

Compact 999.6 nm Actively Q-Switched Yb³⁺:LuAlO₃ Laser for Laser-Induced Breakdown Spectroscopy

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Abstract

Compact actively Q-switched diode-pumped lasers based on Yb³⁺-materials are of practical importance for wide range of scientific, industrial and biomedical applications. The aim of this work was to study the Yb³⁺:LuAlO₃ actively Q-switched laser.

One of the most promising crystalline hosts for rare-earth ions are Perovskite-like aluminate crystals. Yttrium aluminate crystal YAlO₃ (YAP) is a well-known host with good thermal and mechanical properties (thermal conductivity for undoped crystal is about 11 W/m·K and about 8 W/m·K for Yb(5 at.%):YAP) similar to those of YAG. The reduction in the thermal conductivity of doped laser crystal in comparison with host materials is small in the case of ions with close atomic mass and ionic radii such as for Yb³⁺ and Lu³⁺. This feature makes LuAlO₃ (LuAP) more promising host crystal for doping by Yb³⁺ ions in contrast to YAP especially for high output power laser systems.

In our work, for the first time to the best of our knowledge actively Q-switching laser operation of Yb³⁺:LuAP single crystal was demonstrated. The maximum average output power of 4.9 W at 50 kHz pulse repetition frequency (PRF) with opt.-to-opt. efficiency of 21 % was obtained with 30 % OC transmittance. Output power as high as 3.3 W with 333 μJ-laser pulses with duration of about 11.5 ns was demonstrated at 10 kHz PRF the corresponding pulse peak power was 29 kW. 97 μJ second harmonic pulses obtained with 29 % conversion efficiency at 10 kHz PRF.

Performed investigations show high potential of Yb³⁺:LuAP crystals as active elements of compact diode pumped actively Q-switched lasers due to high stimulated emission cross-section ($\approx 3.74 \cdot 10^{-20}$ cm²) at 999.6 nm wavelength and significant reduction of heat load on the active element when pumping around 980 nm and generation around 999 nm.

Keywords: Q-switched laser, ytterbium ions, diode pumping, lutetium aluminate crystals, second harmonic generation.

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Компактный лазер на кристалле $\text{Yb}^{3+}:\text{LuAlO}_3$ с активной модуляцией добротности резонатора, излучающий на длине волны 999,6 нм для применения в лазерно-искровой эмиссионной спектроскопии

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Компактные лазеры с активной модуляцией добротности резонатора, построенные на материалах, легированных ионами Yb^{3+} , представляют практический интерес для широкого ряда научных, промышленных и биомедицинских применений. Целью данной работы являлось исследование режима активной модуляции добротности резонатора лазера на кристалле $\text{Yb}^{3+}:\text{LuAlO}_3$.

Одними из наиболее перспективных матриц для легирования ионами редкоземельных элементов являются кристаллы алюминатов со структурой перовскита. Кристаллы иттриевого алюмината YAlO_3 (YAP) широко известны в качестве матриц благодаря хорошим термомеханическим свойствам (теплопроводность нелегированного кристалла около 11 Вт/м·К и около 8 Вт/м·К для $\text{Yb}(5 \text{ ат.}\%):\text{YAP}$), близким к кристаллам YAG. Снижение теплопроводности лазерного кристалла при легировании по сравнению с чистой матрицей мало в случае незначительно отличающихся атомных масс и ионных радиусов как в случае с ионами Yb^{3+} и Lu^{3+} . Данная особенность делает кристалл LuAlO_3 (LuAP) значительно более перспективной матрицей для ионов Yb^{3+} по сравнению с YAP особенно в случае лазерных систем с высокой средней выходной мощностью.

