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Laser study of the time dependence of induced absorption in a liquid excited by uranium fission fragments

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***Abstract*. The results of 1.05-μm laser measurements of optical properties of the $POCl_3-SnCl_4-UO_2^{2+}$ liquid containing vapour bubbles generated by the fission fragments of ^{235}U nuclei formed upon irradiation of this liquid by neutrons in a BARS-6 pulsed reactor are presented. The average size of a bubble and its mean lifetime are obtained from experiments. The effect of pressure on the bubble size is studied. It is found that the lifetime of a bubble increases with increasing the concentration of molecular chlorine in the liquid due to its radiolysis induced by fission fragments.**

Keywords: *liquid Nd laser, phosphorus oxychloride, nuclear pumping, fission fragments, ^{235}U, fission fragment tracks, vapour bubble characteristics.*

1. Introduction

The interest towards research in the field of direct transformation of nuclear fission energy into laser radiation energy is explained by the fact that it may eventually be used for developing quite powerful and economical sources of laser radiation [1]. Over the last few years, investigations have been carried out at the Institute of Physics and Power Engineering with a view to develop a nuclear pumped liquid laser. A liquid medium containing the fissionable element uranium enriched with the isotope ^{235}U and laser-active centres, namely, Nd^{3+} ions, was developed for such a laser based on phosphorus oxychloride ($POCl_3-SnCl_4$) [1]. Lasing upon optical pumping was attained in this medium, but it has not been possible so far to attain lasing upon pumping by ^{235}U fission fragments. One of the reasons behind this may be a sharp increase in the losses upon excitation of the laser medium by fission fragments because these fragments form vapour bubbles as they slow down in the liquid [2, 3]. In order to verify this fact and to measure these induced additional losses, experiments were carried out for measuring the pulse energy of a probe laser radiation passing through a 'cold' liquid containing uranium, and through the same liquid pumped by fission fragments.

In the first experiments [2], it was shown that additional losses do occur in a liquid pumped by fission fragments, but their magnitude (6×10^{-3} cm^{-1}) does not exceed typical active losses in optically pumped inorganic liquid lasers. A free-running inorganic liquid Nd laser was used as a probe laser. The pulse duration (120 μs) of the probe laser was nearly equal to the duration (140 μs) of the pump pulse. Therefore, the induced additional loss of light in a medium obtained experimentally should be treated as the loss averaged over a pump pulse. These results were used to formulate a model of light scattering by short-lived vapour bubbles produced on the tracks of the fission fragments in the liquid. It follows from the model that additional loss must be directly proportional to the specific rate of energy release by ^{235}U fission fragments in the liquid during pulsed pumping.

In this work, we shall study the dynamics of losses induced by the fission fragments in a liquid irradiated in a pulsed reactor, and verify and develop our previous models of light scattering at vapour bubbles formed on the tracks of fission fragments in a liquid. Experiments were made using an inorganic liquid single-pulse probe laser with a pulse duration of 40 ns.

2. Experimental

Measurements were made in a BARS-6 fast-pulsed self-quenching reactor. The experimental setup is shown in Fig. 1. A cell (*1*) with the liquid under study and a polyethylene neutron moderator (*2*) were placed in close proximity to the active reactor zones in the bench hall. The probe liquid Nd laser (*3*) was installed behind a biological shield (*14*) in the experimental hall at a distance of about 18 m from the experimental setup. The Nd-laser radiation (*3*) was directed to the bench hall with the help of deflecting mirrors (*4*) through telescopic systems (*5*) and (*6*) along the optical path coinciding with the beam from the alignment He–Ne laser (*13*). The telescopic systems considerably reduced the divergence of the alignment He–Ne laser as well as the probe liquid Nd laser. A photodiode (*17*) was used for measuring the amplitude and shape of the probe liquid laser radiation pulse, and also for controlling the synchronisation between the probe pulse and the neutron pulse from the reactor. The duration of the neutron pulse from the reactor was 80 μs, while that of the slowed neutron pulse was 190 μs. The energy release in the active zone of the reactor was 6 MJ per pulse, the pulse

