MODELING OF THERMOACOUSTIC AND EVAPORATION PRESSURE SIGNALS IN ABSORBING LIQUIDS IRRADIATED WITH NANOSECOND LASER PULSES

ZUBKO A.E.[*], SAMOKHIN A.A.[†]

[*] Prokhorov General Physics Institute, RussianAcademy of Sciences(GPI RAS)
Vavilov Str., 38, 119991, Moscow, Russia
e-mail: zubko.aleksey11@gmail.com

[†]Prokhorov General Physics Institute, RussianAcademy of Sciences(GPI RAS)
Vavilov Str., 38, 119991, Moscow, Russia
e-mail: asam40@mail.ru

Keywords: nanosecond laser ablation, absorbing liquids, thermoacoustic signal, surface vaporization.

Summary. Pressure generation in absorbing liquids irradiated with nanosecond laser pulses is mainly due to thermoacoustic and evaporation mechanisms provided the laser intensity is not too high. Despite many years of investigations there are some unresolved problems which concern to surface and bulk (explosive) evaporations regimes as well as non-equilibrium superheated liquid behavior in near-critical region. In the present paper pressure behavior in water irradiated with nanosecond laser pulses in the cases of 1 μm and 10 μm absorption length is investigated in the framework of one-dimensional continual approach. It is shown, in particular, how external atmospheric pressure can modify the generated vaporization pressure signals compared with the vacuum case.

1 INTRODUCTION

Laser action on absorbing liquids is investigated for many decades (see *e.g.,* [1-13] and references therein). However, some important aspects of the problem remain unclear. This relates, in particular, to the surface and bulk (explosive) vaporization regimes as well as non-equilibrium superheated liquid behavior in near-critical region. In [12, 13] applicability limits of the surface evaporation model were determined in the case of water irradiated nanosecond laser pulses with wavelength corresponding to 1 μm and 10 μm absorption length. It was mentioned also that the experimental behavior of vaporization pressure comparable in amplitude with thermoacoustic pressure signals [1, 8, 12] differs significantly from the theoretical case where no external atmospheric pressure is taken into account.

In the present paper vaporization pressure signals are calculated taking into account phenomenologically external atmospheric pressure effect which prevents intense vaporization if saturation pressure P_S is lower than atmospheric pressure P_{ext}. In sec. 2 and 3 formulation of the problem and discussion of the obtain results are given. Concluding remarks are given in the fine section 4.

2010 Mathematics Subject Classification: 35K05, 35Q79, 80A22.
Key words and Phrases: Nanosecond laser ablation, Absorbing liquids, Thermoacoustic signal, Surface vaporization.

2 FORMULATION OF THE PRESSURE GENERATION PROBLEM

Generation of acoustic perturbations in irradiated absorbing liquids is usually described with the help of the following equations [12, 13]

$$\frac{\partial \rho}{\partial t} + u\frac{\partial \rho}{\partial z} + \rho\frac{\partial u}{\partial z} = 0 \tag{1}$$

$$\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial z} + \frac{1}{\rho}\frac{\partial P}{\partial z} = 0 \tag{2}$$

$$\rho C_P\left(\frac{\partial T}{\partial t} + u\frac{\partial T}{\partial z}\right) = \frac{\partial}{\partial z}\left(æ\frac{\partial T}{\partial z}\right) + Q \tag{3}$$

$$Q = I(t)\,\alpha\,exp(-\alpha z) \qquad\qquad \rho = \rho(T) \tag{4}$$

where ρ – density, u – velocity, P – pressure, T – temperature, C_P – heat capacity at constant pressure,æ – heat conduction coefficient, Q – absorbed density heat power,α – absorption coefficient. The laser pulse intensity is approximated with the expression

$$I(t) = \begin{cases} I_m \sin(1.14\,t/\Delta t)^4, & 0 \le t \le \Delta t\ \pi/1.14 \\ 0, & other \end{cases}$$

where I_m – maximum value of absorbed laser intensity, Δt – FWHM of laser pulse. For this form $I(t)$ one has the relations $E = I_m \cdot \Delta t \cdot 1.03 = (\partial I/\partial t)_{max} \cdot \Delta t^2 \cdot 0.7$.

