Chapter 8
Studies on thermo-optic characteristics of high repetition rate dye laser

8.1 Introduction

It is well known that a fraction of the absorbed pump radiation by the dye gain medium is converted into heat [8.1], within a confined area, through nonradiative deactivation of molecules and Stokes shift. Thus, a major limitation on achieving stable, spectrally narrow and spatially coherent radiation from high repetition rate dye laser lies in the formation of refractive index gradients due to non-uniform heating [8.2] by the pump radiation in the region of optical gain. These thermal problems accumulate during high repetition rate pumping, and have severe consequences on the yield of the dye laser. Therefore, studies on thermo-optic properties of dye laser are essential, particularly under high repetition rate pumping. In this chapter, theoretical investigation of dye solution temperature distribution in the dye cell and spectral deviation in the presence of thermal field is presented. Dye laser characteristics in the presence of thermal field under high repetition rate excitation are experimentally investigated. Studies on optical characteristic of a high repetition rate dye laser have been carried out by dye solution bulk temperature alteration as well as its stabilization. Mathematical treatment for spectral intensity fluctuations by inhomogeneous medium is outlined.

8.2 Theoretical analysis of thermo-optics of a high repetition rate dye laser

8.2.1 Theoretical analysis

The optical pump beam is absorbed in the dye medium following the exponential absorption. Hence the maximum absorption predominantly lies close to the pump beam entrance window of the dye cell. This results in the change of density and hence changes in the refractive index of the gain medium. Also, a statistical fluctuation
of the pump beam flux induces spatially random fluctuations in temperature of the gain medium. The temperature $T$ at any point in the dye medium is

$$T = \bar{T} + T', \quad (8.1)$$

where $\bar{T}$ is mean temperature and $T'$ is fluctuating component, about the mean value. Therefore, the refractive index of medium also fluctuates accordingly i.e.

$$n = \bar{n} + n' \quad (8.2)$$

where $\bar{n}$ is the mean refractive index, and $n'$ is the fluctuations in $n$ around $\bar{n}$. The variation in $n'$ is much smaller than $\bar{n}$. Therefore, instantaneous refractive index in the presence of thermal field can be approximated as,

$$n = \bar{n} + \frac{dn}{dT} \Delta T \quad (8.3)$$

where $\frac{dn}{dT}$ is the temperature gradient of the refractive index and $\Delta T$ is the temperature fluctuation.

The dye laser characteristics are affected by the refractive index of the materials and physical change in the length of the mechanical mounts, housing the optics. If $n$ is the refractive index of the cavity and $l$ is the geometrical length of the cavity, then optical length $L$ is given by,

$$L = n_a l_a + n_d l_d + n_g l_g \quad (8.4)$$

where $a$, $d$ and $g$ stands for air, dye and glass medium, respectively.

The change in wavelength due to change in refractive index and geometrical length is approximated as

$$\Delta \lambda = \frac{2}{k} (l \Delta n + n \Delta l) \quad (8.5)$$

$$\Rightarrow \Delta \lambda = \frac{2 ml}{k} \left( \frac{\Delta n}{n} + \frac{\Delta l}{l} \right) = \lambda \left( \frac{\Delta n}{n} + \frac{\Delta l}{l} \right) \quad (8.6)$$
The wavelength variation due to change in the refractive index alone is

$$\frac{\Delta \lambda}{\lambda} = \left(\frac{\Delta n}{n}\right), \quad (8.7)$$

$$\Rightarrow \frac{1}{\lambda} \frac{d \lambda}{dT} = \frac{1}{n} \frac{dn}{dT}, \quad (8.8)$$

As

$$\Delta n = \frac{1}{l} \Delta L = \frac{1}{l} \left(l_a \Delta n_a + l_d \Delta n_d + l_g \Delta n_g\right)$$

$$\Rightarrow \frac{1}{\lambda} \frac{d \lambda}{dT} = \frac{1}{L} \left(l_a \frac{dn_a}{dT} + l_d \frac{dn_d}{dT} + l_g \frac{dn_g}{dT}\right) \quad (8.9)$$

$$\Rightarrow \frac{1}{\lambda} \frac{d \lambda}{dT} = \frac{1}{l} \frac{dl}{dT} = \frac{dL}{dT} = \frac{dL}{dT} = \alpha_m \quad (8.10)$$

where $\frac{dn_a}{dT}$, $\frac{dn_d}{dT}$, and $\frac{dn_g}{dT}$ are the index gradient of air, dye and glass medium, respectively. The variation of refractive index of the dye medium, $n_d$ with temperature is identical to that of solvent.

The wavelength variation of the dye laser, due to variation of geometrical length of the cavity is

$$\Delta \lambda \approx \frac{\Delta l}{l} \quad \Rightarrow \frac{1}{\lambda} \frac{d \lambda}{dT} = \frac{1}{l} \frac{dl}{dT} = \alpha_m \quad (8.11)$$

where $\alpha_m$ is the thermal expansion coefficient of the materials involved. Therefore, change in dye laser wavelength can be approximated as

$$\frac{1}{\lambda} \frac{d \lambda}{dT} = \frac{1}{l} \left(l_a \frac{dn_a}{dT} + l_d \frac{dn_d}{dT} + l_g \frac{dn_g}{dT}\right) + \alpha_m \quad (8.12)$$

$$\Rightarrow \Delta \lambda = \frac{1}{\lambda} \left(l_a \frac{dn_a}{dT} + l_d \frac{dn_d}{dT} + l_g \frac{dn_g}{dT}\right) \Delta T \quad (8.13)$$

$$\Rightarrow \Delta \lambda = \lambda \left[\frac{1}{l} \left(l_a \frac{dn_a}{dT} + l_d \frac{dn_d}{dT} + l_g \frac{dn_g}{dT}\right) + \alpha_m\right] \Delta T \quad (8.14)$$
The most sensitive part of the dye laser affected by temperature is the gain medium refractive index [8.1]. Under this approximation, the dye laser wavelength is

\[ \lambda \approx \lambda_0 + \frac{\lambda_0}{n_d} \frac{dn_d}{dT} \Delta T \]  

