

Multiline distributed feedback dye laser endorses Wien's displacement law

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Abstract Peak spectral intensity shifting in a distributed feedback dye laser (DFDL) supporting multiple lines in 550 to 570 nm range is reported. A 3 mM solution of Rh6G in ethanol is pumped with a complex interference pattern of multiple Q-switched and mode-locked frequency-doubled Nd:YAG laser pulses for obtaining numerous discrete laser lines to study the impact of temperature buildup on resonant frequency shifting characteristics. Multiple pulses pumped DFDL are operated at five and nine wavelengths in two separate arrangements and spectra recorded without changing exciting pulses intensities and angles. Higher intensity lines are found to shift during operation from longer to shorter wavelengths due to gradual temperature rise in the dye solution. Laser lines associated with relatively higher intensities shift from lower to higher frequency parts of emission spectrum due to accumulative temperature rise. This temperature-dependent peak intensity drift occurrence becomes faster by reducing the pumping laser interpulse periods. This paper reports gradual heat accumulation stipulated spectral intensity shifts of laser lines in multiline DFDL, which appears to be compliant with Wien's displacement law.

1 Introduction

DFB lasers were extensively pursued in the last three decades of the twentieth century. Kogelnik and Shank [1], for the first time, in May 1972 reported their famous *Coupled Wave Theory* of distributed feedback dye lasers. DFB laser action was noted by Derkacheva and Krymova [2], 3 years back, in May 1969, in their experiment of four photon resonant parametric interactions in lasers by using dyes. The photoinduced refractive index increase in PMMA was reported by Jomlinson et al. [3] in June 1970. Until February 1971, DFB laser action was not completely described as a result of refractive index and gain modulation of the dye solution, which was confirmed by Kogelnik and Shank [4]. A distributed feedback dye laser (DFDL) produces a single narrow line; however, if the dye solution is pumped with multiple pairs of pulses, the dye laser produces multiple lines. A generalized model of multiple wavelength lines has already been reported elsewhere [5, 6]. Unlike conventional lasers, distributed dye lasers do not require discrete external mirrors. A positive feedback is provided by a distributed type of volume grating that is induced in the gain medium by the interference of two beams of wavelength λ_p at an angle of 2θ between them. The distributed grating can provide positive feedback only if the wave scattered by periodic structures is reversed in direction at the Bragg angle. The bilateral Bragg scattering results in two coupled waves of the same frequency by traveling in opposite directions. When the periodic structure is sufficiently dense with reasonable contrast and gain coefficient, then the scattered radiation will resonate at λ_o to initiate the distributed feedback laser action if the periodic gain structure satisfies the Bragg condition.

$$\Lambda = m_B \frac{\lambda_o}{2n_s} \quad (1)$$

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where Λ is the periodicity of grating, m_B is the Bragg index, λ_o is the DFDL wavelength, and n_s is the refractive index of the solution. Alternatively, periodicity may be defined in terms of excitation wavelength λ_p and half angle θ_H between the interfering beams [7].

$$\Lambda = \frac{\lambda_p}{2 \sin \theta_H} \quad (2)$$

Combining (1) and (2)

$$\lambda_o = \frac{n_s \lambda_p}{m_B \sin \theta_H} \quad (3)$$

The excitation light intensity varies at the dye cell with the cosine of z with a period Λ causing a refractive index and gain modulation. The field intensity distribution may be given as:

$$I = I_o \left(1 + \cos \frac{\pi z}{\Lambda} \right) \quad (4)$$

Superposition of multiple laser pulses leads to an induction of multiple volume dynamic gratings by supporting distinct laser lines. Inspection of laser lines shows that their spectral intensities change with a gradual temperature accumulation. Wien's displacement law holds good for blackbody in equilibrium. It states that the maximum intensity emitted by a blackbody occurs at a wavelength which is inversely proportional to the temperature [8]. Researchers claim [9] that the Wien's displacement law needs a correction for non-equilibrium systems like sonoluminescence and lasers. The work to be described in this paper shows that in case of multiple simultaneous wavelength lasers the Wien's displacement law holds favorable.

