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Highly Yb-doped oxides for thin-disc lasers

K. Petermann[a,*], D. Fagundes-Peters[a], J. Johannsen[a], M. Mond[a], V. Peters[a], J.J. Romero[a], S. Kutovoi[b], J. Speiser[c], A. Giesen[c]

[a]*Institute of Laser-Physics, University of Hamburg, Germany*
[b]*General Physics Institute, Moscow, Russia*
[c]*IFSW, University of Stuttgart, Germany*

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Abstract

For the construction of thin-disc lasers, active media with high RE-dopant concentrations and high thermal conductivity are required. YAG is well known for its excellent thermal properties, which are surpassed only by the sesquioxides Sc_2O_3, Y_2O_3, and Lu_2O_3. Among these, Lu_2O_3 is the most promising oxide because of its high Yb-doping capability at nearly no drop of the heat conductivity. Another important parameter is the quantum efficiency, which in the case of high Yb concentrations may decrease strongly due to energy migration between the Yb ions and finally deexcitation at certain impurities. To overcome this drawback either crystals of extreme purity have to be grown or crystals with structures have to be used, where for reduced energy migration the minimum Yb–Yb separation is quite large like in $KY(WO_4)_2$ (KYW) and $LaSc_3(BO_3)_4$ (LSB). Both routes have been investigated and successfully applied in thin-disc lasers.

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Keywords: Solid state lasers; Oxides; Borates; Tungstates; Yb-doping; Fluorescence lifetime; Quantum efficiency

1. Introduction

In the past, thin-disc lasers have turned out to provide high output powers at high beam quality. Both properties are of great importance for material processing like cutting and welding. The active medium in these lasers is typically Yb-doped YAG because of its excellent optical properties and high thermal conductivity.

In order to get sufficient absorption of the pump radiation in the thin crystal plates (100–200 μm) quite high Yb concentrations are necessary. Yb^{3+} exhibits a very simple energy level scheme with the ground state and only one excited state. However, resonant migration of the excitation energy between the Yb ions occurs. The higher the

*Corresponding author. Fax: +49 40 8998 5195.
E-mail address: petermann@physnet.uni-hamburg.de (K. Petermann).

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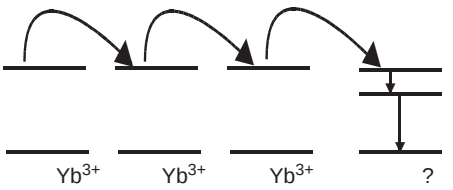

Fig. 1. Energy migration in highly Yb-doped crystals.

concentration of the Yb ions the faster this energy migration. As a consequence, a certain portion of the excitation energy may also be transferred to impurities in the crystal lattice resulting in radiative or nonradiative deexcitation and thus decreasing the Yb-fluorescence quantum efficiency (see Fig. 1). This quenching effect may be so strong that the quantum efficiency drops by many orders of magnitude. With fully concentrated as-grown YbAG crystals ($Yb_3Al_5O_{12}$), quantum efficiencies as low as 10^{-5} have been observed experimentally.

Two possibilities exist to minimize this quenching effect:

1. Reduction of the number of dangerous impurities.
2. Reduction of the energy migration by using crystal structures with large Yb–Yb separation.

Both methods have been investigated and successfully applied in thin-disc lasers.

2. Results

2.1. Crystals under investigation

To investigate the quenching effect in detail, a series of Yb-doped YAG crystals with concentrations up to 100% has been grown by the well-known Czochralski technique using 5 N starting materials. Since high-purity iridium does not exist, all crystals have been grown from 6 N powder-metallurgical rhenium crucibles under reducing growth conditions (N_2/5% H_2). Thus, the resulting crystals are of extreme purity.

Furthermore, Lu_2O_3 crystals have been grown by the HEM technique (heat exchange method) also from rhenium crucibles. This sesquioxide is difficult to grow due to its high melting temperature of 2430 °C, but it is a very promising material due to its superior heat conductivity, especially when doped with Yb.