(8.15)

where \( \lambda_0 \) is the wavelength in the homogeneous medium (i.e. at \( \Delta T = 0 \)). In this way, dye laser output wavelength is affected significantly by the fluctuations in refractive index and temperature gradient of refractive index of the medium.

### 8.2.2 Numerical analysis

The numerical analysis of typical parameters of the dye laser is presented below for spectral variation with temperature. Table 8.1 summarizes the typical parameter of the materials used for the analysis.

**Table 8.1:** Typical parameter of the materials [8.3]

<table>
<thead>
<tr>
<th>Materials parameters</th>
<th>Ethanol</th>
<th>BK-7 glass</th>
<th>Air</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 ( n_d )</td>
<td>1.360</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2 ( \frac{dn_d}{dT} )</td>
<td>-4.38 *10^{-4} /°C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3 ( C_p )</td>
<td>2.438 J/g-C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4 ( \rho )</td>
<td>0.789 g/cm³</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5 ( n_g )</td>
<td>1.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6 ( \frac{dn_g}{dT} )</td>
<td>\approx 1.8*10^{-6} /°C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7 ( n_a )</td>
<td>1.000</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8 ( \frac{dn_a}{dT} )</td>
<td>\approx 1*10^{-6} /°C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9 ( \alpha_m ) (SS)</td>
<td>0.5*10^{-6} /°C</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The change in temperature of the medium can cause the variation in refractive index and consequently changes the dye laser wavelength. The changes in wavelength per
unit wavelength per degree Celsius change in the temperature, for the typically estimated cavity optical path length of 16.0 cm, is

\[
\frac{1}{\lambda} \frac{d\lambda}{dT} = \frac{1}{L} \left( l_a \frac{dn_a}{dT} \right) = -\frac{1}{16} \cdot 2 \cdot 4.38 \cdot 10^{-4} / C = -0.547 \cdot 10^{-4} / C \tag{8.16}
\]

Thus, the change of the dye laser wavelength, at \( \lambda = 576.00 \text{ nm} \), per degree rise in temperature becomes,

\[
\frac{d\lambda}{dT} = 0.0315 \text{ nm/} \degree C \tag{8.17}
\]

And the corresponding frequency change, i.e.

\[
\left[ \frac{d\nu}{dT} \right] = -28 \text{ GHz/} \degree C \tag{8.18}
\]

The variation of the geometrical length of the cavity due to temperature rise can cause the wavelength shift i.e.

\[
\frac{1}{\lambda} \frac{d\lambda}{dT} = \frac{1}{l} \frac{dl}{dT} = \alpha_l = 0.5 \cdot 10^{-6} / \degree C \tag{8.19}
\]

\[
\frac{d\lambda}{dT} = \alpha_l \lambda = 0.0029 \text{ nm/} \degree C \tag{8.20}
\]

And the corresponding frequency change, i.e.

\[
\frac{d\nu}{dT} = -2.62 \text{ GHz/} \degree C \tag{8.21}
\]

The variation of refractive index of air of the cavity can cause variation of the wavelength of dye laser, i.e.

\[
\frac{1}{\lambda} \frac{d\lambda}{dT} = \frac{1}{L} l_a \frac{dn_a}{dT} = \frac{11}{16} \cdot 1 \cdot 10^{-6} / C = 0.69 \cdot 10^{-6} / \degree C \tag{8.22}
\]

\[
\frac{d\lambda}{dT} = 576.0 \cdot 0.69 \cdot 10^{-6} \text{ nm/} C = 0.000397 \text{ nm/} \degree C \tag{8.23}
\]

Corresponding frequency change becomes
\[ \frac{dv}{dT} = -0.36 \text{ GHz/} ^{\circ}\text{C} \]  

(8.24)

Thus, frequency change of the gain medium by the temperature is much larger than the other components of the cavity.