2 Experimental setup

In order to study the effect of delay between two pairs of the pump beams in multiple or nonlinear gratings, the second harmonic of a passively Q-switched and mode-locked Nd:YAG laser was set up to give off 1.5 mJ of green output. The pump laser was charged just above the threshold to reduce the number of oscillating modes. After transverse and longitudinal mode selection, the net output energy was less than 1 mJ. Pump pulses at beam splitter had a total energy of $700 \pm 10 \mu\text{J}$ with which two superimposed DFDLs were operated simultaneously on the same location in the dye solution. Gratings produced in the dye cell were shifted according to the time and space within a temporal and spatial coherence of the pumping laser. Powers of two pumping pulse pairs were 135 ± 10 and $187 \pm 10 \mu\text{J}$, respectively. The angle between the two main pumping beams was 0.19° which is less than the divergence of the DFDL. We used 10 % of the main beam to trigger the streak camera,

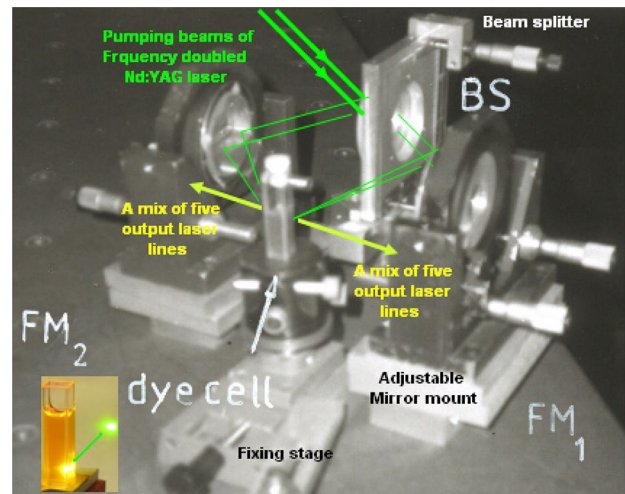


Fig. 1 Experimental setup for five lines DFDL operation

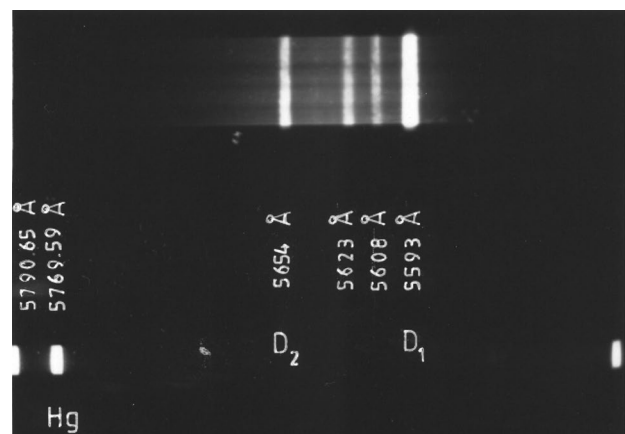


Fig. 2 A spectrum of four simultaneous laser lines

and 15–16 % was lost in the conveying optics. The net ratio of pump beams for two pairs at the dye cell was 1.2:1. The experimental work was carried out in two phases. In the first phase, two DFDLs were aligned individually with a comparatively large optical path difference (delay) between the pumping pairs and then operated simultaneously to obtain a multiwavelength operation. The experimental arrangement may consist of a number of configurations, by using mirrors and reflection or transmission gratings. We chose the most common Shank-type geometry shown in Fig. 1.

When the dye solution is pumped through two pairs of pulses, then two gratings are induced on dye medium. Interaction of two dynamic gratings leads to an induction of five superimposed gratings, supporting five distinct laser lines which are separated by 15–16 angstrom intervals. A spectrum of four equally spaced spectral lines, calibrated by mercury lamp, is shown in Fig. 2.

