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Terrace-Microsphere Lasers:
Spherical Cavity Lasers for Multi-Wavelength Emission

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ABSTRACT

We have successfully made terrace-microspheres for laser emission: micrometer size spherical cavity laser having terrace shaped pumping light entrance. “Terrace-microsphere” is a high refractive index glass sphere (n_D=1.93) of 30μm in diameter with terrace portion of organic-inorganic materials. The glass sphere is in BaO-SiO$_2$-TiO$_2$ glass system and contain a few ppm of Nd^{3+}. Organic-inorganic hybrid materials of refractive index n_D=1.45 were prepared by sol-gel technique using 3-methacryloxypropyltrimethoxysilane and tetramethoxysilane as starting materials. To make terrace portion, a pico-liter of sol droplet was supplied with a micro-capillary into the boundary between a glass sphere and a Teflon sheet. The sol-derived part attached to a sphere showed the flat portion like a terrace structure. The terrace-microspheres were pumped with a tunable CW Ti:sapphire laser (λ=700nm-850nm) for choosing the suitable pumping wavelengths to WGMs. Pumping the terrace portion at around 800nm wavelength, strong resonances due to WGMs were demonstrated. The resonances originated from Raman scattering and Nd^{3+} fluorescence were observed at 840-880nm and 880-940nm wavelength region respectively. Consequently, we can show the potential application for a multi-wavelength laser (about 100 lines) at the extended wavelength range (840-940nm) in the near-infrared. Stimulated Raman emission of WGMs was performed with threshold of 4mW.

Keywords: terrace-microspheres, organic-inorganic hybrid materials, whispering gallery modes, glass microspheres, spherical cavity lasers, multi-wavelength emission, stimulated Raman scattering

1. INTRODUCTION

Microcavity-based Raman lasers are highly attractive for extending the available wavelength range of the existing laser source. Recently, spherical Raman lasers were performed with high-quality factor (high-*Q*) silica glass microspheres of several tens of micrometer in diameters by optical coupling with a fiber-taper[1]. To excite high-Q silica microspheres (refractive index n_D=1.458), we needed to couple pumping light into them effectively. Various techniques for achieving sufficient optical coupling have been tried[2] using prism couplers[3], side-polished fibers[4], fiber-tapers[5] and so on. The fiber-tapers of few-micrometer diameters are remarkable for optical coupling to microspheres, but they require to be drawn delicately from silica-based optical fibers. Silica glass spheres should be pumped with silica-glass fiber-taper, for

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matching their refractive index and modes between those of microspheres and those of fiber tapers. Moreover, the fiber-tapers are practically intolerable because of the lack of their mechanical strength. Thus, other efficient optical coupling techniques into microspheres are urgent issue for meeting the practical requirements. On the other hand, glass microspheres of multi-component glasses such as those made from conventional optical glasses are also attractive because of wide variety of compositions along with wide range of their optical and physical properties. Optical coupling using silica-glass based fiber-taper in not suitable for pumping those multi-component glass spheres, because their refractive-index difference is not affordable for their mode matching.

In our previous works, micrometer-size spherical cavity laser having a terrace-shaped pumping-light entrance was reported[6-8]. On high-index glass microspheres (n_D=1.93) of several tens of micrometers in diameter, terrace-shaped light entrance was formed with commercially available silicone oligomer (n_D=1.49). We called it the "terrace-microsphere". Glass spheres of suitable sizes for pumping were chosen and stimulated Raman emission of whispering gallery modes (WGMs) was demonstrated by pumping the spot at terrace portion with CW Ar+ laser (514.5nm wavelength).

In this paper, a terrace was fabricated on high-index glass spheres using a micro-capillary tube as a supplier of a picoliter organic-inorganic hybrid sol, and pumping experiments were carried out using a tunable CW Ti:sapphire laser (λ=700nm-850nm) for choosing the suitable pumping wavelengths to WGMs of a terrace-microspheres. High-index glass spheres containing a few ppm Nd^{3+} were used in pumping experiments to investigate the double effects of Raman scattering due to high-index glass matrix and fluorescence due to Nd^{3+} simultaneously.

