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PHYSICS OF LASER CRYSTALS

Radiation Resistance of Yb:$LaSc_3(BO_3)_4$ and Yb:$LuYSiO_5$ Laser Crystals

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Abstract—The optical transmission (OT) spectra of Yb:$LaSc_3(BO_3)_4$ and Yb:$LuYSiO_5$ laser crystals have been analyzed before and after irradiation from a ^{60}Co source with doses up to 45 Mrad. The OT spectra of the Yb:$LuYSiO_5$ crystal are found to be the same (within the measurement error) before and after irradiation. The irradiation of the 10 at.%Yb:$LaSc_3(BO_3)_4$ crystal significantly changes its OT spectra in a wide spectral range (330 to 700 nm). A 975-nm laser based on a previously irradiated 4 at.%Yb:$LuYSiO_5$ crystal has exhibited a differential efficiency of 23% under diode pumping. The up-conversion luminescence spectra in the visible range of the crystals under study have been explained.

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1. INTRODUCTION

During the last 10–20 years, breakthrough progress has been observed in the laser technique due to the transition from lamp to diode pumping. Diode-pumped lasers exhibit high efficiency, reliability, compactness, etc. These solid-state selectively pumped lasers are sensitive to intracavity losses (in particular, to the losses caused by radiative impact on crystals of active elements). In this context, fundamental analysis of the radiation resistance of crystalline laser elements for diode-pumped lasers must be performed. The diode-pumped lasers used in spacecraft, measuring complexes of particle accelerators, and in controlled fusion studies undergo radiation impact. Therefore, an urgent problem is to search for and choose highly efficient radiation-resistant laser crystals.

One of the authors of this paper took part in the first studies in this field for lamp-pumped lasers based on garnet crystals activated with Nd^{3+} ions [1]. In particular, a Nd:Cr:GSGG-based laser, irradiated by a ^{60}Co gamma source to a dose of 10 Mrad, turned out to be insensitive to ionizing irradiation, in contrast to the Nd:YAG crystal, whose efficiency decreased by an order of magnitude at a much smaller irradiation dose (1 Mrad). The results of subsequent investigations confirmed that irradiation even to an enormous dose of 1000 Mrad only slightly (by about 5%) reduced the lasing efficiency of Nd:Cr:GSGG crystal. It was also shown that irradiation with 25-MeV protons up to a dose of 100 Mrad or with 2-MeV electrons up to a dose of 18 Mrad did not lead to degradation of the Nd:Cr:GSGG crystal [2]. It was concluded in both publications that Nd:Cr:GSGG is a radiation-resistant laser material and, therefore, is more suitable than Nd:$Y_3Al_5O_{12}$ (Nd:YAG) for space applications. The stability of the lasing characteristics of gamma-irradiated Nd:Cr:GSGG-based laser elements is due to the constancy of passive losses at a lasing wavelength of 1.06 μm, whereas in Nd:YAG crystals these losses significantly increase under irradiation.

An analysis of the photoinduced absorption of Nd:Cr:GSGG and Cr:GSGG crystals showed that they absorb in a wide spectral range: 300 to 370 nm. This process is related to the absorption from the excited state of the $^4T_2$ and $^4F_{3/2}$ activators of the Cr^{3+} and Nd^{3+} ions, respectively [3, 4].

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The absorption spectra of γ-irradiated $Y_3Al_5O_{12}$ crystals activated with a rare-earth element (Pr^{3+}, Nd^{3+}, Eu^{3+}, Tb^{3+}, Yb^{3+}, Lu^{3+}, Er^{3+}, or Ho^{3+}) were investigated in [5]. The irradiation sharply enhances the absorption in the range of 300 to 500 nm, which is related either to the formation of Pr^{4+} and Tb^{4+} tetravalent ions or to the reduction of some of Eu^{3+} and Yb^{3+} ions to the divalent state. The absorption bands were assigned to the centers based on O^- oxygen ions, arising under γ-irradiation.

