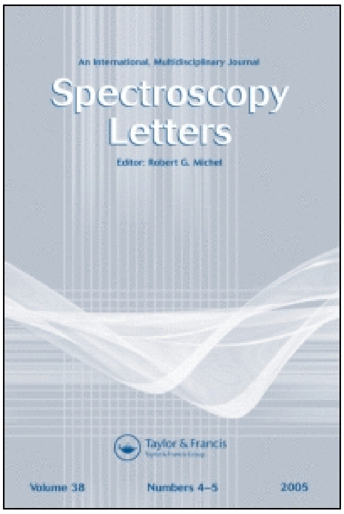
This article was downloaded by:
On: *30 January 2011*
Access details: *Access Details: Free Access*
Publisher *Taylor & Francis*
Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



**Spectroscopy Letters**
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713597299

# ENERGY TRANSFER MECHANISMS IN LASER DYE MIXTURES

S. R. Inamdar[a]; B. G. Mulimani[a]; M. I. Savadatti[a]; A. V. Sapre[b]; T. Mukherjee[b]
[a] Laser Spectroscopy (DRDO/KU), Department of Physics, Karnatak University, Dharwad, India [b] Radiation Chemistry & Chemical Dynamics Division, Chemistry Division, Bhabha Atomic Research Centre, Bombay, India

Online publication date: 13 June 2002

**To cite this Article** Inamdar, S. R. , Mulimani, B. G. , Savadatti, M. I. , Sapre, A. V. and Mukherjee, T.(2002) 'ENERGY TRANSFER MECHANISMS IN LASER DYE MIXTURES', Spectroscopy Letters, 35: 2, 293 — 307
**To link to this Article: DOI:** 10.1081/SL-120003814
**URL:** http://dx.doi.org/10.1081/SL-120003814

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: http://www.informaworld.com/terms-and-conditions-of-access.pdf

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

SPECTROSCOPY LETTERS, 35(2), 293–307 (2002)

Downloaded At: 03:09 30 January 2011

# ENERGY TRANSFER MECHANISMS IN LASER DYE MIXTURES

**S. R. Inamdar,[1,]* B. G. Mulimani,[1] M. I. Savadatti,[1]**
**A. V. Sapre,[2] and T. Mukherjee[2]**

[1]Laser Spectroscopy (DRDO/KU), Department of
Physics, Karnatak University, Dharwad 580 003, India
[2]Radiation Chemistry & Chemical Dynamics Division,
Chemistry Division, Bhabha Atomic Research Centre,
Trombay, Bombay 400 085, India

## ABSTRACT

Measurements of fluorescence intensities in optically thick (lasing) and thin (non-lasing) solutions and fluorescence lifetimes of donor in binary mixtures of laser dyes have been made. Various energy transfer parameters such as energy transfer rate parameters, critical transfer radii and mean diffusion lengths for four pairs of dyes have been determined. Fluorescence intensities measured for optically thick solutions were corrected to account for radiative transfer and direct absorption of excitation by the acceptor. Energy transfer rate parameters obtained from intensity as well as lifetime measurements strongly support the dominance of energy transfer via resonance mechanism.

---

*Corresponding author. E-mail: him_lax3@yahoo.com

Copyright © 2002 by Marcel Dekker, Inc.

*Key Words*: Energy transfer; Rate constant; Fluorescence lifetime; Critical transfer radius; Mean diffusion length; Half-quenching concentration

## INTRODUCTION

Energy transfer (ET) has been studied extensively in many solvent-solute[1–3] and solute-solute[4–6] systems. It deals with excitation of one molecule (donor), optically or otherwise, which then transfers its excitation energy to the second molecule (acceptor) that in turn luminesces. The prerequisite for such a system to work efficiently is substantial overlap of absorption spectrum of the acceptor with the emission spectrum of the donor. Study of energy transfer has been found to be useful in improving the performance of the dye lasers and in the operation of energy transfer dye lasers (ETDLs) simultaneously at two bands[7,8]. There have been many attempts to understand the energy transfer mechanisms operating in such binary mixtures in different concentration regimes[5,9,10] and to quantify the contributions of radiative and nonradiative ET mechanisms[11]. Conte and Martinho[11] have developed a theory to account for direct absorption of excitation by the acceptor and radiative transfer from donor to acceptor. As part of our ongoing programme on photophysical characterization of laser dyes, we have analyzed the influence of these factors on the energy transfer rate parameters for four binary mixtures of dyes in the light of above theory. This is important from the point of view of understanding the mechanisms of ET operating in binary mixtures that are generally employed in ETDLs (optically thick solutions) and non-lasing (optically thin) solutions. All the four mixtures described in the present paper have been shown to be efficient ETDLs with enhancement in output power, improvement in tunability, concentration- and pump-threshold[12,13].

Downloaded At: 03:09 30 January 2011

## THEORY

The principal energy transfer mechanisms that have been established[14] as operating in binary mixtures are: (i) radiative transfer—where the photon emitted by the excited donor (D*) is absorbed by the acceptor (A) which fluoresces, (ii) resonance transfer (Forster mechanism)—where the excitation energy of the donor is transferred to the acceptor over a distance through resonant dipole-dipole interaction and (iii) collisional transfer—where the excitation energy of the donor is transferred to the acceptor molecule on collision.

