# Propagation characteristics and wavelength tuning of amplified spontaneous emission from dye-doped polymer

K. Geetha, M. Rajesh, V. P. N. Nampoori, C. P. G. Vallabhan, and P. Radhakrishnan

The propagation characteristics of amplified spontaneous emission (ASE) through a rhodamine 6 G-doped polymethyl methacrylate freestanding film waveguide were studied. This was done by shifting the excitation stripe horizontally along a transversely pumped waveguide. By this method, we could tune the ASE wavelength. The maximum tunability thus obtained was ~18 nm with a pump stripe length of 6 mm. © 2006 Optical Society of America

*OCIS codes:* 230.7370, 300.2140, 140.3580, 250.5460.

## 1. Introduction

Organic dye lasers have become a subject of intensive research ever since Sorokin and Lankard reported the stimulated emission from an organic dye solution.[1] The use of solid matrix for dye lasers eliminates many of the common problems associated with liquid systems such as concentration variation due to the evaporation of solvents. The first attempts to develop solid-state dye lasers were reported in the late 1960s.[2,3] Most of the recent work has been done using either polymers[4,5] or silica gels[6,7] as the host media. Polymeric materials in particular offer advantages such as ease of processing, which permits fabrication of devices of virtually any shape and potentially very low cost. Organic polymers such as poly(methylmethacrylate)[8,9] (PMMA) or poly(acrylic acid)[10] (PA) have been widely used as host material for laser dyes.

Dye-doped solid-state waveguide lasers provide low-cost, high-efficiency tunable coherent light sources. Waveguide structures provides long gain length and optical confinement, which offer reduction of the lasing threshold. Conventionally, wavelength tuning is achieved by the use of distributed Bragg reflectors as coupling mirrors in resonant cavities.[11] In external cavity solid-state lasers, wide-range wavelength tuning has been achieved by the use of a grating as the dispersion element. A tunability of approximately 53 nm was obtained with a multiple prism-grating solid-state dye laser where the gain medium was a rhodamine 6G (Rh6G)-doped modified PMMA with trapezoidal geometry.[12] Multimode lasing and wideband tuning in sol–gel distributed feedback waveguide lasers have also been reported.[13,14] In these lasers, tuning was achieved either by varying the period of gain modulation[13,14] or by varying the temperature of the gain medium, thereby varying its refractive index.[15] By using the first method, a tunability of more than 30 nm was achieved. By using the latter method, with a Rh6G-doped sol-gel silica waveguide, 17 nm tunability was obtained by varying the temperature from 21 °C to 58 °C while with Rh6G-doped PMMA waveguide, 6 nm tunability was obtained by varying temperature from 22 °C to 98 °C.

For most dyes, the absorption and emission bands overlap and the short wavelength fluorescence is reabsorbed. For such dyes the observed fluorescence peak is shifted to longer wavelengths, at higher concentrations. Thus wavelength tuning can also be achieved by changing the dye concentration.[16]

In high-gain structures, under strong pumping conditions, consideration of the phenomenon of amplified spontaneous emission (ASE) becomes very important for understanding the behavior of such systems. ASE is a phenomenon where the spontaneously emitted light gets amplified as it propagates along the gain medium, and even without feedback the emitted radiation exhibits laserlike properties

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Received 6 June 2005; revised 12 September 2005; accepted 14 September 2005.

0003-6935/06/040764-06$15.00/0

ch as low divergence and spectral narrowing. A hnique for tuning the ASE wavelength by control-g the waveguide thickness has been reported for ugated polymer films[17] as well as for dye-doped id-state waveguides.[18] A wide-range tunability of und 30 nm was observed in these two cases.

Organic dyes exhibit a concentration-dependent shift of the peak emission wavelength.[16] Similar shift was observed for the spectra taken from nsversely pumped dye-doped fibers and wave-ides when the excitation spot position was shifted ng the length of the fiber or waveguide by trans-ing either the source or the waveguide.[19–21] Using imilar technique, we investigated the wavelength ning in a transversely pumped Rh6G-doped PMMA n waveguide by observing the wavelength changes ASE. The cross section of the pump beam was in e form of a stripe. The position of excitation stripe s shifted in such a way that the ASE from the end the pump stripe is guided along different lengths of e waveguide. A tunability of ~18 nm was obtained en the distance between the waveguide edge and e pump stripe edge was varied from 0 to 15 mm. e main advantage of the present technique is that bulk optics are used for tuning. By simultaneous ectral measurement from both ends of the pumped veguide, simultaneous tuning of dual wavelength ission can also be achieved, details of which will be blished elsewhere.