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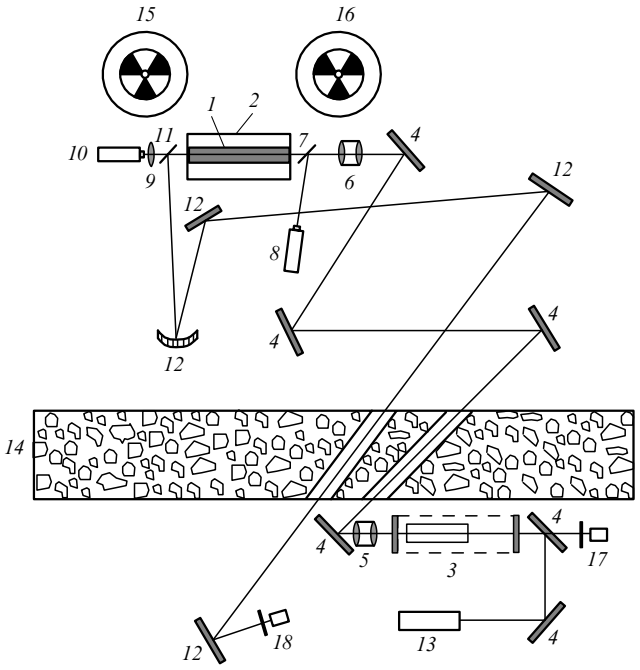

Figure 1. Optical scheme of the experiment: (*1*) cell with liquid; (*2*) polyethylene neutron moderator; (*3*) master oscillator; (*4*, *12*) aluminium mirrors; (*5*, *6*) telescopic lens system; (*7*, *11*) beamsplitters; (*8*, *10*) power meters; (*9*) quartz lens ($f = 48$ mm); (*13*) alignment He–Ne laser; (*14*) biological shield; (*15*, *16*) active zones of the BARS-6 reactor; (*17*, *18*) photodiodes.

repetition rate was one pulse per day, while the energy release in the cell was 5 J cm^{-3} per pulse. The energy meter (*8*) detected the probe laser radiation at the input to the cell (*1*), while the meter (*10*) measured the energy of the radiation passing through cell (*1*) containing the liquid under study. The uranium-containing sample of the $POCl_3 - SnCl_4 - {}^{235}UO_2^{2+} - Ln^{3+}$ liquid was prepared by using uranium enriched with 95 % ^{235}U isotope. This liquid was an imitation of the laser liquid in which Nd^{3+} is replaced by Ln^{3+} to eliminate the influence of amplification light at 1.05 μm on the results of the experiment. The liquid was poured into a hermetically sealed optically transparent quartz cell of length 30 cm and diameter 1.2 cm with plane–parallel end windows. The concentration of ^{235}U nuclei in the sample was equal to 6×10^{19} cm^{-3}. The linear attenuation coefficient of intensity of light at 1.05 μm was measured during passage of light through the 'cold' liquid and was found to be 0.01 cm^{-1}.

The probe liquid ($POCl_3 - SnCl_4 - Nd^{3+}$ mixture) laser operated in a single-pulse regime. Its FWHM pulse duration was 40 ns, and the lasing wavelength was 1.052 μm.

