Multimode laser emission from dye doped polymer optical fiber

Mavila Sheeba,[1,*] Kannampuzha J. Thomas,[1] Mandamparambil Rajesh,[3] Vadakkedathu P. N. Nampoori,[1] Chakkalakkal P. G. Vallabhan,[2] and Padmanabhan Radhakrishnan[1]

[1]International School of Photonics, Cochin University of Science and Technology, Cochin 682 022, India
[2]Center of Excellence in Lasers and Optoelectronic Sciences, Cochin University of Science and Technology, Cochin 682 022, India
[3]School of Engineering and Mathematical Sciences, City University, Northampton Square, London EC1V 0HB, UK

*Corresponding author: sheebanambiar@gmail.com

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Multimode laser emission is observed in a polymer optical fiber doped with a mixture of Rhodamine 6G (Rh 6G) and Rhodamine B (Rh B) dyes. Tuning of laser emission is achieved by using the mixture of dyes due to the energy transfer occurring from donor molecule (Rh 6G) to acceptor molecule (Rh B). The dye doped poly(methyl methacrylate)-based polymer optical fiber is pumped axially at one end of the fiber using a 532 nm pulsed laser beam from a Nd:YAG laser and the fluorescence emission is collected from the other end. At low pump energy levels, fluorescence emission is observed. When the energy is increased beyond a threshold value, laser emission occurs with a multimode structure. The optical feedback for the gain medium is provided by the cylindrical surface of the optical fiber, which acts as a cavity. This fact is confirmed by the mode spacing dependence on the diameter of the fiber.

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1. Introduction

Stimulated emission from organic dye molecules in solution by laser excitation was first reported by Sorokin and Lankard [1,2] and was subsequently studied by Schafer *et al.* [3], Spaeth and Bortfeld [4], and McFarland [5]. Solid-state dye lasers first demonstrated by Soffer and McFarland [6] in 1967 and then by Peterson and Snavely [7] in 1968, have great advantages over liquid dye lasers by being nonvolatile, nonflammable, nontoxic, compact, and mechanically stable. Organic dye doped polymers have been widely investigated as gain media in solid-state dye lasers [8–13]. Dye molecules that have large absorption and induced emission cross sections due to allowed pi–pi transitions are ideal active dopants for the generation and amplification of intense light pulse [14]. Polymer optical fibers (POFs) have attracted much attention during the past two decades because of their unique characteristics, such as flexibility, easiness in handling, and relative low cost in coupling [14,15]. With the development of POF, increased research activities have also been carried out in the field of active POF amplifiers and lasers [14–17]. Muto *et al.* [18] investigated a dye doped step index polymer fiber laser and Gvishi *et al.* [19] reported lasing action in Rhodamine 6G (Rh 6G) doped solgel glass fiber. Also, Kuriki *et al.* [20,21] reported lasing action of graded index POFs containing dyes such as Rhodamine B (Rh B), Rh 6G, and Perylene Orange. Also, photopumped narrow line laser emission is demonstrated using freestanding polymer films [22], and cylindrical microcavities are formed by conjugated polymer thin films, dye doped polymers, and dendrimer doped polymers [23–25].

We report multimode laser emission from an axially pumped POF made of poly(methyl methacrylate) (PMMA) doped with a mixture of Rh 6G and Rh B dyes. Here we use a mixture of dyes instead of a

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single dye to study the extended tunable laser emission from the mixed dye system. The excitation of dye lasers through energy transfer processes provides one of the means of extending the lasing wavelength region [26,27]. Energy transfer of Rh 6G:Rh B dye mixtures in a PMMA matrix was studied and revealed the nonradiative type energy transfer occurring from Rh 6G to Rh B [28]. The main mechanisms that have been proposed for such an energy transfer are (1) radiative transfer, i.e., absorption of donor emission by an acceptor; (2) diffusion controlled collisional transfer; and (3) resonance transfer via dipole–dipole interaction [29,30]. The origin of resonance transfer is the long-range dipole–dipole Coulomb interaction [31]. The probability of such an energy transfer is large if the emission spectrum of the donor strongly overlaps the absorption spectrum of the acceptor. Such a strong overlap of the emission spectrum of Rh 6G (donor) with the absorption spectrum of Rh B (acceptor) has been reported [32]. Radiationless energy transfer in the laser dye mixture consisting of Rh 6G (donor) and Rh B (acceptor) has also been studied using fluorescence lifetime measurements [33].

