PREPARATION OF MICROMETER-SIZE SUPER-SPHERICAL GLASSES FOR OPTICAL RESONATOR

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Micrometer-size super-spherical glass (μ-SSG) particles were prepared for the optical resonator from Eu_2O_3-doped Na_2O-CaO-SiO_2 glass fine particles. The glass particles were heat-treated on the glassy-carbon substrate to have a super-spherical shape. The obtained glass particles had super-spherical shape with very smooth surface. The luminescence spectra from this μ-SSG were measured by introducing CW-Ar^+ ion laser light of 514.5nm into its equatorial plane to excite doped Eu^{3+} ions, and the resonance signals due to Whispering Gallery Modes were observed on the luminescence spectra of Eu^{3+}:$^5D_0 \rightarrow ^7F_j$ transitions in 570-640 nm range. The Q factor was estimated to be $>10^3$, and ensured that the prepared μ-SSG has a perfect circle plane to act as an optical resonator.

(Key words: super-spherical glass, optical resonator, Whispering Gallery Mode, and europium oxide)

1. Introduction

A micrometer-size glass sphere is known to act as an optical resonator. Photons are effectively encapsulated and the resonance modes called as “Whispering Gallery Mode (WGM)” are induced [1]. Various optical devices using WGM in spheres have been proposed; a light emitting source of multi-wavelengths, the stimulated Raman amplifier [2, 3], the probes for the near-field optical microscope [4], the components of photonic crystals [5] and so on. There has been reported a lot of works on WGMs in microspheres: liquid-droplets and/or plastic particles containing organic dyes [6, 7], dye-doped organic/inorganic hybrid particles [8, 9], neodymium-doped glass particles [10], fused silica particles [3] and so on. In these materials, the glasses have wide optical windows from ultra-violet to infrared region, and a wide variety of the optical constants. Further, glass materials show high photo-stability compared with organic materials, and great potentiality to be utilized as the resonator matrix materials.

The size of the particles is ensured theoretically to be one of the most important factors to attain high efficiencies of resonator called by Q-factor. Hill and Benner proposed that micrometer-size particle is favorable [11].

The partly truncated spheres called as ‘super-spherical particles’ are also capable to induce WGM resonance inside, because this shape has spherical part to hold resonance mode. If Hill and Benner’s proposal is applied to this shape, the micrometer-size super-spherical particles can become quite fascinating resonators. Recently, our group has developed the novel preparation method of micrometer-size super-spherical glasses (μ-SSG). Using this preparation method, quite smooth spherical surface can be obtained and no additional treatment is required. We prepared μ-SSG using Na_2O-CaO-SiO_2 glass and found that this μ-SSG particle satisfies the optical constant of the solid immersion lens (SIL) to attain optical super-resolution using the evanescent wave. We also confirmed that the obtained μ-SSG can realize super-resolution, and was named μ-SIL [12].

In this study, we prepared μ-SSG containing luminescent materials Eu_2O_3 using Na_2O-CaO-SiO_2 glass, and investigated their optical functionality of WGM resonator.

2. Experimental

2.1. Sample Preparation

A glass sample with the compositions of $20Na_2O$-10CaO-$70SiO_2$ containing $0.5Eu_2O_3$ (in mol%) was prepared by the conventional melting and quenching method. Reagent grade raw materials, Eu_2O_3,

Na_2CO_3, $CaCO_3$, and SiO_2 were mixed and melted in a platinum crucible at 1350ºC for 1hour. In order to ensure the glass homogeneity, the glass was crushed and re-melted for another 1hour. After second melting, the glass melt was poured onto a graphite plate and immediately transferred into the annealing furnace preheated to 535ºC (T_g+20ºC). The glass sample was held at an annealing temperature for 1hour and cooled to room temperature by 1ºC/min.