2. EXPERIMENTS

2.1. Fabrication of the terrace-microspheres

Commercially available high refractive index (n_D=1.93) glass microspheres of 30μm in diameter (Union Co.) were used for the fabrication of terrace-microspheres. Typical composition of the glass sphere is $38.0BaO\text{-}11.5SiO_2\text{-}38.5TiO_2\text{-}6.7CaO\text{-}5.3ZnO$ in the $BaO\text{-}SiO_2\text{-}TiO_2$ glass system containing a few ppm of Nd^{3+} [9]. They were made by the flame spray technique, where small pieces of glass cullet were melted in the flame and glass spheres of smooth surface were formed by surface tension. Organic-inorganic hybrid materials were prepared by sol-gel technique using 3-methacryloxypropyl-trimethoxysilane (MOPS) and tetramethoxysilane (TMOS) as starting materials. MOPS and TMOS are chosen for controlling the sol properties such as hydrophobic-hydrophilic properties, viscosity, polymerization rate and so on. The 25MOPS-75TMOS sol in the MOPS-TMOS binary system were hydrolyzed and polymerized in hydrochloric acid solution. A micro-capillary supplier controlled with a micro manipulator was used for the terrace formation as illustrated in Fig. 1. The glass micro-capillary was made by using capillary-puller equipment and the tip diameter was about 1μm. A picoliter of MOPS-TMOS sol was supplied into extremely narrow space between a sphere and Teflon sheet caused by capillary force. A glass sphere sank slightly into sol, and then the sol was rapidly solidified. After gelling terrace-microspheres were dried at room temperature, and subsequently heated at 100°C for 60min.

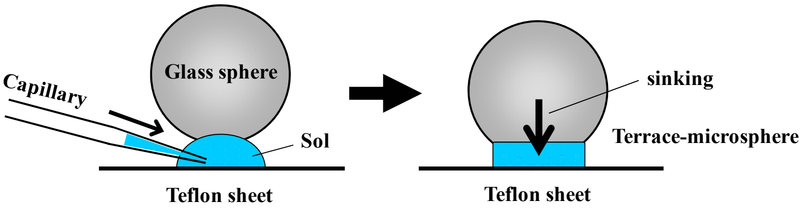


Fig. 1. Schematic illustration of a micro-capillary technique for fabricating terrace-microspheres.

2.2. Pumping of the terrace-microspheres

As a preliminary measurement, emission spectra of the terrace-microspheres were measured by a micro-Raman scattering spectrometer (JASCO, Model NRS-2100) for reconfirmation of performance at visible range and suitable irradiation point. Pumping wavelength was 514.5nm (CW Ar^+ laser), with spectral resolution of $1cm^{-1}$. The incident laser beam was collimated to about 1μm-diameter spot.

Emission spectra of the terrace-microspheres were measured by experimental set-up, as illustrated in Fig. 2. A tunable CW Ti:sapphire laser (λ=700nm~850nm) was used for choosing the suitable pumping wavelengths to WGMs of the microspheres. The incident laser beam was band-passed (750-820nm) and collimated to about 2μm-diameter spot by an objective lens (×100, NA0.8). Terrace-microspheres were fixed on the edge of a slide glass by an electrostatic force. Emission below 835nm wavelength was removed by a long-pass filter. The emission from a terrace-microsphere was sent to a monochromator (JASCO, CT-25C) through an optical fiber and analyzed by a CCD detector (ANDOR, iDus DU401A). The pumping wavelengths were checked by a monochromator (OceanOptics, USB4000).

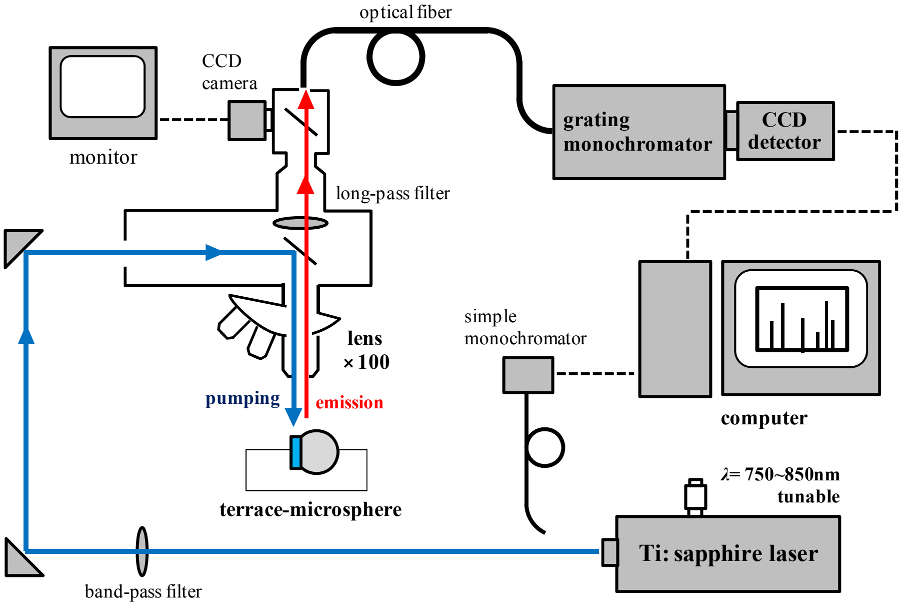


Fig. 2. Schematic illustration of setup for pumping terrace-microspheres.