From equations (1-4) in linear approximation it follows [3]

$$P(t) = P_1 + \frac{\beta}{C_P}\left(æ\frac{\partial T_1}{\partial t} + \frac{1}{\alpha}\frac{\partial I}{\partial t}\right) = P_1 + P_{ta} \tag{5}$$

where β – thermal expansion coefficient while P_1 and P_{ta} denote surface evaporation and thermoacoustic pressure signals. In equation it is also supposed that at $z = z_1$ the temperature gradient is zero and the generated acoustic wavelength is longer than the absorption and heat diffusion lengths [3].

Vaporization process is determined by the following boundary conditions

$$æ\left(\frac{\partial T}{\partial z}\right)_{z_1} = \rho L v \qquad\qquad z_1 = \int_0^t (v + u_1)dt \tag{6}$$

$$P_S(T) = P_B\, exp\left(A\left(1 - \frac{T_B}{T}\right)\right) P_{ext} = P_S(T_B) \tag{7}$$

$$P_1 = 0{,}56\,(P_S(T_1) - P_{ext}) \cdot h(T_1 - T_B) \tag{8}$$

$$v = 0{,}82\sqrt{\frac{m}{2\pi kT}}\frac{1}{\rho}(P_S(T_1) - P_{ext}) \cdot h(T_1 - T_B) \tag{9}$$

$$h(T_1 - T_B) = \begin{cases} 1 - (y^2/2 + y + 1)\,exp(-y), & y \ge 0 \\ 0, & y < 0 \end{cases} \qquad y = 5.32\,(T_1 - T_B)/\Delta T \tag{10}$$

where P_S – saturated pressure, L – latent heat of evaporation, v – vaporization front velocity,

T_B – boiling temperature for pressure $P_{ext} = P_B$. Accommodation coefficient is equal to unity [14-15]. Effect of atmospheric pressure is taken into account phenomenologically with the help of factor $h(T)$ and substruction P_{ext} in equations (8-9) which tend to vacuum vaporization case with Mach number $M = 1$ [7] at sufficiently large values of P_S compared with P_{ext}. The factor $h(T)$ changes from zero at $T_1 = T_b$ to unity at $T_1 - T_b >> \Delta T$ and $h(\Delta T) = 0.9$.

Applicability limits of the considered model are determined by the condition that the subsurface temperature maximum does not succeed the superheating limit temperature $T_{lim} = 0.9\ T_C$. These limits were investigated in ref. [12-13] for different laser action regime and it was shown that nonlinear effect due to density variation with temperature is not very important in this case. At the present paper the same calculation procedure is used as in ref. [12-13] with the same initial temperature $T_0 = 20$°C. In metallic liquid (Hg) with high values of α the laser energy absorption was considered as a surface effect and absorbed laser intensity was inserted in boundary condition with simultaneous putting $Q = 0$ in equation (4). Numerical values of some parameters used in calculation are shown in table 1 where temperature dependence parameters are taken at $T = 20$°C.

Name	Symbol, dimension	H_2O	Hg
FWHM of laser pulse	Δt, ns	200, 300	35
Absorption coefficient	α, cm^{-1}	$1.2\cdot10^4$, $1.15\cdot10^3$	$>10^5$
Liquid density	ρ, g/cm^3	1.0	13.5
Heat capacity at constant pressure	C_P, J/(g·K)	4.18	0.137
Latent heat	L, J/g	2260	282
Boiling temperature	T_B, °C	100	357
Critical temperature	T_C, °C	374	1480
Threshold temperature interval	ΔT, °C	40	70
Exponentialcoefficient	A	12.7	11.6
Thermal conduction coefficient	χ, cm^2/s	$0.16\cdot10^{-2}$	$5.8\cdot10^{-2}$
Thermal expansion coefficient	β, K^{-1}	$3.7\cdot10^{-4}$	$1.8\cdot10^{-4}$

Table 1.

3 RESULTS AND DISCUSSION

Due to lack of sufficient information on absolute pressure values in experiments [8, 12] only the cases where the thermoacoustic and vaporization signals are comparable in magnitudes is considered here. Effect of external pressure P_{ext} on these signals behavior is shown on fig.1 for the cases of $E = 0.04$ J/cm^2, $\alpha = 1.2\cdot10^4$ cm^{-1} (a) and $E = 0.33$ J/cm^2, $\alpha = 1.15\cdot10^3$ cm^{-1} (b) together with partial amplitude pressure dependence on laser fluence (c,d). It is clear from fig.1 that the threshold effect gives rise to some delay and shortening of pressure signals as compared with vacuum vaporization case.

