A Simple and Reliable Dye Laser System for Spectroscopic Investigations

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Efficient spectral narrowing and accurately reproducible tuning of a dye laser has been achieved by the combination of a low-loss narrow-band interference filter and a solid-quartz-plate-Fabry-Perot-etalon. Bandwidths as small as $5 \cdot 10^{-3}$ Å were obtained by this simple system increasing the spectral density by a factor of at least $2 \cdot 10^3$.

1. Introduction

For most experimental applications where organic dye lasers are used as light sources, it is necessary to narrow the spectral output which is up to a few hundred Ångstroms wide. Spectral narrowing down to $0.5-1$ Å can be achieved by using a diffraction grating as one of the resonator mirrors [1], a set of prisms [2] or an interference filter inside the laser cavity [3]. Further reduction of the emission bandwidth is possible if an additional dispersive element is introduced into the cavity. BRADLEY et al. [4] obtained a spectral width of the dye laser emission of about 0.01 Å by using a Fabry-Perot interferometer. The same value is reported by WALTHER and HALL [5], who inserted a two stage polarisation interference filter, similar to the device described by LYOT [6], into the cavity.

The spectral resolution of all known dispersion elements is limited by the divergence of the incident light beams. For flashlamp pumped dye lasers, divergences up to a few mrad are possible because the pumplight can produce optical inhomogeneities in the dye solution. Since interference filters have a very small beam divergence sensitivity, they are superior to all other configurations for spectral narrowing [7, 8].

In this paper we want to describe a very simple and reliable dye laser setup which is able to produce laser emission with a bandwidth of less than 0.005 Å. This small spectral width could be reached by using only a two stage wavelength selector consisting of an interference filter and a Fabry-Perot interferometer inside the laser cavity.

2. Construction and Performance of the Wavelength Selector System

A detailed description of the basic concept of the laser system was already given in [9]. The Brewster angled quartz dye cell, 2 inches long, 3 mm diameter, was opticaly pumped by a 2 inch Xenon flashlamp (ILC, 3F2) inside an elliptical pumping resonator. Pump energies as low as 8.5 J produced a $5-10$ mJ laser output of $300-500$ nsec duration.

Rhodamine 6G was used as a dye. The most intensive narrow-band laser emission in the spectral range between 5800 Å and 6000 Å was achieved with a 2.5×10^{-4} M solution of the dye in methanol *.

For most of our experiments we used a plane mirror resonator with a length of 55 cm. The laser mirrors, dielectric multilayers of reflectivity greater than 99.8% and 82% respectively were evaporated on quartz wedge plates of 5 degrees angle. By varying the reflectivity of the partially transparent mirror between 82% and 50%, an increase in laser output power of a factor of 3 was observed without serious change in laser line width.

The spectral width of the broad band emission centered at 5900 Å covered a range of about 60 Å. Without serious loss in output power, the bandwidth could be narrowed down to about 1 Å by inserting an interference filter of the Fabry-Perot type into the cavity. Frequency tuning of the emission over a spectral range of several nanometers was achieved by tilting the filter relative to the axis of the resonator. As the influence of the beam divergence increases with the growing angle of incidence, it is advisable to choose a suitable filter for the desired wavelength, which has to be tilted to less than 10 degrees. This filter was mounted on a turntable, which allowed reproducible positioning to better than 0.001 degrees of arc.

For further narrowing of the oscillation bandwidth an interferometer of the Fabry-Perot type was intro-

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* If water was used as a solvent for the dye instead of methanol, the center of the emission band shifted by nearly 200 Å to the red part of the spectrum. To prevent dimerisation of the rhodamine 6G, it was necessary to add 1.5% Triton X 100 to the solution [10]. Laser output energies were nearly the same for water and methanol solutions, but the spectral width was smaller by a factor of two for the water solutions. This may be mainly due to the lower heating of the water by the flash light. This smaller emission bandwidth should be advantageous for spectral narrowing of the output. Further work concerning this point is in progress.

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duced within the cavity. To avoid stimulation of more than one laser line at pumping energies high above threshold, it was necesary to employ an interference filter whose bandwidth was less than the free spectral range of the Fabry-Perot interferometer.

When this condition was fulfilled the experimental results showed that the laser emission consisted of only one narrow line for all angular positions of the two filter elements. As our laboratories for optical coatings were able to produce interference filters of half-width as low as 6 Å and peak transmission $T = 84\%$, Fabry-Perots with optical mirror separations between 0.3 and 0.4 mm could be used. With such a two stage filter combination, laser emission bandwidths smaller than 10^{-2} Å could be realized.

