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Model of a nuclear-pumped liquid optical quantum amplifier

A A Seregin, E A Seregina

***Abstract*. A model of a nuclear-pumped liquid optical quantum amplifier is constructed, which allows the analysis of its operation for different parameters of the amplifier, different compositions of the active medium and pump pulses. The amplification characteristics of a liquid amplifier with a neodymium- and uranium-containing phosphorus oxychloride laser medium are calculated. It is shown that amplification in this medium can be obtained using the BARS-6 reactor neutron pumping.**

Keywords: *active media, liquid optical amplifier, nuclear pumping.*

1. Introduction

High-power yet inexpensive sources of laser radiation are required for inertial nuclear fusion, industrial isotope separation, and several other large-scale projects. These sources may well prove to be nuclear-pumped pulsed lasers and optical quantum amplifiers, in which the fission energy of atomic nuclei is directly converted to the energy of laser radiation [1, 2].

The quest for efficient laser media for nuclear pumping is the most important problem in the development of lasers and amplifiers of this type. At the Institute of Physics and Power Engineering, a technology has been elaborated for obtaining a neodymium- and uranium-containing liquid laser medium on the basis of phosphorus oxychloride ($POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$) with the parameters required for lasing upon optical or nuclear pumping [3]. The model of a nuclear-pumped liquid laser was developed for this medium in Ref. [4], which was used to calculate the dependences of the basic laser characteristics on the setup parameters, the medium composition, and the pump pulse.

In this paper, we propose the model of a nuclear-pumped liquid quantum amplifier.

2. Nuclear-pumped liquid optical quantum amplifier

The simplest scheme of a nuclear-pumped liquid optical quantum amplifier is shown in Fig. 1. The main element in this scheme is a cell filled with a liquid working medium, which consists of a matrix and of active and fission elements. The active elements may be ions of rare-earth elements (neodymium, europium, etc.) and the fission elements may be ^{233}U, ^{235}U, or ^{239}Pu, i.e., the elements which are easily fissionable by slow neutrons, do not absorb at the wavelength of amplified radiation, and do not quench the upper working level of the active element. The matrix may be a material that contains the active and fission elements at required concentrations, does not absorb at the amplification wavelength, does not quench the upper working level of the active element, and weakly absorbs slow neutrons.

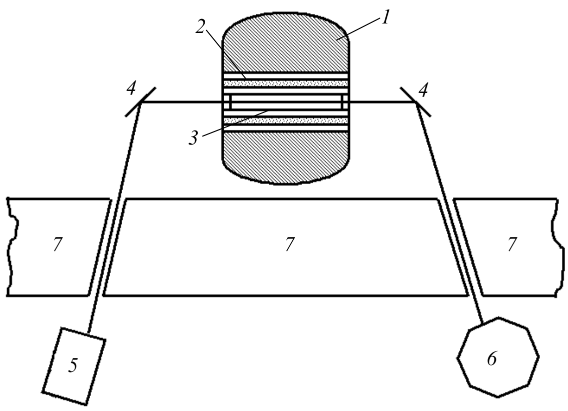

Figure 1. Principal scheme of a nuclear-pumped liquid optical quantum amplifier: (*1*) reactor core; (*2*) polyethylene moderator of neutrons; (*3*) cell with a liquid laser uranium-containing medium; (*4*) mirror; (*5*) generator of amplified radiation; (*6*) detector of the amplified radiation; (*7*) concrete neutron shield.

