International Journal of ChemTech Research
CODEN (USA): IJCRGG ISSN: 0974-4290
Vol.8, No.3, pp 1088-1095, 2015

Tunable Distributed Feedback Dye Laser using Neutral Red and Crystal Violet Dye Mixture

G.V. Vijayaraghavan, M. Basheer Ahamed*

Department of Physics, B.S. Abdur Rahman University, Chennai – 600 048, India

Abstract: The energy transfer distributed feedback dye laser (ETDFDL) characteristics are studied both theoretically and experimentally in a mixture of Neutral Red (NR) and Crystal Violet (CV) dyes pumped by Q- switched Nd,YAG laser. The radiative and non-radiative (Forster type) energy transfer process in a dye mixture of Neutral Red (donor) and Crystal Violet (acceptor) in ethanol under Q-switched Nd,YAG laser excitation are investigated. It is found that most of the pump power absorbed by NR is transferred to CV as a useful pump power. Theoretical calculations are also done to find the energy transfer parameters viz. Critical transfer radius (R_o), Critical concentration (C_o), Half Quenching concentration ($C_{1/2}$) and Foster type transfer rate (k_f).The characteristics of energy transfer distributed feedback dye laser(ETDFDL), on input pump power and acceptor concentrations are studied in detail. The tunability of NR-CV dye mixture is achieved experimentally over a spectral range of 590 – 668 nm using prism dye cell arrangement. The output energy of DFDL is measured at the emission peaks of donor and acceptor for varied pump power and acceptor concentrations.
Keywords, Distributed feedback lasers, Dye lasers, Ultrafast processes.

1. Introduction

In recent years, distributed feedback dye lasers (DFDL) have attracted many researchers because of its potential applications in the field of science and technology [1 – 7]. Generally DFDL have higher efficiency broader tuning range and lower amplified spontaneous emission (ASE) and generates 20-100 times ultra short pulses [8, 9]. Literature surveys reveal that the mixture of co doping of one dye with other dye in liquid medium with one acting as the donor and the other acting as the acceptor enhances the laser efficiency and the tunability of the wavelength of the acceptor due to the energy transfer process [10 –16]. The principle of this process for the dyes includes radiative and nonradiative energy transfer [17, 18]. The mechanism of the nonradiative energy transfer in donor- acceptor pairs can be classified into two types namely diffusion controlled energy transfer and resonance energy transfer. The energy transfer dye lasers (ETDLs) are more efficient because of high gain and low pump power requirements [19-21].

In the present work, a theoretical model [9] is taken to study the characteristics of energy transfer distributed feedback laser (ETDFDL) using Neutral Red (NR) and Crystal Violet (CV) dye mixture in the liquid medium as a function of pump power, donor and acceptor concentration by both radiative and non-radiative (Forster) transfer. The experimental results obtained from ETDFDL using NR and CV dye mixtures are also discussed in detail.

2 Materials used

The Neutral Red and Crystal Violet laser dyes are purchased from Central Drug House, Mumbai. The solvent ethanol is used in the study of spectroscopic data. The general formulae and structures are of Neutral

Red (C.I. 50040, eurhodin) and Crystal Violet (C.I. 42555, triarylmethane) are illustrated in Fig.1 The absorption and Fluorescence spectra are taken using 0.01mM concentration of NR and CV dyes using UV/ VIS spectrometer (Perkin Elmer-LS25) and fluorescence spectrometer (Perkin Elmer-LS45) respectively.