## 2 Experiment

periments were conducted on freestanding thin ms of PMMA doped with Rh6G. PMMA, which is e most frequently used polymer host for dye lasers, ows the best optical transparency in the visible ectral range. RhG6, the best known of all laser es, has been frequently investigated in solid-state e lasers in a variety of solid hosts, on account of its gh fluorescence quantum yield, low intersystem ssing rate, and low excited-state absorption at th pump and lasing wavelengths. The samples re prepared by dissolving PMMA and Rh6G in thylethylketone, with 0.5 mM dye concentration. ms of 50 μm thickness were tape cast on glass ets from this solution. When the solvent is fully porated, freestanding films could be peeled off the ss sheet. The films were then cut into the size m × 2 cm.

The samples were transversely pumped using ns pulses from a frequency doubled Nd:YAG laser 2 nm, 10 Hz). A set of calibrated neutral density ers were used for varying the pump energy. The mp energy was varied from 0.03 to 9.1 mJ/pulse. e beam was focused into a narrow stripe of approx-ately 50 μm width. The stripe length was varied ng an adjustable slit.

When ASE occurs in a long narrow stripe, most of the ht is emitted at the ends of the stripe. The emission m the sample was collected by a fiber and directed to 0.5 m spectrometer with a cooled CCD array. The stance between the collecting fiber and the veguide edge was 1 cm. A slit was kept in front of

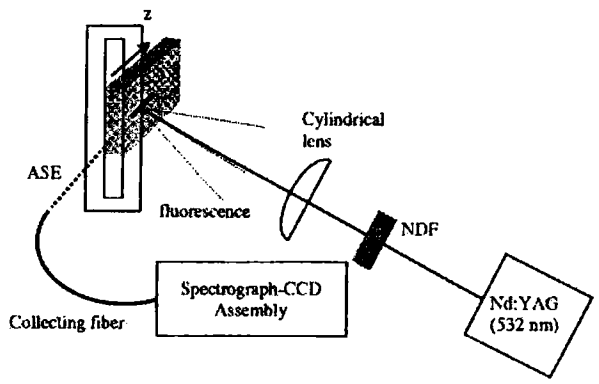


Fig. 1. Excitation and light collecting schemes.

the fiber so that the emission from the front and back surfaces was blocked. The sample was kept on a translator so as to enable horizontal shifting of the excitation stripe along the length of the waveguide. Initially the excitation stripe was formed at the edge ($z = 0$) of the waveguide and the emission spectra for various pump energies were recorded. Then the stripe position was shifted, thereby varying $z$—the distance between one end of the pump stripe and the observation edge of the film (Fig. 1). Observations were made for stripe lengths of 2, 4, and 6 mm.

## 3. Results and Discussion

### A. Observation of Amplified Spontaneous Emission

First we recorded the emission spectra for $z = 0$, at various pump intensities with a constant pump stripe of length 2 mm (Fig. 2).

At low pump energies, the emission spectrum is broad. But above the pump energy of 0.31 mJ/pulse, the peak of the spectrum at approximately 565 nm grew strongly. At $E = 9.1$ mJ/pulse, a narrow spec-

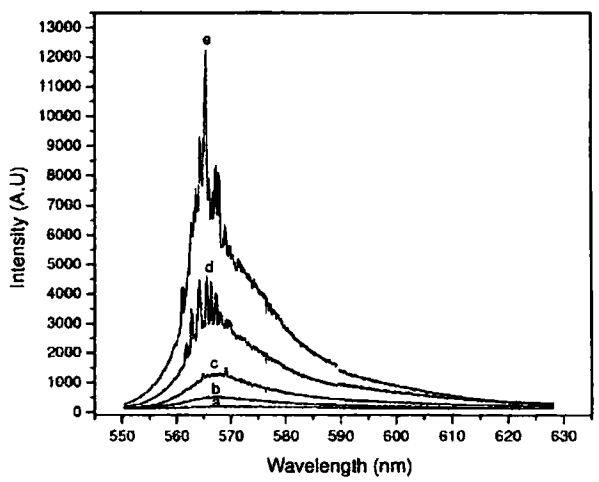


Fig. 2. Emission from the waveguide edge for stripe length 2 mm, for various pump energies: (a) 0.031 mJ/pulse; (b) 0.10 mJ/pulse; (c) 0.31 mJ/pulse; (d) 1 mJ/pulse; (e) 9.1 mJ/pulse.