The obtained glass was crushed and classified by using a filter paper and a sieve in a 0.2mass% $(NaPO_3)_6$ aqueous solution in order to obtain the glass particles with the size from 5 to 20μm. Their particles were washed on the filter paper by pure water, and trapped in ethanol. Then the particles were dispersed on the finely polished glassy carbon (g-carbon) substrate using CCl_4. The particles on g-carbon were heated up to 800ºC and held for 30min under the atmosphere of H_2/N_2=1/5, and then cooled to the room temperature. The heating rate and cooling rate were 10ºC/min.

2.2. SEM Observation and Luminescence Measurements

The obtained glass particles were observed by the scanning electron microscope (SEM) to know the particle shape (the sphericity and the contact angle of the truncated part) and the surface roughness.

The luminescence spectra were measured from the prepared glass particles and the mother glass by using a microscopic Raman spectrometer (Jasco, NRS-2100, Japan). The glass particle was attached on the end face of the optical telecommunication fiber (Figure 1). CW-Ar^+ laser beam with the wavelength 514.5nm was used as an excited source. The spectral resolution of the monochrometer was 1cm^{-1}. The incident laser light was collimated by the object lens into the beam spot of about 5μm. The laser power was changed from 2 to 25mW. The sizes of the examined particles were measured from the captured images by CCD camera equipped in Raman microscope.

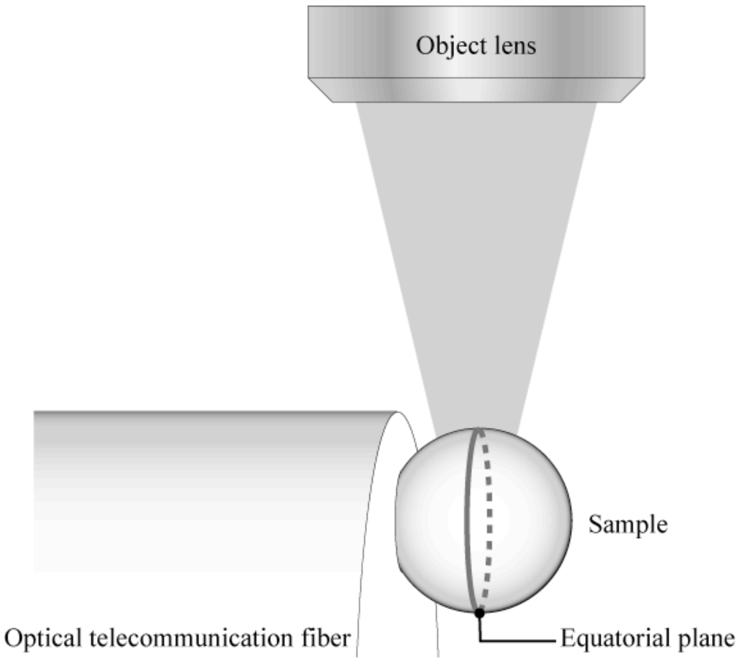

Fig.1. Schematic illustration of the luminescence measurement set up of the μ-SSG.

3. Results and Discussion

3.1 The appearance of Eu_2O_3-doped μ-SSG

Figure 2 shows SEM photographs of the prepared glass particles. All of the obtained glass particles with the diameter 1-40μm were found to have super-spherical shape (μ-SSG, Fig. 2(a)), and the particle surfaces were very smooth. The average contact angle of μ-SSGs is 134 ± 3º. The doping of Eu_2O_3 did not affect the contact angles of the truncated part compared with the non-doped particles reported in the previous study [12]. Figure 2(b) shows the top view of the particle. It is found that the equatorial circle is almost perfect.

