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Laser emission from transversely pumped dye-doped free-standing polymer film

K Geetha[1], M Rajesh[1], V P N Nampoori[1], C P G Vallabhan[1,2] and P Radhakrishnan[1]

[1] International School of Photonics, Cochin University of Science and Technology, Cochin 682022, India

[2] Centre of Excellence in Lasers and Optoelectronic Sciences, Cochin University of Science and Technology, Cochin 682022, India

E-mail: geetha@cusat.ac.in

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Abstract

We present a compact solid-state laser based on leaky mode propagation from a dye-doped polymer free-standing film waveguide. The edge emitted spectrum clearly indicated the existence of periodic resonant modes. The reflections from the lateral faces of the free-standing film provided the optical feedback thus giving rise to a Fabry–Perot like optical cavity. This together with the guidance through the gain medium gave rise to intense narrow emission lines. For a pump energy of 1.82 mJ/pulse, an intense line with FWHM ∼0.4 nm was observed at 576.5 nm.

Keywords: polymer film, laser emission, mode, waveguide

1. Introduction

The first attempts to develop solid-state dye lasers were reported in the late 1960s [1, 2]. Ever since, there have been intensive efforts to achieve the incorporation of organic dyes in solid matrices that might replace conventional liquid dye lasers. Most of the recent work has been done using either polymers [3, 4] or silica gels [5, 6] as the host media. Polymeric materials in particular offer advantages such as ease of processing, which permits the fabrication of devices of virtually any shape and potentially at very low cost. The combination of the tunability and high efficiency of laser dyes with the high power density that can be easily achieved in waveguide structures makes devices based on dye-doped polymer waveguides and fibres very promising [7–11]. Wide range tunable laser emission has been reported from polymer thin films in optically pumped distributed feedback schemes [7, 12]. Laser emission from conjugated polymers and dendrimer doped polymers has also been reported during the past few years [13–17]. There have been numerous investigations on laser emission from polymer planar microcavities [18, 19] and polymer microring lasers as well [20].

In this paper we report the observation of multimode laser emission from a transversely pumped free-standing polymer film (of poly methyl methacrylate) doped with the laser dye rhodamine 6G. Since the film is free standing—surrounded by air at both sides—the reflections from the lateral faces of the sample provided the optical feedback for laser action. This was evident from the lasing mode-spacing dependence on the film thickness. The leaky mode emission from the film waveguide showed a planar microcavity-like behaviour due to Fabry–Perot effects. Selective mode excitation was also observed with an increase in pump power.

2. Experiment

Experiments were conducted on free-standing thin films of poly methyl methacrylate (PMMA) doped with Rhodamine 6G. PMMA, which is the most frequently used polymer host for dye lasers, shows the best optical transparency in the visible spectral range. Rhodamine 6G (Rh6G), the best known of all laser dyes, has been frequently investigated in solid-state dye lasers in a variety of solid hosts, on account of its high fluorescence quantum yield, low intersystem crossing rate and low excited state absorption at both pump and lasing

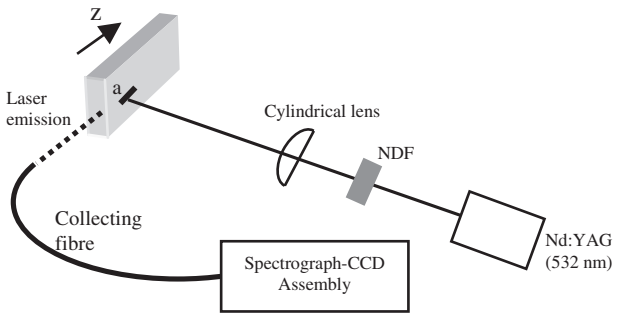

Figure 1. Excitation and light collecting scheme.

wavelengths. The samples were prepared by dissolving PMMA and Rh6G in methyl ethyl ketone, with 1.5 mM dye concentration. Films of 50 μm thickness were tape cast on glass sheets from this solution. When the solvent was fully evaporated, high-quality free-standing films could be peeled off the glass sheet. The lateral faces of the films obtained were of good optical finish such that no further polishing was required. The films were then cut into the size 4 cm $\times$ 2 cm.

