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Hideo Ohtaka<sup>a</sup>, Toshiki Koyama<sup>a</sup> & Yoshio Taniguchi<sup>a</sup>

<sup>a</sup> Faculty of Textile Science and Technology, Shinshu University, Ueda, Nagano, 386-8567, JAPAN

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## Effects of Microparticle Dispersion on Laser Emission from Dye-Doped Microspheres

HIDEO OHTAKA, TOSHIKI KOYAMA and YOSHIO TANIGUCHI

*Faculty of Textile Science and Technology, Shinshu University, Ueda,  
Nagano 386-8567, JAPAN*

Effects of dispersion of SiO<sub>2</sub> microparticles (SOP, 5.8  $\mu\text{m}$  in diameter) into Rhodamine 6G-doped microsphere on photo-pumped laser emission have been investigated. The microparticles with different refractive index dispersed in the microsphere scattered pump laser (337 nm) in the microsphere cavity. In the microsphere including SOP, emission peaks of lasing in whispering gallery mode (WGM) appeared when it was pumped at the energy density above the threshold of 0.6  $\text{mJ pulse}^{-1}\text{cm}^{-2}$ . And their full widths at half maximum (FWHM) of the peaks were about 0.5 nm. Threshold for laser emission in WGM from R6G/pH-SiO<sub>2</sub>/SOP was lower than that from the microsphere without SOP. It was demonstrated that microparticles dispersion into the microsphere cavity is clearly effective on reduction of the threshold for the WGM laser emission.

**Keywords:** laser emission; microsphere cavity; microparticle; scattering pumping light; dye-doped laser; whispering gallery mode

### INTRODUCTION

Microsphere lasers are expected for novel laser devices such as a submicron-sized illuminant, a dot laser array, and so on. Photo-pumped laser emission from dye-doped phenyl hybridized-SiO<sub>2</sub> (pH-SiO<sub>2</sub>) microspheres with a micrometer-sized diameter prepared in the sol-gel technique was previously reported [1]. Lower threshold and higher efficiency of the dye-doped microsphere laser are achieved by lower reabsorption of dye and higher excitation probability in a resonance cavity. As one method for improvement of excitation probability, the scattering of pumping light by dispersion of microparticles into a microspherical cavity formed by a laser dye solution was reported [2]. In addition, strongly scattering media that formed of a colloidal suspension of titanium dioxide microparticles in a dye solution was used as a pseudo-cavity for the

multi-mode laser oscillation without any external cavity [3, 4]. As another effect of dispersion of microparticles into laser dye solution, the single-mode laser oscillation by the distributed feedback using a layer of microparticles which have the same diameter in the dye solution cell was observed [5].

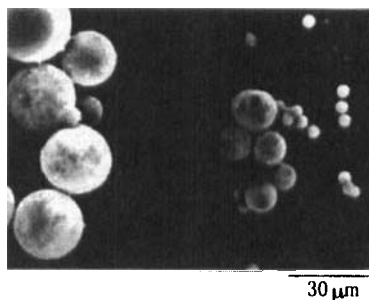
The present paper describes the demonstration of effects of dispersion of  $\text{SiO}_2$  microparticles into Rhodamine 6G-doped pH- $\text{SiO}_2$  microspheres on lasing behavior.

## EXPERIMENT

The preparation of Rhodamine 6G-doped pH- $\text{SiO}_2$  microspheres including  $\text{SiO}_2$  microparticles (R6G/pH- $\text{SiO}_2$ /SOP) was referred to the Shibata's method [1]. A mixture of Rhodamine 6G (R6G) (0.02 g), phenyltriethoxysilane (2 ml), and pH 3 HCl aqueous solution (3.2 ml) was stirred for 24 hours at room temperature followed by addition of  $\text{SiO}_2$  microparticles (SOP) of 5.8  $\mu\text{m}$  in diameter (0.1 g). Then the mixture was added dropwise to a 100 ml NaOH aqueous solution (pH 12.8) with stirring, and was kept for 6 hours. The resultant solution containing R6G/pH- $\text{SiO}_2$ /SOP was dialyzed for 2 days in pure water, followed by filtration to remove smaller microspheres than 6  $\mu\text{m}$  in diameter and non-reacted SOP. The amount of doping of the dye was determined by measuring of absorbance of R6G EtOH solutions extracted from R6G/pH- $\text{SiO}_2$ /SOP. For measurement of fluorescence and emission spectra, the microspheres were mounted on a non-fluorescent glass substrate in mono-dispersion. Emission light obtained by pumping with an  $\text{N}_2$  gas laser ( $\lambda_{\text{em}} = 337$  nm) was detected by an image intensified CCD camera (DH520-18U-01 ANDOR) attached to a monochromator (MS1271<sup>TM</sup> ORIEL Inst.).

