Effect of liquid flow on spectral properties of a dye laser pumped by a copper vapor laser

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Abstract: A dye laser pumped by a high repetition rate copper vapor laser (CVL) shows flow-induced variations in wavelength and bandwidth. A brief description of bandwidth deviations in a flowing rhodamine 6G dye laser, transversely pumped by CVL, is presented.

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Tunable lasers based on organic dyes have various applications such as resonance ionization mass spectroscopy (RIMS) [1, 2], atomic vapor laser isotope separation (AVLIS) [3-5], ultrafast pulse amplification [6], medical applications [6-8], and more. For these purposes, various types of dye laser sources have been developed and a number of critical parameters affecting their performance have been addressed [7]. Absorption of the pump beams in the dye solution and indirect heating, such as residual absorption in the dye cell windows, cause lack of thermal uniformity [7, 9, 10] in the medium. The amount of heat deposited inside the medium dissipates through conduction and convection, to maintain the thermal equilibrium. Non-uniform heating creates temperature gradients, which in turn produce appreciable refractive index gradients in the medium. This is known as thermal lensing and results in divergence or blooming of the output beam. Further, increase in the environmental temperature can also induce increases in beam divergence, decrease in output power, and shifting of the resonant laser frequency [11, 12]. In some cases, this may lead to early termination of the dye laser pulse particularly in high intensity or high frequency pulsed operations. Therefore, temperature control is essential for stable and efficient operation of dye lasers. Replacing the dye solution, using dye circulation systems incorporating a heat exchanger, minimize the thermal effects [6, 7]. Constricting the flow path in the dye cell across the pumping region increases the fluid velocity in the region of the dye laser axis with the objective to replace the dye molecules between successive pump laser pulses [5, 13 –17]. This leads to maximum velocity fluctuations in the thin boundary layers of the dye solution at the front window of the dye cell rather than at the center of the dye cell [18]. In this region the velocity is small where as the pump power and velocity fluctuations are large which seriously affects the divergence and optical path length of the laser radiation passing through it. Therefore, the thermal gradients in the active dye medium in conjunction with the dye flow affects the optical homogeneity of the active medium, which in turn affects the beam divergence, laser linewidth [13] and induces dynamic linewidth instabilities [19, 20]. In this paper, a succinct description of linewidth variations in a rhodamine 6G dye laser, transversely pumped by a high repetition rate CVL, in the presence of gain medium flow, is presented.

It is well known that the environmental temperature modify the output power, beam divergence, linewidth, and frequency stability of the dye lasers [12]. The dye laser output power declines in a monotonous manner, pulse duration decreased, and beam divergence increases, by increasing the cavity temperature [12]. The environmental temperature changes the axial mode separation and hence induces shifts in the output wavelength [10, 12]. The change in cavity temperature can causes a wavelength shifts of 4.5 GHz (0.0045 nm) per degree C. This is attributed to variations in the physical disposition of optical cavity components, especially the grating, and the output coupler mirror.

In pulsed dye lasers the temperature variations are local and very fast [7, 21], resulting in non – uniformity in the temperature distribution at the gain medium thus leading to refractive index gradients. Pulse to pulse and intra pulse variation of pump beam fluxes, due to evolving intensity, and divergence, can also cause fluctuations in the laser bandwidth. Therefore, pulsed dye lasers suffer from pulse to pulse variations in wavelength and bandwidth. The output characteristics of a high repetition rate pulsed dye laser has both short-term fluctuations and long-term drift. Long-term drifts are due to changes in the environmental temperature of the resonator and changes in the optical path length due to mechanical displacements. Deviations of the wavelength and bandwidth also depends on the pump beam induced temperature gradients [7, 13], shift in relative positions of optical mounts [7, 13] and characteristics of the pump beam [22].

