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EFFECT OF EXCITATION MIGRATION AND UPCONVERSION IN HIGHLY ERBIUM-DOPED GLASS MICRO-SPHERE LASERS

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Abstract. *Excitation migration and upconversion process in the Erbium-doped silica-alumina glass microsphere lasers with concentrations of 2500-4000 ppm were investigated in detail. The experiment shows that under 976 nm excitation, the intense of up-conversion emission at 523, 546 and 657 nm, corresponding to the transitions* $^2H_{11/2} \rightarrow ^4I_{15/}$, $^4S_{3/2} \rightarrow ^4I_{15/2}$, *and* $^4F_{9/2} \rightarrow ^4I_{15/2}$, *respectively, depends on the erbium content, migration of excitation and pump power. The excitation migration has strongly influenced on the threshold and Red shift of lasing wavelength and migration-assisted up-conversion process leads to degraded amplification performance of micro-sphere lasers made by silica-alumina glasses with different contents of Er-ions.*

I. INTRODUCTION

Recently, the investigations of up-conversion processes have attracted great interest in connection with the development of Erbium-doped glasses with high doping concentration of Erbium ions (10^{20}-10^{21} cm^{-3}) for planar amplifiers, and micro-cavity lasers operating at 1550 nm [1-3]. In other hand, the conversion of infrared light to visible light through energy up-conversion in rare-earth doped glasses, due to the possibility of infrared-pumped visible lasers, has a great potential application in areas such as optical data storage, lasers, sensors, and optical displays [4-6]. The increase of Erbium concentration leads to degraded amplifier performance owing to migration-assisted up-conversion processes [7-8]. To characterize the performance of a high –concentration optical amplifiers, model accounting for the up-conversion of excitation on homogeneously distributed (homogeneous up-conversion, or HUC) and clustered erbium ions (pair-induced quenching, or PIQ) have been used. The earlier models describing the up-conversion in Er-doped fiber assumed that the migration is so mush faster than the up-conversion that it smoothes out the excitation distribution even though all the ions are randomly distributed. This approximation resulted in a linear dependence of the up-conversion rate on the population inversion. Philipsen and Bjarklev proposed the Monte-Carlo technique for investigation of the up-conversion phenomenon in [9], that for randomly distributed ions, the up-conversion rate is a non-linear function of the population inversion and that it is accelerated by migration. In this description, the ensemble of all the excited ions was divided into two sub-ensembles: equidistant isolated ions and the clustered ones. However, a detailed microscopic study of Erbium-doped glasses by means of X-ray absorption fine structure spectroscopy has found no evidence of short-range pair clustering of Erbium ions [10]. Therefore, more accurate physical models have to be used for fitting experimental results [11].

In this work we experimentally investigate the excitation migration and up-conversion processes in the very high-concentration Erbium-doped silica-alumina glasses in the form of fiber and micropherе bulk. The experiment shows that under 976 nm laser diode (LD) excitation, the intense of up-conversion emission at 523, 546 and 657 nm, corresponding to the transitions $^2H_{11/2} \rightarrow ^4I_{15/}$, $^4S_{3/2} \rightarrow ^4I_{15/2}$, and $^4F_{9/2} \rightarrow ^4I_{15/2}$, respectively, depends on the Erbium content, migration of excitation and pump power. The excitation migration and up-conversion process in the highly Er-doped glasses were verified by increase of threshold, Red shift of laser emission wavelength at the threshold and non-linear decrease of lasing power of microsphere lasers based on silica-alumina glasses with different contents of Er-ions.

II. BACKGROUND

II.1. Micro- and macroscopic characterization of excitation migration and up-conversion process

Migration of excitation and up-conversion process in the frame of microscopic characterization are based on excited state absorption (ESA) and the non-radiative excitation transition in pairs of optical centers. According to microscopic characterization of migration and up-conversion processes in [12], the rate of conversion transition is inversely proportional to the six power of distance between ions and the oscillator strength of corresponding transitions from ground and excited states.