## RESULT AND DISCUSSION

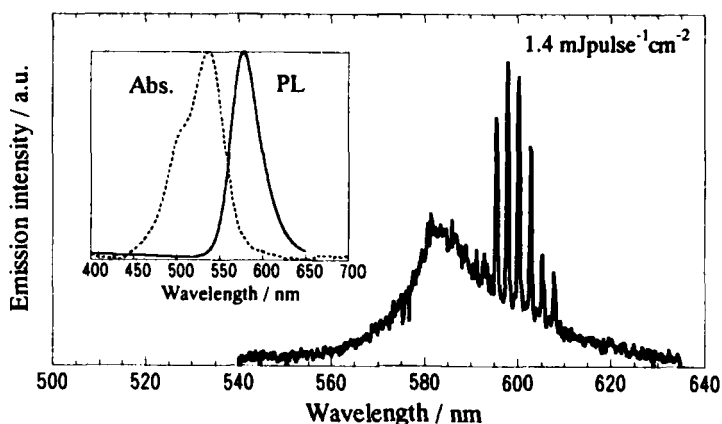
A SEM image of R6G/pH- $\text{SiO}_2$ /SOP was shown in Figure 1. In spite of embedding SOP, sphericity of R6G/pH- $\text{SiO}_2$ /SOP was kept. The diameter of



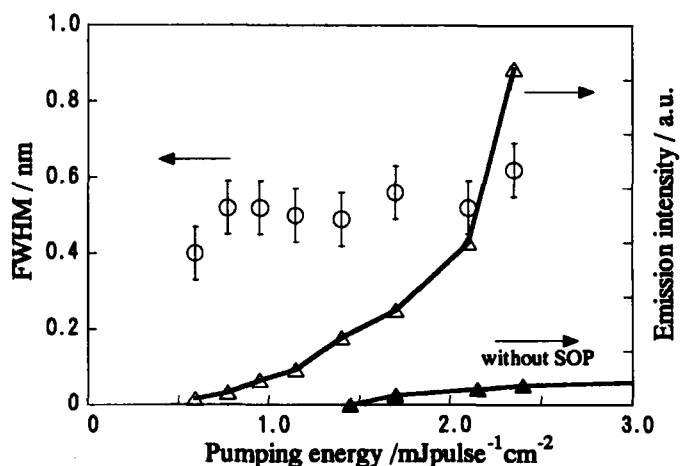
**FIGURE 1** SEM image of R6G/pH- $\text{SiO}_2$ /SOP.

R6G/pH-SiO<sub>2</sub>/SOP distributed from 6  $\mu\text{m}$  to 80  $\mu\text{m}$ , and was averaged to 30  $\mu\text{m}$ . If the reaction solution was not mixed with SOP, the diameter of R6G/pH-SiO<sub>2</sub> was averaged to 15  $\mu\text{m}$ . Because of SOP acted as a nucleus for formation of the microsphere, larger microsphere could be obtained than that in the absence of SOP. R6G was doped into R6G/pH-SiO<sub>2</sub>/SOP at content of about  $1.00 \times 10^{-5} \text{ mol g}^{-1}$ , and mainly maintained monomeric state more than dimeric state in which fluorescence is quenched.

Figure 2 showed the typical emission spectrum from R6G/pH-SiO<sub>2</sub>/SOP pumped with an N<sub>2</sub> gas laser at energy density of  $1.4 \text{ mJ pulse}^{-1} \text{ cm}^{-2}$ . Several sharp emission peaks with a full width at half maximum (FWHM) of about 0.5 nm were appeared at around 600 nm. The peak-intervals were 2.3 - 2.4 nm. When supposing that the diameter of the microsphere is 33.9  $\mu\text{m}$ , the calculated values of the oscillation wavelength agreed with the observed values in Figure 2, and were supported that the lasing is in the whispering gallery mode (WGM). Input-output properties of a lasing peak at 600 nm were showed in Figure 3. The threshold of pumping energy for the WGM lasing was  $0.6 \text{ mJ pulse}^{-1} \text{ cm}^{-2}$ . Above the threshold, the intensity of the emission remarkably increased with increasing the pumping energy. But FWHM was constant at about 0.5 nm. When comparing with R6G/pH-SiO<sub>2</sub> which not included SOP, FWHM for R6G/pH-SiO<sub>2</sub>/SOP was the same but the lasing threshold was reduced from 1.4 to  $0.6 \text{ mJ pulse}^{-1} \text{ cm}^{-2}$ . From the reduction of the threshold for the WGM lasing, it could be demonstrated that the excitation probability of the laser dye in R6G/pH-SiO<sub>2</sub>/SOP increased with scattering of the pumping light on the embedded microparticles.



**FIGURE 2** Emission spectrum of R6G/pH-SiO<sub>2</sub>/SOP microsphere pumped by N<sub>2</sub> gas laser at  $1.4 \text{ mJ pulse}^{-1} \text{ cm}^{-2}$ . Inset: Absorption and photoluminescence (PL) spectra of R6G/pH-SiO<sub>2</sub>/SOP



**FIGURE 3** Dependence of FWHM and emission intensity of WGM peaks for R6G/pH-SiO<sub>2</sub>/SOP (open) and R6G/pH-SiO<sub>2</sub> (close).

### Conclusion

Dispersion of the SiO<sub>2</sub> microparticles into the microsphere cavity for the WGM lasing brought clearly effects on the reduction of threshold and the improvement of the emission efficiency. Further improvements will be achieved by optimization of diameter, refractive index, and arrangement of the embedding microparticles into the microsphere cavity.

### Acknowledgement

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