8.3 Dye laser characteristics during gain medium bulk temperature variation

8.3.1 Introduction

The output of dye laser is influenced by the thermal, mechanical and environmental properties [8.1, 8.4]. Duarte and Piper [8.5] noticed that the variation in laboratory temperature by 2-3 \(^{\circ}\text{C}\) due to fluctuations in the external weather conditions shifted the dye laser frequency. Consequently, Duarte [8.6] investigated the behavior of output characteristics of dye laser, particularly output power, beam divergence, bandwidth, and frequency stability, by placing the dye laser cavity in an electrically heated oven and varied the cavity temperature in the 20-35 \(^{\circ}\text{C}\) range. Bernhardt and Rasmussen [8.7] demonstrated the operating characteristics of CVL pumped dye laser and summarized the laser properties in terms of materials (like stainless, invar, methanol-ethanol, water, fused silica, air) thermal properties along with their corresponding frequency changes. The temperature of the dye gain medium is a sensitive parameter that influences the optical properties of the dye laser [8.8-8.9]. Number of solvents such as organic solvents, normal water, and heavy water were used to improve the thermo-optical characteristics of a high repetition rate dye laser [8.1, 8.10-8.12]. In these studies [8.10-8.11], thermo-optic properties such as quantum yield of fluorescence and photo degradation of the dye molecules under high repetition rate excitation by CVL were evaluated. El-Kashef [8.13-8.14] had theoretically and experimentally investigated the dye solution refractive index and its thermo-optic constants extensively, through high precision interferometric technique. Widely differing results on macroscopic and microscopic parameter of the dye solvents were
reported for thermal analysis of dye laser solutions media [8.13]. Amit et al [8.2] have studied the thermal properties of dye laser medium, pumped by CVL (4 kHz pulse repetition frequency). They investigated the temperature gradients generated in the dye cell by CVL, through the transient angular deflection and blooming of a probing He-Ne laser beam after passing throughout the dye medium. They found that the steady temperature gradients extended from 1 to 2 mm in the upstream direction relative to the pump laser impact position to the entire cell length in the downstream direction. The probing He-Ne laser beam experiences about 10-20 mrad angular deflection, depending on the pump laser energy and the dye solution flow velocity.

To the best of our knowledge, literature are deficient in studies on thermo-optic properties of narrow spectral width high repetition rate dye laser for the gain medium temperature above the room/cavity temperature. Therefore, a comprehensive investigation was carried out on the output characteristics such as spectral width, wavelength, average output power, beam divergence and pulse width of a narrow spectral width high repetition rate dye laser, during the dye gain medium bulk temperature alteration from nearly 23-35 °C ranges. The gain medium bulk temperature was altered by supplying additional heat through the dye solution. A PC based data acquisition system for the temperature measurement was developed to record the bulk temperature of the solution at the dye cell. Dye laser Characteristics was investigated by high resolution spectroscopy based Fabry-Perot (FP) etalon and a composite image generation technique. The thermal inhomogeneity in the gain medium was visualized through composite images.

8.3.2 Experimental details

The dye laser used in the experiment consists of an output coupler mirror (20% reflectivity), a dye cell, intra-cavity etalon (FSR 20 GHz, finesse 13) and a grating
(2400 lines/mm) in conjunction with a tuning mirror. The grating at nearly grazing incidence with intra-cavity etalon was used to provide narrow spectral width emission. This is a very convenient design to provide narrow spectral width with minimum number of optical components. The Rh6G dye in ethanol solvent was used as the gain medium. A CVL (λ = 510.6 nm, average power 4 W, 60 ns pulse duration, 5.6 kHz pulse repetition frequency) was used as the optical excitation source. The schematic layout of the dye laser setup is shown in Fig.8.1 (a), while photograph of arrangement of components of the dye laser is shown in Fig.8.1 (b). The pump beam was transversely line focused onto the flowing type dye cell [8.15] through a combination of spherical (focal length, 40 cm) and cylindrical lens (focal length, 6 cm). An image of fringe generated by Fabry-Perot etalon was acquired through an imaging lens and a CCD camera based setup. The output power was measured using USB based power meter (Ophir). The dye laser beam divergences were estimated from width of the far-field intensity distributions.

![Figure 8.1: Schematic of the (a) dye laser layout, (b) photograph of setup](image)

### 8.3.3 Results and discussion

The dye solution bulk temperature was monitored at the dye cell for the investigation of characteristics of dye laser under high repetition rate excitation in the presence of thermal field. The temperature of the dye solution, just at exit of the liquid
through the dye cell, was observed for more than an hour. The gain medium bulk temperature was raised by putting dye solution cooling mechanism off. The heat generated by the mechanical pump motor, associated with the dye circulation system, slowly increase the temperature of the circulating dye solution. It was observed that temperature of the dye solution was raised ~ 0.5 °C per minute and dye solution bulk temperature of 35 °C was attained in nearly 23 minutes. The dye solution bulk temperature was again observed in the presence of gain medium optical excitation by CVL till the temperature attained 35 °C. When the cooling set temperature was switched from 35 to 20 °C, the temperature of the dye solution goes down rapidly. Fig. 8.2 shows variation of the dye solution bulk temperature with time during (a) cooling off, (b) cooling on. In the presence of optical excitation of the gain medium, there is no change in the slope of the temperature rise of the dye solution.

Figure 8.2: Variation of the dye solution temperature with time during (a) cooling off, and (b) cooling on

This is because of the fact that the thermal load added by optical excitation is negligible as compared to the external heat. The dye laser output parameter such as average optical power, divergence, pulse shape, spectral width and wavelength were investigated during the dye solution temperature rise from 23.0 to 35.0 °C and then cooling back to 20.0 °C.
The average power of the dye laser declines monotonically with temperature rise of the dye solution. Fig.8.3 shows variation of dye laser average power with temperature. The decrease in dye laser average power was approximately 3% per degree rise in the dye solution temperature. The observed deterioration in the output average power was about 36% for a 12 °C rise in the dye gain medium bulk temperature [8.16]. Fig.8.4 (a) shows typical dye laser FP fringe at 23.3 °C temperature. The separation and fringe width measurement showed narrow spectral width (1.13 GHz) of the dye laser. A composite image of the fringes were generated during the temperature rise in the 23-35 °C range and cooling from 35.0-20.0 °C. Fig.8.4 (b) shows composite image of the line scan across the diameter of the fringe during temperature alteration. The region A is labeled for the number of fringes taken during the temperature rise while region B for during the cooling down, in the Fig.8.4 (b). The region C, in the Fig.8.4 (b), is the spectral patterns taken at nearly 20 °C, after recovering the medium from large thermal inhomogeneities.