The macroscopic approach consists in the application of rate equations to populations of energy levels and the temporal variation of number density for a metastable level is usually described by following equation:

$$\frac{\partial N_2}{\partial t} = -AN_2 + Bu(\nu)N_1 - C_{up}N_2^2 \qquad (1)$$

Here, the first term is the number of spontaneously emitted photons, the second term is the number of pumping transitions from ground state, and the third term is the number of up-conversion transition, N_1 and N_2 are number densities for ground and metastable levels, A is the rate of spontaneous emission, $u(\nu)$ is the density of pumping radiation, B is the Einstein coefficient for absorption. It is usually supposed that the number of up-conversion transition is proportional to the square of the population at metastable level, N_2, because two excited atoms are needed for up-conversion transition. The proportionality coefficient, C_{up}, is considered as a macroscopic quantitative characteristic of the up-conversion process. In general, the C_{up} parameter is a function of time and may depend on the concentration of Er-ions, pumping and time recording of luminescence, when a pulse excitation is used. In an experiment, the researcher always measures macroscopic parameters, which are results of the averaging of individual pair properties and up-conversion coefficient, C_{up}, is constant.

II.2. Experiments

In our experiments, we have used SiO_2:Al_2O_3 (silica-alumina) glasses doped with Er and Y/Er with Er-concentration varied from 2500 ppm to 4000 ppm (Table 1). The Er-doped silica-alumina glasses with a fiber form have been used for ensuring sufficient pump power density into the doping area. The Er-doped fiber was a commercial silica-alumina fiber with a typical core/cladding diameter ratio of 9/125 μm and Er-doped area diameter of 3 μm. The sol-gel glass samples with the composition (mol %) $90SiO_2$- $6Al_2O_3$: $4Y_2O_3$: xEr_2O_3, where x = 0.125 and/or 0.2 (it is equivalent to 0.25 -0.4 mol% of Er^{3+}-ion in glass matrix). The Er-sample bulk has a spherical form with diameter varied from 90 to 120 μm. The method for making multi-component and micro-sphere Er-doped glasses was shown in our previous work [13]. We choose 976 nm- laser diode with adjustable output power in the range from 0 to 170 mW in single-mode emission (SDLO-2564-170) for excitation of Erbium ions dispersed into the both kinds of samples. In the case of spherical samples, we had used two different half-taper optical fibers for the pumping and the collecting light emission from the samples. The taper fibers were fabricated by chemical etching a standard single-mode fiber. The waist diameters of half-taper fiber were from 1 to 4 μm, which may be optimized to phase matching and coupling to the fundamental Whispering Gallery Modes (WGMs). The spectra of infrared light were measured by Optical Spectrum Analyzer (OSA) Advantest Q8384 made in Japan of 0.01 nm resolution and the up-conversion spectra were analyzed by monochromator MicroSpec 2300i, silicon CCD camera with resolution of 0.1 nm and sensitivity of - 90 dBm.

Table 1. Concentration of Er^{3+} and Y^{3+} ions

# Sample	Glass	Sample form	Concentration (ppm)	
			Er^{3+}	Y^{3+}
1	Silica-alumina	Fiber	2500	-
2	Silica-alumina	Fiber	4000	-
3	Silica-alumina	Micro-Sphere	4000	-
4	Silica-alumina (Solgel)	Micro-Sphere	2500	80000
5	Silica-alumina (Solgel)	Micro-Sphere	4000	80000