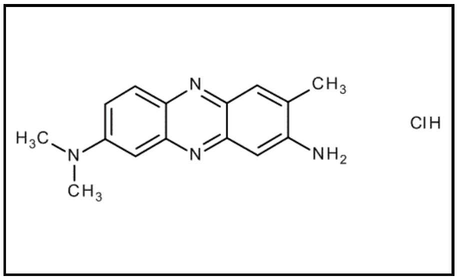


Fig. (a) Molecular Formula, $C_{15}H_{17}ClN_4$, Molecular weight, 288.78 g mol^{-1}

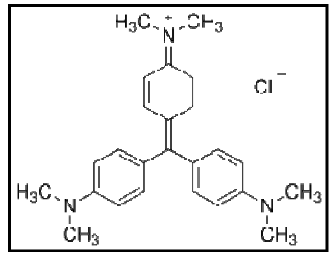


Fig. (b) Molecular Formula, $C_{25}H_{30}N_3Cl$, Molecular weight, 407.98 g mol^{-1}

Fig.1 Molecular formulae and chemical structures of (a) Neutral Red and (b) Crystal Violet

3. Theoretical studies

3.1 Simulation Results

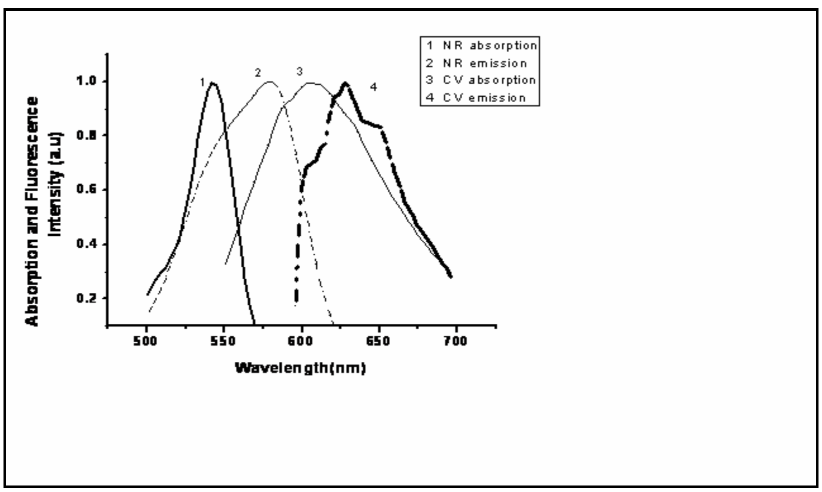


Fig.2 Schematic diagram of absorption and fluorescence spectra of Neutral Red and Crystal Violet.

The absorption and fluorescence spectrum of dye mixtures are carried out and as shown in Fig.2. From the spectrum, it is observed that there is an overlap between absorption and fluorescence of donor or acceptor themselves which indicates the formation of donor-donor (DD) or acceptor-acceptor (AA) transportation. This suggests its suitability for the energy transfer between these two dyes. The critical transfer radium (R_o), critical concentration (C_o) and half quenching concentration ($C_{1/2}$) are calculated by using the spectral datas of NR and CV dyes. The spectral parameter of NR and CV dyes are calculated by using absorption and fluorescence spectra, which is shown in Table 1. The theoretical studies are done using the parameters (in Table1) obtained experimentally from the observed absorption and fluorescence spectra of NR and CV. The dyes NR and CV are chosen as active medium and second harmonic of Q-switched Nd,YAG laser (532nm, 6 ns pulse duration, 10Hz repetition rate) is used as pump source. The donor dye molecules are found to emit DFDL in the wavelength range 580-633 nm with its peak emission at 582 nm.Similarly, the acceptor dye molecules in the dye mixture emits laser emission in the wavelength range of 615 to 668 nm with its peak at 625 nm. Figs.3-4 shows the threshold of the first and other pulses and its starting time as a function of pump power acceptor concentrations. The figures indicate that there is an increase in pump power that causes increase output power of DFDL with decrease in pulse width. Similarly, the increase in the acceptor concentration decreases the donor peak power as shown in the fig. 5. Fig. 6 illustrates that as the acceptor concentration increases, the number of DFDL pulses and the peak power increases with decrease of pulse width.