As a crystal with quite large rare-earth distances, $LaSc_3(BO_3)_4$ (LSB) was also grown by the Czochralski technique with Yb concentrations up to 30%. In the LSB structure [1] the La sites are separated by Sc ions or BO_3 groups which hinder the energy migration between the doping ions considerably. Strong evidence for reduced interaction between the laser active ions was found in Nd-doped LSB, where quite high doping concentrations can be realized without strong cross relaxation, that is with low fluorescence quenching [2]. $KY(WO_4)_2$ also belongs to this class of oxides. Here, pairs of RE sites are separated by WO_4 groups, which causes the excellent laser properties of this material if doped with Yb (see Section 2.3).

In Table 1 some relevant physical and crystal growth data of the above laser crystals are collected. As can be seen from this table, the growth methods and material specific parameters are totally different, which does not make it easy to compare the laser potential of these materials. Lu_2O_3 exhibits the highest heat conductivity, but on the other hand the cation density is also highest and thus the Yb–Yb separation is lowest. The material with the lowest RE cation density (but lowest heat conductivity) is LSB. However, it can be doped only with up to 30% Yb in contrast to all other investigated oxides.

2.2. Lifetime measurements

One of the most important parameters for a high-power solid-state laser material is the quantum efficiency η_q of the laser active ions defined as the ratio of the radiative transition rate to the sum of the radiative and nonradiative rate:

$$\eta_q = \frac{W_{rad}}{W_{rad} + W_{nrad}} = \frac{\tau}{\tau_{rad}}.$$

So, by measuring the fluorescence lifetime τ of the Yb ions after pulsed excitation and comparing this value with the pure radiative lifetime τ_{rad}, the quantum efficiency can easily be determined. Here, for investigation of the concentration quenching it is assumed that the low concentration lifetime

Table 1
Material parameters of the investigated laser crystals

	Yb:Lu_2O_3	Yb:YAG	Yb:KYW	Yb:LSB
Growth method	HEM	Cz	Flux	Cz
Temperature (°C)	2430	1930	1020	≈1600
Crucible	Re	Ir (Re)	Pt	Ir
Yb-distribution coefficient	0.9	1.0	≈1.0	≈1 (Sc 1.1, La 0.7)
Max. doping level (%)	⩽100	⩽100	⩽100	<30
Structure	Cubic	Cubic	Monoclinic	Monoclinic
Space group	Ia3—T_h^7	Ia3d—O_h^{10}	C2/c—C_{2h}^6	C2/c—C_{2h}^6
Heat conduct. ($W\,m^{-1}\,K^{-1}$)				
Undoped	12.5	11.0	3.3	2.8 || z
Yb-doped	11.0 (2.7%)	6.8 (5%)		
RE-density ($10^{21}\,cm^{-3}$)	29	14	6.4	5.1

corresponds to τ_{rad}, that is to 100% quantum efficiency.

At high doping concentrations the measured lifetimes are usually too long due to reabsorption of the emitted fluorescence. This effect is called "radiation trapping" and has to be eliminated by special measuring techniques. One technique is to dilute the powdered sample in an index matching fluid like ethylene glycol, thus eliminating the reabsorption. This was done in the case of Yb:KYW [3]. Another technique is to excite a bulk crystal or better a thin crystal plate through an aperture with various pinhole diameters and measure the fluorescence decay from the same side. The extrapolation of the lifetime data to zero diameter yields an accurate lifetime value without any radiation trapping. This method was successfully tested by measuring the same Yb:KYW crystals as used in Ref. [3]. For the 0.2% sample 232 μs and for the 10% sample 240 μs lifetimes have been obtained in good agreement with the lifetime of 233 μs for both crystals published in Ref. [3]. So, this new method seems to be reliable and all lifetime measurements presented in the paper have been performed in this way.

