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Effect of Solvents on the Dipole Moments and Fluorescence Quantum Yield of Rhodamine Dyes

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Abstract

This study aims to investigate the effects of solvents of various polarities on the electronic absorption and fluorescence spectra of RhB and Rh6G. The singlet−state excited dipole moments (μe) and ground state dipole moments (μg) were estimated from the equations of Bakshiev -Kawski and Chamma−Viallet using the variation of Stokes shift along with the solvent's dielectric constant (ε) and refractive indexes (n). The observed singlet−state excited dipole moments were found to be larger than the ground−state ones. Moreover, the obtained fluorescence quantum yield values were influenced by the environment of the fluorescing molecule. Consequently, the concentration of the dye solution, excited singlet state absorption and aggregate of dye molecules has been found to affect the values of the fluorescence quantum yield.

Keywords: *Absorption, Fluorescence, Ground state and excited state dipole moment, Laser dye, Photo physics, Solvent effect.*

1. Introduction

The rhodamine dyes (RhB and Rh6G) are xanthene dyes with a large variety of technical applications [1]. In photochemistry, they are used as photo sensitizer [2-4] and quantum counter [5], as well as the active medium for tunable laser radiation in the visible region of light spectrum. They are red to violet in color [6]. The fluorescence quantum yield (Q_f) is one of the key photophysical quantities amenable to direct experimental determination. The quantum yield of fluorescence is a measure of the rate of non radiative transitions that compete with the emission of light. Knowledge of the fluorescence quantum efficiency of organic dyes as concentration dependent is essential in selecting efficient laser media. Conventional measurements require the use of accurate luminescence standard samples and comparison of the given sample with a standard for which the fluorescence yield is known [5].

Knowledge of the environmental effects on the spectra and the quantum yields of fluorescence is necessary for the utilization of these fluorescence materials to their maximum potential. The environmental factors that affect the phenomena of fluorescence are interactions with solvent and other dissolved compounds, temperature, pH and the concentration of fluorescence materials. The absorption and emission spectra as well as the quantum yields of the fluorescent molecules are influenced by these four parameters. Solvent effects on the absorption and emission spectra of various dyes have been investigated by several authors [7-10]. Prior to discussing the results for fluorescence efficiency values of RhB and Rh6G, there are a number of other measurement and calculation aspects that should be considered.

The study of the dipole moment of an electronically excited state of a molecule is an important property that provides information on the electronic and geometrical structure of the molecule in the short-lived state.

The methods of determining the singlet excited-state dipole moment are based on the spectral shift caused either externally by electrochromism or internally by solvatochromism. The solvatochromic method is based on the shift of absorption and fluorescence maxima in solvents of different polarities. Experimental and theoretical studies on ground state (μg) and excited-state (μe) dipole moments in a variety of organic fluorescent compounds such as coumarins, indoles, purines, fluorescein and laser dyes are reported [11-18].

The knowledge of absorption and fluorescence characteristics of these dyes in various solvents at different temperatures helps in understanding their use as laser dyes. The excited state dipole moment of a dye is an important parameter as it provides information about the change in electronic charge distribution due to excitation. Prior knowledge of the dipole moments in excited states is often useful in the design of non−linear optical material. The solvatochromic technique gives important information about electronic transitions, which is quite useful for the assigning of $n \rightarrow \pi^*$ or $\pi \rightarrow \pi^*$ transitions. It is an established way of determining the dipole moments in ground and excited states for short-lived states based on a linear correlation between the wave numbers of the UV- VIS spectra of solute and solvent polarity function of ε (dielectric constant) and n (refractive index) of the solvent.

In this paper, the absorption and emission spectra of RhB and Rh6G dyes with different solvents at various concentrations and the singlet−state excited dipole moments (μe) and ground state dipole moments (μg), as well as the values of the fluorescence quantum yield, were estimated.

