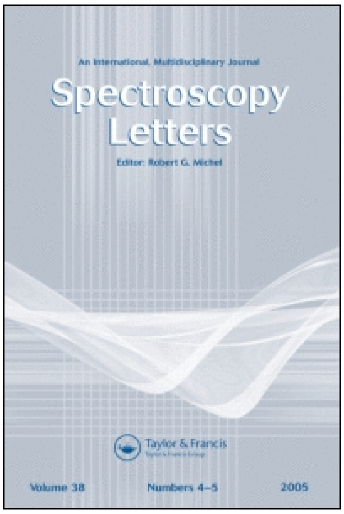

Spectroscopy Letters
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713597299

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To cite this Article Welling, H. , Schröder, H. W. and Wellegehausen, B.(1975) 'Frequency Stability and Linewidth of Single Mode cw Dye Lasers', Spectroscopy Letters, 8: 9, 685 — 695
To link to this Article: DOI: 10.1080/00387017508067373
URL: http://dx.doi.org/10.1080/00387017508067373

SPECTROSCOPY LETTERS, 8(9), 685-695 (1975)

FREQUENCY STABILITY AND LINEWIDTH OF SINGLE MODE CW DYE LASERS

Key words: CW Dye Laser, Frequency Stability, Linewidth

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INTRODUCTION

Dye lasers are as all lasers narrowband light sources, but in addition dye lasers offer the essential feature of being tunable over a wide spectral range. It is a natural consequence that in most applications of the dye laser advantage is taken of the combination of tunability and high monochromaticity. However high monochromaticity in cw dye lasers is more difficult to achieve due to the broad gain profile, the particular guidance of the liquid laser medium in the laser resonator and due to the special requirements of the optical pump mechanism.

The spectral behavior of a laser is determined by fast frequency fluctuations around a mean frequency and by slow drifts of the mean frequency. It can be assumed that the fast frequency fluctuations are of statistical character, which results in an emission line with a Gaussian profile. The linewidth of the Gaussian line will be used to characterize the short term frequency stability. Slow frequency drifts, in general thermally induced, determine the long term frequency stability. Spectroscopic investigations performed with long observation periods have to be made with dye lasers of high long term frequency stability, which can be obtained by aid of an effective temperature stabilization. Highest frequency stabilities can be achieved by frequency locking the dye laser to a stable reference. Stabilization methods are by all means useful but not specific for dye lasers. Since this topic has already been discussed by many authors[1,2], only a short review on stabilization methods already used for cw dye lasers will be given at the end of the article.

Short term frequency disturbing effects specific for dye lasers are primarily caused by an intensive local heat generation in the laser active region mainly due to the optical pump process. The strong heat generation requires a high flow velocity of the active material to avoid optical inhomogeneities. As a direct or indirect consequence of this heat generation and of the high flow velocity, disturbances are observed which overshadow line broadening effects discussed for other types of lasers[3]. This explains why the linewidth of a free running cw dye laser is about two orders of magnitude greater than for comparable gas lasers. The line broadening mechanisms will be discussed in the following section for the cuvette dye laser, where the dye solution flows through a channel with optical windows at the laser active zone. Furtheron in the article line broadening effects for dye lasers with free jet streams will be reviewed.

LINEWIDTH OF CW DYE LASERS WITH DYE CELLS

To estimate the linewidth of a cw dye laser with a dye cell, mechanical instabilities of the laser system and fluctuations of the temperature and the density in the active volume have to be considered. The methods to reduce mechanical disturbances are well known[5] but it should be noted that a reduction of these disturbances for cw dye lasers is difficult compared with gas lasers, because the flow is generally turbulent due to the high flow velocity and can be a large source of mechanical disturbances.

The problem of the thermal fluctuations in the active volume, which may yield a significant contribution to the linewidth, needs special consideration. The sources of temperature fluctuations originate primarily from disturbances of the balance between heating and cooling in the active volume. The heating is caused by radiationless transitions connected with the pump process and the triplet absorption, the cooling takes place predominantly by the flow of the dye solution. Disturbances of the balance result from fluctuations of the dye laser output and fluctuations of the pump power and the flow velocity. These velocity fluctuations arise from the turbulent flow in the dye cell and from pressure fluctuations in the flow system. Thermal fluctuations are of particular importance for cuvette dye lasers, because the velocity has low values close to the window surfaces, which results in a strong heating and consequently in strong temperature fluctuations in this region.

