

# Effect of reactor radiation on the operation of a neodymium inorganic liquid laser

A.F. Dobrovolskii, D.V. Kabakov, A.A. Seregin, E.A. Seregina, G.V. Tikhonov

**Abstract.** The radiation parameters of a neodymium liquid laser based on phosphorus oxychloride are measured upon irradiation of the laser medium by neutrons and gamma-rays from a BARS-6 two-zone reactor. This irradiation increases the laser energy by 20 %–30 % compared to the laser energy in the absence of irradiation. The lasing threshold is observed to decrease with increasing the irradiation dose. In the case of simultaneous optical and nuclear pumping of the laser medium, the free-running regime of laser operation converts to the regime of high-power pulses.

**Keywords:** inorganic liquid laser, neodymium, irradiation, pulsed reactor.

## 1. Introduction

At present, work is in progress on the development of nuclear-optically pumped lasers [1–3], in which the regions of nuclear pumping and lasing are spatially separated. The region of nuclear pumping (converter) plays the role of a ‘nuclear lamp’ converting the nuclear fission energy into the light energy, which then pumps the active medium of a conventional laser. Because the active medium of such lasers is located, as a rule, in the field of high-power reactor radiation, it is of interest to study the effect of nuclear irradiation on condensed media which can be used in nuclear-optically pumped lasers.

To date, there are publications on investigations of the effect of nuclear irradiation on the operation of lasers based on ruby,  $\text{Nd}^{3+}:\text{YAG}$  crystals, and  $\text{Nd}^{3+}$ -doped glasses [4, 5], as well as on the operation of semiconductor lasers [6]. These researches showed that semiconductor lasers are most sensitive to irradiation. Their parameters begin to deteriorate at the neutron fluence of  $10^{13}$  neutron  $\text{cm}^{-2}$  [6]. A decrease in the output energy and an increase in the threshold pump energy of ruby lasers start after irradiation with a fluence of  $10^{15}$  neutron  $\text{cm}^{-2}$ . Such deteriorations in lasers based on  $\text{Nd}^{3+}:\text{YAG}$  crystals and  $\text{Nd}^{3+}$ -doped

glasses also appeared at the fluence of  $10^{15}$  neutron  $\text{cm}^{-2}$ , but they proceeded more slowly than in ruby lasers [4, 5]. A rather high radiation resistance of glass with  $\text{Nd}^{3+}$  ions allows one to use it as an active medium for nuclear-optically pumped lasers [2, 3]. At the same time, solid-state laser media under ionizing irradiation accumulate radiation-induced defects, which considerably reduce the service life of these lasers. Therefore, in our opinion, more promising for application in nuclear-optical lasers are condensed media based on inorganic liquids, which combine high dopant concentrations and the possibility of using the laser medium circulation to remove radiolysis products [7]. The inorganic liquid lasers surpass the neodymium glass lasers in energy characteristics. However, the behavior of such liquid lasers under ionizing irradiation has not yet been studied.

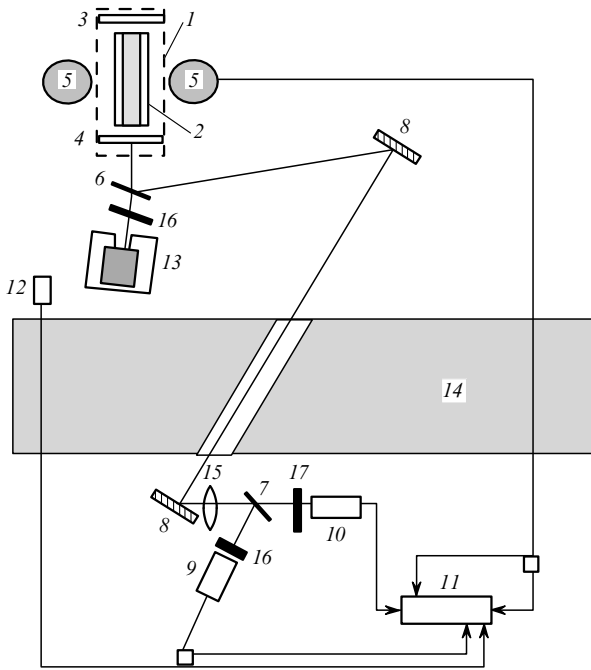
In this work, we study the effect of high-power nuclear radiation on the active medium and parameters of a neodymium laser based on the  $\text{POCl}_3\text{–SnCl}_4\text{–Nd}^{3+}$  inorganic liquid.

