Quasi-single mode laser output from a terrace structure added on a Nd3+-doped tellurite-glass microsphere prepared using localized laser heating

(Article begins on next page)

Quasi-single mode laser output from a terrace structure added on a Nd^{3+}-doped tellurite-glass microsphere prepared using localized laser heating

Tetsuo Kishi,[1,*] Tsutaru Kumagai,[1] Shogo Shibuya,[1] Francesco Prudenzano,[2] Tetsuji Yano,[1] and Shuichi Shibata[1]

[1]*Department of Chemistry and Materials Science, Tokyo Institute of Technology, 2-12-1 Ookayama Meguro-ku Tokyo, 152-8550, Japan*

[2]*Politecnico di Bari, DEI - Dipartimento di Ingegneria Elettrica e dell'Informazione, Via E. Orabona 4, Bari, 70125, Italy*

**tkishi@ceram.titech.ac.jp*

Abstract: A Nd^{3+}-doped tellurite-glass terrace microsphere was fabricated, and its laser characteristics using free-space pumping were investigated. A localized laser heating technique was used for preparing the 29-µm-diameter microsphere. The uncoated sphere exhibited many laser lines with 1.3-mW threshold. Fewer laser lines were observed after terrace formation. The terrace microsphere's lasing threshold was 0.6–2.4 mW depending on the pumping position in the terrace. These results indicate that the terrace structure can modify the modes of a microsphere laser and decrease the laser threshold due to an increase in the coupling efficiency between the cavity and free-space beam.

OCIS codes: (140.3530) Lasers, neodymium; (140.4780) Optical resonators; (160.2750) Glass and other amorphous materials.

References and links

1. M. Kuwata-Gonokami and K. Takeda, "Polymer whispering gallery mode lasers," Opt. Mater. **9**(1-4), 12–17 (1998).
2. S. Shibata, T. Yano, and H. Segawa, "Sol-gel-derived spheres for spherical microcavity," Acc. Chem. Res. **40**(9), 913–920 (2007).
3. C. G. B. Garrett, W. Kaiser, and W. L. Bond, "Stimulated emission into optical whispering modes of spheres," Phys. Rev. **124**(6), 1807–1809 (1961).
4. I. S. Grudinin and L. Maleki, "Ultralow-threshold Raman lasing with CaF_2 resonators," Opt. Lett. **32**(2), 166–168 (2007).
5. V. Sandoghdar, F. Treussart, J. Hare, V. Lefèvre-Seguin, J. Raimond, and S. Haroche, "Very low threshold whispering-gallery-mode microsphere laser," Phys. Rev. A **54**(3), R1777–R1780 (1996).
6. K. Miura, K. Tanaka, and K. Hirao, "Laser oscillation of a Nd^{3+}-doped fluoride glass microsphere," J. Mater. Sci. Lett. **15**(21), 1854–1857 (1996).
7. K. Sasagawa, K. Kusawake, J. Ohta, and M. Nunoshita, "Nd-doped tellurite glass microsphere laser," Proc. SPIE **4969**, 255–262 (2003).
8. M. Cai, O. Painter, K. J. Vahala, and P. C. Sercel, "Fiber-coupled microsphere laser," Opt. Lett. **25**(19), 1430–1432 (2000).
9. G. R. Elliott, G. S. Murugan, J. S. Wilkinson, M. N. Zervas, and D. W. Hewak, "Chalcogenide glass microsphere laser," Opt. Express **18**(25), 26720–26727 (2010).
10. G. S. Murugan, M. N. Zervas, Y. Panitchob, and J. S. Wilkinson, "Integrated Nd-doped borosilicate glass microsphere laser," Opt. Lett. **36**(1), 73–75 (2011).
11. L. Collot, V. Lefèvre-Seguin, M. Brune, J. M. Raimond, and S. Haroche, "Very high-Q whispering-gallery mode resonances observed on fused silica microspheres," Europhys. Lett. **23**(5), 327–334 (1993).
12. S. M. Spillane, T. J. Kippenberg, and K. J. Vahala, "Ultralow-threshold Raman laser using a spherical dielectric microcavity," Nature **415**(6872), 621–623 (2002).
13. S. Shibata, S. Ashida, H. Segawa, and T. Yano, "Coated microsphere as spherical cavity Raman laser," J. Sol-Gel Sci. Technol. **40**(2-3), 379–384 (2006).

