BIULETYN WAT
ROK XLV, NR 8, 1996

Gamma-induced sensibilization of lasing properties of $Y_3Al_5O_{12}$ single crystals doped with Er^{3+} ions

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Abstract. The influence of gamma-irradiation on optical and lasing properties of YAG:Er single crystals was studied. For laser rods from YAG:Er crystals which weren't thermally annealed, the increase of laser output energy was observed after irradiation. The mechanisms of radiation sensibilization are discussed.
Keywords: ionizing radiation, gamma-induced sensibilization process, doping, colour centers.
Universal Decimal Classification: 548.55

1. Introduction

It is a well-established fact that colour centers (CCs) produced by ionizing radiation or short-wave radiation of the pump lamp strongly affect the normal operation of lasers based on the $Y_3Al_5O_{12}$ (YAG) crystals. The investigation of influence of ionizing radiation on the YAG crystal properties is of a special importance for lasers used in space [1]. The large number of publications including reviews and monographs are devoted to the problem of the radiation-stimulated changes of YAG:Nd crystal properties [1–5]. It should be noted, that irradiation of laser crystals may have the positive effect. The gamma-induced sensibilization process in Er^{3+}:CaF_2 crystal was observed by P.A. Forrestier and D.F. Simpson [2]. The improvement of generation characteristics of YAG:Nd lasers was reported as the effect of small dose of gamma and electron radiation [6].

YAG crystals with activators, such as Er^{3+}, are widely used in laser engineering. However, we found only one paper devoted to the gamma-radiation influence on the YAG:Er crystal optical properties [7].

In the present work the effects of gamma radiation on optical characteristics of YAG:Er crystals are described and generation characteristics of active elements made from YAG:Er irradiated with gamma quanta are examined. For a comparison, data about influence of gamma-radiation on optical characteristics of YAG:Nd crystals are presented.

2. Experimental

The examined crystals were pulled by Czochralski technique from iridium crucibles in N_2 atmosphere . The contents of Nd in the crystals was of the order of 1 at.%. The content of erbium in the YAG:Er crystals was equal to 33 at.%, that corresponds to the formula $Y_2ErAl_5O_{12}$. The detailed description of the applied growth process of the crystals is presented in paper [8].

The irradiating gamma quanta were emitted by a ^{60}Co emitter with exposure dose power of 170 R/s and the samples absorbed doses of the order of 10^2–10^5 Gy.

The crystal samples prepared to study the gamma irradiation effects on their optical properties were plane-parallel plates of 1–3 mm high, cut-out perpendicularly to the growth axis in the plane (111). Absorption spectra of the crystals were measured in spectral range 0.2–1.1 μm by means of the spectrometer LAMBDA-2. Additional absorption (AA) induced by irradiation was assumed as:

$$\Delta K = \frac{1}{d}\ln\frac{T_1}{T_2} \tag{1}$$

where: d — the thickness of the sample, T_1, T_2 — the transmissions of the sample before and after irradiation.

To examine gamma radiation effects on lasing characteristics of YAG:Er crystals, an active rod of 5 mm in diameter and 85 mm in length was used. The rod was not subjected to preliminary annealing and had no antireflection coatings on its end faces. The detailed characteristics of the rod, denoted as E21, are published in [8].

To measure generation characteristics of YAG:Er rods a plane-parallel resonator of 19 cm in length was used. The transmission of output mirrors was equal to 30% for YAG:Er ones. The studies were carried out in the ellipsoidal reflective head, made of gold-covered brass. The pump was a xenon lamp of diameter of 4 mm and 0.5–2.5 kV power with 160 μF capacitor battery. Pulse duration of the lamp equals about 580 μs. The laser light was detected with high-sensitive HgCdTe photoconductor and time characteristics of the lamp were observed with use of Si photodiode. The energy of laser pulses was measured by Gen-Tec radiometer with ED--500 gauge head.

3. Results and discussion

3.1. Optical properties

The absorption spectrum of YAG-Er is presented in Fig.1a. The detail description of YAG-Er absorption was made in [8]. After gamma irradiation a wide, complex AA band appears within the range of 48000–12000 cm^{-1}. The peaks of absorption are placed near of 42000 cm^{-1}, 32000 cm^{-1}, 25000 cm^{-1} and 16000 cm^{-1} (240, 310, 400, 625 nm). With the increase of gamma irradiation dose from 10^2 to 10^4 Gy the AA bands grow larger and larger and become saturated for doses of 10^4–10^6 Gy. The AA spectrum of YAG:Er crystals is also shown in Fig.1a (scale on the right hand side).