## 2. Experimental

The experiments were performed upon irradiation of the laser medium by neutrons and  $\gamma$ -rays of a BARS-6 two-zone pulsed reactor (IPPE, Obninsk). The scheme of the experiment is shown in Fig. 1. In the middle between reactor zones (5), we placed liquid laser (1), which consisted of laser head (2), enclosed in a cylindrical polyethylene moderator of neutrons, and cavity mirrors (3) and (4). The power supply unit of the xenon pump lamp was located in the measuring room separated from the reactor room by biological shielding (14). Cavity mirrors (3) and (4) had reflection coefficients of 0.99 and 0.75 and the curvature radii equal to 3 m and infinity, respectively. The cavity was 40 cm long. The output energy was measured with calorimeter (13). A part of radiation was reflected from dielectric mirror (6) with the reflection coefficient  $R = 0.13$  at the wavelength  $\lambda = 1.06$   $\mu\text{m}$  and was directed by mirrors (8) to the measuring room. Then, radiation passed through long-focus lens (15), was reflected from dielectric mirror (7) ( $R = 0.99$  at  $1.064$   $\mu\text{m}$ ), passed through an IKS-5 filter, and was focused on the entrance window of pin photodiode (9). The pump lamp radiation intensity was recorded with FD-24 photodiode (10). The reactor radiation intensity was measured using fission chamber (12), which was enclosed in the same polyethylene moderator of neutrons as that used for the laser head. Pulses from photodiodes (9) and (10) and fission chamber (12) were fed to the entrance channels of a Tektronix TDS

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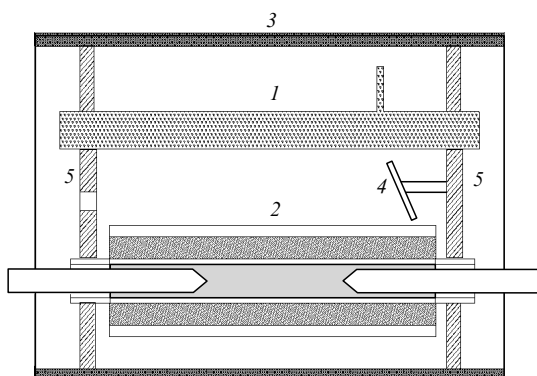
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**Figure 1.** Scheme of the experimental setup: (1) laser; (2) laser head surrounded by a polyethylene moderator of neutrons; (3, 4) cavity mirrors; (5) active zones of the reactor; (6, 7) dielectric mirrors with  $R = 0.13$  and  $0.99$ , respectively, at  $\lambda = 1.06 \mu\text{m}$ ; (8) metal mirrors; (9) pin photodiode; (10) FD-24 photodiode; (11) Tektronix TDS 1012 storage oscilloscope; (12) fission chamber; (13) IMO-2N calorimeter; (14) biological shielding; (15) lens ( $f = 42 \text{ cm}$ ); (16) IKS-5 light filter; (17) bandpass filter with the transmission band of  $360\text{--}580 \text{ nm}$ .

1012 storage oscilloscope operating online with a PC. The recording system was triggered with the help of a low-threshold surface barrier detector of fast neutrons and  $\gamma$ -rays emitted by the reactor.

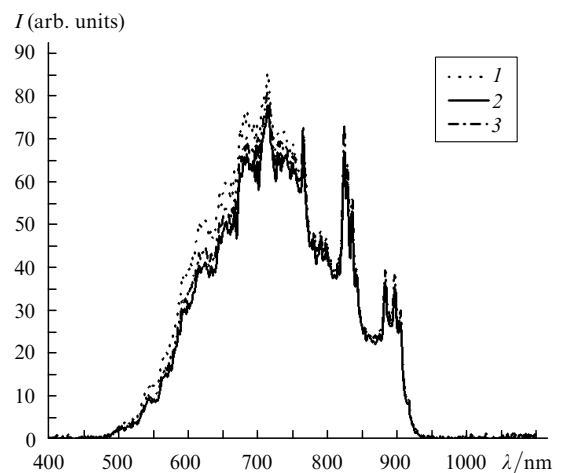
Let us consider the head of the liquid laser (Fig. 2) in more detail. Cylindrical ceramic silvered reflector (3) contains laser cell with liquid (1), xenon pump lamp (2) surrounded by a filter, and mirror (4), which directs the lamp radiation through an aperture in teflon holder (5) to the recording setup. We used an INP-3-7/80A pulsed xenon lamp. To suppress short-wavelength radiation and reduce heating of the laser matrix, the xenon lamp was surrounded