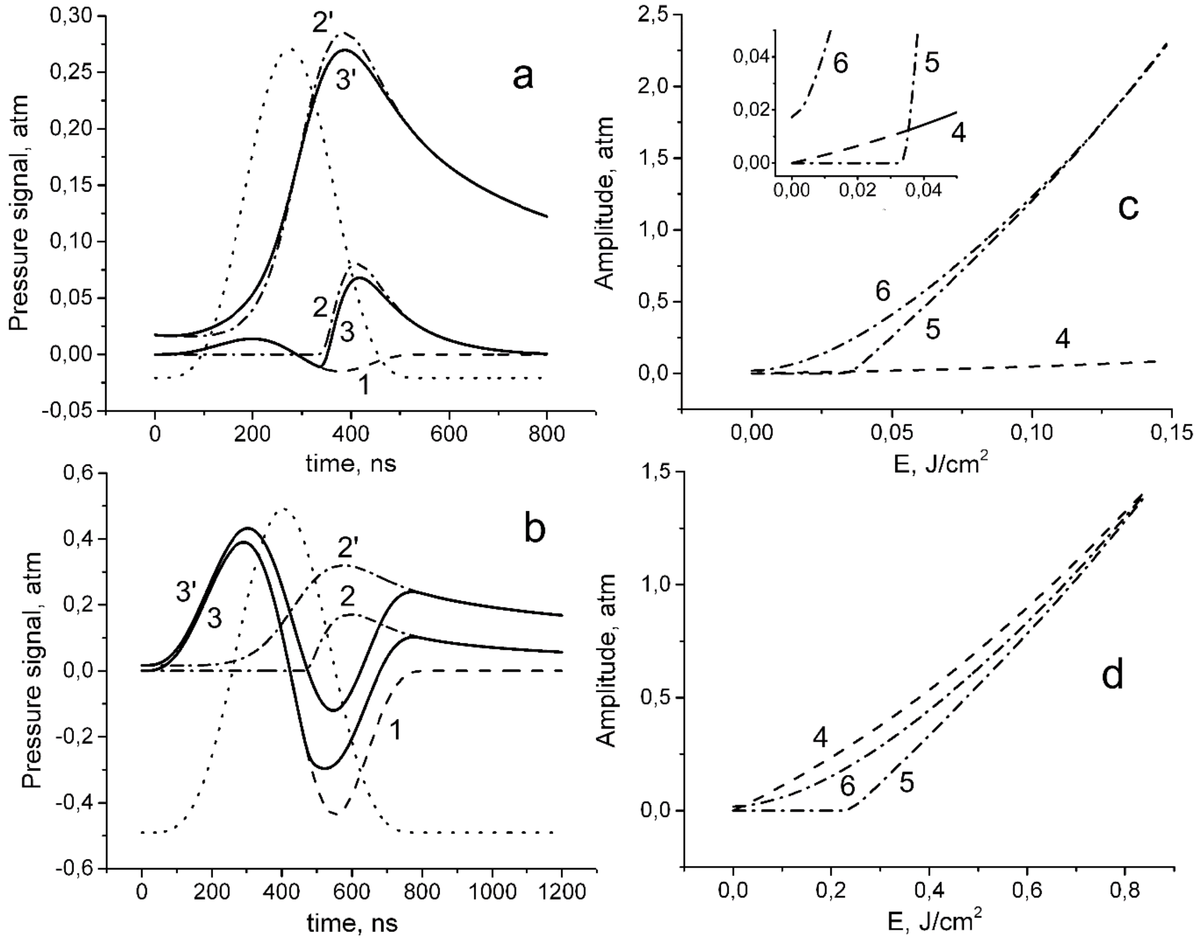

Fig.1. Pressure behavior (a,b) of thermoacoustic signal (curve 1), vaporization signal in atmosphere (2), vacuum vaporization signal (2'), total (1+2) pressure signal (3) and total (1+2') pressure signal (3'), is given by doted line correspondence to normalised laser intensity for $\alpha = 1.2\cdot10^4$ cm^{-1}, $E = 0.04$J/cm^2 (a) and for $\alpha = 1.15\cdot10^3$ cm^{-1}, $E = 0.33$ J/cm^2 (b). Partial amplitude dependence on E for thermoacoustic signal (curve 4), vaporization signal in atmosphere (5), vacuum vaporization signal (6) for$\alpha = 1.2\cdot10^4$ cm^{-1} (c) and for $\alpha = 1.15\cdot10^3$ cm^{-1} (d).

