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Luminescent and lasing characteristics of heavily doped Yb^{3+} : $KY(WO_4)_2$ crystals

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***Abstract*. The luminescence decay times are measured taking into account reabsorption for $KY(WO_4)_2$: Yb (KYW : Yb) crystals with atomic concentrations of active ions from 0.2 % to 30 %. The radiative lifetime of Yb^{3+} ions was measured to be 233 μs. The cw output power of 1.46 and 1.62 W was achieved with the slope efficiency 52 % and 47 % for Yb : KYW lasers with the atomic concentration of Yb^{3+} ions equal to 10 % and 30 %, respectively. Using a semiconductor mirror with a saturable absorber (SESAM) in the passive mode-locking regime, pulses of duration 194 and 180 fs were obtained at wavelengths of 1042 and 1039 nm for crystals with Yb^{3+} concentrations equal to 10 % and 30 %, respectively, the average output power being 0.63 and 0.75 W.**

Keywords: *solid-state laser, luminescence, highly concentrated active media.*

1. Introduction

Tungstate crystals Yb^{3+} : $KY(WO_4)_2$ (Yb : KYW) and Yb^{3+} : $KGd(WO_4)_2$ (Yb : KGW) doped with Yb^{3+} ions attract great recent interest as active media for diode-pumped lasers emitting in the 1-μm spectral region [1, 2]. The advantages of these materials are the intense absorption band at 980 nm which allows their pumping with commercial InGaAs laser diodes; a broad gain band comparable with that of Yb^{3+} ions in glasses, which is required for laser tuning and generating ultrashort pulses; high stimulated-emission cross sections; and a small quantum defect (the difference between the energies of absorbed and laser photons) equal to 4 % – 5 %, which provides low thermal losses.

The pulse duration and the average output power of the first femtosecond Yb : KGW laser were 176 fs and 1.1 W, respectively [3]. For the output power of 0.2 W, the pulse duration of this laser was even shorter (112 fs). An Yb : KYW laser pumped by a high-quality 2-W beam from a diode laser produced 101-fs pulses with the average power of 100 mW [4]. A femtosecond Yb : KYW laser with the 35 % efficiency was studied in [5]. The average output power of a thin disc Yb : KYW laser a emitting 240-fs pulses was 22 W [6]. Diode-pumped Kerr mode-locked Yb : KYW lasers were described in papers [7, 8]. The shortest pulse duration for these lasers was 71 fs for the average output power of 120 mW [7]. The atomic concentration of Yb^{3+} ions in all these cases did not exceed 10 %.

For high-power thin disc femtosecond lasers, the Yb : KYW crystals are of greater interest than Yb : KGW because they have a better optical quality at high concentration of Yb^{3+} ions {up to 100 % in $KYb^{3+}(WO_4)_2$ (KYbW) [9] crystals}. The use of such crystals in a thin-disc laser [10] allows one to improve the cooling efficiency and simplify the pump scheme. The latter considerably alleviates the requirements to the pump radiation quality. Therefore, the study of the laser properties of heavily doped Yb : KYW crystals for obtaining lasing, in particular, emission of ultrashort pulses is of considerable interest.

Radiation losses in heavily doped crystals can be caused by the concentration quenching of fluorescence. The lifetime of the upper laser level of Yb^{3+} measured at room temperature is of about 200 μs for KYbW [9] and 300 – 400 μs for Yb : KYW (1 % – 20 %) [11]. It is well known that reabsorption can strongly affect the measured lifetime for ytterbium-doped materials because of a strong overlap of the absorption and emission bands [12]. This is especially important to take into account for tungstate crystals doped with ytterbium ions, where the bands are overlapped much stronger than, for example, in Yb : YAG. The relatively high refractive index of KYW ($n \approx 2.0$) also increases reabsorption even in optically thin samples due to the total internal reflection. Therefore, to exclude radiation reabsorption for accurate measurements of luminescence lifetimes in such materials, especially in crystals heavily doped with Yb^{3+} ions, special methods should be used, which are described in papers [9, 12].