The liquid optical quantum amplifier operates in the following way. During the burst from a pulsed nuclear reactor, the flux of fast neutrons is incident on the cell with a uranium-containing working medium surrounded by a layer of polyethylene, which moderates the neutrons to thermal energies. The flux of thermal neutrons induces the fission of ^{235}U nuclei in the cell. The ^{235}U nuclear fission fragments propagating through the liquid working medium ionise and excite its molecules, atoms, and ions, including the active ions. Because the excited-state lifetime of the latter is 200 – 300 μs, a large number of excited ions of the active elements can be accumulated during the neutron burst, which lasts for 100 – 200 μs. If an electromagnetic radiation pulse with a photon energy equal to the energy of transition from the long-lived upper level to the lower level is passed through the excited laser medium within 300 – 500 μs of the onset of

A A Seregin, E A Seregina A I Leipunski Physics and Power Engineering Institute, State Scientific Centre of the Russian Federation, ul. Bondarenko 1, 249020 Obninsk, Kaluzhskaya oblast, Russia;
e-mail: sereg@obninsk.ru

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the reactor burst, it is possible to obtain the amplification of radiation.

The laser amplification can be simply described using the rate equations of a two-level model. We restrict ourselves to the case when the incident radiation is a short pulse of duration τ, with $\tau_1 \ll \tau \ll \tau_2$, where τ_1 and τ_2 are the lifetimes of the lower and upper levels. The differential equation for the energy flux of the output pulse $\Gamma(L)$ (L is the amplifier length) has the form [5]

$$\frac{\mathrm{d}\Gamma}{\mathrm{d}z} = g_0\Gamma_\mathrm{s}\left[1-\exp\left(-\frac{\Gamma}{\Gamma_\mathrm{s}}\right)\right] - \mu\Gamma, \qquad (1)$$

where $g_0 = \sigma N_2$ is the unsaturated gain; σ is the cross section for the stimulated transition; N_2 is the population of the upper working level; $\Gamma_\mathrm{s} = hv/\sigma$ is the saturation energy flux; μ is the loss coefficient for the medium; and hv is the transition energy. Eqn (1) should be solved in the $0 - L$ interval with the boundary condition $\Gamma(0) = \Gamma_\mathrm{in}$, where Γ_in is the energy flux of the input pulse. To characterise the amplification, it is convenient to introduce the gain $\kappa = \Gamma(L)/\Gamma_\mathrm{in}$, which is a function of N_2, Γ_in, and μ. Therefore, the population N_2 and the coefficient μ should be known to calculate the gain for a given Γ_in. Note that the dependence of the gain on the losses in the medium was first investigated in Ref. [6].

3. Liquid optical quantum amplifier pumped by neutrons from the pulsed BARS-6 reactor

As mentioned above, a liquid neodymium- and uranium-containing laser medium on the basis of phosphorus oxychloride ($POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Nd^{3+}$) was synthesised in the Institute of Physics and Power Engineering. Experiments to detect gain were performed with this medium. The experimental setup involving neutron pumping from the pulsed BARS-6 reactor is shown in Fig. 2. The active elements in this working medium are neodymium ions, which accomplish amplification. The energy level diagram for neodymium ions is well known [5]. The long-lived excited state in a neodymium ion is the ${}^4F_{3/2}$ state, whose lifetime is equal to 150 – 200 μs, depending on the preparation technology of the working medium. Since the lifetimes of levels 1 and 3 are short compared to the lifetime of level 2 ($\tau_1 \approx \tau_3 \approx 10^{-9}$ s, $\tau_2 \approx 10^{-4}$ s) and the input pulse has a duration of the order of 10^{-6} s, the time dependence of the populations densities can be calculated from simplified rate equations for a four-level laser [5]:

$$\begin{aligned} &\frac{\mathrm{d}N_2}{\mathrm{d}t} = W(t)N_\mathrm{g} - BqN_2 - \frac{N_2}{\tau_2}, \\ &\frac{\mathrm{d}q}{\mathrm{d}t} = \left(\upsilon BN_2 - \frac{1}{\tau_\mathrm{c}}\right)q, \\ &N_\mathrm{Nd} = N_\mathrm{g} + N_2, \\ &N_1 = N_3 = 0 \end{aligned} \qquad (2)$$

with the initial conditions

$$N_2(0) = 0 \text{ и } q(0) = q_0,$$

where q_0 is the minimal number of photons in the resonator required for the onset of lasing; N_1, N_2, and N_3 are the neodymium ion populations for the corresponding levels;