Режим активной модуляции добротности лазера на кристалле $\text{Yb}^{3+}:\text{LuAP}$ исследован впервые в нашей работе. Максимальная средняя выходная мощность 4,9 Вт получена при частоте следования импульсов 50 кГц и оптической эффективности 21 % с применением выходного зеркала пропусканием 30 %. Выходная мощность 3,3 Вт, длительность импульса 11,5 нс получены при частоте следования импульсов 10 кГц, энергия импульса составила 333 мкДж, пиковая мощность 29 кВт. Импульсы энергией 97 мкДж при частоте следования 10 кГц получены на частоте второй гармоники с эффективностью преобразования 29 %.

Проведенные исследования показывают, что благодаря высокому поперечному сечению стимулированного излучения ($\approx 3,74 \cdot 10^{-20} \text{ см}^2$) на длине волны 999,6 нм, а также существенному снижению тепловой нагрузки на активный элемент при накачке в области 980 нм и генерации в области 999 нм, кристаллы $\text{Yb}^{3+}:\text{LuAP}$ весьма перспективны в качестве активных элементов компактных твердотельных лазеров с диодной накачкой, работающих в режиме активной модуляции добротности.

Ключевые слова: модуляция добротности, ионы иттербия, диодная накачка, кристалл лютециевого алюмината, генерация второй гармоники.

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Introduction

Compact diode-pumped Q-switched lasers with pulse repetition rate of tens kilohertz are of practical importance for materials processing, spectroscopy, range finding applications. The increasing demand for laser in materials processing can be attributed to several unique advantages like high processing speed, the ability of automation technological process, non-contact processing, elimination of subsequent finishing operation, reduced processing or operational cost, improved product quality, greater material utilization, and minimum heat affected zone [1–4].

Currently the most popular active media for such commercial systems are Nd³⁺-doped crystals (YAG, YVO₄ or YLF). Neodymium doped media have a number of advantages like 4-level laser scheme and high stimulated emission (SE) cross section (~10⁻¹⁹ cm²) that allow efficient laser action with comparatively low pump and laser beam intracavity intensities [5]. These systems can provide high single pulse energy [6] without damage of the intracavity optics but high quantum defect of Nd³⁺ ions leads to significant thermo-optic aberrations [7] that restrict the possibilities of power scaling, especially at high repetition rates. In order to obtain relatively high output powers, sophisticated cooling systems and powerful 808 nm AlGaAs diodes are used [8], which results in the drop of the cost efficiency. In contrast, Yb³⁺-based laser system have 3-level laser scheme and lower SE cross section [9] that result in a comparatively high pump and intracavity laser beam intensities for efficient laser operation. But this disadvantages almost zeroing when we operate with high pulse repetition frequency (PRF) and thus with reduced single pulse energy. Moreover, the utilization of Yb-doped material could improve the performance and cost of Q-switched laser system because of the substantial reduction of the thermal effects due to low quantum defect and high availability of InGaAs diodes.

It is evident that the issue of development of laser media with advanced thermo-optical properties and lasing performances remains relevant. One of the most promising crystalline hosts for satisfying these conditions are Perovskite-like aluminate crystals. Yttrium aluminate crystal YAlO₃ (YAP) is a well-known host for rare-earth ions [10–17]. The wide interest to YAP crystal is explained by its good thermal and mechanical properties similar to those of YAG [18]. Previously investigated Yb³⁺-doped yttrium aluminate crystal demonstrate

high termo-optical properties (thermal conductivity for undoped crystal is about 11 W/m·K and about 8 W/m·K for Yb(5 at.):YAP [18, 19]) and wide absorption and stimulated emission cross-section spectra [20] that makes this crystal a promising material for high power lasers and amplifiers emitting in the 1 μm-spectral range.

The reduction in the thermal conductivity of doped laser crystal in comparison with host materials is small in the case of ions with close atomic mass and ionic radii such as for Yb³⁺ and Lu³⁺ [21–23]. This feature makes LuAP more promising host crystal for doping by Yb³⁺ ions in contrast to YAP especially for high output power laser systems.

Here we report on the crystal growth, spectroscopy, CW and Q-switched laser operation of the Yb³⁺-doped isostructural LuAlO₃ single crystal as a novel promising material for high power diode-pumped actively Q-switched lasers for the first time to the best of our knowledge.