For our first experiments we used a Fabry-Perot interferometer built up of two quartz plates coated with dielectric mirrors (reflectivity $R \approx 82\%$) separated by a spacer of 0.3 mm length. As the parallel alignment of the mirrors of such an arrangement is rather sensitive to small vibrations and temperature variations, frequent checking of the adjustment was necessary. This procedure was rather wearisome because the Fabry-Perot had to be removed from the laser cavity. Therefore, we tried to develop a sturdy and compact device to overcome these difficulties. A high quality quartz plate (0.25 m thick, 20 mm diameter, optical flatness better than $\lambda/50$, wedge angle less than 5/100″) was choosen as a spacer for the Fabry-Perot. Both sides of the plate were coated with a seven layer dielectric coating of reflectivity about 85%. Extensive measurements with both types of Fabry-Perots showed no essential difference in laser line width and output power between the solid etalon and the air filled gap. Besides the impossibility of mirror misalignment, another advantage of the solid etalon is due to the small temperature coefficient of expansion of quartz. Temperature variations of 0.1 °C of the 250 μm etalon result in a transmission wavelength change as small as 3×10^{-4} Å. Therefore, expensive temperature stabilisation was not necessary. On account of its small dimensions and its rigid construction, this interferometer has proven to be rather suitable for spectral narrowing of dye laser emission. These characteristics should be advantageous especially in the case of N_2-laser pumped dye laser, where short cavities have to be realized. Fine adjustment for scanning was possible within a 3 Å interval by tilting the Fabry-Perot. The mechanics of the turntable allowed reproducible angular positioning of the Fabry-Perot to better than 8 seconds of arc corresponding to a reproducibility of the output wavelength within a 4×10^{-3} Å interval for an angular position of the Fabry-Perot near normal incidence ($\varphi_0 \sim 1°$). Meanwhile we have developed still more sensitive mechanical devices for tilting of the Fabry-Perot whose angular position can be determined to within 1 second of arc, permitting definite wavelength setting within a 5×10^{-4} Å intervall.

3. Experimental Results

a) Laser Output

Results of the laser output power and emission line width measurements are summarized in Table 1. The smaller loss of output power offered by the 6 nm filter compared to the 0.6 nm filter is compensated by the problem of possible multiple line excitation if the laser is tuned. Therefore, the 0.6 nm halfwidth filter is preferable for spectroscopic investigations.

Although the total output power decreased by a factor of 2 or 3 if the interference filter or the combination of the interference filter and the Fabry-Perot were inserted into the cavity, the spectral density of the emission was strongly increased. The emitted intensity per frequency interval of 1 Å was raised by a factor of 21 by insertion of the interference filter alone and by a factor of a least 2000 by insertion of the interference filter and the Fabry-Perot interferometer. This efficiency is comparable to the results obtained by BRADLEY et al. [4] using an echelle grating and a Fabry-Perot etalon. The main advantages of an interference filter are high values of angular dispersion and negligible demands with regard to the adjustment, whereas with the grating used as one of the laser mirrors, accurate orientation and high stability of the rotation axis are necessary.

Table 1. Laser spectral output (Rhodamine 6G, 2.5×10^{-4} M solution in methanol, pump energy 8.5 Joule, reflectivity of the laser mirrors 99.8% and 50%).

Laser cavity filter	Spectral half-width (FWHM) [Å]	Peak power [kW]	Spectral peak power density [kW/Å]
No frequency selective elements in the cavity	61	17.5	0.29
Interference filter (FWHM = 60 Å, peak transmission 96%)	7	11.4	1.63
Interference filter + Fabry-Perot etalon	$< 10^{-2}$	7.6	> 760
Interference filter (FWHM = 6 Å, peak transmission 84%)	1	6.1	6.1
Interference filter + Fabry-Perot etalon	$< 10^{-2}$	5.9	> 590

b) Spectral Bandwidth Measurement

To determine the bandwidth of emission, the laser output was analyzed by a FabryPerot interferometer (reflectivity of the plates 95%, spacer 65 mm), whose fringe system was photographed (Fig. 1). When pumped near the dye laser threshold, excitation of a single longitudinal mode was possible whose emission line width was smaller than 2×10^{-3} Å. This value is the measured width uncorrected for finite resolving power of the analyzing interferometer.