#238546 Received 13 May 2015; revised 22 Jul 2015; accepted 26 Jul 2015; published 29 Jul 2015

14. H. Uehara, T. Yano, and S. Shibata, "Terrace formation with a picoliter sol–gel droplet for spherical cavity Raman laser," J. Sol-Gel Sci. Technol. **58**(1), 319–325 (2011).
15. M. Weber, "Science and technology of laser glass," J. Non-Cryst. Solids **123**(1-3), 208–222 (1990).
16. J. S. Wang, E. M. Vogel, and E. Snitzer, "Tellurite glass: a new candidate for fiber devices," Opt. Mater. **3**(3), 187–203 (1994).
17. J. Wu, S. Jiang, T. Qua, M. Kuwata-Gonokami, and N. Peyghambarian, "2 μm lasing from highly thulium doped tellurite glass microsphere," Appl. Phys. Lett. **87**(21), 211118 (2005).
18. Y. Ruan, K. Boyd, H. Ji, A. Francois, H. Ebendorff-Heidepriem, J. Munch, and T. M. Monro, "Tellurite microspheres for nanoparticle sensing and novel light sources," Opt. Express **22**(10), 11995–12006 (2014).
19. T. Kishi, T. Kumagai, T. Yano, and S. Shibata, "On-chip fabrication of air-bubble-containing Nd3+-doped tellurite glass microsphere for laser emission," AIP Adv. **2**(4), 042169 (2012).
20. T. Kumagai, T. Kishi, and T. Yano, "Low threshold lasing of bubble containing glass microspheres by non-whispering gallery mode excitation over a wide wavelength range," J. Appl. Phys. **117**(11), 113104 (2015).
21. P. R. Conwell, P. W. Barber, and C. K. Rushforth, "Resonant spectra of dielectric spheres," J. Opt. Soc. Am. A **1**(1), 62–67 (1984).
22. Y. P. Rakovich and J. F. Donegan, "Photonic atoms and molecules," Laser Photonics Rev. **4**(2), 179–191 (2010).
23. V. Datsyuk, "Some characteristics of resonant electromagnetic modes in a dielectric sphere," Appl. Phys. B **54**(2), 184–187 (1992).
24. R. Chen, H. D. Sun, "Single mode lasing from hybrid hemispherical microresonators," Sci. Rep. **2**, 244 (2012).
25. X. Peng, F. Song, S. B. Jiang, N. Peyghambarian, M. Kuwata-Gonokami, and L. Xu, "Fiber-taper-coupled L-band Er^{3+}-doped tellurite glass microsphere laser," Appl. Phys. Lett. **82**(10), 1497–1499 (2003).
26. Z. P. Cai, H. Y. Xu, G. M. Stéphan, P. Féron, and M. Mortier, "Red-shift in Er:ZBLALiP whispering gallery mode laser," Opt. Commun. **229**(1-6), 311–315 (2004).

1. Introduction

Micrometer-sized spherical particles made from optically transparent materials can confine light through multiple total internal reflections at the surface and lead to whispering-gallery-mode (WGM) resonances with a high quality (Q) factor and small mode volume (V). Therefore, such particles can be employed to realize nonlinear optical effects at a low incident power and lasers with very low thresholds. Laser oscillation has been demonstrated using various materials such as dye-doped spheres [1], organic–inorganic hybrid materials [2], crystals [3,4], and glasses [5–14]. Spheres made from glass materials offer some of the highest Q factors because of their transparency over a wide wavelength range, with reports of silica microspheres having Q factors as high as 10^9 [11]. Rare-earth ion-doped glass microsphere lasers have been reported using silica [5], fluoride [6], tellurite [7], phosphate [8], chalcogenide [9], and silicate [11] glasses. Furthermore, Raman lasers, which require very high pumping intensities, based on microspheres made from silica with tapered-fiber coupling [12] and $BaO–SiO_2–TiO_2$ glass terrace-microsphere with free-space coupling [13] have been reported