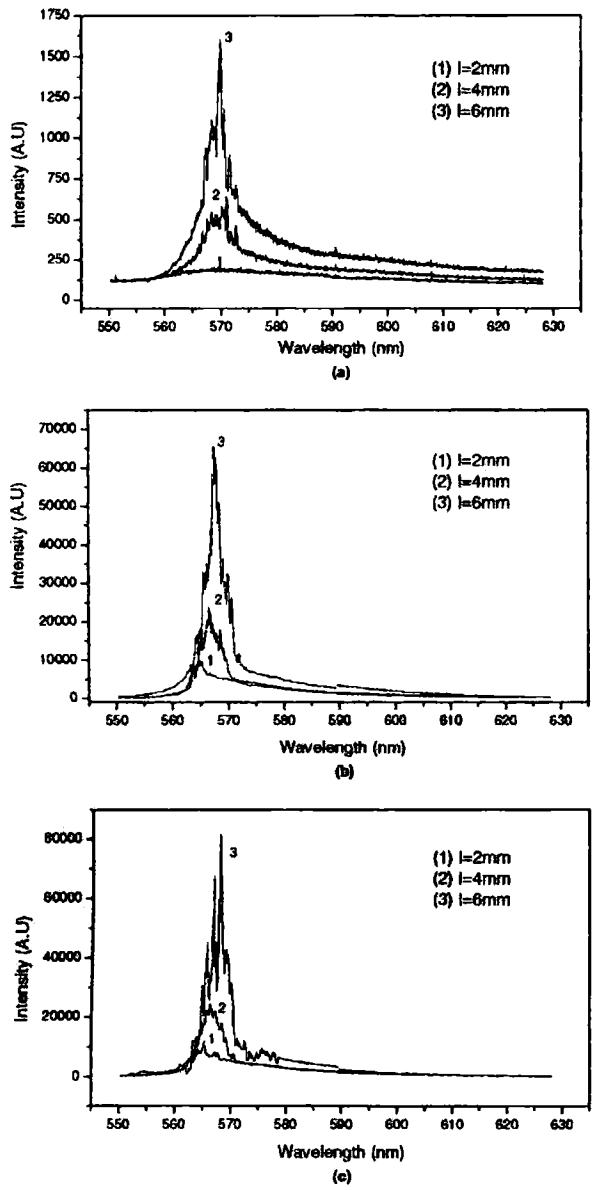


Fig. 3. Spectral narrowing with increase in stripe length for various pump energies: (a) $E$ = 0.031 mJ/pulse; (b) $E$ = 3.1 mJ/pulse; (c) $E$ = 9.1 mJ/pulse.

trum centered at 565.4 nm with a FWHM of only ~5 nm was obtained.

Spectral narrowing of the emitted radiation can be due to several reasons. To confirm the phenomenon responsible for spectral narrowing, we varied the length of the excitation stripe. If ASE occurs, spectra should be broad at short stripe lengths and should become narrow as the excitation length increases.[22,23]

Figure 3 shows the spectra recorded for various pump powers for stripe lengths 2, 4, and 6 mm. As the pump stripe length increases, the threshold for ASE decreases. Also, for a given pump energy, the FWHM decreases with increase in stripe length (see Table 1), with a slight shift in the peak wavelength.

Table 1. FWHM of Spectral Emission for Various Pump Energies and Beam Stripe Lengths

| Pump Energy (mJ/pulse) | FWHM (nm) | | |
|---|---|---|---|
| | $l$ = 2 mm | $l$ = 4 mm | $l$ = 6 mm |
| 0.031 | ~50 | 6.6 | 3.4 |
| 3.1 | 8.1 | 5.6 | 2.5 |
| 9.1 | 5.1 | 4.6 | 1.0 |

### B. Gain Measurement

According to the method introduced by Shaklee *et al.*,[24] the relation between the output light variation with excitation length and the gain is given by

$$I_0(l) = \frac{I_s A}{g}[\exp(gl) - 1], \quad (1)$$

where $I_s$ is the spontaneous emission rate per unit volume; $A$ is a constant related to the spontaneous emission cross section; $g$ is the net gain given by $g' - \alpha$, where $g'$ is the gain due to stimulated emission; $\alpha$ is the optical loss; and $l$ is the length of the pump stripe.