Crystal growth

LuAP is a biaxial crystal of the “distorted perovskite” type (space group D_{2h}¹⁶-Pbnm). Unlike the stable aluminates based on large-size rare-earths (RAlO₃, R = Gd-Er), the end-member orthorhombic aluminates of smaller-size rare-earths (RAlO₃, R = Tm, Yb, Lu) are considered as metastable compounds, i. e., they have no stability regions in the subsolidus and cannot be sintered employing traditional solid state reaction techniques. In the flux growth, TmAlO₃ and YbAlO₃ phases have been observed together with the corresponding garnet phases, while LuAlO₃ could not be obtained; instead of this, Lu₃Al₅O₁₂ and Lu₂O₃ phases have been recognized [24]. The first reported LuAlO₃ single crystals were grown from the melt by Czochralski method [25]. Phase equilibria studies in the Lu₂O₃-Al₂O₃ system [26–29] have shown that the range of stability of the perovskite phase is quite narrow and its nucleation may occur only on cooling from the molten state, from temperatures well above the liquidus temperature. Based on solidification behavior of the LuAP melts, the schemes were developed for single crystal growth, the details of which can be found in [34]. LuAP:Yb single crystals for the present studies were grown by the vertical Bridgman method (or vertical directional crystallization) [30, 31] under Ar/H₂ atmosphere (5 vol% of H₂) using molybdenum containers 14 mm in diameter. High purity Lu₂O₃,

Yb₂O₃ and crystalline Al₂O₃ were used as starting components; the selected concentrations of Yb were 2, 5 and 10 at.%. Due to a very small size mismatch between Yb³⁺ ($r_{\text{VIII}} = 0.985 \text{ \AA}$) and Lu³⁺ ($r_{\text{VIII}} = 0.977 \text{ \AA}$) ions [32] (around 0.8 % with respect to Lu³⁺), the distribution coefficient of Yb³⁺ ions in LuAlO₃ is close to unit and practically all Yb ions amount added to the melts is being incorporated into the lattice.

Spectroscopy

Polarized absorption spectra of Yb³⁺(2 at.%)LuAP (corresponding ytterbium concentration was $4.02 \times 10^{20} \text{ cm}^{-3}$) at room temperature were registered by a Varian CARY-5000 spectrophotometer. Absorption cross-section spectra for three light polarizations parallel to the a, b and c crystallographic axes are shown in Figure 1.

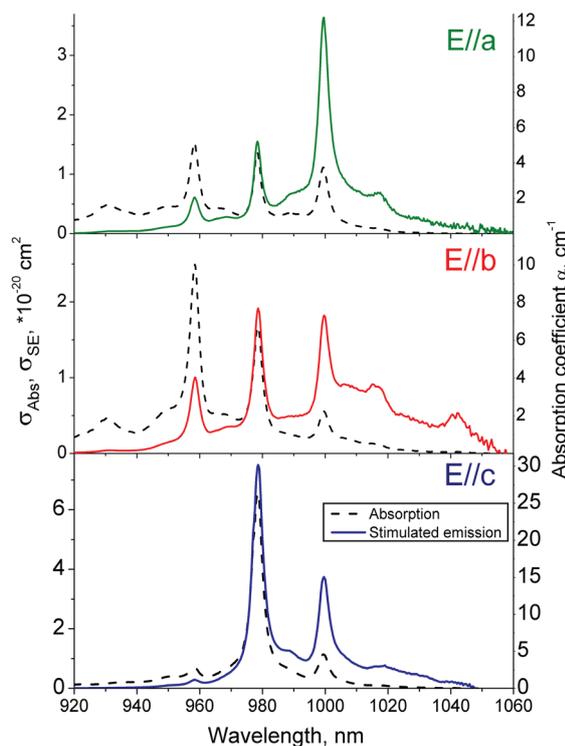


Figure 1 – Polarized absorption and stimulated emission cross-section spectra of Yb³⁺:LuAlO₃ crystal (the spectra were obtained for Yb³⁺(2 at.%)LuAlO₃)

Strong absorption is found for E//c light polarization with the peak absorption cross-section at 978.5 nm of about $6.6 \times 10^{-20} \text{ cm}^2$ and spectral bandwidth FWHM of 4 nm.

