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Influence of Ionizing Radiation on Performance of Nd:YAG Lasers

For crystals annealed in oxidizing atmosphere the influence of the dose on the shape of additional absorption spectrum of Nd:YAG samples after gamma and proton (energy of the order of 26 MeV) irradiation's was described. Changes in optical output of Nd:YAG laser after different value of gamma's dose were determined. The Nd:YAG crystals were also irradiated with 1 MeV electrons coming from Van de Graaf accelerator. Changes induced in the defect structure of the irradiated crystals were described.

Keywords: keyword, keyword, keyword

1. Introduction

Studies of the influence of the irradiation by various types of ionizing particles on the efficiency of solid-state lasers are motivated by a need of using them, in many applications, e.g. as range-finders, altimeters and others, in intense external radiation fields. For example, laser devices working in cosmic space are irradiated by electrons, protons and gamma rays of various energies, including the highest energy cosmic rays. During, say, five years of stay in the space, dependently on the orbit of the spacecraft the exposition dose acting on such device may reach values above 100 krad (10^3 Gy) and, for extraterrestrial missions, even an order or two orders of magnitude larger. Also, not meaningless is the question: how effective could various laser optical devices work after a nuclear explosion?

On the other hand, a positive influence of the ionizing radiation on the laser performance is also known. In particular, such phenomenon was observed in lasers based on Cr,Tm,Ho:$Y_3Al_5O_{12}$ and Er:$Y_3Al_5O_{12}$ crystals (KACZMAREK et al. 1995, MATKOVSKII et al. 1996).

The irradiation of a laser with Nd: $Y_3Al_5O_{12}$ (ND:YAG) crystal by a large radiation dose from a nuclear reactor (neutrons, electrons) causes a significant drop of slope efficiency, leading even to the total break-down of the emission (Bedilov et al. 1981). On the other hand, the irradiation of the same laser by smaller doses, of an order of 10 Gy for gamma quanta or 10^{11} electrons/cm^2, may increase the output energy of the laser by about 17 % (Bedilov et al. 1994). Authors of this paper suggest, that during the work of the laser in the radiation field, an additional pumping of the active ions (Nd^{3+}) takes place. Additionally, some extra defects (color centers - CC) can be acquired, for which their recombination energy, under the action of the optical pump, is transferred to the active ions. Thus, a weekly bound structure and complex defects, which appear during growth and further processing of the laser material, are cured by the irradiation. A significant increase of the output energy of a Nd:YAG laser, which was observed during irradiation by small doses of electrons (as compared to the effect of gamma irradiation) can be attributed to the more effective cure of the genetic structure imperfections.

ROSE et al. 1995 studied changes of optical output of Nd:LiYF$_4$ and Nd: YAG crystals (with a thickness of 1 mm) pumped longitudinally by laser diodes at 800 nm before and after irradiation by gamma's and 30-50 MeV protons. It was found, that both types of the radiation produce the same kinds of the color center lowering the output energy of lasers by absorption of the optical radiation emitted at the wavelength for which the generation takes place. Additionally, for the doses of the order of 10^3 Gy , an influence of the CC lead to a lowering of the slope efficiency of the laser only in the case of continuous pumping, while for the pulsed-mode pumping, no changes of the slope efficiency of Nd: YAG and Nd: LiYF$_4$ lasers were observed.

BEDILOV et al. 1994 and ROSE et al. 1995 performed their studies of the optical outputs of Nd: YAG lasers at room temperature. STELMAKH et al. 1992 studied the radiation-induced spectra at low temperatures, (down to 77 K). An increase of the intensity of the AA bands was observed with the decrease of the temperature at which the irradiation was performed. Authors of this work, analyzing the temperature dependence of basic absorption bands found also, that in the creation and relaxation processes of CC in Nd: YAG crystals two different kinds of the electron capture centers participate at least, of which one of those are Fe^{3+} ions.

