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Population kinetics of laser levels of neodymium ions in $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ excited by fission fragments

E.A. Seregina, A.F. Dobrovol'skii, D.V. Kabakov, A.A. Seregin, G.V. Tikhonov

***Abstract*. The population kinetics of laser levels of neodymium ions in the uranium-containing $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ laser liquid irradiated in a BARS-6 pulsed reactor is studied experimentally and theoretically. The relations between the populations of the upper and lower laser levels are studied at different time moments with respect to the pump pulse. An inverse population of laser levels is observed and found to be directly proportional to the pump power. The efficiency of pumping of the upper laser level of neodymium ions in the laser medium by uranium fission fragments is determined, and quantitative information is obtained on the inverse population, gain, and their dependences on the pump power.**

Keywords: *uranium- and neodymium-containing liquid, phosphorus oxychloride, nuclear pumping by fission fragments, population kinetics, inverse population.*

1. Introduction

The development of new laser technologies and their commercial application require various inexpensive sources of laser radiation. In the future, some of such sources can be nuclear-pumped lasers and laser amplifiers, in which the nuclear fission energy is transformed into the energy of laser radiation [1, 2]. Today there are known even more than thirty various gas media lasing under pumping by fission fragments [3], and the principal possibility of energy conversion from chain fission reaction into laser radiation is shown [4]. However, the obtained parameters of the fission energy conversion into laser radiation of available gas media do not satisfy specialists. This stimulates the search for new laser media in which the kinetic energy of fission fragments is converted into laser radiation more efficiently.

One of the ways in this direction is to use liquid laser media instead of gases. Laser-active liquids are more promising than gaseous media. In liquids the kinetic energy of fission fragments is converted into the energy of excited active ions almost ten times more efficiently than in gases. Liquid laser-active media have extremely high energetics. In particular, optical pumping of a laser liquid based on phosphorus oxychloride ($POCl_3 - SnCl_4 - Nd^{3+}$) allowed one to achieve an output energy of ~ 6 J for a 150-μs pulse from a cubic centimetre of the active medium [5]. These advantages of nuclear-pumped liquid lasers can be realised only in the case of successful solution of two problems. First, it is necessary to find an uranium-containing laser material with a high radiation resistance, a low absorption at the generation wavelength, a low viscosity (for better circulation), and without precipitation both when operating and in storage. Second, one must obtain lasing in this uranium-containing medium under irradiation by a reactor, which cannot be done without understanding physical processes proceeding in liquid laser media upon nuclear pumping.

We were the first to develop and patent [6] a technology to synthesise a new-generation uranium-containing laser liquid ($POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$), which satisfies all the above requirements and has a concentration of neodymium ions up to 5×10^{20} cm^{-3} and an uranium-235 concentration up to 5×10^{19} cm^{-3}. This material has already demonstrated lasing under optical pumping. Thus, the problem of creating an uranium-containing laser liquid for nuclear pumping is solved. However, lasing in this medium under nuclear pimping has not yet been achieved.

Our previous experimental studies [7] and model calculations [8] showed that the efficiency of conversion of the kinetic energy of fission fragments into the excitation energy of neodymium ions in the inorganic laser liquid $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ is high enough to create a nuclear-pumped liquid laser operating in a four-level scheme. In the case of optical pumping, the four-level approximation is quite satisfactory to describe the operation of neodymium lasers. However, under nuclear pumping, laser levels are excited via a different, much more complicated mechanism. Hence, in this case, it is very important to study the population kinetics and efficiency not only for the upper but also for the lower laser levels of the neodymium ion.

In this paper, we consider the population kinetics of the lower and upper laser levels of neodymium ions upon nuclear pumping and determine the time domain of existence of inverse population and its dependence on the energy deposition by fission fragments in the laser medium.

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2. Theoretical simulation of population of Nd^{3+} laser levels under nuclear pumping

To experimentally study the population kinetics of the upper and lower laser levels of neodymium ions in a laser medium (Fig. 1) excited by fission fragments, we propose a method similar to the luminescence dumping method described in literature [9, 10]. The essence of the method consists in measuring a change caused in the intensity of the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition of the neodymium ion in a nuclear-pumped medium by the propagation of a probe laser beam with a wavelength resonant with the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ laser transition.

Figure 1 shows the energy level diagram of the Nd^{3+} ion. The population of the ith level N_i of the Nd^{3+} ion in an unexcited medium is described by the Boltzmann distribution

$$N_i(T) = N_t \exp\left[-\Delta E_i/(kT)\right], \tag{1}$$

where N_t is the concentration of neodymium ions in the medium; ΔE_i is the energy difference between the ith level and the ground state; k is the Boltzmann constant; and T is the medium temperature in kelvins. The energy of the upper $^4F_{3/2}$ laser level of the neodymium ion in the studied medium can be determined from the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ luminescence spectra. The lower level energy is difficult to measure directly due to a high absorption of the medium in the IR spectral region. At the same time, Fig. 1 shows that, knowing the experimental energies of the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ and $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transitions, one can determine the position of the $^4I_{11/2}$ level. It was found that these energies in $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ laser liquids are 1.389 and 1.179 eV, respectively, and, hence, the energy of the lower laser level is 0.21 eV. Thus, the population of the lower $^4I_{11/2}$ laser level at a temperature of 293 K is $3.24 \times 10^{-4} N_t$, while the population of the upper laser level is more than 20 orders of magnitude smaller than N_t, i.e. the upper level at room temperature is almost completely empty compared to the lower level. When a probe laser beam with a wavelength resonant with the $^4I_{11/2} \rightarrow {}^4F_{3/2}$ transition propagates through an unexcited medium, a part of this beam is absorbed and excites neodymium ions from the $^4I_{11/2}$ state to the upper excited $^4F_{3/2}$ state. This causes a spontaneous luminescence from the upper laser level to all the lower-lying levels including the ground state. The luminescence from the upper laser level to the ground state induced by a probe laser beam can be measured with appropriate devices.