The time dependent frequency deviations $\delta\nu(t)$ caused by temperature fluctuations $\delta T(t,l)$ along the optical path in the active volume are

$$\delta\nu(t) = \frac{\nu_0}{L_0}\int_0^L \left(\frac{\partial n}{\partial t}\right)_{\bar{T}(l)} \delta T(t,l)\, dl \quad , \qquad (1)$$

where ν_0 is the laser frequency, L_0 and L are the resonator length and the length of the active volume and $(\partial n/\partial T)_{\bar{T}(l)}$ is the thermal coefficient of the index of refraction at the mean temperature $\bar{T}$ (l). The linewidths due to the various disturbing effects can be calculated if the temperature fluctuations in Eq. (1) are related to the corresponding disturbing influences. Some calculated values will be given in Table 1.

The thermal effects are reduced by using high flow velocities in the cell, but extreme flow velocities should be avoided, because they generate mechanical disturbances which are difficult to eliminate. In addition turbulent flows introduce density fluctuations, which depend quadratically on the flow velocity. The cooling along the laser active volume depends strongly on the flow velocity distribution. A favourable velocity distribution with only a small area of low velocities near the windows is obtained if the active volume is placed close to the channel entrance[6]. Another possibility to keep the thermal linewidth small, is to reduce the magnitude of the fluctuations, this being particularly important for pressure fluctuations, because they generate simultaneously mechanical disturbances and density fluctuations. Pressure fluctuations can be decreased by an effective buffer system in the flow circuit. Velocity fluctuations are unavoidable in cells

TABLE 1

Estimation of the Influence of Fluctuations on the Linewidth

Fluctuating parameter	Fluctuations (r.m.s.)	Linewidth	
		aqueous solution	methanol solution
Density fluctuations due to pressure fluctuations in the flow circuit	o,o1 atm	o.4 MHz	o.4 MHz
Velocity fluctuations due to pressure fluctuations in the flow circuit (pump power 2 W)	o,o1 atm	o.3 MHz	4 MHz
Direct temperature fluctuations in the flow circuit	o,o1 °C	1.5 MHz	1o MHz
Pump power fluctuations	2o mW	o.6 MHz	8 MHz
Dye laser output power fluctuations	2o mW	1 MHz	13 MHz
Thickness fluctuations of a jet stream due to pressure fluctuations in the flow circuit (Ref. 11).	o.o1 atm	23 MHz	-

with turbulent flow, but by using smoothly polished channel walls care can be taken that no additional turbulence effects occur.

A reduction of the linewidth can also be achieved by using long resonators, because the linewidth is inversely proportional to the resonator length. This method was used by Green et. al.[7].

However in long resonators it is more difficult to guarantee single mode operation. To make this simple method of linewidth reduction more attractive some guidelines shall be given how dispersive elements can be used most effectively[8]. The active volume should be placed close to one of the end reflectors of the resonator to obtain a large frequency difference between the oscillating mode and modes with maximum gain due to the spatial hole burning effect[9]. If Fabry-Pérot-etalons are used as mode selecting elements they should have an optical thickness $e = d/q$ where d is the distance to the near end reflector and q is a small whole number. Under this condition modes with maximum gain due to the spatial hole burning effect are kept away from the transmission maxima of the etalon. A laser with prisms as selective elements should be operated at the edge of the stability range of the resonator[1o] to obtain a high selection power of the prisms.

Table 1 summarizes the influence of several fluctuation terms on the linewidth of a rhodamine 6 G dye laser with an aqueous solution at room temperature and with a methanol solution. (Concentration: $3\cdot 1o^{-4}$ molar; resonator length: o,5 m; pump beam radius: 14 µm; mean flow velocity: 1o m/s; the velocity distribution was taken from Ref. 6, Fig. 3 B). The estimates have been made in a similar way as in Ref. 6, Eq. 7 for the influence of pump power fluctuations. No estimates have been made for turbulent velocity and density fluctuations because of incomplete knowledge about these fluctuations. The smaller values of the linewidth in aqueous solutions can be related in first order to the low thermal coefficient of the index of refraction. A further decrease of the thermal linewidth can be obtained by cooling down the aqueous solution below room temperature, as the thermal coefficient of water depends strongly on the temperature[6]. Smallest measured linewidths (around 1 MHz[6,12]) in cuvette dye lasers were achieved with aqueous solutions.