**Figure 2.** Laser head scheme: (1) quartz cell with a laser liquid; (2) xenon lamp with a liquid filter (1% aqueous solution of  $\text{K}_2\text{CrO}_4$ ); (3) cylindrical reflector; (4) mirror; (5) teflon holder.

by a filter of 1% aqueous solution of  $\text{K}_2\text{CrO}_4$  with a thickness of 1.5 mm. Time variations in the pump lamp radiation intensity were recorded using an FD-24 photodiode connected to the multichannel storage oscilloscope. In addition, we studied the effect of the lamp voltage and the neutron fluence on the spectrum and the emission intensity of the xenon lamp with the filter. The emission spectrum of this lamp was measured with an AvaSpec 2048FT spectrometer.

Figure 3 shows the emission spectra of the pump lamp with the  $\text{K}_2\text{CrO}_4$  filter, which were recorded in the absence of the cell with a laser liquid. One can see that, at the same lamp voltage, the emission intensity decreases at the instant of the reactor pulse, while the xenon emission spectral profile in the region of the filter transmission changes insignificantly. Simultaneously with the measurement of the spectrum, we measured the time dependences of the lamp emission intensity using an FD-24 photodiode, which also revealed a noticeable decrease in the intensity at the instant of the reactor pulse. The same changes in the emission spectrum and intensity were observed with increasing the lamp voltage from 1.9 to 1.7 kV. Our methodical studies showed that the intense nuclear irradiation of the INP-3-3/80A pulsed high-pressure xenon lamp decreases its emission intensity. Therefore, in addition to the energy and oscillogram of the laser pulse, we simultaneously recorded the oscillogram of the emission intensity of the xenon pump lamp. In all the experiments, the distance between the centres of the reactor active zones was 700 mm. The neutron fluence rate in the region of the laser element was  $(1.8 \pm 0.1) \times 10^{17} \text{ neutron cm}^{-2} \text{ s}^{-1}$ , and the neutron pulse duration was 180  $\mu\text{s}$ .



**Figure 3.** Emission spectra of the xenon pump lamp with a filter (1% aqueous solution of  $\text{K}_2\text{CrO}_4$ ) recorded before (1), synchronously with (2), and an hour after (3) the reactor pulse.

The active laser element was a quartz cell with plane-parallel end windows filled with the  $\text{POCl}_3\text{--SnCl}_4\text{--Nd}^{3+}$  laser liquid through a special nozzle. This liquid was prepared by us using the previously developed technique [8] and had the following characteristics: the neodymium concentration of  $1.8 \times 10^{20} \text{ cm}^{-3}$ , the lifetime of the upper laser level of the neodymium ion equal to 250  $\mu\text{s}$ , and the linear attenuation coefficient below  $10^{-2} \text{ cm}^{-1}$ . The laser cell had a length of 15 cm and an inner diameter of 7 mm.

The working length of the active element irradiated by the pump lamp was 10 cm.

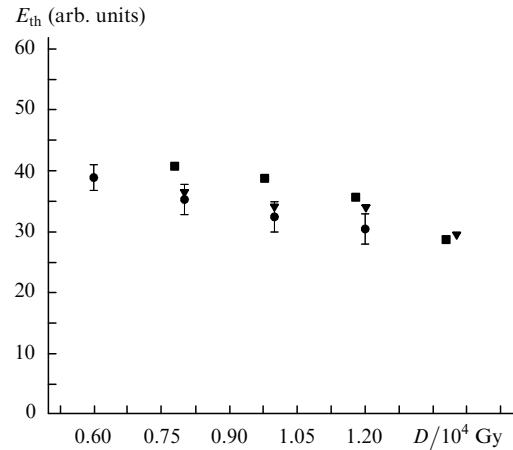
In addition, both before and several times during the experiments, we measured the background noise induced on the IMO-2N calorimeter by reactor radiation, which was found to be  $2.5 \pm 0.5$  mJ when both zones of the reactor operated. The corrections for the background noise were made when processing the data on the laser output energy measured at the instant of ionizing irradiation.

