

In-Band Pumped Continuous-Wave Lasers Based on Ho:KY(WO₄)₂ Crystal and Ho:KGdYbY(WO₄)₂ Epitaxial Layer

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Abstract

2 μm lasers are in demand for a number of practical applications, such as environmental monitoring, remote sensing, medicine, material processing, and are also used as a pump sources for optical parametric generators. Crystals of double potassium tungstates doped with ions of rare-earth elements were shown to be promising materials both for the creation of classical solid-state lasers and waveguide lasers. The aim of this work was to develop a tunable pump laser in the spectral region of 1.9 μm based on double tungstate crystals doped with thulium ions and to study the lasing characteristics of a Ho:KY(WO₄)₂ crystal and a Ho:KGdYbY(WO₄)₂ single-crystal epitaxial layer under in-band pumping.

With a Ho(1at.%):KY(WO₄)₂ crystal, continuous wave low-threshold lasing with an output power of 85 mW with a slope efficiency of 54 % at 2074 nm was achieved. For the first time to our knowledge, continuous wave laser generation in a waveguide configuration is realized in a single-crystal layer of potassium tungstate doped with holmium ions grown by liquid-phase epitaxy. The maximum output power at a wavelength of 2055 nm was 16.5 mW.

Keywords: holmium, thulium, potassium rare-earth tungstates, continuous-wave laser, epitaxial layer.

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Непрерывные лазеры на кристалле $\text{Ho:KY}(\text{WO}_4)_2$ и монокристаллическом слое $\text{Ho:KGdYbY}(\text{WO}_4)_2$ при резонансной накачке

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Лазерные источники, излучающие в спектральной области около 2 мкм, востребованы для ряда практических применений, таких как экологический мониторинг окружающей среды, дистанционное зондирование Земли, медицина, обработка материалов, а также используются в качестве источников возбуждающего излучения параметрических генераторов света. Кристаллы двойных калиевых вольфраматов, активированные ионами редкоземельных элементов, показали себя перспективными материалами как для создания классических твердотельных, так и волноводных лазеров. Целью настоящей работы являлось создание перестраиваемого лазера накачки, излучающего в спектральной области 1,9 мкм, на основе активированных ионами тулия кристаллов двойных вольфраматов и исследование генерационных характеристик кристалла $\text{Ho:KY}(\text{WO}_4)_2$ и монокристаллического эпитаксиального слоя $\text{Ho:KGdYbY}(\text{WO}_4)_2$ при резонансной накачке в полосу поглощения $^5\text{I}_8 \rightarrow ^5\text{I}_7$.

В лазере на основе кристалла $\text{Ho}(1\text{ ат. \%}): \text{KY}(\text{WO}_4)_2$ получена непрерывная низкопороговая генерация с выходной мощностью 85 мВт при дифференциальной эффективности 54 % на длине волны 2074 нм. Впервые реализована непрерывная генерация в волноводном режиме в монокристаллическом слое калиевого вольфрамата, активированного ионами гольмия, выращенного методом жидкофазной эпитаксии. Максимальная выходная мощность на длине волны 2055 нм составила 16,5 мВт.

Ключевые слова: ионы гольмия, ионы тулия, калий-редкоземельные вольфраматы, непрерывный лазер, эпитаксиальный монокристаллический слой.

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Introduction

Attention to anisotropic crystals of potassium-rare-earth double tungstates with a general chemical formula of $KRe(WO_4)_2$ (where $Re = Gd, Y, Lu$) doped with active rare-earth ions is associated with unique combination of their optical and spectroscopic properties which in total provide high laser efficiency, broad tuning range and nonlinear Raman conversion of laser emission in the spectral range 0.5–2.1 μm [1, 2]. For laser generation in the spectral range near 2 μm active laser media doped with Tm^{3+} and Ho^{3+} ions are used. These ions demonstrate a complex energy level structure that leads to energy transfer by up-conversion and cross relaxation mechanisms with efficiency depending on concentration of impurity ions. The advantage of thulium-doped crystals is the ability to pump them by the emission of AlGaAs laser diodes near 800 nm. Efficient cross-relaxation process at Tm^{3+} concentration higher than 5 at.% results in high quantum efficiency of pumping close to 2 [3]. And broad emission band enables to continuously tune laser wavelength in the range of about 200 nm [4]. Ho^{3+} ions have high absorption and emission cross sections, however the absorption band suitable for pumping lies in the spectral range near 1.9 μm . Pumping scheme where Tm-laser is used as a pump source for Ho-laser decreases the influence of up-conversion processes and reduces thermo-optical distortions in laser crystal due to small quantum

defect between pump (1.9 μm) and laser (2.1 μm) emission.

