Concentration quenching of luminescence lifetime in ytterbium-doped potassium double tungstate waveguide amplifiers

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Integrated optical amplifiers employing ytterbium-doped potassium double tungstates exhibit an ultra-high peak gain exceeding 1000 dB/cm, in addition to a broad gain of up to 150 dB/cm over a 55-nm wavelength range. Here we report a study of the luminescence lifetime in samples spanning a broad range of dopant concentrations (1.2–57.5 at.%). By use of the pinhole method, elongation of the luminescence lifetime due to radiation trapping is avoided, providing a more accurate analysis of concentration quenching of the luminescence lifetime, which directly influences the waveguide amplifier performance and can, in principle, inhibit scaling of the gain with increasing dopant concentration.

Introduction

Rare-earth-ion-doped gain media are widely applied as solid-state and fiber lasers and amplifiers, thus it is of great interest to simulate their behavior in order to optimize their performance. Among the parameters that are required for investigating these devices by rate-equation models is the luminescence lifetime, therefore it is important to accurately measure the luminescence lifetime for the proper simulation of these devices.

With increasing rare-earth-ion concentration, the probability of undesired effects such as energy-transfer processes among rare-earth ions, resulting in luminescence lifetime quenching [1], and radiation trapping, resulting in luminescence lifetime elongation [2], increases. Since these processes occur simultaneously, it is beneficial to isolate them for a proper understanding of the system.

Trivalent ytterbium has only one excited state within the 4*f* subshell and, therefore, parasitic spectroscopic processes such as excited-state absorption and energy-transfer upconversion are absent. Nevertheless, energy migration to and transfer to impurities, crystal defects, or color centers, as well as radiation trapping are observed at high Yb^{3+} concentrations due to the large overlap between absorption and emission cross-sections. Here we present a study of the luminescence lifetime, employing the pinhole method [3], of several ytterbium-doped potassium double tungstates of the composition $KY_{1-x-y-z}Gd_xLu_yYb_z(WO_4)_2$. The effects of radiation trapping and concentration quenching in several samples covering concentrations from 1.2 at.% to 57.5at.% are evaluated.

Experimental

Thin $KY_{1-x-y-z}Gd_xLu_yYb_z(WO_4)_2$ layers were fabricated by liquid phase epitaxy on pure KYW substrates as described elsewhere [4]. In Table 1 the composition and thickness of each sample are presented.

Table 1 Samples, film thicknesses, and compositions.

Sample #	Film thickness [µm]	Y [at.%]	Gd [at.%]	Lu [at.%]	Yb [at.%]
1	36.5	98.8	-	-	1.2
2	28	98.3	-	-	1.7
3	27	60	13	24.6	2.4
4	20	75	10	-	15
5	15	-	47.7	34.8	17.5
6	42.5	67	13	-	20
7	20	-	46.2	21.3	32.5
8	24	-	44.7	47.5	47.5
9	16.8	-	42.5	-	57.5

Single-exponential decay curves of luminescence from the Yb^{3+}-doped layers were recorded by pumping each film with a switched diode laser operating at 976 nm. The square pump pulses had a 200 Hz frequency and 50% duty cycle. Luminescent emission in the 1020 nm band was collected perpendicularly from the sample by use of a multimode glass fiber (NA = 0.24). The collecting fiber was placed as close as possible to the point of pump-light launching and right over the film surface in order to minimize reabsorption, as shown in Fig. 1. In order to quantify the effect of radiation trapping in our samples, the distance of the collecting fiber from the sample was varied from 0 mm (just over the film surface) to 2.5 mm in steps of 500 µm, indicated as the distance *d* in Fig. 1. An example of lifetime elongation can be examined in Fig. 2, where the slope of luminescence decay (in logarithmic scale) increases as the distance of the collecting fiber from the sample surface increases.

Lifetimes were computed from the decay curves for all Yb^{3+} concentrations and fiber positions; these results are shown in Fig. 3. It is evident that even for the lowest Yb^{3+} concentration, lifetime values depend strongly on the area of observation, determined by the collecting cone and distance of the fiber. As shown by Kühn *et al.* [3], there is a linear dependence of lifetime values on the radius of the area studied, as indicated by the fitted line in each figure. It is mainly due to the fact that luminescence emitted from a smaller area contains a smaller amount of light re-absorption compared to a larger area, which leads to shorter lifetimes.