2. Experimental Procedure

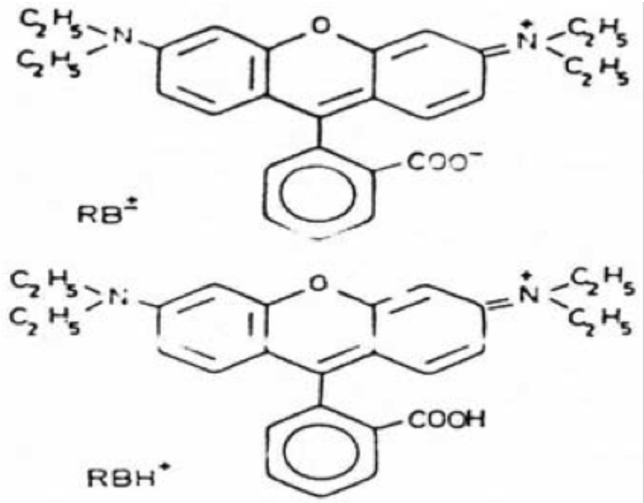
Rhodamine B ($C_{28}H_{31}N_2O_3Cl$) {Eastman Kodak Com.} with molecular weight 479.02 g/M and rhodamine 6 G ($C_{28}H_{31}N_2O_3Cl$) {Eastman Kodak Com.} with molecular weight 479.02 g/M were used for this research; their molecular structures are as shown in ***Figure 1***. RhB and Rh6G solutions at different concentrations varying from 10^{-6}-10^{-4} M/l were prepared from chloro-form, methanol and dimethyl sulfoxide (all analytical grad, Fluka Com.) using the relation reported in equation (1):

$$m = C*V*M \tag{1}$$

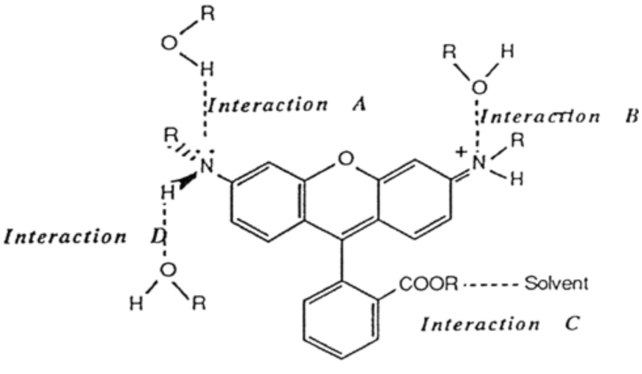
Where m is the mass of dye in grams (gm); C is the final concentration in mole/liter (mol/L); V is the volume of the solvents in liter (L); and M is the molecular weight of the dye in gram/mole (gm/mol).

The absorption and florescence spectra were measured using Shimadzu spectro-fluorimeter model RF-500 illustrated in ***Figure 2***. The quantum efficiencies were measured by the comparative method [7] using Rh6G in methanol with a quantum efficiency of 94% as standard solutions.

Absorption and emission spectra of RhB and Rh6G dyes in solvents of different dielectric constant (ε) and refractive index (*n*) values were also measured. The emission spectra were measured by exciting the sample at its maximum absorption.



