Up-conversion luminescence and C-band laser in Er$^{3+}$-doped fluorozirconate glass microsphere resonator

(Invited paper)

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Abstract: Up-conversion luminescence and C-band microsphere laser output is reported for an Er$^{3+}$-doped ZrF$_4$-BaF$_2$-YF$_3$-AlF$_3$ (ZBYA) fluorozirconate glass microsphere. The microsphere was fabricated by heating a ZBYA glass filament using a CO$_2$ laser beam. The fabrication process accurately and repeatably produces microspheres of 68 μm diameter. The input and output laser light was coupled to the microsphere using a tapered optical fiber. The coupling position between the tapered fiber and microsphere was adjusted using a sophisticated three-dimensional translation stage. The up-conversion luminescence emission, single-mode and multi-mode laser at C-band (1530 to 1565 nm) were observed when pumped using a 980 nm laser.

Index Terms: Up-conversion luminescence, microsphere resonator, fluorozirconate glass, C-band laser.

1. Introduction

In recent years, microcavity resonators have become attractive for fundamental research especially in lasers physics, including microdisks [1], microrods [2] and microspheres [3] etc. The advantages of these include high Quality factors (Q-factors) and small mode volumes which make them ideal structures for producing low threshold and narrow line-width laser output, and are widely used in the fields of optical communication, bio-medical sensing, and cavity quantum electrodynamics [4, 5]. Microcavity resonators rely on the existence of Whispering-Gallery-Modes (WGMs), for which the electromagnetic wave becomes trapped by repeated internal reflection as it propagates around the inside surface of the cavity [6]. Among microcavities in general, microspheres have a number of advantages such as simple fabrication and a high Q value [7-9]. So far microsphere lasers have been successfully fabricated in silica [10], tellurite [11], chalcogenide [12], borosilicate [13], phosphate [14], germanate [15] and fluoride glasses [16]. Generally, the quality of the microspheres mainly depends on the substrate material and the fabrication method. Common microsphere light coupling methods include via Mie scattering and evanescent field [7]. The Mie scattering coupling method utilises a free-space laser beam focused on to the edge of the sphere, but this includes significant losses which reduces the coupling efficiency. Evanescent field coupling is most effectively achieved to date utilising a micro-nano tapered optical fiber [17]. Pump photons can be efficiently coupled into the sphere via the evanescent field when the microsphere is positioned close to the taper. In this case, it is necessary that the gap between the taper and the microsphere be adjusted to achieve an optimal coupling state.

The transition $^4$I$_{13/2}$$→^4$I$_{15/2}$ in Er$^{3+}$ ions gives rise to C-band laser output, consequently these devices have a wide range of applications in telecommunication [18] and have been implemented in a wide variety of structures [19-21]. Additionally, the Er$^{3+}$-doped active medium also has potential for fabricating green lasers, which have significant potential for use in underwater optical communications e.g. in marine defense systems [22]. As early as 2000, Er$^{3+}$-doped microsphere lasers were fabricated in ZBLAN glass and these operated at a wavelength of 1.56 μm [23]. Cai et al reported an Er$^{3+}$-doped ZBLALIP whispering gallery mode laser operating at 1550 nm in 2004 [24].

ZrF$_4$-BaF$_2$-YF$_3$-AlF$_3$ (ZBYA) fluorozirconate glasses have very attractive characteristics which favour their use in lasers including low phonon energy (~ 580 cm$^{-1}$), a wide transmission window and better performance in terms of resistance to moisture compared with conventional ZBLAN glass [25]. The luminescent properties of Er$^{3+}$ doped ZBYA glass have been extensively studied, indicating that ZBYA glass is a potential candidate for use as a visible to infrared range laser [26-28]. However, it remains difficult to fabricate low-loss optical fiber using ZBYA glass. Therefore, ZBYA glass has been chosen as the material to fabricate the microsphere in the investigation described in this article and to thus provide the Er$^{3+}$ doped C-band laser output. A Tm$^{3+}$-doped ZBYA glass microsphere laser operating in the 2.0 μm wavelength region was initially reported by some of the authors of this article early in 2019 [29].

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In this investigation, Er\textsuperscript{3+}-doped ZBYA glass samples have been successfully fabricated and their transmission and fluorescence emission spectra have been measured. 0.1 mol% Er\textsuperscript{3+}-doped ZBYA glass was chosen to fabricate microsphere combined with heating provided from a CO\textsubscript{2} laser. 980 nm pump laser light was coupled into the microsphere via a tapered optical fiber. Multi- and single-mode laser outputs at 1.56 µm are observed. The visible (green) up-conversion luminescence spectrum in the microsphere has also been observed and the intensity shows an increasing trend with increasing pump power.