![Figure 8.3: Variation of dye laser average power with temperature](image-url)
Figure 8.4: Typical dye laser (a) FP fringe at 23.3 °C, (b) composite image of the line scan across the diameter of the fringe during temperature alteration

The composite image generated from successive FP fringes, clearly indicates the variation in diameter of the fringes while raising the gain medium bulk temperature. While cooling from 35.0-20.0 °C, the homogeneity of the medium was significantly perturbed, as a result only random noises patterns were visible, denoted as region B in the Fig.8.4 (b). The analysis of the spectral width and wavelength, during the temperature rise of the gain medium, has been carried out. Fig.8.5 (a) shows variation of dye laser spectral width with time, in the temperature 23-35 °C range. The dye laser spectral width fluctuations increase slightly with the enhancement of the medium volume temperature. Fig.8.5 (b) shows variation of dye laser wavelength with number of fringes taken in the temperature 23-35 °C range. The dye laser wavelength changes from 575.6967 to 575.7534 nm, in this temperature range. Apart from the shift, fluctuation of the wavelength was also observed. Fig.8.6 shows the variation of horizontal and vertical dye laser beam divergence with time, in the temperature ranges...
23-35 °C and 35-23 °C. It changes from 6.0 to 8.3 mrad in the horizontal direction, while 1.2 to 1.5 mrad in the vertical direction, with pulse to pulse fluctuations.

Figure 8.5: Typical variation of dye laser (a) spectral width with time, and (b) wavelength with number of fringes taken, in the temperature 23-35 °C range.
Fig. 8.7 shows the variation of the pulse width and total counts under the widths with number of pulses taken during the temperature range 23.0 - 35.0 °C. The dye laser pulse width increases from 34.6 to 36.9 ns. The total counts under the pulse width nearly changes from 8850 to 7650 counts in this temperature range.

**Figure 8.6**: Variation of the dye laser horizontal and vertical divergence with time during the gain medium temperature change

**Figure 8.7**: Typical variation of pulse width and total counts under the pulse width with number of pulses taken during the temperature rise in 23.0-35.0 °C range
A very few studies [8.6, 8.17] reported on effect of temperature variation on dye laser characteristics. Duarte [8.6] had studied the variation of dye laser, pumped by low repetition rate nitrogen laser, outputs such as output peak power, beam divergence, linewidth and frequency stability by varying the cavity temperature in 20.0 - 35.0 °C. Peters and Mathews [8.17] have investigated the low repetition rate nitrogen laser pumped dye laser peak power and divergence angle by cooling the dye solution temperature from 28.0 to 13.0 °C. They found that the dye laser peak power increased while beam divergence decreased during cooling process. Though the effect of temperature on pulsed dye laser is acknowledged, high repetition rate dye laser still needs attention on consequence of gain medium thermal inhomogeneities on the output characteristics. During the present course of investigation, studies on the deterioration in the performance by temperature alteration from 23.0 to 35.0 °C range, while the cavity temperature was kept constant at normal room temperature was carried out. In this way, optical characteristics of a high repetition rate dye laser have been investigated by varying the temperature of a highly sensitive laser element, dye gain medium. Indeed, wavelength of dye laser in presence of temperature-dominated inhomogeneous medium depends on the refractive index and temperature gradient of the refractive index. Shift in the observed dye laser wavelength with bulk temperature rise is a consequence of variation of the spatial refractive index gradient of the gain medium. The increase of the dye medium bulk temperature changes the effective refractive index gradient of the medium, which in turn red shifts the wavelength of the dye laser. The fluctuations in the wavelengths are due to the transient fluctuations in the refractive index gradients. The dye laser spectral width, effectively, replicated the band-pass of the dispersive components of the cavity. This is indeed the reason why there is no appreciable change in the spectral width, as the dispersive elements, grating and
etalon, are distant from the heating zone. Spectral widths were influenced by the pump beam penetration depth fluctuations and by the inhomogeneity of the gain medium. These subsequently cause the variation of geometrical divergence of emission passing through it and hence angle of incidence on grating.