In the last decade, numerous research teams in different countries have searched for radiation-resistant oxide and fluoride crystals as materials for scintillation detectors operating under high radiation in the central part of the international compact muon solenoid (CMS) in Switzerland (CMS experiments at LHC, CERN, Switzerland). Orthosilicate crystals based on lutetium and yttrium (R_2SiO_5, $R=Y$, Lu, Ce) are the most radiation-resistant scintillators; color centers are not formed in them under gamma irradiation from a ^{60}Co source to a dose of 68 Mrad or irradiation by a 155-MeV proton beam with a fluence of 4.4×10^{12} cm^2 [6]. Therefore, we chose the $LuYSiO_5$ laser host as an object of our study on the suggestion on its high radiation resistance, which is determined by the crystal structure with the densest filling of the unit-cell volume with ions in different oxidation states: Lu^{3+}, Si^{4+}, and O^{2-}. This crystal has a disordered structure because of the incorporation of Lu and Y ions into the crystal lattice. The disorder broadens the luminescence band of Yb ions, which is important for lasing in the mode of broadband wavelength tuning and generation of ultrashort radiation pulses.

The highly efficient $LaSc_3(BO_3)_4$ host was chosen for diode-pumped lasers; however, its radiation resistance was not investigated previously. The Yb:LSB crystal allows for high concentration of Yb ions (25%); therefore, it can efficiently be used in the form of a thin disk in high-power lasers.

In this paper, we report the results of analyzing the radiation resistance, up-conversion luminescence, and lasing properties of the $Yb{:}LuYSiO_5$ (Yb:LYSO, oxyorthosilicate) and $Yb{:}LaSc_3(BO_3)_4$ (Yb:LSB) crystals.

2. CRYSTAL GROWTH

Laser crystals 10- and 25-at.%Yb:LSB and 4- and 25-at.%Yb:LYSO were grown by the Czochralski method from iridium crucibles 40 mm in diameter (Fig. 1). The corresponding metal oxides of high purity grade (Sc_2O_3, La_2O_3, B_2O_3, Yb_2O_3, Y_2O_3, Lu_2O_3, and SiO_2) were used as starting materials for charge preparation. Before weighing, the oxides of rare-earth elements were placed in platinum vessels

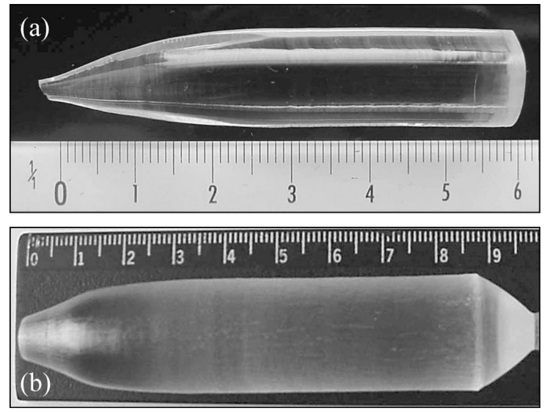

Fig. 1. (a) 10-at.%$Yb{:}LaSc_3(BO_3)_4$ and (b) 25-at.%$Yb{:}LuYSiO_5$ crystals.

and calcinated to remove water in air at $t=1000°C$ for 10 h. Boron oxide was annealed in a vacuum drying box at a temperature of 350°C for 10 h. The charge components were weighted and carefully mixed. After the mixing, the charge was pressed into pellets, which were sintered in platinum vessels at 900°C in air for 10 h. The thus prepared charge pellets were molten in an iridium crucible. After the complete charge melting, a seed crystal was immersed in the melt. At the melt crystallization onset, the crystal growth on the seed was controlled by a computer system. The crystal pulling rate was 2 mm h^{-1}. The seed rotation rate was 10 rpm.

The grown 10-at.%Yb:LSB crystal was used in this study to investigate the radiation resistance; previously, 0.3-mm-thick elements were prepared from this crystal for a disk diode-pumped laser [7, 8].