III. RESULTS AND DISCUSSIONS

Figure 1 demonstrates a simplified energy level diagram of the visible up-conversion emissions from the Er^{3+}-ions in the glass matrix of silica-alumina by a 976nm laser diode excitation. The Er^{3+}-ion was populated on $^4I_{11/2}$level through the ground state absorption by this scheme: $^4I_{15/2}$ + pumped photon (p-photon) $\rightarrow$ $^4I_{11/2}$. Then the excited state absorption (ESA) occurred by two ions at $^4I_{11/2}$ populated one Er^{3+}ion to the $^4F_{7/2}$ level and other one to ground state ($^4I_{11/2}$(A) + $^4I_{11/2}$(B) $\rightarrow$$^4I_{15/2}$ (A) + $^4F_{7/2}$ (B)), and a nonradiative decay from $^4F_{7/2}$to $^2H_{11/2}$,$^4S_{3/2}$, and $^4F_{9/2}$levels. Finally, the green and red up-conversion emissions centered at about 523, 546 and 660 nm were produced by the emitting transition of $^2H_{11/2} \rightarrow {}^4I_{15/2}$, $^4S_{3/2} \rightarrow {}^4I_{15/2}$ and $^4F_{9/2} \rightarrow {}^4I_{15/2}$ of Er^{3+}, respectively.

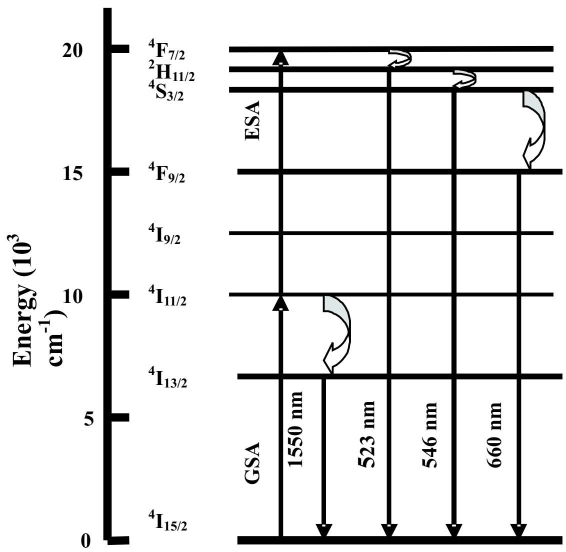


Fig. 1. Energy level diagram of green and red up-conversion emissions from the Er^{3+} ions distributed into SiO_2:Al_2O_3 glasses by a 976 nm laser diode excitation.

Figure 2a shows the spectra of up-conversion emissions in the wavelength range of 500-670 nm for the Er^{3+} ions in the silica-alumina glass matrix excited by 976nm laser diode. The green up-conversion emission spectra exhibited two green emission bands (figure 2b) centered at about 523 and 546 nm, which were attributed to the $^2H_{11/2} \rightarrow {}^4I_{15/}$, and $^4S_{3/2} \rightarrow {}^4I_{15/2}$ of Er^{3+}ions in the silica-alumina glasses, respectively. The energy gap between the $^2H_{11/2}$ and $^4S_{3/2}$ levels (in the range of 760-900 cm^{-1} depending on the host glass matrix) could be obtained from the green up-conversion emission spectra. At the quasithermal equilibrium at room temperature the fluorescence intensity of the transitions of $^4S_{3/2} \rightarrow {}^4I_{15/2}$ (at wavelength of 546nm) is more than of $^2H_{11/2} \rightarrow {}^4I_{15/2}$ (at 523nm). In general, the fluorescence intensity ratio (FIR) of the two emissions varied by temperature [14].

Figure 3 shows a plot of the up-conversion emission intensity at wavelength peaks of 523 and 546 nm as a function of pumping power of 976nm laser diode. This experimental result is good evidence to support for the Monte-Carlo technique proposed by Philipsen and Bjarklev in [9] for investigation of the up-conversion phenomenon, that the up-conversion rate is a non-linear function of the population inversion and that it is accelerated by migration of excitation. This result is promising for making green light laser from erbium-doped glasses.

