SYNOPSIS

There are many applications where broadly tunable pulsed coherent radiation, having narrow bandwidth and high average optical powers are needed. Solid-state dye lasers are emerging as compact tunable light sources, however, so far best performance is limited to low pulse-repetition-frequency operation while high repetition rate operation still eludes operational stability. These days, optical parametric oscillators (OPOs) are emerging as compact, state-of-the-art all-solid-state, tunable light sources, alternative to the conventional organic dye laser. However, the delicate requirement of phase matching, system complexity and thermal issues at high pulse repetition rate operation limit their application potential. Thus, in present scenario, dye laser based on organic dye molecules in liquid solvent as gain medium, is the most comprehensively utilized pulsed tunable laser. Continuous wavelength tunability, efficient narrow bandwidth operation, wide spectral coverage, and simplicity have made a dye laser an indispensable tool in many areas of laser research, scientific, industrial and medical applications. Narrow bandwidth high repetition rate (~ kHz) dye lasers, tunable in the visible region of spectrum, are exclusively used in application like atomic vapor laser isotope separation (AVLIS) scheme. Dye laser pumped by copper vapour laser (CVL), operating typically in the range of 5-10 kHz in the visible region, capable of delivering high average power, is extremely appropriate for this purpose. This thesis work is a study on CVL pumped dye laser system.

In a dye laser, since absorption of the pump beam radiation follow the exponential law, the maximum absorption predominantly lies close to the pump beam entrance window of the dye cell. A fraction of the pump radiation is converted into heat, within a confined area (i.e. in the pumped region) through non-radiative processes. The thermal energy deposited causes change in density, as a result, the dye
laser gain medium suffers from refractive index nonuniformities which leads to refraction/deflection of the dye laser beam passing through it. These thermal effects degrade the optical quality of the medium, reduce the output energy, and change the spectral characteristics. Inhomogeneity in the gain medium created by thermal field is severe, particularly for excitation at high pulse repetition frequency, if the dye medium is static. The simplest technique to minimize these thermal effects in the gain medium and to improve the optical quality of the medium is to rapidly exchange the irradiated dye molecules exposed to the pump laser, before the arrival of the next pulse. Replacement of the dye molecules is easily implemented by flowing of the dye gain medium. The mass flow rate is chosen so as to replace the optically pumped dye molecules after each excitation pulse. Thus, a high repetition rate dye laser requires large flow velocity across the optical pump region. A large liquid flow velocity can be accomplished by linearly constricting the dye cell cross-sectional area, near the dye laser axis. The flow related issues such as dye cell geometry, stagnant boundary layers, flow velocity profiles, cavitations, eddies etc. degrade the optical quality of the lasing medium. Thus, the issues related to the dye cell geometry and the dye gain medium flows are of the main concern for a high repetition rate dye laser. These aspects have not been paid sufficient attention in the past. Thus, a need was felt to design dye cell and studies spectral stability of high repetition rate dye laser. The research work presented in this thesis is primarily directed to extend the understanding of narrow bandwidth high repetition rate dye laser and thereby developing technology to improve its spectral stability. The thesis is organized in nine chapters. In almost all the chapters, after brief review of the topic under investigation, experimental data generated during the course of investigation is presented at length, followed by discussion of results and summary.
Chapter 1: Review on high repetition rate dye laser: Gain medium

Chapter 1 begins with an overview of dye lasers, significance of high repetition rate dye lasers, and issues related to high repetition rate dye lasers. The characteristics of a dye laser gain medium are described. The physics of dye laser gain medium which deals with the physical properties of the laser dyes, and important classes (families) of laser dyes that are widely used for laser application, are presented. Spectroscopic properties related to the energy levels, and the possible absorption and emission processes that occur in the typical laser dyes and which are of importance in dye laser action, are discussed. Photo-physical properties of Rhodamine 6G (Rh6G) dye, which is used as the gain medium for the high repetition rate dye laser, are briefly described. A review of the environmental factors such as solvent, concentration and temperature effects on the absorption characteristics of Rh6G dye is presented. These studies help in understanding of the photo-physics of the Rh6G dye laser.

The subsequent chapters describe the studies on the investigated aspects of a high repetition rate dye laser such as experimental methods and diagnostic tools for analysis of the output of the dye laser, characteristics of high repetition rate excitation source, pulsed dye laser resonator, dye circulation system, dye cell, gain medium flow, and finally results of the experiments performed, after putting together these components.

Chapter 2: Diagnostic techniques for high repetition rate dye laser

Chapter 2 covers the diagnostic tools for data visualization/representation and formulation of the experimental technique for dye laser spectral characteristics investigation. A novel method for dye laser output data presentation was proposed and realized. The composite image generation technique, for representing the spectral characteristics of dye laser, was used for the real-time analysis of the dye laser
performances. Methodical formulation of the measurement technique for spectral parameters such as wavelength, bandwidth and mode structure etc., were put into practice for dye laser spectral investigations. A diagnostic technique was established for the analysis of the spectral distribution of a high repetition rate dye laser by deriving explicit relationship of the parameters with ring diameter of the Fabry-Perot interference pattern and reference He-Ne laser. An expression for the number of axial modes present and their separation, by knowing the diameter of Fabry-Perot pattern, was realized. The experimental technique for spectral description of the dye laser beam is also discussed in this chapter. The spectral contents and the performance of the dye laser were investigated using high resolution spectroscopy based on Fabry-Perot interferometric technique and composite image generation software. All major instruments involved for the dye laser diagnostics are briefly described.