It is well known that radiation trapping strongly affects the measured lifetime of Yb-doped materials because of significant overlap of

the absorption and emission bands [33, 34]. The comparatively high index of refraction of LuAP ($n_0 = 1.923$) also increases the probability of reabsorption even in optically thin samples because of the total internal reflection. Thus the special methods discussed in the literature [33, 34] should be used to determine the luminescence lifetime accurately. In our experiments we used a fine powder of Yb:LuAP crystal immersed in glycerin. The diameter of the powder particles was measured to be approximately 30–40 μm , several times lower than absorption length of the most heavily doped Yb³⁺(10 at.%)LuAP crystal (75 μm at 978.5 nm). The Yb ions contents in the samples were 2, 5 and 10 at.%. The samples were excited by 20 ns pulses at 978.5 nm and luminescence kinetics was registered 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 decays (see Figure 2b). Starting from certain powder content, the lifetime remained constant despite further dilution (Figure 2a), thus indicating that reabsorption effects became negligible. Emission lifetime for 10, 5, and 2 at.% Yb-doped crystals was measured to be $310 \pm 10 \mu\text{s}$, $380 \pm 10 \mu\text{s}$ and $475 \pm 10 \mu\text{s}$, respectively (see Figure 2c). Taking into account that similar concentration quenching was observed for Yb:YAP starting from about 4 at.% of Yb doping concentration [35], we believe that the measured value of $(475 \pm 5) \mu\text{s}$ corresponds to the radiative lifetime of Yb³⁺-ions in LuAP.

The stimulated-emission cross sections were calculated by use of the modified reciprocity method in which it is not necessary to know the Stark level structure of the Yb³⁺ manifolds (²F_{5/2} and ²F_{7/2}) [36]:

$$\sigma_{SE}^{\alpha}(\lambda) = \frac{3 \cdot \exp(-hc/(kT\lambda))}{8\pi n^2 \tau_{rad} \cdot c \cdot \sum_{\beta} \int \lambda^{-4} \sigma_{ABS}^{\beta}(\lambda) \exp(-hc/(kT\lambda)) d\lambda} \sigma_{ABS}^{\alpha}(\lambda), \quad (1)$$

where τ_{rad} is the radiation lifetime of an active center; c is the light velocity; h and k are Planck and Boltzmann constants, respectively; T is the crystal temperature; n is the refractive index of a crystal; α and β denote the polarization state; and σ_{ABS} is the ground-state absorption cross section.

The stimulated-emission cross section spectra calculated with this method are presented in Figure 1.