2. Experimental Details

A range of concentration of 0.05 mol% to 3 mol% of Er\textsuperscript{3+} ions were successfully doped into ZBYA glass which was prepared using a standard melt-quenching technique. High-purity raw materials powder samples were weighed and mixed evenly to form 50ZrF\textsubscript{4} - 33BaF\textsubscript{2} - (10-x)YF\textsubscript{3} - 7AlF\textsubscript{3} - xErF\textsubscript{3} in the ratio, (x=0.05, 0.1, 0.2, 0.5, 1, 2, 3). Each ground sample was placed in a platinum crucible and heated at 850 °C in an electric furnace for 2 hours. The melt was subsequently poured onto a preheated copper plate for annealing for 3 hours which was maintained at the glass transition temperature (370 °C). The fabricated glass samples were then polished and cut using an automatic grinder for later testing.

The properties of the Er\textsuperscript{3+}-doped ZBYA glass samples were characterized in terms of their transmission spectra, emission spectra and lifetime decay curves. The transmission spectra were measured using a Perkin-Elmer Lambda 750 UV spectrophotometer in the range λ ~ 200-2300 nm and a Perkin Elmer FT-IR spectrometer in the range λ ~ 2300-10000 nm. The fluorescence spectra were measured using a Hitachi F-4600 fluorescence spectrophotometer in the range λ ~ 500-900 nm and Zolix Omni-λ3015 infrared monochromator in the range λ ~ 1400-1700 nm. In the fluorescence decay measurements, the samples were excited with 980 nm pulsed laser (Horizon OPO), providing pulses of 6 µs duration and a repetition rate of 10 Hz. A spectrometer (Synergy 1000M) and a digital phosphor oscilloscope (DP04104B) were used to record the fluorescence decay curves.

The ZBYA microspheres were made from glass filaments with diameters of 5-10 µm using a circular ZnSe-lens-focused CO\textsubscript{2} laser beam (power ∼3 W) which was directed onto a section of the ZBYA filament. A small weight attached to the bottom of the filament upon heating facilitated the formation of a very thin tapered region (diameter circa 3 µm), which acts as the stem of the microsphere. The CO\textsubscript{2} laser was then used to cut the fiber and the remaining glass at the tip was reheated. The surface tension of the molten glass at the fiber tip when subjected to high temperature causes the fiber tip to assume a spherical morphology under the effect of gravity [30]. The diameters of the microsphere range from 40 µm to 120 µm. The diameter of the microsphere in this investigation was 68 µm, and the microscopic image confirming this is shown in inset in Fig 1. The model of the CO\textsubscript{2} laser was SYNRAD 48-2KWM-AP which was controlled by a SYNRAD UC-2000 laser controller.

The experimental setup diagram showing the measurement system of the microcavity laser is presented in Fig 1. A λ ~ 980 nm laser diode (MCSPL-980, MC Fiber Optics, China) provided pump light which was coupled into the microsphere through a tapered optical fiber to produce WGMs within the microsphere. The coupling position between the tapered fiber and microsphere was adjusted using a sophisticated three-dimensional accurate translation stage and observed using two 20× microscope eyepieces attached to separate identical CCD cameras. The tapered optical fiber was fabricated by heating and stretching a strand of 1060XP single-mode fiber on both sides using a ceramic microheater (CMH-7-19, NTT-AT) as the heat source. The diameter of the taper achieved in this investigation was about 1.5 µm. The output spectrum was measured using an optical spectrum analyzer (OSA) (AQ-6374, Yokogawa, Japan).

![Experimental setup diagram](image)

**Fig. 1.** The experimental setup for measurement of the laser output spectrum. Inset are the images of the microsphere with the diameter of 68 µm and the green up-conversion light in the microsphere.
3. Results

First the transmission and emission spectra of the Er$^{3+}$-doped ZBYA glass samples are presented in Fig 2. From the transmission spectrum of the 0.1 mol% Er$^{3+}$-doped ZBYA glass sample, one can see that the transmittance of ZBYA glass covers the ultraviolet, visible and mid-infrared wavelength bands, and the upper infrared cutoff wavelength is around $\lambda\sim9000$ nm. The absorption peaks located at 377 nm, 520 nm, 650 nm, 797 nm, 980 nm and 1530 nm can be attributed to the transitions from the ground state level to $^4G_{11/2}$, $^2H_{11/2}$, $^4F_{9/2}$, $^4I_{11/2}$, and $^4I_{13/2}$ levels, respectively. A small absorption located around 3400 nm may be caused by the stretching vibration of OH in the glass samples. The relatively strong absorption peak located at the wavelength of 980 nm means that a laser diode centred on this wavelength value was chosen as the excitation (pump) source. Under excitation from the 980 nm laser diode, the luminescence emission from $\lambda\sim500$-900 nm and $\lambda\sim1400$-1700 nm are shown in Fig 2 (b), (c), (d) respectively. The observed emission peaks are located at 528 nm, 550 nm, 666 nm, 800 nm, 846 nm and 1534 nm. The fluorescence emission intensity of 528 nm, 666 nm, 800 nm and 846 nm were much lower than that recorded at 550 nm. When the concentration of the Er$^{3+}$ ions was increased from 0.05 mol% to 2 mol%, the intensity of all luminescent emission peaks increased. However, when the doping concentration of Er$^{3+}$ was increased further to 3 mol%, the luminescence intensity at 528 nm, 666 nm and 846 nm began to decrease, indicating the presence of concentration quenching at these bands which commences at a doping concentration between 2 mol% and 3 mol%. No such quenching was observed at the 1534 nm wavelength value.

