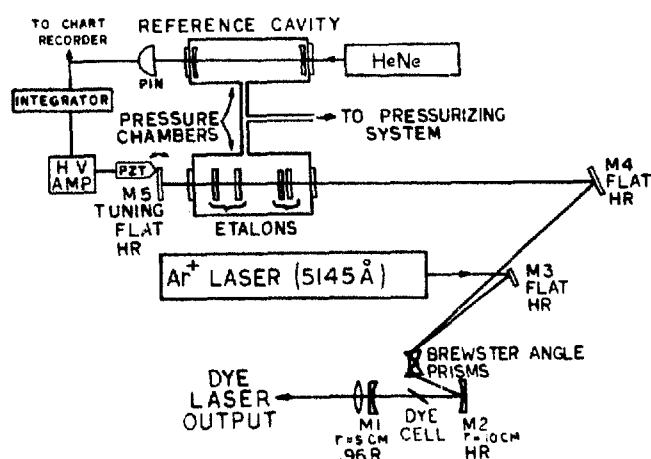


Fig. 1. Schematic diagram of dye laser spectrometer.

these configurations are appealing since they have intracavity regions in which the dye laser beam is collimated and remains fixed as the dye laser frequency is tuned, making these seemingly ideal regions in which to locate the mode selecting etalons. The 3 mirror configuration of fig. 1 was found to be significantly better than these other configurations, for reasons not fully understood. However, in the case of the modified 3-mirror configuration, spurious interference effects were observed [3], and found to be caused by the AR coated lens. Several high quality AR coated lenses of different focal lengths were tried, all behaving similarly.

The dye laser, pumped by the 514.5 nm output of a Spectra Physics Model 170 argon-ion laser, uses a  $3 \times 10^{-4}$  molar concentration of rhodamine 6G in water with 6% by volume Ammonyx LO added. The dye flows at about  $14 \text{ msec}^{-1}$  through a closed cell of thickness 1.0 mm when oriented at the Brewster angle. The pump beam is coupled into the cavity by steering mirror  $M_3$  and two high refractive index Brewster angle prisms. The dye laser cavity comprises mirrors  $M_1$ ,  $M_2$ ,  $M_4$  and  $M_5$ . For spatial mode stability an adjustable aperture is positioned in front of  $M_1$ . All the cavity components are firmly mounted on a heavy table for mechanical stability.

The tuning arm of the cavity containing the intracavity etalons is made long (about 3 m) for two purposes. Firstly, frequency fluctuations caused by changes in the laser cavity length (due to mechanical instabilities in the length of the laser attributed largely to pressure fluctuations in the dye cell) become smaller in proportion to the length of the cavity. Secondly,

as previously mentioned, the smaller longitudinal mode spacing (about 50 MHz) provides for more continuous frequency tuning.

The etalons used to limit the laser to single mode operation have plate spacings of 1 mm and 19 mm. The surfaces of the plates include a  $30^\circ$  wedge and are flat to  $1/20$  wave. Each has a 20% reflecting dielectric coating on one side and an anti-reflection coating on the other. The plates are spaced by invar rods. It is relatively easy to polish the spacers to within a few wavelengths of parallelity by making use of the etalon interference ring pattern which can be seen even with only 20% reflective coatings. Fine adjustments for parallelity can be made if one of the etalon plates is held in place by three adjustable spring loaded pressure pads. The etalon plates can then easily be set to a parallelity of  $\frac{1}{5}$  wave  $\text{cm}^{-1}$ , which is sufficient for use in the laser.

Wedge etalon plates, AR coated on both sides, are used for the windows of the pressure chamber. The windows are orientated at a slight angle of incidence to the intracavity beam and not at Brewster's angle, since for angles other than normal incidence the difference in refractive index on the two sides of each window introduces an angular displacement of the intracavity beam which changes during a pressure scan, resulting in a misalignment of the cavity and eventual extinction of laser action.

The introduction of the pressure chamber and etalons into the cavity substantially raises the threshold of the laser. With a 96% reflecting output mirror ( $M_1$  in fig. 1), the dye laser threshold is raised from typically 500 mW without the pressure chamber and etalons to 1.5 W after their introduction. The increased losses include walk-off losses within the etalons, reflections from the eight AR coated surfaces and some scattering losses. Operating at a power level of 3 W at 514.5 nm, 30 mW of single mode output can be achieved with an amplitude stability better than  $\pm 20\%$ . Such output powers are sufficient to saturate a wide range of molecular transitions [4].