Downloaded At: 03:09 30 January 2011

If $I_D^0$ and $I_D$ are the experimentally determined fluorescence intensities of donor in the absence and in the presence of acceptor respectively, then,

$$I_D^0/I_D = 1 + k_{et}\,\tau_D^0\,[A] \tag{1}$$

where the square bracket denotes concentration. $\tau_D^0$ is the fluorescence lifetime of donor in the absence of acceptor and $k_{et}$ the energy transfer rate parameter, which includes contribution from all the three mechanisms mentioned above. With a view to give a better description of non-radiative transfer process, Conte and Martinho[11] developed theoretical expressions to account for radiative transfer and direct absorption of the excitation by the acceptor, which become important at high concentrations where non-radiative transfer and trapping of radiation are dominant. They showed that,

$$I_D(\nu_{exc},\, \nu_{em}^D) = I_D^t(\nu_{exc},\, \nu_{em}^D)\,\frac{a(\nu_{exc}) + a'(\nu_{exc},\, \nu_{em}^D)}{a(\nu_{exc})}, \tag{2}$$

where $I_D^t(\nu_{exc},\, \nu_{em}^D)$ is the actual donor fluorescence intensity observed at donor's emission wavenumber $\nu_{em}^D$ when excited by wavenumber $\nu_{exc}$. $I_D$ $(\nu_{exc},\, \nu_{em}^D)$ is the intensity which would result if radiative transfer and direct absorption of the excitation by the acceptor are absent and

$$a(\nu_{exc}) = \mu_D(\nu_{exc})[D]\sin\beta \tag{3}$$

$$a'(\nu_{exc},\nu_{em}^D) = [\mu_A(\nu_{exc})\sin\beta + \mu_A(\nu_{em}^D)\sin\alpha]\,[A]; \tag{4}$$

$\mu_D(\nu_{exc})$ and $\mu_A(\nu_{exc})$ being the molar absorptivities of donor and acceptor molecules at the excitation wavenumber $\nu_{exc}$, respectively. $\mu_A(\nu_{em}^D)$ is the molar absorptivity of A at wavenumber of donor emission, $\alpha$ is the angle subtended by the incident ray with the surface of the solution and $\beta$ the angle formed by the emitted radiation with the surface of solution.

The decrease in fluorescence lifetime of donor with increasing concentration of the acceptor is a direct evidence of the non-radiative ET process. The measured fluorescence lifetime of donor in the absence ($\tau_D^0$) and in the presence ($\tau_D$) of varying concentrations of acceptor are generally found to obey Stern-Volmer relationship:

$$\tau_D^0/\tau_D = 1 + k_{et}\,[A]\tau_D^0, \tag{5}$$

where $k_{et}$ is the excitation transfer rate parameter for ET due to resonance mechanism. The plot of $\tau_D{}^0/\tau_D$ versus [A] yields $k_{et}$. The same plot can be used to determine the concentration of A at which the donor lifetime becomes equal to $\tau_D{}^0/2$. This value of [A], known as half quenching concentration $[A]_{1/2}$, may be used to determine the critical transfer radius, $R_0$, defined as distance of separation of donor and acceptor molecules for which energy transfer from $D^* \rightarrow A$ and emission from $D^*$, are equally probable[15]. It is expressed as

$$R_0 = 7.35/[A]_{1/2}^{1/3}. \tag{6}$$

$R_0$ can also be calculated from spectral data using the relation

$$R_0 = \frac{9000\,(\ln 10)\,K^2 \phi_D}{128\pi^5 n^4 N} \quad \frac{F_D(\nu)\,\varepsilon_A(\nu)}{\nu^4} d\nu, \tag{7}$$

where $F_D(\nu)$ is the fluorescence spectrum of the donor, $\varepsilon_A(\nu)$ is the absorption spectrum of the acceptor, (both measured on the wavenumber scale), $\phi_D$ is the fluorescence quantum yield of donor, n is refractive index of the medium and N is Avogadro's number. K denotes the orientation factor and is $(2/3)^{1/2}$ for random orientation of molecules.

Often, in literature, in the study of ET, the values of $R_0$ are compared with the mean diffusion length 'd' of donor and acceptor in binary mixtures to identify the ET mechanism. This mean diffusion length is defined as[16]

$$d = (2D\tau_D^0)^{1/2} \tag{8}$$

with $D = D_D + D_A$. Here $D_D$ and $D_A$ are the diffusion coefficient of donor and acceptor, respectively. D is given by Stokes-Einstein relation

$$D = k_B T/6\pi\eta r,$$

with $\eta$ the viscosity of the medium, r the radius of the diffusing molecule, $k_B$ the Boltzmann's constant and T the absolute temperature.

Comparing d and $R_0$ the studies of ET can be divided into three regimes[16]:

(i) a diffusion controlled Stern-Volmer region where $d > R_0$,
(ii) an intermediate region where $d \sim R_0$,
(iii) resonance transfer or Forster region where $d < R_0$.