### 3. Experimental results and discussion

In experiments with the reactor, the laser active liquid  $\text{POCl}_3\text{-SnCl}_4\text{-Nd}^{3+}$  was irradiated by neutrons and  $\gamma$ -rays. Because neutrons used in nuclear-optically pumped lasers are presumably slowed down to thermal energies, we used the same neutron fluxes. For this purpose, the laser head with the liquid active element was surrounded by a polyethylene moderator. Our estimates showed that the main contribution to the excitation of the  $\text{POCl}_3\text{-SnCl}_4\text{-Nd}^{3+}$  active liquid was made by  $\gamma$ -rays and recoil nuclei in the  $^{35}\text{Cl}(n,\gamma)^{36}\text{Cl}$  reaction. Hereafter, the excitation of the medium by neutrons or  $\gamma$ -rays means the primary irradiation by these fluxes. In our experiments, the energy release in the active laser medium per one reactor pulse was  $2.7 \pm 0.1$  J  $\text{cm}^{-2}$ , which corresponded to an absorbed dose of  $1.6 \times 10^3$  Gy.

Thus, the active medium of the laser was pumped by a xenon lamp and simultaneously irradiated by reactor pulses. Table 1 lists the output energies  $E_g$  of the  $\text{POCl}_3\text{-SnCl}_4\text{-Nd}^{3+}$  laser averaged over six reactor pulses irradiating the active medium and the energies  $E_g$  in the case of a 200- $\mu\text{s}$  delay of the optical pump pulse with respect to the reactor pulse (averaged over two reactor pulses). The measurements were performed before, during, and an hour after the reactor pulse. One can see from Table 1 that the laser energy at the instant of the reactor pulse was somewhat higher than before the pulse and considerably higher than an hour after it. On average, at the same pump energies, this excess was 20 % with respect to the laser energy before the reactor pulse and 35 % with respect to the energy measured an hour after it.

Figure 4 shows the dependence of the threshold pump energy on the dose of ionizing radiation absorbed by the laser liquid. Before each reactor pulse, we performed a series of measurements of the laser output and pump energies. Figure 4 also presents the errors in the lasing threshold determined from the spread of data measured before each reactor pulse. One can see that the threshold pump energy  $E_{th}$  tends to decrease with increasing the absorbed irradiation dose. This tendency points, first, to a high radiation resistance of the active medium of the phosphorus oxychloride laser, and, second, to the possible appearance of additional excitation sources in the irradiated liquid. Such sources can be long-lived unstable isotopes formed in the



**Figure 4.** Dependence of the threshold pump energy  $E_{th}$  on the irradiation dose  $D$  measured before ( $\bullet$ ), at the instant of ( $\blacksquare$ ), and an hour after ( $\blacktriangledown$ ) the reactor pulse.

medium due to radiative neutron capture (for example,  $^{36}\text{Cl}$ ,  $^{121}\text{Sn}$ , and  $^{147}\text{Nd}$ ) and radiolysis products.

Figures 5a and b demonstrate typical oscillograms of the laser output and pump pulses recorded before and at the instant of a reactor pulse. The lasing pattern changes upon nuclear irradiation. From free-running lasing, which, in our case, has a spike character (Fig. 5a), the laser switches to the regime of individual high-power pulses (Fig. 5b). This lasing pattern was observed in all the cases when the reactor and pump pulses were synchronised. If the optical pump pulse was delayed by 200  $\mu\text{s}$  with respect to the reactor pulse, the lasing behaviour was different (Fig. 6). Comparing Figs 5a and 6, we can see that, in this case, the lasing pattern during the reactor pulse better resembles the free-running regime than the regime of individual high-power pulses. This lasing character upon irradiation of the medium by reactor pulses can be qualitatively explained by the appearance in the active medium of radiation-induced light absorbers with passive  $Q$ -switch properties, i.e. with the ability of switching from a saturated to a transparent state.