Thermoacoustical and vaporization pressure signals calculated at absorbed laser fluence $E = 37$ mJ/cm^2 for laser pulse width 200 ns are shown of fig.2b. The total pressure behavior qualitatively resembles the experimental curves in fig.2a, 2c in contrast to the vacuum vaporization case shown in [12, 13] where calculated vaporization signals is considerably wider than experimental. Evolution of pressure signals at $E = 38$ mJ/cm^2 in fig.2d also is qualitative accordance with experimental curves in fig.2c.

Experimental pressure curves from [8] are shown in fig.3a, 3c together with calculated pressure curves fig.3b, 3d for the cases where vaporization peak pressure is slightly lower and higher than thermoacoustic pressure peak. The qualitative differences between experiment and theory in fig.3 are mainly the same as in fig.2: calculated pressure peaks are wider and delayed compared with experiment. Diminishing of threshold region ΔT leads to diminishing of the delay (gap) between thermoacoustic and vaporization signals, but the vaporization signal width remains almost the same and somewhat wider the in experiment.

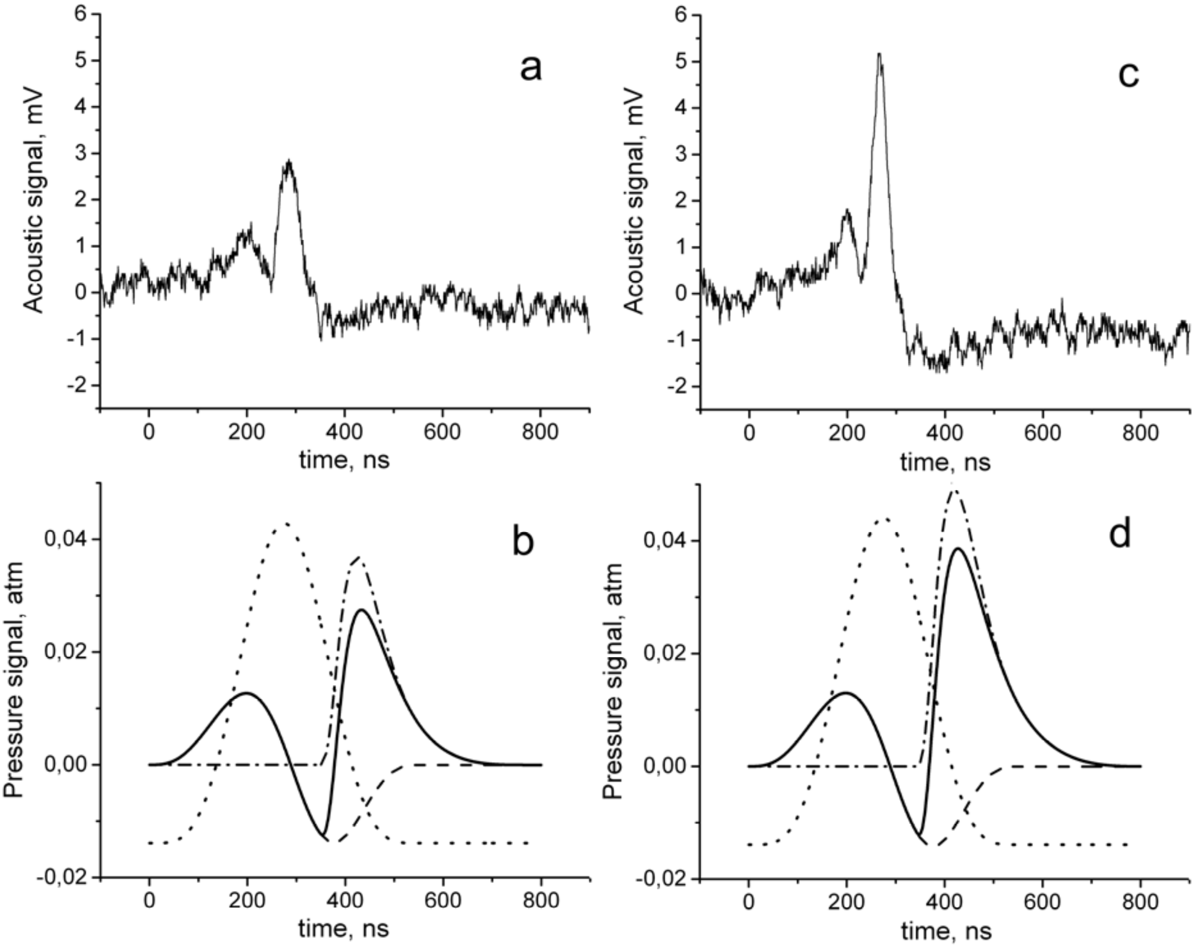

Fig.2. Experimental (a,c) and theoretical (b,d) pressure curves for $\alpha = 1.2\cdot10^4$ cm^{-1} in the case where thermoacoustic and vaporization peaks are comparable. $E = 37$ mJ/cm^2 (b) and $E = 38$ mJ/cm^2 (d). In (b,d): solid line – total pressure signal, doted – laser pulse intensity, dashed – thermoacoustic signal, dash-doted – vaporization signal in atmosphere.

In the case of absorption length $\alpha = 1.15\cdot10^3$ cm^{-1} experimental and theoretical pressure curves (fig.4b) are rather different. It should be mentioned that in the considered model case vaporization peak does not exceed the thermoacoustic one so that the experimental regime fig.4a [1] is probably out of the applicability limit of the surface evaporation model determined by achievement of superheating limit temperature $T_{lim} = 309$˚C in the subsurface region. This condition leads to limiting value of $E = 0.85$ J/cm^2 with corresponding surface and maximum temperature values $T_1 = 140$˚C and $T_m = 305$˚C, respectively. Discussion on the explosive boiling regime (see *e.g.,* [5,9-11] and references therein) is out the scope of the present paper.

During laser ablation of metallic liquids the surface temperature T_1 differs but slightly from the subsurface temperature maximum T_m because of higher values of absorption and heat conduction coefficients than in dielectric liquids. In this case thermoacoustic signal in linear approximation is proportional to $\partial T_1/\partial t$ and not to $\partial I/\partial t$ from equation (5). Dependence of vaporization recoil pressure $P_1(T_1)$ in metallic and dielectric liquids is the same for corresponding thermophysical parameters.

