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Optical properties of doped Potassium Gadolinium Tungstate single crystals

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ABSTRACT

Single crystals of double tungstates find applications as laser materials having very good parameters. One of the intensively investigated materials is $KGd(WO_4)_2$ doped with rare earth elements. Single crystals of $KGd(WO_4)_2$ were grown with the use of Top Seeded Solution Growth technique from $K_2W_2O_7$ solvent. The crystals have low absorption losses and show high lasing efficiency. Optical investigations of as grown KGW:Nd single crystals confirmed their good optical quality and high absorption coefficient near 810 nm, what in connection with strong luminescence near 1067 nm allows fabrication of diode pumped microchip lasers working both in CW and giant pulse regime. Absorption and luminescence spectra of Nd^{3+} doped KGW single crystals are presented. Laser action was obtained in form of 128.5 kHz train of 100 ns giant pulses due to YAG:Cr^{4+} passive Q-switch.

keywords: potassium gadolinium tungstate, high temperature solution growth, diode pumped laser, microlaser.

1. INTRODUCTION

In recent years double tungstates have been intensively investigated[1,2,3] due to their excellent properties allowing their applications in many optoelectronic devices. Especially solid state lasers with laser diode selective pumping establish a new class of lasers with particular properties. The spectral matching of diode radiation to the absorption bands of doped double tungstates makes it possible to obtain high pumping efficiency, single mode operation, and miniaturization of lasers. Two main double tungstates which find applications in diode pumped lasers fabrication are rare earth ions doped $KGd(WO_4)_2$ (KGW)[4] and $KY(WO_4)_2$ (KYW)[5] single crystals. Owing to the possibility of Gd or Y ions substitution with rare earth ions in a very broad range, one can create single crystals having properties suitable for chosen applications. In this paper we report on crystal growth and characterization of KGW:Nd single crystals containing different amounts of neodymium ions. Absorption and luminescence spectra of uniform KGW:Nd single crystals grown from high temperature solutions were measured.

2. KGW SINGLE CRYSTAL GROWTH

Monoclinic KGW with space group C2/c, as many other double tungstates, reveals high temperature irreversible phase transition at 1005°C.[6] This property imposes necessity of crystallization of low temperature KGW phase from high temperature solutions which enable lowering the temperature of crystallization below the temperature of the phase transition. $K_2W_2O_7$, a well-known compound commonly used in crystallization of double tungstates containing potassium, was chosen as a solvent. $K_2W_2O_7$ has relatively low viscosity,[4] what is very important factor in flux growth, and does not introduce any additional elements to the melt (so called self-flux). The solutions for crystallization were prepared by melting together proper amounts of K_2CO_3 p.a., WO_3 99.998%, Gd_2O_3 99.99%, and Nd_2O_3 99.99% to obtain 20 mol.% solution of $KGd_{1-x}Nd_x(WO_4)_2$ in $K_2W_2O_7$. Crystal growth was carried out from platinum crucibles on KGW seeds oriented in [010] direction under conditions of small temperature gradients (5°C/cm). Two-zone resistance furnace controlled by two independent Eurotherm 906S regulators/programmers was used in our experiments. High heat capacity of the furnace in connection with good temperature control (linear changes of temperature at a rate 0.1°C/hr) provided stable thermal conditions of crystallization.

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Top Seeded Solution Growth (TSSG) technique in connection with pulling growing crystals up at a rate of 2 mm/day allowed to grow uniform KGW:Nd single crystals having, contrary to crystals grown without pulling in Ref. 4, only one crystallographic plane (010) on the bottom of the growing crystal. Owing to this the as grown crystals had uniform distribution of neodymium. Such uniformity was impossible to obtain when crystallization occurred on several crystallographic planes having different coefficients of dopant distribution. KGW:Nd single crystals containing 1, 3 and 8 at.% of neodymium were grown. In Fig.1 typical KGW:Nd single crystal grown by TSSG with flat (010) bottom is presented.