Measurements were synchronised with a neutron pulse reactor and were carried out in several stages. In the first stage, the radiation-induced background energy E_b on the energy meters (*8*) and (*10*) was measured. The probe laser pulse energy E_2 passing through the cell containing the liquid pumped by fission fragments was measured in the subsequent stages. Measurements were made at different instants of time in the reactor pulse with an interval of about 50 μs in one pulse-one measurement mode. Six measurements made it possible to cover a time interval of 300 μs and to obtain data about the behaviour of additional losses of laser radiation during pulsed pumping of the liquid by fission fragments. Measurements of energy E_2 and energy E_1 of the pulse passing through the cell containing the 'cold' liquid led to an additional loss factor μ_a:

$$\mu_a(t) = L^{-1} \ln\left(\frac{E_1}{E_2 - E_b}\right), \quad (1)$$

where L is the length of the cell. Figure 2 shows the results of the experiments. One can see that the loss factor really increases in an excited liquid and its increase correlates with the increase in energy contribution. This can be seen especially clearly at the initial stage of pumping of the liquid by fission fragments.

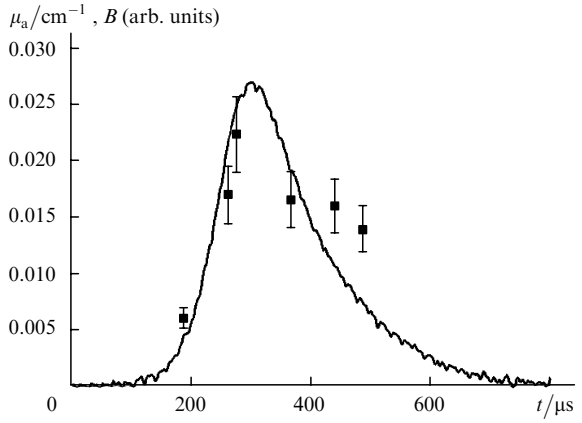

Figure 2. Time dependences of the additional loss factor μ_a (points) and the energy contribution rate (solid curve) for an energy release of 5 J cm^{-3}.

3. Experimental results and analysis

It was mentioned above that the most probable reason for the attenuation of electromagnetic radiation passing through an excited uranium-containing liquid is its scattering by the vapour bubbles formed in the liquid. According to the thermal model of formation and evolution of vapour bubbles [2, 3], the kinetic energy of fission fragments (172 MeV) is transformed into thermal energy when they are stopped in a liquid medium in 10^{-12} s. This energy is distributed over a localised cylindrical region of the medium with a radius 2.9×10^{-7} cm, volume 1.2×10^{-15} cm^{-3} and length 5.2×10^{-3} cm, equal to the mean free path of a fragment in phosphorus oxychloride. The released energy is quite sufficient to convert the liquid into vapour, heat it to a temperature of more than 5000 K and create a pressure of more than 25000 atm inside the bubble. Heating causes an explosion of the bubble and its radius and volume increase to 3.7×10^{-6} cm and 2.2×10^{-13} cm^3 respectively over a period of 10^{-10} s. After the explosion, the bubble begins to cool down and disappears in a period of the order of 10^{-8} s. The process of cooling is the slowest stage in the evolution of a bubble and therefore determines its lifetime.

The formation of vapour bubbles in the active medium results in its additional scattering and absorption of the electromagnetic radiation and consequently leads to an additional time-dependent loss factor μ_a which can be written explicitly in the form

$$\mu_a(t) = \frac{\sigma}{\Delta}\int_t^{t+\Delta} dt_1 \int_{t_1-\tau}^{t_1} N_f(t_2)dt_2, \quad (2)$$

where σ is the sum of cross sections for scattering and absorption of an electromagnetic wave at a bubble; $\Delta = 4\times10^{-8}$ s is the probing laser pulse duration; $\tau \approx 10^{-8}$ s is the lifetime of the bubble; and N_f is the rate of fission of ^{235}U nuclei in the cell. Our estimates show that the absorption cross section is several orders of magnitude smaller than the scattering cross section, and hence we shall disregard this cross section in the subsequent analysis.

Because the duration of a fission pulse is 190 μs, i.e., much larger than the laser pulse duration or the bubble lifetime, expression (2) can be simplified and written in the form

$$\mu_a(t) = \sigma\tau N_f(t - \tau/2). \quad (3)$$

It follows from this relation that, first, the additional loss associated with scattering at the bubbles will correlate with the rate of change of fissions and, second, will be determined by the product of cross section of scattering at the bubble and its lifetime.