Figure 1 shows a schematic diagram of the experimental set up. The samples were transversely pumped using 10 ns pulses from a frequency doubled Nd:YAG laser (532 nm, 10 Hz). A set of calibrated neutral density filters was used for varying the pump energy. The pump energy was varied from 0.02 mJ/pulse to 1.82 mJ/pulse. The beam was focused into a narrow stripe of length 6 mm and approximately 50 μm width. The film waveguide was kept on a translator so as to enable horizontal shifting of the excitation stripe along the length of the waveguide. The direction of translation of the excitation stripe is indicated by the arrow in figure 1. The distance between one end of the pump stripe (shown as point 'a' in figure 1) and the edge of the waveguide from where the emission is collected, is measured as z. The emission from the sample was collected by a fibre and directed to a

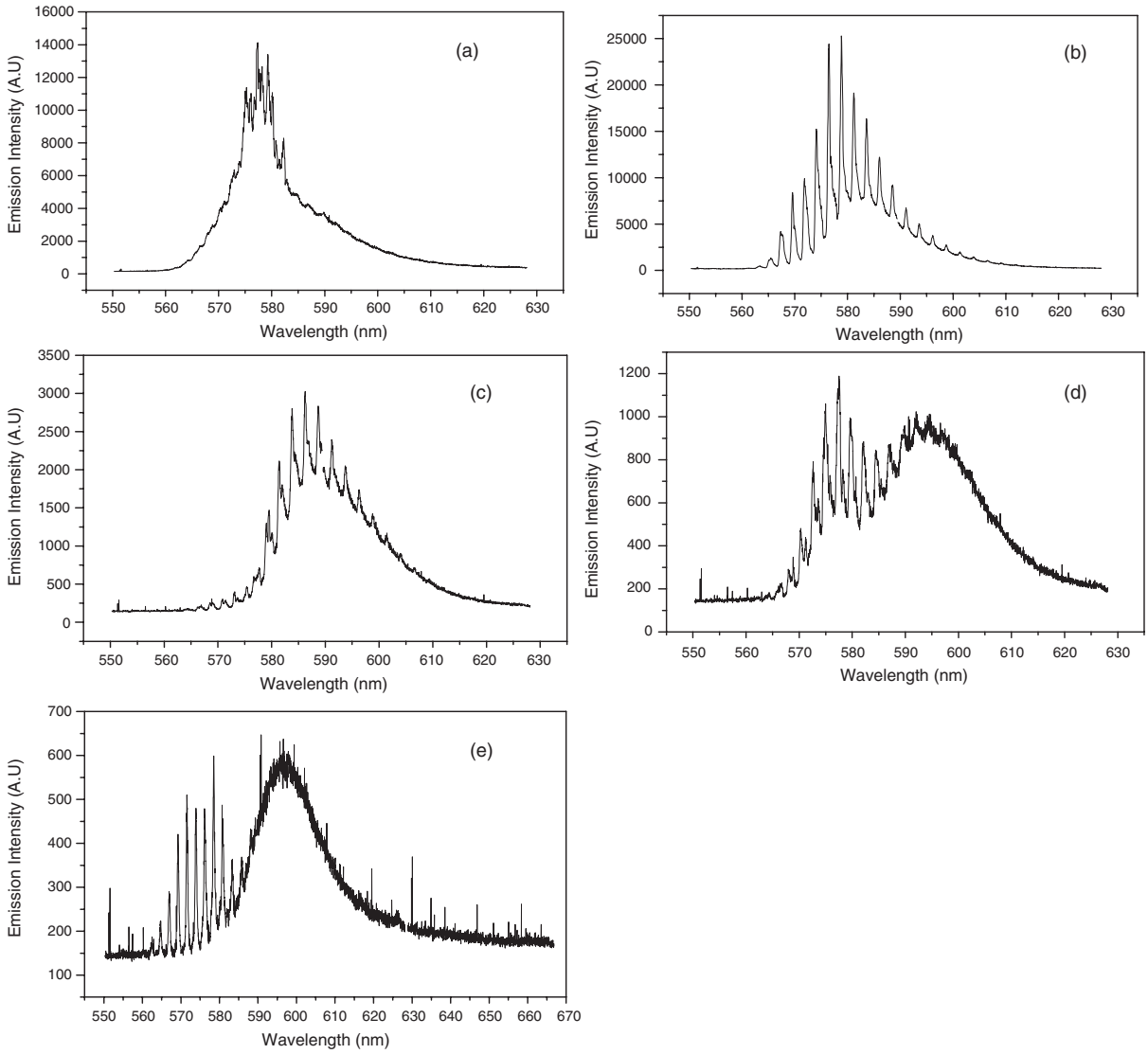

Figure 2. Emission from 50 μm thick Rh6G-doped PMMA film at pump energy 0.2 mJ/pulse for (a) $z = 0$, (b) $z = 5$ mm, (c) $z = 10$ mm, (d) $z = 15$ mm and (e) $z = 20$ mm.