A single spectrum analyzer trace of the dye laser in single mode operation is shown in fig. 2a, and fig. 2b shows the frequency stability over a one-second exposure. The long term frequency stability of the single mode can be seen to be of order  $\pm 60$  MHz. This is much less than the Doppler width at room temperature of most atomic and molecular transitions within the wavelength region of the dye laser.

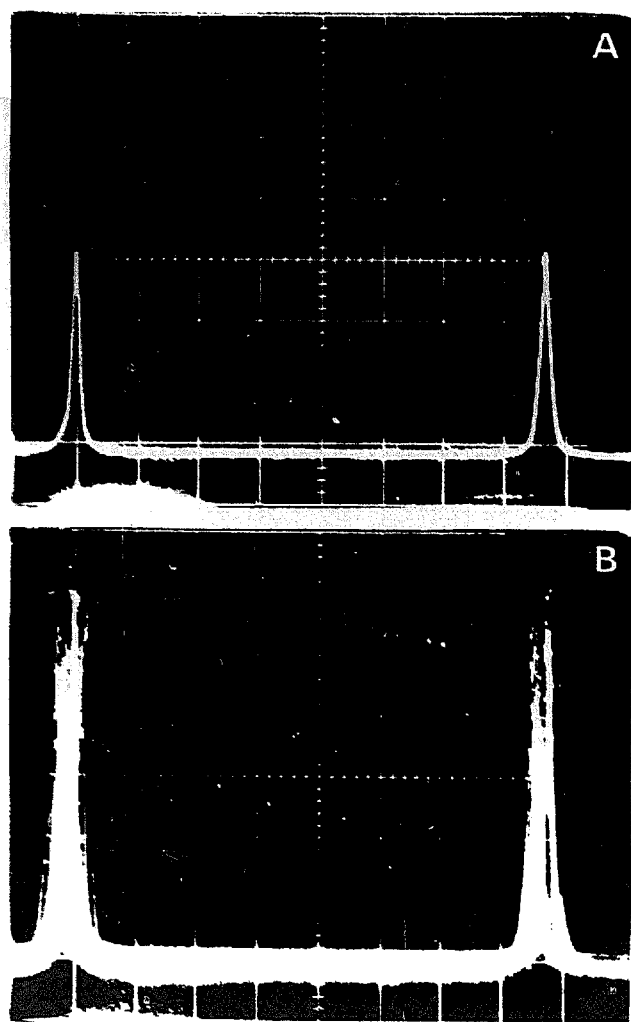


Fig. 2. Output of spectrum analyzer (2 GHz FSR). (a) Single sweep output; (b) 1 sec. exposure.

The vacuum system for the intracavity pressure chamber allows for the chamber to be evacuated and subsequently for air or sulfur hexafluoride ( $\text{SF}_6$ ) to be titrated in at a uniform rate.  $\text{SF}_6$  gas is completely safe to use, and is superior to air for pressure scanning in increasing the tuning range per unit pressure change by a factor of almost three. It is quite feasible to scan the laser over many nm by overpressuring the pressure chamber. For a scan of 0.59 nm between the sodium D lines, for example, a pressure change of only 1.3 atm of  $\text{SF}_6$  is necessary.

A high finesse quartz spaced confocal reference cavity of FSR 375 MHz is used to provide frequency markers for a chart recorder as well as to step the tun-

ing mirror ( $M_5$ ) in synchronism with the frequency tuning of the etalons. The reference cavity is enclosed in a pressure chamber maintained at the same pressure as the intracavity etalon chamber (as shown in fig. 1). Since from eq. (2) the frequencies of the reference cavity transmission maxima sweep at the same rate as those of the intracavity etalons, detection of a stabilized single mode He-Ne laser through the reference cavity provides precise frequency markers spaced by the FSR of the reference cavity. This is a useful feature of the pressure scanning technique: the frequency markers provide for accurate measurement of frequency intervals after allowance has been made for the difference in frequency between the dye and He-Ne lasers. The separation of two adjacent frequency markers corresponds to a change ( $\Delta\nu_d$ ) in the dye laser frequency of

$$\Delta\nu_d = (\text{FSR})_{\text{ref. cavity}} \times \nu_d / \nu_{\text{He-Ne}} \quad (4)$$

The frequency interval  $\Delta\nu_d$  was calculated to be  $402 \pm 2$  MHz from a frequency scan between the sodium D lines.