Coupling technology is, in fact, indispensable for pumping and reading high-Q modes in microspheres. Previous studies have demonstrated the use of various optical coupling technologies such as prisms [5,7,11], side-polished fibers [4,9], and fiber tapers [8,12] for achieving low-threshold lasing due to the excitation of high-Q modes. Generally, free-space coupling, which is robust and requires less rigorous experimental conditions than the evanescent couplers mentioned above, is inefficient because of a large mismatch between the excitation beam and WGMs and lacks high spatial mode selectivity [12]. However, we previously demonstrated Raman lasing in a high refractive index microsphere via free-space pumping through a terrace structure with a lower refractive index [13,14]. The terrace microsphere is a successful example of the highly effective excitation of a high refractive index microsphere laser and will enable a wide variety of glasses to be utilized for micrometer-sized optical resonators.

Tellurite glass doped with rare-earth ions has great potential for use as a laser material because of its lower phonon energy and higher stimulated-emission cross section [7,15,16]. The chemical durability of tellurite glass is also much better than that of fluoride glass. In addition, because the linear and nonlinear refractive indices of tellurite glass are high, this glass has considerable promise as a resonator matrix for a range of optical devices. The most

#238546 Received 13 May 2015; revised 22 Jul 2015; accepted 26 Jul 2015; published 29 Jul 2015

common technique for fabrication of tellurite glass microspheres is pouring molten glass onto a spinning plate in a furnace [17], or heating glass fiber tapers using a resistive heater [7] or a CO_2 laser [18]. Previously, we fabricated bubble-containing and bubble-free Nd^{3+}-doped tellurite glass microspheres on a substrate using a localized laser heating technique [19,20]. The microspheres exhibited laser emissions corresponding to the optical transition from $^4F_{3/2}$ to $^4I_{11/2}$ at a wavelength of approximately 1,060 nm via free-space pumping at a wavelength of approximately 800 nm.

Herein, we describe the fabrication of Nd^{3+}-doped tellurite-glass terrace microspheres and their laser characteristics using free-space coupling. A localized laser heating technique was employed to obtain the Nd^{3+}-doped tellurite glass microsphere [19,20], and a terrace portion was formed on the microsphere using a microcapillary tube technique [14]. Laser emission spectra for the tellurite microsphere before and after terrace formation were obtained to determine the effects of terracing on laser characteristics. The dependence of the lasing threshold and wavelength on the pumping position within the terrace portion was also investigated.

2. Experimental

Glass with the composition $10K_2O$–$10WO_3$–$80TeO_2$ (in mol%) doped with 1.0 mol% Nd_2O_3 was prepared using a conventional melt-quenching method. The refractive index of the glass at 633 nm was 2.05. A localized laser heating technique was used for microsphere production. Crushed glass particles were placed on a silica substrate and irradiated using a high-power continuous-wave (CW) Ti:sapphire laser. The crushed glass melted and formed into a sphere during irradiation with a sufficient power density. The sphere was quenched when laser irradiation was halted. This microsphere fabrication process is described in detail in a previous study [20]. The laser actions of microspheres prepared on silica substrates were confirmed by excitation measurements using free-space coupling.

Using a microcapillary method, terrace portions were then formed on microspheres that exhibited laser oscillations [14]. An organic–inorganic hybrid material (refractive index n_D: 1.45) was prepared via a sol–gel technique using 3-methacryloxypropyl-trimethoxysilane (MOPS) and tetramethoxysilane (TMOS) as starting materials. The 25MOPS–75TMOS sol was hydrolyzed and polymerized in hydrochloric acid solution (pH = 2) at room temperature. A small amount of the MOPS–TMOS sol was then supplied into the space between the microsphere and a Teflon® sheet using a glass capillary with a 1-µm-diameter tip. After gelation, the sample was dried at room temperature and subsequently heated at 100 °C for 60 min to remove remaining water or alcohol.