The thermal energy deposited in the dye gain medium by the optical pump beam causes maximum change in density and hence perturbations in refractive index within the thin boundary layers of the medium, next to the pump beam entrance window. As the temperature in the thermo and hydrodynamic boundary layers/sub-layers is much larger than the dye solution bulk temperature, while in this region, the velocity is much smaller whereas the pump intensity is largest. This leads to maximum spatial and temperature gradient of the refractive index in the thin boundary layers of the dye solution. The refractive index gradient in this narrow boundary region is significantly affected by pump power and velocity fluctuations, rather than the dye solution bulk temperature. However, temperature of the solution, around the thermal and hydrodynamic boundary region, tries to adjust the gradients. The divergence characteristics of dye laser emission differ in longitudinal and transverse to the pump beam penetration direction in the gain medium [8.8-8.9]. This is also evident from our experimental measurements, which shows that the dye laser has different beam divergence characteristics in horizontal and vertical directions. The temperature induced inhomogeneities of the medium changes the divergence angles [8.2]. Investigation of the dye laser spectral and divergence characteristics during the bulk temperature rise (23.0-35.0 °C) and sudden cooling of the bulk medium (from 35.0-20.0 °C) clearly showed inhomogeneity generated in the pumping region of the medium. During the cooling, the degree of inhomogeneity was too high and hence unable to measure the spectral variations. These inhomogeneities were also replicated in the dye
laser beam divergence measurements during this range of temperatures variations. The maximum deflection of the laser beam occurs during the cooling, which was also clearly visible by naked eye at the time of experimentation. The decline in average power along the dye laser axis with temperature rise is a consequence of collective effect of increased deflection angles of the beam as well as channelization of energy through nonradiative routes. The dye laser pulse profile depends on the number of round trips, within the available gain time. Inhomogeneities of the gain medium affect the optical path length of the emission propagating through it. The fluctuations in the optical path length by thermal inhomogeneity have an effect on transit time, and hence pulse profile, of the light passing through it. The increased effective path length by refraction/deflection in the dye medium slightly influence the pulse duration during the increased inhomogeneity by the temperature. The counts under the pulse width are a measure of photon flux. The decrease in optical average power with temperature rise is a sign of decreasing photons flux at the detector. Thus, decrease in average power, counts under the pulse width with temperature rise of the bulk medium is an assessment of decrease in quantum yield due to the increase in non-radiative rates as well as fractional photons lost from the cavity. The present observation [8.16] of average power declination and almost no change in bandwidth is well in conformity with the earlier measurements [8.6, 8.17].

8.3.4 Summary

In summary, an effect of dye gain medium bulk temperature on the optical properties of a CVL pumped narrow bandwidth dye laser is investigated. A data acquisition system is developed to record the temperature of the dye solution at the dye cell. The dye gain medium bulk temperature of about 12.0 °C was raised in nearly 23
minutes and cooled down from 35.0-20.0 ºC within 3 minutes. The dye laser spectral width is almost unaffected while wavelength changes from 575.6967 to 575.7434 nm by the temperature variation in 23.0-35.0 ºC range. The pulse width slightly increased, whereas counts under the pulse width decreased by approximately 1200 counts in the 23.0-35.0 ºC range of temperature. The dye laser horizontal beam divergence increased almost by 2.5 mrad, while the vertical beam divergence by 0.3 mrad, for an increase of the dye solution bulk temperature by 12.0 ºC. For these ranges of temperature variations, the dye laser average power declined by approximately 36%.

8.4 Dye laser characteristics during gain medium bulk temperature stabilization

It is well known that the temperature of the dye gain medium brings about considerable changes not only in the dye photo physics but also on the lasing characteristics. In this section, optical characteristics of high repetition rate dye laser over the period of time, by stabilizing dye solution bulk temperature within ±0.1ºC, was investigated. For the completeness, a review of the work reported in literature on the spectral stability of dye lasers by means of temperature stabilization is briefly presented.

In past, studies [8.18-8.19] on the performance of CVL pumped narrow linewidth dye laser, by keeping the dye solution temperature fluctuation within 0.01 ºC at the dye solution reservoir, were reported. The recent studies [8.20] also reports on the beam divergence, pointing stability, linewidth and wavelength stability of CVL pumped narrow linewidth dye laser with [8.22, 8.23], over a few minutes. All these studies [8.18-8.21] have been performed in different perspectives, in terms of dye laser system mechanical structure and the dye solution temperature stability. In another laser system, studies of spectral stability had been reported for a few minutes with the
temperature of dye laser solution controlled within ±0.1 °C [8.22]. In similar narrow linewidth laser system [8.7] where all optical mounts were attached to thick invar base and the dye solution temperature was kept stable to within ±0.1 °C. In all these studies, the temperature was controlled from ±0.01 °C to ±0.1 °C in the dye solution reservoir. Additionally, the dye laser spectral stability was reported [8.21] for few minutes both for coarse control of temperature 23±2 °C and fine control of temperature 23±0.1 °C of the dye solution. It was reported that the linewidth varied between 100 and 770 MHz for a observation period of 30 seconds, with dye solution temperature controlled within 23 ± 2 °C, while linewidth variation was less than equal to 100 MHz for few minutes, with dye solution temperature controlled to 23.0 ± 0.1 °C. In another study [8.22], it was reported that the linewidth variation was from 200 to 300 MHz over a period of 10 minutes, by controlling the dye solution temperature ± 0.1 °C. In this fashion, attention was drawn to the importance of dye solution temperature controlled either ± 0.1 °C or ± 0.01 °C, and significance of thermal stability for better spectral stability of the dye laser. In the present course of investigation, not only the dye laser spectral stability but also average optical power and beam divergence characteristics were investigated, over the long period of time more than hours, by controlling the dye solution bulk temperature ± 0.1 °C. Experimental details and diagnostic techniques were same as described in section 8.3.2. In this investigation, the temperature of the dye solution was controlled within 23.3 ± 0.1 °C by PID controller. The room and cavity temperatures were also stabilized to near 23.3 °C. Fig.8.8 shows variation of cavity and gain medium bulk temperature with time. The temperature of the dye laser cavity changes slightly from 23.4 to 22.8 °C and hence was fairly stable over the period of more than an hour. The gain medium bulk temperature was also steady over the same period.
The dye laser spectral variations were investigated through fringe capturing and composite image generation techniques. For a long-term spectral stability investigation, a composite image of a line scan across the diameter of the FP fringe pattern was generated. Fig.8.9 shows typical Fabry-Perot (a) fringe, (b) composite image of fringes taken during the observation period. The measured spectral width of the dye laser emission was 0.824 GHz. Fig.8.9 (b) shows typical composite image of 1000 successive FP fringes during the observation period of more than an hour. Measurement of dye laser spectral width and wavelength was carried out from this composite image data. Fig.8.10 shows the variation of dye laser spectral width and wavelength with time. The short-term (in 1 minute) fluctuations in the spectral width were within ±75 MHz, while its central value drifted from 0.824 MHz to 1.124 GHz over the observation period of 75 minutes. Fig.8.10 shows variation of dye laser wavelength with time.
Figure 8.9: Typical Fabry-Perot (a) fringe, (b) composite image of the successive fringe

