Laser emission from self-assembled active photonic crystal matrix

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Abstract. Three-dimensionally ordered photonic crystals were grown using self-assembly technique from Rhodamine-B dye doped polystyrene micro-spheres resulting in a stop band at 611 nm overlapping the emission spectrum of the dye. When excited at a wavelength away from the stop band, using a frequency-doubled Nd:YAG laser, the crystal showed angle-dependent suppression of spontaneous emission of the dye in the wavelength range of the photonic stop band and enhancement at the band edge, in reflection and transmission geometries. Spectral narrowing, a sharp threshold and a highly directional emission, all indicative of stimulated emission, were observed from the active photonic crystal matrix.

Keywords: photonic crystals, self-assembly, emission, lasing.

1 INTRODUCTION

Artificial photonic crystals are gaining in popularity due to their increasing use in the production of optical devices. These structures contain periodically arranged dielectrics with lattice constants in the range of visible and near-IR wavelengths. Among a variety of applications of the photonic crystals, an important one is the fabrication of low threshold lasers [1]. In this connection, there have been several reports where the photonic crystals are used to control and modify the spontaneous emission of radiative materials [2-5]. The spontaneous emission of an emitter depends strongly upon its environment and it can be enhanced or inhibited depending on the allowed density of states in its environment. In photonic crystals, the photon density of states is lowered along the photonic stop band direction and enhanced at the band edges. Due to this property, the emission intensity of any embedded emitter in a photonic crystal environment redistributes and results in a modified emission spectrum. Different active materials, such as semiconductor quantum dots [2], rare earth ions [3] and laser dyes [4, 5], have been implanted into the 3D photonic crystal matrix and modified emission has been studied in earlier works. An important requirement for these studies is the overlap between the emission spectrum of the emitter and the photonic stop band, while its absorption band is required to be far from the range of photonic stop band.

In this Letter, the results from the laser-induced emission studies on dyed 3D colloidal photonic crystals are presented. The rhodamine-B (RhB) dye molecules are present uniformly inside each polystyrene (PS) sphere, constituting active building blocks of the crystal, and hence these dyed crystals are quite different from the crystals used in similar studies earlier, where the passive photonic crystals were either infiltrated with the dye solution [5, 6] or kept

inside a cuvette filled with the dye solution [4, 7, 8]. By an appropriate choice of the colloidal diameter, the stop band wavelength is made to overlap with the emission spectrum of RhB. The excitation wavelength of the emission studies lies outside the wavelength range of the stop band. The significance of this active matrix in obtaining a clear spectral narrowing in the stimulated emission spectrum, along with a sharp laser threshold and highly directional emission, is brought out in this work.

2 RESULTS AND DISCUSSION

The photonic crystals are grown within 3 hr by inward-growing self-assembly technique [9] using commercially available RhB-doped PS colloidal spheres of diameter 302 nm (PS-RhB). The self-assembly technique provides face-centered cubic lattice arrangement to the polymer spheres with (111) plane parallel to the substrate. The hexagonally arranged spheres in the (111) plane of the crystal can be seen in the scanning electron microscope (SEM) image of the crystal in Fig. 1(a). The inset in this figure is the photograph of a crystal grown to an area of 2.5 cm x 2.5 cm with the white circle indicating the void region unavoidable in this self-assembling technique. Figure 1(b) is the cross-sectional view of the crystal where the extent of layers is visible. The crystal has a uniform thickness with more than 25 ordered layers.
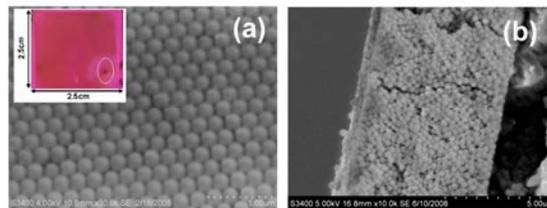
Fig. 1. SEM images of PS-RhB photonic crystal (a) (111) plane (scale bar 1 µm); inset is a photograph of the crystal (b) Cross-sectional view (scale bar 5 µm).

The PS-RhB photonic crystal has a maximum reflectance of 71% at 611 nm, measured at an incident angle of 8°. The reflection spectrum has a full width at half maximum (FWHM) of 46 nm. The Fabry-Perot oscillations on either side of the reflection peak provide an estimate of 28 layers for the ordered crystal [10]. The reflection spectrum of the crystal shows the expected shift towards the lower wavelength range for larger angles of incidence, indicating pseudo band gap nature. The peak reflectance and the FWHM of the reflection spectrum remain constant when measured along incident directions of 8° to 45°. This indicates a well-ordered photonic crystal with fewer defects.
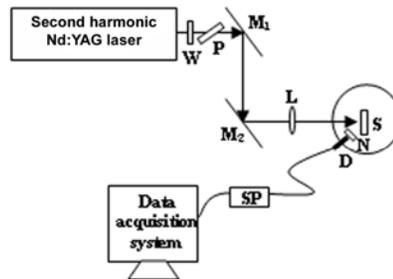
Fig. 2. Schematic of the experimental set-up used in laser-induced emission studies. W is a half wave plate, P is a polarizer plate at 55°, M_1 and M_2 are mirrors, L is a lens with a focal length of 10 cm, S is the crystal, N is the 532 nm cutoff notch filter and D is the detector connected to an optical fiber based spectrophotometer (SP).