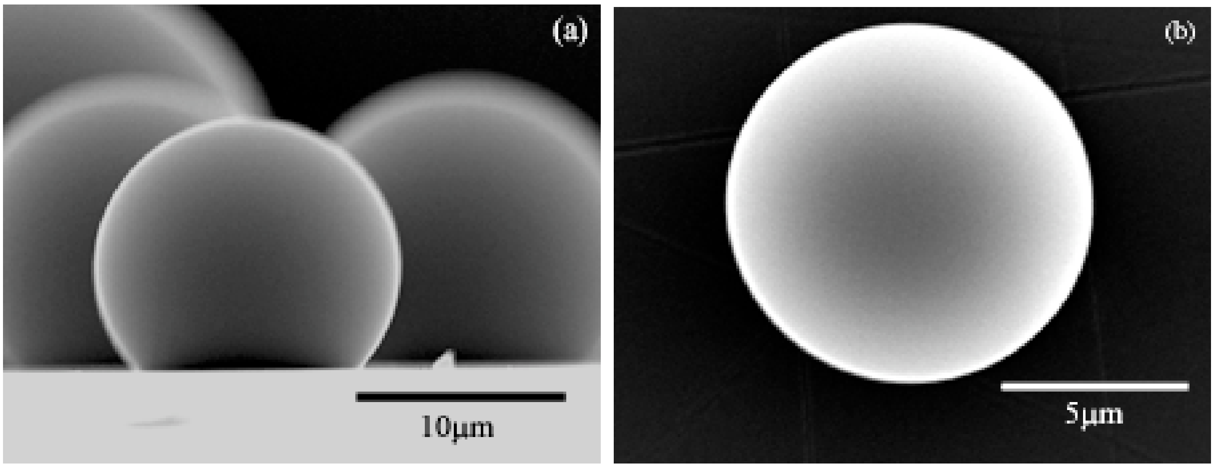

Fig.2. SEM photographs of the obtained glass particles. (a) the side view and (b) the top view.

3.2 Fluorescence Spectra Measurement

Figure 3 shows the luminescent spectra from the prepared μ-SSG with diameter of 6.8μm and the bulk glass. The spectrum of the bulk glass shows the characteristic spectral shape to the transition of $^5D_0 \rightarrow {}^7F_j$ (j=0,1,2) [13]. On the other hand, the spectra of the μ-SSG contain the regularly positioned sharp signals, and their intensities increased with the increasing laser power.

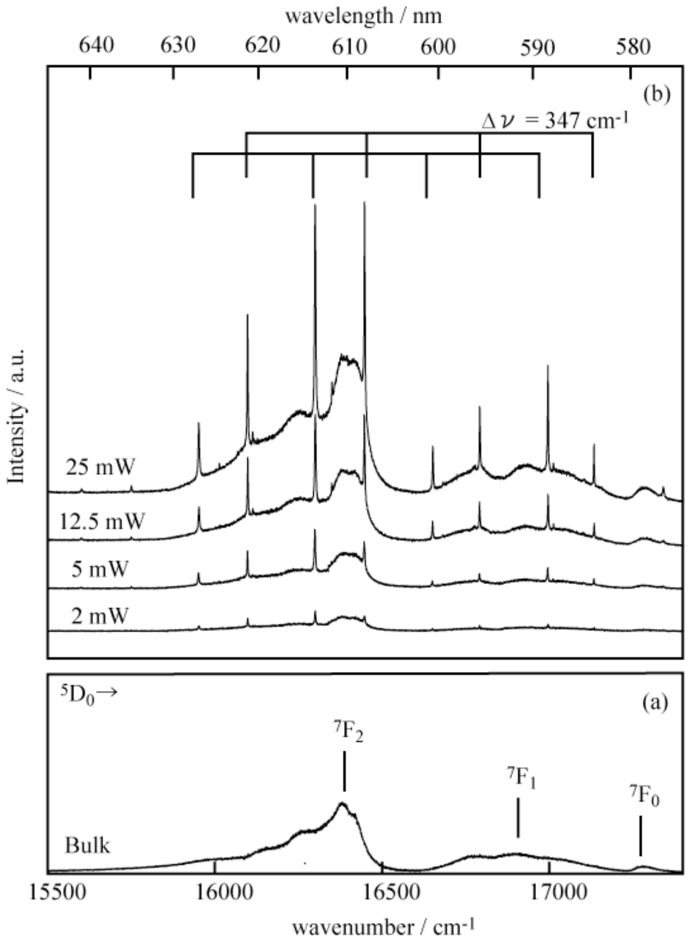

Fig. 3. Luminescence spectra of (a) the bulk glass and (b) the glass particle (6.8μm diameter) by microscopic Raman scattering spectroscopy. The excitation wavelength was 514.5nm, and laser power was 15mW for the bulk glass and 2, 5, 12.5, and 25mW for the glass particle.

