Characterization of a pulsed rhodamine/ethanol dye laser

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Abstract. We present here the implementation and characterization of a rhodamine dye laser pumped by a pulsed, high-power solid-state laser. The employed dye consisted of a solution of rhodamine 6G in ethanol at different concentrations. To maximize the laser efficiency the second harmonic of a pulsed Nd: YAG was used to collinearly pump the cavity. In particular, the effect of rhodamine concentration and pump energy on the laser emission were investigated. The laser emission obtained was found to have, on average, a width at half maximum (FWHM) of 12 nm. It was found, for example, that the wavelength of the output line varies linearly with the dye's concentration. Under the employed experimental conditions, maximum efficiency of 12% is obtained with a concentration of $2.5 \times 10^{-7}$ mol/l rhodamine in alcohol.

1. Introduction

The dye lasers employ an organic compound as the active medium. When an external pump (a flashlamp or a laser) illuminates the dye it produces population inversion, followed by stimulated emission that, in turn, generates the laser output. These type of lasers are widely used because they can be tuned over a large range of wavelengths. Adding to its versatility, the active medium can be also solid or gaseous. The liquid active form is more convenient as it offers better optical quality and cooling can be easily achieved simply by re-circulating it. Furthermore, contrary to what happens to a solid medium which can be permanently damaged if exposed to excessive irradiation, the liquid medium is self-healing. In principle the dye lasers can offer as much output power as solid-state lasers due to the fact that they have similar density of states. Finally, the cost of the liquid active medium is much less than that of a solid-state laser [1].

In comparison with other types of lasers, dye lasers can easily produce ultra-short time pulses due to its wide emission spectrum. The first attempts to produce ultra-short pulses with a dye laser employed a train of pulse from a mode-locked solid state laser as the pumping source. It has been demonstrated that under some circumstances the optical resonant cavity can be eliminated altogether and the that dye cell can take its place, provided it has parallel faces. The pulse width attained in this way are of the order of 10 to 30 ps [2].

The organic molecules employed in dye lasers have molecular weights within the range 175-830 g/mol. The lasers that use these compounds have a range of emission spanning from the ultraviolet to the near infrared [3]. Figure 1 shows the range of emission of three types of different organic dyes, namely: coumarins, xanthenes and cyanines.
Rhodamine is a family of fluorescent organic compounds based in xanthene and can be considered a derivative of fluorescein. The emission wavelength of a rhodamine-based laser is in the range 570-610 nm. The dye laser is an economic alternative to expensive tuning lasers due to the fact that it can also be tuned to several wavelengths. A dye laser can be used in applications such as: laser-induced fluorescence (LIF) that is widely used to probe molecular structures, investigation of absorption and luminescence in semiconductors and dielectrics. Another possible application is the dye laser resonant absorption photography (DLRAP) in which the expanded, collimated beam of a dye laser is selectively absorbed by the different atomic or molecular species in the plume of an ablation plasma. The latter technique represents the convolution of two different techniques: absorption spectroscopy and shadow photography [4]. Finally, a prototype like the one reported here can be used in experimental teaching of photonics courses at the undergraduate and postgraduate levels [5].

2. Experimental set-up
One of the best configurations employed in dye lasers is the co-linear (or longitudinal) one [6]. Figure 2 shows the schematics of the rhodamine 6G dye laser employed in the present work. Here the pump beam was the second harmonic (532 nm) of a Nd:YAG laser that was separated from its companion fundamental by means of a dichroic mirror (M1, in the figure). The pulsed Nd: YAG laser used was a Continuum Inlite III model with a pulse width of 7ns, maximum energy of 25mJ at 532nm and a repetition rate of 1-10Hz. In the present configuration the pump beam enters the resonator's cell through a narrow-band filter (M2) centered at 532 nm with a bandwidth of 10 nm, that acts as a total-reflection mirror as it reflects almost all of the dye's emission but lets the pump beam pass through. The dye cell employed was made of quartz and had a base of 1×1 cm and height of 4 cm.

By playing with the concentration of the solution and the thickness of the cell's wall it was possible to obtain a low-divergence beam, typically of the order of 3-5 mrad. It was found that it is a good practice to aim the pump beam slightly skewed from normal incidence in order to separate the beams of the pump laser and the dye laser. The resonator employed was of the Fabry-Perot type, without additional tuning elements, made of two reflecting mirrors: one with reflectivity 99% (M2) and the other (M3) with a reflectivity of 20% in the visible range. The separation between the mirrors was kept fixed at 11.6 cm.

The energy of the pump beam was regulated through the delay of the Q-switch of the Nd:YAG laser. To measure the energy of the dye laser's beam a pyroelectric power meter (Spectra-Physics

![Figure 1. Range of emission of three different types of dyes: coumarins, xanthenes and cyanines. The latter can reach wavelengths as far as 1100 nm.](image-url)
model 407A) was placed after the mirror M3, as indicated in Fig. 2. Two beam splitters, BS1 and BS2, deviated part of the light toward two photodiodes, PD1 and PD2, that were employed to measure the time delay between the pump and output beams, respectively. The photodiodes were fitted with neutral-density attenuators to improve their response and their signals were recorded by a 500 MHz digital oscilloscope (Tektronix TDS 3034).

