

Influence of the medium on the fluorescence of copper vapor laser pumped rhodamine 6G dye: introduction, experimental details, and stationary case

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Abstract: The physical conditions of the medium surrounding the dye molecules significantly affect the fluorescence properties of an organic dye. The fluorescence intensity, wavelength and its linewidth are altered by the refractive index gradient induced by the temperature and the flow of the dye solution. The peak wavelength and its full width at half maximum (FWHM) vary with the Reynolds number of the fluid flow. This is the first to two sequential communications that report on the observed fluorescence characteristics of rhodamine 6G, in ethylene glycol solvent, in a stationary dye solution and at different Reynolds number of fluid flow under copper vapor laser (CVL) excitation. The first part includes the introduction, experimental details, and results of the stationary case. The second part includes the dynamic case and the discussion.

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An organic dye, characterized by a strong absorption band in the visible region of the electromagnetic spectrum, is the active component of the amplifying medium for dye lasers [1]. Lasing action is achieved by modifying the fluorescence of the dyes. Presence of any kind of impurities or foreign element contiguous to it significantly affects the inherent properties. Scattering has always been considered detrimental to laser action because it alters the direction and spatial coherence of the photons. There are many nonradiative processes that can compete effectively with the light emission and also reduce the quantum efficiency of the dyes [1]. It is known that absorption of the pump light in the dye solution causes thermal heating. When the heating is non-uniform, the non-uniformity gives rise to temperature gradients [2] (and hence refractive index gradients) in the solution that may cause optical in-homogeneities. There are many instances where the states of the dye molecules affect the rates of nonradiative processes and other characteristics to a degree that cannot be neglected. Studies on the effect of excited-state absorption [3], self-absorption of molecular fluorescence [4], influence of the solvent on the spectral and stimulated emission characteristics [5], influence of solvation on the lasing characteristics of dye solutions [6], influence of the solution concentration on the efficiency of dye laser [7], effect of laser dye deterioration on performance [8], refractive indices and the thermo-optical properties of the dye laser solvents [9] have been done in detail. Laser linewidth instabilities, due to thermal gradients and medium inhomogeneities have also been studied [10]. To minimize the thermal distortion of the medium, the active dye volume exposed by the pump laser needs to be rapidly replaced between successive pump laser pulses [10, 11]. The dye solution must be flowed at sufficient velocities across the pump region for this purpose. This

often results in chaos and hence the degradation of the optical quality of the medium [2, 12, 13]. Spontaneously emitted photons, which will finally be converted into the laser photons, are affected by the physical state of the solvent molecules surrounding the active dye molecules.

Studies have been reported on the temperature dependent emission characteristics of dyes and its effect on the lasing performance [8, 9, 14]. A recent report by Singh *et al.* [15] on the stability of output characteristics (particularly on wavelength and bandwidth) of a high repetition rate dye laser shows that the flow of the dye solution across the pump region significantly affects its performance in the laminar region of fluid flow. The speed of the flow of the liquid in which dye is dissolved may also be having an effect on fluorescence width, peak wavelength, and its intensity. However, to the best of our knowledge, no study of the fluorescence characteristics of dyes in a flowing dye solution has been carried out. This is very important for the development of high repetition rate high average power dye lasers. In this paper, we report the observations on the fluorescence characteristics of rhodamine 6G in ethylene glycol solvent in a stationary solution and at different flow conditions of the fluid transversely pumped by a high repetition rate CVL.

The experimental set-up consists of a dye cell containing laser grade rhodamine 6G dye dissolved in spectroscopic grade ethylene glycol solvent (1.0 mM concentration), and a plug-and-play miniature fiber optic spectrometer (USB 2000, Ocean Optics Inc.). A CVL ($\lambda = 510.6$ nm and 578.2 nm, 60 ns pulse duration, 5.6 kHz repetition rate, plane-parallel resonator, 47 mm beam diameter) is used as the pump source. The pump beam was transversely line focused on to the dye cell through a cylindrical lens. The spectrometer accepts light input via an optical fiber. The CCD detector of the spectrometer had a minimum integration time of 1ms. A dedicated software developed by Vora [16] was used to record the laser spectra continuously and to make composite on-line image simultaneously for the duration of observation. A dye cell, used in the experiment was connected with a dye solution reservoir in closed loop. The dye solution cooling and pumping system consisted of a closed reservoir, a pump, a compressor, a heat exchanger, a filter, a by-pass valve, a temperature sensor, a temperature controller and a digital flow meter. To adjust the flow at different flow rates, a part of the solution was diverted through an adjustable