3. RADIATION RESISTANCE

The radiation resistance of the 10-at.%Yb:LSB and 25-at.%Yb:LYSO crystals was measured on samples with lengths of 18 and 13 mm, respectively. The samples were irradiated by a ^{60}Co radiation source (maximum power of approximately 4 krad min^{-1}). The crystal irradiation dose was 45 Mrad. The OT spectra were recorded before and immediately after the irradiation on a Shimadzu-UV3600 spectrophotometer.

An analysis of the OT spectra shows that the irradiation with a dose of 45 Mrad does not affect the optical transmission of the 25-at.%Yb:LYSO crystal. On the contrary, the transmission of the 10-at.%Yb:LSB crystal changes to a great extent under irradiation up to a dose of 45 Mrad in a wide spectral range: 330 to 700 nm (Fig. 2). The absorption-band edge undergoes a significant redshift (by 200 to 300 nm), which indicates a low radiation resistance of the Yb:LSB

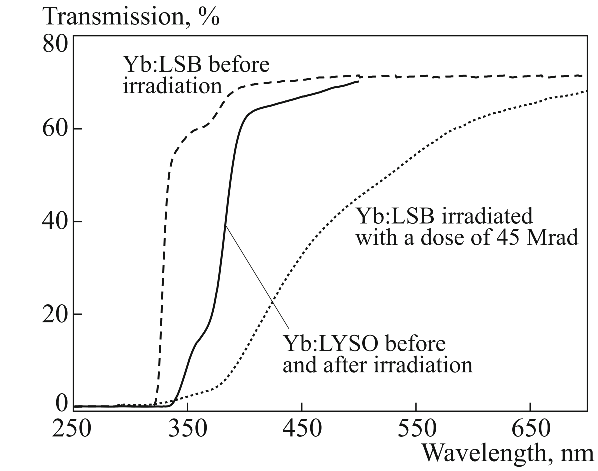

Fig. 2. Transmission spectra of 13-mm-long Yb:LSB and Yb:LYSO crystals before and after irradiation from a ^{60}Co radioactive source with an absorbed dose of 45 Mrad.

crystal. The illuminated Yb:LSB crystal was kept for a long time in darkness at room temperature, and only after five years of storage long-lived color centers disappeared and the sample became colorless again.

4. LASING

The 10-at.%Yb:LSB crystal, whose radiation resistance was analyzed in this study, was previously investigated in the laser experiment with a 300-μm-thick disk element [7, 8]. In this configuration, the output power of 1050-nm laser beam is 37 W under diode pumping at a wavelength of 974 nm with a power of 117 W, which corresponds to a conversion efficiency of 32%.

Laser experiments were performed on elements in the form of polished plane-parallel 2-mm-thick plates, with the crystallographic axis X oriented parallel to the surface, cut from a 4-at.%Yb:LYSO crystal. This geometry is characterized by strong absorption anisotropy in dependence of crystallographic orientation (Fig. 3).

A schematic of the experimental setup is shown in Fig. 4. The laser cavity is formed by two mirrors, one of which is a flat output mirror for lasing radiation with a reflectance of 95%. The other mirror is dichroic: it transmits pump radiation and completely reflects off lasing radiation. This mirror had a radius of curvature of 100 mm. A 2-mm-long Yb:LYSO active element with a cross section of 5×4 mm^2, having no antireflection coatings on its surfaces, was mounted on a copper heat sink using an indium foil.

A fiber-coupled diode laser module DLM-30 (IRE-Polus, Russia) with a fiber core diameter of 110 μm was used as a pump source. The pump radiation with a wavelength of about 975 nm and a spectral width of 6 nm was focused by a lens in the active element crystal into a spot 120 μm in diameter. Pumping could be performed both in the pulsed-periodic mode with pulse-width control and in the cw mode. The radiation power was measured by an OPHIR NOVA-II meter (Israel).