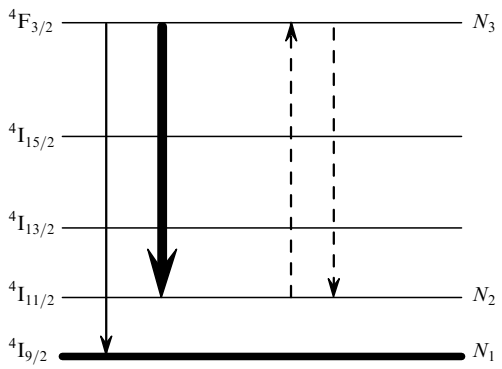

Figure 1. Energy level diagram of the Nd^{3+} ion . The thick arrow shows the laser transition, the thin arrow presents the studied transition, and the dashed arrows show the transitions corresponding to the absorption or amplification of the probe laser radiation.

The excitation of a medium by fission fragments causes spontaneous luminescence of neodymium ions. As a probe laser beam propagates through the medium, the population of the upper $^4F_{3/2}$ laser level rapidly changes due to either absorption or amplification of radiation with the wavelength $\lambda = 1053$ nm. This results in a change in the luminescence intensity at the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition. If this intensity does not change, this means that the populations of the upper and lower levels are equal to each other. Thus, the change in the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ luminescence intensity at the instant of propagation of the resonance laser beam allows one to estimate the population ratio of the upper ($^4F_{3/2}$) and lower ($^4I_{11/2}$) laser levels of the neodymium ion in the $POCl_3 - SnCl_4 - UO_2^{2+} - Nd^{3+}$ laser liquid excited by fission fragments. These populations are described by the system of equations

$$\begin{aligned} \frac{dN_3}{dt} &= w_{13}(N_1 - N_3) + \sigma I(N_2 - N_3) - \frac{N_3}{\tau_3}, \\ \frac{dN_2}{dt} &= w_{12}(N_1 - N_2) - \sigma I(N_2 - N_3) + \frac{N_3}{\tau_{32}} - \frac{N_2 - N_{20}}{\tau_2}, \end{aligned} \tag{2}$$

where N_3 and N_2 are the concentrations of neodymium ions at the upper and lower laser levels, respectively; N_{20} is the equilibrium concentration of neodymium ions at the lower level [see Eqn (1)]; w_{12} and w_{13} are the nuclear pump rates of the upper and lower levels; σ is the stimulated emission cross section at the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition; I is the photon flux density in the laser pulse; τ_2 and τ_3 are the lifetimes of the lower and upper laser levels; and $1/\tau_{32}$ is the probability of the radiative $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition.

The nuclear pump rate of the lower laser level can be written in the form [8]

$$w_{12}(t) = \frac{\delta_2 \varepsilon_f I_f(t)}{N_t h\nu_{21}}, \tag{3}$$

and the pump rate for the upper level is

$$w_{13}(t) = \frac{\delta_3 \varepsilon_f I_f(t)}{N_t h\nu_{31}}. \tag{4}$$

In these expressionsx $\varepsilon_f = 172$ MeV is the energy released in the fission of one uranium-235 nucleus; δ_2 and δ_3 are the efficiencies of pumping of the lower and upper levels; $h\nu_{21}$ and $h\nu_{31}$ are the energies of transitions from the ground $^4I_{9/2}$ level to the $^4I_{11/2}$ and $^4F_{3/2}$ levels, respectively; and $I_f(t)$ is the intensity of uranium fission in 1 cm^3 of a laser liquid, which, for a two-zone pulsed reactor, is well described by the expression [8]

$$I_f(t) = \frac{E_r \varkappa}{2\theta_{las} V} \exp\left(\frac{\theta_r^2}{4\pi\theta_{las}^2} - \frac{t - t_m}{\theta_{las}}\right) \times \left\{1 + \operatorname{erf}\left[\frac{\sqrt{\pi}(t - t_m)}{\theta_{las}} - \frac{\theta_r}{2\theta_{las}\sqrt{\pi}}\right]\right\}. \tag{5}$$

Here, $E_r = 2 \times 10^{17}$ is the number of fissions per pulse in the two zones of the reactor; V is the volume of liquid in the cell; $\varkappa$ is the experimentally measured geometric factor dependent on the cell position with respect to the reactors; $t_m = 165$ μs is the time for which the reactor pulse reaches

the maximum; $\theta_r = 80$ μs is the neutron pulse duration in the active zone of the reactor; $\theta_{las} = 140$ μs is the neutron pulse duration after neutron moderation in the region of the cell with the uranium-containing laser liquid; and $\mathrm{erf}(x)$ is the error function.

When solving the system of equations (2), it is necessary to take into account that $N_1 + N_2 + N_3 = N_t$, $N_2(0) = N_{20}$ and $N_3(0) = 0$. Then, system (2) is transformed into the system of equations

$$\frac{dN_3}{dt} = w_{13}(N_t - N_2 - 2N_3) + \sigma I(N_2 - N_3) - \frac{N_3}{\tau_3},$$

$$\frac{dN_2}{dt} = w_{12}(N_t - 2N_2 - N_3) - \sigma I(N_2 - N_3) + \frac{N_3}{\tau_{32}} - \frac{N_2 - N_{20}}{\tau_2}. \quad (6)$$

System (6) cannot be solved analytically and was solved numerically. Figure 2 shows the populations of the lower ($^4I_{11/2}$) and upper ($^4F_{3/2}$) laser levels of the neodymium ion in the $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ laser liquid calculated in the absence of nuclear pumping. In calculations, we used the following typical parameters of a liquid laser medium: $N_t = 1.8 \times 10^{20}$ cm^{-3}, $N_{20} = 1.2 \times 10^{-4} N_t$, $\sigma = 8 \times 10^{-20}$ cm^2, $\tau_3 = 200$ μs, $\tau_{32} = 830$ μs, and $\tau_2 = 0.01$ μs. The stimulated emission cross section σ and the radiative lifetime τ_{32} are taken from [11]. One can see from Fig. 2 that the laser pulse with an energy density of 1 J cm^{-2} excites a part of neodymium ions from the lower to the upper laser level due to the resonance absorption of light. At the instant of pulse propagation, the concentration of ions sharply decreases at the lower level and increases at the upper level. Then, the equilibrium at the lower level is rapidly restored due to the short lifetime τ_2, and the concentration of neodymium ions at the upper level begins to decrease according to its lifetime, which, in this case, is 200 μs.