a- Rhodamine B



b- Rhodamine 6G

Figure 1. *The molecular structure of a) Rhodamine 6G and b) Rhodamine B.*

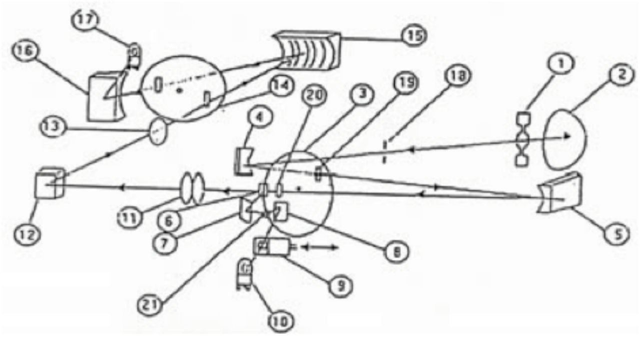


Figure 2. *Schematic of the Shimadzu spectro-fluorimeter model RF-500 apparatus. (1) Xenon lamp; (2) Condenser ellipsoidal mirror (SiO_2 coated); (3) Excitation slit assembly; (4) Concave mirror; (5) Concave diffraction grating (for excitation); (6) Beam splitter quartz plate; (7) Teflon reflector No.1; (8) Teflon reflector No.2; (9) Optical attenuator; (10) Monitor photomultiplier, R212-09; (11) A couple of light-collecting lenses; (12) Cell; (13) Light-collecting lens; 14) Emission slit assembly; 15) Concave diffraction (for emission); (16) Concave mirror; (17) Photometric photomultiplier R452-01; (18) Focus; (19) Entrance slit; (20) Exit slit; (21) Light beam balancing aperture.*

3. Results and Discussion

Figures 3, ***4***, ***5*** and ***6*** show the recorded absorbance and fluorescence spectra of RhB and Rh6G dyes in chloroform, methanol and dimethyl sulfoxide respectively.