To verify the migration of excitation and up-conversion process in the highly Er-doped silica-alumina glasses, we investigated the lasing characteristics of micro-sphere lasers based on silica-alumina glasses doped 2500 ppm and/or 4000 ppm of Er^{3+} ions. We

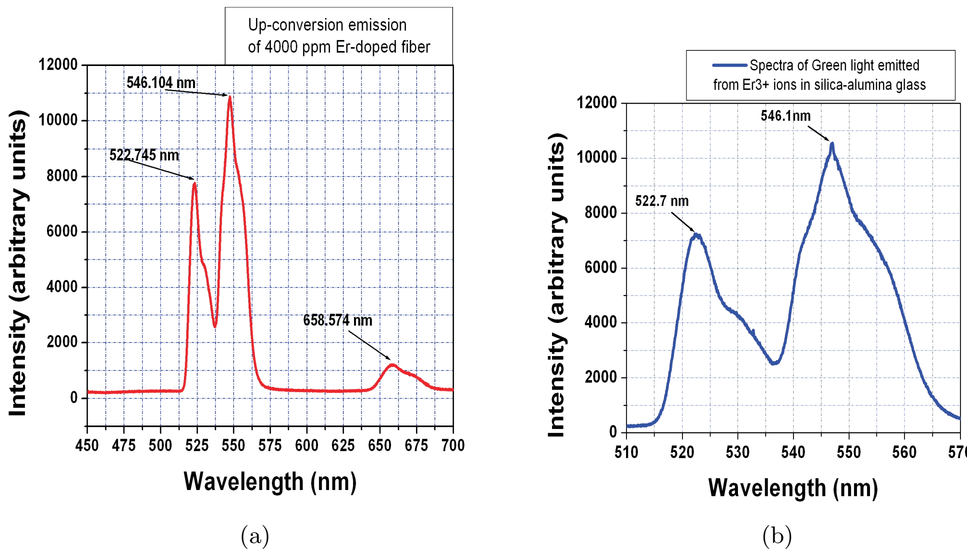


Fig. 2. Up-converion emission spectra in the wavelength range of 500- 600 nm (a) and green light emissions (b) for the Er^{3+}-doped silica-alumina glasses with 4000 ppm-concentration of Er-ions

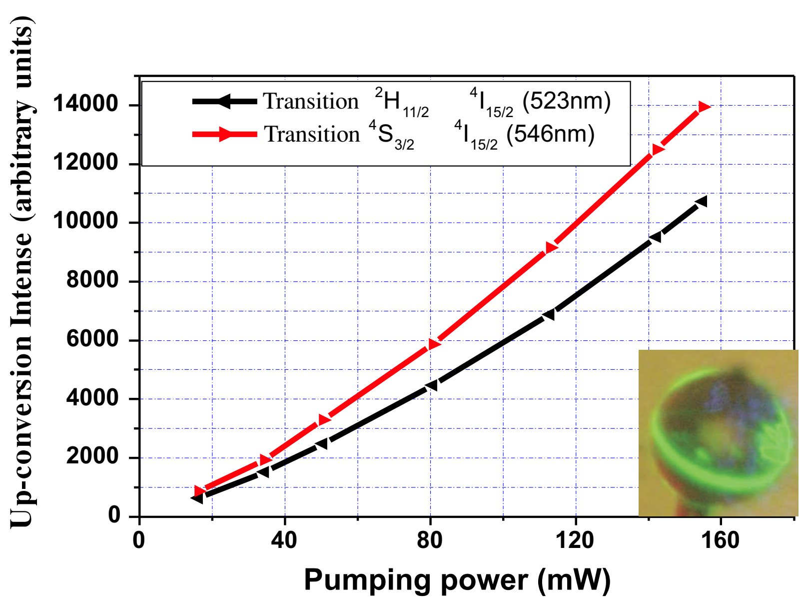
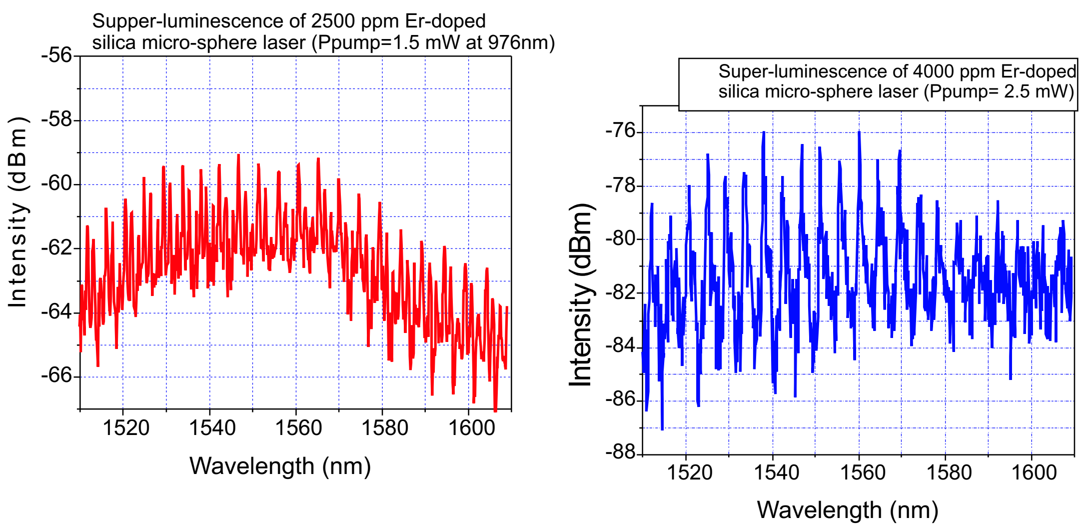