One can notice, that the gamma-induced absorption spectrum of YAG:Nd crystals has got the similar form as that of YAG:Er crystals and nearly the same peaks can be distinguished in both spectra.

The analysis of the published reports on radiation-induced colouration of YAG and YAG:Nd crystals [5, 9] shows that the form and intensity of AA spectrum depend on growth conditions of crystals (method and atmosphere of growth, purity of starting material, addition of an activator etc.). At the same time in some papers [10, 11] an existence of specified AA bands placed in 40000–42000 cm^{-1}, ~32000 cm^{-1} and ~22000–26000 cm^{-1} regions was reported. The bands within 40000–42000 cm^{-1} range are usually explained as caused by the absorption of uncontrolled Fe^{3+} impurities. The band placed near 32000 cm^{-1} is attributed mostly to the absorption of Fe^{2+} ions or CC connected with oxygen vacancies. The absorption bands within 22000–26000 cm^{-1} range are interpreted as the effect of O^- hole centers localized near defects of cation sub-lattice.

Consequently, it can be assumed that the AA bands observed in gamma-irradiated YAG:Er crystals are connected with CC produced by gamma-induced recharging of growth defects that may be uncontrolled impurities of Fe ions, oxygen vacancies or defects of cation sub-lattice (e.g. rare-earth ions instead of Al^{3+} ions). Note, that the precise definition of nature of the CC demands more detailed investigations.

To measure thermal stability of optical property changes caused by irradiation, an isochronous annealing of irradiated samples was performed in temperatures 293--673 K (15 min. duration at each temperature with steps of 20–30 K). It was found, that AA value in all the considered bands decreases accordingly to the same dependence of the rise of temperature. However, the complete return to the previous, what means before irradiation, optical features of the crystals were not reached after annealing.