Shank *et al.*[25] measured the single pass gain of dye laser by comparing the intensities of ASE in single and double cell lengths. By writing an equation similar to that of Eq. (1) for a pump stripe of length $l/2$, we get

$$I_0(l/2) = \frac{I_s A}{g}[\exp(gl/2) - 1]. \quad (2)$$

From Eqs. (1) and (2), the net gain can be obtained as

$$g = \frac{2}{l}\ln\left[\frac{I_0(l)}{I_0(l/2)} - 1\right]. \quad (3)$$

Equation (3) applies only for pump powers up to the onset of saturation The gain at 570 nm for a pump energy of 0.31 mJ/pulse calculated as per Eq. (3) was obtained to be 5.36 $cm^{-1}$.

### C. Amplified Spontaneous Emission Propagation Through the Waveguide and Wavelength Tuning

It is well known that by varying the dye concentration, wavelength tuning can be attained in dye lasers.[16] For RhG6, there is a spectral region of overlap between the absorption and emission bands. As a result, the short wavelength emission from the dye molecules gets absorbed by itself and is reemitted at a longer wavelength. Due to this self-absorption and reemission process, the peak emission wavelength shows a redshift with an increase in concentration.[14] This property is utilized in the concentration dependent tuning of the emitted radiation. Increasing the path length through the dye-doped sample is somewhat similar to an increment in dye concentration. A

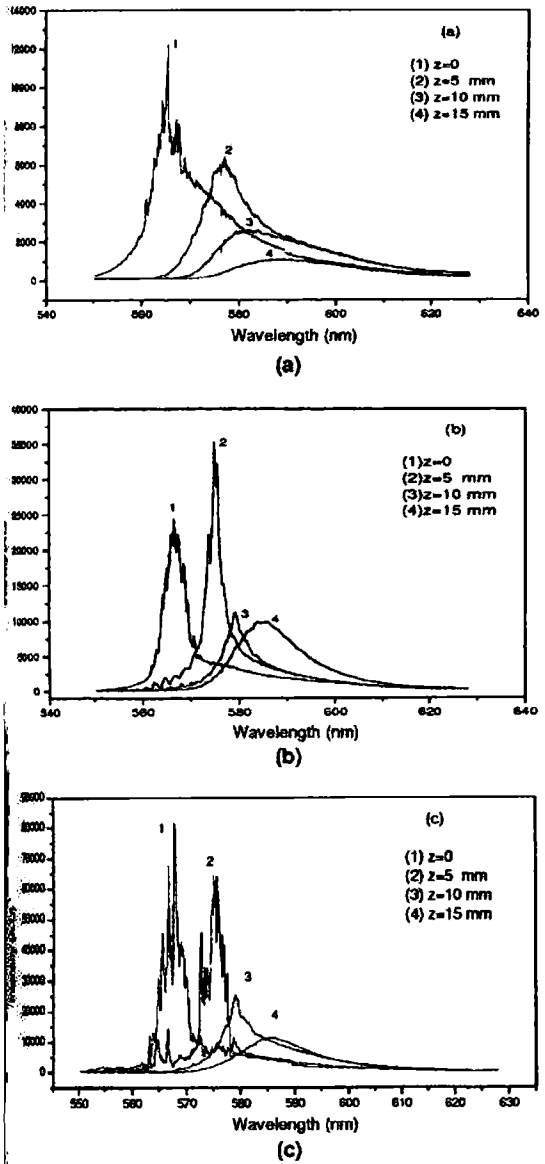


Fig. 4. Evolution of ASE as it is guided through various lengths through the waveguide for stripe lengths: (a) $l$ = 2 mm, (b) $l$ = 4 mm, and (c) $l$ = 6 mm for pump energy 9.1 mJ/pulse.

similar redshift is observed when the path length through the waveguide or fiber is increased.[19–21]

When irradiated with a narrow stripe, the ASE will be maximum at the stripe ends, along the transverse direction. We changed the position of the excitation stripe by translating the waveguide horizontally across the exciting beam. Now the ASE emitted at the end of the stripe is made to traverse through the sample. By shifting the position of the exciting stripe, we are varying the propagation length of the emission through the waveguide. Emitted light is collected exactly in a similar manner as described above. Figure 4 shows the spectra obtained for various $z$ values at a pump energy of 9.1 mJ/pulse for various stripe lengths.