0.5 m spectrograph (SpectraPro-500i) coupled with a cooled CCD array. The distance between the collecting fibre and the waveguide edge was 1 cm. Emission spectra for various values of z were recorded. Initially the excitation stripe was formed at the edge of the waveguide ($z = 0$). The excitation stripe position was shifted by translating the waveguide horizontally across the pump beam, thereby varying z.

3. Results and discussion

Figure 2 shows the emission spectra recorded for different values of z at a pump energy 0.2 mJ/pulse. The spectrum shown in figure 2(b) clearly indicates the existence of resonant modes. The average mode spacing is ∼2.3 nm. The strongest emission lines occurred at 576.5 and 578.8 nm with an FWHM ∼0.6 nm. Such a spectral narrowing can be attributed to laser emission.

Laser emission requires an external feedback. In our case, there were no external mirrors to provide the feedback. Since the pumped polymer film was a free-standing one, the lateral faces of the film acted as mirrors, thus giving rise to a Fabry–Perot-type optical cavity whose length corresponds to the film thickness. Owing to the sample geometry and pumping scheme, the longitudinal modes of this cavity are analogous to the transverse modes of the waveguide. In the case of a thin stripe excitation, the stimulated emission occurs in the direction along the stripe [14]. Both the stimulated emission along with the propagation in the guiding gain medium and the feedback at the lateral faces induce high gain for laser action, though reflections at the lateral faces are much smaller than that of conventional cavity mirrors. The fine structure pattern in the spectra shown in figure 2 can be attributed to the axial modes of the Fabry–Perot cavity formed by two surfaces of the planar waveguide. In other words the planar waveguide can be thought of as being formed by a number of serially connected Fabry–Perot etalons. This can be verified from the mode spacing.

The mode spacing at λ can be calculated using the equation which describes Fabry–Perot cavities, namely,

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

where λ is the wavelength of the strongest emission line, n is the refractive index and L is the length of the resonator cavity.

In the present case, the length of the Fabry–Perot cavity corresponds to the thickness of the polymer film. Putting the values for λ, n and L as 576 nm, 1.49 and 50 μm respectively, we get the mode spacing as 2.226 nm, which is same as the observed value 2.3 nm.

The emission spectrum for a 130 μm thick film with the same dye concentration as in the case of 50 μm thick film is given in figure 3, for $z = 10$ mm.

The obtained mode spacing (0.9 nm) agrees with the calculated value (0.86 nm). These observations confirm that the observed equally spaced fine structures in the emission spectra are Fabry–Perot-type modes of the optical cavity formed by the film thickness, which are also the transverse modes of the film waveguide.

A similar phenomenon was observed by Yokoyama *et al* [21] in laser dye-doped dendrimer solution. The observed

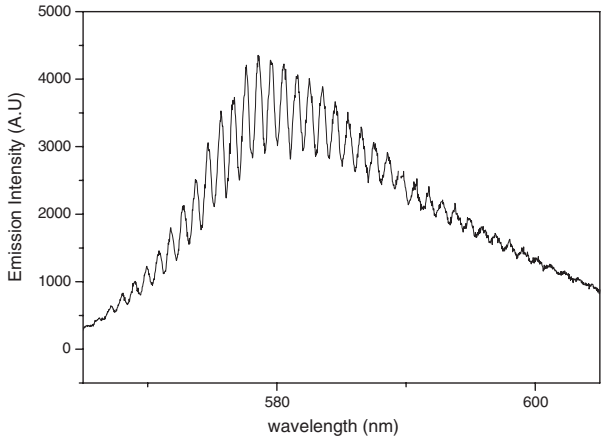

Figure 3. Emission from a 130 μm thick sample at pump energy 0.2 mJ/pulse for $z = 10$ mm.

results can also be compared with typical emission from Fabry–Perot microcavity lasers [22]. The Fabry–Perot-like behaviour of an asymmetric thin-film waveguide was explained to be quite similar to that of thin-film microcavities [23]. The present case can be considered as the Fabry–Perot behaviour of a thick symmetric waveguide. The reflections at the film–air interface serve the purpose of metallic mirrors or Bragg reflectors used in microcavity thin film lasers [18].