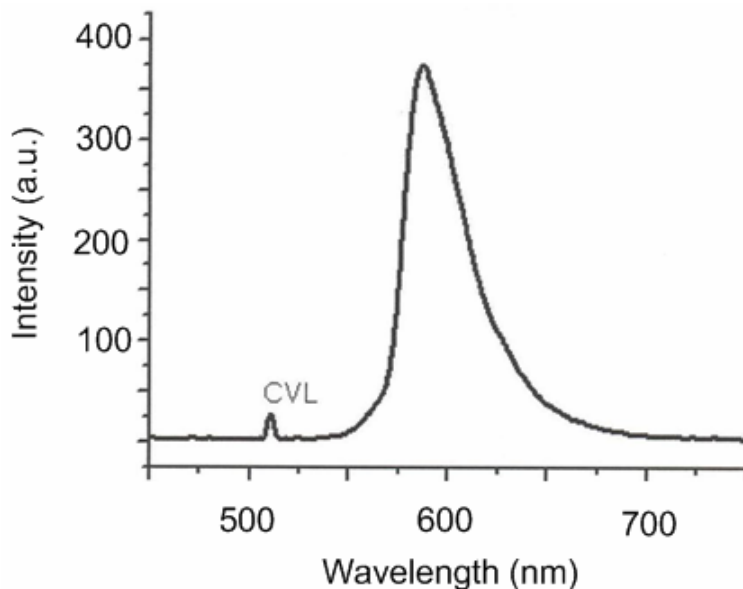


Fig. 1. Typical fluorescence spectrum of rhodamine 6G, in ethylene glycol, in a stationary solution.

by-pass valve again to the reservoir. The other part of the solution flowed through the filter system to the dye cell and returned to the dye solution reservoir. The flow of the dye solution could be adjusted to any value from 0 to 20 liters per minute (l/min.). The temperature of the solution was maintained during the experiment at 22 ± 0.5 degrees C.

The study on fluorescence properties of rhodamine 6 G in ethylene glycol solvent was carried out in two modes. In the first mode, the dye solution is stationary. Fig.1 shows the typical fluorescence spectrum of the rhodamine 6G in ethylene glycol solvent for a stationary dye solution. The small sharp vertical line, to the left of the main spectrum, is the green line (510.6 nm) from the CVL. The observed fluorescence peak wavelength is 587.45 nm and has an FWHM of 35.73 nm. The high repetition rate CVL heats up the medium, and changes the ambient temperature of the dye solution. As a result, the spectrum changes. A number of spectra over a long time (~ few minutes) have been recorded to see the thermal effects on peak wavelength and its width. Fig.2 shows the composite (stacked) picture of 1000 fluorescence spectra. Any variation from the top, where acquiring the spectra starts, to the bottom, gives visual information about any kind of departure taking place in the time domain. Each spectrum was recorded with an integration time of 1ms, which was the minimum exposure time set by the CCD camera, and also by the minimum time required to process the data by sending it into the dynamically allocated memory of the PC. The total time taken in acquiring 1000 spectra and generating their composite picture simultaneously was 5 minutes. The peak wavelength, the peak intensity, and width of the

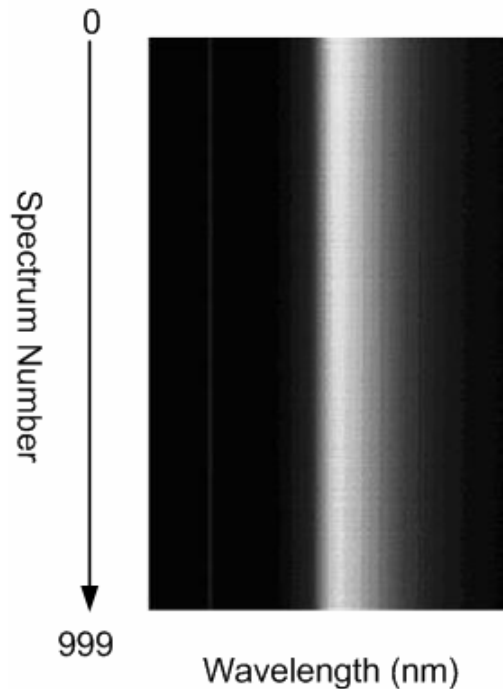


Fig. 2. The composite (stacked) picture of line scan of fluorescence spectrum.

spectra, from the composite image, were also recorded. Fig.3 shows the FWHM of 1000 spectra (from 0 to 999). The fluorescence FWHM changes with time over the observation period. It has minimum value 34.74 nm, maximum value 36.64 nm over the observation period of 5 min. Fig.4 shows the variation in peak wavelength intensity of the fluorescence in stationary solution over the observation period. The intensity decreases with time over the observation period. Fig.5

shows the variation of the peak emission wavelength in a stationary solution over the observation period. The emission peak wavelength also shows slight change with time.