Figure 1 shows a schematic of the experimental setup for obtaining the emission spectrum from a terrace microsphere. The terrace microsphere was pumped using a tunable CW Ti:sapphire laser (wavelength: 800–810 nm). An incident beam was focused through an objective lens (100x, numerical aperture: 0.8) on a 2-µm-diameter spot. The terrace microsphere was fixed on the edge of a silica glass using the sol. The emission from the entire terrace portion of the microsphere was collected via the objective lens and sent to a monochromator (JASCO, CT-25C) through a multimode optical fiber and detected using a charge-coupled device (ANDOR, iDus DU401A). A long-pass filter was inserted prior to the monochromator for removing emissions with wavelengths <835 nm. The spectral resolution of the monochromator was 0.1 nm.

#238546 Received 13 May 2015; revised 22 Jul 2015; accepted 26 Jul 2015; published 29 Jul 2015

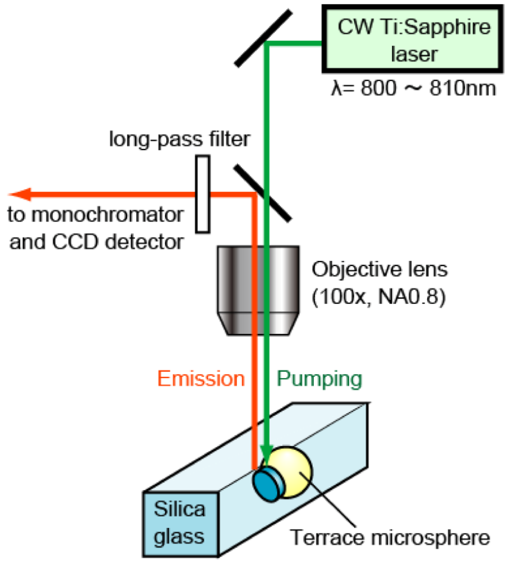

Fig. 1. Schematic of the experimental apparatus used to pump the terrace microsphere and obtain its emission spectrum.

3. Results and discussion

The tellurite glass microsphere used in this study is shown in Fig. 2(a). The dimensions of the terrace microsphere are illustrated in Fig. 2(b). The microsphere was 29 μm in diameter, and the terrace portion was 19 μm in diameter and 4 μm in height.

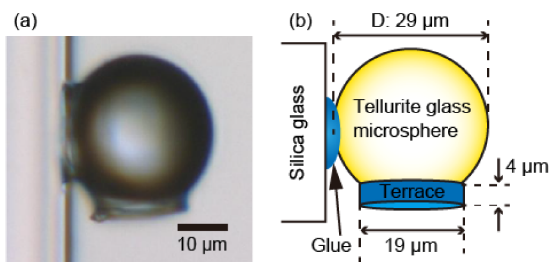

Fig. 2. (a) Optical photograph of a terrace microsphere attached to a silica glass substrate. (b) Schematic of the terrace microsphere.

Emission spectra from a microsphere before and after terrace formation for various pumping intensities are shown in Fig. 3. The pumping positions shown in Figs. 3(a) and 3(b) were the edge of the uncoated sphere and that of the terrace portion, respectively. In addition, the pumping wavelength was tuned to obtain the highest emission intensity: 803.5 nm for Fig. 3(a) and 805.3 nm for Fig. 3(b). The emission spectra shown in Figs. 3(a) and 3(b) include several sharp peaks and many broad weak peaks. Regardless of the pumping positions, the spectral shapes were nearly identical for the uncoated and terrace microspheres. The sharp peaks were attributed to laser emission because they had obvious thresholds as the excitation power increased (see Fig. 4). The Q values estimated from the full width at half maximum of the peaks below the thresholds were approximately 5×10^3 for the terrace microsphere and approximately 1×10^4 for the uncoated sphere, whose Q value was limited by the measurement system. Furthermore, as the excitation power increased, the number of laser lines increased, and the peak wavelength shifted to longer wavelengths. The uncoated microsphere laser became multimodal just above the threshold. Notably, the number of laser