The emission of the PS-RhB photonic crystal is studied by optically pumping it with the second harmonic of Nd: YAG laser at 532 nm, with a repetition rate of 10 Hz and average pulse duration of 6 ns. The schematic of the experimental set-up is shown in Fig. 2 where the crystal (S) is fixed at the center of a circular stage and the detector (D) is allowed to rotate around the crystal over 360°. The half-wave plate (W) and the polarizer plate (P) are used in the path to control the pump power.

The emission of the PS-RhB photonic crystal recorded in the reflection and the transmission modes of the photonic crystal are shown in Fig. 3(a) and Fig. 3(b), respectively, for certain chosen angles of the detector. The emission recorded at 60° from the direction of excitation beam is considered as the intrinsic emission of the dye (shown with a thin line) in the reflection mode since the stop band (reflection spectrum) and the emission spectrum do not overlap at angles greater than 45°. The thick line in Fig. 3(a-i) is the emission spectrum of PS-RhB photonic crystal at 8° which shows a dip as compared to the intrinsic emission of the dye. The position of the dip in the emission spectrum exactly matches with the peak of the reflection spectrum (dotted curve), indicated with an arrow in the figure. Due to the presence of stop band, there are no states available for emission to emerge from the crystal and this wavelength reflects back into the structure from the Bragg planes present along that direction. The dip in the emission spectrum blue shifted when the collection angle of the detector is increased from 8° to 30° as expected from the pseudo band gap nature of the PS-RhB photonic crystal. The position of the dip is indicated with an arrow in each figure. At 45°, the stop band of the photonic crystal has moved far away from the range of its emission spectrum, and hence no effect of stop band is observed on the emission at this angle, as can be seen in Fig. 3(a-iv).
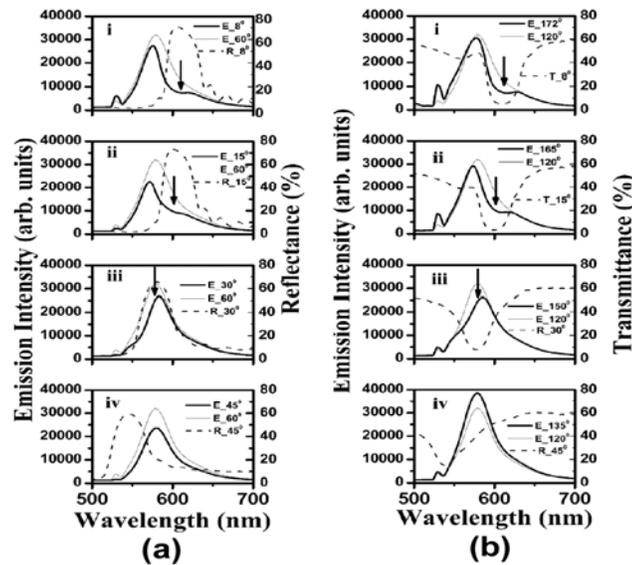
Fig. 3. Spontaneous emission of PS-RhB photonic crystal at different angles is shown with thick line in (a) reflection mode and (b) transmission mode. The dotted line is the reflection spectrum in (a) and transmission spectrum in (b). The thin line in all the figures is the intrinsic emission of the dye. The measurement is done for laser pulse energy of ~ 8.3 mJ.

The emission spectrum of the crystal for some angles in transmission mode of the crystal is shown in Fig. 3(b). An effect similar to that in reflection mode is observed in this case. For

the transmission mode, emission at 120° is considered as the intrinsic emission. The minimum in the transmission spectrum and the dip in the emission spectrum occur at the same wavelength. In each sub-figure of Fig. 3, the emission of the dye is inhibited in the photonic stop band range and enhanced at the band edge, depending upon the number of allowed density of states. The enhancement is clearer in the case of transmission. Similar enhancement in transmission mode was observed by Eradat et al. [4] in the case of SiO_2 photonic crystal placed in a cuvette filled with the dye solution. The dip in the emission spectrum as an effect of the photonic stop band is confirmed in the present work by recording the emission from RhB dye solution, colloidal PS-RhB solution and PS in toluene solution at different angles. No dip is observed in the emission spectra of these solutions and the spectral features remain unchanged at all angles.