![Experimental set up employed for implementation of the rhodamine dye laser. Key: M1 dichroic mirror, M2 narrow-band filter, M3 partly-reflecting mirror, BS1-BS2 beam splitters, PD1-PD2 photodiodes.](image)

**Figure 2.** Experimental set up employed for implementation of the rhodamine dye laser. Key: M1 dichroic mirror, M2 narrow-band filter, M3 partly-reflecting mirror, BS1-BS2 beam splitters, PD1-PD2 photodiodes.

3. Characterization of the laser emission

The absorption spectra of the dye medium were recorded by means of a miniature spectrometer (Ocean Optics USB4000) coupled to a quartz fibre optics with an internal diameter of 50 μm. Several spectra were recorded in order to map the absorption bands as a function of the solution concentration. Figure 3 shows the spectra obtained for different concentrations of rhodamine in high-purity ethanol. It is evident from Fig. 3 how the larger the rhodamine content is the wider the absorption band is, for example, the half maximum width of the absorption band increases from 80.6 nm for the $5 \times 10^{-6}$ mol/l concentration to 116 nm for the concentration $20 \times 10^{-6}$ mol/l.

![Absorption spectra of the dye solution at different concentrations.](image)

**Figure 3.** Absorption spectra of the dye solution at different concentrations.
It is also evident that the wavelength of the pump laser, 532 nm, is well inside the absorption bands of the concentrations employed. This confirms that the choice of pump beam was the optimal one. In rhodamine solution in alcohol at high concentrations of the solute, dimerization is practically negligible, while at low concentrations it is more significant; and because the dimers formed are not fluorescent, the absorption bands decrease when the concentration decreases [1].

Figure 4-a shows the emission spectra of the dye at different concentrations of the rhodamine/ethanol solution. The figure shows that the concentration has a clear effect on the wavelength of the light emitted: the peak of the wavelength varied from 560 nm for a $0.25\times10^{-6}$ mol/l concentration to 585 nm for the $10\times10^{-6}$ mol/l one, in other words, as the concentration increases so does the wavelength of the emitted light. The shift in the laser emission bands as a function of concentration is due to the change in the absorption bands shown in figure 3, given that the fluorescence spectrum of an organic dye is, practically, the mirror image of its absorption spectrum. In fact, Fig. 4-b shows that this variation of the peak wavelength with the dye concentration is completely linear. The laser emission obtained was found to have, on average, a width at half maximum (FWHM) of 12 nm. The efficiency of the laser was obtained by taking the ratio of the energies of the pump laser and the dye lasers. It was found that for the concentration of $2.5\times10^{-7}$ mol/l the energy efficiency of our laser was 12%. This is to be compared with the efficiency of over 50% reported by Igarashi et al [7] but the simplicity and lower cost of the present implementation should be factored in the comparison.

In a series of tests in which the concentration was kept fixed at $0.25\times10^{-6}$ mol/l, a comparison was made of both the energy of the incoming pump beam and that of the output beam. All the tests were made at a laser frequency of 10 Hz. The results obtained are shown in Figure 5. This figure shows that the output energy does not vary linearly to the pump energy but rather that the output energy fits a polynomial of the pump energy, at least, in the range investigated. The observed output energy values have a dispersion that is not larger than 10% showing that the output beam is very stable.

Another characteristic that was investigated was the relative delay between the pump and the output beams. To implement this analysis a small fraction of both beams was deviated toward the photodiodes by means of beam splitters as shown in Fig. 2. The photodiodes are fast enough (1 ns risetime) to faithfully record the corresponding times. Figure 6 shows that as the pump beam's energy is increased the delay between the two beams decreases. The plot in this figure shows that the delay decreases exponentially with increasing beam energy and reaches a plateau at 15 mJ. The delay is the total time the dye laser takes to absorb the pump beam, to fluoresce and emit the output beam.
Figure 5. Comparison between the pump and output energies. Fixed concentration $2.5 \times 10^{-7}$ mol/l.

Figure 6 shows that at high pump-beam energy (16.5 mJ, for example) the delay between the two beams is shorter (15 ns in this example) than at lower energies; this is to be compared with the 40 ns delay obtained for 1.5 mJ pump energy. Sorokin et al. [8] carried out a detailed investigation on the temporal relationship between the pump and the output beams through a comparison of measured and calculated risetimes for a ruby laser pump and a chloro-aluminum phthalocyanine (CAP) dye laser. They found that the risetime of their dye laser was one tenth of that of the pump, furthermore, that the population inversion rate was directly proportional to the pumping energy.

It is interesting to note that the linear relationship that exists between concentration and wavelength (Fig. 4) is not replicated in the relationship between the pump energy and the delay, the latter is not linear. Also interesting is the asymptotic trend observed at pump energies higher than 17 mJ.

Figure 6. Relative delay between the pump and output beams. Fixed dye concentration, $0.25 \times 10^{-6}$ mol/l.
4. Conclusions
A low cost dye laser was designed and characterized. Even though the present prototype employs an expensive high power pump, cost is nevertheless much less than that of the other instrument that has the same functionality: optical parametric oscillation (OPO) laser. It was found that, under the conditions employed in this work, the optimal dye concentration was $0.25 \times 10^{-6}$ mol/l with an energy efficiency of 12%. One advantage of the present prototype was its great stability, in spite of the fact that no agitation of the liquid was employed. Our design does not require additional tuning elements inside the resonant cavity, due to the fact that the bandwidth of the output pulse for the rhodamine/ethanol solution employed, $\sim 12$ nm, was already very narrow. In this work it was found that the delay between the pump and output beams is steadily reduced as the pump energy is increased but it exhibits an asymptotic behavior for very high energies.

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