To obtain a higher number of laser lines, the number of pumping pulses was increased from 4–6 to produce nine laser lines. Frequency-doubled, Q-switched and mode-locked Nd:YAG laser output of 511 μJ pulses was split up into three beams to produce three pairs of pump pulses, so that it excites the dye solution to produce nine laser lines as shown in Fig. 3.

When the dye solution is pumped through three pairs of pulses, three gratings are induced on dye medium. Interaction of three dynamic gratings leads to the induction of nine superimposed gratings, supporting nine distinct wavelength laser lines which are separated by 7–10 angstrom intervals depending on incident half angles. A spectrum of nine equally spaced lines, calibrated by mercury lamp, is shown in Fig. 4.

The number of simultaneous output laser lines depends on the pairs of pumping pulses, derived from the same

excitation laser by amplitude phases or wavefront divisions, so as the delay between pumping pulses is lesser than temporal and spatial coherence lengths of the pump laser.

3 Results and discussions

Rhodamine 6G (Rh6G) has a niche emission window around 587 nm. Solution of Rh6G (brown) in ethanol has a maximum absorption from 500 to 580 nm and a maximum emission at 570–600 nm. It supports laser action at 580–590 nm as shown in Fig. 5.

Simultaneous oscillation of multiple lines requires precise incidence angles, delays and molar concentration to achieve the discrete super-continuum. Multiwavelength operation of DFDL has exhibited two new results, which are simultaneous oscillation of five and nine lines as described elsewhere [5, 6], and a spectral shift of high intensity lines due to temperature effects.

Wavelengths of higher intensities in blackbody emission spectrum depend on the temperature. Radiation intensity at wavelength λ (μm) and temperature T (K) is given by Plank's law.

$$I_{\lambda h}(\lambda, T) = \frac{2\pi h(\lambda\nu)^2}{\lambda^5(e^{14387.69/\lambda T} - 1)} \tag{5}$$

Blackbody radiations, at specific wavelengths, exhibit maximum intensity under the famous Wien's displacement law

$$\lambda T = 2897.8 \mu\text{m K} \tag{6}$$

Peak of sunshine occurs at 0.5 μm, earth radiations occur at 9.72 μm, and human body emits at 9.35 μm. When heater approaches 1500 K, it emits enough radiations to appear white to the human eye. A typical spectrum of blackbody