Figure 8.10: Typical variation of bandwidth and wavelength with time

It was analyzed that the short-term fluctuations in wavelength were from 575.7713 to 575.7791 nm. The dye laser wavelength drift were from 575.7791 to 575.7662 nm over the observation period of more than an hour. Further, fluctuation of spectral width at a particular wavelength was analyzed. Fig.8.11 (a) shows variation of dye laser spectral width with wavelength over the period of observation.
Fig. 8.11 (b) shows variation of dye laser average power with time. The measured dye laser output average optical power was 30 mW. The fluctuations of average power about the mean were within 3%.

In conclusion, optical stability of CVL pumped narrow bandwidth dye laser was experimentally investigated over the period for more than an hour. It was observed that the short-term spectral width varies within ±75 MHz, while in a long-term, more than an hour, it was drifted by about 180 MHz. The short-term wavelength fluctuations were within 0.0065 nm, while in a long-term, more than an hour, it was drifted by about 0.0105 nm. Dye laser average power was fairly stable.

8.5 Theoretical studies for dye laser intensity fluctuations

It was observed that dye laser has output fluctuations were present even with very precise temperature control of the dye solution. In this section, attempts were made to correlate microscopic parameter of the medium, responsible for intensity fluctuations.

The propagation of waves through random media is a subject matter that had been of considerable theoretical and practical interest for a long time, as is evident from
the number of books and papers written on the subject [8.23-8.32]. However, most of the works were on the problem of wave propagation and scattering in the atmosphere, the ocean, and in biological media, which are, in general, randomly varying in time and space so that the amplitude and phase of the waves may also fluctuate randomly in time and space.

Jannson et al [8.33] derived the expression for the spectral intensity of the field, produced by scattering of radiation, in the far zone. They showed that the spectral intensity depends on the spatial correlation function of the field and degree of spatial coherence of the incident field. The field generated by scattering of light from a quasi-homogeneous source on a quasi-homogeneous random medium was investigated by Visser et al [8.34]. The far field generated by scattering of a plane monochromatic wave incident on a quasi-homogeneous, random medium, was explained with the help of correlation function of the scattering potential of the random scatterer [8.34]. A theory was developed [8.35] for the fluctuations in the phase and amplitude of a laser beam probing a locally homogeneous and anisotropic medium with the help of correlation functions, which are related to the corresponding stochastic properties of the scattering medium.

Most of the theoretical and experimental works were related to the coherent/incoherent or partially coherent beam propagation through homogeneous, quasi-homogeneous, random and/or turbulent media. All of these models considered propagation and scattering of light through macroscopic gaseous media. To the best of our knowledge, no report is available on the spectral intensity variation by the correlation function of the refractive index fluctuation of the microscopic liquid media, particularly dye emission through inhomogeneous liquid gain medium. In this section, analytical expression for the spectral intensity variation of the radiation scattered by the
correlation function of the refractive index fluctuations of the inhomogeneous liquid medium is derived.

The physical properties of the medium is generally characterized by the dielectric constant $\epsilon$. Thus, from the Maxwell’s equation

$$\text{div} \, \vec{\varepsilon} \, \vec{E} = \varepsilon \, \text{div} \, \vec{E} + \vec{E} \cdot \text{grad} \, \varepsilon = 0 \tag{8.25}$$

We have

$$\text{div} \, \vec{E} = -\frac{\vec{E} \cdot \text{grad} \, \varepsilon}{\varepsilon} = -\vec{E} \cdot \text{grad} \, (\log \varepsilon) \tag{8.24}$$

Using $\varepsilon = n^2$, simplification of Maxwell’s equations gives

$$\nabla^2 \vec{E} + k^2 n^2 \vec{E} + 2 \vec{E} \cdot \text{grad} \left( \vec{E} \cdot \text{grad} \log n \right) = 0 \tag{8.27}$$

where $n$ is refractive index, $k = \frac{\omega}{c}$ the wave number associated with the frequency $\omega$, $c$ is the speed of light in vacuum. The refractive index is expressed as sum of mean and a fluctuating part i.e. the instantaneous refractive index is given by

$$n = \overline{n} + n_i \tag{8.28}$$

where $n_i$ is the deviation of $n$ from its mean value, $\overline{n}$. Therefore,

$$\nabla^2 \vec{E} + k^2 \left( \overline{n} + n_i \right)^2 \vec{E} + 2 \vec{E} \cdot \text{grad} \left( \overline{n} \cdot \text{grad} \log (\overline{n} + n_i) \right) = 0 \tag{8.29}$$

$$\nabla^2 \vec{E} + k^2 \overline{n}^2 \vec{E} = -2 \vec{E} \cdot \text{grad} \left( \overline{n} \cdot \text{grad} \log (\overline{n} + n_i) \right) - k^2 \left( n_i^2 + 2 n_i \overline{n} \right) \overline{E} \tag{8.30}$$