A stable narrow linewidth dye laser, pumped by a high repetition rate copper vapor laser, requires control of the dye flow, apart from temperature control, through the dye cell to eliminate thermal heating and flow turbulence in the active volume. The dye cell geometry, which stabilizes the flow, also significantly affects the stability of dye laser [23]. The microscopic velocity fluctuations and thermal gradients near the solid liquid interface, through the laser active zone, are affected by dye flow rates. Readjustment of dye flow often results in the appearance of a second cavity mode in the dye laser output, greatly increased frequency jitter (leading to increased effective linewidth), and significant intensity fluctuations. Linewidth instabilities are diminished considerably by reducing the turbulences in the medium. Therefore, inhomogeneity in the gain medium induced by the dye flow affects the linewidth and its stability. No instability in linewidth was found in the case of TEA CO_2 laser oscillators [24], where the lasing medium is homogeneous. On the other hand, in a dye laser pumped by a high repetition rate CVL, a highly stable narrow linewidth operation could only be achieved with an accurate and precise control of flow of the dye through dye cell [13]. For a relatively short duration pump pulse like that from a CVL, the fluid can be assumed to be stationary during the excitation. The refractive index of the medium, which is temperature dependent, changes due to absorption of the pump beam during the pulse. These changes result in optical path length changes in the resonator. In addition, the refractive index non-uniformities of the dye medium, due to the flow, lead to increased losses, and changes in the divergence of the beam passing through it, thus affecting the laser linewidth.

The flow of the dye solution through the dye laser axis has a strong influence on the stability of the output characteristics of a dye laser transversely pumped by a high repetition rate CVL [25]. The Reynolds number (R), which characterizes the liquid flow, is proportional to the average flow velocity, the hydraulic diameter and inverse of the kinematic viscosity. In our earlier study on stability of wavelength and linewidth, the flows of the dye were kept at Reynolds number of 221, 681, and 1012. The dye laser output consists of three axial modes, in which two side modes also compete for gain hence its intensity fluctuates. Out of the three modes, the central mode is relatively stable over a fairly long time [25]. The linewidth, wavelength and its fluctuation are large at R = 221 and decrease slightly at R = 681. There was also a fast variation in the separation of the modes, which makes it difficult to distinguish them at low flow rate. The individual modes that appear indistinguishable at low flow rate are clearly distinguishable at higher flow rates. The number of modes and mode separation varies from shot to shot, depending on the strength of instantaneous fluctuations present in the medium. Fig. 1 shows the typical Fabry–Pérot interference of the dye laser beam at a Reynolds number of 1012. Fig. 2 shows the variation in linewidth with Reynolds number. Incresed flow of the dye solution decreases the linewidth and its fluctuations. Fig. 3 shows the variation of $\Delta\lambda/\lambda$ with Reynolds number. The vertical bar shows fluctuations over the observation period from the mean value. Both $\Delta\lambda$ and $\Delta\lambda/\lambda$ decrease with flow of the dye solution in an experiment where no change except the flow of the dye solution takes place. A linear fit of the observed linewidth gives a correlation coefficient of 0.95048, and a standard deviation of 1.74236 whereas $\Delta\lambda/\lambda$ has a correlation coefficient of 0.93814 and a standard deviation of 1.94991. Also the slop of the variations of $\Delta\lambda$ and $\Delta\lambda/\lambda$ with flow is different. These descriptions suggest that dye flow not only affects the linewidth but also the wavelength of dye laser.

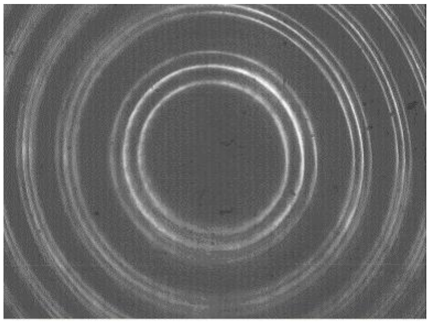

Fig. 1. Fabry-Pérot interferergram of the laser emission at R = 1012.

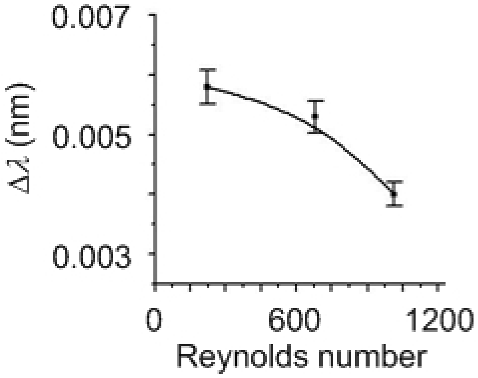

Fig. 2. Laser linewidth $\Delta\lambda$ as a function of R.

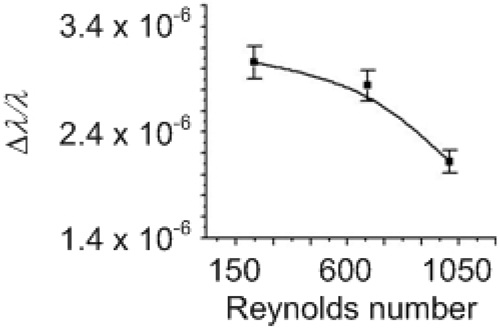

Fig. 3. $\Delta\lambda/\lambda$ as a function of R.