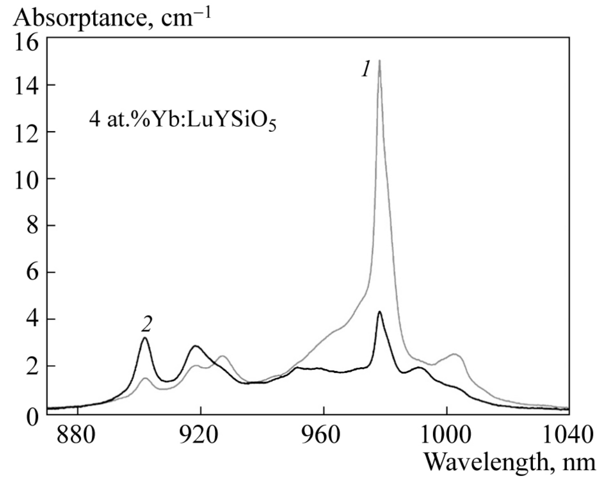

Fig. 3. Absorptance of the 4-at.%Yb:LuYSiO$_5$ crystal at room temperature for the $E \parallel X$ (*1*) and $E \perp X$ (*2*) polarizations.

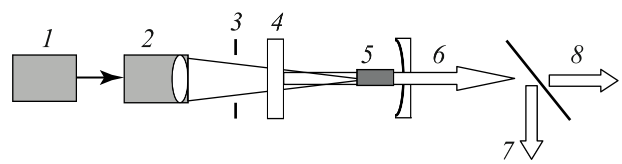

Fig. 4. Schematic of the laser system: pump laser (*1*), focusing lens (*2*), mechanical chopper (*3*), dichroic mirror (*4*), active element (*5*), output mirror (*6*), monochromator (*7*), and power meter (*8*).

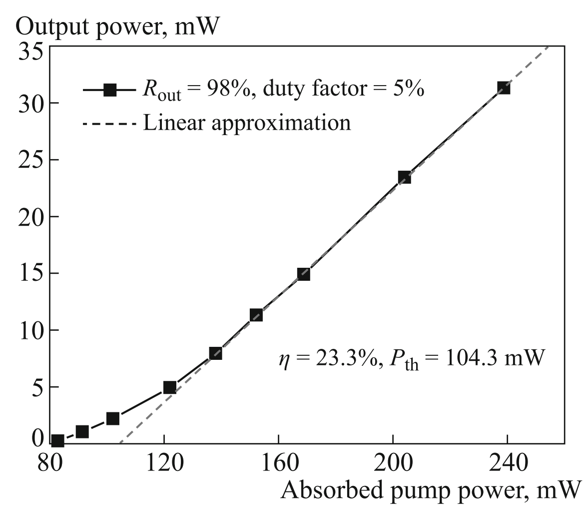

Fig. 5. Dependence of the output laser power on the absorbed pump power for 4-at.%Yb:LuYSiO$_5$ at the output-mirror reflectance $R_{out} = 98\%$.

Pumping in the pulsed-periodic mode was carried out at a pulse repetition rate of 42 Hz and on–off time ratio of 1:20. The average laser power was measured as a function of the average pump power absorbed in the active element. Figure 5 shows the dependence of the output laser power on the absorbed pump power for the 4 at.%Yb:LYSO laser element.

The laser based on the radiation-resistant 4-at.%Yb:LYSO crystal had an output power of 35 mW under diode pumping with a wavelength of 975 nm and power of 240 mW, which corresponds to a differential efficiency of 23%.

5. UP-CONVERSION LUMINESCENCE OF Yb:LYSO AND Yb:LSB

The laser experiment on the 4-at.%Yb:LYSO crystal under diode pumping at a wavelength of 975 nm, with the pump beam focused into a spot 120 μm in diameter, revealed intense luminescence in visible spectral range (Fig. 6).

