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Effects of the solvent environments on the ASE from coumarin 503

K.H. Ibnaouf*

Al Imam Mohammad Ibn Saud Islamic University (IMSIU), College of Science, Physics Department, P.O. Box 90950, Riyadh 11623, Saudi Arabia

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ABSTRACT

In this work, the spectral and the amplified spontaneous emission (ASE) characteristics of coumarin 503 have been investigated in different solvents and concentrations. The absorption and fluorescence spectra of coumarin 503 showed only one band; this indicates no dimer or excimer formation, respectively. Under certain concentrations, solvents and pump power energies, coumarin 503 exhibits dual ASE peaks; (475 nm and 495 nm) even though for the same solutions there was only one absorption or fluorescence band. The shorter wavelength (475 nm) coincides to fluorescence band while the longer wavelength (LW) at 495 is an abnormal peak. This abnormal peak (495 nm) could be attributed to the combination of two molecules in an excited state and the solvent acting as a bridge between them under high power excitation.

1. Introduction

Coumarin dyes are a class of laser dyes widely used to produce lasers in the blue and green region of the spectrum. They are derived from coumarin by substitution with an amino or hydroxyl group. When the dye molecules are pumped with an intense light source (flash lamp or laser), they are excited to produce fluorescence, ASE or laser [1–5].

When the combination takes place between two organic molecules both in the ground state, this is called a dimer. When an excited organic molecule combines with another molecule in the ground state, however, this is called an excimer. Similarly, when an excited molecule interacts with a different molecule (solvent or other solute) in the ground state, an exciplex is obtained. All of these molecular species have been studied for a long time [6]. However, a new molecular species has been proposed, wherein two excited molecules combine together with the assistance of the solvent, this is called a superexciplex. Such a superexciplex has been observed only for very few dye solutions under certain concentrations and pump power energies [7–17].

In this context, we reported the amplified spontaneous emission (ASE) bands from a coumarin 503 solutions under Nd:YAG laser excitation (355 nm). Coumarin 503 under certain concentrations exhibited two ASE bands (longer (LW) and shorter (SW) wavelength) in some solvents that having intermediate polarities. However, the dual ASE is strongly dependent on the solvents, solute concentration and pump power energy for coumarin dye. The longer wavelength ASE band is attributed to the interaction between the dye and the solvent in the excited state, and the solvent plays an important role.

2. Experimental

Coumarin 503 dye was brought from Exciton Co. (USA) and used as received. The molecular structure of which is given in Fig. 1. Coumarin 503 was dissolved in different solvents (spectroscopic grade with purity 99.8%) for a wide range of concentrations. The absorption spectra were recorded using a Perkin Elmer lambda 950 spectrophotometers over the range from 100 to 1100 nm, and the fluorescence spectra were recorded using a Perkin Elmer LS 55 spectrofluorometer which has a scan range of 200–1000 mm, at room temperature. The excitation wavelength was 355 nm.

The excitation source was third harmonic (355 nm) of a Nd:YAG laser. A pulse width was 6 ns was used. A quartz cylindrical lens of focal length of 5 cm was used to focus the UV laser pulse. The focused pulse is transversely applied to excite the coumarin solution. The cuvette was kept tilted to avoid feedback. See Fig. 2. Under suitable pump power and concentration of coumarin 503, we could achieve amplified spontaneous emission (ASE) beam coming out as a cone of light. The emanated light was collected by a 1-mm entrance slit of a CCD camera, to obtain the spectral features of the amplified spontaneous emission (ASE).

* Tel.: +966 552866400; fax: +966 2586679.
E-mail address: kheo90@gmail.com

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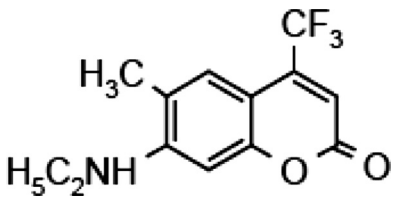

Fig. 1. Molecular structures of polymer of coumarin 503.

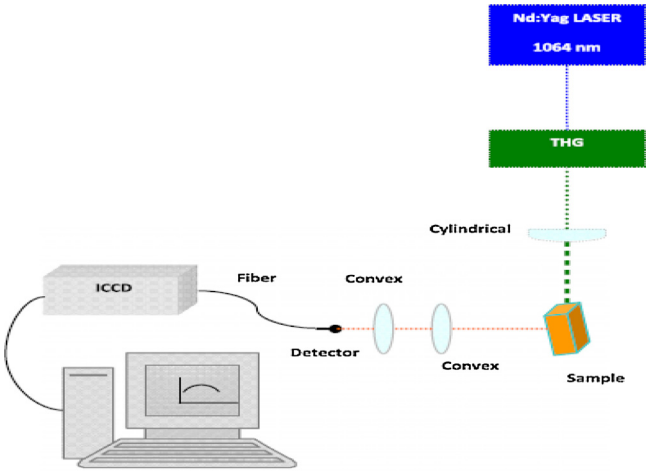

Fig. 2. Experimental arrangement for transverse excitation of the coumarin 503 in solution.