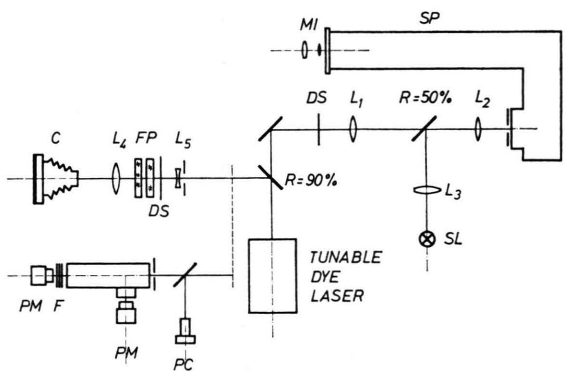

Fig. 1. Schematic diagram of the experimental setup. (MI microscope, SP spectrograph for coarse adjustment, SL sodium vapor lamp, DS diffusing screen, F filter, PC photocell, PM photomultiplier, SAC sodium absorption cell, FP Fabry-Perot interferometer, C camera, $L_1 \ldots L_5$ lenses.)

As expected, the line width increased if the pump energy was raised because more longitudinal modes were above threshold. A typical result, showing simultaneous excitation of two modes, is shown in Figure 2 a **. In the center part of the fringe system, the two modes which have a spectral distance of 3×10^{-3} Å are easily resolved. Investigations with a shorter laser resonator (about 26 cm length) confirm the asignment of the lines to different modes of the laser cavity. Figure 2 b shows the interferogram of three modes with spectral distance 6.5×10^{-3} Å which were simultaneously excited in the shorter cavity.

In Fig. 3 the spectral line width of the dye laser with the interference filter and the Fabry-Perot interferometer as frequency selective elements, is plotted as a function of the pump energy. In all cases in which more than one longitudinal mode is excited, the line width is mainly determined by the mode distance of the laser cavity.

** Figs. 2 a, b see p. 604 a.

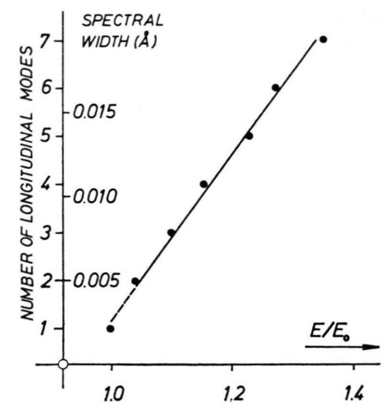

Fig. 3. Spectral linewidth of the dye laser narrowed by a 6 Å interference filter and a Fabry-Perot (0.3 mm spacing of the plates, reflectivity of the mirrors 82%) as a function of pump energy at threshold.

The wavelength stability of the laser emission was tested by measuring the diameters of corresponding fringes in the Fabry-Perot interference patterns photographed for different laser pulses. These measurements demonstrated that the wavelength variations over a time of 20 minutes were less than 2.5×10^{-3} Å. These results were confirmed by absorption and fluorescence experiments with sodium vapor to be described below.

c) Sodium D_2 Line Absorption and Fluorescence

To test the capability of the described laser setup for spectroscopic investigations, the absorption and fluorescence excitation of sodium vapor was studied with the rhodamine 6G laser as a light source. By varying the heating current of the cell, the sodium vapor density could be changed between 2 and 500 ng/cm^3, corresponding to maximum temperature from 145 to 285 °C. The absorption or fluorescence signal caused by the laser beam in the cell was recorded by a photomultiplier shielded with a gray filter and a wide-band interference filter for rejection of laser pump light (Figure 1). To eliminate fluctuations between laser pulses and variations of the output energy as a function of the wavelength tuning, part of the incident light (≈20%) was monitored by a separate photocell. Both signals could be observed on a 50 MHz-dual-beam oscilloscope. Scanning the dye laser emission stepwise across the D_2-resonance line at 5890 Å, the absorption and fluorescence line profiles shown in Fig. 4 were recorded.

In Fig. 4 we plotted the intensity ratio between the light absorbed or emited by the sodium vapor and the laser output intensity as a function of the angular position φ of the Fabry-Perot interferometer inside the cavity. The halfwidth of the recorded line profiles corresponded to a wavelength interval of 0.04 Å. This value agrees very well with the known hyperfine splitting of the D_2-line and the Doppler-width corresponding to the cell temperature. For each measuring point 40 different laser pulses were monitored. The error bars given in the graph are 3 times the standard deviations of the mean value. The deviations from the mean profile are small for all 40 scans across the D_2-line demonstrating the high stability and reproducible tunability of the laser during the entire measuring time of about 1 hour.