Table 1. Spectral parameters of Neutral Red and Crystal Violet used in the rate equation model

Symbols	Meaning of the symbol	Numerical Value
τ_d	Donor lifetime	3.0807ns
τ_a	Acceptor lifetime	2.509ns
R_0	Critical Transfer Radius	39.68Å
C_o	Critical concentration	$6.89 \times 10^{18} cm^{-3}$
$C_{1/2}$	Half quenching concentration	$3.45 \times 10^{18} cm^{-3}$
k_F	Forster type transfer rate	$2.53 \times 10^{-10}\ cm^3/s$
σ_{pd}	Absorption cross section of donor dye at the pumping wavelength (532nm)	$0.356 \times 10^{-16} cm^{-2}$
σ_{pa}	Absorption cross section of acceptor dye at the pumping wavelength (532nm)	$0.335 \times 10^{-16} cm^{-2}$
σ_{edl}	Emission cross section of donor dye molecules at the lasing wavelength ($\lambda_l = 582$nm) of donor	$5.5177 \times 10^{-16} cm^{-2}$
σ_{aal}	Ground-state absorption cross-section of the acceptor dye molecules at the lasing wavelength (λ_l=582 nm)	$0.168 \times 10^{-16} cm^{-2}$
σ_{eal}	Emission cross section of donor dye molecules at the lasing wavelength ($\lambda_l = 582$nm) of donor	$4.95 \times 10^{-16} cm^{-2}$
σ_{eaa}	Emission cross section of acceptor dye molecules at the lasing wavelength (λ =625nm)	$5.468 \times 10^{-16} cm^{-2}$
L	Length of the transversely excited region	0.9cm
b	Height of the transversely excited region	0.02cm
c	Speed of light	$3 \times 10^{10} cm\ s^{-1}$
n	Refractive index of the solvent	1.3628
n_p	Refractive index of the prism	1.52
V	Visibility of the interference pattern	0.4
S	Spectral factor contributing spontaneous emission	10^4
Ω_d Ω_a	Factor determining the fraction of the spontaneously emitted photons by excited donor and acceptor dye molecules respectively.	1.227×10^{-9} 0.39×10^{-7}

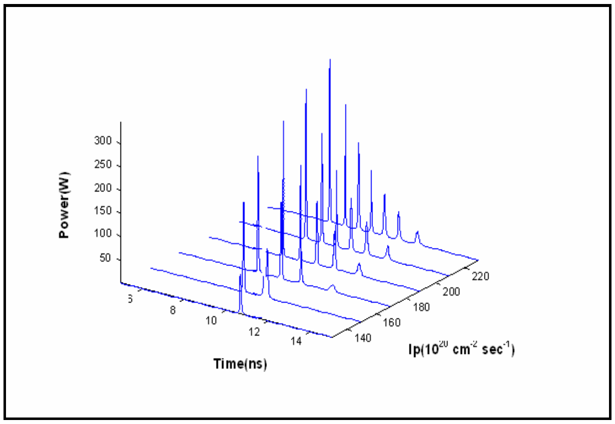


Fig.3 Variation in pulse width and peak output power of donor DFDL at fixed donor (N_d=1.8 $\times 10^{18} cm^{-3}$) and acceptor concentration (N_a = 0.6 $\times 10^{18}\ cm^{-3}$) for different pump intensities.