#238546 Received 13 May 2015; revised 22 Jul 2015; accepted 26 Jul 2015; published 29 Jul 2015

emission peaks was reduced after terrace formation, and the terrace microsphere laser achieved single mode emission over a wide excitation power range. This result implies that mode selection with free-space coupling may be achieved using a terrace structure. Figure 3 also shows spectra based on Q_{sca} values calculated using Mie theory [21,22] for a 28.82-μm microsphere with a refractive index of 2.025. The peaks in the spectrum of the uncoated microsphere agree well with the theoretically estimated values for a WGM resonator. The laser emission peaks in the uncoated sphere are due to WGMs with mode orders l of 1, 2, and 3. However, the laser emission line pattern for the terrace microsphere did not match WGMs of the uncoated sphere. This result indicates that modified WGMs existed in the terrace microsphere. The difference in refractive index between the tellurite glass (2.05) and the hybrid polymer (1.45) optically perturbs WGMs in the sphere. The relative refractive index decreases to 1.41 at the terrace portion. Lower relative refractive index increases radiation loss of the WGMs. The radiation loss of WGM with higher l is larger than that with lower l [23]. It is possible that multimodal emission in the terrace sphere was suppressed because the decrease in the Q values of the higher-order modes due to the increase of loss was greater than that for l = 1, which has the highest Q value in the same mode number n. Therefore, the modes in the terrace structure may have lower Q values when compared with the high-Q WGMs. Such suppression of sidebands using a robust structure with free-space coupling is attractive for laser light sources required for practical applications in laser spectroscopy and metrology [24].