2. Experiment

The dye doped polymer fibers used for the present studies are based on PMMA. PMMA is chosen as the host as it has good optical quality and is compatible with most of the organic dyes used as dopants. The dyes used as dopants are Rh 6G and Rh B, which have high quantum yield, a low intersystem crossing rate, low excited state absorption at both pump and lasing wavelengths, and reasonably good photostability. The refractive index of methylmethacrylate (MMA) is ~1.41. When polymerized, the index of the polymer will increase up to 1.49 due to volume reduction during liquid to solid phase transition.

3. Fabrication of Dye Doped Polymer Optical Fiber

Commercially available MMA will contain inhibitors such as hydroquinone. Inhibitors are used to transport MMA without polymerization. Inhibitors are removed by repeatedly washing the monomer with 5% NaOH solution followed by flushing with distilled water. The remaining water is removed by adding suitable drying agents such as $CaCl_2$. The monomer is purified by distillation under reduced pressure. The preform for fiber fabrication is [34] made by polymerizing the purified MMA. The monomer is mixed with benzoyl peroxide (0.01 M) as initiator, *n*-butyl mercaptan (0.03M) as the chain transfer agent along with Rh 6G and Rh B in sealed tubes. The uniform mixing of the dye–monomer mixture is ensured by stirring well with a magnetic stirrer followed by filtration. These tubes are then placed in a constant temperature bath for three days at a temperature of 80 °C until it hardens and later they are kept in a furnace at 100 °C for 24 h. The prepared preform is now ready for drawing the fiber in a custom-made fiber drawing tower. By using a preform feeder, the preform is lowered into the furnace and fiber is drawn

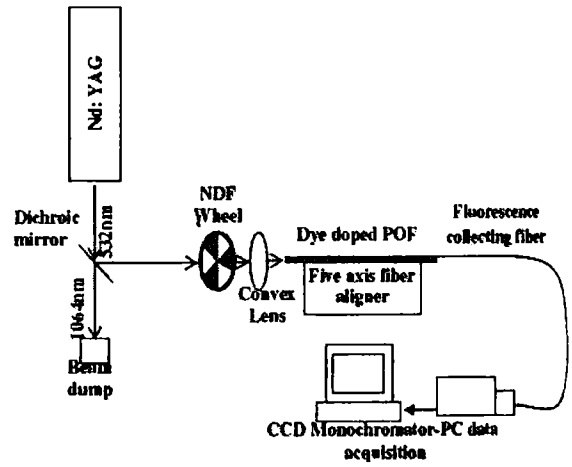


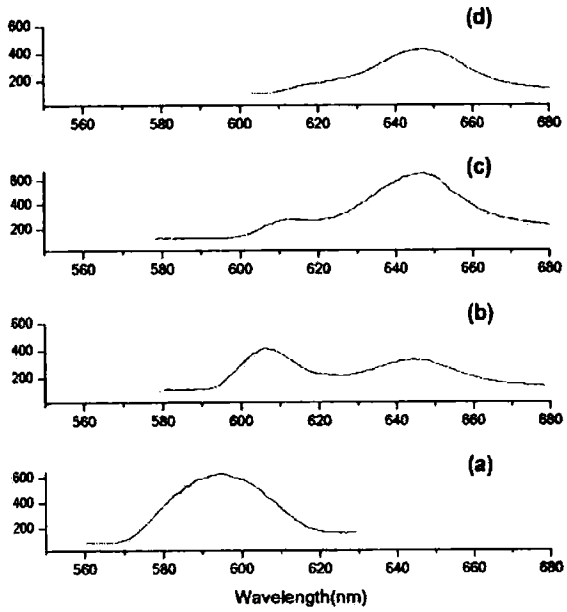
Fig. 1. Experimental setup to record the fluorescence emission from the fiber end. Pumping is done axially.

at a temperature of 180 °C. The fiber diameter can be varied by adjusting the feed rate of the preform and the draw rate of the fiber. For investigation of the tuning of multimode laser emission due to energy transfer process, four fiber samples are used with the following dye concentrations: (1) Rh 6G (0.25 mM), (2) Rh 6G (0.25 mM), and Rh B (0.11 mM), (3) Rh 6G (0.25 mM) and Rh B (0.25 mM), and (4) Rh B (0.25 mM). For this study we used fibers with diameters of 335, 405, and 510 μm. The maximum variation observed in the measurement of fiber diameter is ±2 μm. The effect of the length of the dye doped fiber on laser emission is studied using different lengths of the fiber, namely, 2, 4.5, 7, 9, and 12 cm.