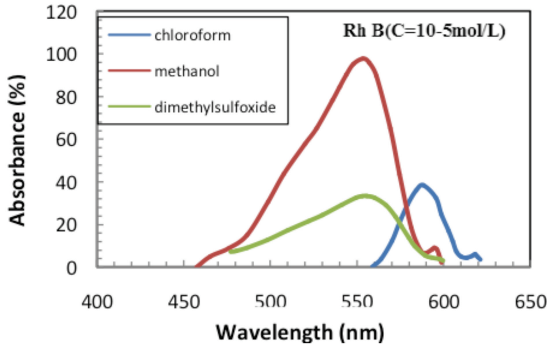


Figure 3. *The absorbance spectra obtained for RhB in different solvents.*

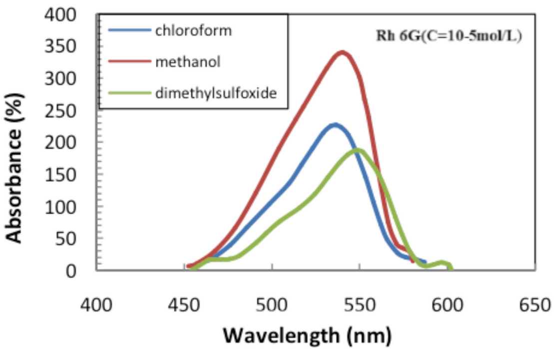


Figure 4. *The absorbance spectra obtained for Rh6G in different solvents.*

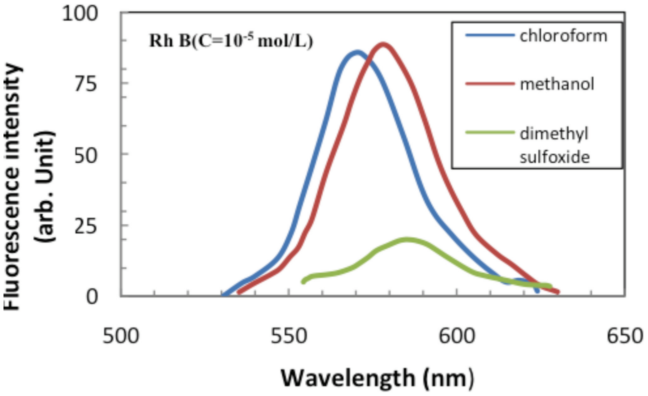


Figure 5. *The fluorescence spectra obtained for RhB in different solvents.*

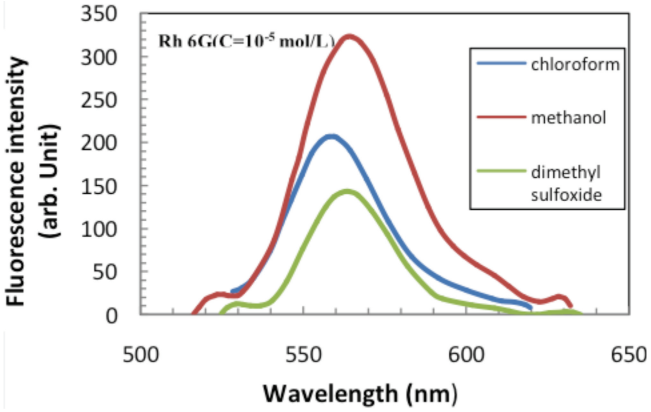


Figure 6. *The fluorescence spectra obtained for Rh6G in different solvents.*

Figures 7 and ***8*** represent the peak fluorescence wavelength (pfw) of RhB and Rh6G as a function of concentration in chloroform, methanol, and dimethyl sulfoxide as the respective solutions. A concentration-dependent change in the fluorescence spectra was observed. The fluorescence spectrum for the highest concentration differs strongly from that recorded at the lowest concentrations. These differences were significant as could be seen from the red shift of fluorescence and enhancement of the half width of the fluorescence spectrum with increasing concentrations (***Figures 7*** & ***8***). The observed spectral shift can be used as a guide to ascertain the interaction of the dye with the solvent media. It can be seen that the pfw increases with concentration and the fluorescence spectra of the dye decreases (that is, are shifted to smaller energies) against the increase in dye concentration. Bindhu *et al.* [19], employing the transient thermal lens technique (TL), found out that the value of the pfw increases with varying concentrations of Rh6G laser dye in alcohol and ethylene glycol solutions.

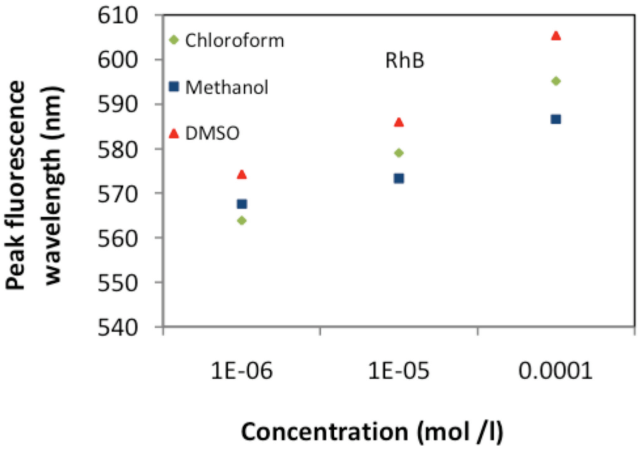


Figure 7. *The variation of peak fluorescence wavelength (pfw) with concentration for RhB in different solvents.*

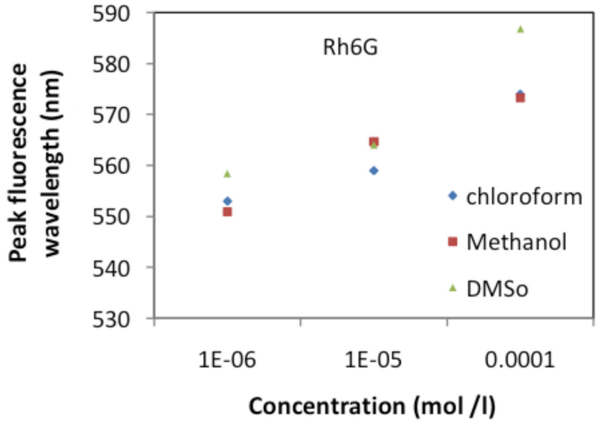


Figure 8. *Variation of the peak fluorescence wavelength (pfw) with a concentration for Rh6G in different solvents.*

The observed absorption and emission maxima, wave numbers, Stokes shift ($v_a - v_f$) and ½($v_a + v_f$) of the molecules were also studied. The charge transfer band shows a shift in the absorption spectra on changing the solvent while the emission spectra shows a larger shift compared to the absorption spectra as illustrated in ***Table 1***.