In this paper, we measured the luminescence lifetimes of Yb^{3+} ions in single crystals and finely dispersed Yb : KYW powders in ethylene glycol. Powders were used to avoid reabsorption, and ethylene glycol – to equalise the refractive indices for reducing the effect of total internal reflection. We

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also studied cw lasing and passive mode locking for Yb : KYW crystals at concentrations of Yb^{3+} ions 10 % and 30 %. The aim of our experiments was to compare the laser characteristics of heavily doped Yb : KYW crystals with those of widely used materials with the concentration of active ions equal to 10 %, which is necessary for studying possible negative effects and restrictions caused by the high concentration of Yb^{3+} ions.

2. Measurements of the luminescence lifetime

The scheme of the experimental setup is shown in Fig. 1. Luminescence was excited by ~ 20-ns, 981-nm pulses from an optical parametric oscillator which was pumped by the third harmonic of a *Q*-switched Nd : YAG laser. Luminescence was collected from a sample surface irradiated by laser pulses, passed through a monochromator, and was detected with a germanium photodiode with a time constant of 500 ns and a digital oscilloscope with the 500-MHz pass band. The results were averaged over 20 measurements for each sample, the measurement error being no more than 5 %.

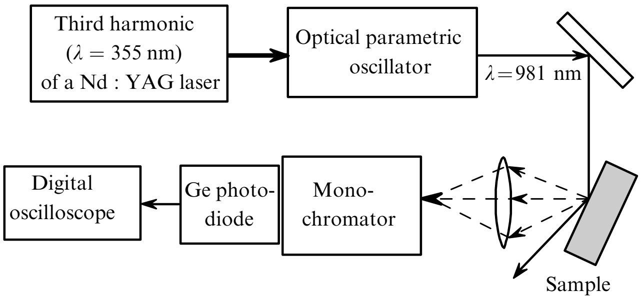

Figure 1. Scheme of the experimental setup for measuring the luminescence lifetime of Yb : KYW crystals.

Yb : KYW single crystals were grown by the modified Czochralski method. We studied samples with the concentrations of ytterbium ions 0.2 %, 2 %, 5 %, 10 %, and 30 %. All the samples, except the crystal with the ytterbium concentration 0.2 %, were prepared in the form of a powder suspension in liquid ethylene glycol. Powders were prepared by grinding crystals in a mortar. The diameter of powder particles did not exceed 50 µm, which is smaller than the absorption length (at the 1/e level) even for crystals with the concentration of ytterbium ions equal to 30 % (about 70 µm at $\lambda = 981$ nm). The use of ethylene glycol increased the angle of total internal reflection for a KYW crystal up to 44° compared to 30° in air. In addition, we measured powders and crystals in air, including thin (70 – 150 µm) plates.

The luminescence decay in all the samples occurred exponentially. The data presented in Figs 2 and 3 show that the lifetime of Yb^{3+} ions considerably decreases with decreasing the ytterbium concentration in the crystal (Fig. 2) or the weight concentration of Yb : KYW powders in the suspension (Fig. 3). This suggest that the influence of reabsorption is strong. The luminescence lifetime of an Yb : KYW sample (0.2 %) in ethylene glycol was measured to be 233 µs. We assume that this lifetime corresponds to the radiative lifetime of Yb^{3+} in KYW.

The luminescence lifetimes for suspensions of powders in ethylene glycol as functions of the weight concentration of

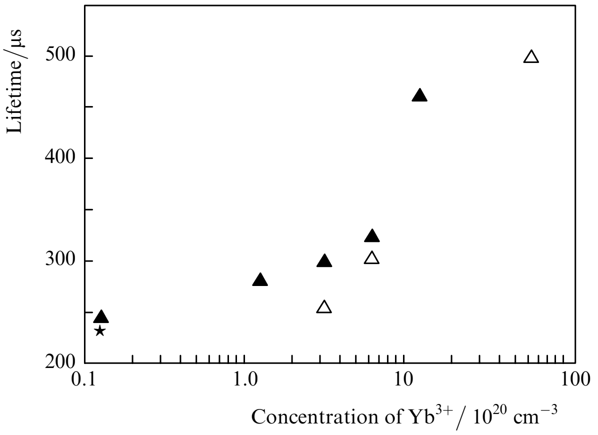

Figure 2. Luminescence lifetime for 3-mm (▲) and 70-µm thick (△) Yb : KYW crystals and the Yb : KYW crystal powder (0.2 %) in ethylene glycol (★).