In the case of nuclear pumping, the situation is more complicated. Figure 3 shows the time dependences of the upper level population N_3, of the equilibrium population N_{20} of the lower laser level, and of the lower level population $N_2 - N_{20}$ induced by fission fragments, which were calculated for the pump energy densities $E_{in} = 7$ and 14 J cm^{-3}. Here E_{in} is the specific energy deposition equal to the kinetic energy of fission fragments absorbed in 1 cm^3 of a liquid. The efficiency δ of pumping of the upper laser level was taken to be 1 %. As can be seen from Fig. 3, in the case of pumping by fission fragments, the relation between the populations of the lower and upper laser levels changes at two successive points on the t axis. These points divide the plots into three successive time regions. In the first region, the population N_2 of the lower laser level exceeds the population N_3 of the upper laser level. In the second region, N_2 becomes smaller than N_3, which corresponds to the inverse population. In the third region, N_2 is again higher than N_3. The calculation results given in Fig. 3 show that the region of inverse population extends with increasing the energy deposition by fission fragments in the laser medium. The first of these points at the t axis allows one to determine the minimum pump threshold needed to obtain inversion in the laser liquid. This threshold for the used liquid was found to be 1.6 J cm^{-3}. This minimum pump energy density

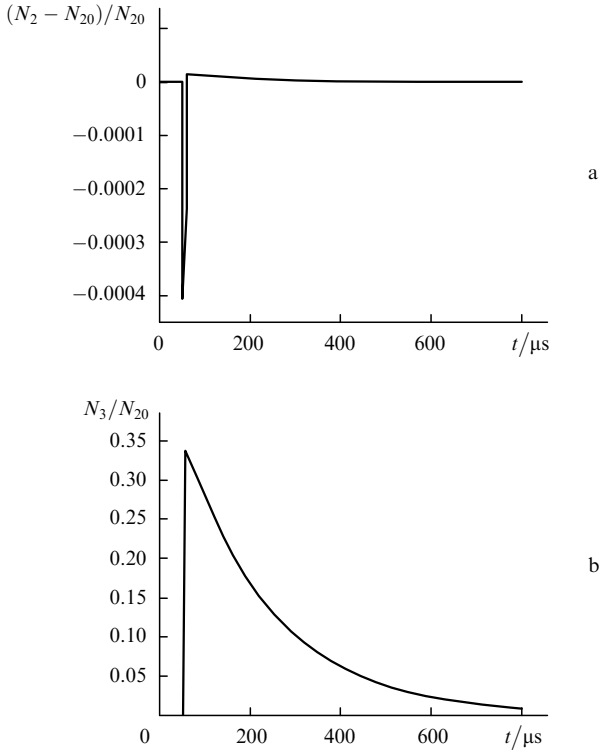

Figure 2. Calculated relative populations of the lower (a) and upper (b) laser levels of Nd^{3+} in the laser medium upon propagation of a probe pulse with a wavelength of 1053 nm, a duration of 10 μs, and an energy density of 1 J cm^{-2}.

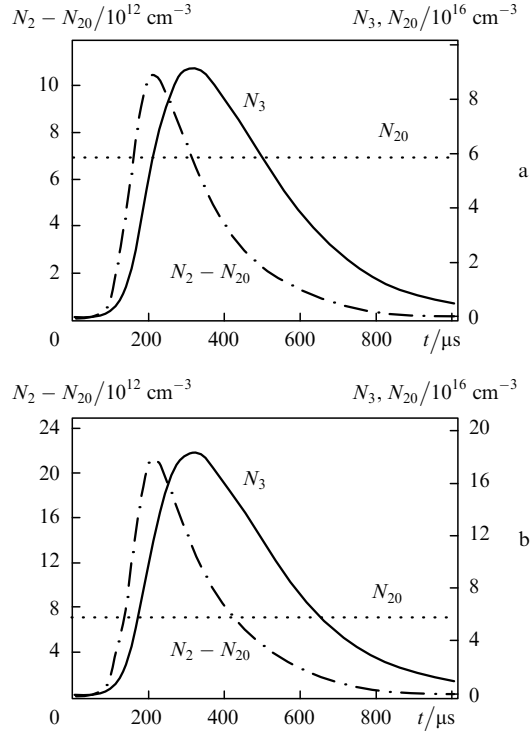

Figure 3. Calculated populations of the lower ($N_2 - N_{20}$) and upper (N_3) laser levels in the liquid excited by fission fragments with energy depositions of 7 (a) and 14 J cm^{-3} (b) at $N_t = 1.8 \times 10^{20}$ cm^{-3}, $N_{20} = 5.8 \times 10^{16}$ cm^{-3}, and $\delta = 1$ %.

corresponds to the minimum theoretical lasing threshold in the ideal case of the absence of losses on the walls of the cell with the laser liquid and on the cavity mirrors, as well as of losses related to the light absorption and scattering in the laser medium.

Figure 4 presents the populations of the upper and lower laser levels calculated in the case of propagation of a probe pulse with an energy density of 1 J cm^{-2} and different delays with respect to the onset of the pulse of pumping by fission fragments with an energy deposition of 7 J cm^{-3}. If the probe pulse propagates with a delay of 150 μs with respect to the time moment $t = 0$ (i.e. in the region where $N_2 > N_3$), the absorption of the probe light sharply decreases the population of the lower laser level [curve (*1*) in Fig. 4a] and increases the population of the upper laser level [curve (*1*) in Fig. 4b]. When the probe pulse delay is 320 μs, we observe the opposite pattern: the nonequilibrium population of the lower level [curve (*2*) in Fig. 4a] increases and the population of the upper laser level [curve (*2*) in Fig. 4b] sharply decreases.

