

Electronic Supporting Information

Low Threshold Photonic Crystal Laser Based on Rhodamine Dye Doped High Gain Polymer

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1. Preparation of monodisperse PS microparticles

Monodisperse PS microparticles with the size of 265 nm were synthesized following the emulsion polymerization.¹ Styrene (113.1 mmol) was dispersed into 100 mL deionized water. A minute amount of surfactant sodium dodecyl sulfonate (SDS, 0.38 mmol) and buffer agent of ammonium bicarbonate (0.89 mmol) was dissolved. The reaction mixture was kept at 70 °C for half an hour and bubbled with N₂. After adding an aqueous solution of potassium persulfate (KPS, 0.15 mmol), the polymerization was carried out at 80 °C for 10 hours with continuous stirring.

2. Preparation of PS colloidal crystals (PS CCs)

The PS CCs were prepared by vertical deposition method.² Firstly, glass substrates were sonicated in acetone and ethanol for 15 min and cleaned with piranha solution (7:3 H₂SO₄/H₂O₂) at 90 °C for 1 h. **Caution:** Piranha solution is strong oxidant and should be handled with care. The glass substrates were rinsed with deionized water and then dried in vacuum oven. Secondly, the treated glass substrates were vertically dipped in container with PS colloidal solution of 0.18 wt % solid density, which was kept in oven at 65 °C. PS CCs were deposited on the surfaces of the glass substrates for 48-72 hours, which was confirmed by the presence of brilliant color on the substrate. PS CC on one side of substrate was scraped off, while that on the other side is left for characterization and resonating cavity construction.

3. Quality factors (Q) of the resonating cavity were determined by equation S1:

$$Q = \frac{\lambda}{\Delta\lambda} \quad (S1)$$

λ is the lasing wavelength and $\Delta\lambda$ is the full-width-at-half-maximum of the lasing peak.