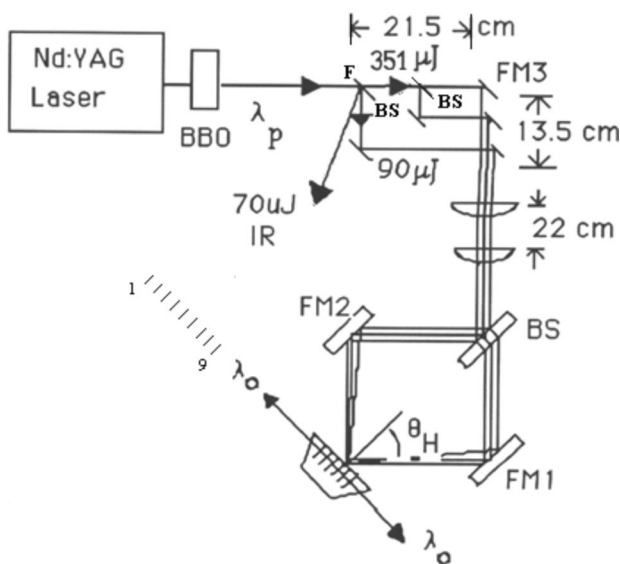


Fig. 3 Experimental setup for nine lines DFDL operation

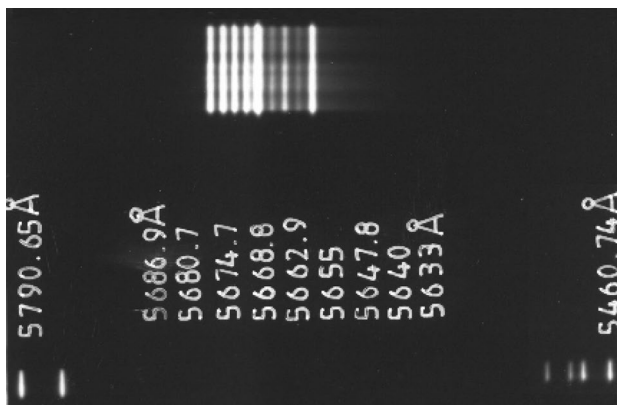


Fig. 4 A spectrum of nine laser lines

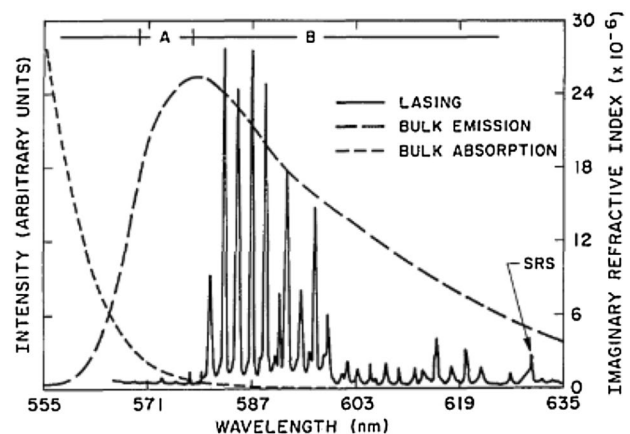


Fig. 5 Absorption, emission and lasing spectra of Rh6G in ethanol [10]

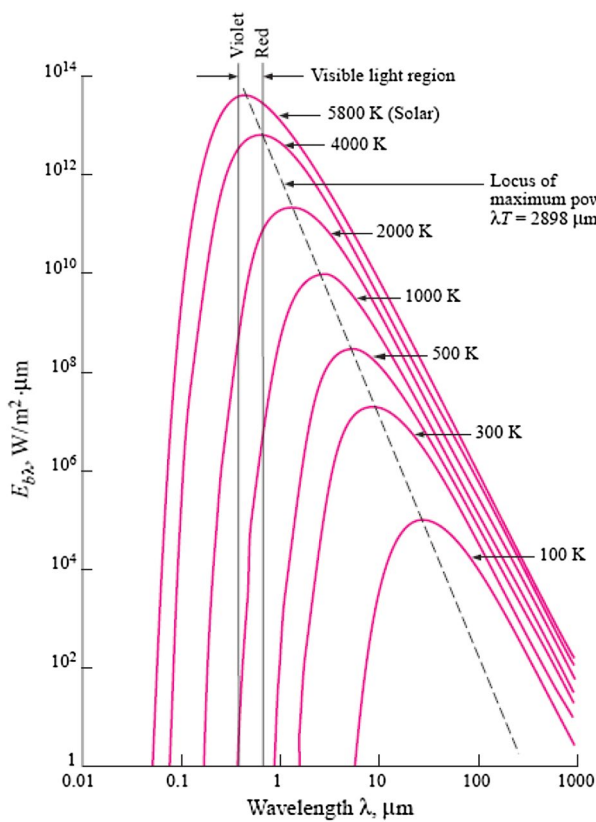


Fig. 6 Wien's displacement law [11]

radiation, exhibiting blue shift of maximum intensity radiations, is shown in Fig. 6.

When the temperature of a blackbody rises from 100 to 6000 K, the wavelength associated with maximum energy shifts from 50 to 0.4 μm . At extremely high temperatures during nuclear explosions, the X-rays and gamma rays are also emitted. Earth at low temperature emits longer wavelengths compared to the human body, engine and solar radiations.

Laser and sonoluminescence are typical examples of non-equilibrium blackbody radiation sources. An atomic system, giving off spontaneous emission, may be regarded at thermal equilibrium to derive Einstein coefficients, but it goes into non-equilibrium state during stimulated emission due to a population inversion in multilevel atomic or molecular systems. Laser-pumped laser wavelength is longer than the pumping source which is 520 and 560 to 575 nm in this case. The wavelength shift between pumping and laser lines is recognized as the Stokes' shift which may range from 20 to 30 nm as shown in Fig. 7.