If we collect light by keeping a narrow slit in between the fibre and the waveguide edge, these structures vanish. On placing a narrow slit, we restrict the collection of emission along the waveguide axis. In the absence of a slit, light emitted at different angles from the waveguide surface can also be collected (note that we are using a plastic fibre with 0.98 mm core diameter and NA 0.5 for collecting the emitted light). So the observed emission can be considered as the leaky modes of the film waveguide.

Leaky mode emission and microcavity-like behaviour due to the Fabry–Perot effect from asymmetric luminescent film waveguides have been studied in [23]. The corresponding emission was tilted towards the substrate side of the sample. The free-standing polymer film in the present experiment is a symmetric waveguide, and the higher-order transverse modes of the waveguide leak into air at different angles. The dependence of the peak intensity and spectral width of the leaky mode self-trapped exciton emission on the observation direction has been studied in anatase thin films [24]. We also noticed that the intensity of the peaks depends on the collecting angle. However, a study of the exact dependence of emission parameters on the observation angle can be done only by modifying the present experimental set up, which is currently under progress.

The mode patterns are observed only when there is a separation between one end of the pump stripe and the waveguide edge from which emission is collected. As seen from figure 2, the emission spectra comprises equally spaced mode structures, superposed over the amplified spontaneous emission (ASE). For a pump stripe of length 2 mm, the ASE threshold was observed to be 0.126 mJ/pulse. The threshold decreased for increased stripe lengths. For $z = 0$, i.e., when the pump stripe is placed right up to the edge