The smaller variation in the absorption shift with solvent indicates that the ground state energy distribution is not significantly affected, possibly due to the less polar nature of the dyes in the ground state than in the excited state. The values of solvent dielectric constant and refractive index (ε, n) can be correlated to Stokes shift (the shift between the ground state and excited state transitions of dyes), i.e. the large Stokes shift is also indicative of the charge transfer transition. The large magnitude of the Stokes shift indicates that the excited state geometry could be different from that of the ground state. Generally speaking, increases in the Stokes shift value are a function of an increase in solvent polarity, which indicates an increase in the dipole moment of the excitation state. In such cases, the relaxed excited singlet−state (S_1) will be energetically stabilized compared to the ground state (S_0) and a significant red shift of fluorescence will be observed. A variety of equations have been proposed [20, 21] to describe the effects of the physical properties of a solvent on the emission spectra of fluorophores. In all these treatments, the solvent is regarded as a continuum containing the fluorophore. The interactions between the solvent and fluorophore molecules affect the energy difference between the ground state and the excited state. This energy difference is a property of the refractive index (n) and dielectric constant (ε) of the solvent. Within the zero-order approximation, the solute-solvent interaction is considered to be primarily of dipole-dipole nature including dispersion interactions. This article presents different solvent parameters such as dielectric constant (ε), refractive index (*n*) [***Table 2***] and spectral parameters (e.g. Stokes shift) related to dipole moments as suggested by Kawski [21], Bakshiev[22] and Chamma−Viallet [23].

TABLE 1. Solvatochromic data of RhB and Rh6G in different solvents

Dye	Solvent	λ_a (nm)	λ_f (nm)	v_a (cm^{-1})	v_f (cm^{-1})	$\Delta v = v_a - v_f$(cm^{-1})	$\Delta v = v_a + v_f$(cm^{-1})
RhB C=10^{-5} M/L	Chloroform	563.79	579	17736.80	17271.15	465.65	17712.62
	Methanol	556.1	573.3	17982.37	17442.87	539.5	17534.94
	Dimethylsulfoxide	555.4	586	18005.04	17064.84	940.2	17503.97
R6G C=10^{-5} M/L	Ehloroform	563.8	578.9	17736.26	17271.148	465.11	18255.91
	Methanol	536.15	559.9	18651.49	17860.33	791.16	18112.73
	Dimethylsulfoxide	540	564.75	18518.51	17706.94	811.57	1793605

A simplified quantum mechanical second order perturbation theory of absorption (ν_a) and fluorescence (ν_f) band shift in different solvents of varying permittivity (ε) and refractive index (n) relative to the band position of a solute molecule was obtained based on the following equations[24]:

$$\nu_a - \nu_f = m_1 F_1(\varepsilon,n) + \text{const} \tag{2}$$

$$1/2(\nu_a + \nu_f) = - m_2 F_2(\varepsilon,n) + \text{const} \tag{3}$$

Where ν_a and ν_f are the 0-0 absorption and fluorescence maxima wave number is in cm^{-1}, respectively, the expression for $F1(\varepsilon, n)$ [Bakshiev's polarity function] and $F2(\varepsilon, n)$ [Kawski and Chamma−Viallet's polarity function] are given as:

$$F_1(\varepsilon,n) = [(2n^2 + 1)/(n^2 + 2)][\{(\varepsilon - 1)/(\varepsilon + 2)\} - \{(n^2 - 1)/(n^2 + 2)\}] \tag{4}$$

$$F_2(\varepsilon,n) = (1/2)\, F_1 + [(3/2)\{(n^4 - 1)/2(n^2 + 2)^2\}] \tag{5}$$

with

$$m_1 = 2(\mu_e - \mu_g)^2/hca^3 \tag{6}$$

$$m_2 = 2(\mu_e^2 - \mu_g^2)/hca^3 \tag{7}$$

μ_e and μ_g are the permanent dipole moments in the excited and ground states of solute molecules respectively, h is Planck's constant, c is the speed of light in a vacuum and a is Onsager's cavity radius. The parameters m_1 and m_2 can be determined from absorption and fluorescence band shifts. If the ground state and excited state are parallel then by using the above equations, the values of μ_e and μ_g can be obtained from equations 6 and 7:

$$\mu_g = \{(m_2 - m_1)/2\}(hca^3/2m_1)^{1/2} \tag{8}$$

$$\mu_e = \{(m_1 + m_2)/2\}(hca^3/2m_1)^{1/2} \tag{9}$$

The ratio of the dipole moment in excited state to ground state is equal to:

$$(\mu_e/\mu_g) = (m_1 + m_2)/(m_1 - m_2) \tag{10}$$

The parameters m_1 and m_2 occurring in the differences ($\nu_a - \nu_f$) and the sum ½($\nu_a + \nu_f$) of the wave−numbers are linear functions of the solvent polarity parameters, $F1$ (ε, n) and $F2$ (ε,n) respectively and can be determined from the slopes of the linear fits (***Table 2***). Onsager's cavity radii 'a' can be obtained from the molecular volume of molecules, given by Suppan's equation [25]:

$$a = \sqrt[3]{\frac{3M}{4\pi\rho N}} \tag{11}$$

Where M is the molecular weight of the dye; ρ is the density of the dye, assuming 1.0 gm/cm^3; N is the Avogadro's number, which was used in calculating the radii of RhB and Rh6G in this research work and was found to be equal to 0.575 nm for both RhB and Rh6G since both dyes have the same molecular weight.

Figures 9-12 show graphs of spectral shifts ($\nu_a - \nu_f$) and ½($\nu_a + \nu_f$) in cm^{-1} of RhB and Rh6G with polarity function F_1 (ε, n) and F_2 (ε, n) respectively.

TABLE 2. Calculated values of polarity functions for RhB and Rh6G dyes.

Solvents	ε	n	$F_1(\varepsilon,n)$	$F_2(\varepsilon,n)$
Chloroform	4.81	1.442	0.37	0.49
Methanol	33.000	1.374	0.856	0.651
Dimethylsulfoxide	47.24	1.478	0.842	0.744

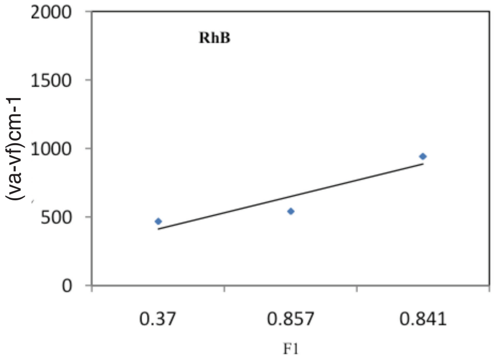


Figure 9. *The linear fit of (ν_a-ν_f) vs. F_1 (ε, n) of RhB in different solvents.*

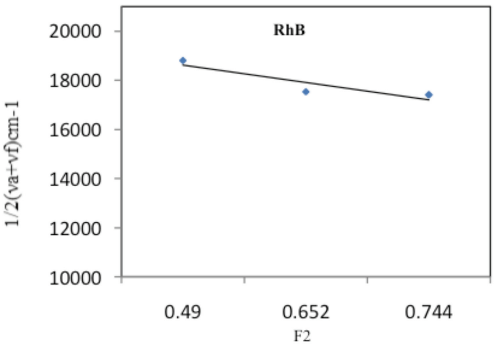


Figure 10. *The linear fit of 1/2(ν_a+ν_f) vs. F_2 (ε, n) of RhB in different solvents.*