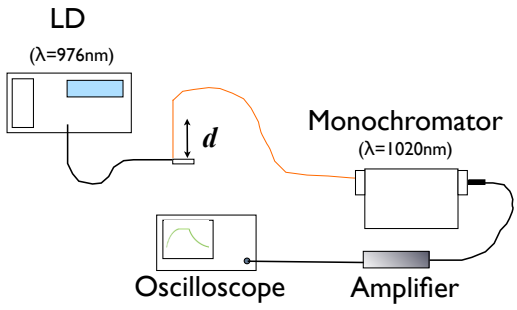

Fig. 1. Schematic of the experimental setup employed to measure the luminescence.

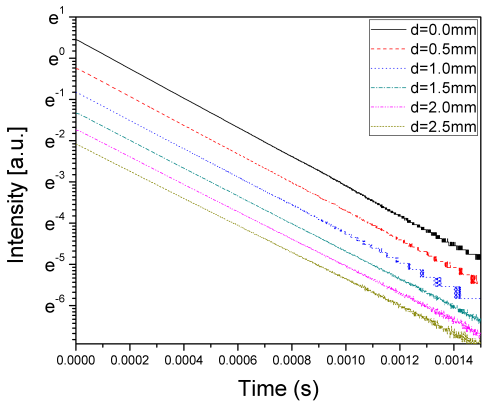

Fig. 2. Luminescence decay curves from sample # 6 (Yb^{3+} concentration 20at.%) at different collecting fiber heights.