Downloaded At: 03:09 30 January 2011

## MATERIALS AND METHODS

Absorption spectra were recorded on a Hitachi (UV/vis) spectrophotometer (Model 150-20). Fluorescence spectra were recorded on a fluorescence spectrophotometer set up in our lab (Fig. 1) comprising a 400 W Hg lamp with a monochromator $M_1$, used as the excitation source and a quartz cylindrical lens to focus the light onto a 1 sq.cm. quartz cell. The cell holder was so mounted that it could be adjusted with ease for both front surface and perpendicular configurations. A scanning monochromator and a photomultiplier collected the fluorescence emission and the spectra were recorded on an X-Y recorder (Riken-Denshi Model F-3DG). This experimental arrangement was specifically designed for the present study and calibrated by recording the known fluorescence spectra of standard dyes, rhodamine 6G, fluorescein, etc. As compared to commercial fluorescence spectrophotometers this set-up has the flexibility of recording spectra in front surface configuration, which is essential for solutions at higher concentrations.

Fluorescence lifetimes of donor without and with different concentrations of acceptor were measured using Edinburgh's Model 199 Fluorescence Time Domain Spectrometer.

The dyes chosen for the present study, coumarin 1 (C1), coumarin 102 (C102) and uranine (Ur) were obtained from Exciton Chemical Company,

Downloaded At: 03:09 30 January 2011

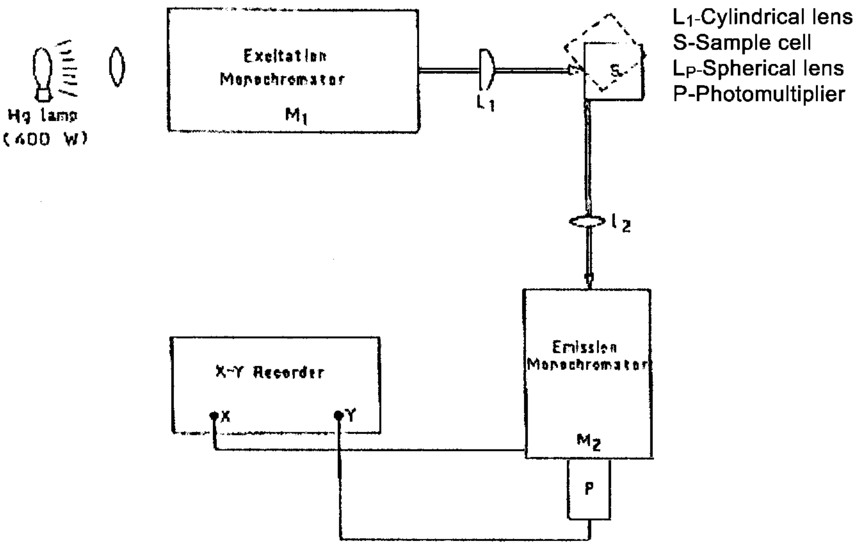


*Figure 1.* Experimental setup to record fluorescence spectra.

USA and acridine orange (AO) from E. Merck, Germany, and were used without further purification. Ethanol (Fluka, Germany) was used as a solvent in all the experiments.

The following four combinations of dyes were chosen for the present study:

| Pair | Donor (D) | Acceptor (A) |
|---|---|---|
| I | coumarin 1(C1) | uranine (Ur) |
| II | coumarin 102 (C102) | uranine (Ur) |
| III | coumarin 1 (C1) | acridine orange (AO) |
| IV | coumarin 102 (C102) | acridine orange (AO) |

C1, C102 and Ur are laser dyes and AO is useful as biological stain. AO is very important for the present study as it does not lase on its own when pumped by nitrogen laser but lases as energy transfer dye laser in combination with C1, C102 and C339 as donors[13,17]. It has also been used as a fluorescent intercalator in the study of fluorescence quenching by DNA molecule[18].

Downloaded At: 03:09 30 January 2011

## RESULTS AND DISCUSSION

The absorption spectra for all these dyes and their mixtures were recorded at low concentration ($5 \times 10^{-5}$ M). The donors exhibit good absorption at the excitation wavelength, 365 nm, whereas acceptors weakly absorb (Fig. 2).

Note that the individual absorption bands of D and A are present in the mixture too, indicating absence of complex formation in the mixture at these concentrations.

The fluorescence spectra of the individual dyes reveal the fact that coumarins exhibit almost no red shift due to concentration whereas uranine and acridine orange show appreciable red shift with increasing concentration indicating self-absorption. The fluorescence spectra of mixtures were recorded in perpendicular as well as front surface configurations.

### (1) Perpendicular Configuration

The fluorescence spectra of dye mixtures (Pairs I through IV) with concentrations of donor fixed at $1 \times 10^{-4}$ M and varying concentrations of acceptor (optically thin solutions) were recorded in the perpendicular configuration. Figure 3 depicts the fluorescence spectra for mixtures of C1 and Ur (Pair I).