3. RESULTS

A typical SEM image of terrace-microspheres is shown in Fig. 3 (a). Surface of the glass spheres are smooth and they have well-looking terrace shapes. The terrace portion was fabricated by the micro-capillary technique. The size of terrace-structure is illustrated in Fig. 3 (b). Refractive index of the 25MOPS-75TMOS hybrid material was n_D=1.45 after curing.

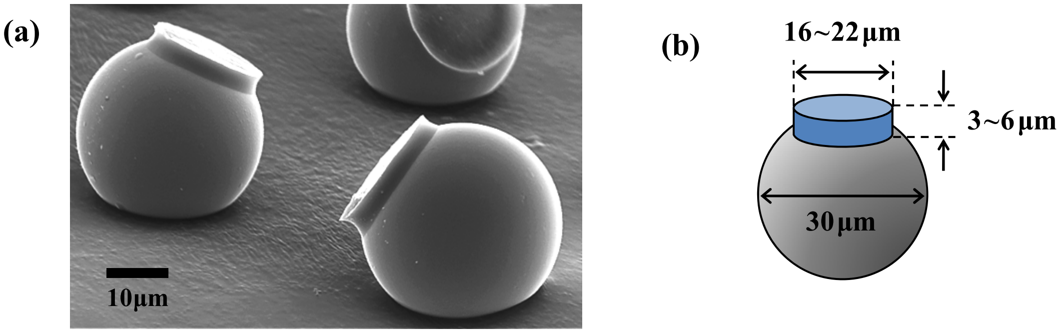


Fig. 3. (a) SEM image of terrace-microspheres. (b) Schematic illustration of a terrace-microsphere.

Figure 4 shows the results of the preliminary measurement at visible range pumping. Resonant Raman spectra from terrace-microsphere with various irradiation points are shown in Fig. 4 (a). The corresponding laser pumping spots (from A to E) are illustrated in the inset figure. The pumping laser intensity was 10mW. Strong resonant peaks due to WGMs were observed, especially, when the terrace portion (the spot B) was irradiated. In Fig. 4 (b), emission intensities of a terrace-microsphere are plotted against the pumping power of a CW Ar$^+$ laser. At pumping power of about 4mW the slope of emission intensity vs. pumping power plot increased clearly. Extrapolation of the line showed the threshold of 2.5mW.