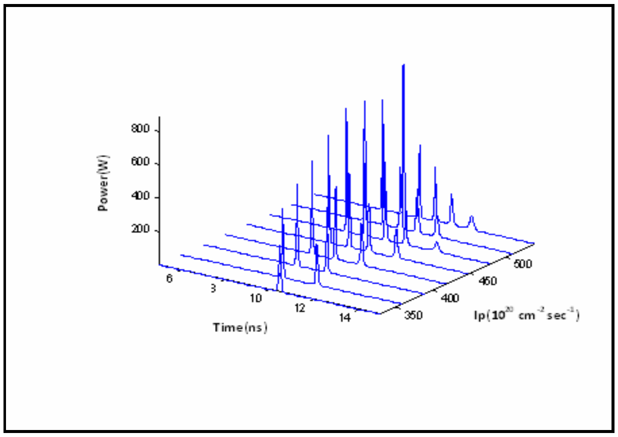


Fig. 4 Variation in pulse width and peak output power of acceptor DFDL at fixed donor (N_d=1.8 ×10^{18}cm^{-3}) and acceptor concentration (N_a = 0.6 ×10^{18} cm^{-3}) for different pump intensities.

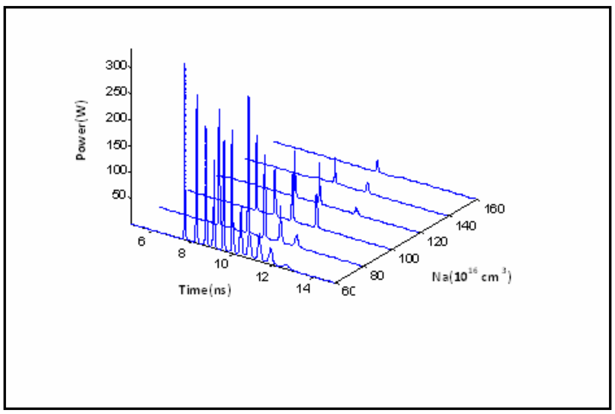


Fig.5 Variation in pulse width and peak output power of donor DFDL at fixed donor concentration (N_d= 1.8 ×10^{18}cm^{-3}) and pump intensity (I_p= 0.6 ×10^{22}cm^{-2}s^{-1}) for different acceptor concentrations.

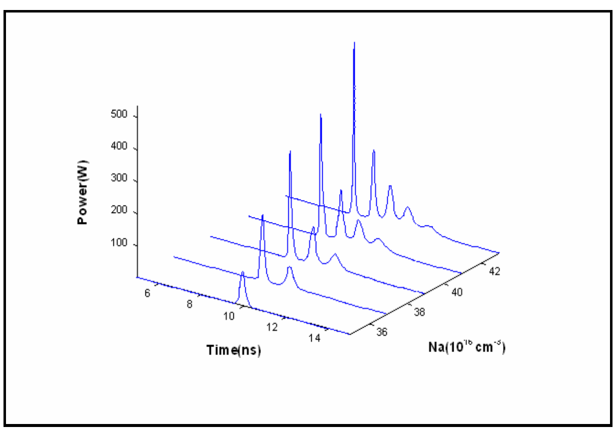
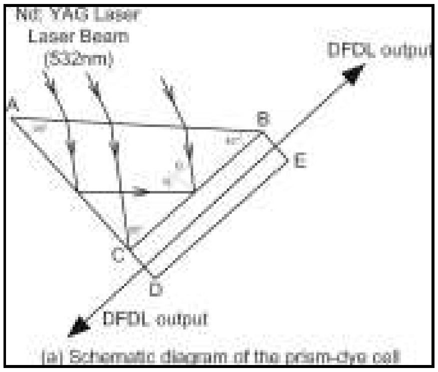


Fig. 6 Variation in pulse width and peak output power of acceptor DFDL at fixed donor concentration (N_d= 1.8 ×10^{18}cm^{-3}) and pump intensity (I_p= 0.6 ×10^{22}cm^{-2}) for different acceptor concentrations.