AKHMADULIN et al. described possible defects, which can be created in YAG crystals by γ–irradiation after earlier heating in the oxidizing or reducing atmospheres. They are visible in the absorption spectrum of Nd: YAG crystals, giving AA-bands at λ_1=310 nm, λ_2=240-260 nm and λ_3=407 nm. The ESR studies of these crystals have shown, that bands λ_1 and λ_2 are connected with ions Fe^{2+} and Fe^{3+}, respectively. Additionally, in the band λ_2 a sub-band λ_{23} =258 nm was observed and it was prescribed to the presence of Fe^{2+} ions. Also sub-bands λ_{21}=246 nm and λ_{22}= 261 nm were seen, and they were attributed to Fe^{3+} ions. In the case of the crystal annealed in the oxidizing atmosphere, band λ_3 was attributed to the hole centers O^-, which are created by the photo-ionization of an electron or, to the hole vacancies V_0. In the case of the crystal annealed in the reducing atmosphere, band λ_3 was attributed to the presence of F centers, i.e. to those with two electrons localized in the oxygen vacancy.

To avoid changes of the optical properties of irradiated samples, Nd: YAG crystals were doped by Cr^{3+} ions (ROSE et al. 1995) or Ce^{3+} ions (ASHUROV et al. 1985), which in suitable concentrations can lower the sensitivity of YAG crystals to the irradiation.

The aim of the present work is to study the Nd: YAG crystals being subjected to irradiation by ^{60}Co gamma rays at doses within the range of 10^2-10^7 Gy as well as by 1 MeV electrons and 26 MeV protons. Effects of the radiation dose as well as influence of the kind of the annealing process on the character and type of the created radiation defects is studied. The laser optical outputs were measured for Nd: YAG crystals annealed in both, the oxidizing (air) or in the reducing atmosphere.

2. Experimental

For the present study, crystals obtained by Czochralski method in iridium crucibles in the nitrogen atmosphere (ITME), were used. Concentration of Nd^{3+} ions in the samples was ~1at.%. In Ce, Nd: YAG crystals, Ce concentration was ~0.05at.%.

2.1. Spectroscopic studies

To study of radiation-induced changes in optical properties of the Nd: YAG crystals, polished in both sides, parallel-plate samples of thickness from 0.5 to 3 mm were prepared. They were cut perpendicularly to the growth axis in the plane of (111). Optical transmission spectra were measured before and after each irradiation or thermal annealing processing of the samples using LAMBDA-2 PERKIN-ELMER, ACTA VII BECKMAN and FTIR 1725 PERKIN-ELMER spectrophotometers. The induced absorption (AA) was calculated from the formula:

$$\Delta K = 1/d \ln(T1/T2) \tag{1}$$

where K is absorption coefficient, d is the sample thickness, T1, T2 are optical transmissions of the samples before and after the irradiation (or heating), respectively.

For luminescence studies, the third additional surface was polished. The luminescence spectrum of γ-irradiated sample was measured in the range from 700 to 2000 nm and compared with such for an unirradiated crystal.

The samples prepared for the thermoluminescence (TL) studies, with the thickness lower that 1 mm and with the diameters up to 6 mm, were not polished. Measurements of the TL of „as grown" crystals as well as those for γ-irradiated ones were performed in the temperature range from 70 to 400°C by means of a roundabout-type WAWA-TLD RA'95 analyzer.

2.2. Irradiation of the crystals

For „as grown" crystals as well as those annealed an influence of the UV irradiation by a xenon lamp on the kind of the CC was investigated by irradiating of the samples placed in the laser resonator by a sequence of 10 light pulses from the xenon pump lamp. The separation time between pulses was 15 sec. Gamma irradiation's were performed using ^{60}Co sources. For the electron irradiation a 1 MeV beam from the Van de Graaf accelerator was used, while for 26 MeV protons a beam from compact isochronous proton cyclotron was applied.

The dose of gamma irradiation was varied from 10^2 to 10^7 Gy, fluencies of electrons from 10^{14} to $5*10^{16}$ particles/ cm^2 and for protons from $5*10^{12}$ to 10^{16} particles/cm^2.

2.3 Heating procedures

All the samples were irradiated for "as grown" as well as after annealing at 1400^{o}C in the oxidizing atmosphere and at 1200^{o}C in the reducing atmosphere. Annealing was performed for three regimes: 1-thermal relaxation was performed by annealing in air at 400^{o}C for 3 hours , 2-annealing in the oxidizing atmosphere by heating-up at 1400^{o}C for 3 hours in air and, 3-annealing in the reducing atmosphere by heating-up in the mixture of hydrogen and nitrogen at 1200^{o}C for 1.5 h in steps of 0.5 h.