4. ¹H NMR of lactone form of *t*-Bu RhB

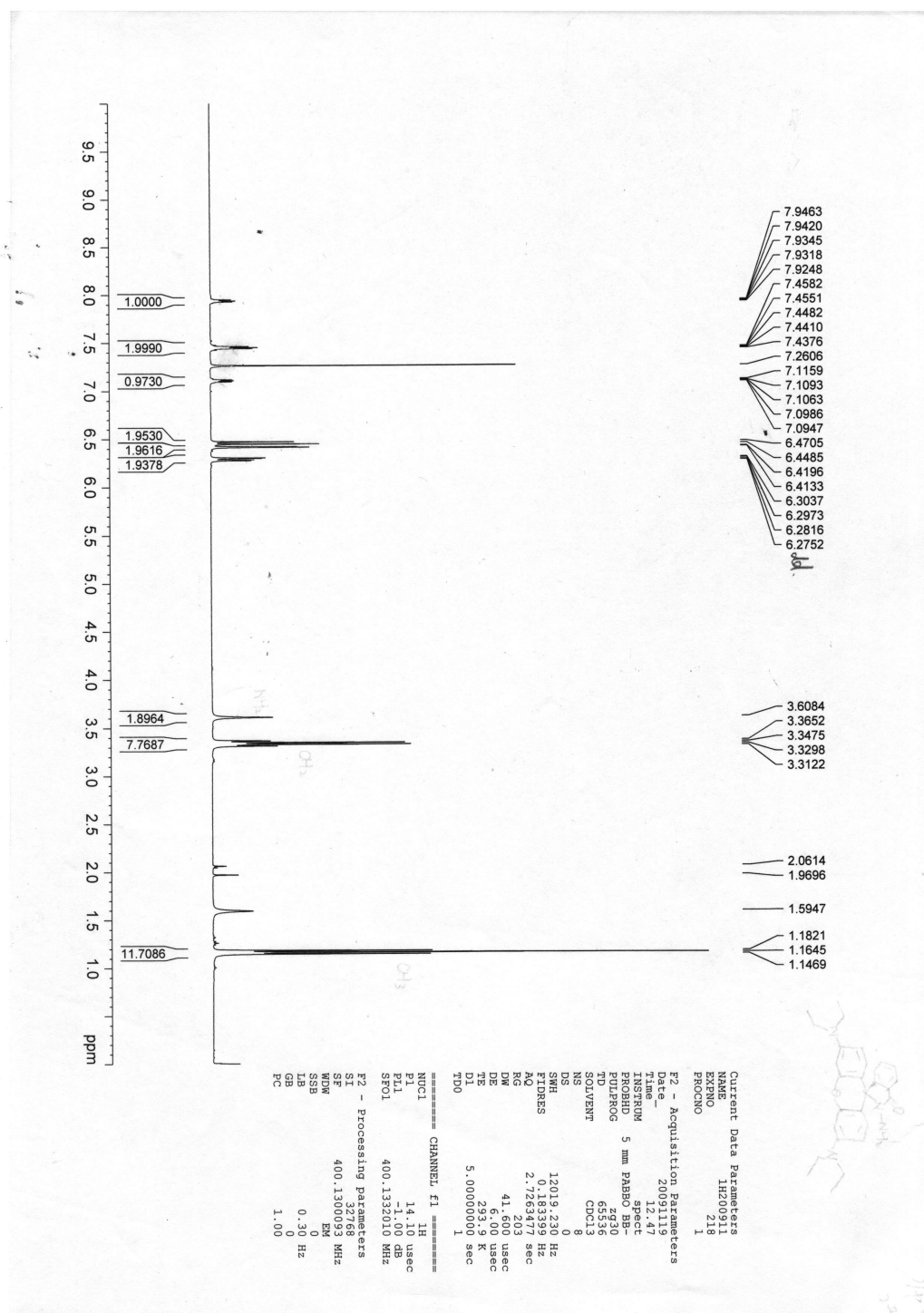


Fig. S1 ¹H NMR of the lactone form *t*-Bu RhB.

5. Chemical structures of the components of the photopolymerizable resin

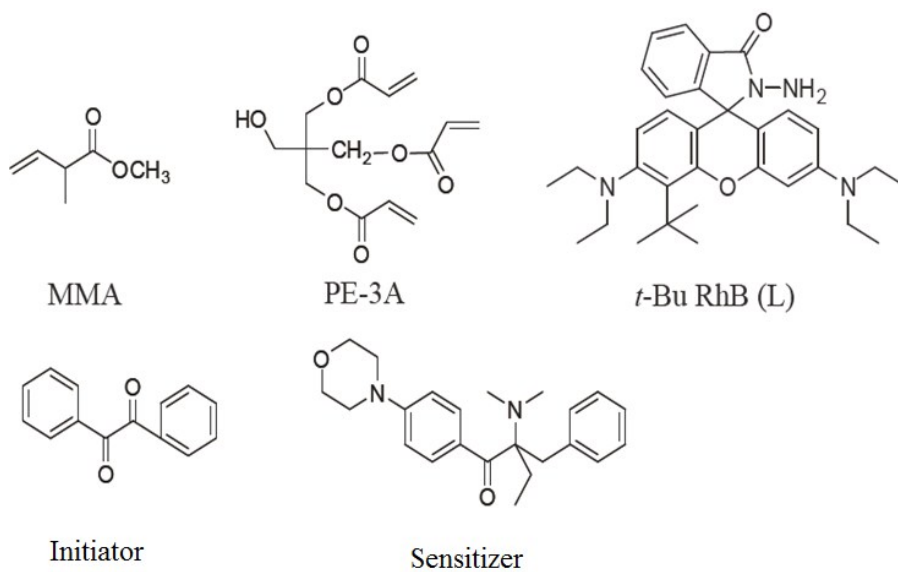


Fig. S2 Chemical structures of the components of the photopolymerizable resins.