Stokes' shift is found from longer to shorter wavelengths in sonoluminescence as acoustic waves compress bubble to produce blue luminescence. Wien's displacement law applies well despite Stokes' shift in multiple wavelength lasers. The 530 nm Nd:YAG laser pumping light produces

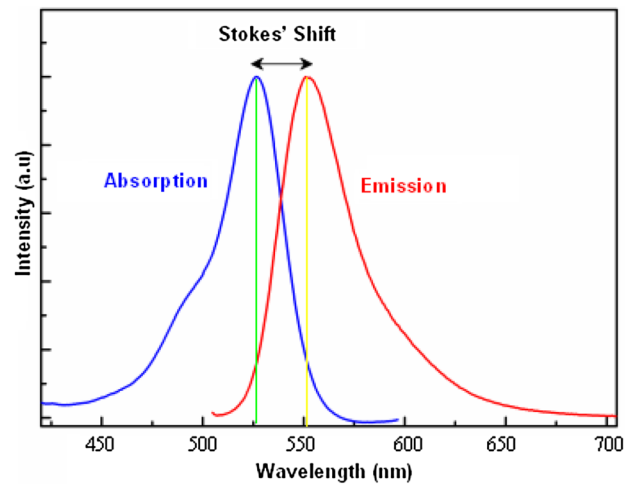


Fig. 7 Rh6G stokes' shift

multiple lines from 560 to 570 nm wavelength. Radiation intensity in the blackbody interval, say $(\lambda, \Delta\lambda)$, is given as:

$$I_{\lambda h}(\lambda, \lambda + \Delta\lambda) = \int_{\lambda}^{\lambda + \Delta\lambda} \frac{2\pi h(xv)^2}{x^5 (e^{14387.69/xT} - 1)} dx \quad (7)$$

Differentiation of (7) leads to

$$\frac{\partial I_{\lambda h}(\lambda, \lambda + \Delta\lambda)}{\partial \lambda} = \frac{\partial}{\partial \lambda} \int_{\lambda}^{\lambda + \Delta\lambda} \frac{2\pi h(xv)^2}{x^5 (e^{14387.69/xT} - 1)} dx = 0 \quad (8)$$

Two wavelengths separated by $\Delta\lambda$ on either side of peak emission wavelength under Leibniz rule may be written as [9]

$$(x + \Delta\lambda)^5 (e^{14387.69/(\lambda + \Delta\lambda)T} - 1) = x^5 (e^{14387.69/\lambda T} - 1) \quad (9)$$

In terms of bandwidth, the temperature may be given as:

$$T = \frac{14387.69}{5} \left(\frac{\Delta\lambda}{\lambda(\lambda + \Delta\lambda)} \right) \left(\frac{1}{\ln(\lambda + \Delta\lambda)/\lambda} \right) \quad (10)$$

In terms of λ_1 and λ_2 , (10) may be rewritten as

$$T = \frac{14387.69}{5} \left(\frac{\lambda_2 - \lambda_1}{\lambda_1 \lambda_2} \right) \left(\frac{1}{\ln(\lambda_2/\lambda_1)} \right) \quad (11)$$

In terms of central wavelengths, Eq. (10) may be rewritten as

$$T = \frac{14387.69}{5} \left(\frac{4\Delta\lambda}{4\lambda_c - \Delta\lambda^2} \right) \left(\frac{1}{\ln[(\lambda_c + \Delta\lambda/2)/(\lambda_c - \Delta\lambda/2)]} \right) \quad (12)$$

To investigate the effect of dye cell's medium temperature on the emission characteristics of distributed feedback dye lasers, the dye cell was pumped by a pair of pulses derived

from a Q-switched Nd:YAG laser. The DF DL produced a single line and its wavelength changed with an increase in the temperature of the dye cell as shown in Fig. 8.

