Spectroscopy and femtosecond laser performance of Yb$^{3+}$:YAlO$_3$ crystal

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Present widespread use of efficient InGaAs laser diodes and favorable electronic structure of ytterbium ions stimulate interest in Yb$^{3+}$:doped solid-state materials for diode-pumped high-power lasers emitting in the spectral range near 1 µm. Broad gain bandwidth of these media has stimulated their applications for generation of ultrashort pulses in mode-locked regime.

So far, the highest average output power of 60 W with ultrashort pulses has been obtained with a thin-disk Yb:YAG laser, passively mode-locked with a semiconductor saturable-absorber mirror (SESAM) [1]. The thermal conductivity of YAG is about 13 W/(mK). However, the emission bandwidth of Yb:YAG limits the pulse duration to only 700–800 fs in a thin-disk laser, or to 340 fs in a bulk laser [2]. Much shorter pulse durations were reported for a number of crystals with broader emission bands, such as Yb-doped YVO$_4$, CaGdAlO$_4$, Y$_2$SiO$_5$, KGd(WO$_4$)$_2$, KY(WO$_4$)$_2$, KLu(WO$_4$)$_2$, SrY$_2$(SiO$_4$)$_3$O, CaGa$_2$O$_4$(BO$_3$)$_3$, Sr$_2$Y(BO$_3$)$_3$, etc. [3–12], as well as phosphate and silicate glasses [13]. However, these materials have low thermal conductivity of approximately 2–5 W/(mK), which limits their potential for high-power operation. With Yb-doped CaF$_2$, which has thermal conductivity of 9.7 W/(mK), average power up to 880 mW with pulses as short as 150 fs was reported [14]; however, a rather low absorption cross-section of this crystal makes it not suitable for thin-disk lasers. Sesquioxides, such as Lu$_2$O$_3$ and Sc$_2$O$_3$, that have high thermal conductivity of more than 12 W/(mK) are particularly promising materials to date. 370 fs pulses with 20.5 W average power with a mode-locked Yb:Lu$_2$O$_3$ thin-disk laser were recently demonstrated [15]. Here we publish results on spectroscopy and efficient cw and mode-locked laser operation of a Yb$^{3+}$:YAlO$_3$ single crystal as promising material for thin-disk laser applications.

Perovskite-like yttrium aluminate, YAlO$_3$ (YAP), is a biaxial crystal with the orthorhombic $P_{2b}$ space group. It has high thermal conductivity of 11 W/(mK) [16]. Some spectroscopic properties as well as an energy-level diagram of Yb:YAP crystals with Yb$^{3+}$ concentration of 2 at. % were reported already in 1971 [17]. Unpolarized absorption and emission spectra of ytterbium-doped yttrium aluminate and comparison with other ytterbium-doped materials were made in [18]. More recently, comparison of spectroscopic parameters of Yb$^{3+}$:doped YAP and YAG single crystals was carried out, and self-absorption phenomena was studied [19,20]. It was shown that Yb$^{3+}$(15 at. %):YAP crystal is a potential candidate for compact, efficient thin-chip lasers; in addition, its broad emission band allows the generation of ultrashort pulses. The latest description and analysis of Yb:YAP crystal growth, structural characterization, and spectroscopic properties were published in [21]. In this study, Boulon et al. reported experimental decay-time dependence on Yb$^{3+}$-dopant concentrations, and room-temperature absorption and emission cross section spectra along the a axis were given. Radiative lifetime of an upper-laser level was estimated there to be 0.6 ms, which has a large discrepancy with the decay time published in [19].

In our study polarized absorption spectra of Yb$^{3+}$(0.6 at. %):YAP at room temperature were measured by a Varian Cary-5000 spectrophotometer. The content of ytterbium ions in crystal was measured by means of a Tescan VEGA II LMU scanning electron microscope with Oxford INCA Energy 350 energy dispersive x-ray analyzer and was estimated to be $1.17 \times 10^{20}$ cm$^{-3}$. The cross-section spectra for three polarizations are shown in Fig. 1. Strong absorption is found for E//c light polarization. A peak absorption cross-section at 978.2 nm is about $6.6 \times 10^{-20}$ cm$^2$, however, with comparatively narrow bandwidth of about 4 nm. For a and b polarizations maximal absorption cross sections near 978.5 nm are $0.97 \times 10^{-20}$ cm$^2$ and $1.38 \times 10^{-20}$ cm$^2$, respectively. The stimulated emission cross sections are also given in Fig. 1. They were calculated by use of a modified
reciprocity method [22]. Lifetime measurements were carried out with fine powdered Yb/H2O8490.6 at.%/H2O850:YAP crystal immersed in glycerol in order to suppress reabsorption [23]. The sample was excited by about 20 ns pulses at 982 nm. The emitted luminescence was measured with the use of a 0.3 m monochromator, fast Ge-photodiode with a rise time of <20 ns and a 500 MHz digital storage oscilloscope. All the samples exhibited single exponential decay (see inset in Fig. 2). With the decrease of weight content of crystalline powder in suspension measured lifetime also decreased (see Fig. 2). After considerable dilution of the powder and thus elimination of self-trapping effect the emission lifetime of ytterbium excited state in YAP crystal was determined to be 500±10 μs. This value is even smaller than the results published in [21] (0.6 μs), which is to our belief owing to more careful reduction of reabsorption influence.