2.4 Laser performance investigations

Both, irradiation and annealing processes were performed also using the Nd:YAG crystal rod of the length L = 45,63 mm, and diameter Φ = 4mm. The rod was introduced in the system of the Nd: YAG laser working in free-running emission regime. Additionally, the Ce, Nd: YAG (0.05 at. % Ce and 1 at. % Nd) rod with the dimensions L= 55 mm, Φ=4mm was investigated. These rods had no antireflective covers on its end surfaces and were naturally air-cooled.

For a study of the laser output the 21 cm parallel-plate resonator was used. The rod was placed inside the ellipsoidal reflector, made of brass covered by gold. Transmission of the enter mirror was 36.6 % for the wavelength of λ=1.06 μm. As the optical pump the arc xenon lamp was used of the diameter 4 mm, with the battery of 2×25 μF as a power supply. The pump energy was varied in the range from 7 to 50 J. The pump pulse duration was about 300 microseconds.

Radiation of the laser was detected by means of the pyroelectric head. The output energy of the laser pulses was measured by means of the Gen-Tec radiometer equipped with the measuring head of the type ED-500.

3. Results and discussion

3.1. Optical investigations

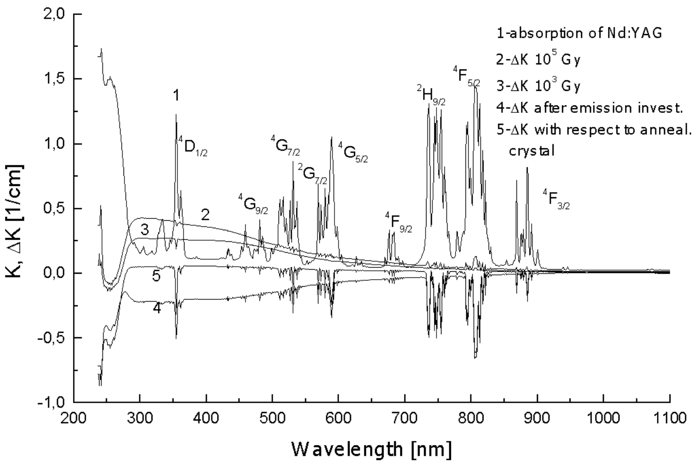

Fig. 1: Absorption (curve 1) and AA bands in Nd: YAG rod after γ-irradiation with doses of 10^5 (curve 2), 10^3 (curve 3) Gy and UV (curve 4) during laser emission. Curve 5 presents the difference in absorption of the rod after emission process with respect to annealing at 1400°C.

Measurements of the transmission, luminescence and TL before and after irradiation by gamma's, electrons and protons were performed for 13 samples of thickness about 3 mm (labeled further as " Y1,...Y13" counting from the seed) with the diameter of about 16 mm. There were cut from the same piece of the Nd: YAG single crystal from the start (Y1) to the end (Y13) of crystal growth surfaces.

Also, after each annealing process and optical output measurement of the laser, the optical transmission of the rod of L=45.63 mm length was measured.

3.1.1 „As grown" and annealed in reducing atmosphere Nd: YAG crystals

Absorption and the effect of gamma interaction on the absorption of Nd: YAG rod is shown in Fig.1. Similarly to the UV case, at least three CC at 253, 297 and 360 nm, respectively, are seen. Optical characteristics of the crystal return to its original state before the irradiation, after annealing for 3 hours at 400°C. The extension of these bands is larger than in the UV case and exceeds of 1.06 μm.

As one can see from the Fig. 1. UV radiation of a xenon pump lamp changes valency of Nd^{3+} ions inside YAG lattice.