In these emission studies, a broad spectrum is observed for spontaneous emission with its peak at 598 nm and FWHM of 65 nm at lower excitation powers. Figure 4(a) shows the emission spectrum of the PS-RhB photonic crystal recorded at 22° at different pump powers. When the pump power is increased, the FWHM of the spectrum reduces to 9 nm with its peak at 587 nm along this direction. The open circle curve in the figure is the reflection spectrum of the PS-RhB crystal along 22°. Intriguingly, the spectral narrowing is still within the stop band governed by the reflection spectrum. The reason for the preferential peak is associated to the spectral dependence of emission cross-section in RhB dye. The emission cross-section of RhB in ethanol has a peak at 580 nm [11] which can be the natural wavelength for lasing of RhB in a structure such as a self-assembled photonic crystal. The slight difference in wavelength between this value (580 nm) and our experimental observation (587 nm) is attributed to the changes in RhB characteristics, such as its absorption and emission cross-sections, brought about by its chemical combination with the PS polymer in the colloids used in this work. The sharp narrowing of the stimulated emission spectrum is found to be associated with a slightly enhanced emission close to the band edge of the reflection spectrum at this angle (indicated with a dotted vertical line in Fig. 4(a)). A certain extent of spectral narrowing is also observed along a few more directions such as at 18° and at 151° (in transmission mode) but the narrowing is not significant for these angles even for larger pump powers. Since the reflectance of the PS-RhB photonic crystal is almost the same for all the angles from 8° to 45°, tunable narrowing was expected. However, this was not observed and the spectral narrowing was preferentially present only at 587 nm. In Fig. 4(b), the integrated emission is plotted as a function of the input pump power. A clear evidence of threshold in the switch-over from spontaneous to stimulated emission is seen in this figure at a recording angle of 22° at a power of 74 mW (with peak intensity of 158 MW/cm^2).
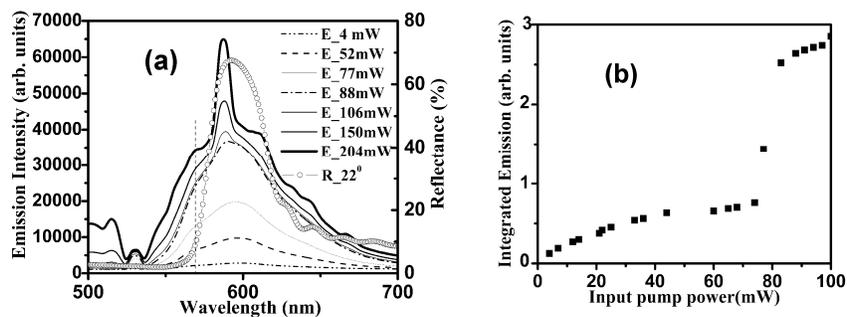
Fig. 4. (a) Emission spectrum of PS-RhB photonic crystal along 22° for different pump powers. The open circle curve is the reflection spectrum of the crystal at 22°. (b) Plot between integrated emitted power and input pump power showing the threshold of lasing at 74 mW.

In an earlier report, lasing was achieved at a threshold intensity of 13.2 GW/cm^2 by infiltrating an inverse photonic crystal with Rhodamine-6G dye solution [6]. Infiltration of dye solution into the voids will precipitate the dye on the walls of the voids. Frolov *et al.* [7] and Shkunov *et al.* [8] achieved lasing in colloidal photonic crystals, at a threshold of 6 MW/cm^2 and at 8 MW/cm^2 respectively, by keeping the passive polymer photonic crystal inside a cuvette filled with the dye solution. In these cases, lasing due to cavity effects from the parallel faces of the cuvette cannot be ruled out. The threshold obtained in the present work cannot be directly compared with these reports since the quantity of the dye is extremely small (concentration of 0.09 wt %) and the building block of the crystal is different with the gain medium being present in each colloidal sphere. Li *et al.* in 2007 reported the spectral narrowing in dye doped polymeric photonic crystals [12] where the FWHM of the emission spectrum reduced to 15 nm but a clear threshold for spectral narrowing was not observed as in the present case. The photostability characteristics of a dye entrapped in a colloidal sphere are different from the dyes infiltrated through solution chemistry [13].

3 CONCLUSION

Three dimensionally ordered, very-high-quality RhB-doped polymeric photonic crystals were fabricated using a simple self-assembly technique within 3 hr. The spontaneous emission of the embedded dye was studied along different directions of the crystal and for different pump powers. Due to the reduced density of states at the stop band wavelengths, the structure does not allow the emission to transmit effectively out of the crystal at these wavelengths, and it appears as an angle-dependent dip in emission. This suppression in the emission spectrum at the stop band is compensated as an enhancement at the band edge wavelength of the crystal because of the presence of more allowed states. This modification in the spontaneous emission of the dye is observed both in reflection and transmission modes of the crystal. To our knowledge, this is the first report of lasing in an active dyed photonic crystal matrix with a reflectance of only 71% and with a very small concentration of the dye. The spontaneous emission of the RhB dye narrowed down along 22° from the direction of pumping with a selective preference for emission peak at 587 nm at a threshold power of 74 mW. This wavelength is very close to the peak in emission cross-section of the conventional RhB dye. Hence in this work, the micro cavity photonic crystal laser with a lasing wavelength at 587 nm is demonstrated through the three properties of spectral narrowing, threshold effect and preferential directionality.

Acknowledgments

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