We observe a redshift for the emission peak with increase in length of propagation. Even after propagation through the sample, we get a narrow emission spectrum, especially for longer stripe lengths. Table 2 gives the values for FWHM and peak emission wavelengths for various pump stripe positions, for the three different stripe lengths corresponding to a pump energy of 9.1 mJ/pulse.

We see that for a stripe length of 2 mm, we get narrow emission peaked at 565.4 for propagation distances $z$ = 0 and at 577.2 nm for $z$ = 5 mm. The spectral width increases from ~5 to 11 nm when $z$ changes from 0 to 5 mm. Thus the ASE peak wavelength can be continuously tuned over 12 nm by changing the $z$ value from 0 to 5 mm (see Table 1). For further propagation lengths, the emission becomes very broad and ASE character of the emission is lost. With longer pump stripe lengths, the tunability range gets enhanced to around 18 nm. The tuning range can be effectively further enhanced by collecting emission from both ends of the waveguide simultaneously. Details of this will appear elsewhere. ASE can be tuned from 566.6 to 585.4 and 568.1 to 586.1 for stripe lengths 4 and 6 mm, respectively. It should also be noted that during the tuning the spectral width generally increases up to 16 nm. An exception is observed in the case of stripe length 4 mm at $z$ = 5 mm, for which the intensity corresponding to $z$ = 5 mm is greater than that corresponding to $z$ = 0. A similar tendency is observed in the case of stripe length $z$ = 6 mm for which the peak intensities corresponding to $z$ = 0 and $z$ = 5 mm are comparable. This is a clear indication of the existence of gain in the medium. Detailed studies in this direction are in progress.

One of the important points to be noted is that the

Table 2. FWHM and Peak Wavelength for Different $z$ Values at Pump Energy 9.1 mJ/pulse

| | Stripe Length = 2 mm | | Stripe Length = 4 mm | | Stripe Length = 6 mm | |
|---|---|---|---|---|---|---|
| Z (mm) | FWHM (nm) | Peak λ (nm) | FWHM (nm) | Peak λ (nm) | FWHM (nm) | Peak λ (nm) |
| 0 | 5.1 | 565.4 | 4.6 | 566.6 | 1.0 | 568.1 |
| 5 | 11.4 | 577.2 | 1.8 | 574.7 | 2.4 | 575.7 |
| 10 | 25.3 | 582.3 | 5.9 | 579.2 | 5.9 | 579.3 |
| 15 | 26.3 | 589.6 | 15.8 | 585.4 | 16.4 | 586.2 |