Table 1 contains the values of the product $\sigma\tau$ calculated from the experimental data by using formula (3). One can see that the values of $\sigma\tau$ are quite close for the first four experimental points, but differ sharply for the last two points. This may serve as an indication that certain processes that did not exist at the initial stages occur at the final stage of pumping. For a given fission rate, we equate the experimental and theoretical values of μ_a at the first point and determine the value of $\sigma\tau$ using formula (3). Knowing $\sigma\tau$, we determine the value of μ_a from formula (3). The obtained dependence $\mu_a(t)$ is shown by the dotted curve in Fig. 3. One can see that the theoretical curve matches with the experimental data on the whole, but there are some noticeable disparities. This is due to the fact that certain factors were disregarded while calculating μ_a. First, fission of uranium nuclei causes a heating (and, consequently, expansion) of the liquid as a result of which its pressure in the cell increases. Second, heating of the liquid changes its surface tension. Finally, radiolysis (rupture of molecules of the medium) of phosphorus oxychloride due to the action of fission fragments results in the formation of chlorine molecules whose emergence inside a bubble increases its lifetime. The inclusion of these factors requires a detailed analysis of scattering of light at a bubble and dependence of the bubble size on pressure.

The highest uncertainty in formula (3) is associated with the scattering cross section σ which depends on the shape of a bubble as well as the ratio of its diameter to the wavelength λ of the electromagnetic radiation propagating in an excited liquid. In order to determine the cross section of scattering at a cylindrical bubble, we follow the results of Ref. [4] according to which the cross section of scattering of an electromagnetic wave at a cylindrical bubble of volume V, averaged over various orientations of the cylinder axis coincides with the cross section of scattering at a spherical bubble of volume

$$V^* = 0.45V.$$

Knowing the volume of the cylindrical bubble, we can determine the volume, and hence the radius R, of the corresponding spherical bubble. Thus the problem of scattering at a cylinder has been reduced to the familiar problem of scattering at a transparent sphere [5]. The analytic expressions for the cross section for scattering of radiation of wavelength λ at a transparent dielectric sphere of radius R have been obtained in two limiting cases, for $R < \lambda/2\pi$ and for $R > \lambda/2\pi$, respectively:

$$\sigma = \frac{2}{3}\pi R^2\left[\frac{2(1-n)}{1+2n^2}\right]^2\left(\frac{R}{\lambda/2\pi}\right)^4, \quad (4)$$

and

$$\sigma = \pi R^2\left\{2 - \frac{8n\lambda}{2\pi R(1+n)^2(1-n)}\sin\left[\frac{2\pi R(1-n)}{\lambda n}\right]\right\}. \quad (5)$$

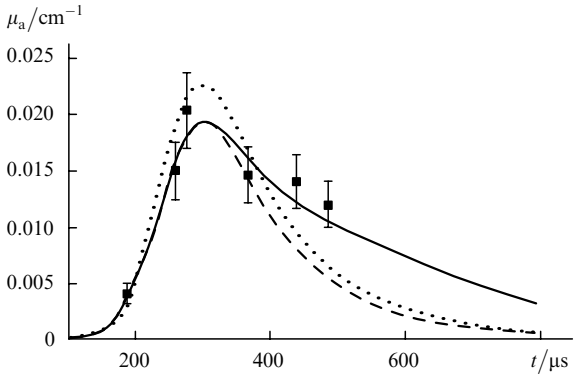

Figure 3. Time dependences of the additional loss factor for an energy release of 5 J cm^{-3}. The points correspond to the experimental values. The dashed curve shows the theoretical values of μ_a calculated by using formula (3) neglecting the pressure increase in the cell, while the dashed curve shows the same quantity taking into account the pressure increase in the cell. The solid curve was obtained by taking into account the pressure increase in the cell and the increase in the lifetime of the bubble.