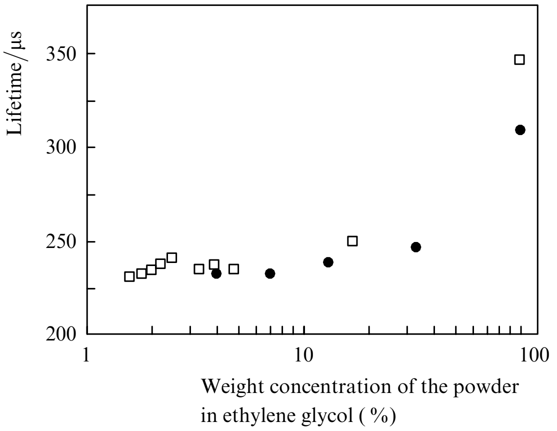

Figure 3. Dependences of the luminescence lifetime of the Yb : KYW (10 %) (●) and Yb : KYW (30 %) (□) crystal powders in ethylene glycol on the weight concentration of the powder in suspension.

Table 1. Luminescence lifetimes measured in Yb : KYW crystals.

Material	Weight concentration of a powder in ethylene glycol (%)	Lifetime/µs
Yb : KYW (30 %)	– (crystal in air)	460
	100 (powder in air)	347
	17	250
	4.8	235
	3.9	237
	3.3	235
	2.5	237
	2.2	235
	2	231
	1.8	234
Yb : KYW (10 %)	– (crystal in air)	327
	100 (powder in air)	309
	33	247
	13	239
	7	232
	4	234
Yb : KYW (0.2 %)	– (crystal in air)	272
	– (crystal in ethylene glycol)	233

Yb : KYW are presented in Fig. 3 and Table 1. The dilution of suspensions equivalent to decreasing the average concentration of Yb^{3+} makes it possible to obtain the limiting luminescence lifetime. This lifetime for all powder samples (2 %, 5 %, 10 %, and 30 %) and for the unground sample with the concentration of ytterbium ions equal to 0.2 % in ethylene glycol was about 233 μs. Thus, no concentration quenching of luminescence was observed even for Yb : KYW crystals with the doping level up to 30 %.

3. Laser experiments

Laser experiments were performed in a resonator shown schematically in Fig. 4. The active elements were Yb : KYW crystals of thickness 2 and 0.8 mm with the ytterbium concentration 10 % and 30 %, respectively, which were oriented at the Brewster angle. The absorption spectrum for the $E||N_m$ polarisation (N_m, N_g, and N_p are the axes of the optical indicatrix) is shown in Fig. 5. The crystals were pumped longitudinally along the crystallographic axis b (N_p) by a 984-nm cw laser diode with a fibre pigtail (the fibre core diameter was 100 μm, NA = 0.22, and the power at the fibre output was 7 W). Because the pump and laser wavelength were close to each other, the transmission of the input mirror at the pump wavelength did not exceed 75 % and the maximum radiation power incident on the crystal was 5 W. The pump radiation was focused into a spot of diameter 110 μm. Because the active crystal was oriented at the Brewster angle, the pump radiation spot on it had the elliptic shape with semiaxes 110 and 220 μm. The waist size of the TEM_{00} mode of the resonator in the active element was 105×210 μm. The active elements were mounted on a copper heat sink, whose temperature was kept at 10 °C.