Temperature-tuned DF DL works on the principle of increasing dye solution refractive index by increasing n_s in (3), which has nothing to do with our actual claim in this paper. To investigate the effect of temperature accumulation in the dye cell on the maximum intensity, wavelength DF DL was operated for five lines. Variation of individual lines' intensities at different times of operations is shown in Fig. 9.

Spectral line intensity shift from larger to smaller wavelengths over periods of operation may be attributed to a gradual temperature buildup phenomenon as reported previously [12]. Wavelength shift with temperature was estimated to 0.2 nm/C, but spectral intensity movement under

Wien's displacement law was noted more pronounced and apparent. To verify this effect, DF DL was operated for nine wavelengths. The spectral lines' intensities are found to correspond to Wien's law as shown in Fig. 10.

The spectral intensity shifting effect seems to be a thermal effect. Continual pumping of small dye solution volume increases its temperature, which leads to fluorescence peak shift from longer to shorter wavelengths. The peak intensity drifting effect occurs in multiple line lasers, but manifests itself by intensity fluctuation in single-frequency lasers.

To investigate the impact of heat accumulation in a single shot of 10–12 pulses of Q-switched and mode-locked Nd:YAG laser, the excitation and output pulses that were recorded on streak camera are shown in Fig. 11.

Streak records give an impression of peak pulse shift, but cannot give any quantitative shift in peak intensity envelopes of the DF DL pulses. The streak records were scanned using a microdensitometer to create a data file. The scan focus was passed through the middle of the both spectra one by one. The data were stored in a computer file for further analysis. A plot of single-wavelength Q-switched and mode-locked Nd:YAG and DF DL pulses is shown in Fig. 12.

It is concluded that the shifting of wavelength associated with the highest intensity in multiwavelength lasers is a Wien's law dependent phenomenon which can play a role in erbium-doped fiber amplifiers and tunable lasers. Wavelength drifting is far less than line intensity shifting during a routine operation. Application of DF B lasers in communication needs cooling to avoid the intensity shifting or gain shifting effect at various wavelengths. The wavelength

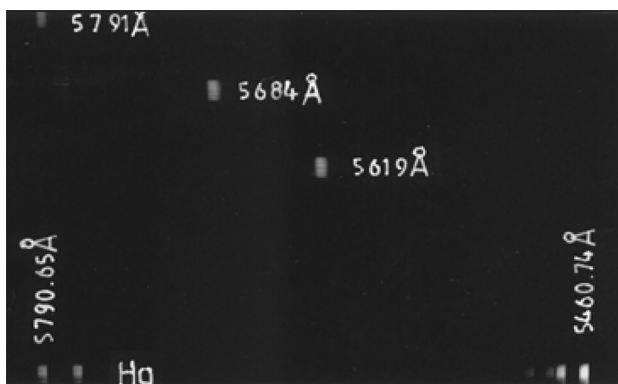
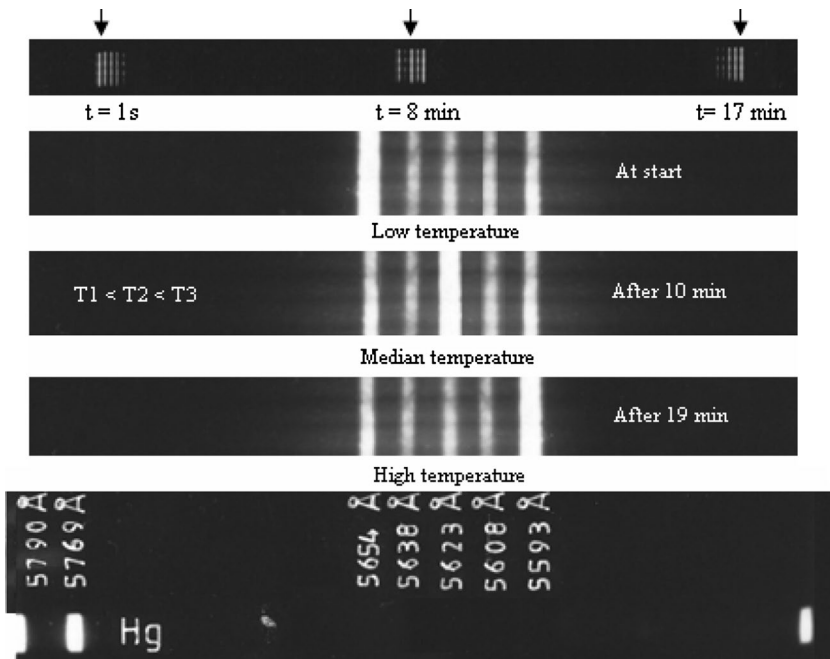