Fig. 3. Non-linearly dependence of Green up-conversion emissions at 523 and 546 nm versus pumping power of 976 nm laser diode. Inset: Image of upconversion green light emission from microsphere laser.

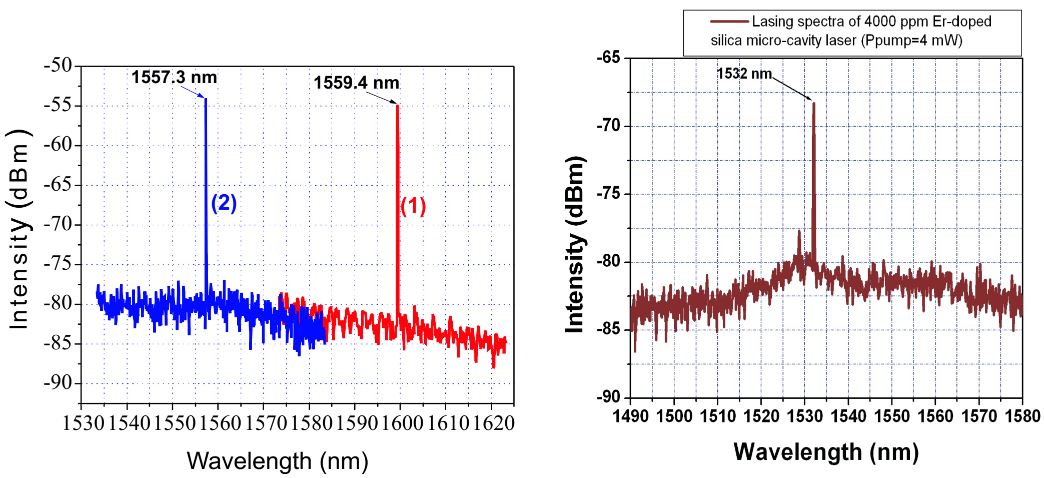
had obtained good results of the WGM emissions from sol-gel Er-doped glass micro-sphere lasers in our previous works [13, 15]. The optimal ratio of Al_2O_3:Er_2O_3 and optimal Er^{3+}-concentration dispersed into sol-gel silica-alumina glasses for micro-sphere lasers were 10:1 and of 2500 ppm, respectively. In comparison with previous studies, we have made the micro-sphere lasers with diameters of 90-120 μm based on Er-doped silica-alumina glass of 4000 ppm of Er^{3+}-ions concentration. Figure 4 demonstrates the supper-luminescence emissions from 2500 ppm and 4000 ppm Er-doped micro-sphere lasers with the pumped power below the threshold of 1.5mW and of 2.5mW at 976nm LD excitation, respectively. We can observe the laser oscillation modes (WGMs) of varied Er-doped silica-alumina microsphere lasers with a diameter of 90-120 μm, when the optical pump power at 976 nm was below the laser threshold. The WGM laser oscillations were in the large wavelength range from 1510 nm to 1610 nm, which was in the both C-band and L-band for telecom. Here the Er-ion concentration was ranging 0.25 - 0.4 mol% of Er_2^{3+}. It is remarkable that with increasing Er- ion concentration from 2500 ppm to 4000 ppm the amplitude of supper-luminescence emissions would be non-linearly decreased (from - 60 dBm to -76 dBm in our cases).