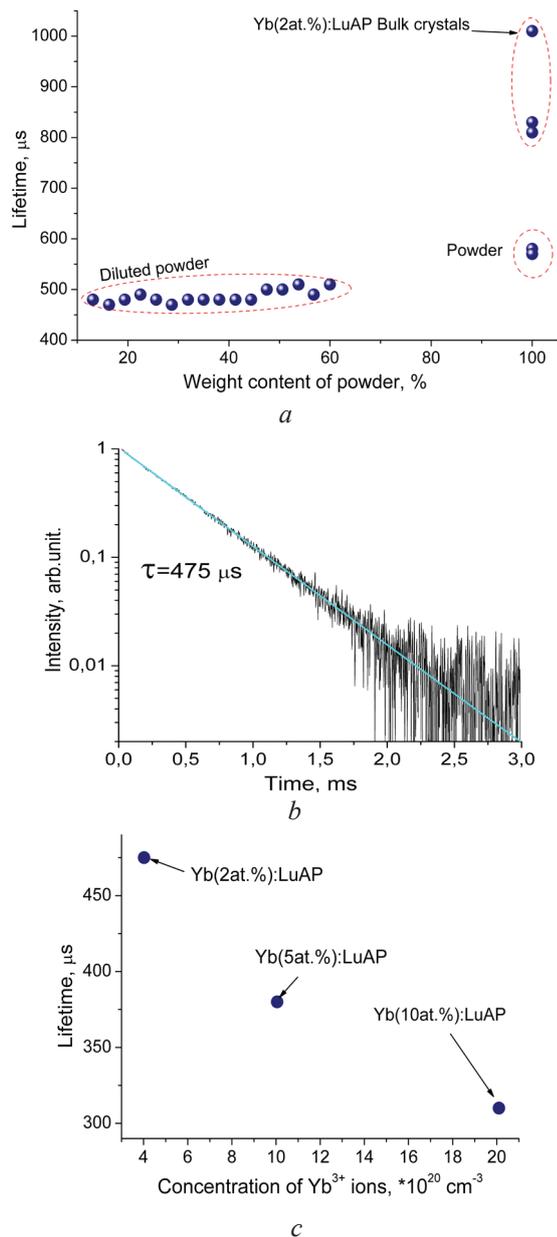


Figure 2 – Measured lifetime for different weight content of Yb(2 at.%):LuAP crystalline powder in glycerin suspension (a). Kinetics of luminescence decay (b) and measured Yb excited state lifetime for LuAP with different concentrations (c)

The most intensive stimulated emission (SE) cross-section band at 999.6 nm has peak value of about $3.74 \times 10^{-20} \text{ cm}^2$ for E//c-polarization. Such a high SE cross-section value is very suitable for actively Q-switched laser operation.

Continuous wave laser experiment

For laser operation the most interesting polarization states in the crystal are E//c and E//b (c and b are crystallographic axes) due to

high stimulated-emission cross sections values. In comparison with Yb-doped YAP, the crystal of Yb:LuAP exhibits slightly higher stimulated-emission cross section, a close radiative lifetime and a comparable stimulated emission bandwidth [20].

For a CW laser experiments a set up with X-folded cavity design was used (see Figure 3). It consisted of two curved mirrors M1 and M2 and two plane mirrors: OC and HR.

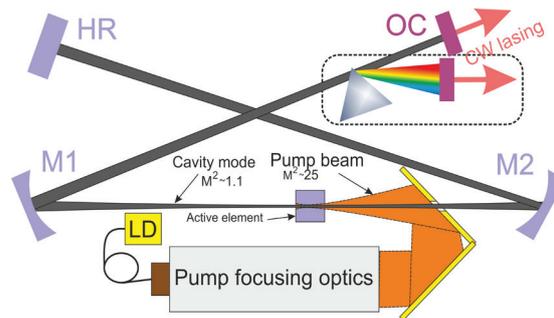


Figure 3 – Experimental setup of continuous wave diode-pumped Yb:LuAP laser: HR-highly reflective mirror, M1, M2-concave mirrors, OC-output coupler, LD-laser diode

The calculated TEM₀₀ mode diameter in the crystal was about 180 μm. As a pump source, a multiple single emitter InGaAs fiber-coupled laser diode (Ø105 μm, NA = 0.15) with a maximum output power of about 25 W was used. An “off-axis” pump layout was used for longitudinal pumping of the active element (see Figure 3). This pump arrangement was successfully tested in our previous work [36–39] and the main advantage of such a pump scheme is that all the cavity mirrors have highly reflecting coating at 900–1100 nm. The pump light was formed by a set of lenses into the spot with a diameter of about 180 μm (1/e²). A 2 mm long Yb(2 at.%):LuAlO₃ crystal was used as a gain medium. The crystal was a-cut to provide E//b and E//c polarized laser output. It was a slab with dimensions 2(a) × 5(b) × 1.5(c) mm³; both 5 × 2 mm² lateral faces were maintained at 15 °C by means of copper plates (indium foil was used to improve thermal contact) and thermo-electrical cooling elements with water-cooled heat sink, while 1.5 × 5 mm² working faces were antireflection coated for pump and laser radiation.