The up-conversion luminescence spectrum of the 4-at.%Yb:LYSO crystal excited by 975-nm light contains narrow luminescence lines at 485.6, 526, 549.6, 669.6, and 828 nm. These luminescence can be explained by the presence of Pr^{3+} impurity ions. However, these lines are not observed under irradiation of the same sample by a 200-keV electron beam in the pulsed cathodoluminescence method, which indicates that impurity centers have a low concentration and are spaced by large distances. At the same time, the high energy density in the pumping focus leads to the up-conversion luminescence of the 4-at.%Yb:LYSO crystal, and this luminescence indicates that the pump energy is partially transformed into visible light (therefore, this energy is not involved in lasing).

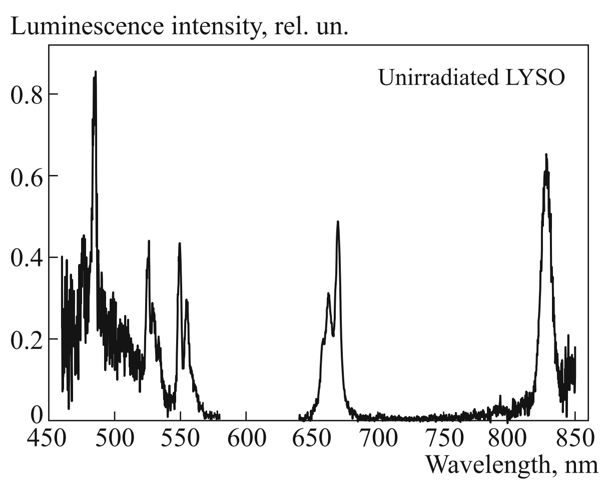

Fig. 6. Up-conversion luminescence spectrum of 4-at.%$Yb:LuYSiO_5$ crystal, excited by 975-nm light.

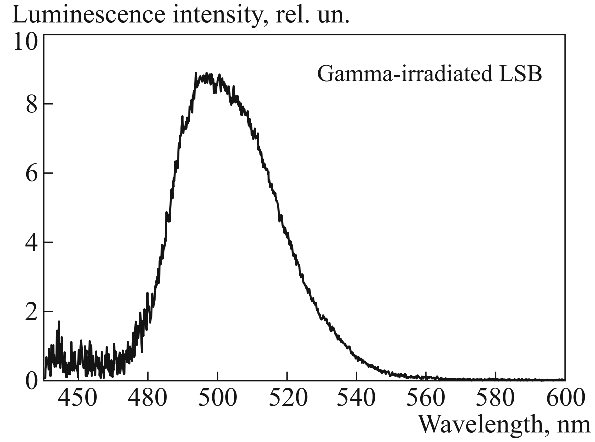

Fig. 7. Up-conversion luminescence spectrum of 25-at.%Yb:LSB crystal excited by 975-nm light.

Broadband luminescence, peaking at about 500 nm and having an FWHM of about 37 nm, is observed in the up-conversion luminescence spectrum of the 25-at.%Yb:LSB crystal excited by 975-nm light (Fig. 7). These values of the peak position and linewidth can be explained by the luminescence of oxygen defects O^- with a charge of -1 in the crystal lattice. In particular, cathodoluminescence in the vicinity of 510 nm was observed for several oxide compounds with different crystal structures. This radiation was explained as luminescence from oxygen defects [9, 10].

6. CONCLUSIONS

The transmission spectra of $Yb:LaSc_3(BO_3)_4$ and $Yb:LuYSiO_5$ laser crystals were analyzed before and after irradiation from a ^{60}Co source with doses up to 45 Mrad. It was established that the OT spectra of the $Yb:LuYSiO_5$ crystal before and after irradiation are identical (within the measurement error). The irradiation of the 10-at.%$Yb:LaSc_3(BO_3)_4$ crystal changes its OT spectra to a great extent in a wide spectral range: 330 to 700 nm. A 975-nm laser based on the 4-at.%$Yb:LuYSiO_5$ crystal exhibited a differential efficiency of 23% under diode pumping. The up-conversion luminescence of 25-at.%Yb:LSB with a peak at 509 nm can be explained by the presence of oxygen defects O^- with a charge of -1 in the crystal lattice.

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