(a) (b)

Fig. 4. Supper-luminescence emissions measured from micro-cavity lasers with diameter of 110 μm under 976nm LD excitation. a) SL emission from 2500 ppm and b) from 4000 ppm Er-doped silica-alumina glasses

Figure 5 shows the single-mode lasing spectra of WGM from microsphere lasers with Er-ion content changed from 2500 ppm to 4000 ppm. In the case of homogeneous bulk 2500 ppm Er-doped glass laser, the threshold was of 2 mW and the lasing wavelength peak was at about 1600 nm. For the 2500 ppm Er-doped glass-coated laser, the threshold was increased to the 2.5mW and the wavelength peak was shifted to the 1557 nm. The peak intensity of output power at laser threshold of both cases was of -55dBm (Fig. 5a). Microsphere laser based on 4000 ppm Er-doped silica-alumina glass has very



(a) (b)

Fig. 5. a) Single-WGM spectra of solgel 2500 ppm Er-doped glass micro-sphere lasers at threshold ($P_{pump} \approx 2$ - 2.5 mW): (1) solgel bulk glass microsphere and (2) solgel coated glass microsphere. b) Single-WGM spectra of 4000 ppm bulk Er-doped glass micro-sphere at threshold pump of 4 mW. Diameter of sphere was of 110 μm for all cases.

high threshold, and in some case the threshold would be increased for two times in comparison with 2500 ppm Er-doped glass lasers and the lasing wavelength peak was shifted to the 1532 nm (Fig. 5b). The lasing power peak at threshold of 4000 ppm Er-doped glass laser was of -68 dBm, which decreased on 13dB (equivalent to 20 times less) in comparison with 2500 ppm Er-doped glass lasers. The obtained results of decreasing light emission intensity of IR-range with increasing Er-ion concentration can be explained by [16]: (i) the higher the surrounding density is, the faster is the up-conversion, that means the rate of up-conversion would be proportional to the concentration of excited ions.(ii) Since the up-conversion is fastest in the regions of the highest Er-ion density, it causes holes in the excitation distribution and migration efficiently fills up those holes by bringing excitations from the lower-density regions, where the up-conversion would be with a lower rate. Hence, the up-conversion is accelerated by migration. In our experiments, the migration of excitation may be directly influenced on the emission spectra of the Er-doped glasses and the migration-assisted up-conversion process leads to degraded amplification performance in the IR-range.

IV. CONCLUSIONS

In summary, we report new experimental results of the up-conversion emissions in the highly Er^{3+}-doped silica-alumina glasses of the form of fiber and spherical bulk. The intensity and emitting wavelength range of visible and IR emissions were investigated in detail based on spectrum characteristics of luminescence and lasing emissions from Er-doped glass micro-cavity lasers. The up-conversion rate is a non-linear function of the population inversion and that it is accelerated by migration of excitation. The IR emission intense had been decreased on 13-16 dB and the lasing wavelength at threshold was shifted to the red-wavelength range, when the Er^{3+}- ion concentration was increased from 2500 ppm to 4000 ppm in micro-cavity Er-doped glass lasers. The obtained results are good evidence of phenomenon of migration-assisted up-conversion process in the highly Er^{3+} -doped silica-alumina glasses.

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