Downloaded At: 03:09 30 January 2011

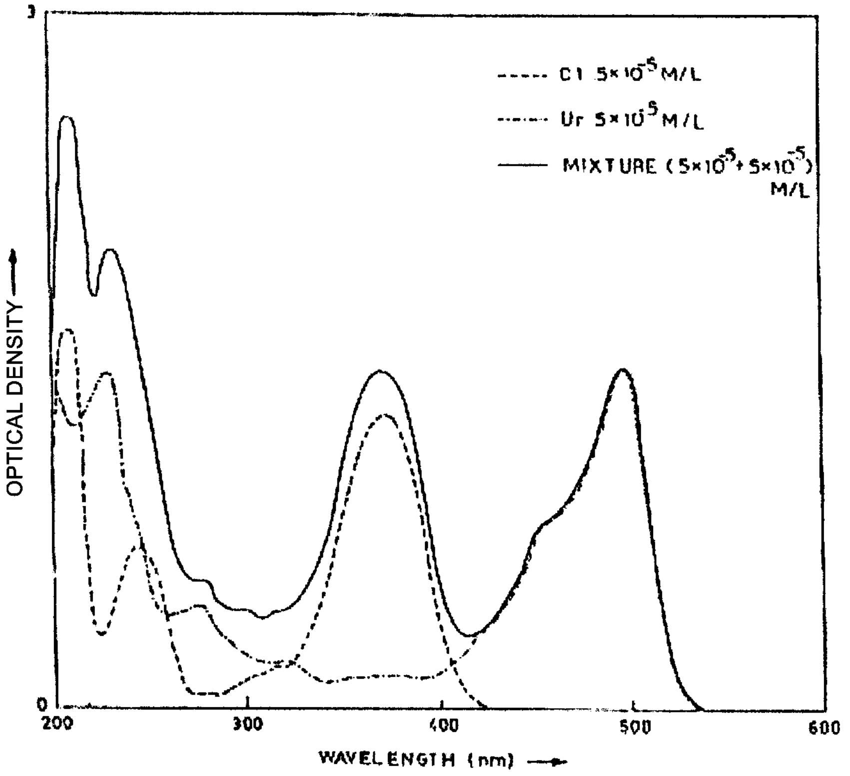


*Figure 2.* Absorption spectra of Cl, Ur, and mixture.

The peak intensities of the measured donor fluorescence emission were corrected for sensitivity of the detector and used to plot $I_D^0/I_D$ vs. [A] (Fig. 4).

The ET rate parameters $k_{et}$ were determined from these linear plots. However, no corrections for radiative transfer and direct absorption of excitation by the acceptor were applied since such corrections are not available for this experimental configuration. Though Eq. (2) is valid for measurements made in reflection (for high O.D. solutions) there are reports of the use of this expression in the low concentration regimes, where they are not valid[19,20].

## (2) Front Surface Configuration

The fluorescence spectra of dye mixtures for all the four pairs (optically thick solutions) with concentration of donor maintained constant at

Downloaded At: 03:09 30 January 2011

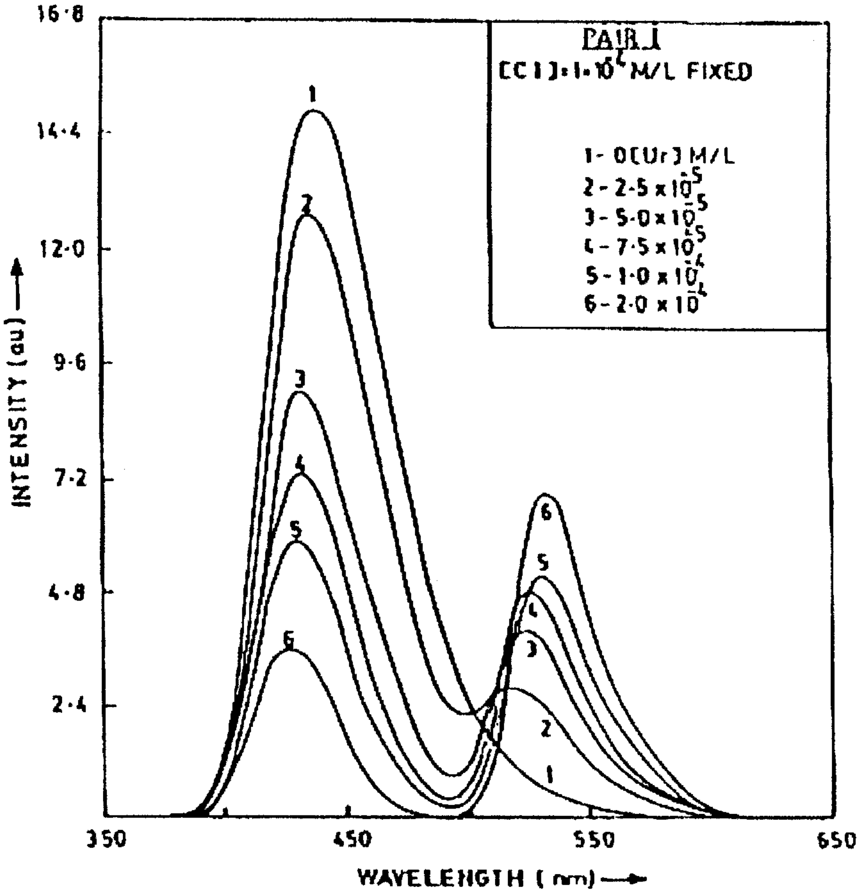


*Figure 3.* Fluorescence spectra of C1 and Ur mixtures recorded in perpendicular configuration (not corrected for spectral sensitivity of detector).