The linewidth of a prism–grating–mirror system of a tunable dye laser is given by [10, 26]

$$\Delta\lambda = \frac{2^{3/2}\lambda}{\pi w}\left[M\left(\frac{d\alpha}{d\lambda}\right)_G + \left(\frac{d\theta}{d\lambda}\right)_P\right]^{-1} \tag{1}$$

where, the terms in the bracket are the dispersion due to the grating (G) and the prisms (P), respectively, λ the wavelength, α the angles of incidence at the grating, θ angle incidence at the prism expander, M is the prismatic beam magnification, and w is the beam waist. This band pass analysis for a grazing incidence grating (GIG) configuration has been used to study the variations in dye laser linewidth. Fig. 4 shows the variation of linewidth with w. The linewidth sharply decreases in the range of 40–100 μm beam size, which is the typical beam size observed in dye lasers. Slight variation in beam size in the gain medium results in a significant change in the linewidth. Fig. 5 shows the variation of $\Delta\lambda / \lambda$ with beam size. The linewidth $\Delta\lambda$ and $\Delta\lambda / \lambda$ illustrate a similar trend of change with both Reynolds number and beam size. However, the variation is more in case of $\Delta\lambda / \lambda$. This behavior can be attributed to the change in wavelength.

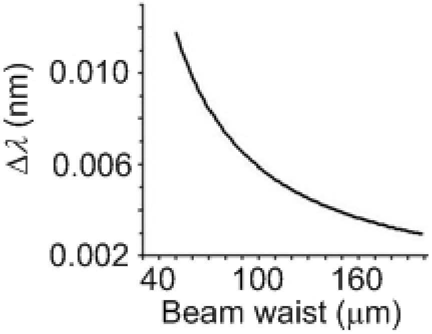

Fig. 4. Laser linewidth $\Delta\lambda$ as a function of w.

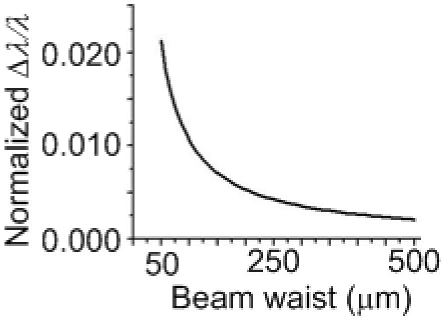

Fig. 5. Normalized $\Delta\lambda/\lambda$ as a function of w.

The bandwidth $\Delta\lambda$ and ratio $\Delta\lambda / \lambda$ vary with both Reynolds number and estimated beam waist. Bandwidth is a function of beam size, intracavity dispersion, and divergence of the emitted radiation whereas the wavelength depends on the optical path length of the resonator. Variations in bandwidth are due to changes in the divergence of the radiation. Wavelength variations are due to changes in optical path length resulting from non-uniform refractive index variation in the active medium. When the flow is increased, the dye medium across the optical axis becomes more uniform and consequently the fluctuations decrease. Initially, the dye flow causes the enhancement of homogeneity of the medium, which in turn reduces the divergence and optical path length. At low flow rate the vortices created near the solid–liquid interface, due to large local variations in fluid velocity, are confined very close to the boundary and diffuse rapidly away into the main body of the fluid. There is sufficient time for these vortices to diffuse out of the thin region, near the solid surface, where they are produced, and to grow into a large region of vortices. These vortices are insufficient to create appreciable turbulence rather they diminish the thermally induced inhomogeneity by diffusion. This will help in reducing the thermal gradients thus optically homogenizing the medium. However, at a high flow rate, the time of diffusion for vortices into a larger region of the fluid are less and they begin to fill in a thin band. In this band the flow is chaotic and irregular, which significantly modify the radiation passing through it [27]. The dye laser axis lies predominantly close to the solid–liquid surface (i.e. near the pump beam entrance window). Therefore, the flow-induced turbulence disturbs the wavelength stability and increases the bandwidth at higher Reynolds number. At a high flow rate (beyond a Reynolds number of 5000) the length scale of turbulences or vortices by cavitations dominated in the medium, which cause a deviation in dye laser linewidth [28]. Therefore, the behavior of a dye laser at low and high flow rate is dissimilar in nature. There could be a critical flow rate above and below which the behavior of the dye laser is different. The thermal and flow-induced refractive index inhomogeneity [29] affects the bandwidth and wavelength of the dye lasers. In other words, the optical inhomogeneities in the gain medium, in addition to the resonator parameters, affect the output spectral properties of the dye laser.