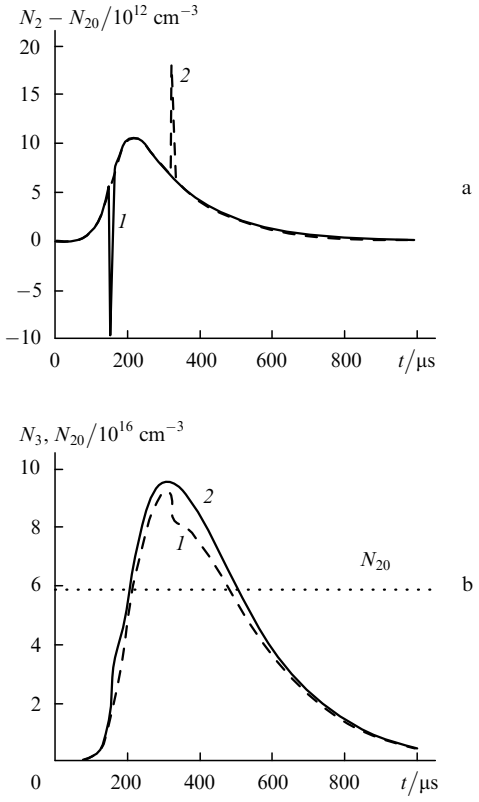

Figure 4. Time dependences of the populations of the lower (a) and upper (b) laser levels in the medium pumped by fission fragments with an energy deposition of 7 J cm^{-3} upon propagation of a probe pulse with delays of 150 (*1*) and 320 (*2*) μs.

It should be noted that the maximum increase in the lower level population with respect to the equilibrium population in the process of nuclear pumping is small and does not exceed one percent.

Thus, our calculations show that the pumping of a neodymium-doped laser liquid by fission fragments with an energy deposition exceeding the minimum theoretical threshold creates an inverse population of laser levels. Since the luminescence intensity is proportional to the upper level population, the changes similar to calculated (Fig. 4) can be observed in the intensity of the luminescence from the upper to the ground level experimentally measured at the instants of propagation of a probe pulse with different time delays with respect to the pump pulse. In this case, according to calculations, the inversion should increase with increasing the specific energy deposition in the medium.

3. Experimental technique

The experimental technique and the results of preliminary experiments were considered in detail in our works [12, 13]. Here, we will briefly describe the scheme of experiments in a BARS-6 reactor (Fig. 5). Hermetically sealed quartz cell (*2*) with the studied medium ($POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$) was placed in polished cone (*3*) and positioned between active zones (*1*) of the pulsed reactor. Since the main contribution to the medium excitation is made by the fission fragments of uranium-235 isotopes, the cell was enclosed in neutron moderator (*4*) in order to increase the number of fissions. The cell with the studied liquid excited by fission fragments was irradiated from the measuring room by a shot of probe laser (*5*) with the wavelength $\lambda = 1053$ nm resonant with the laser transition.

As a probe laser, we used a *Q*-switched $POCl_3 - SnCl_4 - Nd^{3+}$ laser with passive *Q*-switch (*7*) (a Cr^{4+} : YAG crystal). The pumping and recording units were triggered synchronously with the pulse of the BARS-6 reactor. The probe beam energy was measured with calorimeter (*23*), which received a portion of laser radiation from quartz plate (*22*). The laser pulse was recorded with pin-photodiode (*25*). The laser pulse energy was 0.7 ± 0.1 J and the pulse duration was 20 ns. With the use of a system of mirrors, the laser radiation was directed to the cell placed in the reactor room. The luminescence from the cell was directed by cone (*3*) to spherical mirror (*14*) and then by mirrors (*12*) and (*13*) to plane aluminum mirror (*15*) with an aperture of 10 mm in diameter, through which the probe laser beam entered the reactor room. The luminescence reflected from mirror (*15*) was sent by mirrors (*16*), (*17*), and (*18*) to the measuring room, where it was focused by lens (*20*) on the window of FEU-100 photomultiplier (*21*). In front of the FEU window, we placed a band-pass filter that transmitted only the luminescence of neodymium from the upper laser level to the ground state ($\lambda = 880 \pm 50$ nm).

To measure the fission rate during the reactor pulse, we used vacuum fission chamber (*26*) located in a special channel of the neutron moderator in the close vicinity to the studied cell. The pulses from the fission chamber, FEU-100, and pin-photodiode were fed to a recording system consisting of TDS-220 and TDS-1012 two-channel storage oscilloscopes and a PC. The recording system, including detectors, had a rather high time resolution (better than 1 μs) and caused no noticeable distortions in the shape of measured signals.

To calculate the pump power, the temporal intensity distribution of the signal from the fission chamber was normalised to the energy distribution by fission fragments per reactor pulse. The latter was determined by the known amount of uranium in the cell, by the measured number of fissions per pulse, and by the total kinetic energy of fission fragments in a fission event. The number of fissions per pulse was measured using a miniature glass tracking detector and a calibrated uranium foil, which were located