$5 \times 10^{-3}$ M and that of acceptor varied from of $10^{-5}$ to $10^{-3}$ M were recorded in the front surface configuration (with $\alpha = 30°$ and $\beta = 60°$).

Typical fluorescence curves obtained for pair II are shown in Fig. 5. The fluorescence intensities of donor emission were corrected for spectral sensitivity of the detector, for radiative transfer and direct absorption of excitation by the acceptor using Eq. (2). These corrected $I_D$ values were employed to plot $I_D{}^0/I_D$ vs. [A] and the plots were linear in the range of concentrations studied (Fig. 6). The ET rate parameters deduced, along with those for perpendicular configurations are presented in Table 1.

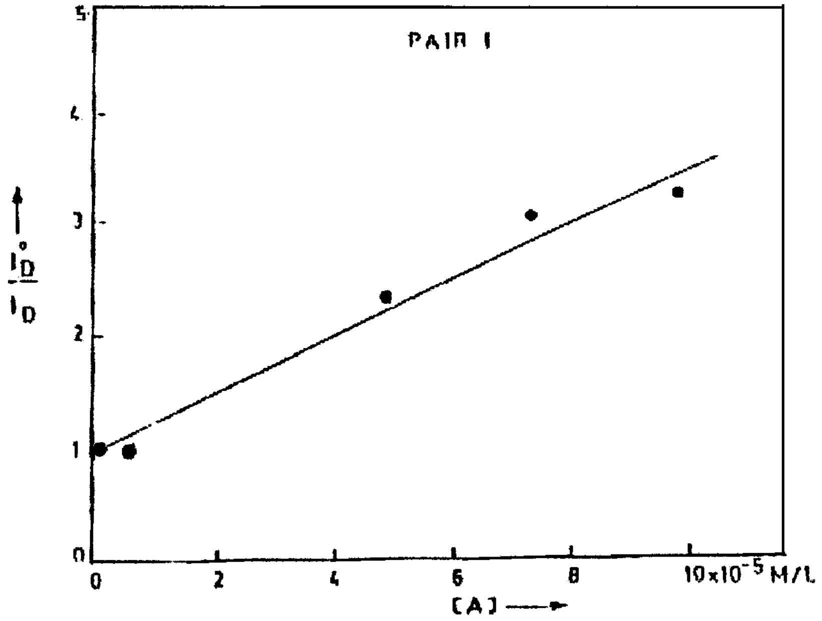


*Figure 4.* Stern-Volmer plot.

### (3) Fluorescence Lifetimes

Fluorescence lifetimes of donors without ($\tau_D^0$) and with ($\tau_D$) acceptor were studied for the first three pairs of dyes. The lifetimes of donors decreased with the increasing concentrations of the acceptors, which is a direct evidence of the non-radiative ET process. Figure 7 shows a fluorescence decay curve of C 1, as an example. The lifetime values obtained for individual dyes agree with the literature values (Table 2).

The plots of $\tau_D^0/\tau_D$ vs. [A] of all the pairs obeyed Stern-Volmer relation. A typical plot for pair III is shown in Fig. 8.

The non-radiative ET rate parameters obtained are shown in Table 3. $R_0$ and d, calculated using Eqs. (7) and (8), are given in Table 4.

## CONCLUSIONS

The values of $R_0$ determined from the Eq. (7) and those determined from experimental data on fluorescence lifetimes are in agreement (Tables 3