The passive mode locking was achieved by using a semiconductor saturable absorber mirror (SESAM [13, 14]). The modulation depth of this SESAM was about 1 %, the saturation energy density was 350 mJ cm^{-2}, and the relaxation time was 100 ns. To provide the negative group-velocity dispersion required for obtaining soliton pulses, a pair of SF10 glass prisms [15] separated by a distance of $\sim$ 48 cm was used [15]. This ensured the group-velocity dispersion of the order of -3000 fs^2 per round trip in the resonator. A stable mode-locking regime was obtained for both crystals when the transmission of the output mirror was 1.6 %.

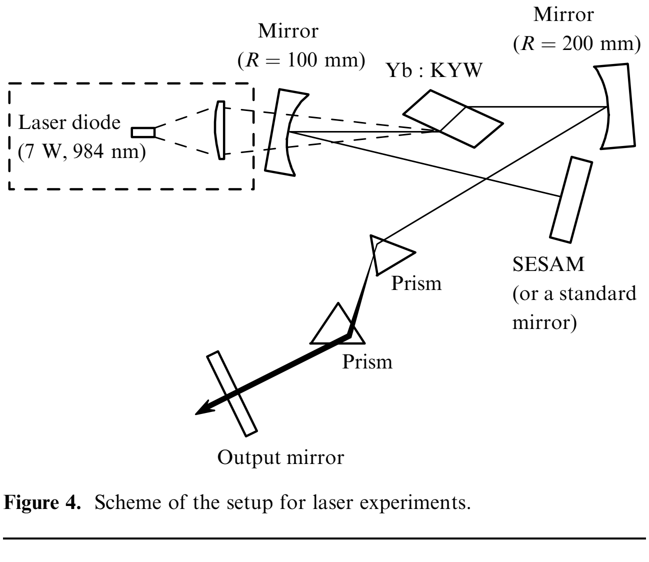

Figure 4. Scheme of the setup for laser experiments.

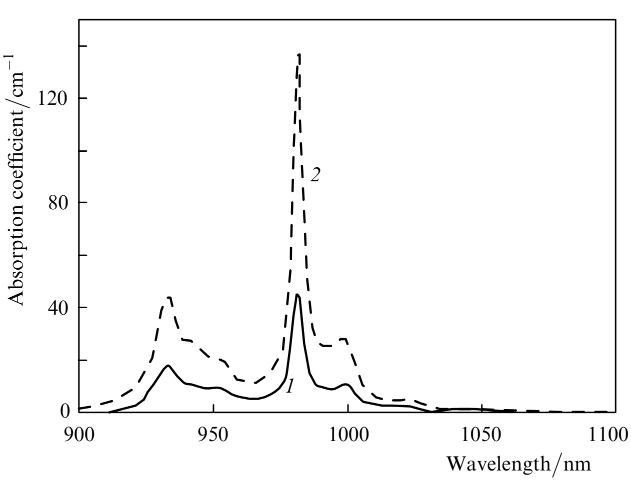

Figure 5. Absorption spectra of the Yb : KYW (10 %) (*1*) and Yb : KYW (30 %) (*2*) crystals for the $E||N_m$ polarisation.

The autocorrelation function and the output spectrum of the Yb : KYW laser are presented in Fig. 6. The average output power of the Yb : KYW laser (30 %) emitting 180-fs, 1040-nm pulses was 0.75 W (Figs 6a, b). The laser pulses were close to transform-limited pulses with the product $\Delta\nu\Delta\tau = 0.320$, the pulse repetition rate was 155 MHz, and the pulse peak power was 23 kW. The average output power of the Yb(10%) : KYW laser emitting 197-fs, 1037-nm, 18-kW pulses was 0.63 W.

The mode-locked laser was tuned with a 'knife' mounted near the output mirror inside the resonator. The tuning range for a crystal with the concentration of ytterbium ions equal to 10 % was 1030 – 1050 nm. A somewhat narrower tuning range (1035 – 1053 nm) was obtained when the concentration of ytterbium ions was 30 %.