According to the Lorenz-Mie theory [11], the interval energy, $\Delta\nu$ in cm^{-1}, between these sharp signals has the proximate relation of

$$\Delta\nu \approx \frac{1}{\pi a}\frac{\tan^{-1}\left(m^2-1\right)^{1/2}}{\left(m^2-1\right)^{1/2}}. \qquad (1)$$

If they are induced by the resonance modes in the sphere, and $\Delta\nu$ is called 'mode spacing'. Here, a is the diameter of the sphere, and m is the relative refractive index of the sphere. In the obtained spectra, 2 series of the resonance modes with $\Delta\nu$=347cm^{-1} are found and shown in Fig. 3. The refractive index m could be calculated as m=1.52 using the values of $\Delta\nu$ and a (=6.8μm), and corresponded well with n_D=1.515 of this glass. This means that the sharp signals are derived from the resonances of the luminescent light from Eu^{3+} in this μ-SSG, and this particle is ensured to work as the WGM oscillator. In Fig. 4, the intensities of this WGM signal against the power of the incident CW-Ar^{+} laser light are plotted. The linear relation can be found to pass near the origin. The spherical oscillator is known to have a low threshold for lasing action, but it can not be said that the observed signals are in lasing mode, because the excitation wavelength of 514.5nm is far from the appropriate excitation wavelength of $^5D_1$ level of Eu^{3+} (the absorption peak is located 533nm). However, the dearly observed oscillation signals even under the excitation of the absorption 'tail' of $^5D_1$ level represent high potentiality of μ-SSG for the micro cavity.

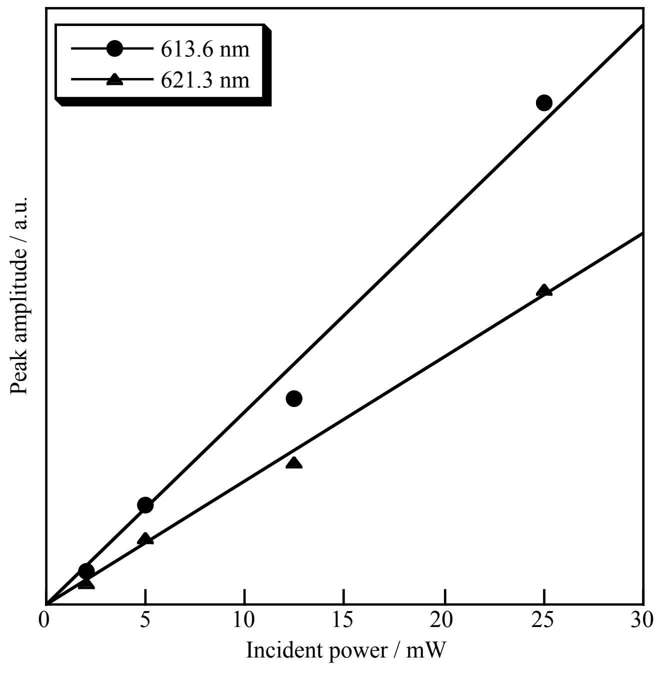

Fig.4. Dependence of the emission intensities of the WGM signals on the incident beam power. Lines were drawn by the least square method.

The quality factor, Q, is the representative to express the potential of the oscillation cavity, and can be estimated simply from the equation Q= $\lambda/\Delta\lambda$, where $\Delta\lambda$ is the half-width at the half maximum of the resonance signals. From the spectra, Q factor was calculated to 10^3-10^4. Taking into account the spectral resolution of the monochromator >1cm^{-1}, Q of the μ-SSG should be, at least lager than 10^3. Thus, the prepared μ-SSG is considered to have the equatorial plane with sufficiently perfect circle and work as an efficient oscillator media. The appropriate choice of the excitation wavelength is required to realize the lasing in this μ-SSG.

4. References

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