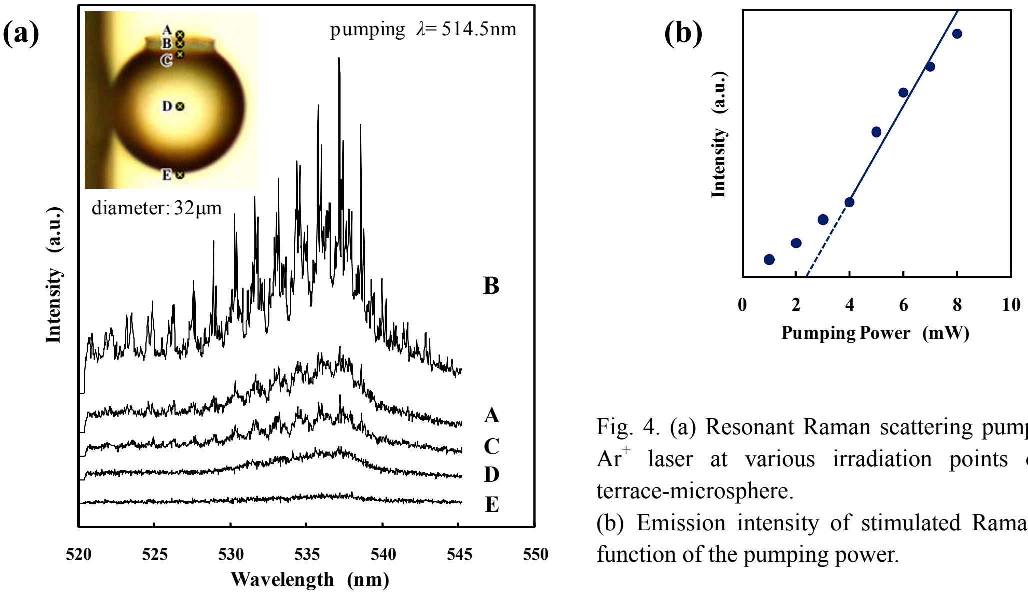
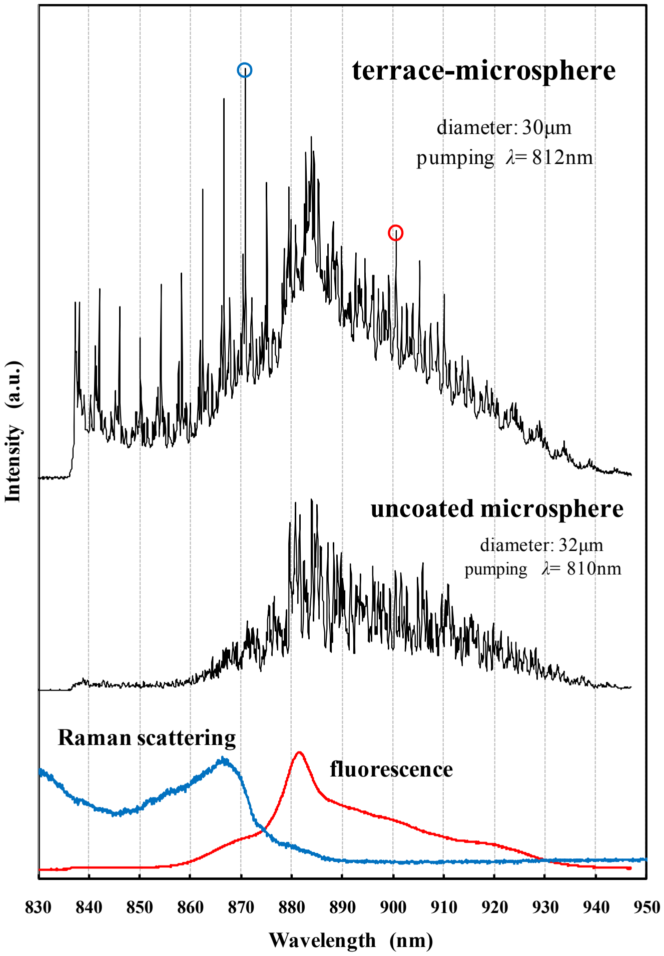


Fig. 4. (a) Resonant Raman scattering pumped by Ar$^+$ laser at various irradiation points of the terrace-microsphere.
(b) Emission intensity of stimulated Raman as a function of the pumping power.

Emission spectra of terrace-microspheres pumped by a tunable CW Ti:sapphire laser are shown in Fig. 5. The upper emission spectrum is that from a terrace-microsphere. Pumping power was 15mW. A terrace-microsphere performed strong resonant peaks due to WGMs in the 835~880nm wavelength range and in the 880~940nm range. Meanwhile, in the pumping an uncoated microsphere, the resonant peaks appeared only in the 860-940nm range. The bottommost spectra are Raman scattering and fluorescence of the BaO-SiO_2-TiO_2 glass containing a few ppm of Nd^{3+} pumped by 810nm wavelength laser light. The broad band below 880nm is Raman scattering derived from BaO-SiO_2-TiO_2 glass, and the band at 860-940nm is a fluorescence originated from Nd^{3+}. In Fig. 6, emission intensities of a terrace-microsphere are plotted against pumping power. Blue and red lines in Fig. 6 correspond to blue and red circles in the upper spectrum in Fig. 5: the resonant peaks due to Raman scattering (λ=870.9nm) and fluorescence (λ=901.1nm). In the Raman scattering plot, the slope of emission intensity vs. pumping power line increased clearly above 4mW. Thus the threshold was 4mW. On the other hand, the slope of fluorescence line changed at about 4mW. The quality factor (Q) of the terrace-microsphere estimated from a FWHM of the spectrum is about 1×10^4, which is limited by the measurement system.