Downloaded At: 03:09 30 January 2011

Downloaded At: 03:09 30 January 2011

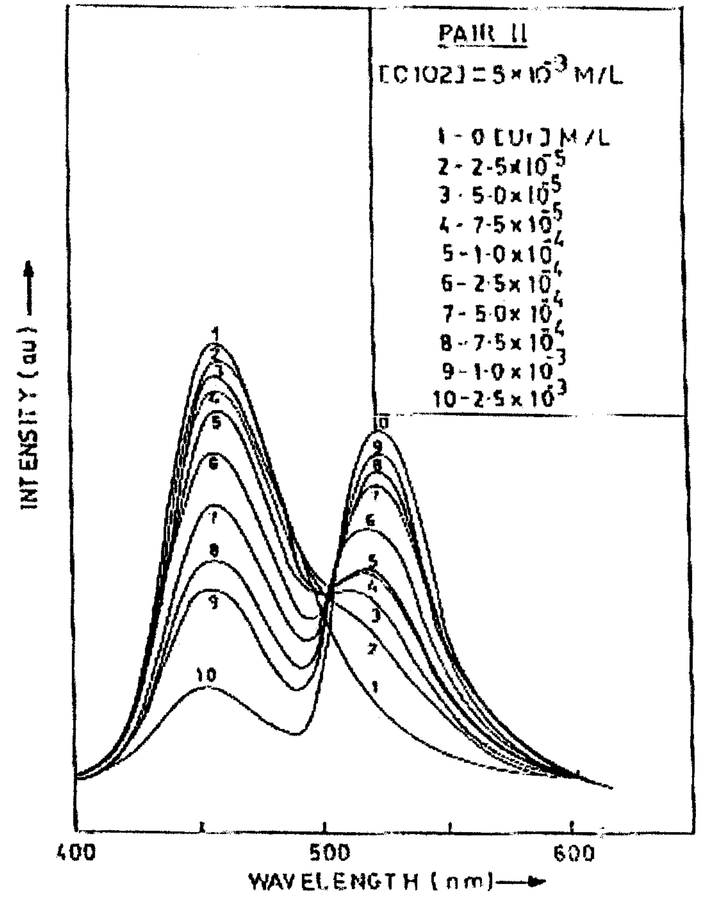


***Figure 5.*** Fluorescence spectra of C102 and Ur mixtures recorded in front surface configuration (not corrected for spectral sensitivity of detector).

and 4) for the first three pairs, and are in the range of 15–60 Å, which clearly implies the dominance of resonance mechanism[21]. Also, the fact that the $R_0$ values for all the four pairs are greater than d, further supports our argument that in all the four binary mixtures resonance transfer via long range dipole-dipole interaction is dominant.

The ET rate parameters are configuration dependent. The difference in the rates obtained from uncorrected and corrected intensities in the front surface configuration is small (Table 1) and indicates that the contribution from radiative transfer and direct absorption by the acceptor is

Downloaded At: 03:09 30 January 2011

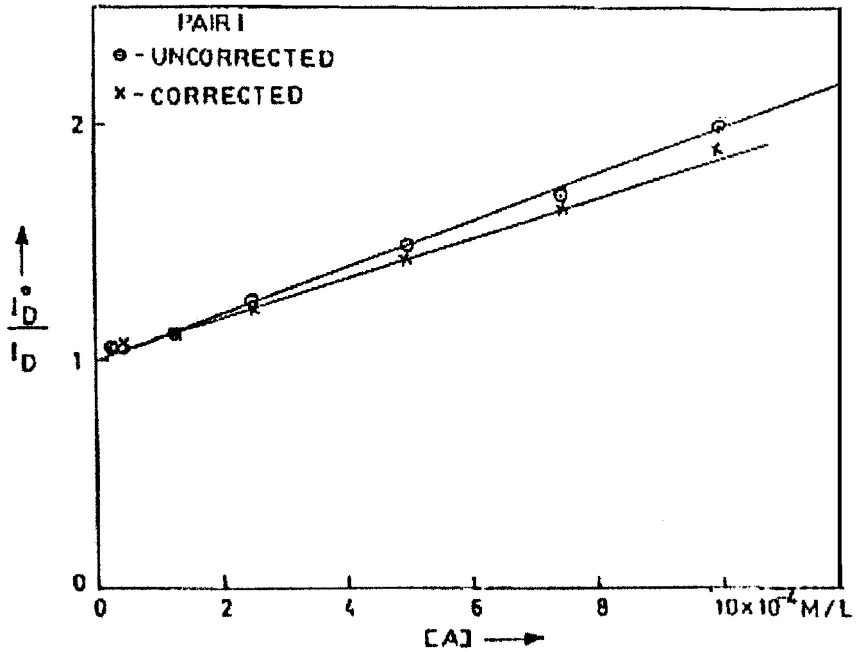


*Figure 6.* Stem-Volmer plot.

not significant at the high concentrations employed. These measurements in the front surface configuration yield rates of the order of $10^{11}\,M^{-1}\,s^{-1}$ whereas those from perpendicular configuration yield an order of magnitude higher ET rates for the same mixtures. The rates determined from

***Table 1.*** Energy Transfer Rate Parameters Obtained from Perpendicular and Front Surface Configuration

| | $k_{et}$ $(10^{11}\,M^{-1}\,s^{-1})$ | | |
|---|---|---|---|
| | | Front Surface Configuration | |
| Pair | Perpendicular Configuration | Uncorrected | Corrected |
| I | 51.8 | 3.12 | 2.61 |
| II | 99.6 | 3.82 | 3.20 |
| III | 78.7 | 1.13 | 1.01 |
| IV | 126.5 | 1.23 | 0.78 |