4. Experimental Studies

4.1 Experimental details

The distributed feedback dye laser set up is obtained by using an isosceles right –angled quartz prism, which is used to create the interference pattern on the surface of the dye cell and is shown in Fig. 7a. The DFDL is pumped by Q-switched Nd,YAG laser (QUANTA RAY Model, LAB-170-10) that emit pulses of 6ns duration at a repetition rate of 10Hz. The pump beam (532nm) is focused by a cylindrical quartz lens of focal length 5cm into a line image, which is incident on the hypotenuse AB of the prism. The light transmitted by hypotenuse of the prism is totally reflected from the side AC of the prism and interferes to form fringes on a dye cell attached to the prism producing periodic modulation of refractive index and also the gain. The output beam of the DFDL obtained from the side BC of the prism is shown in Fig.7b. The feedback is obtained from the Bragg reflection from the periodic structure incorporated throughout the active medium. The pumping beam of wavelength λ_P incident at an angle θ on the medium is given by [14, 22]



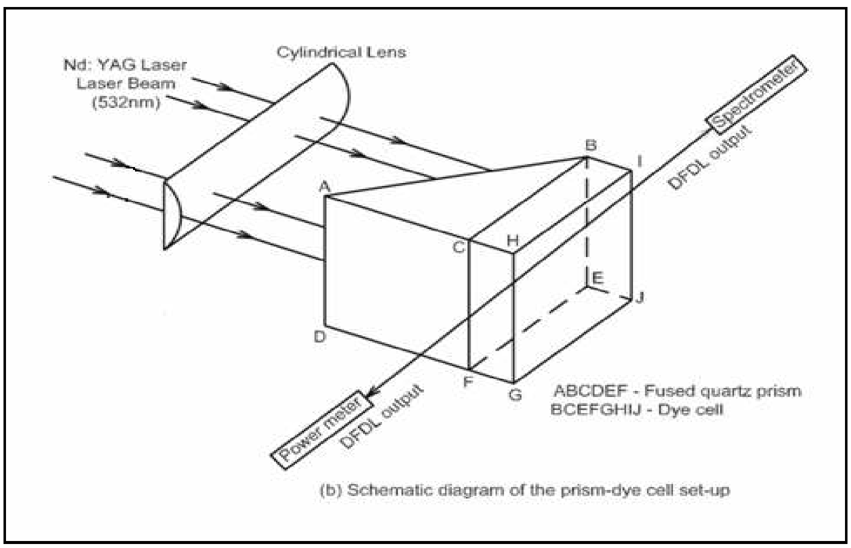


Fig.7 (a) Schematic of Prism dye cell (b) Experimental set-up of DFDL

$\lambda_{DFDL} = n\lambda_P / n_p \sin\theta$

Here n and n_p are the refractive indices of the dye solution and the prism material respectively.

5 Results and Discussion

5.1 DFDL output energy measurements

In order to obtain energy measurements, the experimental conditions are chosen that corresponds to the parameter of the simulation. Q-switched Nd,YAG laser (QUANTA RAY Model, LAB-170-10) is used as the pumping source. The input power is measured using sensor head-I (Model, J-10-LE-YAG) which is connected with the power meter (EPM 2000- Coherent Molectron, USA). Assuming that the total DFDL output power is equal on both sides of the dye cell, the output energy of the DFDL is measured at one end using sensor head-II (Model, J-50-MB-YAG) and is connected to the power meter and the emission wavelength is monitored by a spectrometer (Model, OSM2, USA). The output energy of the DFDL is measured using power meter as a function of input pump power and acceptor concentration. The output energy of donor and acceptor distributed feedback dye laser (DFDL) as a function of pump energy for fixed donor ($N_d = 1.8 \times 10^{18} cm^{-3}$) and acceptor concentration ($N_a = 0.6 \times 10^{18}\ cm^{-3}$) is shown in Fig.8. When the input energy increases, there is an increase in the output energy for the donor dye alone whereas when the acceptor dye combines with the donor dye, enhanced output energy is observed, due to good spectral overlap between the donor and acceptor dye molecules (shown in Fig.8).