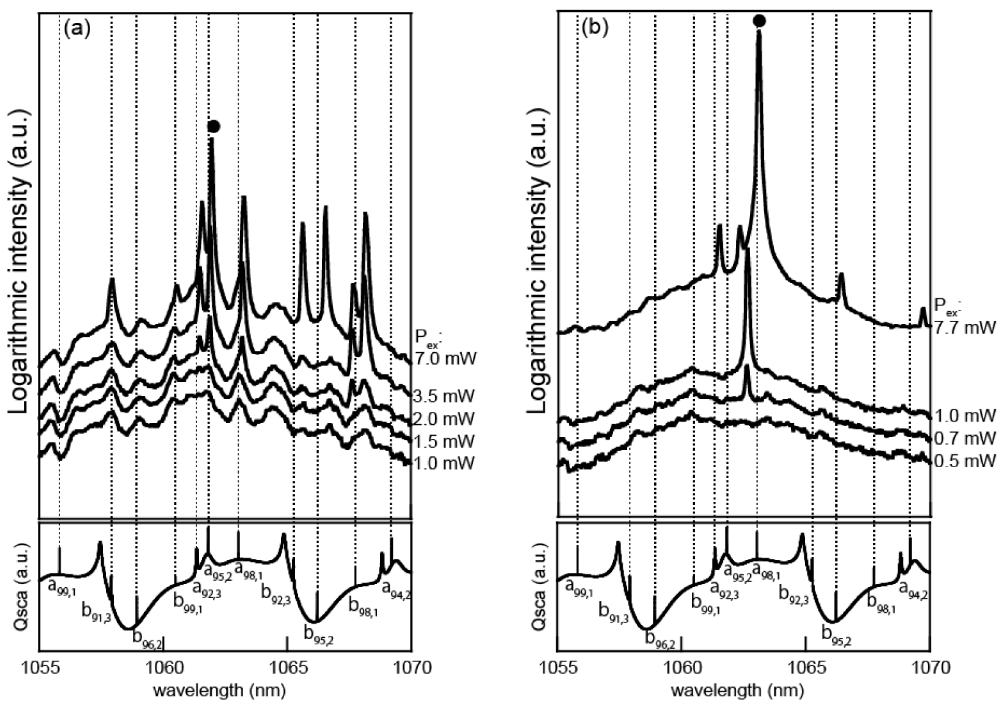

Fig. 3. Emission spectra from (a) uncoated and (b) terrace microspheres at various excitation powers. Pumping positions for (a) and (b) were the edge of the sphere and that of the terrace, respectively, and the pumping wavelengths were 803.5 and 805.3 nm, respectively. The power of the CW Ti:sapphire laser irradiated on each sample is represented by P_{ex}. The bottom spectra are based on Q_{sca} values calculated using Mie theory for a 28.82-μm microsphere. The dotted lines indicate the resonance wavelengths of WGMs. The parameters $a_{n,l}$ and $b_{n,l}$ are the transverse magnetic and electric modes, respectively. The subscripts n and l indicate the mode number and order of each resonance, respectively.

#238546 Received 13 May 2015; revised 22 Jul 2015; accepted 26 Jul 2015; published 29 Jul 2015

Figure 4 shows the emission intensities of the peaks marked with closed circles in Fig. 3 as a function of the excitation power. The pumping positions were the edge of the sphere for Fig. 4(a), center of the terrace for Fig. 4(b), and edge of the terrace for Fig. 4(c). The least-squares fitting of the data revealed that the threshold excitation powers for Figs. 4(a), 4(b), and 4(c) were 1.3, 1.6, and 0.6 mW, respectively. When the edge of the terrace was pumped, the lasing threshold of the terrace microsphere was lower than that of the uncoated sphere, indicating that low-threshold lasing can be achieved by selecting the optimum pumping position for the terrace portion.

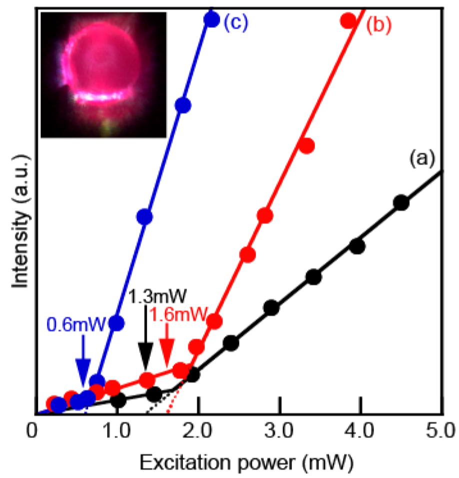

Fig. 4. Plots of emission intensities of (a) uncoated and (b, c) terrace microspheres versus the excitation power of the Ti:sapphire laser. Pumping positions are the edge of the sphere, center of the terrace, and edge of the terrace for (a), (b), and (c), respectively. Solid lines indicate the least-squares fitting results, and the lasing threshold is given by the intersection of curves with the horizontal axis. The inset photograph is the terrace microsphere pumped above the laser threshold.

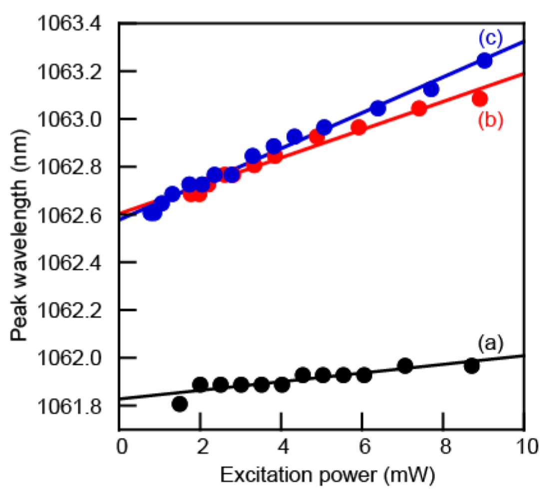

Fig. 5. Peak wavelengths versus excitation power for (a) uncoated and (b, c) terrace microspheres. Solid lines represent the least-squares fitting results. Pumping positions were the (a) edge of the sphere, (b) center of the terrace, and (c) edge of the terrace.