In summary: the inclusion of Reynolds number in the gain medium transform the equilibrium conditions introduced by the resonator and a thermal and flow analyses essentially enlighten the nature of the physical mechanism and length scale responsible for bandwidth changes and short-term wavelength fluctuations in the dye laser.

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References

1. R. V. Ambartzumian and V. S. Letokhov, *Appl. Opt.* **11**, 354 (1972).
2. V. S. Letokhov, *Laser Photoionization Spectroscopy* (Academic, New York, 1987).
3. J. A. Paisner, *Appl. Phys. B* **46**, 253 (1988).
4. J. L. Emmitt, W. F. Krupke, and J. I. Davis, *IEEE J. Quantum Electron.* **QE-20**, 591 (1984).
5. I. L. Bass, R. E. Bonanno, R. P. Hackel, and P. R. Hammond, *Appl. Opt.* **31**, 6993 (1992).
6. C. E. Webb, in *High -Power Dye Lasers*, F. J. Duarte, ed. (Springer, Berlin, 1991) Chapter 5.
7. F. J. Duarte, in *Dye Laser Principles*, F. J. Duarte and L. W. Hillman, eds. (Academic, New York, 1990) Chapter 6.
8. L. Goldman, in *Dye Laser Principles*, F J Duarte and L W Hillman, eds. (Academic, New York, 1990) Chapter 10.
9. M. Amit, G. Bialolenker, D. Levron and Z. Burshtein, *J. Appl. Phys.* **63**, 1293 (1988).
10. F. J Duarte, in *High Power Dye Laser*, F J Duarte, ed. (Springer, New York, 1991) Chapter 2.
11. D. W. Peters and C. W. Mathews, *Appl. Opt.* **19** , 4131 (1980).
12. F. J. Duarte, *IEEE J. Quantum Electron.* **QE-19**, 1345 (1983).
13. F. J. Duarte and J A Piper, *Appl. Opt.* **23**, 1391 (1984).
14. A. N. Zherikin, V. S. Letokhov, V. I. Mishin, V. P. Belyaev, A. N. Evtyunin, and M. A. Lesnoĭ, *Sov. J. Quantum Electron.* **11**, 806 (1981).
15. S. Lavi, M. Amit, G. Bialolanker, E. Miron, and L. A. Levin, *Appl. Opt.* **24**, 1905 (1985).
16. R. S. Hargrove and T. Kan, *IEEE J. Quantum Electron.* **QE-16**, 1108 (1980).
17. M. Broyer, J. Chevaleyre, G. Delacretaz and L. Wöste, *Appl. Phys. B* **35**, 31 (1984).
18. H. W. Schröder, H. Welling and B. Wellegehausen, *Appl. Phys.* **1**, 343 (1973).
19. F. J. Duarte, J. J. Ehrlich, S. P. Patterson, S. D. Russel, J. E. Adams, *Appl. Opt.* 27, 843 (1988).
20. F. J. Duarte and R. W. Conrad, *Appl. Opt.* **26**, 2567 (1987).
21. M. E. Lusty and M H Dunn, *Appl. Phys. B* **44**, 193 (1987).
22. O. Prakash, S. K. Dixit, R. Jain, S. V. Nakhe, and R Bhatnagar, *Opt. Commun.* **176**, 177 (2000).
23. N. Singh, *J. Phys. D: Appl. Phys.* **39,** 2084 (2006).
24. F. J. Duarte, *Appl. Opt.* **24**, 1244 (1985).
25. N. Singh and H. S. Vora, *Appl. Phys. B* **82**, 71 (2005).
26. F. J. Duarte and J. A. Piper, *Appl. Opt.* **20**, 2113 (1981).
27. T. H. Chyba, E. C. Gage, R. Ghosh, P. Lett, and L. Mandel, *Opt. Lett.* **12**, 422 (1987).
28. Y. Maruyama, M. Kato, A. Sugiyama, and T. Arisawa, *Opt. Commun.* **81**, 67 (1991).
29. N. Singh, *Opt. Eng.* **45**, 104204 (2006).