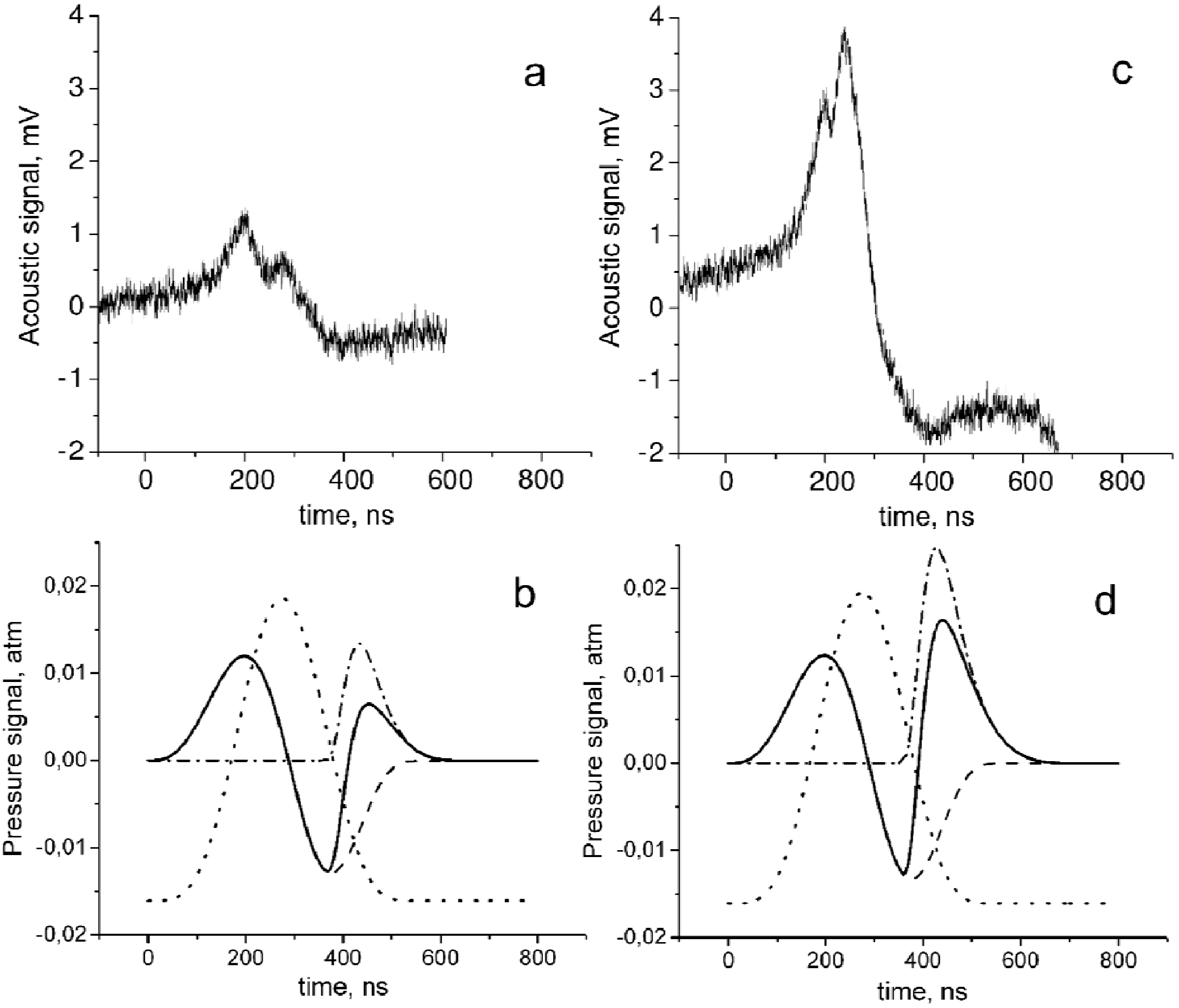

Fig.3. Experimental (a,c) and theoretical (b,d) pressure curves for α = $1.2\cdot10^4$ cm^{-1} in the case where thermoacoustic and vaporization peaks are comparable. E = 35 mJ/cm^2 (b) and E = 36 mJ/cm^2 (d). In (b,d): solid line – total pressure signal, doted – laser pulse intensity, dashed – thermoacoustic signal, dash-doted – vaporization signal in atmosphere.

Irradiation of liquid Hg with 35ns laser pulse gives rise to total pressure recoil shown in fig.5a for three cases with maximum values of T_1= 354, 429 and 500°C exceeding of normal boiling point temperature T_B = 357°C and corresponding to absorption laser fluence E = 30, 36 and 42 mJ/cm^2, respectively. Curve 1 corresponds to the thermoacoustic signal at E = 30 mJ/cm^2 with no vaporization. At higher fluences vaporization peak appears which is closer to thermoacoustic peak than in dielectric case as it is clear fromcomparison of fig.5a with fig.1a, fig.2b,2d, fig.3b,3d. Fig.5b as well as fig.1c shows that at some values of E vaporization peak P_V exceeds thermoacoustic one P_A in contrast to the case of fig.1d where P_V is always greater thenP_A in the applicability limits of the surface evaporation model.

Thus, the theoretical recoil pressure pulse forms in the case of irradiated metal is an agreement with experimental curves obtained in [4] for the intensity where P_V and P_A are comparable with each other.