The dependencies of the laser output power on the absorbed pump power for E//b- and E//c-polarized outputs and different OCs are shown in Figure 4. Absorbed pump power was real-time measured during the laser action.

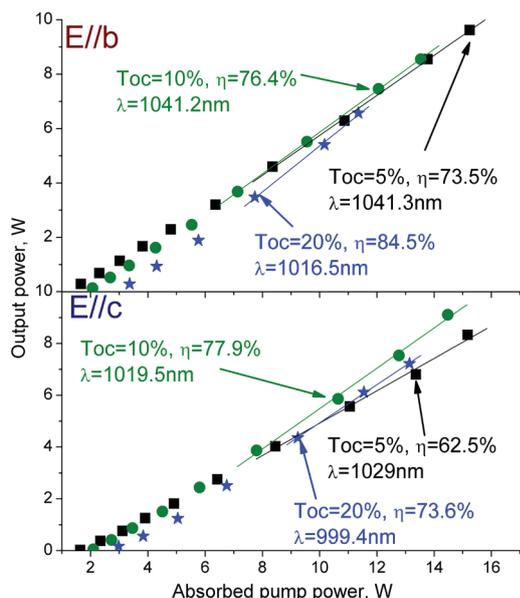


Figure 4 – CW laser performance of Yb:LuAP crystal for different polarizations and output coupler transmittances

The maximum CW output power of 9.6 W at absorbed pump power of 15.2 W with slope efficiency of 73.5 % was demonstrated for E//b polarization with 5 % OC transmittance. With output coupler transmission of 10 % and 20 % the laser output power slightly decreased to 8.6 W and 6.6 W, respectively, while the corresponding slope efficiencies increased to 76.4 % and 84.5 %. Similar output powers were demonstrated for E//c laser output. With 10 % output coupler transmittance 9.1 W of output power was obtained at 14.5 W of absorbed pump power with 77.9 % slope efficiency. Output powers of 8.3 W and 7.2 W with slope efficiencies of 62.5 % and 73.6 % were obtained for 5 % and 20 % OCs, respectively.

Actively Q-switched laser experiment

For actively Q-switched laser experiments a 3-mirror laser cavity was used (Figure 5) consisted of curved mirror and two plane mirrors, Pockels cell, Thin film polarizer and short wave filter. The calculated TEM₀₀ mode diameter in the crystal was about 180 μm. As a pump source, a multiple single emitter InGaAs fiber-coupled laser diode (25 W, Ø105 μm, NA=0.15) was used.

For Q-switched laser experiments was used the same active element as for CW experiments. First we study CW performance with “short” laser cavity (Figure 5). To reduce intracavity energy

density in Q-switched regime 20 % and 30 % output couplers were used. The dependencies of the CW output power versus incident pump power are shown in Figure 6.

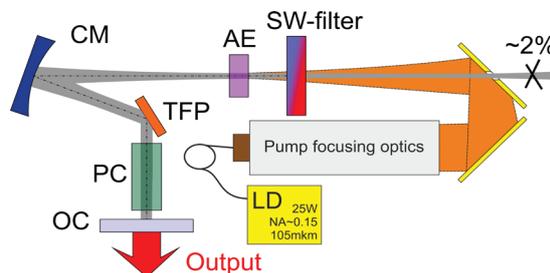


Figure 5 – Experimental setup of actively Q-switched diode-pumped Yb:LuAP laser: CM – concave mirror; AE – active element; SW – short wave filter; TFP – thin film polarizer; PC – Pockels cell; OC – output coupler; LD – laser diode