Laser generation with tungstate crystals doped with holmium ions was demonstrated in a number of papers [5–9]. First experiments with in-band pumping of Ho:KReW were carried out with Ho:KLu(WO₄)₂ crystal in hemispherical laser cavity [5] and among doping levels of 1, 3 and 5 % laser action was obtained only with 3 % doped sample. The absence of generation for 1 % doped crystals was explained by the weak absorption of pump emission. For the next laser experiments the crystals were used with holmium concentration of 3 % and higher. In [6] spectroscopic characteristics of Ho(3at.%):KY(WO₄)₂ crystal were investigated and laser action was obtained in microchip cavity configuration. Comparative investigation of laser properties of the crystals Ho(3at.%):KLu(WO₄)₂, Ho(3at.%):KY(WO₄)₂, and Ho(3at.%):KG(WO₄)₂ in the same experimental conditions [7] did not demonstrate sufficient difference between these materials, maximal slope efficiency of 58.8 % with respect to absorbed pump power under diode pumping was achieved for Ho(3at.%):KY(WO₄)₂. At holmium doping concentration of ≥ 7 at.% in tungstates laser action was absent and it was associated with up-conversion and re-absorption losses [8]. In [9] efficient generation under fiber-laser pumping was demonstrated in Ho(1at.%):KY(WO₄)₂ crystal however laser threshold was high. The results described in [5–9] are summarized in Table 1.

Table 1

Laser characteristics of in-band Ho^{3+} -doped $KRe(WO_4)_2$ crystals

	λ_p , nm	P_{out} , mW	P_{th} , mW	η_{abs} , %	η_{inc} , %	λ_{las} , nm	Ref.
Ho(3%):KLuW	1946	648	≈ 500	54.8		2078	[5]
Ho(3%):KYW	1941	438	521	58.8	9.85	2075	[7]
Ho(3%):KYW	1946	206	190	85		2015	[6]
Ho(1%):KYW	1960	3000	≈ 500	72.5	57.5	2074	[9]

* λ_p – pump wavelength; P_{out} – output power; P_{th} – laser threshold power; η_{abs} – slope efficiency with respect to the absorbed pump power; η_{inc} – slope efficiency with respect to the incident pump power; λ_{las} – laser wavelength

Potassium-rare-earth double tungstates were shown to be promising materials for waveguide laser applications. Laser generation with slope efficiency more than 80 % was demonstrated with Yb(1.8at.%):KY(WO₄)₂ [10] и Tm(8at.%):KY_{0.40}Gd_{0.29}Lu_{0.23}(WO₄)₂ [11]

waveguide lasers. Lasing with holmium doped tungstates in waveguide mode of operation was realized only with Ho(5at.%):KGd(WO₄)₂ channel waveguide which was inscribed by femtosecond laser pulses [12]. Output power up to 212 mW at 2055 nm wavelength was achieved with slope effi-

ciency of 67.2 % with respect to absorbed pump power and 12.1 % with respect to incident pump power.

The aim of this work was to develop a tunable pump laser in the spectral region of 1.9 μm based on double tungstate crystals doped with thulium ions and to study the lasing characteristics of a Ho:KY(WO₄)₂ crystal and a Ho:KGdYbY(WO₄)₂ single-crystal epitaxial layer under in-band pumping.

Growth of crystals and monocrystalline layer

Tm:KLu(WO₄)₂ and Ho:KY(WO₄)₂ crystals were grown in the Institute of Inorganic Chemistry SB RAS by a top seeded solution growth method in the conditions of low thermal gradients. The potassium tungstate K₂W₂O₇ was used as a solvent.