A schematic of the experimental setup for the laser emission studies from dye doped polymer optical fiber is shown in Fig. 1. Dye mixture doped polymer optical fiber is mounted on a five-axis fiber aligner. The fiber is axially pumped using 10 ns pulses from a frequency doubled Nd:YAG laser (532 nm, 10 Hz). A set of calibrated neutral density filters is used to vary the pump energy. The pump beam is focused at the tip of the fiber using a convex lens of appropriate focal length. The emission is collected from the other end of the dye doped fiber using a collecting optical fiber coupled to a monochromator–CCD system (Acton Spectrapro).

4. Results and Discussion

Figure 2 shows a comparison of fluorescence emission from POF doped with Rh 6G, Rh 6G–Rh B dye mixture system and Rh B. In Fig. 2(b), the fluorescence spectrum from the POF doped with Rh 6G (0.25 mM) and Rh B (0.11 mM), shows a redshift in comparison with Rh 6G (0.25 mM) doped POF [Fig. 2(a)] confirming that energy transfer occurs from Rh 6G (donor) to Rh B (acceptor) [26–33]. Figure 2(b) also shows an enhancement of bandwidth up to 60 nm compared to the 30 nm bandwidth of Rh 6G doped POF [Fig. 2(a)]. When both Rh 6G and Rh B are present in equal concentration (0.25 mM), the spectrum shifts more



g. 2. Shift of fluorescence emission peak due to energy transfer ocess in dye mixture doped POF: (a) Rh 6G (0.25 mM), (b) dye ixture Rh 6G (0.25 mM) and Rh B (0.11 mM), (c) dye mixture Rh (0.25 mM) and Rh B (0.25 mM), and (d) Rh B (0.25 mM). Pump ergy is 0.12 mJ/pulse. Length of the POF is 7 cm and diameter 510 μm.

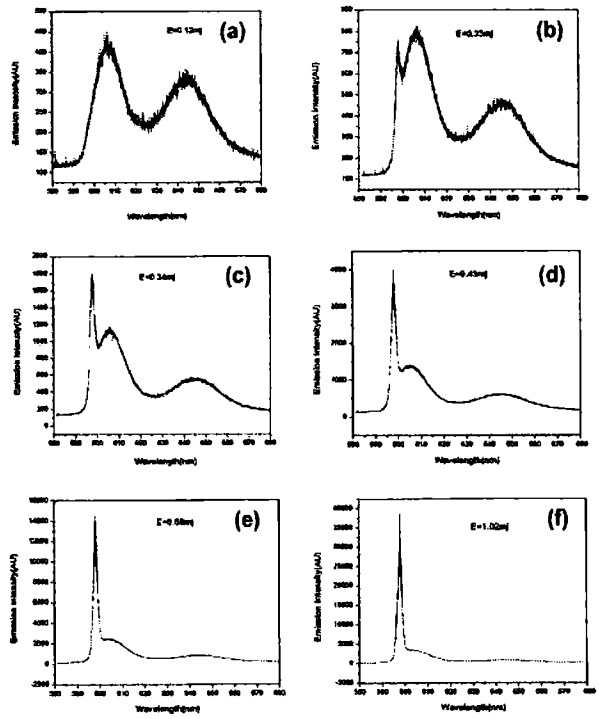


Fig. 3. Emission from 510 μm diameter and 7 cm long dye doped POF at a pump energy of (a) 0.12, (b) 0.23, (c) 0.34, (d) 0.43, (e) 0.68, and (f) 1.02 mJ.

ward the emission region of Rh B [Fig. 2(c)] because aximum energy transfer occurs when both dyes are ken in equal concentration [35]. Figure 2(d) repreents the fluorescence spectrum corresponding to Rh (0.25 mM).

Figure 3 shows the emission spectra of dye mixure doped POF [sample corresponding to Fig. 2(b)] ith variation in pump energy. Figure 3(a) shows a pical fluorescence emission spectrum from the e mixture doped polymer optical fiber when mped using the 532 nm radiation of energy 0.12 J/pulse. As the pump energy is increased further, horescence spectrum narrows due to amplified ontaneous emission (ASE) and at a threshold pump ergy, laser emission with a multimode structure erges [Fig. 3(c)]. The expanded modes are clearly own in Fig. 6. For a fiber with a diameter of 510 μm d a length of 7 cm the threshold energy is observed be 0.34 mJ/pulse.