6. Angel-resolved lasing measurements

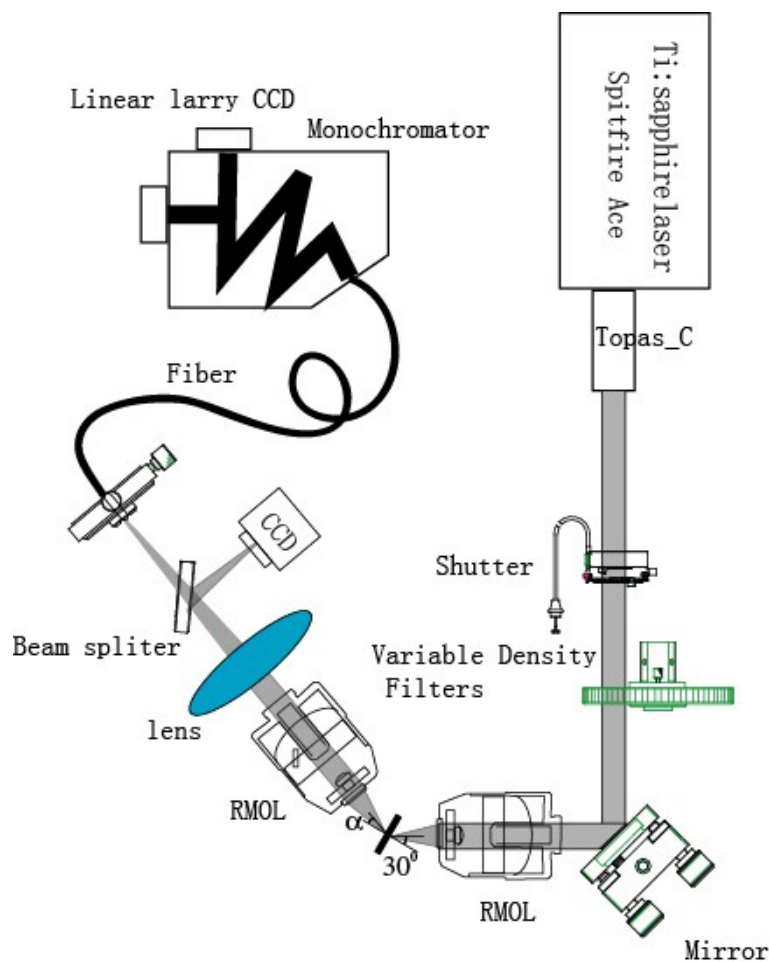


Fig. S3 Experimental setup for the angle-resolved lasing measurement.

Fig. S3 depicts the experimental setup to perform the angle-resolved lasing measurement. The excitation light source was 532 nm femtosecond laser beam produced by an optical parametric amplifier (TOPAS-C), which was pumped by a Ti:sapphire femtosecond (fs) laser amplifier (Spitfire Ace). The excitation laser has a pulse duration of 140 fs and repetition rate of 4 Hz. The average excitation power was 11.2 nW. The pump beam was focused on the sample surface at an incident angle of 30° . The PhC resonating cavity was mounted on a rotating stage, which allows the emission spectra to be measured at different detection angles. The emission spectra were collected by a Reflective Microscope objective lens (20X, NA0.35, SIGMA

KOKI OBLR-20A) and sent through a bundled optical fiber connected to an Omni- λ 300 monochromator (Zolix, 0.01 nm spectral resolution) and the spectral was then detected by an linear array CCD (AmesD7222, Series, Scientific, 200 nm-1100 nm) mounted on the monochromator.

7. SEM images of *t*-Bu-RhB doped PMMA films

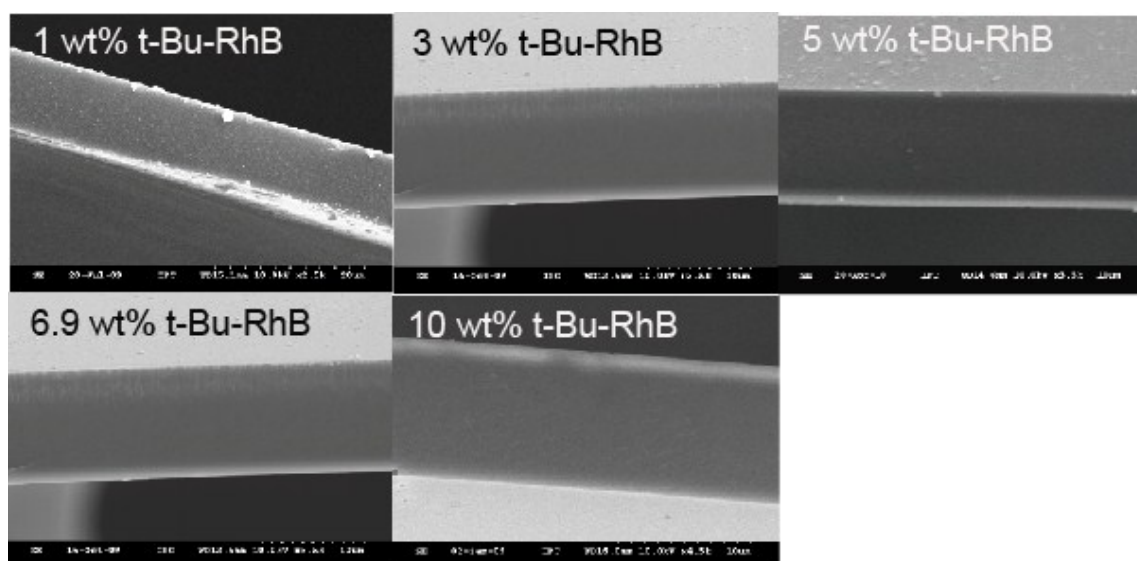


Fig. S4 SEM images of *t*-Bu-RhB doped PMMA films with different concentrations of *t*-Bu-RhB.