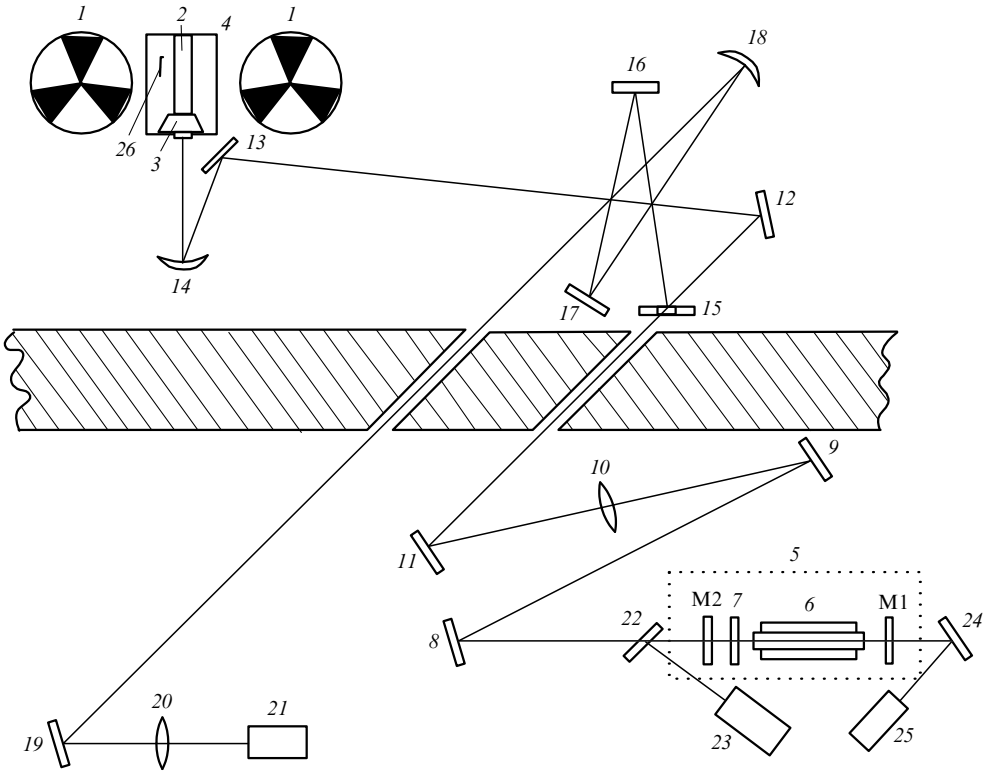

Figure 5. Scheme of the experiment in a BARS-6 reactor: (*1*) active zones of the reactor; (*2*) cell with uranium-containing laser liquid; (*3*) cone; (*4*) neutron moderator; (*5*) probe laser based on the $POCl_3 - SnCl_4 - Nd^{3+}$ liquid; (*6*) pump cavity; (*7*) passive *Q*-switch; (M1, M2) cavity mirrors with reflectivities of 99 % and 17 %, respectively; (*8, 9, 11–13, 16, 17, 19*) plane mirrors; (*10*) lens; (*14*) spherical copper mirror with the curvature radius $R = 4$ m; (*15*) plane aluminium mirror with an aperture in the centre; (*18*) spherical copper mirror with $R = 6$ m; (*20*) lens; (*21*) FEU-100; (*22, 24*) quartz plates; (*23*) IMO-2N calorimeter; (*25*) pin-photodiode; (*26*) fission chamber.

on the cell surface in the channel of the polyethylene moderator.

Upon irradiation of the $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ laser liquid by neutrons of the pulsed reactor, the uranium nuclei decay into two fragment. When the fission fragments are slowed down in the liquid, their energy is spent for ionisation and excitation of particles, including Nd^{3+} ions. In such an excited medium, both the upper and lower laser levels of neodymium ions will be populated. It is expected that the propagation of a high-power resonance probe beam will induce a fast change in the population of the upper laser $^4F_{3/2}$ level due to either absorption or amplification of radiation with $\lambda = 1053$ nm. This must change the intensity of luminescence at the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition, which is measured with appropriate devices. If the intensity does not change, then the populations of the upper and lower laser levels are the same and the liquid remains transparent for the probe radiation.

It is important to note that, in this method, due to the small volume of luminescent liquid (below 0.8 cm^3), which is scanned by 2-cm high cone (*3*), neither the inactive losses nor the losses induced in the medium by fission fragments must be taken into account because they almost do not affect the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ luminescence intensity due to the small distance propagated by light in the laser liquid.

4. Experimental results and discussion

For experiments, we prepared two laser liquids with identical concentrations of neodymium ions ($N_t = 1.8 \times 10^{20}$ cm^{-3}) and different uranium concentrations, 1.8×10^{19} cm^{-3} (No. 1) and 7.0×10^{19} cm^{-3} (No. 2). The laser liquids were synthesised by a previously developed technique [6].

Prior to experiments, we measured the luminescence intensity of a sample placed between the active zones of the reactor under the action of a probe laser pulse. Figure 6 shows typical oscillograms of a probe pulse and of a signal of the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ luminescence. This figure demonstrates a

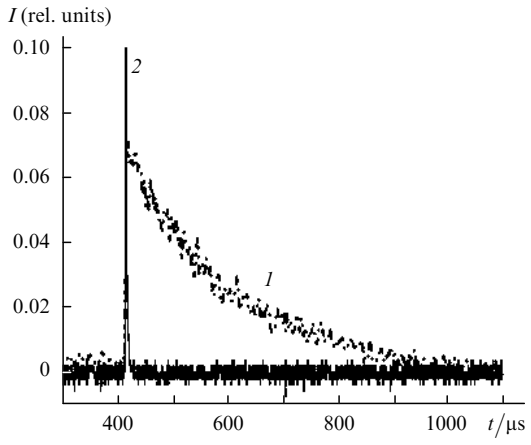

Figure 6. Oscillograms of a luminescence pulse at the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition in the neodymium ion (*1*) and a probe laser pulse (*2*) propagating through the medium in the absence of a reactor pulse.

rather high sensitivity of the recording channel. The energy deposition by fission fragments was 5 ± 0.5 J cm^{-3} in the case of liquid No. 1 and $10-14$ J cm^{-3} in the case of liquid No. 2. In all the cases, we recorded an inverse population of the upper laser level in the region of the maximum spontaneous radioluminescence. The results of investigation of liquid No. 1 are described in [13]. In this study, we investigate in detail liquid No. 2.

Figure 7 presents the luminescence intensity of the neodymium ion at the transition from the upper laser level to the ground state observed in liquid No. 2 excited by fission fragments and irradiated by a probe laser pulse at different instants with respect to the reactor pulse. One can see from Fig. 7a that, in the initial period of the reactor pulse, the luminescence intensity increases under the action of the laser pulse. In this case, the probe pulse passes ahead of the pump pulse maximum by $\Delta t = -115$ μs. However, even at $\Delta t = -55$ μs, the laser pulse causes no changes, i.e. the populations of the of the upper and lower laser levels at this instant become identical due the nuclear pumping (Fig. 7b). As the probe laser pulse shifts to the luminescence pulse maximum, one observes a strong decrease in the luminescence intensity. The time delay of the laser pulse with respect to the pump pulse maximum in Figs 7c and 7d is 165 and 235 μs, respectively. The luminescence intensity decreases under the action of laser pulses until the populations of the upper and lower levels become identical again. In the case of liquid No. 2, this occurs when the laser pulse is delayed from the pump pulse maximum by about 510 μs (Fig. 7e). With a further increase in the delay of the laser pulse (Fig. 7f, $\Delta t = 800$ μs), the upper level population becomes higher than the population of the lower level and the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ luminescence intensity again sharply increases at the instant of the laser pulse propagation.