As $|n_i| \ll 1$, it can be approximated that $\left( n_i^2 + 2 n_i \overline{n} \right) = 2 n_i \overline{n}$

$$\text{grad} \left( \log (\overline{n} + n_i) \right) = \text{grad} \left( \log \overline{n} \right) + \text{grad} \left( \log \left( 1 + \frac{n_i}{n} \right) \right) \tag{8.31}$$

$$\text{grad} \left( \log \overline{n} \right) + \text{grad} \left( \log \left( 1 + \frac{n_i}{n} \right) \right) \approx 0 + \text{grad} \left( \frac{n_i}{n} - \frac{1}{2} \left( \frac{n_i}{n} \right)^2 \right) \tag{8.32}$$

This gives an approximation
\[
\text{grad} (\log n) + \text{grad} \left( \log \left( \frac{n_1}{n} \right) \right) = \frac{1}{n} \text{grad} (n_1) \tag{8.33}
\]

\[
\nabla^2 \vec{E} + k^2 n^2 \vec{E} = -\frac{2}{n} \text{grad} \left( \vec{E} \cdot \text{grad} n_1 \right) - 2n_1 k^2 \vec{E} \tag{8.34}
\]

This gives the reduced scalar field equation,

\[
\nabla^2 u + k^2 u = f \left( \vec{r} \right) \tag{8.35}
\]

where \( u \) can denote any of the field components, and \( f \left( \vec{r} \right) \) the scattering potential, which is given by expression

\[
f \left( \vec{r} \right) = -\frac{2}{n} \text{grad} \left( \vec{E} \cdot \text{grad} n_1 \right) - 2n_1 k^2 \vec{E} \tag{8.36}
\]

It well known that the solution of eq. (8.35) corresponding to outgoing is given by

\[
u(r) = -\frac{1}{4\pi} \int \frac{f(r') e^{ik(r-r')}}{r-r'} dV' \tag{8.37}
\]

where \( \vec{r}' \) is a variable vector ranging over the scattering volume \( V \). It is shown in Appendix A that under approximation eq. (8.37) can be transformed to

\[
u(r) = \frac{1}{4\pi} \frac{e^{ikr}}{r} \int f \left( \vec{r}' \right) e^{-ik \vec{m} \cdot \vec{r}'} dV' \tag{8.38}
\]

Therefore, from eq. (8.36) and eq. (8.37), we get

\[
u(r) = \frac{2k^2}{4\pi} \frac{e^{ikr}}{r} \int n_1 \vec{E} \cdot \text{grad} \left( \vec{E} \cdot \text{grad} n_1 \right) \ e^{-ik \vec{m} \cdot \vec{r}'} dV' \tag{8.39}
\]

\[
u(r) = \frac{n_k^2}{2\pi} \frac{e^{ikr}}{r} \int n_1 \vec{E} \ e^{-ik \vec{m} \cdot \vec{r}'} dV' + \frac{k^2}{2\pi} \frac{e^{ikr}}{r} \int \text{grad} \left( \vec{E} \cdot \text{grad} n_1 \right) e^{-ik \vec{m} \cdot \vec{r}'} dV' \tag{8.40}
\]

Using Gauss' theorem, under vanishing surface integral

\[
\text{grad} \left( \vec{E} \cdot \text{grad} n_1 \right) e^{-ik \vec{m} \cdot \vec{r}'} = \left( \vec{E} \cdot \text{grad} n_1 \right) \text{grad} e^{-ik \vec{m} \cdot \vec{r}'} = -i k \vec{m} e^{-ik \vec{m} \cdot \vec{r}'} \tag{8.41}
\]

Therefore,
\[ u(r) = \frac{n k^2}{2\pi} e^{ikr} \int \frac{n_1(r')}{r} \, E^* \, e^{-ik m \cdot r'} \, dV' + \frac{ik m k^2}{2\pi n} e^{ikr} \int \frac{n_1(r')}{r} \, (\vec{E} \cdot \text{grad} n_1) \, e^{-ik m \cdot r'} \, dV' \]  

(8.42)

\[ = \frac{n k^2}{2\pi r} C_1 + \frac{ik m}{2\pi r n} C_2 \]  

(8.43)

where

\[ C_1 = \int n_1(r') \, E^* \, e^{-ik m \cdot r'} \, dV' \]  

(8.44)

and

\[ C_2 = \int \vec{E} \cdot \text{grad} n_1(r') \, e^{-ik m \cdot r'} \, dV' \]  

(8.45)

Both terms of Eq. (8.43) represent spherical waves whose amplitudes and phases depend on the refractive index fluctuations inside the volume \( V \) (through the random variables \( C_1 \) and \( C_2 \)). Indeed, second term can simply be ignored in calculating the flow of scattered energy.

\[ u(r) \equiv \frac{n k^2}{2\pi r} C_1 \]  

(8.46)

The (spectral) intensity of the scattered field at any point \( \vec{r} \), at frequency \( \omega \), is just the diagonal element of the cross–spectral density of the scattered field and hence is given by

\[ I(r, \omega) = \left( \frac{n k^2}{2\pi r} \right)^2 \int \int \left[ n_1(r_1) n_1^*(r_2) \right] \left( E_1 E_2^* \right) e^{-ik m \cdot (\vec{r}_1 - \vec{r}_2)} \, dV_1 \, dV_2^* \]  

(8.47)

If the refractive index \( n(\vec{r}, \omega) \) of the medium is a random function of position, then scattering potential \( F(\vec{r}, \omega) \) will also be a random function of position, and the corresponding expression for the cross-spectral density and for the spectral intensity of the scattered field are obtained at once by averaging over the ensemble of the scattering potential (denoted by angular bracket with subscript n).
Let $C_n(\vec{r}_1', \vec{r}_2') = C_n(\vec{r}_1' - \vec{r}_2')$ denote the two-point spatial correlation function of the refractive index fluctuations, viz.,

$$C_n(r_1, r_2, \omega) = \langle n_1(\vec{r}_1') n_1(\vec{r}_2') \rangle$$  \hspace{1cm} (8.48)

where the angle bracket represent the statistical average, taken over the ensemble $n_1(r, \omega)$ of the refractive index.