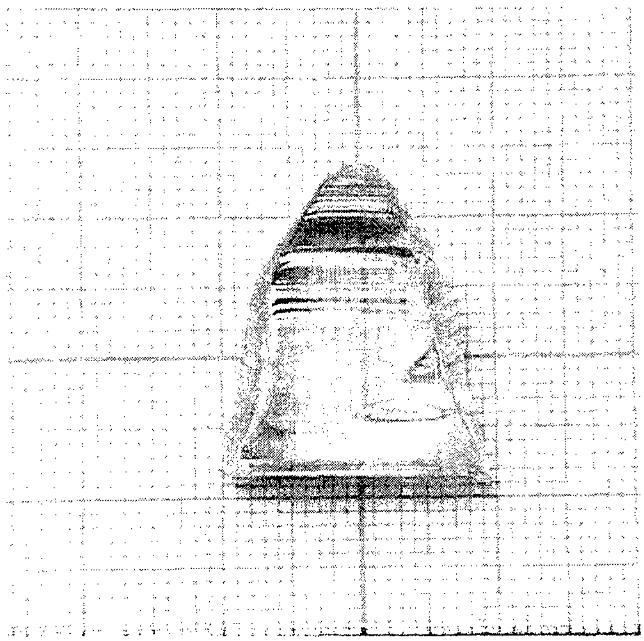


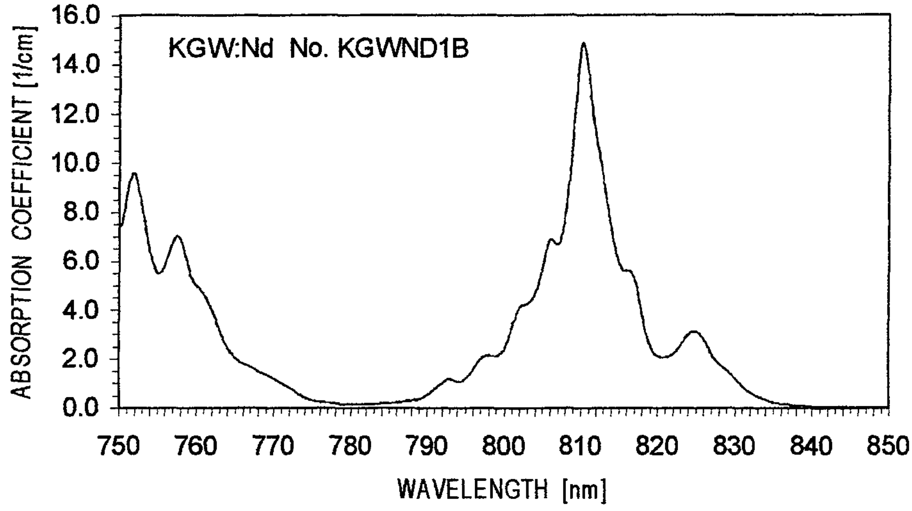
Fig. 1. As-grown [010] KGW:Nd single crystal having flat bottom. Scale in mm.

3. SPECTROSCOPIC INVESTIGATIONS

Plane-parallel plates 1 mm thick were cut from obtained single crystals of KGW:Nd, then graded, and polished. In order to determine absorption coefficient dependence on wavelength $k(\lambda)$ of the examined samples, the transmission measurements as a function of wavelength were performed. The measurements were carried out using the LAMBDA2 PERKIN ELMER spectrophotometer within the spectral range of 200÷1100 nm ($\Delta\lambda$=1 nm), the ACTA MVII BECKMAN spectrophotometer within the range of 1100÷1500 nm ($\Delta\lambda$=1 nm), and the Fourier PERKIN ELMER spectrophotometer 1725-X FT-IR within the range of 1.5÷25 μm ($\Delta 1/\lambda = 1\ cm^{-1}$). On the basis of the transmission $T(\lambda)$ measurements of the samples, an absorption coefficient was calculated with consideration of multiple reflections of radiation inside a sample. The spectral curves of the absorption coefficient for Nd^{3+} doped KGW crystals are shown in Fig. 2.

The measurements of luminescence spectra have been performed in the system with the H20 JOBIN YVON monochromator (focal length 200 mm). In the excitation channel the laser diode emitting at 808 nm was applied. Luminescence excited with laser radiation, after spectral splitting in H20 monochromator with holographic gratings, was registered by means of the LOCK-IN (STANFORD RESEARCH SR510) system with thermoelectrically cooled InGaAs detector. Three luminescence bands located at 900, 1067, 1350 nm were observed. The measurements of Nd^{3+} ions lifetime at the upper laser level ($^4F_{3/2}$) for the samples of KGW:Nd crystals were made by means of direct method with pulse excitation. The investigated medium was excited with radiation pulse duration significantly shorter than the lifetime τ at the excited level. After excitation a population level decay occurs, the evidence of which is fluorescence decay that can be observed. As a source of 808 nm diagnostic pulses SDL2430 laser diode was used. The laser was supplied from the power supply SDL800, controlled by a pulses generator. Generated pulses of 8 μs duration and 0.66 kHz frequency. In the detection channel, perpendicular to the excitation channel, the silicon photodiode was applied and time characteristics of fluorescence decay were registered with the digital oscilloscope LeCROY 9350AM (500 MHz). The results of measurements of fluorescence decay time in the investigated samples are listed in Table 1.