In the configuration intended for obtaining cw lasing, i.e., with a highly reflecting mirror instead of SESAM and without intracavity prisms, the output mirror with the transmission of 4.5 % was used. The maximum output power was 1.46 W at 1040 nm and 1.62 W at 1050 nm with the slope efficiency with respect to the absorbed pump power equal to 52 % and 47 % for Yb : KYW (10 %) and Yb : KYW (30 %) crystals, respectively.

The output characteristics of Yb : KYW lasers operating in the cw regime are presented in Fig. 7. Because of a comparatively small difference (60 – 70 nm) between the pump and laser wavelengths, the losses on a mirror with $R = 100$ mm were about 1 % at the laser wavelength. This is one of the reasons for a lower cw lasing efficiency in both crystals compared to that obtained in [3, 8].

Another reason is the non-optimal overlap of the pump beam (the beam quality is $M^2 \approx 18$) and the TEM_{00} mode of the resonator, because for the beam-waist diameter equal to 110 μm the Rayleigh length (double distance at which the beam diameter increases by a factor of $\sqrt{2}$ compared to the waist diameter) was smaller than the crystal thickness, being about 1.8 mm. A lower lasing efficiency at the ytterbium ion concentration equal to 30 % (compared to that for the concentration of 10 %) can be partially explained by a stronger local heat release and by a decrease in the cooling efficiency because heat removal was performed through thin faces of the active element.