Table 1. Experimental values of induced additional losses.

Number of an experimental point (see Figs 2, 3)	$t/\mu s$	$N_f/10^{13}$ neutron cm^{-3} s^{-1}	$\mu_a/10^{-2}$ cm^{-1}	$\sigma\tau/10^{-17}$ cm^2 s	$\sigma/10^{-9}$ cm^{2*}
1	187	8.42	0.4	4.87	1.62
2	261	39.5	1.5	3.80	1.27
3	275	43.8	2.0	4.66	1.55
4	366	35.2	1.5	4.15	1.83
5	438	19.9	1.4	7.03	2.34
6	486	13.4	1.2	8.96	2.99

*Values of σ are obtained for $\tau \approx 3\times10^{-8}$ s.

In these relations, $n = 1.46$ is the refractive index of the medium. Note that formulas (4) and (5) do not coincide for $R = \lambda/2\pi$, and the scattering cross section must be evaluated numerically in this case. Thus, if we know the dependence of the bubble radius on the pressure P_a in the cell, we can also determine the scattering cross section. In order to obtain this dependence, we use a formula describing the increase in pressure in the cell as a result of heating:

$$P_a = \frac{\varkappa \Delta T}{\beta}, \tag{6}$$

where $\varkappa = 1.16 \times 10^{-3}$ K^{-1} is the coefficient of thermal expansion of phosphorus oxychloride, $\beta = 3 \times 10^{-9}$ Pa^{-1} is the compressibility of phosphorus oxychloride, and ΔT is the increase in temperature due to the deceleration of uranium fission fragments in the liquid. This increase in temperature is proportional to the energy Q_f released by the fission fragments and has the form

$$\Delta T = \frac{Q_f}{c\rho}, \tag{7}$$

where $c = 1.34$ J K^{-1} g^{-1} is the specific heat and $\rho = 1.88$ g cm^{-3} is the density of phosphorus oxychloride. The energy release is described by the expression

$$Q_f = \varepsilon \int_0^t N_f(t')\mathrm{d}t', \tag{8}$$

where $\varepsilon = 172$ MeV is the energy released in a single act of fission, and $N_f(t)$ is the rate of fission in 1 cm^3 which can be expressed in terms of the rate of change in the number $N_n(t)$ of neutrons in 1 cm^3 of the cell:

$$N_f(t) = \sigma_f c_U N_n(t), \tag{9}$$

where $\sigma_f = 583$ b is the cross section of fission of ^{235}U nuclei by thermal neutrons, $c_U = 5 \times 10^{-19}$ cm^{-3} is the concentration of ^{235}U nuclei in the laser medium, and

$$N_n(t) = \frac{E_r k}{2\theta_1} \exp\left(\frac{\theta_r^2}{4\pi\theta_1^2} - \frac{t}{\theta_1}\right)\left[1 + \mathrm{erf}\left(\frac{\sqrt{\pi}t}{\theta_1} - \frac{\theta_r}{2\theta_1\sqrt{\pi}}\right)\right]. \tag{10}$$

In this expression, $E_r = 2 \times 10^{17}$ is the number of fissions in two reactor zones, $k \approx 10^{-4}$ cm^{-2} is a geometrical factor, $\theta_r \approx 80$ μs is the duration of a neutron pulse in the active zone of the reactor, $\theta_1 \approx 100 - 200$ μs is the duration of a neutron pulse in the cell after slowing-down, and erf x is the error function.

By substituting $k = 7.78 \times 10^{-5}$ cm^{-2} and $\theta_1 = 190$ μs in Eqn (10), we can match the theoretical values of N_f with the data obtained in the BARS-6 reactor experiment. A good agreement between the theoretical and experimental values makes it possible to use the functional dependence (9) for subsequent evaluation of other quantities as well as their time dependence.