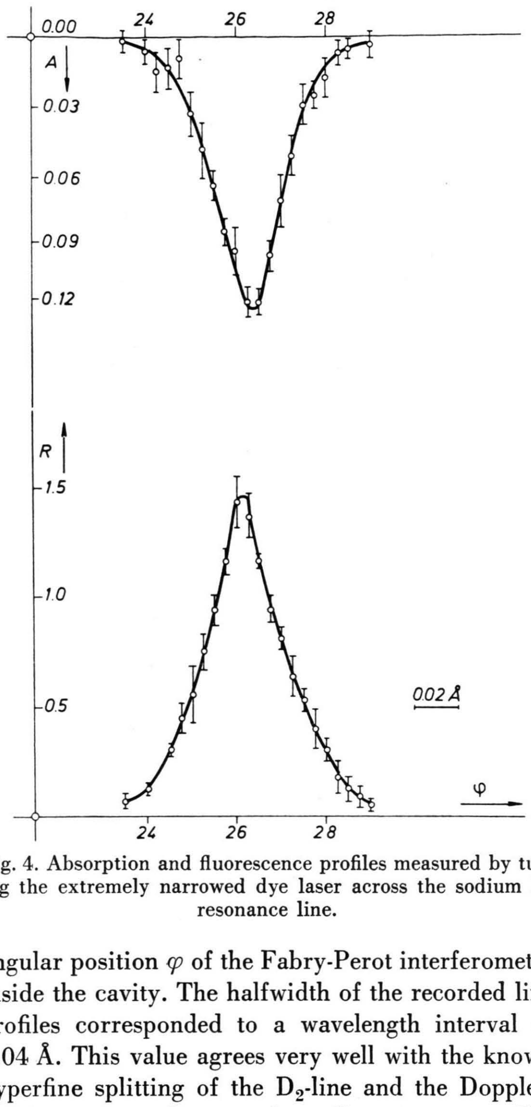

Fig. 4. Absorption and fluorescence profiles measured by tuning the extremely narrowed dye laser across the sodium D_2 resonance line.

A first application of the described laser setup for analytical purposes are reported in [11, 12].

4. Conclusions

The experiments described above have shown that the simple system consisting of the combination of a low-loss, narrow-band interference filter and a solid-quartz plate-Fabry-Perot-etalon is very efficient for spectral narrowing of the dye laser output. Reduction of the emission bandwidth by a factor of nearly 10^{-4} is possible and more than 30% of the total output power can be defined to a spectral width of 10^{-2} Å. On account of its rigid construction very compact wavelength selectors of small dimensions can be realized, which should be interesting especially in the case of N_2-laser pumped dye lasers. First experiments were carried out with rodamine 6G as a dye, meanwhile, similar results were obtained for cresyl violet [13].

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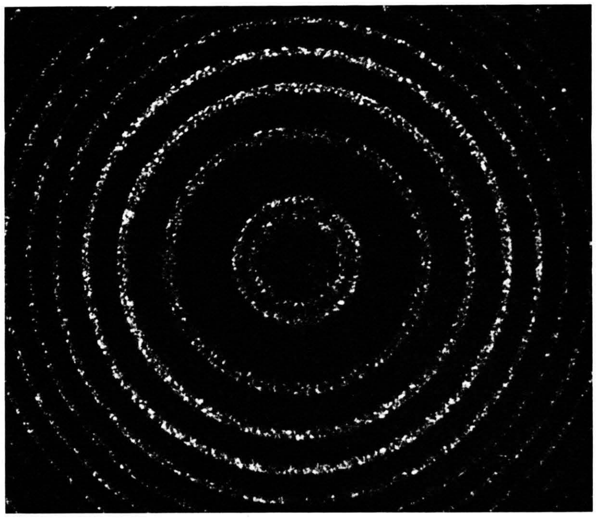

Fig. 2 a

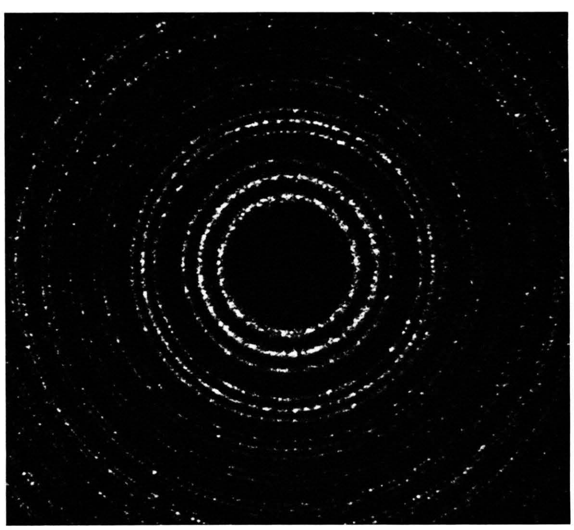

Fig. 2 b

Fig. 2. Longitudinal modes of the dye laser analyzed with a 65 mm Fabry-Perot (free spectral range 0.077 cm^{-1}). a) Two modes simultaneously excited in a 55 cm cavity, mode distance 3×10^{-3} Å. b) Three modes simultaneously excited in a 26 cm cavity, mode distance 6.5×10^{-3} Å.