8. PL spectra of *t*-Bu-RhB doped PMMA films

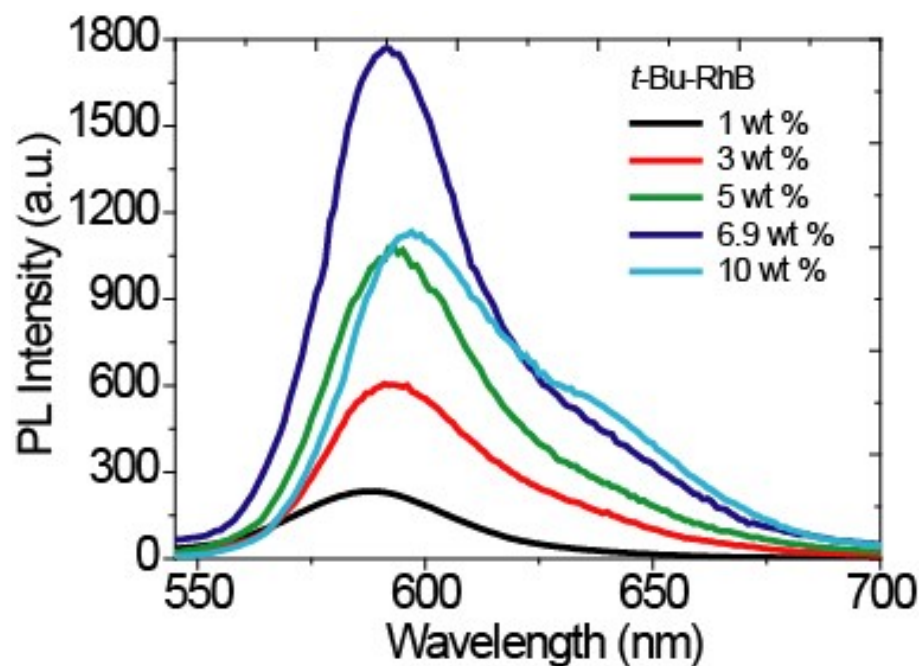


Fig. S5 Photoluminescence spectra of *t*-Bu-RhB doped PMMA films with different *t*-Bu-RhB concentrations.