### 4. Conclusions

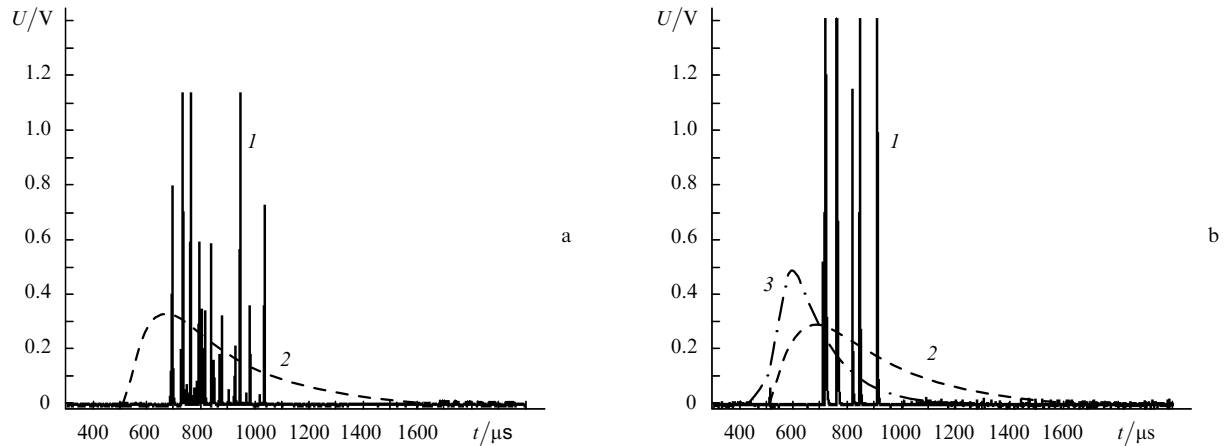
Thus, in this paper, we have obtained the following results. Upon synchronous irradiation of a liquid laser by optical and nuclear radiation, the laser energy increases by 20 % – 30 %. In this case, the threshold pump energy changes slightly and tends to decrease with increasing the absorbed dose of reactor radiation. The free-running regime is observed to transfer to the regime of individual high-power pulses. Finally, in none of the experiments the reactor radiation terminated the laser oscillation.

The obtained results allow us to make the following conclusions:

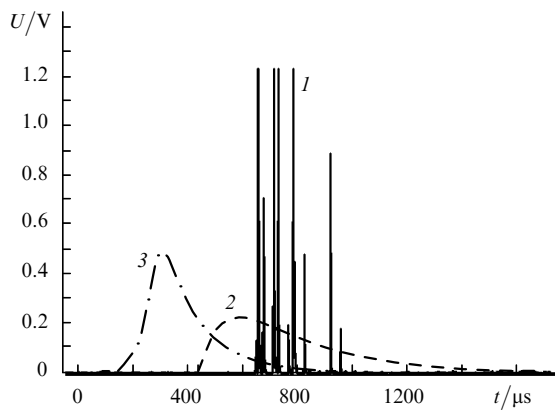
- (i) Inorganic liquid lasers reliably operate under irradi-

**Table 1.** Output energy  $E_g$  and pump energy  $E_p$  of the  $\text{POCl}_3\text{-SnCl}_4\text{-Nd}^{3+}$  laser.

Delay of the optical pump pulse with respect to the reactor pulse/ $\mu\text{s}$	$E_g/\text{mJ}$			$E_p/\text{mJ}$		
	Before the reactor pulse	At the instant of the reactor pulse	An hour after the reactor pulse	Before the reactor pulse	At the instant of the reactor pulse	An hour after the reactor pulse
0	$33.4 \pm 1.0$	$41.7 \pm 4.7$	$25.9 \pm 0.3$	$112.4 \pm 12.0$	$115.7 \pm 10.7$	$97.2 \pm 8.4$
200	$31.5 \pm 2.2$	$36.0 \pm 2.5$	$25.5 \pm 1.8$	$99.0 \pm 7.0$	$98.6 \pm 6.8$	$84.5 \pm 5.9$



**Figure 5.** Oscillograms of the  $\text{POCl}_3\text{-SnCl}_4\text{-Nd}^{3+}$  laser pulses (1) and pump pulses (2) before (a) and at the instant of nuclear irradiation (b); curve (3) corresponds to the pulse from the fission chamber.



**Figure 6.** Oscillograms of the output (1), pump (2), and reactor (3) pulses at a 200- $\mu\text{s}$  delay of the pump pulse with respect to the reactor pulse.

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ation by high-power reactor pulses without deterioration of the generation parameters, which points to a high radiation resistance of phosphorus oxychloride lasers;

(ii) The high-power reactor irradiation increases the liquid laser efficiency;

(iii) The  $\text{POCl}_3\text{-SnCl}_4\text{-Nd}^{3+}$  laser liquid can be recommended as a promising medium for the development of lasers and amplifiers with nuclear-optical pumping.

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