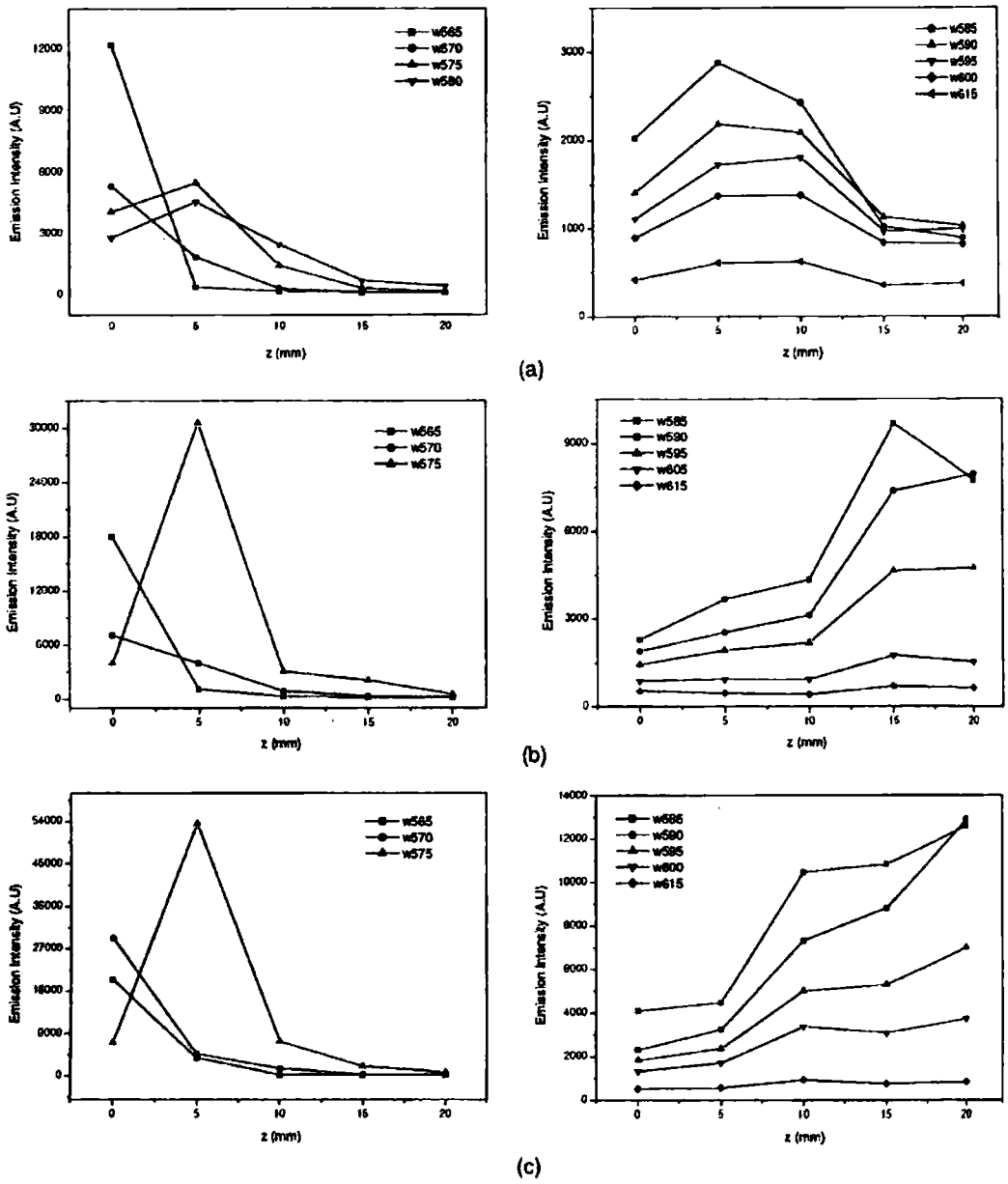


Fig. 5. Emitted power at various wavelengths versus propagation length through the waveguide for stripe lengths: (a) $l = 2$ mm, (b) $l = 4$ mm, and (c) $l = 6$ mm for pump energy 9.1 mJ/pulse.

intensity at longer wavelengths of the ASE gets amplified on traversing through the waveguide. Output intensity at shorter wavelengths show the usual exponential decrease. Figure 5 shows the variation of output power at different wavelengths with propagation length through the waveguide.

As can be clearly seen from Fig. 5, as the propagation distance $z$ increases, the emission intensity shows enhancement in the long wavelength region. This gain in the long wavelength region can be attributed to the gain achieved as the radiation is propagated through the amplifying medium as well as due to the reemission from the interacting dye molecules at longer wavelengths. Such emission enhancement becomes prominent as the stripe length of the pump beam is increased. Increase in the stripe length enhances the interaction length between the pump radiation and the dye molecules. This will result in the gain in the spectral emission in addition to the gain due to reemission in the long wavelength region. Propagation characteristics of emitted radiation through the waveguide cannot be described by the usual Lambert–Beer law with single attenuation or gain coefficient. Due to complex interactions between the radiation and dye molecules in the gain media, one has to describe a space dependent attenuation

gain coefficient. This will be the subject matter of future publication.

## Conclusions

We have studied the propagation characteristics of amplified spontaneous emission through a transversely pumped dye-doped polymer freestanding film waveguide. With the increase in propagation length through the waveguide, the emission peak shows a redshift due to self-absorption and reemission by the dye molecules. As the radiation is propagated through the amplifying medium, we get spectrally narrow output at longer wavelength, due to the gain achieved at these wavelengths. Thus we could tune the ASE wavelength. The maximum tunability obtained by this method was ~18 nm.

The authors acknowledge the financial support from NUFFIC, Netherlands under the MHO assistance to the International School of Photonics. The authors thank A. Denny and Raghu Natarajan, CAMET, Thrissur, for providing the dye-doped samples. K. Geetha is grateful to the Council of Scientific and Industrial Research, New Delhi, for the research fellowship. M. Rajesh, V. P. N. Nampoori, and C. P. G. Vallabhan acknowledge University Grants Commission, New Delhi for financial assistance.

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