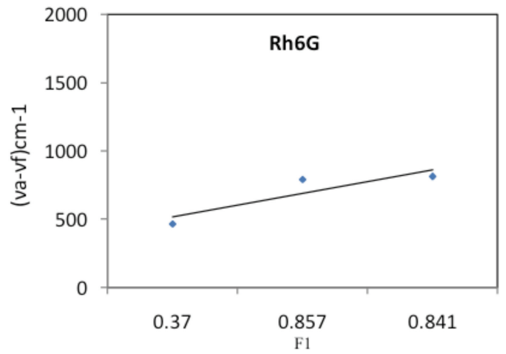


Figure 11. *The linear fit of (v_a-v_f) vs. F_1 (ε, n) of Rh6G in different solvents.*

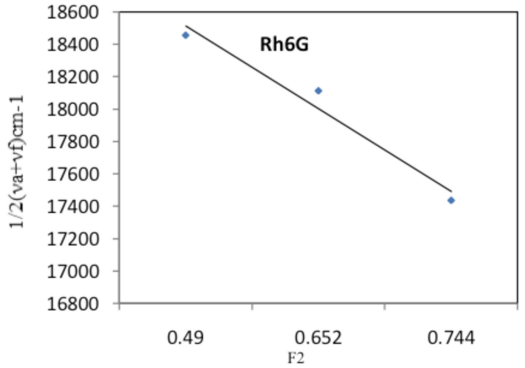


Figure 12. *The linear fit of 1/2(v_a+v_f) vs. F_2 (ε, n) of Rh6G in different solvents.*

The linear behavior of Stokes shift versus solvent polarity function indicates general solvent effects as a function of the dielectric constant and refractive index. The slopes (m_1 and m_2) were taken into account in calculating the ground and excited states dipole moment (μ_g and μ_e) of the two dyes. The change in the dipole moments (μ_g and μ_e) depends on m_1 and m_2 as well as on the radius of the solute which is equal to (in Debye, D) 2.079 and 4.194 D for RhB and 1.75 and 3.56D for R6G, respectively.

The values of change in the dipole moment ($\Delta\mu = \mu_e - \mu_g$) obtained are 2.115D for RhB and 1.805D for R6G. Obviously, the dipole moment in the excited state is significantly larger than the dipole moment in the ground state. A study of the photo-physical properties of coumarin (5BAMC and 4DHMC) molecules also showed that the excited state dipole moment (μe) is greater than the ground state dipole moment (μg). The increase in dipole moment in the excited singlet states ranges between 2.5 to 3 D and it was concluded that these two coumarins are more polar in excited states than in ground states for all the solvents studied.

The dipole moments of a molecule in the ground and excited states are different due to the changes in electron densities in these states. Therefore, it was generally observed that an increase in the stokes shift is a result of a corresponding increase in solvent polarity and an increase in the dipole moment on excitation. In spite of the solvatochromic data being used in identifying the spectra, namely π- π*, n-π* etc, it was observed that increasing solvent polarity will cause the fluorescence emission peak to undergo a bathochromic shift, confirming a π- π* transition. The shift of the fluorescence wavelengths towards longer wavelengths could be a result of the excited state charge distribution in the solute being markedly different from the ground state charge distribution, and as such giving a stronger interaction with polar solvents in the excited state. The experimental variations in the dipole moment values can also be understood in terms of their possible resonance structures.

Figures 13 and ***14*** show the variation of the quantum yield Q_f of RhB and Rh6G dyes in chloroform, methanol, and dimethyl sulfoxide as a function of concentration. It must be noted here that the quantum yield of the dye solution decreases at higher concentrations irrespective of the solvent used. The major mechanisms that may prevent the fluorescence emission at these concentrations are the triplet state absorption and aggregate formation. The rapid decrease in Q_f at higher concentrations can be mainly attributed to the formation of dimers and higher aggregates which have a zero or very small fluorescence quantum yield. The competition between increasing dimer absorption and decreasing monomer absorption with increasing concentration plays a comparable (or even greater) role than direct self-quenching of fluorescence in decreasing quantum efficiency, which is in agreement with early findings [26].