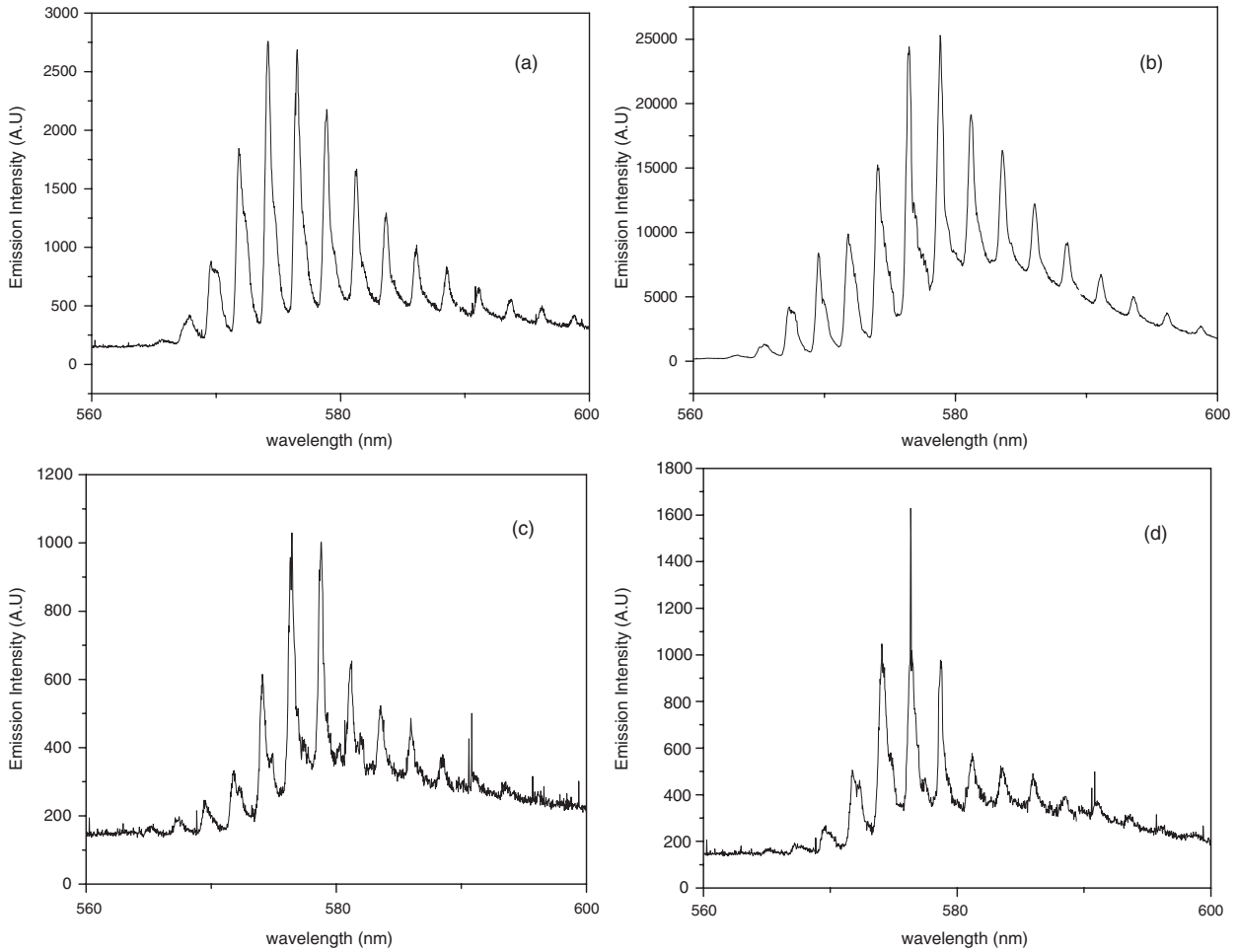

Figure 4. Emission from 50 μm thick film for $z = 5$ mm at pump energy: (a) $E = 0.02$ mJ/pulse; (b) $E = 0.2$ mJ/pulse; (c) $E = 0.63$ mJ/pulse and (d) $E = 1.82$ mJ/pulse. For (c) and (d) the Y-axis values should be multiplied with a factor of 10^2.

of the film, the mode structures are not prominent, except for some fine structures superposed over the ASE. When z is increased from 0, the emitted light is allowed to guide through the film so that the effect of the Fabry–Perot modes become predominant. Modes acquire sufficient gain so that the mode structures become prominent. Now the emission spectrum gets broader and we get intense mode structures over the spectral range 560–600 nm (figure 2(b)). Since the length of the unpumped region increases with increasing z, eventually the intensity of the whole emission together with the mode structures diminishes. The collected spectrum contains both the stimulated emission and ASE. We have noticed that the observed modes are the higher-order transverse modes (leaky modes) of the waveguide. For larger z, these modes are not propagating through the sample; they leak out. The modes which get propagated through the whole length of the unpumped region undergo self-absorption and remission. Hence there is a red-shift for the emitted light and for larger z; the ASE gets separated out of the laser emission as is clear from figures 2(d) and (e). The propagation of fluorescence as well as ASE in a similar sample geometry and pumping scheme has already been studied by the same authors in [25, 26].

Another important observation was the selective mode excitation on increasing the pump energy. Figure 4 shows the emission spectrum at $z = 5$ mm taken for various pump powers.

We see that, as the pump power is increased, the centre lines of the spectrum gain more energy. Though there is an increase in intensity for the whole emission, it is clear from the figures that energy from other modes is coupled to the high gain mode at 576.5 nm as the pump power is increased. The output intensity is very high for pump energies 0.63 and 1.82 mJ/pulse. To avoid the saturation of the CCD sensor, we placed a neutral density filter (with optical density = 2) just in front of the slit of the CCD. The intensity of the mode at 576.5 nm increases rapidly when the pump power reaches 1.82 mJ/pulse and the line width is reduced to ∼0.4 nm.

4. Conclusion

A compact solid-state laser based on dye-doped polymer is demonstrated. Leaky mode laser emission was observed from a transversely pumped free-standing poly methyl methacrylate film doped with rhodamine 6G. The film was 50 μm thick with dimensions 4 cm × 2 cm. Reflections from the lateral faces provided the optical feedback. This together with the guidance through the gain medium gave rise to intense narrow emission lines. For a pump energy of

1.82 mJ/pulse, an intense line with FWHM ∼0.4 nm was observed at 576.5 nm due to energy transfer from other modes.

Acknowledgments

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