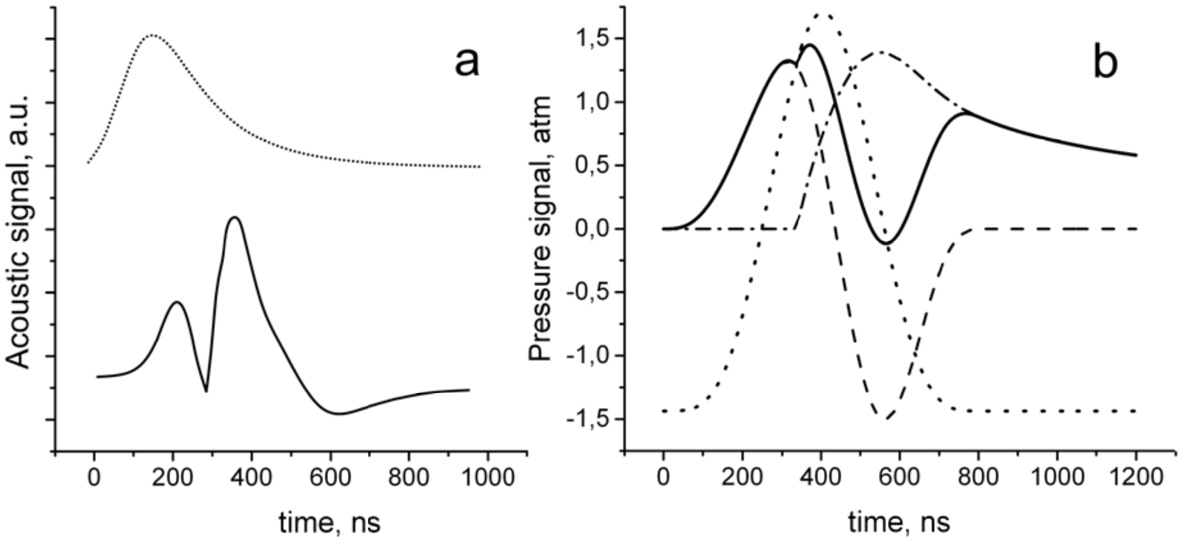

Fig.4. Experimental laser intensity and pressure curves (a), theoretical curves for $\alpha = 1.15 \cdot 10^3$ cm^{-1} (b). In (b): solid line – total pressure signal, doted – laser pulse intensity, dashed – thermoacoustic signal, dash-doted – vaporization signal in atmosphere.

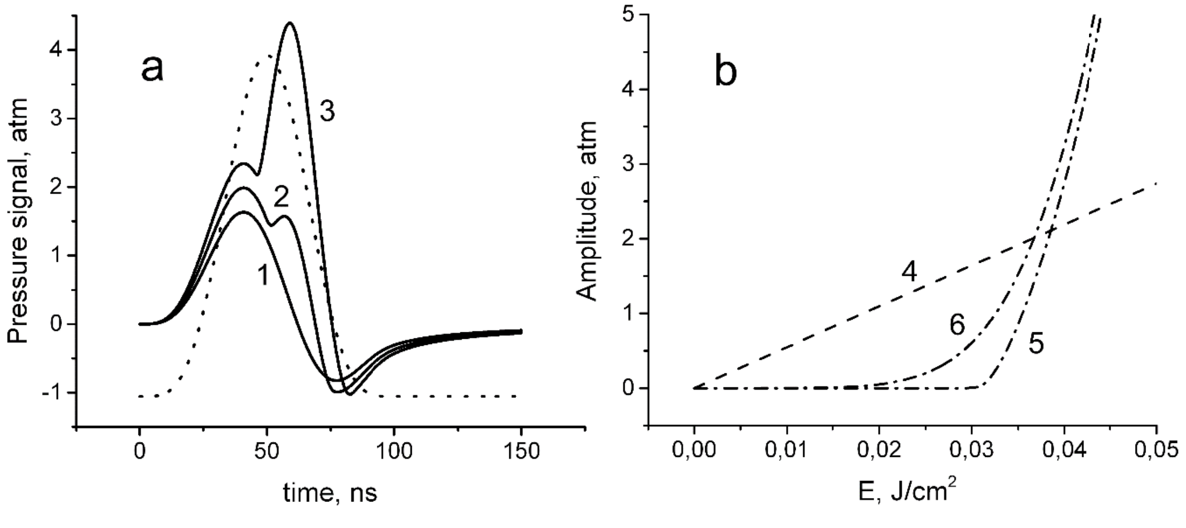

Fig.5. Theoretical (total) pressure curves in irradiated Hg (a) for E = 30 mJ/cm^2 (1), E = 36 mJ/cm^2 (2), E = 42 mJ/cm^2 (3) and partial amplitude dependence on E for thermoacoustic signal (curve 4), vaporization signal in atmosphere (5), vacuum vaporization signal (6).

4 CONCLUDING REMARKS

From the presented calculation it follows that the surface evaporation model of nanosecond laser ablation of dielectric liquids needs further investigations because of significant differences between available experimental and theoretical data. The discrepancy is probably due to variation of laser intensity across the irradiation spot and to the phenomenological approach in the present consideration of atmospheric pressure effect. Role of atmospheric pressure can be investigated experimentally by means of the external pressure diminishing.