As already mentioned, fast energy migration occurs in all highly Yb-doped materials, which may reduce the fluorescence lifetime and thus the quantum efficiency considerably. This is demonstrated in Fig. 2 with Yb:YAG and Yb:Lu_2O_3. In Yb:YAG the lifetime stays nearly constant at 1040 μs up to about 15% Yb ($2.0 \times 10^{21}\,cm^{-3}$) and

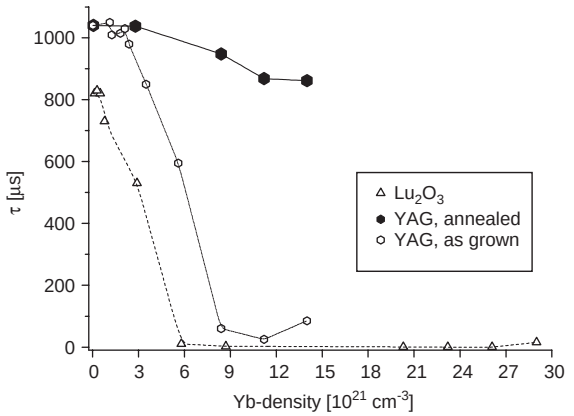

Fig. 2. Fluorescence lifetime of Yb:YAG and Yb:Lu_2O_3 versus Yb concentration.

then drops down to less than 50 μs. In Yb:Lu_2O_3 the situation is even worse. Here, the radiative lifetime of 820 μs is strongly quenched already at very low concentrations of 1–2% Yb ($0.5 \times 10^{21}\,cm^{-3}$) reaching 10 μs at 20% Yb. However, the lifetimes of the annealed Yb:YAG—samples (800–1000 °C in air, one day) drop only slightly from 1040 μs to 860 μs for the fully concentrated YbAG crystal. As was shown earlier [4] colour centres and Yb^{2+} ions created during growth of the crystals are removed by the annealing process. In the as-grown crystals these centres act as nonradiative acceptors besides additional impurities like transition-metal- or other rare-earth-ions.

The strong quenching behaviour of Yb:Lu_2O_3 and Yb:YAG can be correlated with the efficiency of the Yb–Yb energy transfer, which in the case of dipole–dipole interaction scales with the donor–donor distance R_{DD} according to

$$W_{DD} = C_{DD} \times R_{DD}^{-6}.$$

The minimum Yb–Yb distances for all investigated crystals and the R^{-6} values normalized to that of Yb:YAG are given in Table 2. Although the microparameters C_{DD} may differ somewhat for the different crystal lattices, it is obvious that the energy migration in Lu_2O_3 is much stronger than in YAG and much less in KYW and LSB.

The reduced energy migration in KYW is demonstrated by the nearly concentration-independent lifetime shown in Fig. 3 [3,5]. Up to 30% Yb-doping, the lifetime does not change and for the fully concentrated KYbW crystal only a small drop down to 200 μs is observed. From this point of view Yb:KYW is superior to Yb:Lu_2O_3 and Yb:YAG. Another positive effect is that, due to the slow energy migration, higher impurity concentrations can be tolerated in the grown crystal.

The same should be true for Yb:LSB, where the minimum La–La separation is even larger (see Table 2). The lifetime data of Yb:LSB are also given in Fig. 3. Because the monoclinic Huntite structure of LSB is very sensitive to the ionic radii of the constituents, crack-free crystals could only be grown up to a concentration of 25% Yb. The 30% crystal was already totally cracked. Surprisingly, the Yb lifetime increases strongly with the Yb concentration, which most probably is caused by the occupation of the two types of Sc sites (symmetry: distorted octahedra) as well as the La site (symmetry: distorted prism) by the Yb ions. The ionic radius of Yb^{3+} in sixfold oxygen coordination is $r_{Yb} = 0.9$ Å, which is in the middle of the La radius $r_{La} = 1.05$ Å and the Sc radius $r_{Sc} = 0.75$ Å. Therefore it can be expected, that the Yb ions are able to occupy both types of sites. Recent crystal growth experiments have shown, that at low Yb concentrations preferably the two types of Sc sites are occupied and at high concentrations both, the Sc sites and the La sites. By micro-probe analysis it was found, that at the maximum useful concentration (25% Yb) about 30% of the Yb ions are incorporated at the La site and 70% at the Sc sites. The Yb distribution coefficients at these high-concentrations were estimated to be κ(Yb/Sc) ~1.1 and κ(Yb/La) ~0.7; the total distribution coefficient was measured to be about unity.