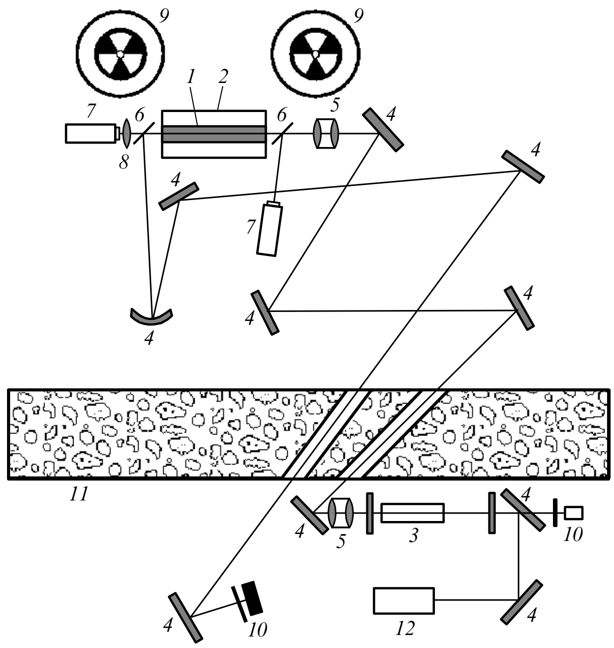

Figure 2. Optical scheme of the experiment: (*1*) cell with a liquid; (*2*) polyethylene moderator of neutrons; (*3*) generator of amplified radiation; (*4*) deflecting aluminium mirrors; (*5*) telescopes; (*6*) beamsplitters; (*7*) laser power meters; (*8*) quartz lens ($f = 48$ mm); (*9*) BARS-6 reactor cores; (*10*) photodiodes; (*11*) concrete neutron shield; (*12*) adjustment laser.

N_g is the neodymium ion population in the ground state; N_Nd is the neodymium ion concentration in the laser liquid; $W(t)$ is the pumping rate; B is the Einstein coefficient for stimulated emission; q is the total number of photons in the resonator; υ is the volume occupied by the mode in the active medium; and τ_c is the average photon lifetime in the resonator.

4. Nuclear pumping by neutrons from the pulsed BARS-6 reactor

In experiments to detect gain with neutron pumping, the cell with a liquid laser medium is surrounded by polyethylene, in which fast neutrons from the BARS-6 reactor are moderated to nearly thermal velocities. These neutrons cause ${}^{235}U$ to fission. As a result, in the cell with the laser medium a specific power is released, which can be approximated by the function [7]

$$N_\mathrm{f}(t) = \frac{\sigma_\mathrm{f} C_\mathrm{U} E_\mathrm{n} k}{2\theta_1}\exp\left(\frac{\theta_\mathrm{r}^2}{4\pi\theta_1^2} - \frac{t}{\theta_1}\right) \times\left[1 + \mathrm{erf}\left(\frac{\sqrt{\pi}t}{\theta_1} - \frac{\theta_\mathrm{r}}{2\theta_1\sqrt{\pi}}\right)\right], \qquad (3)$$

where $\sigma_\mathrm{f} = 583$ barn is the cross section for the ${}^{235}U$ nuclear fission induced by thermal neutrons; $C_\mathrm{U} = 5\times10^{19}$ cm^{-3} is the concentration of ${}^{235}U$ nuclei in the laser medium; $E_\mathrm{n} = 2\times10^{17}$ is the number of fissions in reactor cores; $k \approx 10^{-4}$ cm^{-2} is the geometrical factor; $\theta_\mathrm{r} \approx 50$– 70 μs is the neutron pulse duration in the reactor core; $\theta_1 \approx 100$– 200 μs is the neutron pulse duration in the cell after moderation; and erf$\,x$ is the error function. If it is assumed that $k = 1.1915 \times 10^{-4}$ cm^{-2} and $\theta_1 = 100$ μs, it is possible to fit the calculated values of N_f to the data obtained in experiments on the BARS-6 [8]. Good agreement between the calculated and experimental data permits the use of the function (3) in subsequent calculations of different quantities and their time dependences.