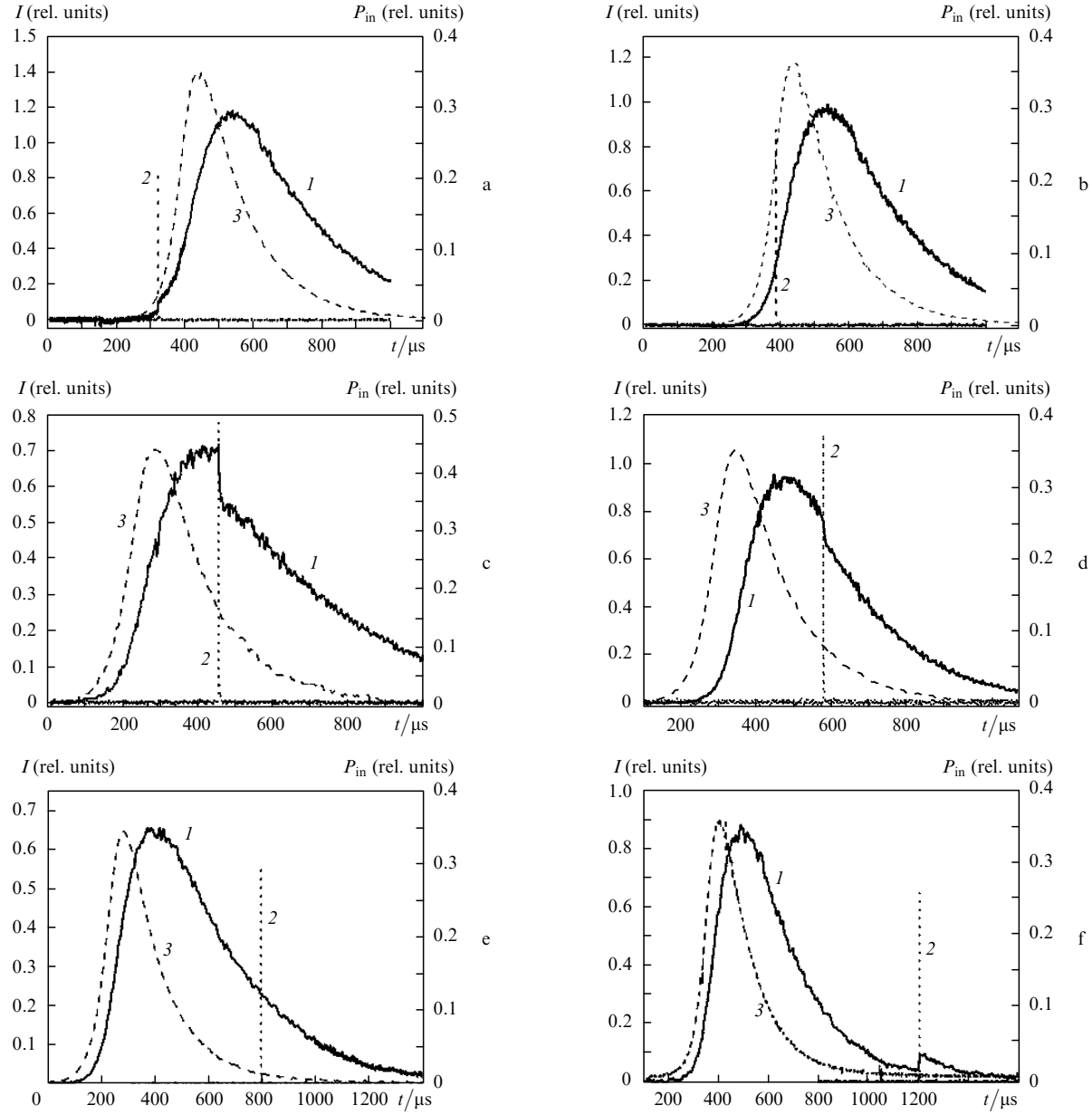

Figure 7. Time dependences of the luminescence intensity (*1*), of the probe laser radiation (*2*), and of the pump power density (*3*) for different instants of probe pulse propagation through a laser liquid.

It is important that the increase in the luminescence intensity observed in our experiments is directly proportional to the increase in the energy deposition by fission fragments. This circumstance indicates that the population of the lower level upon nuclear pumping is low and does not affect the amplification properties of the medium within experimental errors.

Thus, the analysis of experiments with liquids No. 1 and No. 2 showed that an increase in the energy deposition upon nuclear pumping leads to a significant increase in the inverse population. This is accompanied by widening of the time region in which this inverse population exists.

The results of our experiments allow us to obtain additional information on the laser properties of a medium. For this purpose, note that the experimentally measured power density of spontaneous emission of neodymium ions is described by the expression [7]

$$P_{\text{lum}}(t) = N_3(t)E_{\text{ph}}\eta/\tau_3. \tag{7}$$

Here, E_{ph} is the energy of recorded photons and η is the luminescence quantum yield.

Let us analyse the time distribution of the luminescence intensity at the instants when it does not change under the action of a probe laser pulse. This means that the population of the upper level at these time moments (t_e) is approximately equal to the known equilibrium population N_{20} of the lower level, i.e., $N_3(t_e) = N_{20} = 3.24 \times 10^{-4} N_t$. Then, substituting all the known values into expression (7), we can easily calculate the absolute luminescence power density at time moments t_e. Such calculations were fulfilled, and the obtained experimental curves were normalised to the calculated values of P_{lum} at points t_e. For example, the instants t_e in Figs 7b and 7e correspond to the instants at which the probe laser pulse propagates through the medium [curves (*2*)]. The error in the determination of t_e was ± 5 μs. When a probe pulse propagated through the medium five and more microseconds before or after t_e, we recorded either a drop or a spike in the luminescence intensity. The relative error in the calculation of $P_{\text{lum}}(t_e)$ due to this uncertainty in the measurement of t_e did not exceed ± 10 %. Figure 8 shows the experimental distributions of the luminescence power density of neodymium ions at different energy depositions.