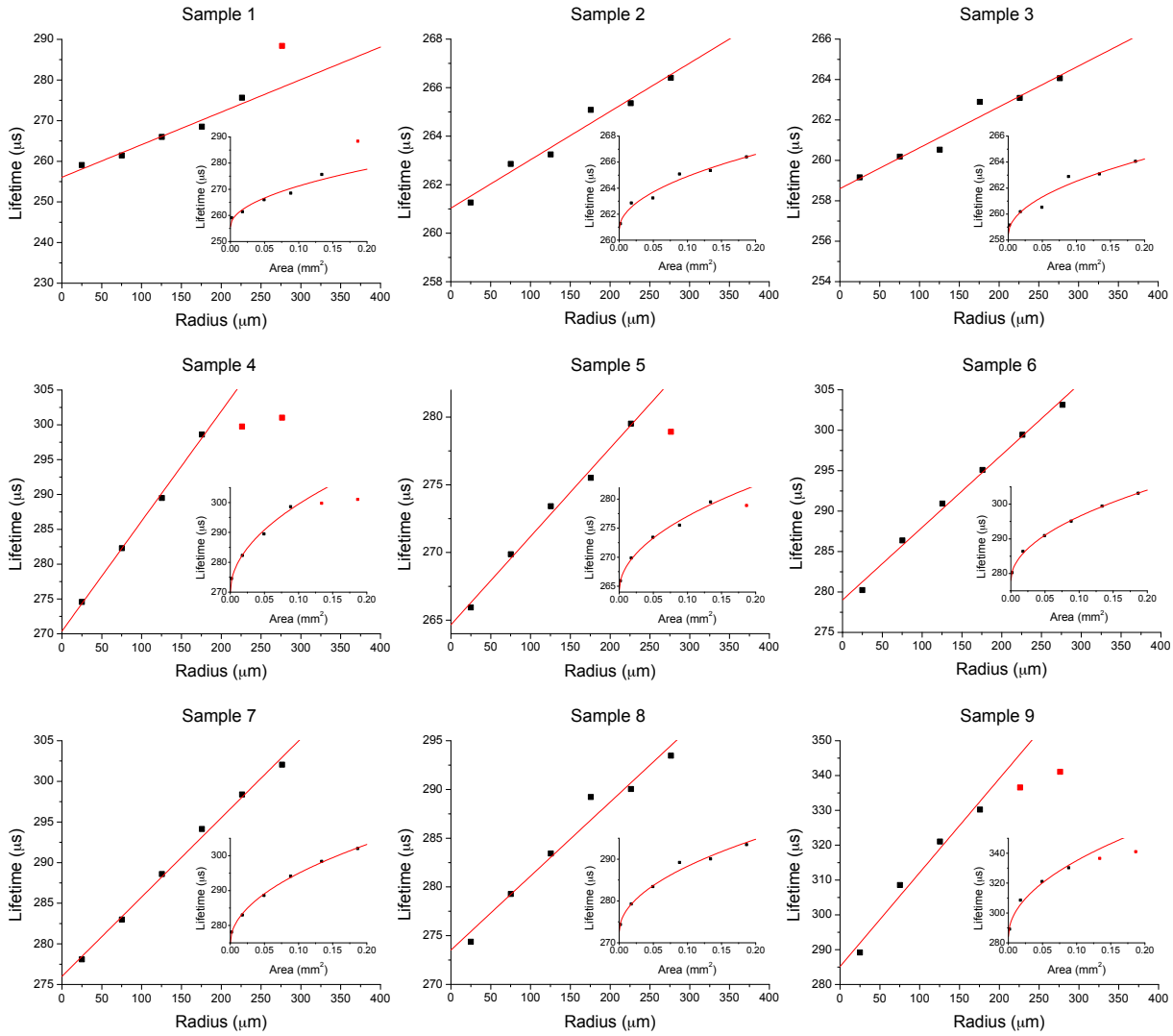

Fig. 3. Luminescence lifetimes of all samples recorded at different distances of the collecting fiber. The graphs show the linear dependence on the radius of observation, and inserts show the square-root dependence of lifetime on the area of observation.

Due to the geometry of the experimental setup, the observed area was not always circular. Because the fiber was close to the pumped end and lifted from the sample surface, part of the acceptance cone was outside the sample area. The dependence of lifetime on calculated observation area is shown in the insets of Fig. 3; the lines are fits using the equation $\tau = a_0 + a_1\sqrt{A}$, where a_0 and a_1 are fit parameters and A is the observation area.

In Fig. 4 the dependence of lifetime on ytterbium concentration is shown for all positions of the collecting fiber. The experimental lifetime values corresponding to the smallest observed area were chosen to fit the equation [1]

$$\tau(N) = \tau_0 \frac{1 + \sigma l N}{1 + \frac{9}{2\pi}\left(\frac{N}{N_0}\right)^2} \qquad (1)$$

$\tau_0 = 261$ μs is the lifetime at low Yb^{3+} concentration, $\sigma = 2.92 \times 10^{-21}$ cm^2 is the absorption cross-section taken from [5], N is the dopant concentration, $l \approx 140$ μm is the average absorption length, and $N_0 \approx 2 \times 10^{22}$ cm^{-3} is the critical concentration for concentration

quenching (1at.% equals 0.63×10^{20} cm^{-3}). The latter two are free parameters to fit the concentration dependence of lifetime, as shown as a continuous line in Fig. 4. The denominator in Eq. (1) represents the contribution of concentration quenching, while luminescence reabsorption is accounted for by the numerator. By withdrawing the numerator from Eq. (1) it is possible to de-convolute the concentration quenching from luminescence reabsorption, resulting in the dashed line in Fig. 4. As seen from this line, concentration quenching is relatively weak as a result of the large distances between adjacent Yb^{3+} ions in the investigated double tungstate crystals, in addition to the simplicity of the electronic level scheme of Yb^{3+}, which inhibits energy-transfer upconversion.

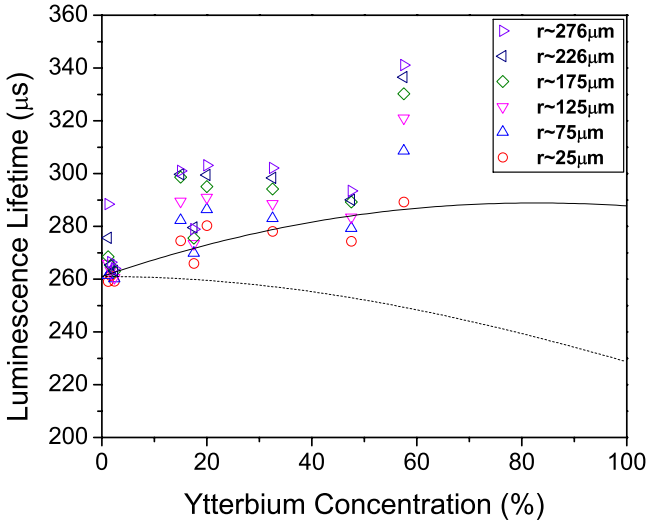

Fig. 4. Experimental lifetime data in $KY_{1-x-y-z}Gd_xLu_yYb_z(WO_4)_2$ for different Yb^{3+} concentrations (at.%) and six different observation areas. The best fit from Eq. (1) to the lifetime measured just above the film surface is shown as the continuous line, while the dashed curve shows the expected lifetimes without luminescence reabsorption, i.e., quantifying the influence of concentration quenching.

Conclusion

A study of concentration quenching of the luminescence lifetime in $KY_{1-x-y-z}Gd_xLu_yYb_z(WO_4)_2$ indicates that for concentrations up to 57.5at.% only weak concentration quenching is present, opening the possibility of using highly doped layers as optical amplifiers and lasers in the absence of detrimental effects often observed at high doping levels. This leads to interesting opportunities concerning the miniaturization of amplifiers in optical circuits.

References

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