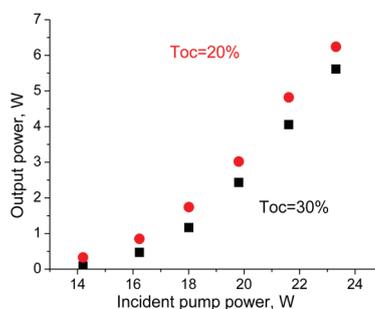


Figure 6 – Dependencies of CW output power versus incident pump power for Yb:LuAP laser (with “short” cavity)

Maximum CW output powers of 6.2 and 5.6 W obtained at 23.3 W of incident pump power for 20 % and 30 % OC transmittances, correspondingly.

The dependencies of the average output power and pulse duration (in actively Q-switched regime) on pulse repetition frequency (PRF) for E//c-polarized output and 30 % output coupler (OC) are shown in Figure 7.

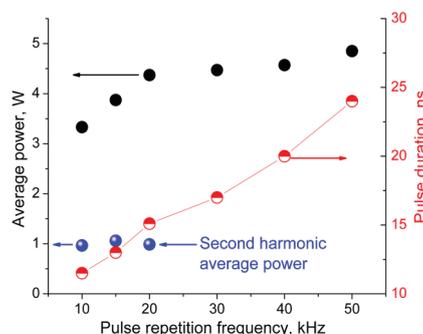


Figure 7 – Average output power and pulse duration vs PRF for actively Q-switched Yb³⁺:LuAP laser

The maximum average output power of 4.9 W at 50 kHz PRF with opt.-to-opt. efficiency of 21 % was demonstrated with 30 % OC transmittance. Output power as high as 3.3 W with pulse duration of about 11.5 ns was demonstrated at 10 kHz PRF with pulse energy of 333 μ J and peak power of 29 kW.

Second harmonic generation experiment

Second harmonic (SH) generation was investigated at low PRF by using 15 mm long LBO (Type I, $\theta=90^\circ$, $\varphi=13^\circ$) nonlinear crystal. Maximum pulse energy of 97 μ J was obtained at 10 kHz PRF. Dependency of SH average power on PRF are shown in Figure 7 for PRF range 10–20 kHz. Fundamental and SH wave spectra shown in Figure 8.

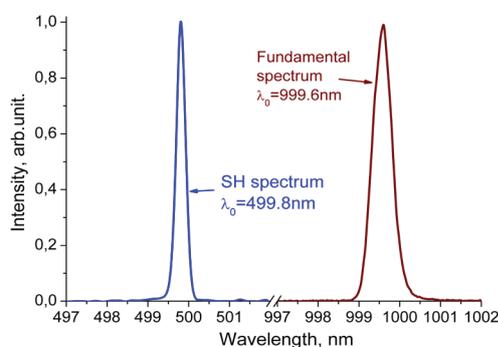


Figure 8 – Fundamental and SH spectra of actively Q-switched and frequency doubled $\text{Yb}^{3+}:\text{LuAP}$ laser

Conclusion

In conclusion, for the first time to the best of our knowledge actively Q-switched laser operation with $\text{Yb}^{3+}:\text{LuAP}$ single crystal was demonstrated. The maximum average output power of 4.9 W at 50 kHz PRF with opt.-to-opt. efficiency of 21 % was demonstrated with 30 % OC transmittance. Output power as high as 3.3 W with pulse duration of about 11.5 ns was demonstrated at 10 kHz PRF with pulse energy of 333 μ J and peak power of 29 kW. 97 μ J second harmonic pulses at 10 kHz PRF obtained with 29 % conversion efficiency.

Performed investigations show high potential of $\text{Yb}^{3+}:\text{LuAP}$ crystals as active elements of compact diode pumped actively Q-switched lasers due to high stimulated emission cross-section ($\approx 3.74 \cdot 10^{-20} \text{ cm}^2$) at 999.6 nm wavelength, which also reduces heat load on the active element taking into account low quantum defect for pumping around 980 nm.

Acknowledgments

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