Tungstate crystals are optically biaxial and their optical properties should be described within the frame of optical indicatrix axes (N_p , N_m and N_g). The spectroscopic characteristics of the Ho:KY(WO₄)₂ crystal are presented in [6]. In this work, the absorption spectra of the Ho:KY(WO₄)₂ crystal (Figure 1) recorded in polarized light by a Varian CARY-5000 spectrophotometer were used to refine the concentration of holmium ions in the studied samples which amounted to 1 at.%. The spectral width of the spectrophotometer slit during measurements was 0.6 nm.

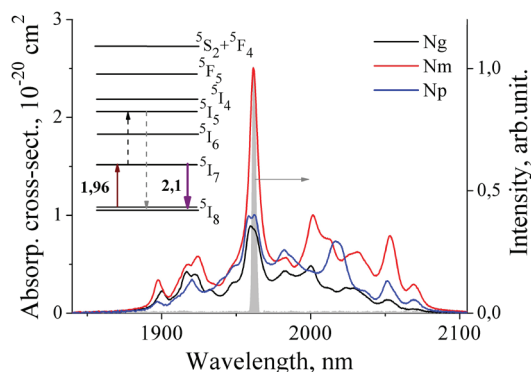


Figure 1 – Absorption spectrum of the Ho:KY(WO₄)₂ crystal at 2 μm and pump laser output spectrum

Polarizations $E//N_m$ and $E//N_p$ show generally higher values of absorption and emission cross section in comparison with $E//N_g$, therefore the crystal was cut along N_g optical axis. The maximum absorption cross-section is $2.5 \cdot 10^{-20} \text{ cm}^2$ at 1961.5 nm for $E//N_m$, the full width at half maximum of absorption band amounts 8.6 nm. It should be noted that the peak absorption cross section of the $^5I_8 \rightarrow ^5I_7$ transition exceeds the value presented in the work [6].

The growth of the monocrystalline layer was performed by a liquid phase epitaxy technique onto an undoped b -cut KY(WO₄)₂ substrate in Scientific and Practical Materials Research Center of the National Academy of Sciences of Belarus. The process of crystallization in the method of liquid-phase epitaxy is carried out due to the supersaturation of the solution-melt near the interface line in the central near-surface area. The fabrication was carried out on a lab-type of an electrical resistance furnace with a two-zone heater using platinum crucibles in a temperature range of 900–920 °C.

The two-section heating circuit made it possible to change the temperature difference along the height of the solution-melt to ensure effective convection mixing and prevent the formation of parasitic phases in the bottom region of the solution-melt. The solution-melt was heated in a crucible above the saturation temperature by 30–50 °C for the homogenization during 12 hours. Then the temperature dropped by one to two degrees below saturation temperature and the rod with a single crystal substrate was placed in the melt solution. The temperature was adjusted so that the crystallization process took place in the metastable region.

The growth rate of the layer was 2.5–3 $\mu\text{m}/\text{min}$ and was determined by the difference between the liquidus temperature and the growth temperature. The full growth-run of the layer was completed in 1.5–2 h. The growth was carried out along the crystallographic axis b which coincides with the axis of the optical indicatrix N_p . The grown Ho(4.8at.%):KGd_{0.1}Yb_{0.112}Y_{0.74}(WO₄)₂ sample was cut along N_g optical indicatrix axis of the crystal and was polished to a $40^{+5} \mu\text{m}$ thickness. The refractive index contrast between the substrate and layer was ≈ 0.003 .

The photograph of the layer made from the end face of the structure with a polarized Polam RP-1 (LOMO) microscope with a magnification of 40x with a CCD camera is presented in Figure 2.

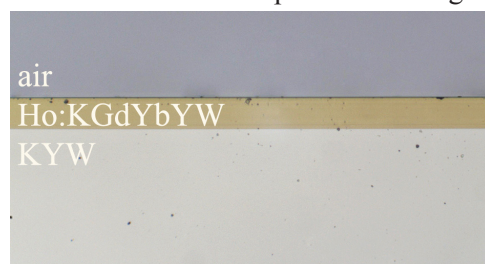


Figure 2 – Microscope photograph of the Ho(4.8at.%):KGd_{0.1}Yb_{0.112}Y_{0.74}(WO₄)₂ epitaxial layer made from the end face

The grown layer was characterized by the absence of inhomogeneities, cracks and significant defects.