The concentration dependence of the lifetime in Yb:LSB (Fig. 3) is unusual and not yet understood. Because of the different local symmetry of both, the Sc and La sites, the lifetimes should be different, too, resulting in a nonexponential fluorescence decay behaviour. This was already experimentally observed. Furthermore,

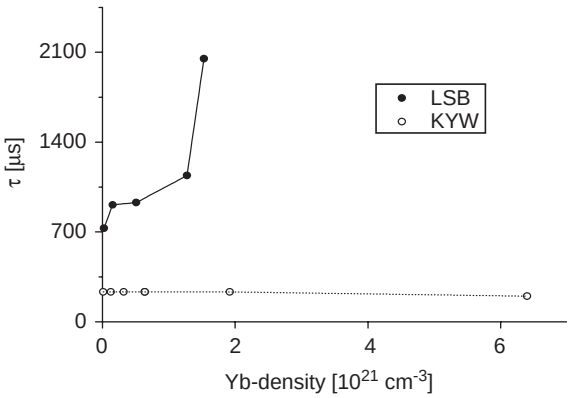

Fig. 3. Fluorescence lifetime of Yb:KYW and Yb:LSB versus Yb concentration; for LSB the Yb concentrations are given only for the La sites.

Table 2
Laser relevant data of the investigated crystals

	Structure	RYb–Yb (Å)	Migration (R^{-6}) (%)	σ_{em} (cm^2)	λ_{em} (nm)	τ_{rad} (μs)
Yb:Lu_2O_3	Cubic	3.43	153	13×10^{-21}	1032	820
Yb:YAG	Cubic	3.68	100	19×10^{-21}	1030	1040
Yb:KYW	Monoclinic	4.75	21	30×10^{-21}	1026	232
Yb:LSB	Monoclinic	6.06 (La)	5	1.1×10^{-21}	1045	730

the concentration dependent site occupancy makes it very difficult to separate the effect of the reduced energy migration since La–La, La–Sc, and Sc–Sc energy transfer will also happen. Because the minimum Sc–Sc-separation is 3.3 Å in contrast to the La–La separation of 6.06 Å, reduced energy migration can only be expected for the Yb ions on the La sites and it raises the question, which Yb ion is the emitting ion. Presently, detailed spectroscopic measurements are performed at low temperatures to analyse the energy transfer between the different Yb sites and to attribute the measured lifetimes to the three available lattice sites. Indeed, first results indicate that in highly concentrated crystals the fluorescence arises from the Yb ions on the La sites and thus a very high quantum efficiency is expected.

Although the spectroscopic properties of Yb:LSB are not yet completely investigated, first laser experiments have shown that Yb:LSB is a very efficient laser material. This will be demonstrated in Section 2.3.

2.3. Laser experiments

Till now, thin-disc laser experiments have been performed with the crystals Yb(9.3%):YAG, Yb(2.6%):KYW, and Yb(2.7%):Lu_2O_3 (the concentrations are Yb/RE site). The experimental setup is described for example in Ref. [6]. In Fig. 4 the input–output curves of Yb:YAG [7] and Yb:KYW [8] are displayed together with the achieved optical efficiencies and the corresponding simulation calculations. For these two laser systems, a quantum efficiency of 100% had to be used in the simulations. This fact coincides with the lifetime measurements shown in Fig. 2 (YAG) and Fig. 3 (KYW), where at these rather low Yb concentrations only a very small quenching effect is observed with Yb:YAG. So, the somewhat higher slope and optical efficiency of Yb:KYW in comparison to Yb:YAG (despite the lower heat conductivity, the lower doping level, and the higher resonator losses) may be interpreted to be due to a slight (a few %) reduction of the quantum efficiency in Yb:YAG.

With Yb:Lu_2O_3 the situation is more obvious. A rather good fit of the input–output curve in Fig. 5 is only possible if a reduced quantum efficiency between 85% and 90% is assumed. This value matches very well the ratio of the measured lifetime of 730 μs for 2.7% Yb doping and the radiative lifetime of 820 μs (low concentration value). The maximum optical efficiency obtained so far is $\eta_{opt} = 34\%$.