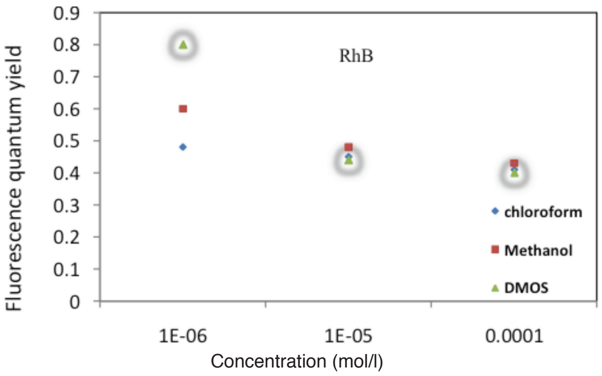


Figure 13. *The variation of Q_f with concentration of RhB dye in different solvents*

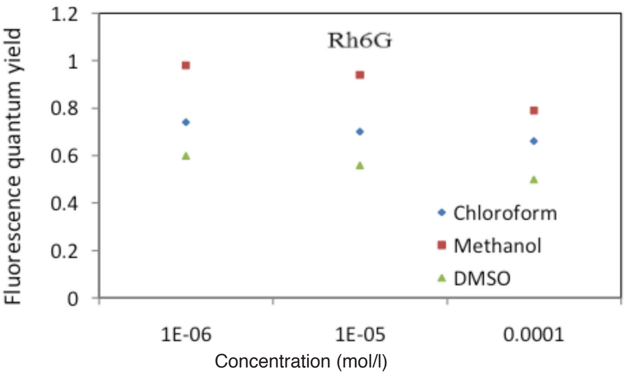


Figure 14. *The variation of Qf with concentration of Rh6G dye in different solvents*

In the present case, a significant reduction in fluorescence quantum yield can be expected due to aggregate formation at higher concentrations.

Conclusion

The following conclusions may be drawn from the above findings:

- Enhancement of the half width of the fluorescence spectrum at higher concentrations.
- The charge transfer band shows a shift in the absorption spectra on changing the solvent, whereas emission spectra shows a larger shift compared to the absorption.
- The large magnitude of Stokes shift indicates that the excited state geometry is different from that of the ground state.
- The interactions between the solvent and fluorophore molecules affect the energy difference between the ground and excited states. This energy difference is a property of the refractive index (n) and dielectric constant (ε) of the solvent.
- The linear behavior of the Stokes shift versus solvent polarity function indicates general solvent effects as a function of dielectric constant and refractive index.
- The dipole moments (μ_g and μ_e) depend not only on m_1 and m_2 but also on the radius of the solute.
- The dipole moment in the excited state is significantly larger than the dipole moment in the ground state and increasing in Stokes shift with rising solvent polarity as a result of an increase in the dipole moment on excitation.
- The quantum yield of the dye solution decreases at higher concentrations irrespective of the solvent used. This is due to the aggregate formation at higher concentrations.

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Nomenclature

RhB : Rhodamine B
Rh6G : Rhodamine 6G
Me : excited state dipole moments
Mg : ground state dipole moments
ε : solvent's dielectric constant
n : refractive index
m : mass of dye
C : concentration
V : volume
M : molecular weight of the dye
Q_f : fluorescence quantum yield
Pfw : peak fluorescence wavelength
TL : thermal lens technique
λ_a : absorption maxima wavelength
λ_f : emission maxima wavelength
ν_a : absorption maxima wave numbers
ν_f : emission maxima wave numbers
S_1 : excited singlet−state
S_0 : ground state
h : Planck's constant
c : speed of light in a vacuum
a : Onsager's cavity radius
ρ : density of dye
N : Avogadro's number
$\Delta\mu$: change in dipole moment
π- π^* : molecular electronic transitions [transition of an electron from a π-bonding orbital to an anti bonding π^* orbital* is denoted as a $\pi \rightarrow \pi^*$ transition.]
$F_1(\varepsilon, n)$: Bakshiev's polarity function
$F_2(\varepsilon, n)$: Kawski and Chamma−Viallet's polarity function