Thermoluminescence measurements also show three type defects in Nd: YAG crystal. For the „as grown" crystal maximum of the TL appears at 158°C and at near 350°C. In the case of gamma irradiated Nd: YAG crystals (dose 10^3 Gy), at the heating rate of 3 K/s there are three peaks of thermoluminescence spectrum, at about 462, 517 and 563 K. In the frame of a simple model of Randall and Wilkins, the peaks are associated with the three different electron traps. To find the trap parameters we used the Randall-Wilkins formula (Randall, Wilkins, 1945):

$$I(T) = \sum_{i=1}^{4} n_{0i} s_i \exp(-\frac{E_i}{k_B T}) \exp(-\frac{s_i}{\beta} \int_{T_o}^{T} \exp(-\frac{E_i}{k_B T}) dT) \qquad (2)$$

where *I(T)* is the thermoluminescence intensity at the temperature T, β is the heating rate, n_0 the initial concentration of filled traps, E the trap depth, s the frequency factor, and k_B is the Boltzmann constant. This expression was fitted to the experimental points for YAG: Nd. A detailed description of the fitting procedure is given by Drozdowski et al. 1999. The results of the fit are summarized in Table 1.

trap	*E* [eV]	ln *s*
1	1.0	23.5
2	1.1	23.1
3	2.6	51.1

Table 1: The depths and frequency factors of the traps in YAG: Nd crystal.

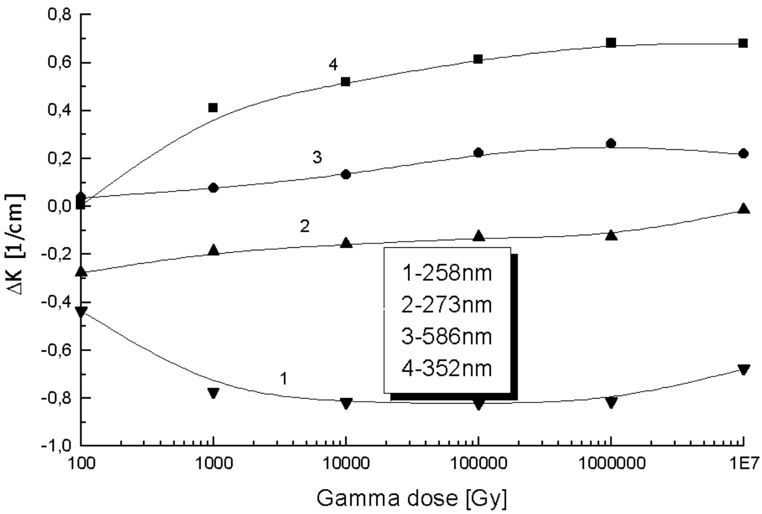

Fig. 2: Changes in AA band of annealed and next γ–irradiated Nd: YAG crystals as a dose function for four values of a wavelength.

The interpretation of these centers is similar to those for UV irradiation, first maximum, at 253 nm, corresponds to recharging of the Fe^{3+} ions , the second one at 297 nm is attributed to the existence of Fe^{2+} ions and the third one at 360 nm, to the CC of the F-type [9]. The same type of effects for the crystals annealed in reducing atmosphere are observed.

Thus, from these measurements one can conclude, that in the Nd: YAG crystal after the ionizing irradiation three different kinds of defects arise at least. They are recharged growth defects as well as new ones, which are the result of the recharging of optically active ions (e.g. Fe^{3+}), existing already in the Nd: YAG crystal lattice.

3.1.2 Nd: YAG crystals annealed in air

The AA values of previously annealed at 1400°C in oxidizing atmosphere Nd: YAG crystal for wavelengths of 258, 273, 352 and 586 nm as a function of the gamma dose is presented in Fig.2. It is seen a saturation effect after reaching the level of 10^6 Gy gamma dose.

The AA for Nd: YAG and pure YAG crystals, previously annealed at 1400° C in the air, and irradiated by protons and electrons are presented in Fig.3. As the irradiation dose increases, the deepening of the center attributed to the recharging of Fe^{3+} ions is observed, but the AA value due to the hole center increases.

3.2. Optical output measurements

Fig.4 illustrates changes in optical output of Nd: YAG rod after γ-irradiation. As it is seen, gamma irradiation causes, in general, worsening the optical output of the Nd: YAG laser, the larger when larger dose is applied (the slope efficiency of the rod decreases). This decrease is permanent when the rod is air cooled in the laser head, as it is in our case. In this case, after some 50 pumping pulses, the thermal equilibrium in the rod is established and the laser output characteristics remain stable.