a)



b)

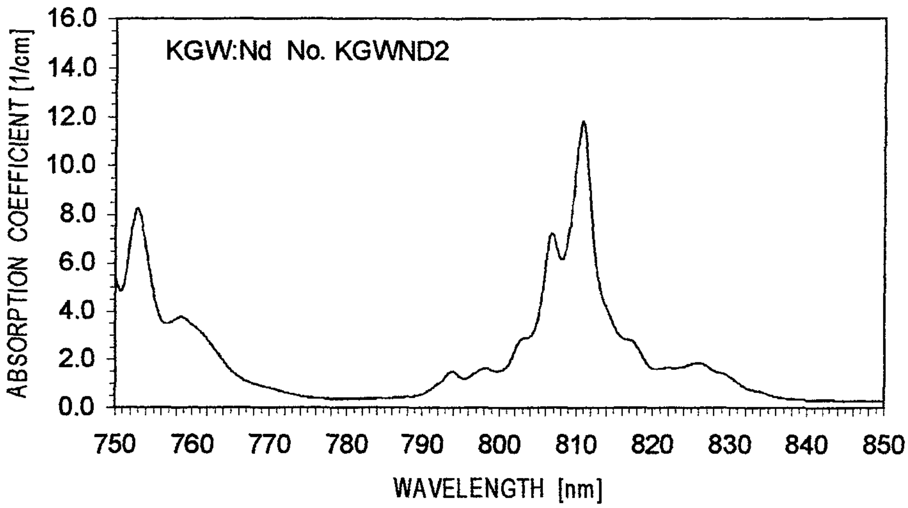


Fig. 2. Absorption spectra of neodymium doped KGW crystals
a) KGW:Nd^{3+} (8 at.% Nd^{3+}), b) KGW:Nd^{3+} (3 at.% Nd^{3+}).

Table 1. Results of measurements of fluorescence decay time (level $^4F_{3/2}$) in the investigated samples of KGW:Nd^{3+} crystals.

Crystal	Fluorescence decay time
KGW:Nd (8 at.% Nd^{3+})	101 ± 2 μs
KGW:Nd (5 at.% Nd^{3+})	115 ± 2 μs
KGW:Nd (3 at.% Nd^{3+})	128 ± 2 μs
KGW:Nd (1 at.% Nd^{3+})	132 ± 2 μs

4. LASER ACTION IN KGW:Nd

Investigations of longitudinally pumped microlasers generating at 1067 nm made of KGW:Nd with various Nd^{3+} ions concentration, were carried out. A schematic of the laser cavity is shown in Fig. 3. The results for KGW:Nd 3 at.% are presented in Fig. 4. Also the generation characteristics of longitudinally pumped microlasers with passive Q-modulation by CW SDL 2362 P3 laser diode were measured.