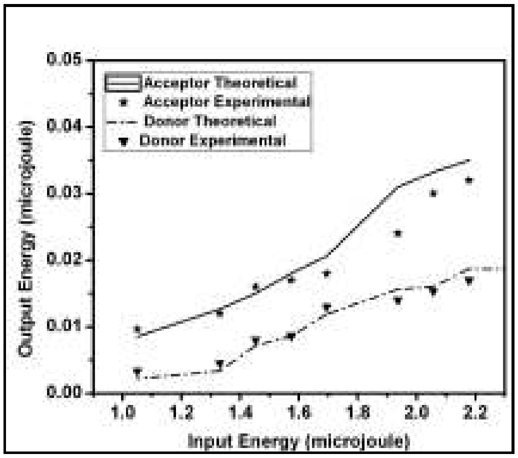


Fig. 8 Donor and acceptor DFDL output energy as a function of pump energy for fixed donor (N_d= 1.8 × 10^{18}cm^{-3}) and acceptor concentration (N_a = 0.6 ×10^{18} cm^{-3})

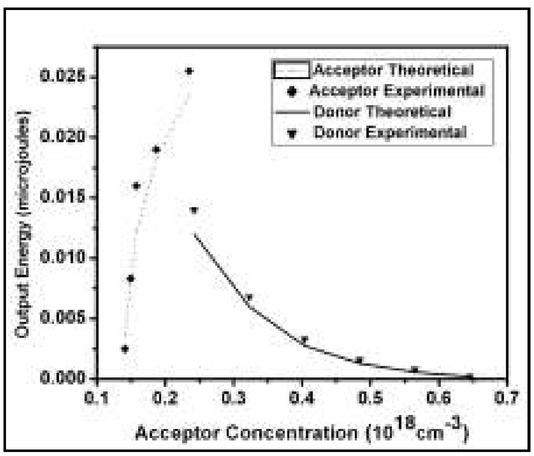


Fig. 9 Donor and acceptor DFDL output energy as a function of acceptor concentration for fixed pump power (I_p= 0.8 ×10^{22}cm^{-2}s^{-1}) and donor concentration (N_d=1.8 ×10^{18}cm^{-3})

Donor and acceptor DFDL output energy as a function of acceptor concentration for fixed pump power (I_p= 0.8 ×10^{22}cm^{-2}s^{-1}) and donor concentration (N_d=1.8 ×10^{18}cm^{-3}) is shown in Fig.9. From the figure, it is observed that when the acceptor concentration increases gradually, there is a decrease in the output energy for the donor DFDL, whereas there is an increase in the output energy in the acceptor DFDL which shows that there is an efficient energy transfer that takes place between the donor and acceptor dye molecules. The experimental values are found to be in good agreement with the theoretical values.

5.2 Tunability of DFDL

3 mM Concentration of Neutral Red solution is taken as a donor medium and its tunability is also studied by changing the angle of interference θ of the pump beam at the surface of the dye medium. It is observed that varying θ between 49^o and 54^o, DFDL is emitted with peak at 54^o. The donor (NR) and acceptor (CV) dye mixture is prepared by adding 2ml of 1mM concentration of acceptor (CV) in 1ml of 3mM concentration of donor (NR). The experiment is repeated and the dye mixture is found to lase in acceptor region for the angle of interference varying between 45^o and 49^o with its peak at 49^o.

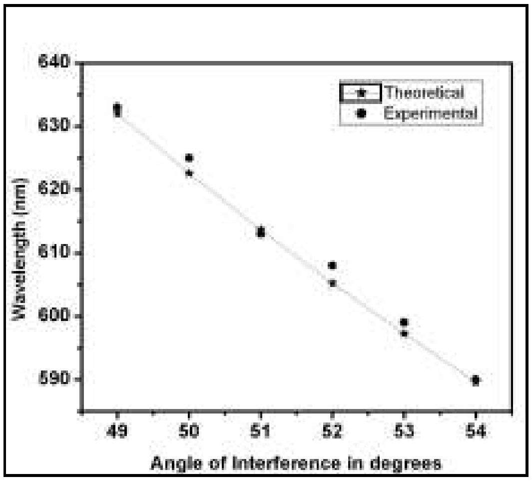


Fig.10 Donor (NR) DFDL as a function of angle of interference (θ)