With Yb:LSB, laser experiments have also been performed for the first time in a linear resonator under TiSa pumping. The measured input–output curve for a 10% Yb-doped crystal is shown in Fig. 6. Although the slope efficiency of $\eta = 64\%$

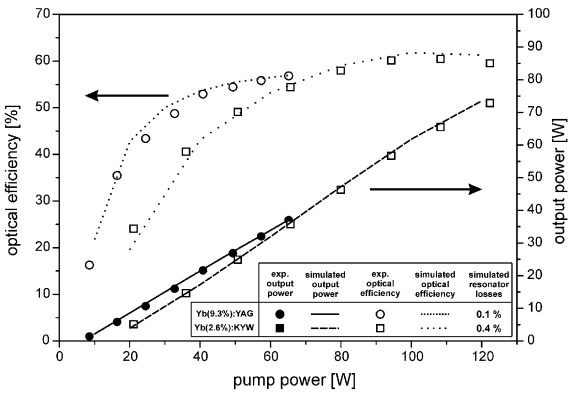

Fig. 4. Input–output curves and optical efficiencies of Yb:YAG and Yb:KYW; the full and dashed lines are calculated curves and the full and open symbols are experimental data.

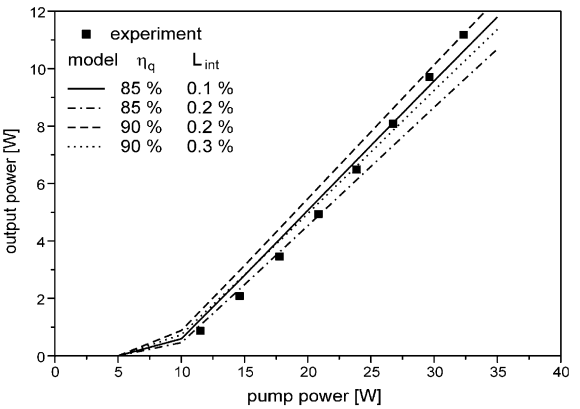

Fig. 5. Input–output curve of Yb:Lu_2O_3 and corresponding simulation calculations; the best fit is obtained with 85–90% quantum efficiency.

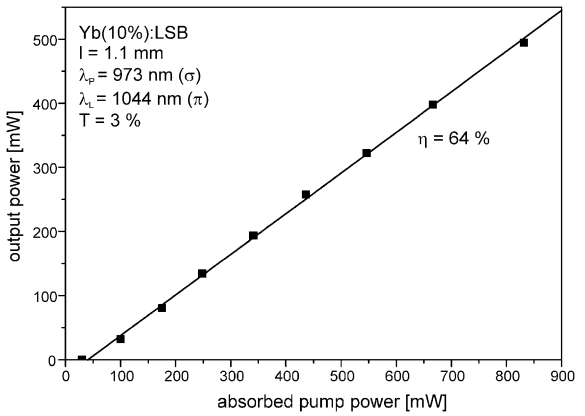

Fig. 6. Input–output curve of Yb:LSB in a linear resonator under TiSa pumping.

cannot be compared directly with the optical efficiencies of the other laser crystals due to the different laser resonators and pumping schemes, this high value for a new laser material is quite promising and points again to a very high quantum efficiency of Yb:LSB. Thin-disc laser experiments are planned for the near future.

3. Summary

The fluorescence lifetime quenching in the investigated laser crystals Yb:Lu_2O_3, Yb:YAG, and Yb:KYW is caused by the migration of excitation energy between the Yb ions and finally the transfer to certain impurities. This effect is strongest in Yb:Lu_2O_3, where the Yb–Yb distance is shortest, and considerably smaller in Yb:KYW. In Yb:LSB the concentration quenching might also be reduced, because the energy migration between the widely separated La sites must be very slow. However, detailed spectroscopic investigations are still necessary and are right now performed to clear up the complicated energy transfer mechanisms in LSB.

Although it is difficult to compare precisely the results of the thin-disc laser experiments with the different laser materials due to the many parameters influencing the performance of the lasers, the same general trend can be observed as with the lifetime data: the highest optical efficiency till now was obtained with Yb:KYW and the lowest with Yb:Lu_2O_3 despite the fact that the heat conductivity in Lu_2O_3 is a factor of four higher than in KYW. However, the emission cross-section is larger in Yb:KYW providing a higher gain in this laser material.

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