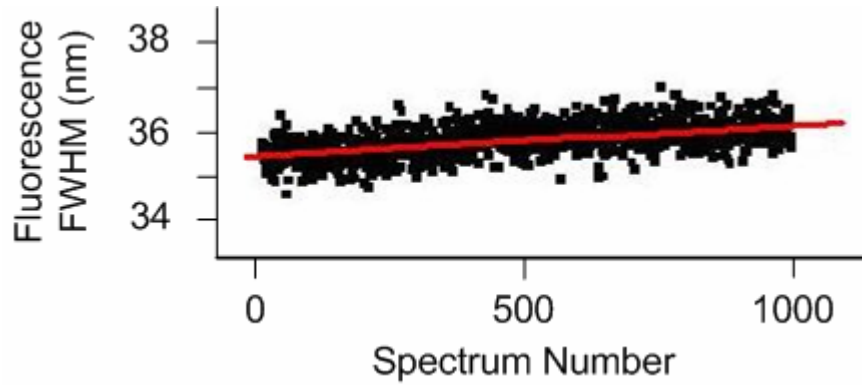


Fig. 3. The variation in bandwidth of 1000 fluorescence spectra (0 to 999) in a stationary dye solution.

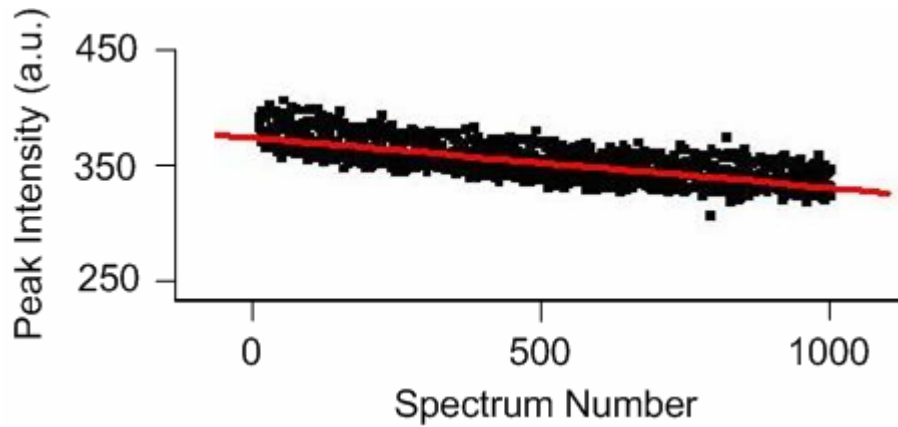


Fig. 4. The variation of the intensity, at the peak wavelength, in a stationary solution over the observation period.

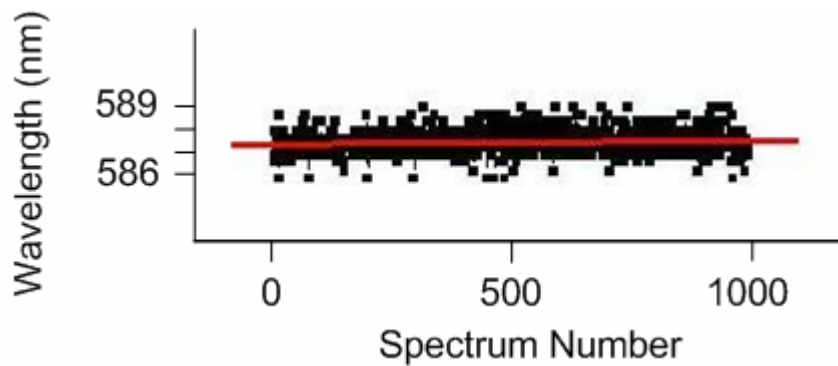


Fig. 5. The variation of peak emission wavelength in a stationary solution over the observation period.

To summarize the results of the stationary dye case: under CVL excitation, at a 5.6 kHz repetition rate, the FWHM of the linewidth increases slightly, with a corresponding decrease in the intensity, accompanied by a slight shift in the peak of the lasing wavelength. The results of the dynamic study and corresponding discussion are given in [17].

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