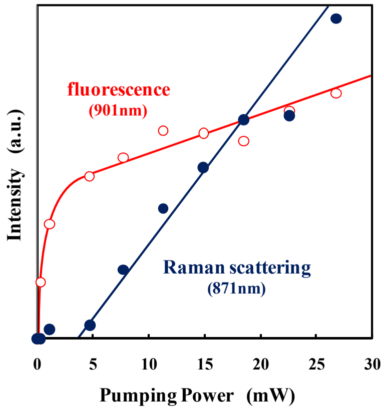


Fig. 6. Emission intensity of a terrace-microsphere as a function of the pumping power. The resonant peaks derived from Raman (870.9nm) and fluorescence (901.1nm) correspond to the plot, marked with blue circle and red one in the top spectrum of Fig. 5, respectively.

Fig. 5. Resonant spectra of a terrace-microsphere and a uncoated microsphere pumped by a Ti:sapphire laser. The bottommost spectra are Raman scattering and fluorescent emitted by the glass sphere at 810.0nm wavelength pumping.

4. DISCUSSION

High-index BaO-SiO_2-TiO_2 glass spheres containing a few ppm of Nd^{3+} were used in the pumping experiments. We planed to investigate the double effects of Raman scattering due to high-index glass matrix and fluorescence due to Nd^{3+} simultaneously. Spontaneous Raman scattering spectra of a BaO-SiO_2-TiO_2 glass and a silica glass were plotted in Fig. 7. Scattering peaks at 300cm^{-1} and 800cm^{-1} were originated from bonds of Ti-O and Si-O in BaO-SiO_2-TiO_2 glass matrix[10, 11]. Since the BaO-SiO_2-TiO_2 high-index glass showed strong Raman scattering in wide range of wavelengths compared with that of a silica glass, a multi-component glass is one of the most favorable candidates for multi-wavelength Raman lasers. As shown in Fig. 4 and 5, the Raman shift at 600-900cm^{-1} band corresponded to laser emissions at 520-545nm wavelengths (Ar^+ laser) and those at 840~890nm wavelengths (Ti:sapphire laser).

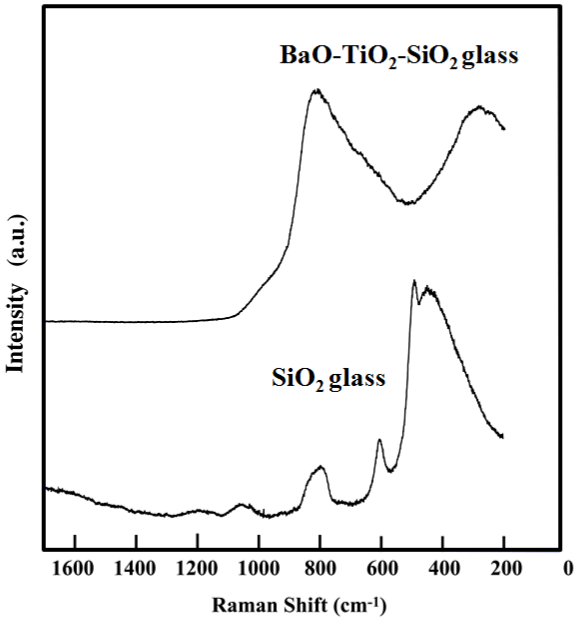


Fig. 7. Spontaneous Raman scattering spectra of BaO-SiO_2-TiO_2 and SiO_2 glasses.

The BaO-SiO_2-TiO_2 glass microspheres also contain a few ppm of Nd^{3+}, which is a well-known active ion for laser emission[12, 13]. An energy level diagram of Nd^{3+} and its fluorescent spectrum in a soda-lime silica glass is shown in Fig. 8 (a) and (b)[14], respectively. Pumping by laser light at around 810nm wavelength (the transition $^4I_{9/2}$ - $^4F_{5/2}$) gave three principal fluorescent bands (wavelengths: 880nm, 1060nm and 1325nm). Transition $^4I_{3/2}$ - $^4I_{9/2}$ gave fluorescence in the 850-940nm wavelengths, which were overlapped with Raman scattering in the 860-890nm wavelength region (see the bottommost blue and red spectra in Fig. 5). It can be seen from the spectrum of uncoated glass sphere in Fig. 5, under the condition of weak optical coupling, only the resonant peaks due to Nd^{3+} fluorescence was induced and there was no resonant Raman peaks in the 840-890nm wavelength region. In terrace-microsphere, however, above 4mW pumping power, stimulated Raman emission was observed. As can be seen in Fig. 6, the change point of the slope in fluorescence-pumping power line seemed to correlate with the threshold of stimulated Raman scattering. Additionally, above 4mW pumping power, the laser emission lines at around 1060nm wavelength (transition ${}^4F_{3/2}$ - ${}^4I_{11/2}$ originated from Nd^{3+}) were also observed.