![Fig. 2. (a) Transmission spectra of Er$^{3+}$-doped ZBYA glass samples in the range of $\lambda\sim200$-10000 nm. (b) fluorescence emission spectra of Er$^{3+}$-doped ZBYA glass samples in the range of $\lambda\sim500$-900 nm and $\lambda \sim1400$-1700 nm.](image)

The energy-level diagram for Er$^{3+}$ ion is shown in Fig 3(a). Under excitation of 980 nm laser, the Er$^{3+}$ ions initially in the ground state $^4I_{15/2}$ absorb photons and the ion transfers to the $^4I_{13/2}$ level via the ground state absorption (GSA) process. The ions then relax to the $^4I_{13/2}$ level after which further excitation from the pump lifts the ion to the $^4F_{9/2}$ level. The Er$^{3+}$ ions in the $^4I_{13/2}$ level continue absorbing 980 nm laser and are excited to $^2H_{11/2}$, resulting in an excited state absorption (ESA) [26]. Ions with electrons located in the upper energy levels can relax directly back to the ground state. The temporal evolution of the emission at a wavelength of 1534 nm under 980 nm excitation is shown in Fig 3(b). The lifetime of the emission from the $^4I_{13/2}$ level transition in the 0.1 mol% Er$^{3+}$-doped ZBYA glass was measured as 11.5 ms, which is longer than the value observed in pure ZBLAN glass (8.69 ms) [31]. The transition from the $^4I_{13/2}$ to the $^4I_{15/2}$ level results in the 1550 nm wavelength laser emission. Generally, the relatively longer decay lifetime is beneficial for reducing the laser oscillation threshold.
Fig. 3. (a) Energy level diagram of Er³⁺ ions under 980 nm laser excitation. (b) Fluorescence decay curves of Er³⁺-doped ZBYA glass samples at 1534 nm.

When the 980 nm laser was coupled into the Er³⁺-doped ZBYA glass microsphere, a multimode laser line at ~1.56 µm was observed with a pump threshold power of 1.34 mW, the actual threshold should be lower than this, but the raised value can be attributed to the minimum output power problem associated with the pump laser diode. Fig 4(a) shows the output light spectrum of the multimode laser centered at ~1.56 µm when the pump power was 14.2 mW. By adjusting the coupling position between the microsphere and the tapered fiber, a single mode laser emission at ~1557 nm appeared, and whose line width was 0.1 nm as observed in the spectrum shown in Fig 4(b). In the experiment of this investigation, the WG lasing modes related to the taper contact position on the microsphere and pump power. The existence of the fundamental WG mode (|m| = 1) makes it relatively easy to absorb more pump energy and thus lowers the lasing threshold. Single mode lasing can therefore be achieved using a lower pump power when the taper is physically contacted at the microsphere equator [32, 33]. Mode competition was observed and the higher order polar laser modes (l > |m|) were excited as the coupling taper was placed far from the equatorial plane in a corresponding region [34]. Fig 4(c) demonstrates the up-conversion luminescence in the Er³⁺-doped ZBYA microsphere when subjected to different pump powers. Photoluminescence peaks located at 525 nm, 550 nm, 650 nm, 800 nm and 850 nm were also observed. Increasing the pump power resulted in a corresponding increase of the luminescent intensity of the emissions at each wavelength. The discontinuity in the spectrum of Fig 4(c) observed at 580 nm was caused by the grating conversion in the spectrometer. At the same time, an intense green up-conversion luminescence in the microsphere was visible to the naked eye, which is also shown photographically in Fig 1 inset. It is shown centered on a wavelength of 550 nm and is labelled in Fig 4(c).

The output power of the microsphere laser as a function of the pump power is shown in Fig 4(d). As the minimum power of the 980 nm laser is 1.34 mW, the laser output was observed from this value immediately without observation of fluorescence. While, the threshold is much lower than the output power obeys a linear relationship with the pump power. The threshold of the 1.5 µm laser in Er³⁺-doped ZBYA microsphere is lower than tellurite glass microsphere (2 mW) [11] and silica glass microsphere (2 mW) [35].
4. Conclusions

The successful realization and characterization of a microsphere laser with an output wavelength at 1.56 µm together with up-conversion luminescence with Er³⁺-doped ZBYA glasses have been experimentally demonstrated. The laser was excited via evanescent wave coupling from a proximal tapered microfiber. Single mode and multimode laser operation with threshold of 1.34 mW were observed using a 980 nm laser diode as a pump source. The result in this work indicated that ZBYA glass has potential to be used as the gain material for communications based lasers operating in the 1.56 µm range and hence have several potential applications in the communications/ICT space. An up-converted emission located at a wavelength of 550 nm was also observed which could provide further applications in several specialized applications including underwater optical communications and sensing.

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