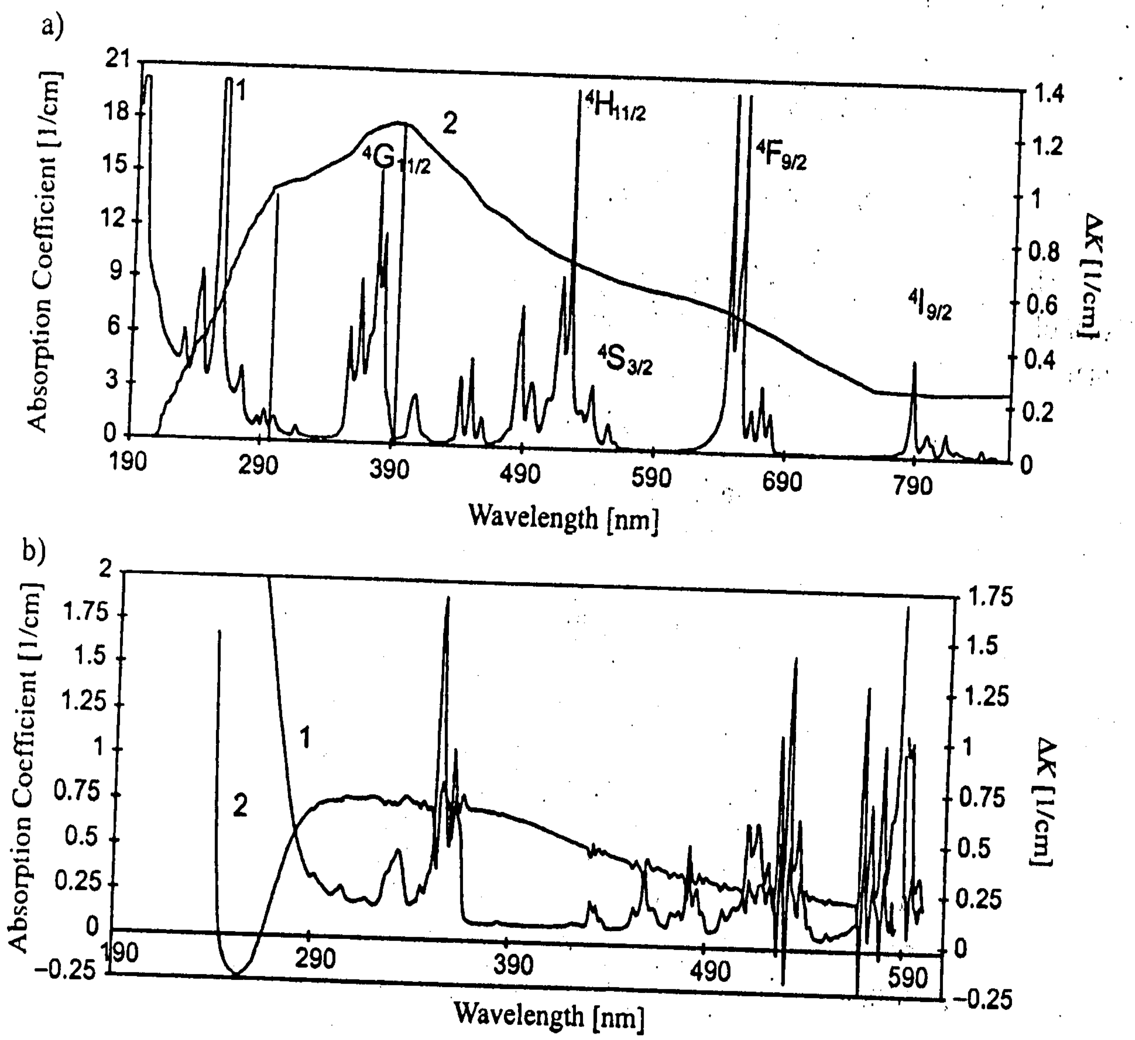

Fig. 1. Absorption spectrum (1) and additional absorption spectrum after gamma irradiation (2) of Er:YAG (a) and Nd:YAG (b) single crystals. Energy of gamma quanta was equal to 1.25 MeV and dose of 10^5 Gy

3.2. Lasing properties of YAG:Er

The lasing properties of YAG:Er crystals significantly change after gamma irradiation. The increase of the laser output energy is observed for YAG:Er after influence of gamma quanta. At the pumping level of 205 J the output energy increases from 75 mJ for an nonirradiated crystal to 135 mJ for the sample irradiated with the dose of 10^5 Gy (Fig. 2). The annealing of the crystals after irradiation at the temperature of 673 K during 3 hours restores the initial characteristics of the laser. The subsequent gamma irradiation again increases the laser power efficiency. The obtained results point out the direct influence of the colouring centers on the processes of formation of the inverse population of the laser levels of YAG:Er crystals.

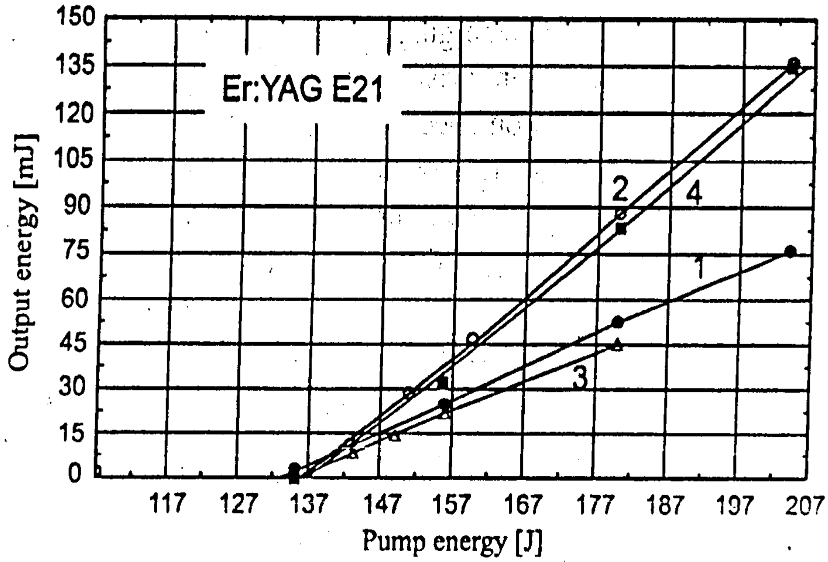

Fig.2. Output energy dependence on pumping energy for Er:YAG laser rod: 1 — before gamma irradiation; 2 — after gamma irradiation with dose of 10^5 Gy; 3 — after annealing in air during 3 hours at the temperature of 673 K of irradiated rod; 4 — after repeated gamma irradiation with dose $5 \cdot 10^4$ Gy.