For a cw laser, experiments set up with a folded cavity design were used. It consisted of two curved mirrors with 100 mm and 300 mm radii of curvature, a plane output coupler (OC), and a plane end mirror highly reflecting at 1020–1100 nm. For the experimental assessment a 3-mm-long 0.6 at.% Yb:YAP Brewster-angled crystal cut perpendicular to the crystallographic a axis was used and was oriented in the cavity for polarization parallel to the b axis, where the stimulated emission cross section is slightly higher than that for c polarization (Fig. 1). For heat dissipation a gain medium was mounted onto a copper heat sink kept at 20°C. A cw fiber-coupled (5=105 μm, NA=0.15) InGaAs laser diode with a maximum output power of about 6 W at 980 nm wavelength was used for longitudinal pumping of the gain medium through the mirror with 100 mm radius of curvature. The pump beam was focused inside a Yb:YAP laser crystal into a 105 μm spot with a confocal length of about 2.5 mm. The cavity-mode diameter for TEM00 transversal mode at the active element was calculated to be 98 μm.

Input-output diagrams for the Yb:YAlO3 CW laser with OC transmissions of 3.5% and 5.5% are presented in Fig. 3. Maximum output power of about 1.2 W at 1040 nm for TEM00 mode with a slope efficiency of 64.5% with respect to the absorbed pump power was obtained with 5.5% OC. For the laser with OC transmission of 3.5% the slope efficiency decreased to 58% with output power of 1.12 W at 1041 nm. Laser thresholds for the 3.5% and 5.5% output couplers were determined to be 0.85 W and 1.05 W of the absorbed pump power, respectively.

For the experiments in a mode-locked regime the same cavity design was used with a SESAM having modulation depth of 1.6% and saturation fluence of 30 μJ/cm² substituted for a high-reflecting mirror and two chirped mirrors, each providing second-order dispersion of nearly −920 fs² in the spectral range from 1030 to 1065 nm, for obtaining negative group-delay dispersion in the cavity (Fig. 4). Stable cw mode locking was obtained for OC transmission of 3.5%. Average output power up to 0.8 W at 1041 nm with pulse duration of 225 fs and pulse repetition

Fig. 1. Polarized absorption and stimulated emission spectra of Yb(0.6 at.%) :YAlO3 crystal.

Fig. 2. Measured lifetime for different weight content of Yb:YAP crystalline powder in glycerol suspension. Inset, kinetics of luminescence decay.

Fig. 3. Average output power of cw Yb:YAP laser versus absorbed pump power for output couplers with different transmissions.
Fig. 4. Experimental setup of passively mode-locked diode-pumped Yb$^{3+}$:YAP laser.

Fig. 5. Intensity autocorrelation (left) with a sech$^2$-fit and optical spectrum (right) of the mode-locked Yb:YAlO$_3$ laser.

rate of 70 MHz occurred, resulting in peak power as high as 44.5 kW. Intensity autocorrelation with a sech$^2$ fit and optical spectrum of the mode-locked Yb:YAP laser are presented in Fig. 5. During autocorrelator scanning a time range of 130 ps occurred after laser pulse, and no other pulses were observed. They were not also seen on the oscilloscope with time resolution less than 1 ns. The time-bandwidth product was estimated to be 0.32, which was very close to the transform limit for soliton pulses ($\tau_c\Delta\nu=0.315$) [24].

In conclusion, for the first time, to our knowledge, efficient cw and mode-locked laser operation with Yb$^{3+}$:YAlO$_3$ single crystal was demonstrated. Polarized absorption and stimulated emission cross-section spectra and luminescence lifetime of Yb:YAP were determined. The cw operation of a Yb:YAP laser at 1040 nm with 1.2 W output power and slope efficiency as high as 64% with respect to the absorbed pump power were achieved. In a mode-locked regime soliton-like pulses with duration of 225 fs and average power as high as 0.8 W around 1041 nm were realized. We believe that the increase of output power and pulse shortening could be achieved by optimization of Yb-dopant concentration and group-velocity dispersion compensation in the cavity. It is prospective to grow samples with higher Yb ions doping concentration and find out the maximum doping level for YAP. Yb:YAlO$_3$ looks very promising for thin-disk ultrafast lasers owing to high thermal conductivity, strong absorption at 985 nm, and comparatively broad emission bandwidth. In the future it also is interesting to measure its nonlinear refractive index and determine the possibility of using Yb:YAP for Kerr-lens mode-locked ultrashort Yb lasers.

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