A similar type of multimode laser emission is obrved in all other samples corresponding to Figs. a), 2(c), and 2(d). Figure 4 shows the tuning of the ultimode laser emission peak with variation in dye ncentration. The lasing wavelength peak of Rh 6G e (0.25 mM) doped POF is found to be at 572 nm. hen a mixture of Rh 6G (0.25 mM) and Rh B .11 mM) is used, the lasing wavelength peak shows lear redshift toward 598 nm [Fig. 3(d)]. From this, is clear that the lasing wavelength is tunable over larger range due to the enhancement in bandwidth hen a mixed dye system is used. When Rh B conntration in the dye mixture system is increased to 0.25 mM, the lasing wavelength peak shifts to 606 nm. A typical multimode laser emission spectrum, at a pump energy of 0.47 mJ/pulse from a POF doped with equal concentration (0.25 mM) of Rh 6G and Rh B dyes, is shown in Fig. 5. The lasing wavelength of Rh B (0.25 mM) is found to be at 610 nm. Since there is a clear overlap between the emission spectrum of Rh 6G and the absorption spectrum of Rh B, the energy transfer occurs from Rh 6G to Rh B

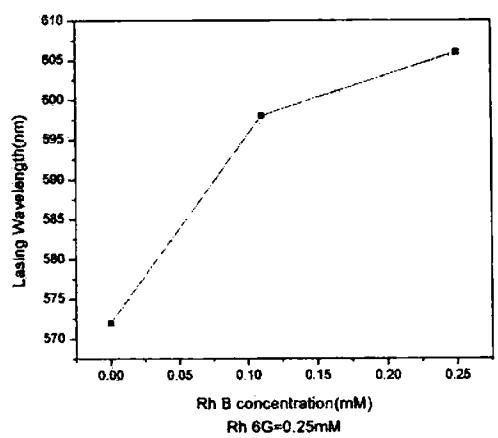


Fig. 4. Tuning of multimode laser emission peak with different dye concentrations. Rh 6G concentration is constant (0.25 mM) in all three samples. Pump energy is 1.37 mJ/pulse. L = 7 cm and D = 510 μm.

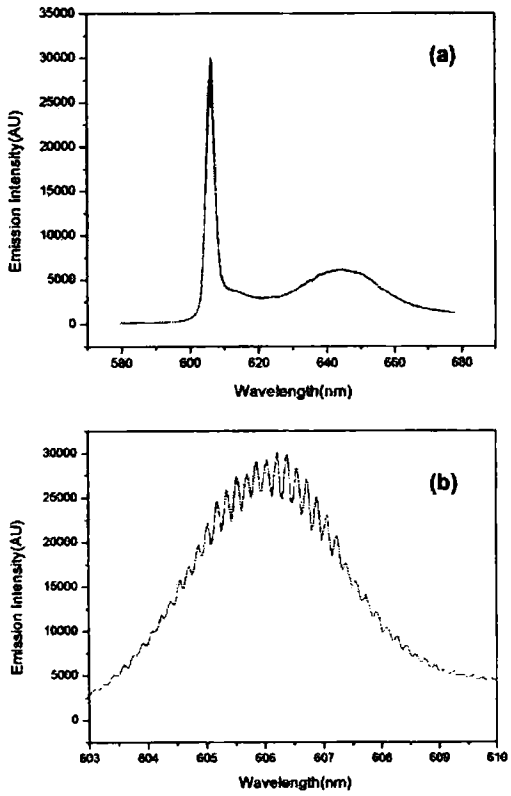


Fig. 5. (a) Typical multimode laser emission spectrum at a pump energy of 0.47 mJ/pulse from a POF doped with equal concentrations (0.25 mM) of Rh 6G and Rh B. (b) Expanded modes in (a).

and the lasing wavelength also shifts towards the emission region of Rh B.