As was mentioned earlier, the degree of synchronism between the etalon tuning and the tilting of the tuning mirror is not critical. Mirror tilting is accomplished using a piezo-electric translator (PZT) driven by a high voltage operational amplifier. It is quite possible to adjust the PZT voltage manually, but for convenience, the output of the reference cavity is integrated to form a "staircase" voltage which is amplified and applied to the PZT as shown in fig. 1.

#### 4. Experiment

The absorption spectrum of iodine ( $\text{I}_2$ ) provides a convenient demonstration of the scanning capabilities of the dye laser spectrometer. The output from the dye laser was directed through a room temperature iodine absorption cell, and a photomultiplier monitored the iodine fluorescence at right angles to the laser beam.

Fig. 3a shows the iodine absorption spectrum measured in fluorescence between the sodium D lines. The spectrum was recorded in five minutes. In fig. 3b a region of about 50 GHz of fig. 3a is shown, scanned at a slower rate with a 1 sec. time constant added to

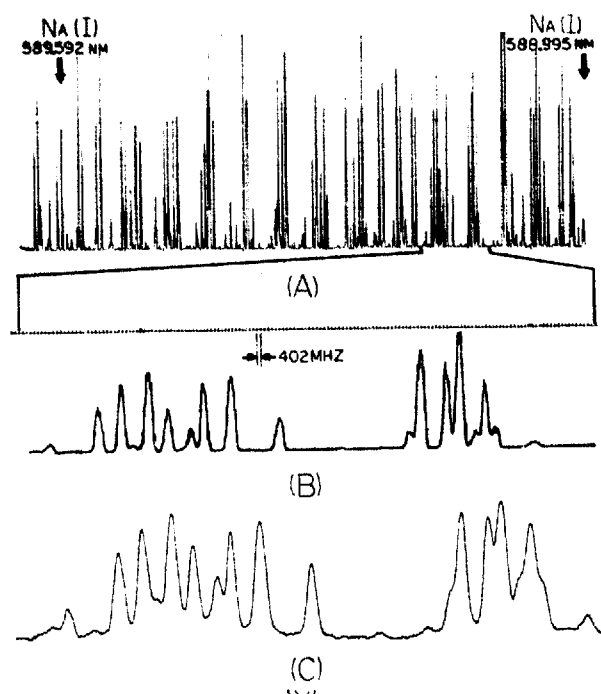


Fig. 3. Iodine absorption spectrum. (a) Taken in fluorescence between the sodium D lines using the dye laser spectrometer; (b) calibrated scan (using 1 sec, time constant) of approximately 50 GHz section of spectrum shown in (a); (c) Same region as shown in (b) but taken in 10th order with a conventional high resolution grating spectrograph (average dispersion  $0.21 \text{ nm cm}^{-1}$ ).

smooth out amplitude and frequency fluctuations. Here, typical absorption linewidths are seen to be about 800 MHz, much greater than the 120 MHz resolution of the spectrometer. This linewidth is a combination of both Doppler broadening (about 550 MHz) and unresolved hyperfine structure.

Fig. 3c is a photoelectric recording of the same region of the iodine absorption spectrum as fig. 3b, taken in the tenth order (an average dispersion of  $0.21 \text{ nm cm}^{-1}$ ) of a 7.3 m Ebert spectrograph†. The comparison of figs. 3b and 3c shows that the dye laser spectrometer gives a faithful representation of the absorption spectrum with slightly higher resolving power.

† The spectrum was taken by Werner Goetz of D.A. Ramsay's group at the Physics Division, National Research Council of Canada, Ottawa, Canada.

By incorporating the etalon pressure scanning technique in frequency tuning a dye laser, we have thus demonstrated the practicality of tuning of a single mode dye laser over many nm. Using the frequency marker, it is possible to set the dye laser accurately to a desired frequency using, for example, the calibrated iodine absorption spectrum.

In optical pumping experiments with molecules having a high fluorescent efficiency, their photoluminescence can be used to identify the pumped transition. However, if a transition cannot be so identified because of a very poor fluorescence efficiency, the dye laser spectrometer is particularly useful, since the desired transition can then be identified from the molecular absorption spectrum. For example, a microwave-optical double resonance experiment on the excited  $A^3\Pi_1$  state of the ICI molecule is planned. The  $A^3\Pi_1 \leftarrow X^1\Sigma^+$  transition from the ground state to be pumped by the dye laser will be first identified from the ICI absorption spectrum since it appears that the resonance fluorescence of the molecule is very faint.

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