Another parameter required for subsequent calculations is the factor μ_a of induced additional losses, which arise primarily upon scattering of an electromagnetic wave from the tracks of uranium fission fragments produced in the laser medium [9, 10]:

$$\mu_a = \sigma_t \int_{t-\tau_0}^{t} N_f(t')\mathrm{d}t', \tag{4}$$

where σ_t is the cross section for the electromagnetic wave scattering by a single track and τ_0 is the average lifetime of a track. Unfortunately, σ_t and τ_0 are hard to calculate, and only μ_a can be determined from experimental data [9]. By assuming that $\tau_0 \approx 10^{-8}$ s (this follows from our estimate and the estimate of Ref. [10]) and using the experimental data of Ref. [9], we obtained $\sigma_t \approx 10^{-9}$ cm^2.

Finally, we give the explicit expression for the rate $W(t)$ of upper laser-level pumping by fission fragments:

$$W(t) = \frac{\delta q_f N_f(t)}{N_{Nd} h\nu}. \tag{5}$$

Here, q_f is the energy released in a single event of ^{235}U nuclear fission; $h\nu = 1.17$ eV is the laser transition energy;

$$\delta = \eta \frac{\tau_s}{\tau_2} \tag{6}$$

is the efficiency of upper laser-level pumping by the fragments [11]; η is the efficiency of conversion of the energy of a fragment to electromagnetic radiation with a photon energy $h\nu$; τ_s is the radiative lifetime; and τ_2 is the lifetime of the working level. We are presently unable to calculate the pumping efficiency δ, but it can be determined experimentally. Experimental data suggest that δ is highest in crystals, is somewhat lower in liquid media, and is still lower in glasses, the concentration of active ions being the same [11].

5. Results of calculations

The solution of the system of equations (2) was obtained for neodymium- and uranium-containing liquid laser medium on the basis of phosphorus oxychloride for the following typical values of the main parameters of the medium: the concentration of uranium nuclei of 5×10^{19} cm^{-3}, the concentration of neodymium atoms of 2×10^{20} cm^{-3}, the stimulated-transition cross section of $\sigma = 8 \times 10^{-20}$ cm^2, the excited-state lifetime of $\tau_2 = 200$ μs. According to experimental data, the efficiency of pumping by fission fragments increases proportionally to the neodymium ion concentration and is equal to 0.005 at the concentration of 10^{20} cm^{-3} [11]. The length of the cell with the laser liquid was taken to be 30 cm, the laser-beam radius was 0.4 cm, the transmittance of the end cell windows was 0.92, and the pump pulse duration was 100 μs.

The system of equations (2) with the above parameter values was solved numerically by the Gear method. The time dependences of the population N_2 of the upper working level and the total loss factor μ for different values of the energy deposition hold the greatest interest. Fig. 3 shows the time dependences of the population for energy depositions of 5, 7.5, and 31 J cm^{-3}. One can see that the population of the upper working level increases in proportion with the energy deposition.

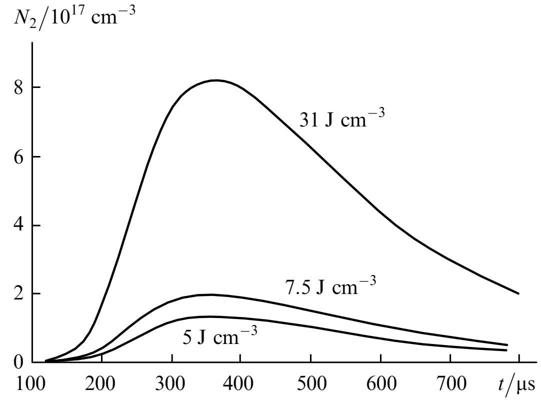

Figure 3. Time dependences of the population N_2 of the upper laser level for different values of the energy deposition in the active medium pumped by neutrons from the BARS-6 reactor.