Figure 6 shows the emission spectra recorded [sample corresponding to Fig. 2(b)] for different diameters of dye doped POF for a length of 7 cm at a pump energy of 1.37 mJ/pulse. The spectrum shown in Fig. 6(a) is a clear indication of the existence of resonant modes. The average mode spacing is 0.23 nm. The strongest mode at 595.3 nm has a FWHM of 0.1 nm. The dye doped fiber can be modeled as a number of serially connected microring cavities. The equations used to describe Fabry–Perot cavities work equally well here, with the substitution of the linear cavity length L by $\pi D/2$ [36–38]. Thus the expected mode spacing $\Delta\lambda$ is given by

$$\Delta\lambda = \frac{\lambda^2}{2nL} = \frac{\lambda^2}{\pi nD}, \tag{1}$$

where λ is the wavelength of the strongest emission line, n is the refractive index, and D is the diameter of the fiber. In the case of Fig. 6(a), substituting the values for λ, n, and D as 595.3 nm, 1.49, and 335 μm respectively, we get the mode spacing as 0.23 nm, which is the same as the observed mode spacing value. Table 1 shows the observed and calculated mode spacing values for fibers having different diam-

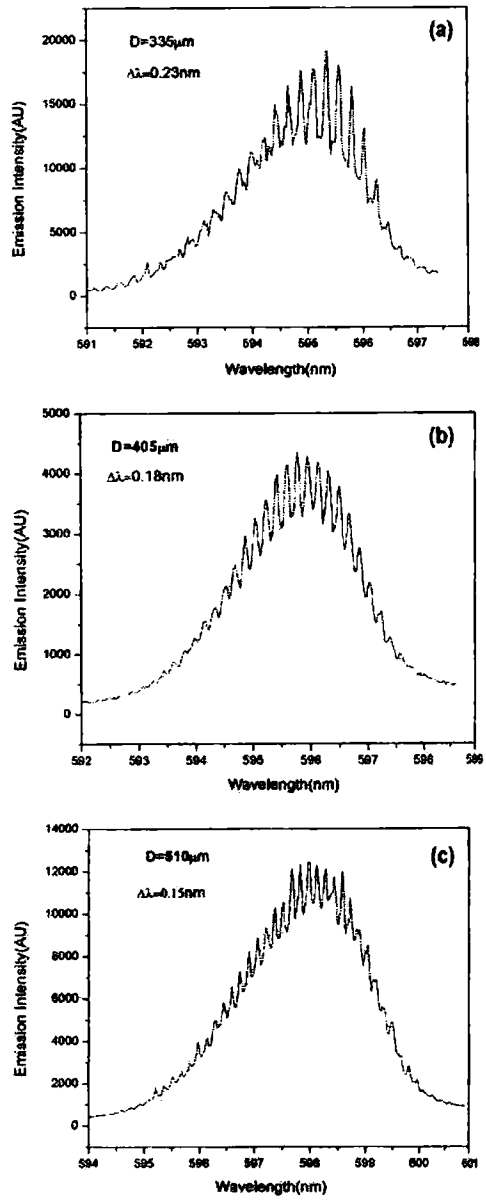


Fig. 6. Multimode laser emission from 7 cm long dye doped fiber at a pump energy 1.37 mJ/pulse. D = (a) 335, (b) 405, and (c) 510 μm.

eters. There is close agreement between the observed mode spacing and the calculated values. Thus as the diameter of the fiber increases, the mode spacing decreases accordingly. This observation is clear evidence of the fact that the observed fine structures in the emission spectra are resonant modes of the cavity formed by the cylindrical surfaces of the dye doped polymer optical fiber.

A similar phenomenon was observed by Geetha *et al.* [22] and Nedumpara *et al.* [39] in dye doped freestanding polymer films and Yokoyama *et al.* [40] in dye doped dendrimer solution. The observed resonant modes in our case can also be compared with the multimode lasing in plastic microring lasers on fibers

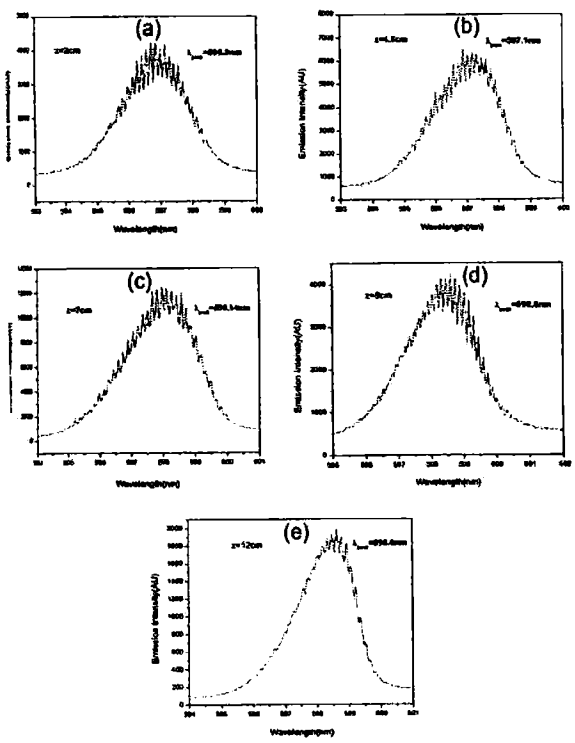
Table 1. Mode Spacing Dependence on Diameter of Fiber Based on the Microring Resonator Model[a]