3. Results and discussion

3.1.1. Absorption and fluorescence spectra

Coumarin 503 was dissolved in ethanol under different concentrations. The absorption spectra of coumarin 503 were recorded as shown in Fig. 3. The spectra were taken for a wide range of concentrations from 5×10^{-5} to 15×10^{-5} g/ml. It was found that, the absorption spectrum has only one band at 393 nm. When the concentration increased, the spectrum profile remained the unchanged regardless of concentration, though the optical density increases with increasing concentration of coumarin 503, without any new band. This indicates that the absence of aggregation in the ground state for these dye solutions over a wide range of concentration used.

The fluorescence spectra of coumarin 503 in ethanol were obtained. The concentrations were taken from 5×10^{-5} to 15×10^{-5} g/ml. The results showed that the fluorescence also has only one band at 492 nm as shown in Fig. 3. The fluorescence spectral profile did not change irrespective of concentration. This indicates that the absence of excimer or exciplex for these dye solutions over a wide range of concentrations used.

Having discussed the effect of the concentrations on the spectral properties of coumarin 503, we may now discuss the influence of the solvents on the absorption and fluorescence spectra. Coumarin 503 was dissolved in different organic solvents having different dielectric constant. The concentration of these solutions was fixed at 5×10^{-5} g/ml. When the dielectric constant of the solvent increased, the absorption and the fluorescence peak of coumarin 503 shifted toward the red. For example, the absorption and the fluorescence of coumarin 503 in hexane were 365 nm and 422 nm, respectively. For dimethylformamide (DMF), the absorption and the fluorescence were located at 398 nm and 495 nm. Table 1 shows the absorption and fluorescence bands for coumarin 503 in different organic solvents. It could seen that the dielectric constant of the solvents play an important role for the absorption and emission bands even though the concentration and all the operation condition were fixed.

Table 1
Spectral properties of coumarin 503 in different solvents.

Solvents	Dielectric constant	Absorption (nm)	Fluorescence (nm)
Hexane	1.88	365	422
Benzene	2.3	377	449
Chloroform	4.81	381	459
n-Butyl acetate	5.1	388	471
Ethanol	24.55	393	492
Dimethylformamide (DMF)	38	398	495

3.1.2. Stokes' shift

Coumarin 503 was dissolved in various organic solvents that have different dielectric constants with a fixed concentration of 5×10^{-5} g/ml. Small changes in the absorption and fluorescence spectra were observed (the fluorescence spectra were recorded at the excitation wavelength of 355 nm). Fig. 4 gives the variation of the Stokes' shift as a function of the dipole factor of the solvent, as defined by Metaga and Tsuno [18]. It can be seen that coumarin 503 in solution undergoes significant changes in the electron delocalization and becomes highly polar in the excited state than in the ground state. The Stokes, shift has a linear variation with the dipole factor, given within square brackets in the expression:

$$\nu_a - \nu_f \approx \left[\frac{(\varepsilon - 1)}{(2\varepsilon + 1)} - \frac{(n^2 - 1)}{(2n^2 + 1)}\right] \frac{(\mu_e - \mu_g)^2}{a^3 hc} \quad (3.1a)$$

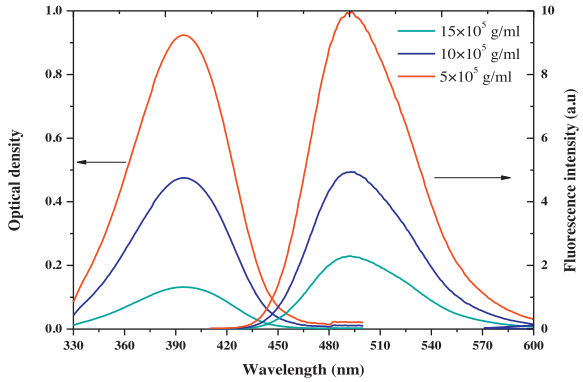

Fig. 3. Absorption and fluorescence spectra of coumarin 503 in ethanol at different concentrations.

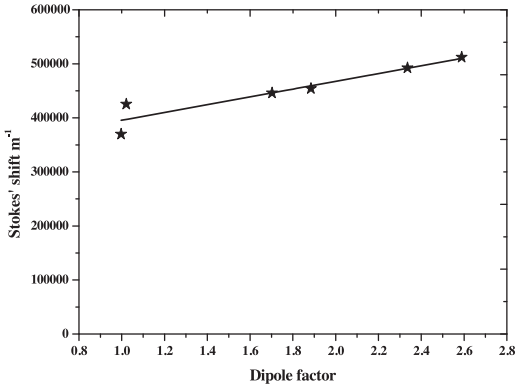

Fig. 4. Variation in the Stokes's shift of coumarin 503 in solution with a dipole factor of various solvents.