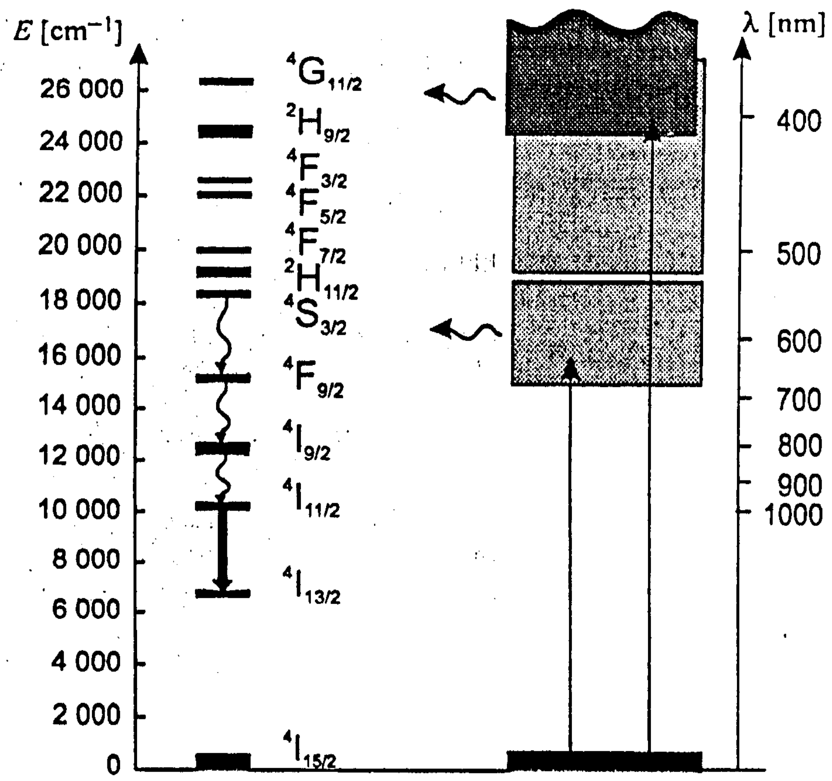

Fig. 3. The scheme of energy transfer from colour centers to Er^{3+} ions in YAG crystals.

After gamma irradiation of the crystals with garnet structure, as it was shown earlier [5], rearrangement and recharging of the defects causing the change of the pumping energy transfer efficiency to the emitting centers take place. We think that in the case of YAG:Er crystals the emerging colouring centers may fulfil the function of sensibilizators providing the increase of the pumping efficiency. Comparison of the AA spectra with the absorption spectrum of Er^{3+} ion (Fig. 1) shows that the AA bands are superimposed with the most intensive bands corresponding to the transitions from the main state $^4I_{15/2}$ to the multiplets of $^2H_{9/2}$, $^2H_{11/2}$ and $^4F_{9/2}$ through which the laser pumping takes place and the sensibilization may occur according to the scheme presented in the Fig. 3.

It should be noted, that in lasers generating in 2–3 μm range the passive losses due to the absorption of the colouring centers are absent because of great spectral distance from the value of the laser radiation frequency to the band of additional absorption.

4. Conclusion

After gamma irradiation of YAG:Er and YAG:Nd a wide complex of AA bands appeared. These bands are connected with CCs caused by gamma-induced recharging of growth defects that may be non-controlled impurities of Fe ions, oxygen vacancies or defects of cation sublattice.

In YAG:Er crystals CCs may fulfill the function of sensibilizer providing the increase of the pumping efficiency and output energy of laser generation at $\lambda = 2.94$ μm [12, 13].

Received March 21, 1996; revised June 4, 1996.

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Wpływ promieniowania gamma na optyczne i laserowe własności monokryształów $Y_3Al_5O_{12}$ domieszkowanych jonami Er^{3+}

Streszczenie. Przedstawiono wpływ promieniowania gamma na optyczne i laserowe własności kryształów granatu itrowo-aluminiowego domieszkowanego trójwartościowym erbem na poziomie 33at.%. Dla termicznie nie odprężonych kryształów otrzymano wzrost różniczkowej sprawności lasera Er:YAG rzędu 100%, powtarzalny po wygrzaniu w temperaturze 673 K i ponownym naświetleniu tą samą dawką kwantów gamma. W celu opisu powyższego zjawiska zaproponowano mechanizm indukowanej przez promieniowanie gamma sensybilizacji jonów (w wyniku bezpromienistych przejść) erbu przez centra barwne stabilne w temperaturze pokojowej.

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Гамма-индуцирована сенсибилизация лазерных свойств монокристаллов $Y_3AL_5O_{12}$ легированных ионами Er^{3+}

Резюме. Исследовалось влияние гамма-излучения на оптические и лазерные свойства монокристаллов Er:YAG. Увеличение выходной энергии лазера было отмечено после гамма--облучения лазерных стержней из Er:YAG, которые не подверагилсь термическому накаливанию. Обсуждается механизм радиационной сенсибилизации в этом случае.