Fig. 8 Spectra of a temperature-tuned DF DL

Fig. 9 Five lines intensity shifting from 565.4 to 559.3 nm wavelengths



intensity drifting phenomenon may not be confused with Doppler effect as the measurements were taken in stationary laboratory environment. In case of Doppler shifting, the wavelength shifts without changing its intensity, but in case of Wien's displacement law for blackbody radiations

as well as multiple line lasers, the spectral lines stay at their own position, while their intensities shift from longer to shorter wavelengths.

Multiwavelength lasers are important for monitoring the multiple gases in real time. Commercially available lasers can provide multiple lines in UV, visible and IR regions. Dye lasers give off multiple lines in visible and NIR (0.8–0.9 μm) regions, fiber or semiconductor DFB lasers in IR (1.35–1.55 μm) and optical parametric oscillators (OPO) in MIR (2–4 μm) regions. OPO need to be tuned to satisfy the frequency difference or sum phase-matching constraints, but DFB dye lasers are capable of supplying simultaneously multiple lines for gas monitoring or industrial control applications. OPO support one line at a time, so they are recognized by their wide tuning ranges. A high-resolution (0.01 cm^{-1}) tunable laser (VHR-TL), with tunability range from 2350 to 3125 cm^{-1} , is more expensive than 10 cm^{-1} range tunable diode laser (TDLAS). TDLAS is powered by a quantum cascade laser (QCL) or vertical cavity surface emitting laser (VCSEL). The heart of VHR-TL is a laser-pumped OPO. VHR-TL powered Blue X-FLR8 gas analyzer provides multiple gas analysis down to 30 ppb level. It can measure multiple gases due to a scan facility in seven windows [13]. We can conceive a multiple IR line DFB laser by pumping suitable luminescent materials or organic dyes [14]. Spectra-Physics Sirah dye laser using Styryl 8 (DMSO) and Styryl 8 (Ethanol) produces MIR wavelengths, which can further be tuned by using OPO technique. The energy transfer technique may also be used to pump dye solutions at a longer wavelength to design longer wavelength lasers [15]. There is a dire need of multiple lines IR laser for environmental monitoring and communication applications. DFB lasers in the market have the drawbacks of spectral intensity shifting as well as minor wavelength drifting which may be harnessed by one of the several techniques stated in the literature [16–18].