9. PL Lifetimes of *t*-Bu-RhB doped PMMA films

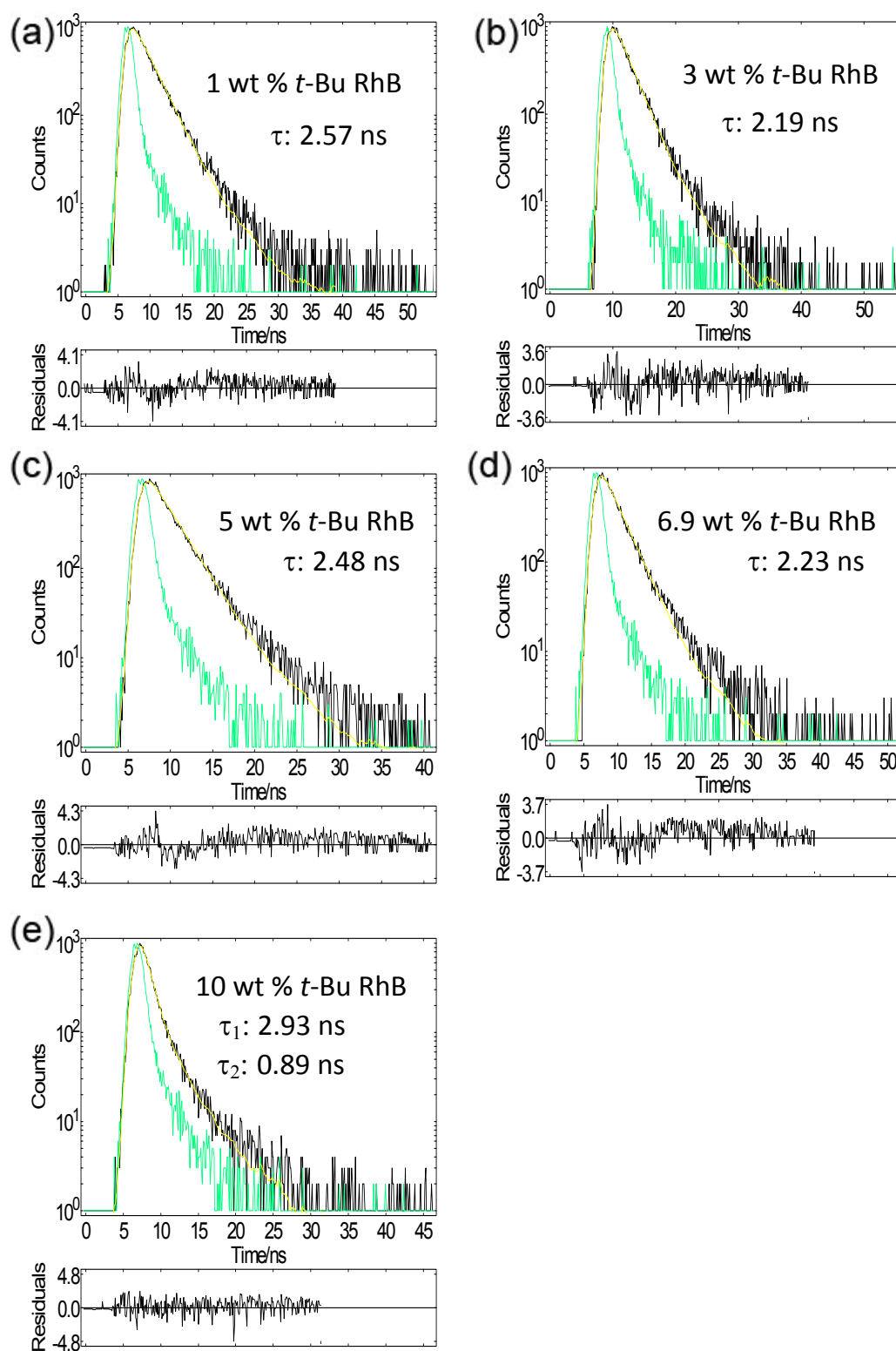


Fig. S6 Photoluminescence lifetime measurement of *t*-Bu-RhB doped PMMA films with (a) 1, (b) 3, (c) 5, (d) 6.9, and (e) 10 wt% *t*-Bu-RhB concentrations.

Photoluminescence lifetime measurements of *t*-Bu-RhB doped PMMA films were

carried out by using single photon count fluorescence spectrometry (F900, Edinburgh Instrument Co. UK). The data were recorded at 592 nm with excitation wavelength at 532 nm. The decay of the excited state of the dye molecules to the ground state could be expressed as equation (S2) for 1, 3, 5, 6.9 wt % *t*-Bu-RhB doped PMMA films:

$$I(t) = A + Be^{(-t/\tau)} \quad (\text{S2})$$

, and equation (S3) for 10 wt % *t*-Bu-RhB doped PMMA film:

$$I(t) = A + Be^{(-t/\tau_1)} + Ce^{(-t/\tau_2)} \quad (\text{S3})$$

10. Transmission spectrum of the photonic crystal resonating cavity

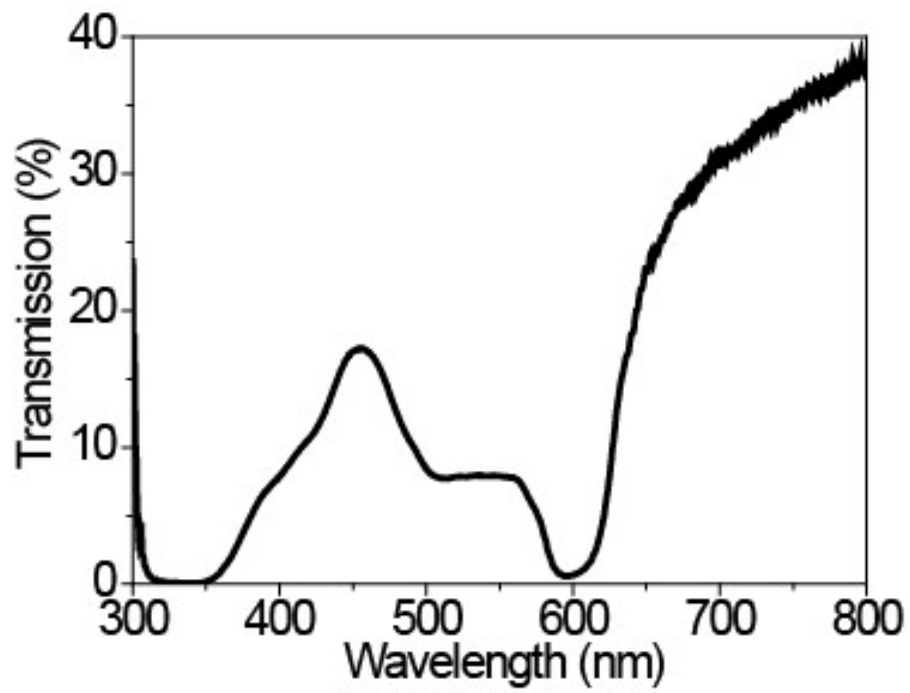


Fig. S7 Transmission spectrum of the photonic crystal resonating cavity.