The wavelengths of the peaks marked with closed circles in Fig. 3 are plotted as a function of the excitation power in Fig. 5. As the excitation power increased, the peak wavelengths linearly shifted to longer wavelengths. Similar red shifts were observed for other laser lines

#238546 Received 13 May 2015; revised 22 Jul 2015; accepted 26 Jul 2015; published 29 Jul 2015

and nonlasing peaks. Notably, the extent of the red shift for the terrace microsphere pumped at the edge was 4–6 times greater than that for the uncoated sphere. In addition, pumping at the edge of the terrace induced a greater shift than pumping at the center of the terrace. Such a peak shift with increasing excitation power in microsphere lasers is known as the thermal effect [9,25,26]. A microsphere laser is an active cavity within which phonons associated with the nonradiative transition in an emission center such as a Nd^{3+} ion deposit heat and increase the temperature of the microsphere. The increase in temperature induces both an expansion of the microsphere and a change in the refractive index. These changes vary the effective cavity length, resulting in a resonant wavelength shift. Therefore, the red shift is related to the pumping photons absorbed by the emission center in the cavity. Assuming that the thermal properties of the terrace microsphere were nearly the same as those of the uncoated sphere, the large wavelength shift in the terrace microsphere implies that the terrace structure improves the coupling efficiency between the free-space beam and modes in the microsphere, resulting in the absorption of a greater quantity of pumping photons by the Nd^{3+} ions in the microsphere.

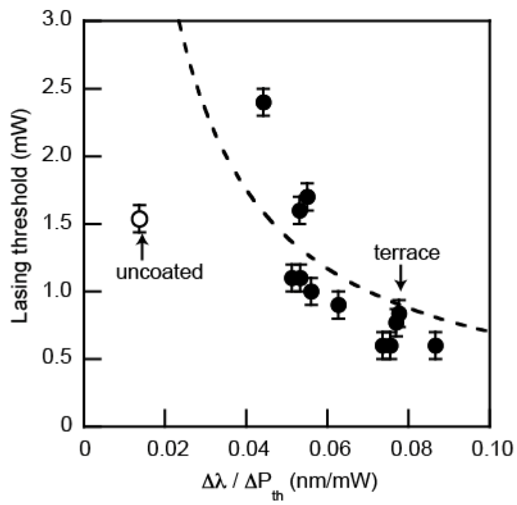

Fig. 6. Lasing threshold versus the ratio of the wavelength shift ($\Delta\lambda$) and increase in the excitation power (ΔP_{ex}). Closed circles indicate the results for the terrace microsphere at various pumping positions in the terrace. The open circle indicates the results for the uncoated sphere pumped at the edge of the sphere. The error bars represent 0.1 mW because of the instability of the excitation laser. The dashed line is a guide for the eye.

4. Conclusion

Laser action from a terrace microsphere laser of Nd^{3+}-doped K_2O–WO_3–TeO_2 glass was investigated. The number of laser lines decreased after terrace formation, and quasi-single mode laser emission was achieved because of modified WGMs in the terrace microsphere using free-space coupling. The resonance peaks were shifted to longer wavelengths with increasing the excitation power. The extent of red shift, which is related to absorbed pumping photons, for terrace microsphere was 4–6 times greater than that for the uncoated sphere. The lasing threshold of the terrace microsphere was 0.6–2.4 mW depending on the absorbed pumping light, which varied with the pumping position in the terrace. These results indicate that the terrace structure enables not only the modification of the modes in a microsphere laser but also a decrease in the laser threshold due to an increase in the coupling efficiency between the cavity and free-space beam. Further studies are underway to reveal the effects of the dimensions of terrace microspheres on laser performance and explore the optimum balance between efficient coupling and degeneracy of the confinement factors in the cavity.

#238546 Received 13 May 2015; revised 22 Jul 2015; accepted 26 Jul 2015; published 29 Jul 2015