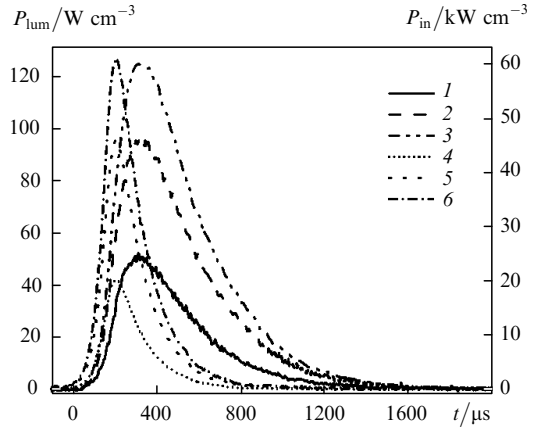

Figure 8. Time dependences of the luminescence power density P_{lum} (*1–3*) and pump power density P_{in} (*4–6*) at $N_t = 1.8 \times 10^{20}$ cm^{-3} for [U] $= 1.8 \times 10^{19}$ cm^{-3}, $\tau = 225$ μs, $\eta = 0.61$, $E_{\text{in}} = 5$ J cm^{-3} (*1, 4*); [U] $= 7.0 \times 10^{19}$ cm^{-3}, $\tau = 205$ μs, $\eta = 0.55$, $E_{\text{in}} = 11$ J cm^{-3} (*2, 5*); and [U] $= 7.0 \times 10^{19}$ cm^{-3}, $\tau = 200$ μs, $\eta = 0.54$, $E_{\text{in}} = 15$ J cm^{-3} (*3, 6*).

Knowing the luminescence and pump powers, we can determine the pump efficiency δ for the upper laser level by the formula [7]

$$\delta = \frac{\int P_{\text{lum}}(t)\mathrm{d}t}{\eta \int P_{\text{in}}(t)\mathrm{d}t}. \tag{8}$$

where P_{in} is the specific power density of pumping by fission fragments. Figure 9 presents the pump efficiencies obtained in experiments in which the propagation of a probe laser pulse did not change the luminescence intensity of the sample. The relative error in calculation of δ is associated mainly with the accuracy of determination of P_{lum} and with the accuracy of measurement of the energy deposition by fission fragments in the medium. According to our estimates, this error in each individual pulse did not exceed 20 %. As can be seen from Fig. 9, the pump efficiency, on average, remains constant and does not depend on the energy deposition. Thus, the average efficiency of pumping of the upper laser level by fission fragments in a laser liquid at $N_t = 1.8 \times 10^{20}$ cm^{-3} is 0.78 ± 0.05 %. Taking into account that the pump efficiency is proportional to the concentration of neodymium ions, the results of this study satisfactorily agree both with the data of subthreshold diagnostics [14, 15] and with the experimental data obtained in a reactor by another method using reference radiation [7].

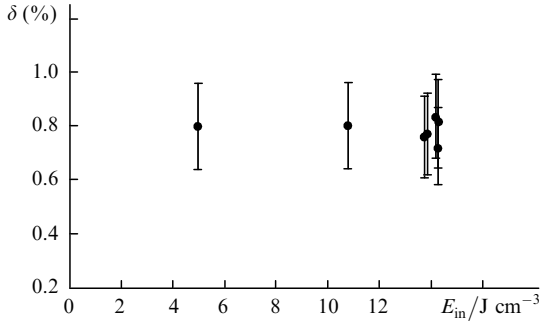

Figure 9. Efficiencies of pumping of the upper laser level of the neodymium ion at different specific energy depositions per reactor pulse for $N_t = 1.8 \times 10^{20}$ cm^{-3}.

From the experimentally found distributions $P_{\text{lum}}(t)$, we can find by formula (7) the distribution $N_3(t)$ of the population of the upper laser level of neodymium ions upon pumping by fission fragments. The results of this calculation are shown in Fig. 10. Using the calculated distributions $N_3(t)$ and taking into account that N_2 is almost unchanged and approximately equal to the equilibrium population N_{20}, we find the time dependence of the linear gain (absorption) coefficient α for a nuclear-pumped laser medium in the form

$$\alpha(t) = \Delta N(t)\sigma, \tag{9}$$

where $\Delta N(t) = N_3(t) - N_2(t)$ is the inverse population density. The cross section of the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ laser transition of the neodymium ion in $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ is $\sigma = 8 \times 10^{-20}$ cm^2. The coefficient α in the negative and positive regions corresponds to the linear absorption coefficient at the

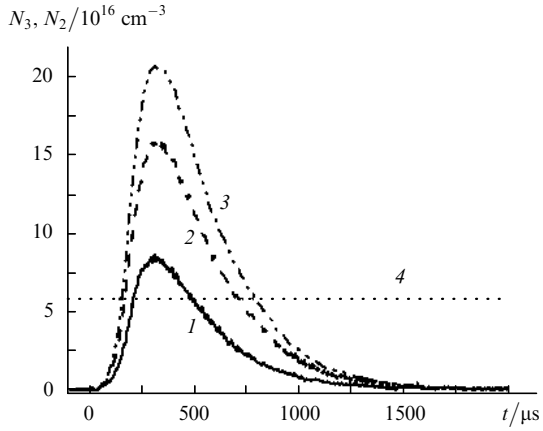

Figure 10. Time distributions of the populations of the upper laser level N_3 (*1–3*) and the lower laser level N_2 (*4*) at $N_t = 1.8 \times 10^{20}$ cm^{-3} for [U] $= 1.8 \times 10^{19}$ cm^{-3}, $\tau = 225$ μs, $\eta = 0.61$, $E_{in} = 5$ J cm^{-3} (*1*), [U] $= 7.0 \times 10^{19}$ cm^{-3}, $\tau = 205$ μs, $\eta = 0.55$, $E_{in} = 11$ J cm^{-3} (*2*) and [U] $= 7.0 \times 10^{19}$ cm^{-3}, $\tau = 200$ μs, $\eta = 0.54$, $E_{in} = 15$ J cm^{-3} (*3*).