11. Lasing spectra of PhC resonating cavities with different *t*-Bu-RhB concentrations

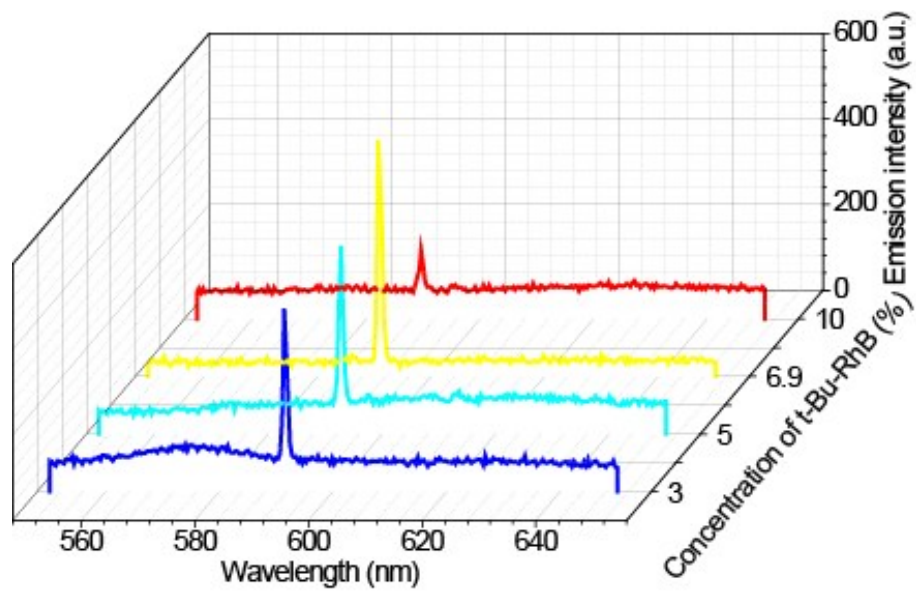


Fig. S8 Lasing spectra of the photonic crystal resonating cavities with different *t*-Bu-RhB concentrations

12. Emission spectra of *t*-Bu-RhB doped PMMA films under Nd:YAG laser excitation

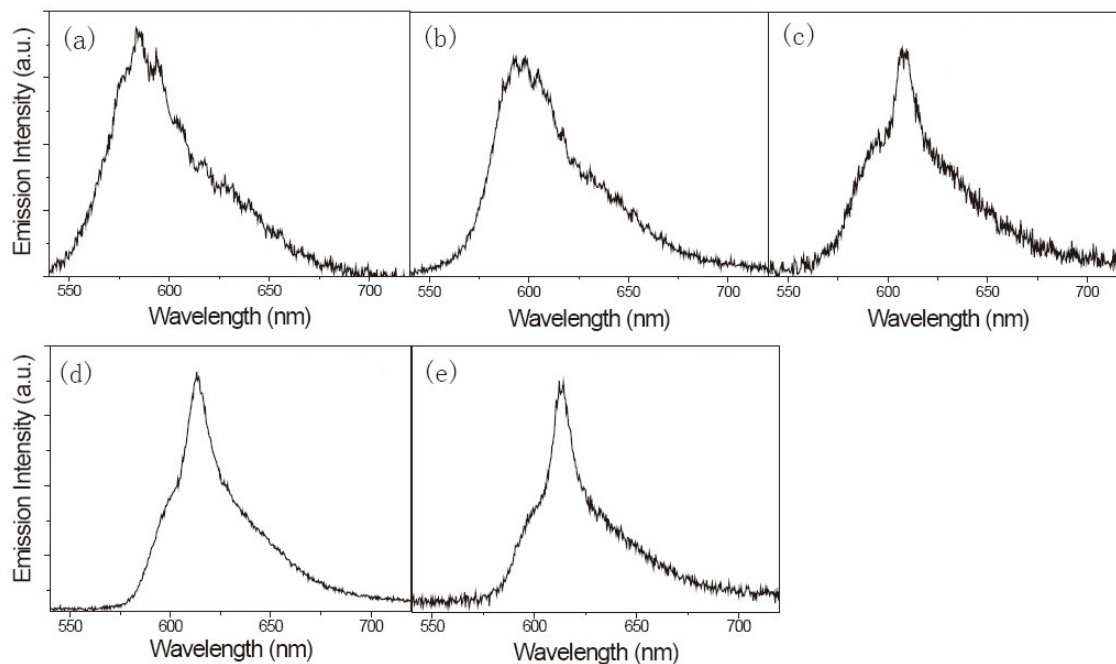


Fig. S9 Emission spectra of *t*-Bu-RhB doped PMMA films under Nd:YAG laser excitation, with *t*-Bu-RhB concentration at (a) 1, (b) 3, (c) 5, (d) 6.9, and (e) 10 wt %, respectively. The pump energy is 20.22 MW/cm².