For a nuclear-pumped liquid optical quantum amplifier, it is vitally important to consider the appearance of the gain under the condition when the total loss coefficient μ increases during pumping. For a nuclear pumping,

$$\mu = \mu_0 + \mu_a, \tag{7}$$

where $\mu_0 = 0.007$ cm^{-1} is the loss coefficient for the medium unperturbed by fission fragments.

Fig. 4 shows the time dependences of the loss coefficients upon nuclear pumping by neutrons for energy depositions of 5, 7.5, and 31 J cm^{-3}. One can see that the transparency of the cell with the laser liquid decreases significantly. In this case, as the energy deposition increases by factors of 1.5 and 6, the loss coefficient increases by factors of 1.3 and 4. Therefore, the increase in the energy deposition results not only in the increase in the population of the upper working level, but in the losses as well. However, the population of the upper working level rises somewhat faster.

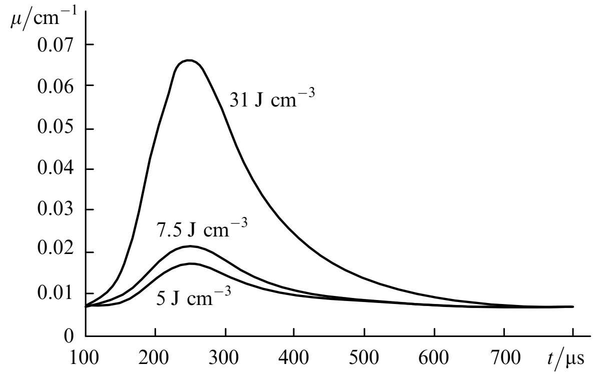

Figure 4. Time dependences of the total loss coefficient in the laser medium for different energy depositions upon pumping by neutrons from the BARS-6 reactor.

Taking into account the calculated populations and the loss coefficients, we found from Eqn (1) the time dependences of the gains for different energy depositions upon nuclear pumping. The calculations showed that the gain was absent for an energy deposition of 5 J cm^{-3} and below.

Fig. 5 shows the time dependence of the gain for an energy deposition of 7.5 J cm^{-3}. One can see that the gain can be already detected in this case and the maximum gain equal to 10 % is reached within 420 μs after the onset of the neutron pulse.

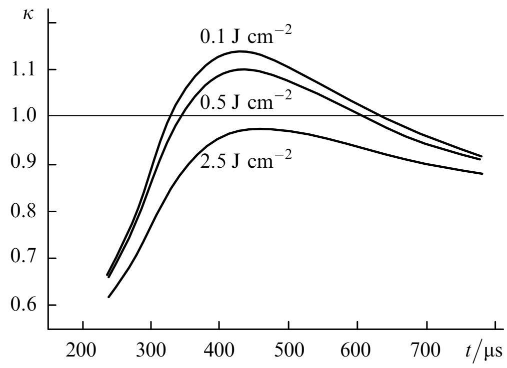

Figure 5. Time dependences of the gain for an energy deposition of 7.5 J cm^{-3} for different input energy fluxes.

Finally, Fig. 6 shows the time dependences of the gain for an energy deposition of 31 J cm^{-3}. One can see that this energy deposition should produce the highest gain.

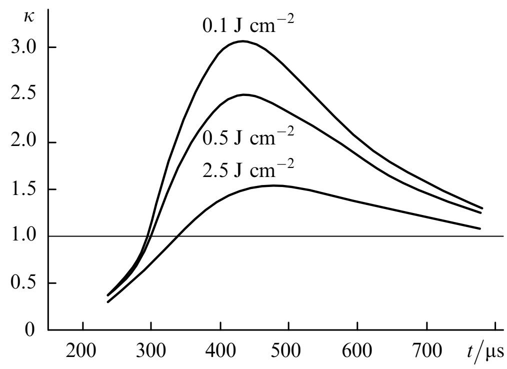

Figure 6. Time dependences of the gain for an energy deposition of 31 J cm^{-3} for different input energy fluxes.

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