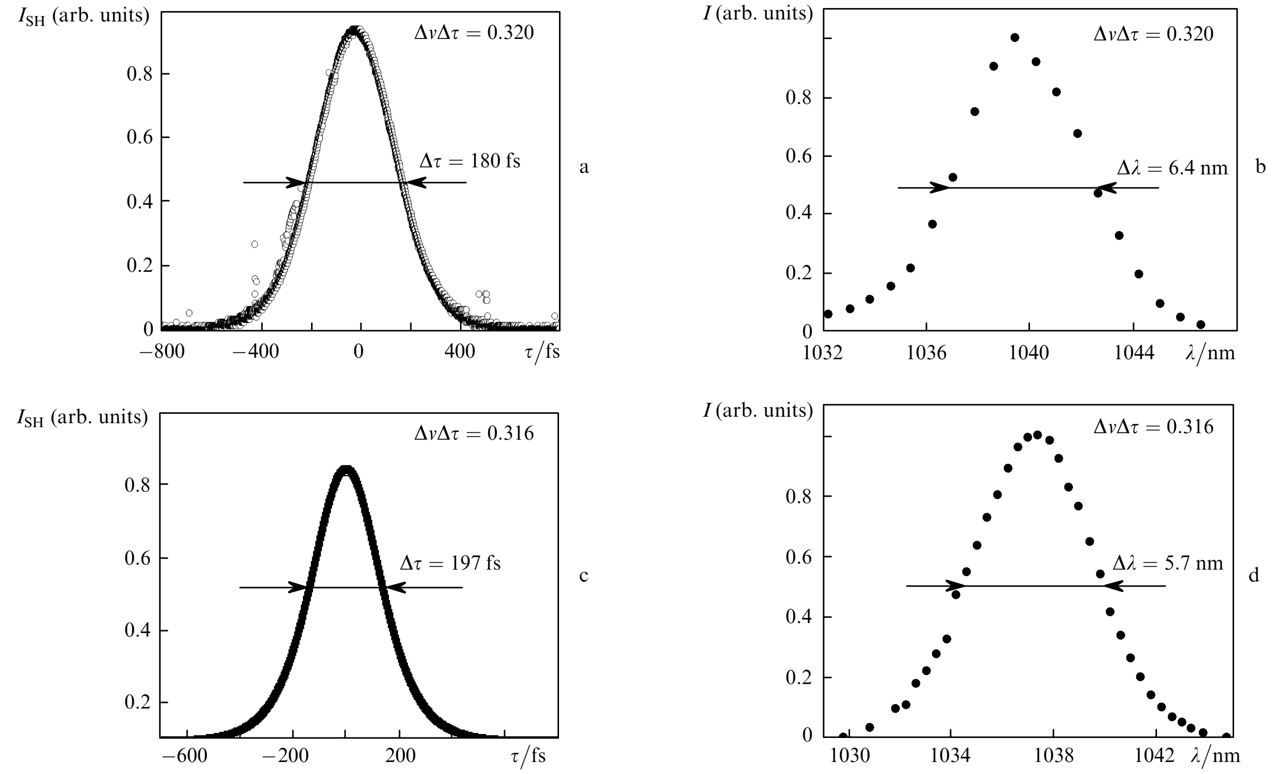

Figure 6. Experimental (points) and calculated (curves) autocorrelation functions (a, c) and emission spectra (b, d) of Yb : KYW lasers with the concentration of ytterbium ions 30 % (a, b) and 10 % (c, d); I_{SH} is the second harmonic intensity; I is the spectral intensity.

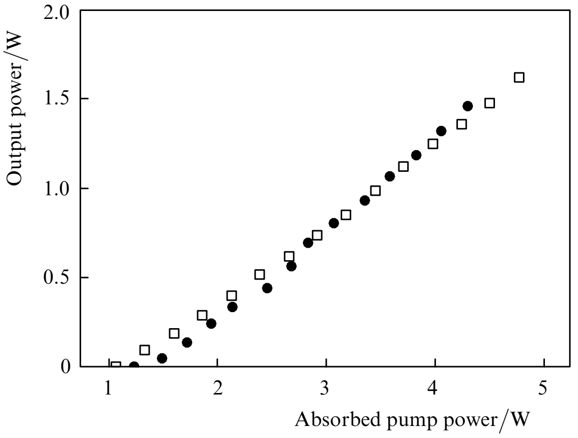

Figure 7. Output characteristics of the Yb : KYW (10 %) (●) and Yb:KYW (30 %) (□) crystal lasers operating in the cw regime at the slope lasing efficiency with respect to the absorbed pump power equal to 52 % (●) and 47 % (□).

The results obtained for cw and mode-locked lasing are presented in Table 2. The pulse durations measured in our experiments exceed those reported in the literature because the resonator parameters (including the dispersion compensation) were not optimised. Nevertheless, these results demonstrate that no considerable deterioration of the lasing parameters of the Yb : KYW laser caused by a high concentration of Yb^{3+} ion was observed both in the cw and mode-locking regimes.

Table 2. Results of laser experiments with Yb : KYW crystals with different concentrations of Yb^{3+} ions.

C (%)	P_1/W	η (%)	P_2/W	τ/fs	f/MHz	$\Delta\lambda$/nm
10	1.46	52	0.63	197	155	1030–1050
30	1.62	47	0.75	180	155	1035–1053

Notes: C is the atomic concentration of Yb ions in KYW; P_1 and P_2 are the average output powers in the cw and mode-locked regimes; η is the lasing efficiency with respect to the absorbed pump power; τ is the pulse duration; f is the pulse repetition rate; $\Delta\lambda$ is the tuning range.

4. Conclusions

We have measured the lifetimes of Yb^{3+} ions in Yb : KYW crystals with different concentrations of ytterbium ions by using crystal powder suspensions in ethylene glycol to suppress reabsorption and total internal reflection of radiation. The lifetime was measured to be 233 μs for the atomic concentration of Yb^{3+} up to 30 %. CW and passively mode-locked lasing was obtained in the Yb : KYW crystal (30 %) pumped by a diode laser. The maximum output cw power was 1.62 W with the slope efficiency with respect to the absorbed pump power equal to 47 %. Laser pulses of duration 180 fs with the average power 0.75 W were obtained in the mode-locked regime. These results show that the increase in the concentration of Yb^{3+} ions up to 30 % does not cause the concentration quenching of luminescence and almost does not affect the lasing characteristics of the Yb : KYW crystal.

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