LINEWIDTH OF JET STREAM DYE LASERS

Dye lasers with a free jet stream as proposed by Runge et. al.[13] avoid any detrimental influence on the laser operation by a window damage and are therefore well suited for high pump power applications[14]. Opposite to the generally turbulent flow in dye cells, jet streams must be laminar in order to achieve a good optical quality. Consequently turbulent velocity and density fluctuations do not arise and mechanical disturbances more easily can be kept small. In addition, the disturbing effect of boundary zones with small velocities is reduced, as these zones are accelerated when leaving the nozzle. Unfortunately a new disturbing effect is introduced, as the jet may show thickness fluctuations caused by pressure fluctuations in the flow system and by surface waves generated by disturbances inside and outside the nozzle.

Calculations and measurements for simple jet stream geometries show[11,15] that the smallest thickness fluctuations can be obtained near the exit of the nozzle, for thin jets, for high viscosity solvents and at optimum flow velocities. The optimum flow velocities depend on the type and quality of the nozzle and increase with increasing viscosity and decreasing jet thickness. The jet thickness should, however, be larger than approximately o,2 mm for rhodamine 6 G, otherwise, to guarantee a reasonable pump light absorption, the concentration of the dye solution has to be so high, that the effect of concentration quenching deteriorates the laser oscillation[16].

Solvents of high viscosity are used to damp strongly surface waves so that even simple nozzles consisting of a tube with razor blades epoxied on one end or a tube pressed to a rectangular cross section at the end are possible[13,17]. Nozzles of this kind, operated with the highly viscous solvent ethyleneglycol, yielded linewidths of several MHz[18]. A nozzle proposed by Wellegehausen et. al. [11] guarantees operation and small linewidth for low viscous solvents too. Linewidths of 9 MHz and 4 MHz for water ammonyx and water ethyleneglycol solutions were obtained, for observation times up to 1 s and a resonator length of o,65 m. It should be mentioned that the effect of pressure fluctuations is an order of magnitude higher than in cuvette dye lasers(see table 1). Therefore special care has to be taken to eliminate pressure fluctuations in the flow system.

FREQUENCY STABILIZATION OF DYE LASERS

The short term stability and in particular the long term stability can considerably be improved by applying active stabilization methods.

Barger et. al.[19] described a stabilization scheme, where the dye laser was frequency-locked to a high finesse Fabry-Pérot cavity. The linewidth was reduced from 5o MHz (free running) to 5o kHz. The long term frequency stability was determined by thermal drifts of the reference cavity (1 to 2 MHz per minute).

A quite sophisticated system for a tunable and stabilized laser was successfully demonstrated by Hall and by Walther[20]. Their frequency locked one dye laser to an atomic reference line. A second dye laser was frequency locked to the first dye laser, but the intermediate frequency in the servo system was variable. This means, that with respect to the absolute frequency of the atomic transition line the second laser could be detuned with the accuracy of the intermediate frequency in the servo system.

CONCLUDING REMARKS

Progress in recent years has remarkably improved the stability of cw dye lasers. Emission linewidths of several MHz for free running dye lasers will please most of the spectroscopists who intend to perform high resolution spectroscopy, where it is desirable to have a probing linewidth of the dye laser below the natural linewidth of the atomic or molecular transition. The question now is, if it is worthwhile to spend more effort in the future to develop dye lasers with higher stabilities. It may be fascinating to use the dye laser as a primary frequency standard in the optical region, because the dye laser, due to its tunability, has the strong advantage, that many atomic or molecular lines are available as reference lines for frequency locking. The position of the dye laser as a promising candidate for an optical frequency standard would be strengthened, if the short term stability would be increased. We are optimistic that by a careful design the basic short term stability of cuvette and jet stream dye lasers can be improved by a factor of ten.

In many present and future applications of the dye laser a small linewidth is not the only critical parameter; rather it will be important to have a combination of several specific properties, as a small linewidth with a wide tunability range in single mode or with a high output power or with a high optical field within the resonator. This makes the linewidth problem more complex. Finally it should be emphasized that considerable effort has to be invested, to extend the frequency range of tunable narrowband dye lasers. The use of new dyes and solvents will not raise principal new questions, however, a wide extension of the frequency range obtained by frequency mixing and doubling in nonlinear optical materials may create a variety of problems to maintain small linewidths.

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