Thus, by introducing the change of variables $\vec{r}_1' - \vec{r}_2' = \vec{\rho}$, and $\vec{r}_1' + \vec{r}_2' = 2 \vec{r}$

$$I(r, \omega) = \{\frac{\pi^2}{2 \pi r^2}\}^2 \int C_n(\vec{\rho}) \left| \vec{E} \right|^2 e^{-ik\vec{m} \cdot \vec{\rho}} dV_{\rho} \hspace{1cm} (8.50)$$

This formula shows explicitly how the spectral intensity of the scattered radiation depends on the correlation function of the refractive index fluctuations and on the field passing through it.

For $\vec{E} = A_0 \exp(i \vec{k} \cdot \vec{r})$

$$I(r, \omega) = \{\frac{\pi A_0^2 k^2}{2 \pi r^2}\}^2 \int C_n(\vec{\rho}) e^{i [\vec{k} - \vec{m}] \cdot \vec{\rho}} dV_{\rho} \hspace{1cm} (8.51)$$

The spectral intensity is in fact Fourier transform of correlation function of the refractive index fluctuations. Thus, by knowing the information about correlation function of the refractive index, intensity fluctuations can be approximated. The exact form of refractive index correlation function depends on the characteristics of inhomogeneities. For example the spatial correlation function of the refractive index of the liquid medium, at higher Reynolds numbers, can be approximated in the form

$$C_n(\vec{\rho}) = C_n(0) \exp \left[ -\beta \left( \frac{\rho}{\rho_0} \right)^2 \right] \hspace{1cm} (8.52)$$

where $\beta$ and $\rho_0$ are positive constants, which depends upon the scale length and nature of the medium inhomogeneity.

Though, this analysis applies to a single frequency component of the field, however, it can be extended to multiple or broad bandwidth too.
A spectral intensity fluctuation from a liquid gain medium of a pulsed dye laser is known from long time. The non-uniform changes in the refractive index gradient of the dye solution lead to the refractive index fluctuations. Variation of the spectral intensity of laser emission in liquid media is directly coupled with spatial correlation function of the refractive index fluctuations and can be used to explain the experimental observations of spectral fluctuations in the high repetition rate dye laser.

In summary, a macroscopic theory of propagation and scattering of light through random media can be used under approximation for the microscopic dye liquid media. The spatial correlation function known for a long time is correlated with the refractive index fluctuations of the dye medium. Analytical expression for the spectral intensity of the scattered radiation and the correlation function of the refractive index of the medium is formulated. It shows how the spectral intensity of the field scattered by random medium depends on the correlation function of the refractive index fluctuations. Experimentally observed spectral intensity variation of the fluorescence emission of the dye and the dye laser emission in liquid media in the presence of thermal and flow field is found to be a corollary of scale length of spatial refractive index unsteadiness. Scattering, which is considered to be detrimental to the propagation of the light, of the spectral intensity of laser emission in liquid media is found to be directly coupled to the spatial correlation function of the refractive index fluctuations.

**Appendix A:** Derivation of an approximation relating to expression (8.37).

We derive here the approximation that is applied in expression (8.38), viz.

We choose the origin of coordinates inside the scattering volume. If the observation point \( \vec{r} \) is at a great distance from the scattering volume \( V \) as compared to the
dimensions of \( V \), then for all \( r' \) the quantity \( |r - r'| \) is almost constant and close to \( r = |r| \). In this case, the quantity \( |r - r'| \) can be expanded in a series of powers i.e.

\[
|r - r'| = r - m \cdot r' + \frac{1}{2r} \left[ r'^2 - \left( m \cdot r' \right)^2 \right] + \cdots ,
\]

(A1)

where \( m = \frac{r}{r} \) is a unit vector directed from the origin of coordinates (chosen within the scattering volume) to the observation point. If the inequality

\[
k \left[ \frac{r'^2 - \left( m \cdot r' \right)^2}{2r} \right] \ll 1.
\]

(A2)

holds for all values of \( r' \), i.e. if the dimensions \( L \) of the scattering volume satisfy the condition

\( A r \gg L^2 \), then

\[
\exp \left( ik |r - r'| \right) = \exp \left[ ik \left( r - m \cdot r' \right) \right]
\]

(A3)

Moreover, in the denominator can be replace \( |r - r'| \) by \( r \). Thus, we have in far filed zone

\[
u(r) = \frac{1}{4\pi} \frac{e^{ikr}}{r} \int_{V} f(r') e^{-ikm \cdot r'} dV'
\]

(A4)

**Publications based on this chapter**

1. Studies on thermo-optic characteristics of a high repetition rate dye laser
   **Nageshwar Singh**, R. Jain, S. K. Dixit, and H. S. Vora

2. Spectral intensity variation by the correlation function of refractive index fluctuations of the liquid medium
   **Nageshwar Singh**