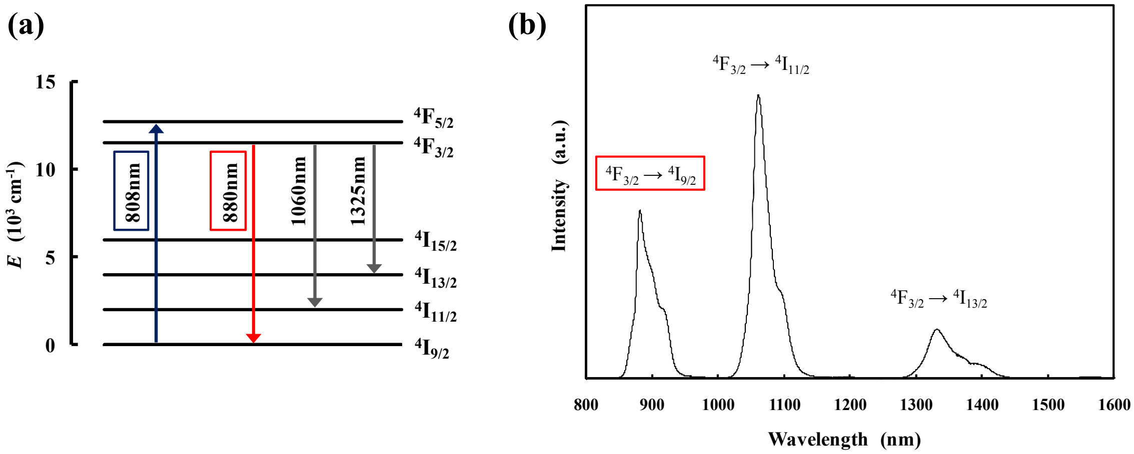


Fig. 8. (a) Energy level diagram of Nd^{3+} ion. Arrow lines show laser transition processes. (b) Fluorescent spectrum of Nd-doped soda-lime silica glass (excitation wavelength is 810nm).

The mode spacing in the spectrum of uncoated microsphere is 4.7nm at around 880nm wavelength (see Fig. 5), which is similar to WGM mode spacing theoretically estimated from sphere radius r, refractive index of glass n_1, index of surrounding medium n_2. The mode spacing of spherical particle is given by[15]

$$\Delta\lambda = \frac{\lambda^2}{2\pi r n_2} \cdot \frac{\tan^{-1}\sqrt{(n_1/n_2)^2 - 1}}{\sqrt{(n_1/n_2)^2 - 1}} \qquad (1)$$

Thus the comb-shaped peaks of uncoated glass sphere in Fig. 5 are attributed to the resonant peaks due to WGMs. Typical mode spacing of terrace-microsphere in Fig. 5, however, is 4.3nm at around 880nm-wavelength (4.6nm at around 900nm-wavelength), which is smaller than theoretically estimated values 5.1nm at around 880nm-wavelength (5.4nm at around 900nm) from r and n_1 of the glass sphere. Remarkable difference in mode spacing of the spectra of a terrace-microsphere was also observed in Fig. 4, in the stimulated Raman spectrum of terrace-microsphere pumped by Ar^+ laser: 1.4nm as experimental mode-spacing, and 1.8nm as theoretical estimated spacing. If the low-index part was formed on the boundary between high-index sphere and surrounding air, the relative index difference decreases. Thus, the decrease of index difference will increase the mode spacing: $\Delta\lambda$ (experiment) > $\Delta\lambda$ (theoretical calculation). The result ($\Delta\lambda$ (experiment) < $\Delta\lambda$ (theoretical calculation)) did not agree with the assumption. Therefore, unfortunately the reason for the difference is still not clear. It should be emphasized, however, that WGM resonant modes were influenced dramatically by forming a terrace-structure onto the sphere surface.

In conclusion, the terrace-microspheres were pumped with a tunable CW Ti:sapphire laser. Pumping the terrace portion at around 800nm wavelength, strong resonances due to WGMs were demonstrated. The stimulated emission originated from Raman scattering with threshold of 4mW and resonant emission due to Nd^{3+} were observed in the 840-880nm and 880-940nm wavelength region, respectively. We can show the potential use of glass spheres for a multi-wavelength laser in the extended wavelength region.

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