Downloaded At: 03:09 30 January 2011

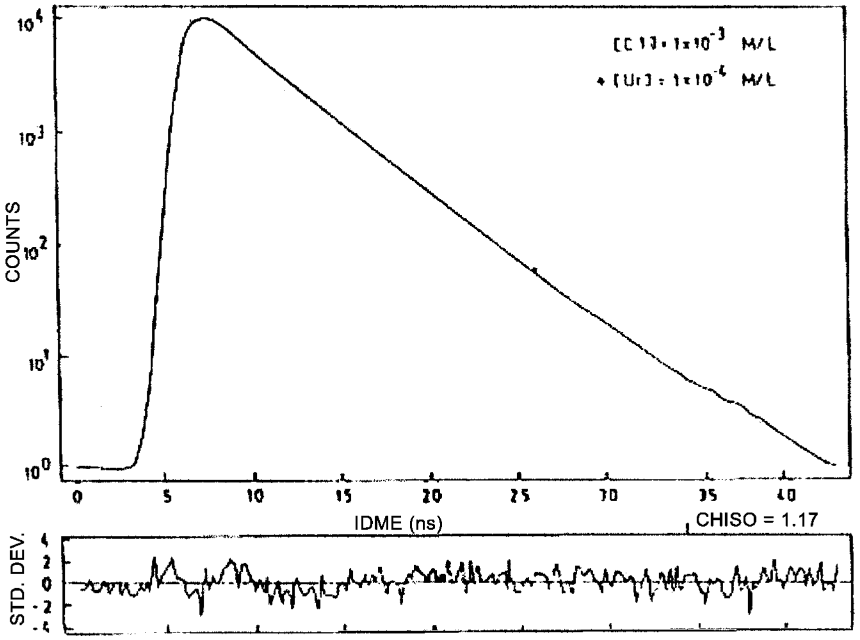


*Figure 7.*

front surface configuration agree with those determined from lifetime data indicating the dominance of non-radiative ET mechanism and are also supported by many reports[22,23]. Nevertheless, the high values for rate parameters obtained from perpendicular configurations may be useful to assess the laser performance since the laser and superradiance intensity values also yield high transfer rates[17,23,24] of the order of $10^{12}$ and $10^{13}\,M^{-1}\,s^{-1}$. It may also be noted that values of the order of $10^{12}\,M^{-1}\,s^{-1}$ for $k_{et}$ have been obtained for coumarin 339+AO pair by examining laser induced fluorescence using $N_2$ laser excitation[17]. The process of radiative

***Table 2.*** Fluorescence Lifetimes of Donors

| | Lifetime (ns) | |
|---|---|---|
| Dye | Measured | Literature[13] |
| C 1 | 3.18 | 3.10 |
| C 102 | 4.36 | 4.50 |

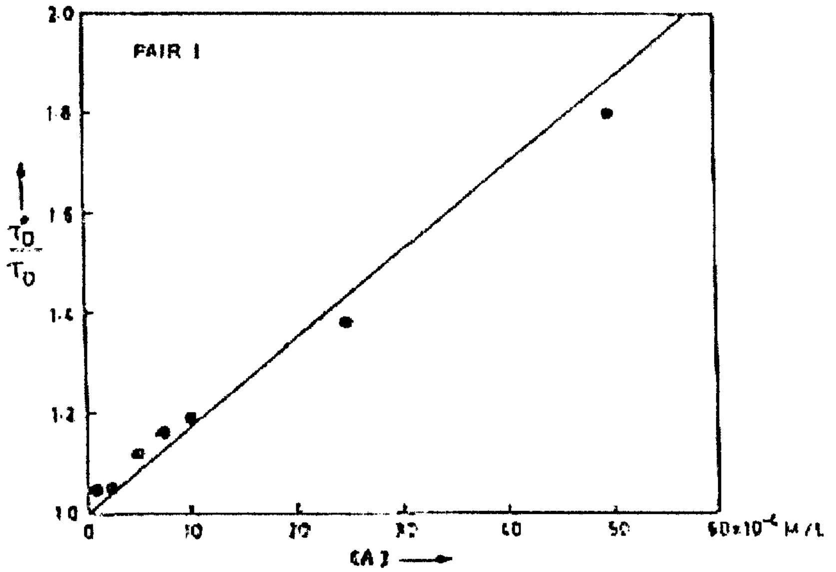


*Figure 8.* Stern-Volmer plot.

*Table 3.* Non-radiative Energy Transfer Parameters Obtained Using Lifetimes

| Pairs | $k_{et}$ ($10^{11}$ $M^{-1}s^{-1}$) | $[A]_{1/2}$ ($10^{-3}$ M) | $R_0$*(A°) |
|---|---|---|---|
| I | 0.54 | 5.77 | 40.98 |
| II | 0.51 | 4.46 | 44.85 |
| III | 0.85 | 3.70 | 47.52 |

*Calculated from Eq. (6).

*Table 4.* Experimental Values of $R_0$ and d

| Pair | $R_0$* (A°) | d** (A°) |
|---|---|---|
| I | 35.22 | 27.70 |
| II | 46.23 | 30.83 |
| III | 41.39 | 28.10 |
| IV | 46.08 | 31.19 |

*Calculated from Eq. (7).

**Calculated from Eq. (8).

Downloaded At: 03:09 30 January 2011

transfer and direct absorption of the excitation by the acceptor do contribute significantly in the perpendicular configuration as indicated by these high $k_{et}$ values. The ET rate parameter determined for collisional transfer is of the order of $10^9\,M^{-1}\,s^{-1}$ and the process does not contribute significantly. The rate parameters determined represent values due to nonradiative ET by resonance transfer process and the dominance of resonance transfer mechanism in high concentration regime in all the four pairs studied is consistent with the theory[11].

## ACKNOWLEDGMENTS

The authors are grateful to the Defence Research & Development Organization, New Delhi and the University Grants Commission, New Delhi, India, for financial support. SRI thanks Dr. N.N. Math for useful discussions.