13. Transmission spectra of the PS CCs with different PBGs

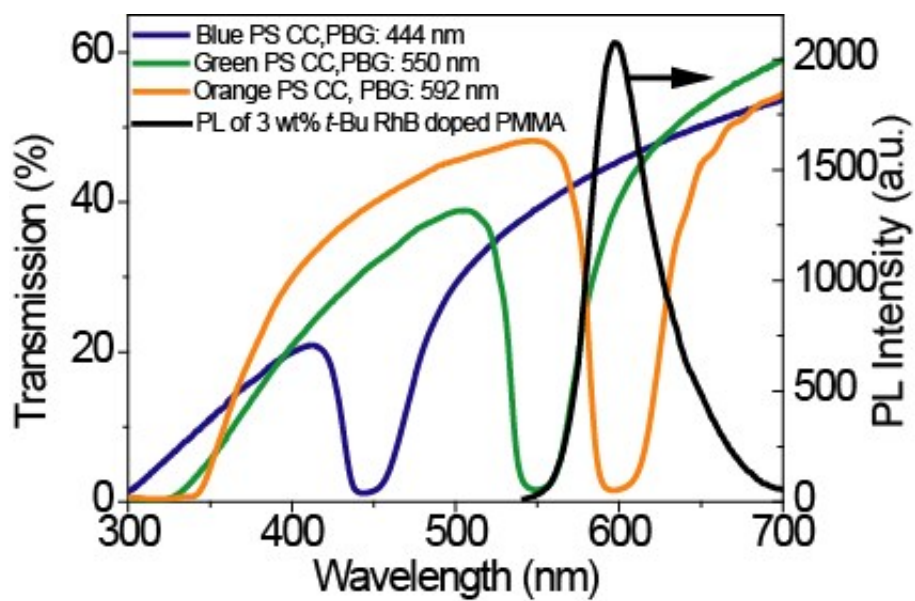


Fig. S10 Transmission spectra of the PS colloidal crystals with PBG of 444nm, 550nm and 592 nm, respectively. Photoluminescence spectrum of the 3 wt % *t*-Bu-RhB doped PMMA film.

14. Emission spectra of 3 wt % *t*-Bu-RhB doped PMMA film

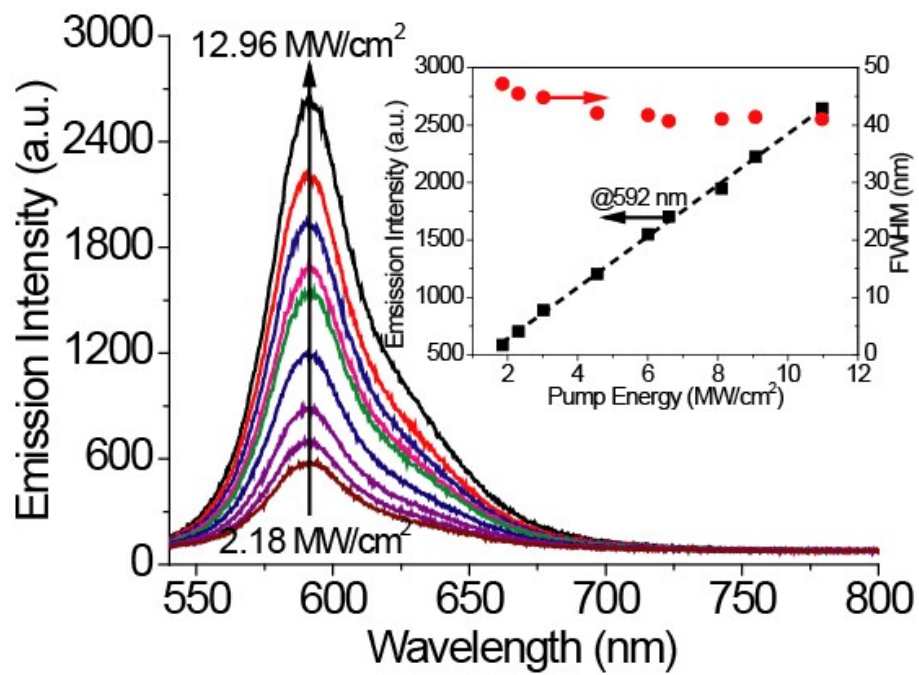


Fig. S11 Emission spectra of the 3 wt % *t*-Bu-RhB doped PMMA film without PC resonating cavity under different optical excitation. The inset is the dependence of the emission intensity and FWHM on the excitation intensity.