In ref. [8] it was mentioned that experimentally observed strongly nonmonotonous modulated pressure amplitude behavior induced by laser amplitude modulation is not probably an accordance with the surface evaporation model. This question has no explicit answer up to now. Possible role of surface vaporization front instabilities [7] is also unclear in nanosecond laser ablation regime similar to those used in [1, 8].

Further experimental and theoretical investigations are also needed for recoil pressure behavior when the explosive boiling process begins. Such investigations can give new information on metastable and unstable liquid behavior in near-critical region.

REFERENCES

[1] M.W. Sigrist and F.K. Kneubuhl, “Laser-generated stress waves in liquids”, *Journal Acoustical Society of America*, **64**(6), 1652-1663 (1978).

[2] L.M. Lyamshev, “Optoacoustic sources of sound”, *Sov. Phys. Usp.*, **24,** 977–995 (1981).

[3] A.A. Samokhin, “The photoacoustic effect induced by phase transitions in an irradiated medium”, *Proceeding of the Institute of General Physics Academy of Sciences of the USSR*, **13**, 66-82 (1990).

[4] I.A. Veselovskii, B.M. Zhiryakov, N.I. Popov, A.A. Samokhin, “The photoacoustic effect and phase transitions in semiconductors and metals irradiated by laser pulses”, *Proceeding of the Institute of General Physics Academy of Sciences of the USSR*, **13**, 179-198 (1990).

[5] A.F. Vitshas, V.V. Korneev, L.P. Menahin, “Konkurencija razlichnyh mehanizmov vzaimodejstvija lazernogo izluchenija s dijelektricheskoj zhidkost'ju” (in Russian), *Izvestija akademii nauk SSSR*, **53**(3), 595-599 (1989).

[6] A. Vogel and V. Venugopalan, “Mechanisms of Pulsed Laser Ablation of Biological Tissues”, *Chem. Rev.*, **103**, 577-644 (2003).

[7] I.N. Kartashov, A.A. Samokhin and I.Yu. Smurov, “Boundary conditions and evaporation front instabilities”, *J. Phys. D: Appl. Phys.*, **38**, 3703–3714 (2005).

[8] A.A. Samokhin and N.N. Il’ichev, “Laser-induced photoacoustic and vaporization pressure signals in absorbing condensed matter: New results”, *Phase Transitions Induced by Short Laser Pulses*, *Nova Science Publishers*, *editor G.A. Shafeev*, 135-154 (2009).

[9] V.I. Mazhukin, A.V. Sharapov, A.A. Samokhin, A. Yu. Ivochkin, “Mathematical modeling of non-equilibriium phase transition in rapidly heated thin liquid film”, *Mathematica Montisnigri*, **XXVII**, 65-90 (2013).

[10] M.Q. Jiang, Y.P. Wei, G. Wilde, and L.H. Dai, “Explosive boiling of a metallic glass superheated by nanosecond pulse laser ablation”, *Applied physics letters*, **106**, 021904 (2015).

[11] Serap Celen, “On mechanism of explosive boiling in nanosecond regime”, *Appl. Phys. B*, 122-168 (2016).

[12] A.E. Zubko, A.A. Samokhin, A.V. Sidorin, “On the use of surface evaporation model during laser ablation of absorbing dielectric liquids” (in Russian), *Engineering Physics*, **5**, 13-19 (2016).

[13] A.E. Zubko, A.A. Samokhin, “Nonlinearities effects on thermoacoustic and evaporation pressure pulses in absorbing liquids during nanosecond laser action” (in Russian), *Engineering Physics*, **8**, 13-18 (2016).

[14] R.E.H. Miles, J.P. Reid and I.J. Riipinen, “Comparison of approaches for measuring the mass accommodation coefficient for the condensation of water and sensitivities to uncertainties in thermophysical properties”, *The Journal of Physical Chemistry A*, **116**, 10810−10825 (2012).

[15] J. Julin, M. Shiraiwa, R.E.H. Miles, J.P. Reid, U. Poschl and I.J. Riipinen, “Mass accommodation of water: bridging the gap between molecular dynamics simulations and kinetic condensation models”, *The Journal of Physical Chemistry A*, **117**, 410−420 (2013).

The results were presented at the 14-th International seminar "Mathematical models &modeling in laser-plasma processes & advanced science technologies" (July 4 - 9, 2016, Moscow, Russia).

Received May, 17 2016.