## REFERENCES

1. Birks, J.B.; Kuchela, K.N. Energy Transfer in Fluorescent Plastic Solutions. Discu. Faraday Soc. **1959**, *27*, 57–63.
2. Birks, J.B.; Cameron, A.J.W. Energy Transfer in Organic Systems I. Photofluorescence of Terphenyl-Toluene Solutions. Proc. Phys. Soc. **1958**, *72*, 53–64.
3. Shailaja, M.K.; Hanagodimath, S.M.; Kadadevarmath, J.S.; Chikkur, G.C. Effect of Solvent Polarity on the Fluorescence Quenching of Organic Liquid Scintillators by Aniline. J. Photosci. **2000**, *6*, 159–163.
4. Berlman, I.B.; Rokni, M.; Goldschmidt, C.R. Lasing in Some Aromatic Couples by Means of Energy Transfer. Appl. Phys. Lett. **1973**, *22*, 458–460.
5. Terada, M.; Ohba, Y. Energy Transfer Mechanisms and Amplified Spontaneous Emission Characteristics of Mixture Solutions. Jap. J. Appl. Phys. **1983**, *22*, 1392–1396.
6. Ebeid, E.M.; Sabry, M.M.F.; E1-Daly, S.A. 1,4-Bis (β-pyridyl-2-vinyl) Benzene (P2VB) and 3,5-Distyryl-Pyrazine (DSP) as Blue Laser Dyes. Laser Chem. **1985**, *5*, 223.
7. Saito, Y.; Nomura, A.; Kano, T. Simultaneous Two-band Laser Emission and Three Primary Color Outputs in the Superradiant Mode from Dye Mixtures. Appl. Phys. Lett. **1988**, *53*, 1903–1904.
8. Saito, Y.; Kato, K.; Nomura, A.; Kano, T. Simultaneous Three Primary Color Laser Emissions from Dye Mixtures. Appl. Phys. Lett. **1990**, *56*, 811–813.

Downloaded At: 03:09 30 January 2011

9. Rivano, V.; Mazzinghi, P.; Burlamacchi, P. Energy Transfer in Slab Dye Lasers. Appl. Phys. **1984**, *B35*, 71–75.
10. Kaschke, M.; Stamm, U.; Vogler, K. Appl. Phys. **1986**, *B39*, 183.
11. Conte, J.C.; Martinho, J.M.G. Radiative Energy Transfer I. General Equations. J. Lumin. **1981**, *22*, 273–284.
12. Savadatti, M.I.; Inamdar, S.R.; Math, N.N.; Mulla, A.D. Energy Transfer Dye Lasers. J. Chem. Soc. Faraday Trans. 2, **1986**, *82*, 2417–2422.
13. Inamdar, S.R.; Math, N.N.; Savadatti, M.I. Fluorescence and Laser Emission from Coumarin-acridine Orange Mixtures. Spectrosc. Lett. **1993**, *26*, 359–374.
14. Birks, J.B. *Photophysics of Aromatic Molecules*; Wiley Inter-Science: NY, 1970; p. 518.
15. Turro, N.J. *Molecular Photochemistry*; W.A. Benjamin, Inc. 1965.
16. Birks, J.B.; Leite, M.S.S.C.P. Energy Transfer in Organic Systems IX. Effect of Diffusion on Transfer Efficiency. J. Phys. **1970**, *B3*, 513–525.
17. Chandrasekhar, K.; Inamdar, S.R.; Patil, D.C.; Math, N.N. Study of Intermolecular Interactions by Laser-Induced Fluorescence Measurements J. Karnatak Univ. – Science **1997**, *41*, 71–77.
18. Brun, A.M.; Harriman, A. Dynamics of Electron Transfer Between Intercalated Polycyclic Molecules, Effect of Interspersed Bases. J. Am. Chem. Soc. **1992**, *114*, 3656–3660.
19. Kushwaha, R.P.S.; Pant, D.D. Energy Transfer from Acriflavin to Eosin in Methanol with Excitation at Three Different Wavelengths. Ind. J. Phys. **1984**, *58B*, 271–283.
20. Kushwaha, R.P.S.; Pant, D.D. Energy Transfer from Acriflavin to Rhodamine 6G. Ind. J. Pure & Appl. Phys. **1984**, *22*, 636–641.
21. Lumb, M.D. *Luminescence Spectroscopy*; Academic Press: NY, 1978.
22. Lei Jie.; Fu Honglang. Radiation of Dye Mixtures Pumped by $N_2$ Laser. Chin. J. Lasers **1988**, *15*, 665–668.
23. Muto, S.; Ito, C.; Inaba, H. Continuously Tunable Energy Transfer Laser Operation in Four-Dye Mixture Systems. Electronics and Communications in Japan **1984**, *66-C*, 120–126.
24. Inamdar, S.R.; Savadatti, M.I. Influence of Experimental Configuration on Dominant Energy Transfer Mechanisms. Pramana – J. Phys. **1991**, *37*, 353–356.

Received May 27, 2001
Accepted December 21, 2001

Downloaded At: 03:09 30 January 2011