The formulas obtained here would be final but for the pressure compensator in the cell, whose role is played by the branch pipe used for filling the cell with the liquid. As a result of expansion of the liquid volume in the cell, a part of the liquid will flow into the branch pipe. According to Ref. [6], the rate of flow v of a liquid through a hole in a vessel with excess pressure P_a is defined by the semiempirical formula

$$v = \phi\left(\frac{2P_a}{\rho}\right)^{1/2}, \tag{11}$$

where ρ is the density of the liquid, $\phi \approx 0.97$ is an experimentally determined factor for water which depends on several factors and, in particular, on viscosity. The volume of liquid flowing through a hole of area S in time dt is defined as

$$\Delta V' = Sv\mathrm{d}t. \tag{12}$$

The volume of the liquid flowing in time t is given by the relation

$$V' = S\phi \int_0^t \left(\frac{2P_a}{\rho}\right)^{1/2} \mathrm{d}t'. \tag{13}$$

Knowing the volume of the liquid flowing out and using formulas (6)–(13), we can write down an equation for calculating the pressure in the cell:

$$P_a = \frac{1}{\beta V_c}\left[\frac{\varkappa V \varepsilon}{c\rho}\int_0^t N_f(t')\mathrm{d}t' - S\phi\int_0^t \left(\frac{2P_a}{\rho}\right)^{1/2}\mathrm{d}t'\right], \tag{14}$$

where V_c is the cell volume. In order to find the solution of this equation, we differentiate it with respect to t and arrive at the differential equation

$$\frac{\mathrm{d}P_a}{\mathrm{d}t} = \frac{1}{\beta V}\left[\frac{\varkappa\varepsilon V}{c\rho}N_f(t) - S\phi\left(\frac{2P_a}{\rho}\right)^{1/2}\right] \tag{15}$$

with the initial condition $P_a = 0$ at $t = 0$. Eqn (15) was solved numerically by the Runge–Kutta method realised using the INTSTP program from the CERN library. The results of solution of Eqn (15) for a cell with a laser liquid are presented in Fig. 4 for various inner radii r of the filling branch pipe. It can be seen that the filling branch pipe really plays the role of compensator of pressure in the cell and considerably lowers it. In the cell used by us, $r = 0.25$ cm and hence, according to the calculations, the pressure in the cell rose by just 1 atm. An increase in pressure in the cell would lead to a decrease in the maximum size of the bubble [3]. Our calculations (Fig. 5) show that the maximum radius of a cylindrical bubble decreases by 3×10^{-7} cm. A decrease in the maximum bubble size lowers its average

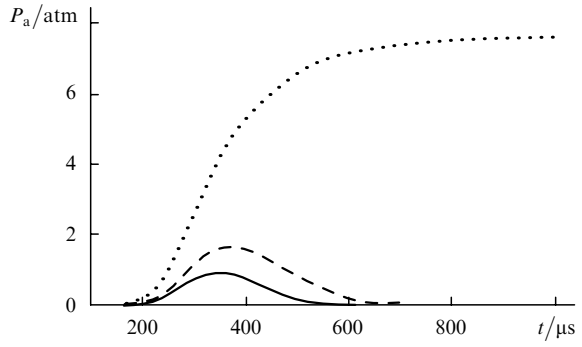

Figure 4. Time dependence of the pressure in the cell for an energy release of 5 J cm^{-3} for a branch pipe radius $r = 0$ (dotted curve), 0.2 cm (dashed curve) and 0.25 cm (solid curve).