Experimental setup

The scheme of the setup for the laser experiments is shown in Figure 3. Output power was determined using a VEGA power meter with a 3A-P thermal

sensor, measurement range 15 μ W–3 W. The wavelength of the laser was measured with an APE waveScan High Resolution Spectrometer (spectral range: 800–2600 nm; spectral slit width: 0.5 nm). Dual Scanning Slit Beam Profiler Thorlabs BP209-IR2/M with wavelength range 900–2700 nm was used for analyzing cross sectional profiles of near-Gaussian laser beams.

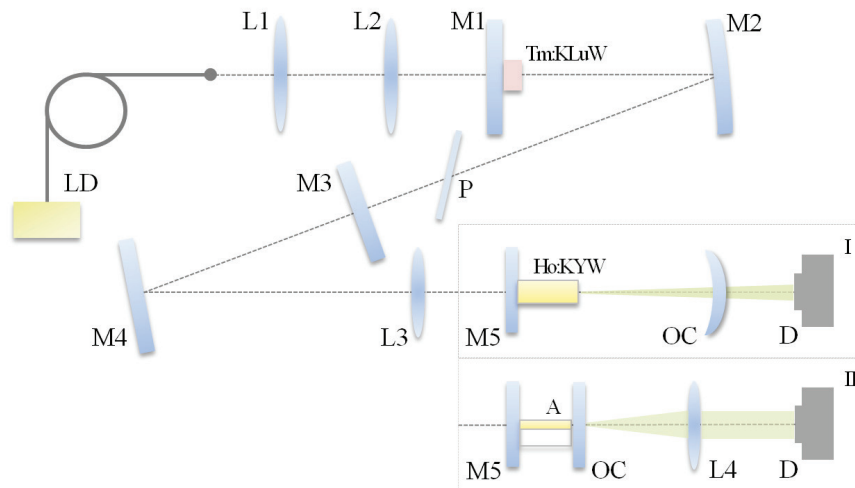


Figure 3 – Setup of the laser: LD – laser diode; L1, L2 – lens assembly for collimation and focusing; M1 – input mirror; Tm:KLuW – active element; M2 – spherical mirror; P – polarization interference filter; M3 – output coupler; M4 – plane bending mirrors; L3 – focusing lens; M5 – input mirror; Ho:KYW and A – active element, OC – output coupler; L4 – collimating lens; D – detector

The pump source for holmium laser medium was a homemade diode-pumped tunable Tm:KLu(WO₄)₂ laser. The wavelength of the laser was tuned to the maximum absorption of the holmium ions ⁵I₈→⁵I₇ in KY(WO₄)₂ (Figure 1) by using polarizing interference filter P placed at Brewster's angle. The active element Tm(6.2at.%):KLu(WO₄)₂ 2 mm long was wrapped in indium foil and fixed on a Peltier-cooled copper heat sink. A fiber-coupled (Ø 100 μ m, N.A. = 0.22) laser diode emitting up to 3 W at 801 nm was used as a pump source. The pump radiation was focused using collimating L1 (80 mm) and focusing L2 (70 mm) lenses into a spot with 95 μ m diameter.

The maximum output power of the thulium laser at 1961.5 nm was reached 600 mW. The radiation was linearly polarized in the $E//N_m$ direction. The lasing spectrum is shown in Figure 1, its full width at half maximum was 2.8 nm. The laser generated the TEM₀₀ mode, $M^2 = 1.2$.

The pump radiation was focused by a spherical lens with 40 mm focal length into a spot with 60 μ m diameter. Investigation of the lasing characteristics of the bulk Ho(1at.%):KY(WO₄)₂ crystal was held in a hemispherical cavity. To ensure efficient absorption of

pump radiation an active element 11 mm long was used. Flat input mirror M5 was coated for high transmission (> 98 %) at 1960 nm and high reflection above 2000 nm. Two spherical concave mirrors OC with transmission at 2050 nm $T_{OC} = 0.6\%$ (with a radius of curvature of 100 mm) and $T_{OC} = 1\%$ (with a radius of curvature of 30 mm) were used as output couplers. The cavity was calculated to provide intracavity laser mode radius inside the crystal of 60 μ m, the cavity length was 105 mm and 34 mm, respectively.