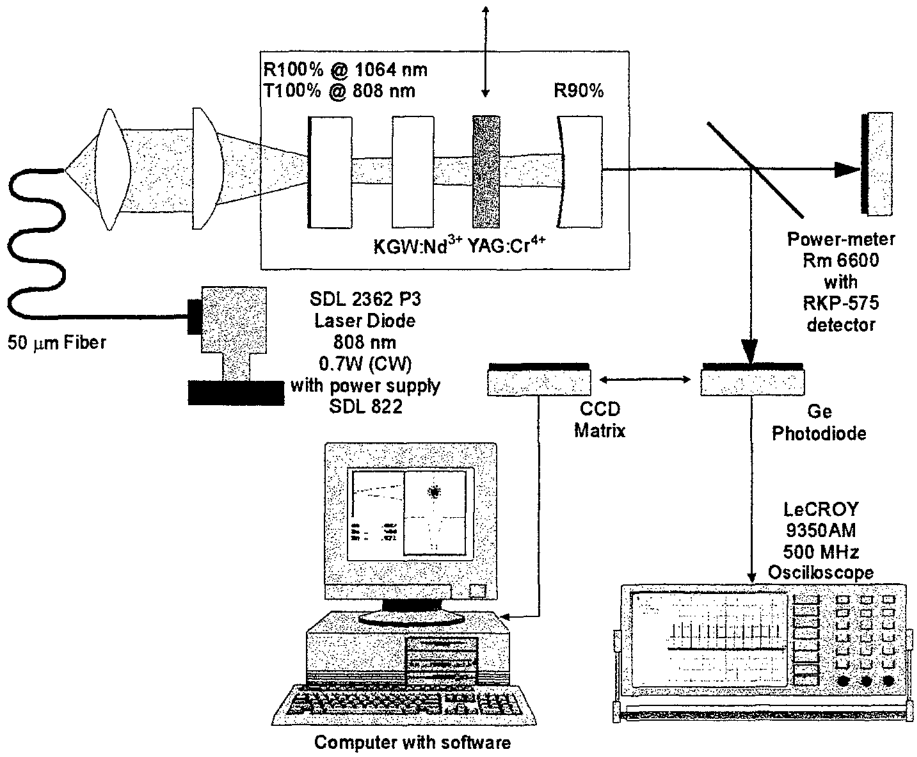


Fig. 3. Schematic of the $YAG:Cr^{4+}$ Q-switched $KGW:Nd^{3+}$ laser cavity

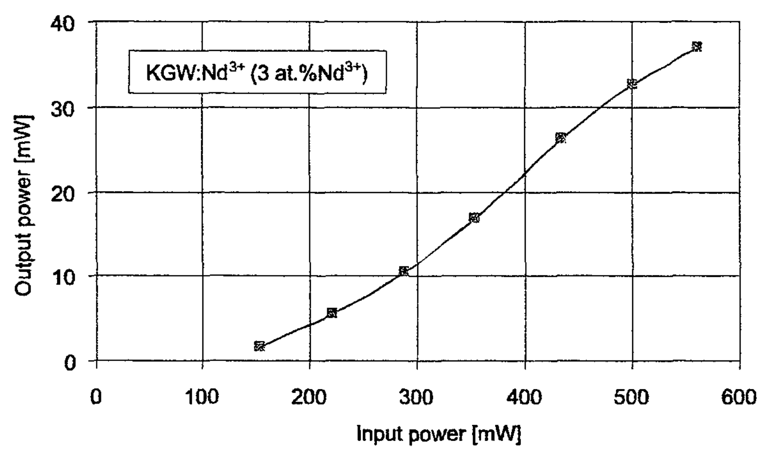


Fig. 4. Generation characteristics of $KGW:Nd^{3+}$ diode pumped laser (output mirror T=3.1%, r=50 mm)

The generation of a train of pulses with duration of 100 ns, frequency of 128.5 kHz and energy 9 μJ has been obtained (see Fig. 5). Q-switched pulses showed intensity and timing jitter depending on the positions of intracavity elements.

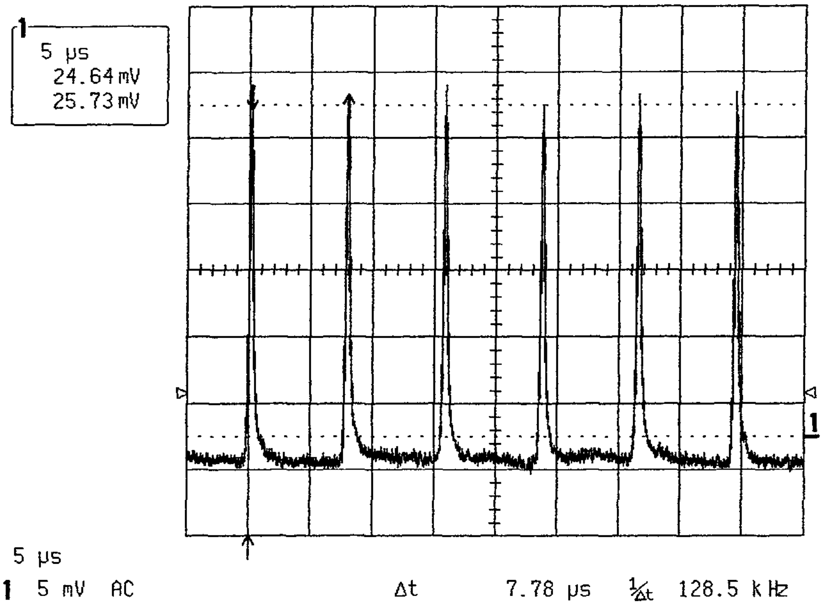


Fig. 5. Train of giant-pulses from YAG:Cr^{4+} passive Q-switched KGW:Nd laser (repetition rate 128.5 kHz, giant-pulse duration 100 ns)

5. CONCLUSIONS

The conditions of growth of KGW:Nd single crystals having various concentrations of dopant were determined. High Temperature Solution Growth technique was used. Investigations of optical and spectroscopic properties of the obtained crystals showed their good optical quality, so they can be applied in laser systems.

Due to advantageous spectroscopic parameters of KGW:Nd crystals and good thermal and mechanical resistance, those crystals were used both in CW as well as giant pulses train generation of the diode pumped microlasers.

ACKNOWLEDGEMENTS

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