15. Beam quality analysis of the lasing emission

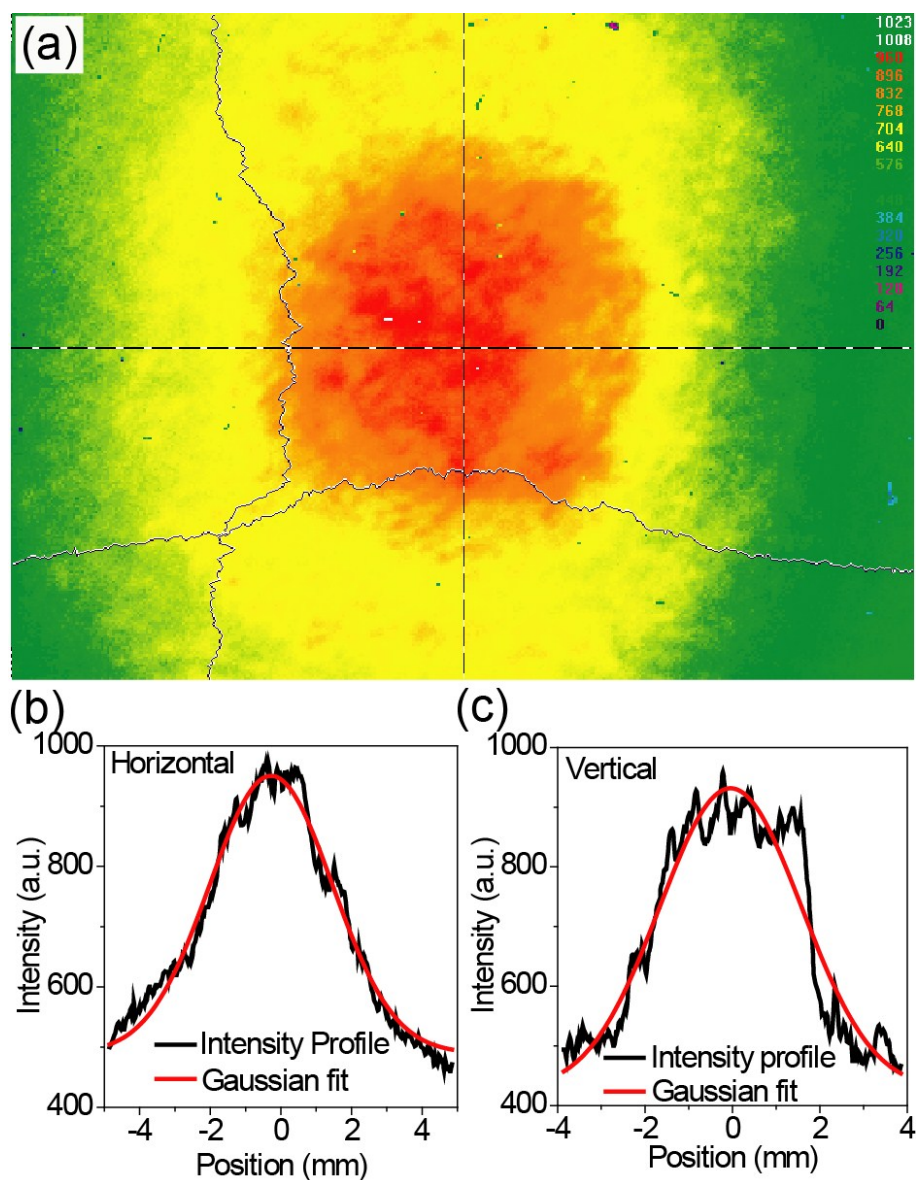


Fig. S12 (a) Spatial profile of the lasing emission shown in Fig. 7(a). Intensity profile and Gaussian fit to the center of the lasing beam from (b) horizontal and (c) vertical direction.

A COHU 7512 silicon CCD camera was used with Spiricon LBA-PC V4.08 software (Spiricon Inc., USA) to directly image the emitted lasing beam profiles and displayed them on a computer monitor. The beam quality of lasing emission was also analyzed by the same software.

The beam quality of the lasing emission at 592 nm was analyzed by a CCD beam profiler. Fig. S10 shows the intensity profile and intensity distribution of the lasing

emission beam. As is shown from the figure, the lasing beam has a nearly round shape with high symmetry. The intensity profile plotted in Fig. S10(b)-(c) indicates a good Gaussian fit to the output intensity distribution. Meanwhile, the Gaussian fit is better in horizontal direction than that in vertical direction. High-quality of the lasing emission was verified by the round-shape lasing spot with Gaussian intensity profile, which is similar to the recently reported result in the vertical-cavity surface-emitting laser.⁴

16. Table S1. The components of the *t*-Bu-RhB doped PMMA films.

Sample No.	<i>t</i> -Bu RhB (wt %)	MMA (wt %)	PE-3A (wt %)	Benzil (wt %)	Sensitizer (wt %)
1	1	49.1	47.9	1	1
2	3	49.3	45.7	1	1
3	5	46.4	46.6	1	1
4	6.9	46.2	44.9	1	1
5	10	44.4	43.6	1	1

References:

- S1 J. X. Wang, Y. Q. Wen, H. L. Ge, Z. W. Sun, Y. M. Zheng, Y. L. Song and L. Jiang, *Macromol. Chem. Phys.*, 2006, **207**, 596-604.
- S2 P. Jiang, J. F. Bertone, K. S. Hwang and V. L. Colvin, *Chem. Mater.*, 1999, **11**, 2132-2140.
- S3 H. Yamada, T. Takamura, Y. Yamada and K. Yano, *Adv. Mater.*, 2009, **21**, 4134-4138.
- S4 B. Guzelturk, Y. Kelestemur, K. Gungor, A. Yeltik, M. Z. Akgul, Y. Wang, R. Chen, C. Dang, H. Sun and H. V. Demir, *Adv. Mater.*, 2015, **27**, 2741-2746.