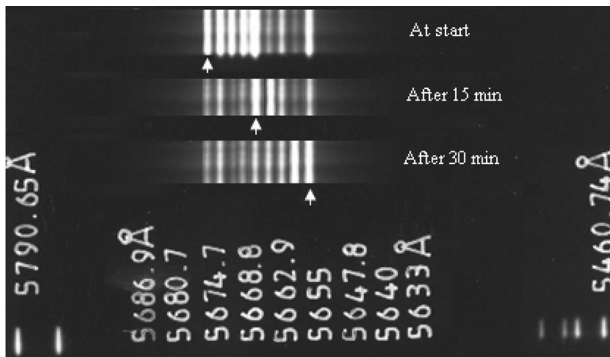


Fig. 10 Nine lines intensity shifting from 568.6 to 563.3 nm wavelengths

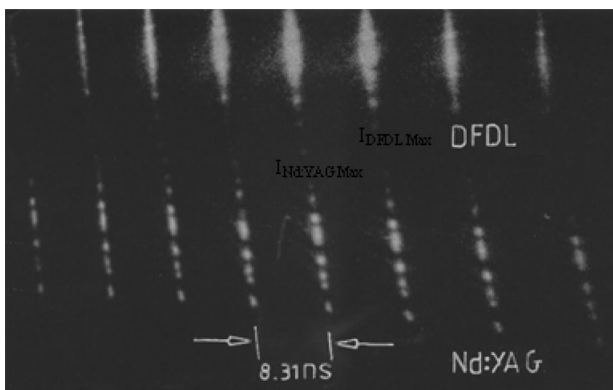
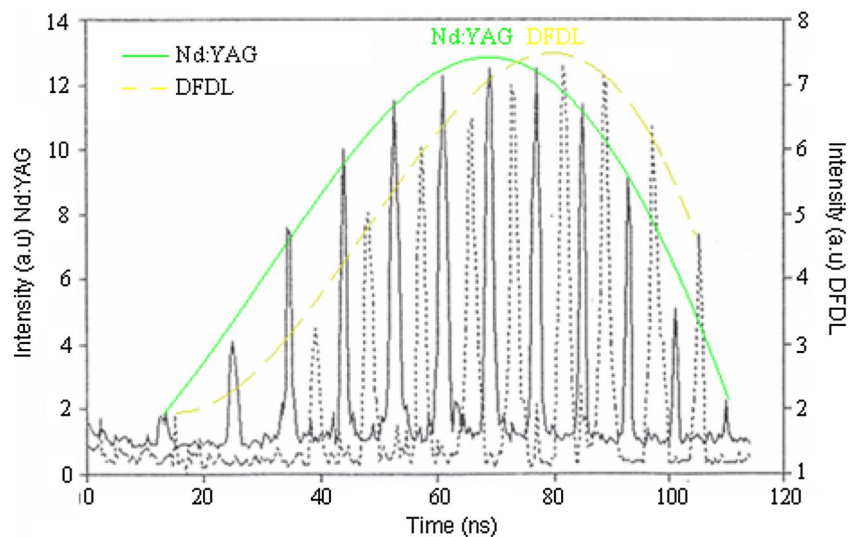


Fig. 11 Streak records of DFDL and Nd:YAG laser lines

Fig. 12 A typical streak record of Q-switched and DFDL mode-locked Nd:YAG laser and corresponding DFDL output for interpulse period of 8.31 ns



4 Conclusions

When distributed feedback dye laser is pumped by the second harmonic of a Q-switched and mode-locked Nd:YAG laser, a discrete coherent light continuum is obtained. DFDL spectral linewidth is narrower than the pump laser, both in Q-switched and mode-locked operations; however, it becomes equal to pump laser when pinhole is inserted into the cavity for transverse mode selection. The effect of gradual heat accumulation is to shift the peak of DFDL pulses envelope during continued operation due to steady temperature rise. Temperature rise can be simulated numerically using a software program. It was found in this study that the dye medium retains nearly 57 % of the earlier pumping pulse energy, which contributes to temperature rise in the dye solution. The oscillation of several lines in simultaneous operation was theorized due to multiple superimposed gratings causing nonlinear behavior. Spectral line shift from longer to shorter wavelengths may be attributed to a temperature rise by a continuous pumping which indirectly endorses the Wien's displacement law that is not considered valid for thermally stabilized single-line lasers.

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Compliance with ethical standards

Conflict of interest The authors whose names are listed above certify that they have no affiliations with or involvement in any organization or entity with any financial interest (such as honoraria; educational grants; participation in speakers' bureaus; membership, employment, consultancies, stock ownership, or other equity interest; and expert testimony or patent-licensing arrangements), or non-financial interest (such as personal or professional relationships, affiliations, knowledge or beliefs) in the subject matter or materials discussed in this manuscript.

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