The cavity of a laser with a monocrystalline layer was formed by flat mirrors that were placed as close as possible to the waveguide end-faces with air gaps of less than 1 mm. Flat mirrors OC having different transmission T_{OC} at the laser wavelength were used as output couplers. The 8 mm long N_g -cut Ho(4.8at.%):KGd_{0,1}Yb_{0,112}Y_{0,74}(WO₄)₂ sample was placed on a copper heat sink without an active cooling system. No anti-reflection coatings were applied to the ends of the sample and no index-matching liquid was used. A spherical lens L4 was placed above the output coupler to collect and collimate the output beam. A dichroic mirror was used to separate the laser and the residual pump radiation.

Results

Output parameters of Ho(1at.%):KY(WO₄)₂ crystal laser were practically the same for output couplers with the transmittance of 0.6 % and 1 % (Figure 4). The maximal output power of 85 mW was obtained with 1 % output coupler, slope efficiency with respect to absorbed pump power was estimated to be 54 % that corresponded to 37 % efficiency with respect to incident pump power. A double pass of pump emission through laser crystal was taken into account for estimated absorbed pump power. Laser threshold was observed at the comparatively low pump power of 60 mW of absorbed power and 80 mW of incident power.

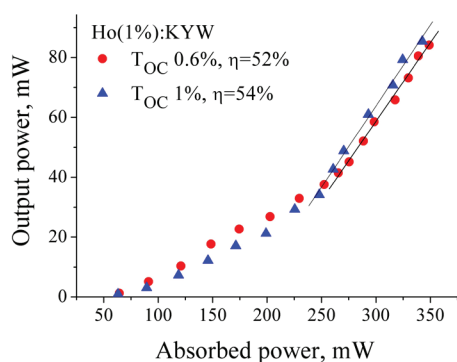


Figure 4 – Input-output characteristics of lasers on Ho(1at.%):KY(WO₄)₂ crystal

Laser emission had linear polarization $E//N_m$ and was observed at 2096 nm wavelength with 0.6 % output coupler while with 1 % OC transmittance laser wavelength was shifted to a shorter wavelength of 2074 nm (Figure 5). Transversal spatial distribution of laser beam intensity was in good agreement with Gaussian approximation (see insert in Figure 5) that evidenced TEM₀₀ laser mode.

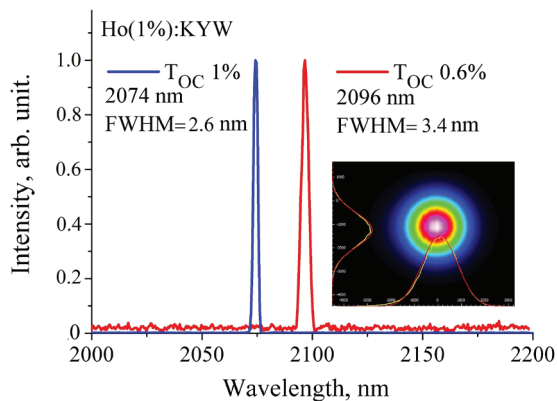


Figure 5 – Output spectrum of the Ho(1at.%):KY(WO₄)₂ laser

Output parameters of waveguide laser based on Ho(4.8at.%):KGd_{0,1}Yb_{0,112}Y_{0,74}(WO₄)₂ single crystal layer were investigated by using output couplers with transmittance of 0.6 %, 4 %, 7 %, and 14 %. The launching efficiency of pump emission in the active layer was experimentally estimated to be near 80 %.