Diameter (D) (μm)	Calculated Mode Spacing ($\Delta\lambda = \lambda^2/\pi nD$) (nm)	Observed Mode Spacing ($\Delta\lambda$) (nm)
335 ± 2	0.23 ± 0.002	0.23
405 ± 2	0.19 ± 0.002	0.18
510 ± 2	0.15 ± 0.002	0.15

[a]Fiber length is 7 cm and pump energy is 1.37 mJ/pulse.

and wires [36–38]. External feedback is necessary to obtain laser emission. In this case there are no external mirrors to give feedback to the gain medium. The optical feedback for the gain medium is provided by the cylindrical surface of the optical fiber, which acts as the cavity. Though the reflections from these cylindrical surfaces are weak compared to the conventional laser cavity mirrors, the stimulated emission along with its propagation through the gain medium resulted in laser emission with a multimode structure.

Figure 7 shows the emission spectra from dye doped POF [sample corresponding to Fig. 2(b)] having different lengths (z in figure) at a pump energy of 1.37 mJ/pulse. When the fiber length is increased, the multimode structure is observed superposed over the ASE up to a length of 9 cm for a fiber having 510 μm diameter. When the fiber length is 12 cm the fine structure degrades and laser emission becomes less prominent. An increase in the length of the fiber will result in the enhancement of the loss factor, which leads to the reduction of the visibility of the mode structure as observed in the present case. Also, as the length of the fiber is increased, redshift in the multimode laser emission can be observed due to the absorption and reemission process taking place within the dye doped fiber [41]; also see Fig. 8.



g. 7. Multimode laser emission from 510 μm diameter dye ped POF at a pump energy of 1.37 mJ/pulse. z = (a) 2, (b) 4.5, 7, (d) 9, and (e) 12 cm.

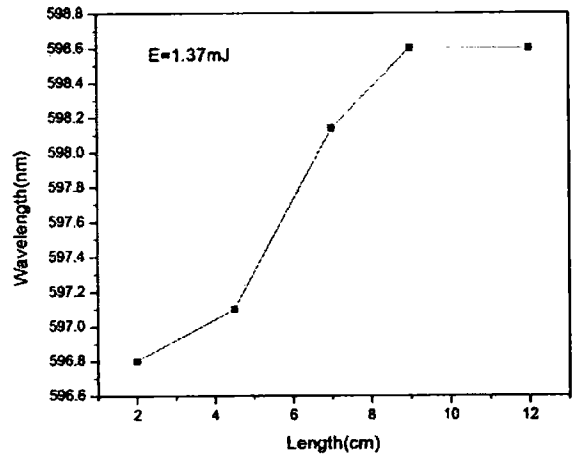


Fig. 8. Variation of laser emission peak with length of the fiber having a diameter of 510 μm at a pump energy of 1.37 mJ/pulse.

5. Conclusion

Multimode laser emission from a dye mixture doped polymer optical fiber is observed when excited by a 532 nm pulsed laser beam from a Nd:YAG laser. Tuning of laser emission is achieved by using a mixture of dyes utilizing the energy transfer that occurs from the donor (Rhodamine 6G) molecule to the acceptor molecule (Rhodamine B). As the energy of the pump beam is increased, fluorescence spectrum narrows due to amplified spontaneous emission and at a threshold energy, laser emission with a multimode structure emerges. When the diameter of the fiber is increased the mode spacing is found to decrease confirming our proposition that the observed modes are the resonant modes of a number of serially connected microring cavities formed by the dye doped POF. As the length of the fiber is increased, redshift in the multimode laser emission can be observed due to the absorption and reemission processes taking place within the dye doped fiber.

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