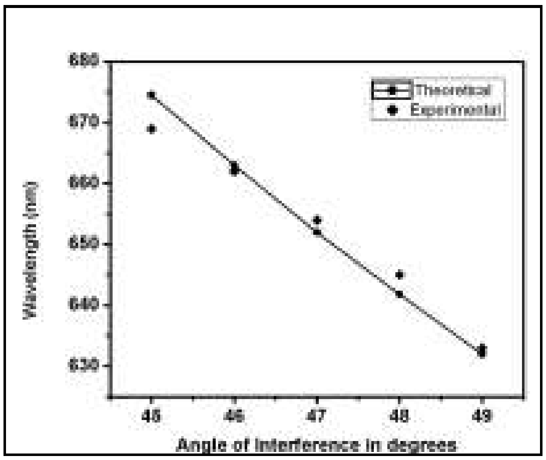


Fig.11 Acceptor (CV) DFDL as a function of angle of interference (θ)

DFDL output is tuned by changing the angle of interference of pump beam and its tunability range is measured using spectrometer (Model, OSM2). From this, it is observed that the tunability range for the donor DFDL alone is 590 to 633 nm and the addition of acceptor is extends up to 669 nm which confirms the wide tuning range when compared with that of other energy transfer dye laser [13 -14]. Fig.10 and 11 show the experimental and theoretical values of the tuning of the donor and acceptor DFDL and the angular tuning of both the donor and acceptor is found to be nearly linear.

6. Conclusion

We have analyzed in detail the energy transfer process between NR and CV dye mixture in ethanol, we found that our results could forecast suitable concentration regions for the wavelength shifts with the acceptor concentration for the dye mixture. We have observed pump power and concentration dependence of Q-switched Nd,YAG laser pumped ETDFDL in both the donor and acceptor theoretically. From the theoretical studies of donor and acceptor DFDL, it has been found that the pulse width of both DFDL pulses decrease with increase of pump power and donor concentration, the pulse width of acceptor DFDL decreases while that of donor DFDL increases. The experiment shows that the energy characteristics are in good agreement with the theoretical results. It was also observed that the continuous tunability of DFDL over yellow to red region (590 to 668 nm). This can be used in laser spectroscopy, atmospheric and underwater sensing.