The maximum output power of laser emission of 16.5 mW with a slope efficiency of 26 % with respect to absorbed pump power was obtained for 7 % output coupler (Figure 6) at the wavelength of 2055 nm (Figure 7). The spatial distribution of laser radiation on the output face of the active element is shown in the insert in Figure 7. In accordance with mathematical stimulation with such a waveguide thickness 4 transverse modes were supported. With other output couplers laser output power did not exceed 3 mW with a slope efficiency of 1–2 % and the laser wavelength was not changed.

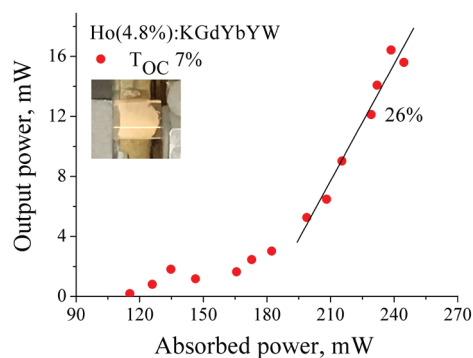


Figure 6 – Input-output characteristics of lasers on Ho(4.8at.%):KGd_{0,1}Yb_{0,112}Y_{0,74}(WO₄)₂ layer. The inset shows a photograph of an active element in a setup with an observed up-conversion glow (top view)

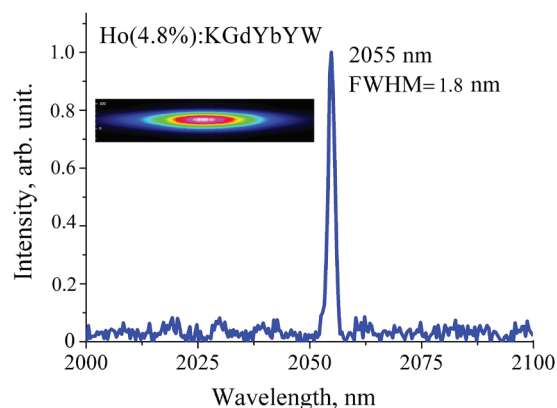


Figure 7 – Output spectrum of the Ho(4.8at.%):KGd_{0,1}Yb_{0,112}Y_{0,74}(WO₄)₂ laser. The inset shows the distribution of radiation on the output face of the active element

In laser experiments yellow up-conversion luminescence in a single crystal layer was observed (insert in Figure 6) which was associated probably with a high concentration of holmium ions as well as with non-resonant (phonon assisted) energy transfer between Ho^{3+} ($^5\text{I}_5$ level) and Yb^{3+} ($^2\text{F}_{5/2}$) ions. Mechanisms of energy transfer between Yb^{3+} and Ho^{3+} ions which leads to the population of upper excited states $^5\text{S}_2$, $^5\text{F}_4$ and $^5\text{F}_5$ of holmium ions are described in [13].

Underin-bandpumpingof $\text{Ho}(1\text{at.}\%):\text{KY}(\text{WO}_4)_2$ crystal weak up-conversion luminescence near 900 nm was detected corresponding to $^5\text{I}_5 \rightarrow ^5\text{I}_8$ transition. Population of $^5\text{I}_5$ level under pumping at 1.96 μm occurs as a result of excited-state absorption from $^5\text{I}_7$ state.

Conclusion

Continuous wave laser generation with $\text{Ho}(1\text{at.}\%):\text{KY}(\text{WO}_4)_2$ crystal and $\text{Ho}(4.8\text{at.}\%):\text{KGd}_{0.1}\text{Yb}_{0.112}\text{Y}_{0.74}(\text{WO}_4)_2$ single crystal layer has been demonstrated under in-band pumping by Tm-laser emission at 1961.5 nm wavelength. With $\text{Ho}(1\text{at.}\%):\text{KY}(\text{WO}_4)_2$ crystal an output power of 85 mW with a slope efficiency of 54 % at 2074 nm was achieved with comparatively low laser threshold of 80 mW of incident pump power.

For the first time to our knowledge continuous wave laser action in a waveguide configuration is realized in a single-crystal layer of potassium rare-earth tungstate doped with holmium ions grown by liquid-phase epitaxy. The laser threshold was reached at 115 mW of incident pump power. The maximum output power of 16.5 mW at a wavelength of 2055 nm was achieved and was limited by pump power.

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