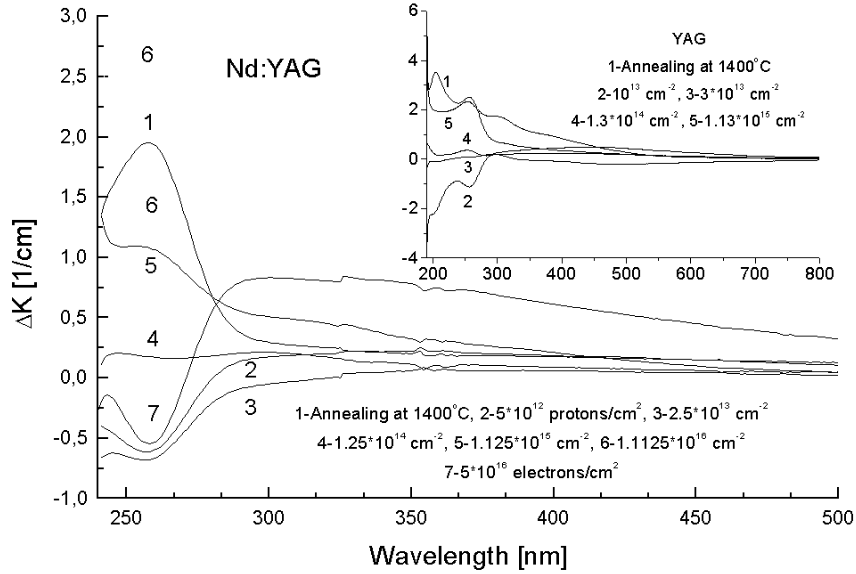

Fig. 3: AA bands of Y8 sample previously annealed (curve 1) and then irradiated by protons with doses: $2\text{-}5\times10^{12}$, $3\text{-}2{,}5\times10^{13}$, $4\text{-}1{,}25\times10^{14}$, $5\text{-}1{,}135\times10^{15}$ and $6\text{-}1{,}1135\times10^{16}$ cm^{-2} (two curves denoted by 6 describes two sides of the sample) and Y10 sample irradiated by electrons (curve 7) with a dose of 5×10^{16} cm^{-2}. Small picture show AA of annealed YAG crystal (curve 1) irradiated by protons with doses of: $2\text{-}10^{13}$, $3\text{-}3\times10^{13}$, $4\text{-}1.3\times10^{14}$ and $5\text{-}1.13\times10^{15}$ cm^{-2}.

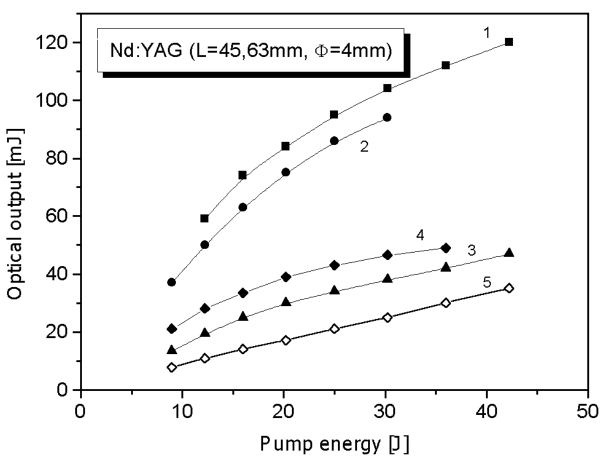

Fig. 4: Free-running laser emission from Nd: YAG: 1 - after annealing at 1400°C in O_2, 2 - after γ-irradiation with a dose of 10^3 Gy, 3 - after γ-irradiation with a dose of 10^5 Gy, 4 - after annealing at 1200°C in N_2+H_2 mixture and 5 - after γ-irradiation of previously annealed rod with 10^5 Gy.

Fig. 5 shows changes in the output energy of the Nd: YAG - gamma irradiated with 10^5 Gy dose - laser, as a function of the number of pumping pulses, with energy of 25 J and with the time interval of 5 sec's. It is seen that the state of the thermal equilibrium of the rod is reached after about 50 excitation pulses.

After gamma irradiation, lower output energies are obtained for the Ce, Nd: YAG (1 at. % Nd and 0.05 at. % Ce) rod as compare to the Nd: YAG one. It means that the concentration of Ce ions is too small in this rod. From the absorption curves it was calculated, that the concentration of Ce in Ce, Nd: YAG crystals was really of the order of 0.01 at. %.