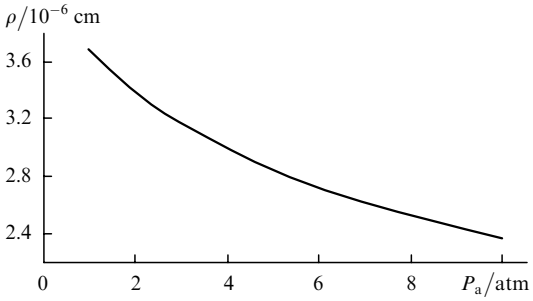

Figure 5. Dependence of the cylindrical radius ρ of a bubble on the pressure P_a in a cell containing phosphorus oxychloride.

size and hence the average cross section for scattering of electromagnetic radiation of wavelength $\lambda = 1.052$ μm as well. Our estimates show that the cross section decreases by 0.35×10^{-9} cm^{-2} under maximum pressure. When this fact is taken into consideration, the matching of the experimental and theoretical results improves (see the dotted curve and points in Fig. 3) with the exception of two points on the extreme right.

These extreme points cannot be explained theoretically even under the assumption that surface tension decreases as a result of heating. In our experiments, the temperature of phosphorus oxychloride rose by 1 K, and hence the surface tension may drop by 1 %–2 %. However, a decrease in surface tension by such an amount increases the scattering cross section also by 1 %–2 %, which is obviously inadequate for explaining the experimental data.

Moreover, the improvement in the agreement between the theoretical and experimental data can be explained by taking into consideration the radiolysis of phosphorus oxychloride under the action of fission fragments. Indeed, radiolysis is accompanied by the formation of chlorine molecules in the cell, their concentration increasing towards the end of the neutron pulse. The formation of molecular chlorine will increase the lifetime of the bubble since some time is required for dissolution of chlorine in phosphorus oxychloride. Unfortunately, we do not know much about this process. However, assuming that the lifetime of a bubble increases towards the end of the neutron pulse, the agreement between theory and experiment improves (see Fig. 3). Thus, the lifetime of the bubble for the fifth and sixth points is 4.5×10^{-8} and 5.7×10^{-8} s respectively. It can be seen that this is slightly longer than the lifetime of the bubble for the first points (3×10^{-8} s).

4. Conclusions

The results obtained in the present work can be summarised as follows. Additional losses occurring in a liquid pumped by ^{235}U fission fragments have been measured experimentally. It is found that these losses mainly correlates with the pump pulse. The thermal model of generation and evolution of a bubble formed as a result of deceleration of fragments in a liquid can be used to explain the source of the additional loss as well as its magnitude. Using this model, we can determine not only the lifetime of a cylindrical bubble being formed ($\sim 3 \times 10^{-8}$ s), but also its geometrical dimensions. Under normal pressure and at room temperature, a bubble has a length 5.2×10^{-3} cm and a radius 2.9×10^{-7} cm at the instant of its formation . After expansion of the bubble, its length does not change but its radius increases to 3.8×10^{-6} cm. The time-averaged length and radius of the bubble, determined from the average cross section of scattering of light with $\lambda = 1.052$ μm are equal to 5.2×10^{-3} and 3.2×10^{-6} cm respectively.

A significant achievement of this research is that a dependence has been established between the geometrical size of the bubble and the pressure in the cell (see Fig. 5). Thus, the maximum radius of the bubble decreases by a factor of 1.2 upon an increase of 1 atm in pressure, and by a factor of 2.4 as the pressure increases by 10 atm. A decrease in the bubble size lowers the scattering cross section by a factor of 1.5 and 4 respectively. Thus, by creating a pressure increase inside the cell, we can really decrease the induced additional loss associated with the slowing down of fission fragments in the liquid.

The effect of radiolysis of phosphorus oxychloride under the action of fission fragments on the lifetime of a bubble has been considered for the first time. However, the process has not been studied in depth and requires a separate analysis.

Finally, becuase the experimentally determined induced losses are almost identical to their theoretical values (see the dotted curve in Fig. 3) used in the calculations of generation parameters of a liquid laser with nuclear pumping [7], the development of a nuclear pumped liquid laser in actual practice remains a distinct possibility.

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