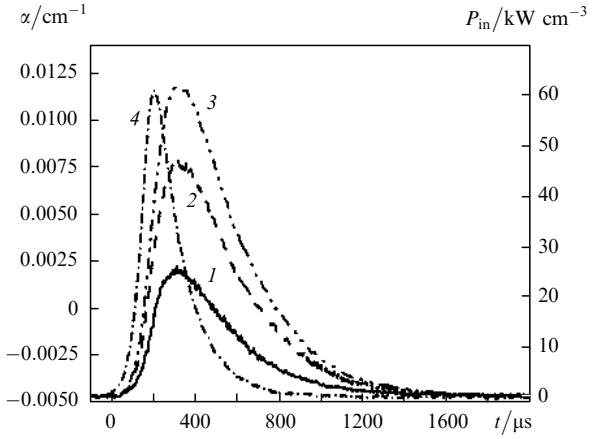

Figure 11. Time dependences of the α coefficient (*1–3*) for different energy contributions at $N_t = 1.8 \times 10^{20}$ cm^{-3} and time dependence of the pump power density P_{in} at $E_{in} = 15$ J cm^{-3} (*4*). [U] $= 1.8 \times 10^{19}$ cm^{-3}, $\tau = 225$ μs, $\eta = 0.61$, $E_{in} = 5$ J cm^{-3} (*1*), [U] $= 7.0 \times 10^{19}$ cm^{-3}, $\tau = 205$ μs, $\eta = 0.55$, $E_{in} = 11$ J cm^{-3} (*2*) and [U] $= 7.0 \times 10^{19}$ cm^{-3}, $\tau = 200$ μs, $\eta = 0.54$, $E_{in} = 15$ J cm^{-3} (*3*).

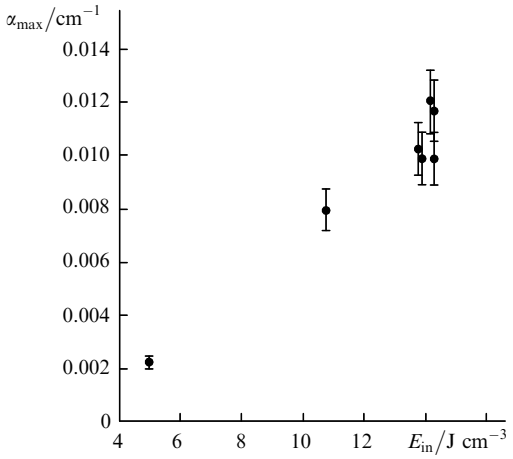

Figure 12. Dependence of the maximum linear gain on the specific energy deposition per reactor pulse at $N_t = 1.8 \times 10^{20}$ cm^{-3}.

wavelength of neodymium emission and to the linear gain coefficient, respectively. Figure 11 demonstrates the time dependences of the coefficient α for liquids with $N_t = 1.8 \times 10^{20}$ cm^{-3} at different energy depositions. In the regions where α is positive, the upper laser level is inversely populated and one observes a drop in the luminescence intensity. In the region of negative α, the probe laser radiation is absorbed by the medium and the luminescence intensity in the experiment abruptly increases. The dependence of the maximum linear gain on the specific energy deposition is shown in Fig. 12. One can see that this dependence is almost linear up to the specific energy deposition of 15 J cm^{-3} per pulse.

5. Conclusions

The process of population of the upper and lower laser levels is theoretically simulated. The minimum theoretical pump threshold that creates an inverse population in neodymium ions in the $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ laser liquid is found to be 1.6 J cm^{-3} at $N_t = 1.8 \times 10^{20}$ cm^{-3}.

The changes in the ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ luminescence intensity of the neodymium ion are measured at the instants when a probe laser beam with a wavelength resonant with the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ laser transition propagates through the medium excited by fission fragments. In the region of the luminescence maximum of the neodymium ion in the $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ laser liquid, we observed an inverse population of the upper laser level with respect to the lower level, which increased proportionally to the energy deposition by fission fragments.

The obtained experimental data were analysed in detail. The theoretical calculations well describe the observed experimental data. It is shown that the population of the lower laser level upon pumping by fission fragments insignificantly affects the inverse population and the threshold characteristics of nuclear-pumped liquid lasers.

Thus, the theoretical analysis and the analysis of the experimental data show that the additional population of the lower ${}^4I_{11/2}$ laser level of the neodymium ion in $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$ due to pumping by fission fragments does not exceed 1 % from the equilibrium population of this level, which is not an obstacle to achieving a lasing threshold in nuclear-pumped liquid lasers. It is important to note that the efficiency of pumping of Nd^{3+} by fission fragments depends, first of all, on the matrix composition, while the inverse population of laser levels and the linear gain increase proportionally to the concentration of neodymium ions and the pump power. In our case, at the concentration of neodymium ions of 1.8×10^{20} cm^{-3} in $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$, the maximum inverse population and the linear gain were, respectively, $(2.05 \pm 0.20) \times 10^{17}$ cm^{-3} and 0.012 ± 0.001 cm^{-1} at the specific energy deposition by fission fragments of 14.5 J cm^{-3}.

The obtained results allow us to suggest directions of development of a nuclear-pumped liquid laser. First, it is necessary to improve the technology of synthesis of uranium- and neodymium-containing laser liquids in order to decrease inactive losses at the wavelength of neodymium laser radiation to a level below 8×10^{-3} cm^{-1} at a neodymium concentration higher than 2.5×10^{20} cm^{-3}. Second, the energy deposition by fission fragments to the medium

must be increased due to both an increase in the concentration of uranium in the laser liquid and an increase of the neutron flux density in the region of the laser cell with respect to the flux density in the active zones of the reactor. However, simultaneously with a higher energy deposition, fission fragments induce higher losses at the lasing wavelength, and hence, the third direction is to study the nature of light scattering upon nuclear pumping and to search for methods of its suppression.

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