3.3. Discussion

After irradiation of the Nd: YAG crystal by UV photons, gamma's, electrons and protons, in the absorption spectrum a broad additional absorption band appears in the range of 200-930 nm , which shape does not depend on the kind of the radiation, but depends on the kind of the atmosphere, in which the crystal was previously annealed.

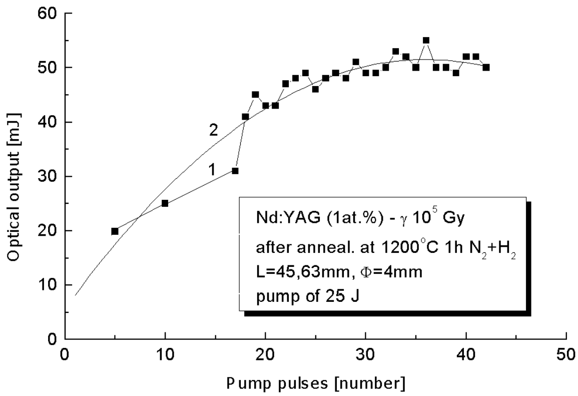

Fig. 5: Change in optical output of γ-irradiated (105 Gy) Nd: YAG laser as a function of pulse amount for pump energy equal to 25 J

Additional absorption peaks at 258, 278 and 350 nm arises (and, for crystal heated in air, at 586 nm). The shape of the AA-band for crystal heated in O2 differs from the corresponding curve for crystal heated in N_2 by the ratio of the amplitudes at 350 nm. Physically, in both cases we are dealing with two different kinds of defects: hole defect O- localized close to the defects of the cation sub-lattice and vacancies Vo, for crystal heated in the air and with F centers for the crystal annealed in nitrogen (KACZMAREK et al. 1996)]. With an increase of the γ–dose from 102 to 107 Gy, fluencies of electrons from 1014 to 1016 particles/cm^2 and of protons from 1012 to 1016 particles/cm^2, values of AA band became higher and higher. For crystals heated in air and next γ-irradiated, AA bands saturate at 106-107 Gy on the level of 0.75 cm-1. At 1016 protons/cm^2 or 5*1016 electrons /cm^2 we have dealing probably with the interaction of the particles with crystal leading to the production of the Frenkel-pairs and saturation effect is not observed.

It should be noted that the AA value for Nd: YAG crystals have similar shape and maximum absorption peak positions to that for pure YAG crystals. One may suppose (see Fig. 3), that defects responsible for these bands are characteristic for crystals with the YAG structure. The positions of the AA maxims in our crystals differs somewhat from their positions given by AKHMADULIN et al. 1992. This is due to the fact, that even when the same growth method is applied, one can get crystals which differ between themselves in some features due to the different real growth conditions (ASHUROV et al. 1985). As was shown by studies of YAG and Nd: YAG crystals performed a year after gamma irradiation

(KACZMAREK et al. 1996), return to starting transmission during their annealing at room temperature is very slow (AA drops about 10 %).

Properties of Nd: YAG laser using a gamma irradiated crystal are different from those for a non-irradiated crystal. Gamma irradiation lowers the slope efficiency of the pulse laser, contrary to results obtained by ROSE et al. 1995. After subsequent pulses output energy of the laser increases to the level, which comes out from the thermal equilibrium of rod being the heated by pumping pulses and air cooled. The output characteristics of the Ce, Nd: YAG laser , when the concentration of Ce is of the order of 0.05 at. % is worse than that for Nd: YAG laser. Behavior of the laser after γ-irradiation is the same type as previously mentioned for Nd: YAG one.

This increase of the laser energy after subsequent pumping pulses suggests that UV contained in the pump spectrum causes heating up the rod and accelerates those relaxation processes which decrease the AA.

The CC which were produced during gamma irradiation of the Nd: YAG crystal, cover the spectral region with the most intensive absorption bands 4I9/2→2G9/2 , 4G7/2, 2G7/2 and 4G3/2 (see Fig. 1). However, the efficiency of the Nd: YAG laser after gamma irradiation drops. This may suggest that in the Nd: YAG crystals, besides of possible sensitization process being a result of the energy transfer from CC to Nd3+ ions, we have dealing also with an increase of the passive losses at the wavelength of the laser emission (ROSE et al. 1995).

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