Chapter 3: Review and studies on Copper Vapour Laser

Chapter 3 gives an overview of CVL, the high repetition rate excitation source for the dye laser. CVL is used as efficient excitation source for tunable dye lasers. Therefore, knowledge of fundamentals of this laser is an essential before its practical application for exciting the dye laser. The characteristics of CVL are reviewed. It covers spectroscopy of a copper atom, energy levels and transition mechanisms, kinetics of CVL, design, assembly details and operating characteristics of CVL. This chapter also covers the theoretical and experimental work done during the present course of investigations on spectral structure of CVL and the effect of electrical input power and buffer gas pressures on the spectral width. Other relevant CVL data such as laser power, pulse shape, etc. are also presented.
Chapter 4: Studies on pulsed dye laser resonator

Chapter 4 of the thesis focuses on the spectral narrowing in a pulsed dye laser resonator. A narrowband dye laser resonator is theoretically and numerically investigated. The dye laser resonator, typically, consists of a partially transmitting output coupler and dispersive optical elements. For a short pulse dye laser, the spectral width of the emission depends mainly on the passive bandwidth of the resonator. Narrow spectral emission through a pulsed dye laser is usually achieved by incorporating a diffraction grating and a beam expander. A prism is used as a compact one-dimensional intra-cavity beam expander. The design parameters of the prism beam expander, and the dispersion of the grating in grazing incidence configuration, are numerically investigated. The dye laser dispersive resonator was simplified to a two-mirror cavity and the transmission coefficient of the partially transmitting output coupler for optimum output dye laser power was numerically and experimentally investigated. A novel technique has been proposed for single mode operation of the dye laser and is experimentally demonstrated.

Chapter 5: Studies on high repetition rate dye laser subsystem

Chapter 5 describes the studies on high repetition rate dye laser subsystems. High repetition rate dye laser subsystems, typically consist of a dye circulation and cooling system for minimization of thermal effects, dye cell for containing gain medium, and a system for characterizing the mass flow through the dye cell. The dye circulation and cooling system was especially designed, fabricated, and employed for the temperature stabilization of the dye gain medium. The mechanical pump, which is one of the key components in any dye solution circulation system, inherently produces vibrations which are transmitted in the flow loop system and hence around dye laser axis. The frequency of the vibrations produced by the dye circulating mechanical pump
was studied. This subsystem plays an important part towards stability of the dye laser. The dye cell used to contain the gain medium is one of the crucial components of the dye laser. The dye cell aspect in the dye laser system is explored in this chapter. A dye cell of rectangular straight channel in the pump region has been conventionally used for high repetition rate dye laser to replace the dye solution between successive pump laser pulses. Therefore, one needs a suitable dye cell with smooth liquid flow in the optical pump region. Several dye cells were designed, some of them were modeled and flow characteristics are numerically investigated.

**Chapter 6: Studies on spectral stability of a high repetition rate dye laser**

Spectral stability of a tunable dye laser is of vital concern in spectroscopic applications. In **Chapter 6**, the spectral stability of CVL pumped dye laser using different dye cells and as a function of Reynolds number of the flow, has been experimentally investigated. Influence of the dye cell geometries and mass flow rates on the stability of a high repetition rate dye laser have not paid much attention so far. During the course of our studies, it was evident that dye solution flow channel and mass flow rates significantly affect the dye laser performance. The spectral stability (wavelength, bandwidth) of dye laser using conventional straight channel, curved (convex-concave), pinched, and spatially profiled convex-plano dye cell in narrow bandwidth resonator were investigated. The methodology adopted for the investigation is spectral stability of the dye laser outputs. It is demonstrated that the geometries of the dye cell as well as mass flow rates significantly affect the spectral stability of a high repetition rate dye laser. Computational analysis of the dye laser gain medium flow has been carried out to explain the observed trends on spectral fluctuations as a function of Reynolds number.
Chapter 7: Studies on fluorescence of Rhodamine 6G dye under high repetition rate excitation

Investigation of the spectral properties of dyes under high repetition rate excitation is of fundamental importance in the utilization of dye as a stable and good optical quality amplifying medium. In Chapter 7, the fluorescence characteristic of Rhodamine 6G dye, under high repetition rate excitation, was experimentally investigated in stationary and flowing dye gain medium. A commercially available spectrometer, along with specially developed diagnostics software described in Chapter 2, was used for the investigation of fluorescence emission fluctuations. An estimation of the peak wavelength, spectral width, and area under the widths of an individual spectrum were carried out from the composite image of the recorded spectra. It was observed that the spectral width broadened and large fluctuations are visible in stationary solution as compared to dye flowing medium.

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

In Chapter 8, theoretical and numerical investigations of the dye gain medium inhomogeneity induced by high repetition rate excitation, and its influence on the spectral emission are described. The thermo-optical properties of a narrow spectral width CVL pumped dye laser were experimentally investigated by changing the dye gain medium temperature from 23-35 °C. The dye laser output parameters such as average optical power, spectral width, wavelength, and beam divergence were experimentally investigated during the dye solution temperature rise from 23 to 35 °C. The dye laser optical characteristics through gain medium bulk temperature stabilization were also experimentally investigated.
Finally, in Chapter 9, a summary of the work carried out during the course of investigations is given. This chapter also provides a brief discussion about the scope of further work in this field.

In summary, the present thesis comprehensively investigates the issues coupled with high repetition rate dye laser, which incorporates studies on inclusive dye laser system i.e. gain medium, resonator, excitation source, diagnostics tools, dye cells, gain medium flow analysis and thermo-optic properties.