References

1. Chen, Lujian, Gao, Fengyu, Bu, Yikun, Jia, Fuqiang, Liu, Chun and Cai, Zhiping, "Tunable distributed feedback lasing from leaky waveguides based on gel-glass dispersed liquid crystal thin films", Mat. Lett., 2011, 65, 3476 – 3478
2. Schneider D., Hartmann S., Benstem T., Dobbertin T., Heithecker D., Metzdorf D., Becker E., Riedl T., Johannes H.-H., Kowalsky W., Weimann T., Wang J., Hinze P., "Wavelength-tunable organic solid-state distributed-feedback laser", Appl Phys B 2003, **77**, 399 - 402.
3. Mele E, Camposeo A, Stabile R, Del Carro P, Di Benedetto F, Persano L, Roberto Cingolani and Dario Pisignano, 'Polymeric distributed feedback lasers by room-temperature nanoimprint lithography', Appl. Phys Lett. 2006, 89, 131109.
4. Chen LJ, Zhou Q, Li SS, Cai ZP, Liu S, Ren XC, "Zirconium-doped hybrid films patterned by soft lithography for distributed-feedback lasers", Appl Phys B, 2010, 101, 207 - 211.
5. He GS, Signorini R, Prasad PN, "Longitudinally two-photon pumped leaky waveguide dye film laser", IEEE J Quantum Electron, 1998, 34, 7 - 13.
6. Zhu XL, Lo D., "Sol–gel glass distributed feedback waveguide laser", Appl Phys Lett 2002, 80, 917–919.
7. Lo D, Shi L, Wang J, Zhang GX, Zhu XL., "Zirconia and zirconia-organically modified silicate distributed feedback waveguide lasers tunable in the visible",Appl Phys Lett 2002,81,2707–9.
8. Bor Z., Racz B., Muller A., "Generation of 6-psec pulses with a nitrogen-laser-pumped distributed-feedback dye laser", Appl. Opt. 1983, 22, 3327 - 3330.
9. Basheer Ahamed M., Palanisamy P.K., "Nd,YAG laser pumped energy transfer distributed feedback dye laser in Rhodamine 6G and Acid blue 7 dye mixture", Opt. Comm. 2002, 213, 67 - 80.
10. Xiaohui Li, Rongwei Fan, Xin Yu, Deying Chen, "Investigation of energy transfer between PM567,Rh610 dye mixture in modified poly (methyl methacrylate)", J. of Lumin., 2014, 145, 202 - 207.
11. Nedumpara R.J., Manu P.J., Vallabhan C.P.G., Nampoori V.P.N., Radhakrishnan P., "Energy transfer studies in dye mixtures in different solvent environments",Opt. Laser Technol. 2008, 40, 953 - 957.
12. Sahare P.D., Sharma V.K., Mohan D., Rupasov A.A.," Energy transfer studies in binary dye solution mixtures, Acriflavine + Rhodamine 6G and Acriflavine + Rhodamine B", Spectrochim. Acta A, 2008, 69, 1257 - 1264.
13. BasheerAhamed M., Ramalingam A., Palanisamy P.K., "Studies on widely tunable ultra-short laser pulses using energy transfer distributed feedback dye laser", J. of Lumin., 2003, 105 (1), 9 - 20.
14. Basheer Ahamed M., Geethu Mani R.G., Vijayaraghavan G., "Tunable Energy Transfer Distributed Feedback Dye Laser Using Pyronin B and Crystal Violet Dye Mixture", Laser Physics, 2012, 22, 1469 - 1475.
15. Yang Y., Qian G.D., Su D.L., Wang Z.Y., Wang M.Q., "Energy transfer mechanism between laser dyes doped in ORMOSILs", Chem. Phys. Lett., 2005 , 402, 389 - 394.
16. Su D.L., Yang Y., Qian G.D., Wang Z.Y., Wang M.Q., "Influence of energy transfer on fluorescence and lasing properties of various laser dyes co-doped in ORMOSILs", Chem. Phys. Lett., 2004, 397, 397 - 401.
17. Kumar G.A., Unnikrishnan N.V., Kumar G.A., Unnikrishnan N.V., "Energy transfer and optical gain studies of FDS, Rh B dye mixture investigated under cw laser excitation", J. Photoch. Photobio. A, 2001, 144, 107 – 117.
18. Azim S.A., Ghazy R., Shaheen M., El-Mekawey F.,"Investigations of energy transfer from some diolefinic laser dyes to Rhodamine 110", J. Photoch. Photobio. A 2000, 133, 185 - 188.
19. Ahamed S.A., Gergerly J.S., Infante D. "Energy transfer organic dye mixture lasers", J. Chem. Phys. 1974, 61, 1584 – 1585.
20. Marason E.G., Opt. Commun., "Energy transfer dye mixture for argon-pumped dye laser operation in the 700 to 800 nm region",1982, 40, 212 – 214.
21. Peterson O.G. and Snavely B.B., 'Stimulated emission from flash lamp-excited organic dyes in polymethyl methacrylate', Appl. Phys. Lett.,1968, 12, 238-240.
22. Chandra S., Takeuchi W., Hartmann S.R., "Prism Dye Laser", Appl. Phys. Lett. 1972, 21, 144 - 146.

****