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Acoustic diagnostics of the explosive boiling up of a transparent liquid on an absorbing substrate induced by two nanosecond laser pulses

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***Abstract*. Photoacoustic pressure signals produced in an absorbing substrate under a layer of a transparent liquid irradiated by two successive nanosecond laser pulses are studied experimentally. The first pulse causes the heating and explosive boiling up of the liquid layer in contact with the irradiated surface, while the second pulse reaches the target at the time when a vapour film has already formed on its surface. The second photoacoustic response contains additional information on heat-and-mass transfer between the liquid and substrate surfaces separated by the vapour film.**

Keywords: *photoacoustic effect, explosive boiling up, heat-and-mass transfer.*

1. Introduction

The study of the interaction of a liquid with another strongly heated solid (or liquid) is of current interest due to its practical importance and the presence of some unsolved fundamental problems related to the dynamics of non-equilibrium phase transformations and accompanying heat-and-mass transfer processes (see, for example, [1 – 5] and references therein).

One of the methods for experimental studying such processes is the detection of acoustooptic signals induced by them, in particular, in the case of additional pulsed irradiation. In this paper, we present the results of measuring pressure signals induced by two nanosecond laser pulses in an absorbing (metal) substrate covered by a layer of a transparent liquid. The absorption of the first pulse leads to the contact heating and explosive boiling up of a liquid layer on the substrate surface. The second laser pulse arrives at the moment when a vapour layer has already formed between the substrate and liquid due to explosive boiling up.

2. Formulation of the problem

Let us recall first of all some fundamental concepts concerning the mechanism of formation of a photoacoustic signal in absorbing media with free and not free interfaces (see, for example, [6, 7]).

The shape of pressure pulses propagating into an irradiated substrate is determined by variations in the temperature T_i and density ρ_i ($i = 1, 2$) of material media in the region of their contact for $z = 0$, where the subscript 1 corresponds to the left half-space (liquid) and the subscript 2 is related to the right half-space (substrate). Before explosive boiling up, the behaviour of temperature is described by the heat conduction equation

$$\frac{\partial T_2}{\partial t} = \chi_2 \frac{\partial^2 T_2}{\partial z^2} + \frac{\alpha}{\rho_2 c_{p2}} I_0 \exp(-\alpha z), \quad z > 0, \tag{1}$$

$$\frac{\partial T_1}{\partial t} = \chi_1 \frac{\partial^2 T_1}{\partial z^2}, \qquad z < 0 \tag{2}$$

with the boundary conditions of the temperature and heat flow continuity at the interface:

$$T_1 = T_2, \quad \kappa_1 \frac{\partial T_1}{\partial z} = \kappa_2 \frac{\partial T_2}{\partial z}, \ \ z = 0, \tag{3}$$

where I_0 is the absorbed laser pulse intensity; α, χ_i, and $\kappa_i = \rho_i c_{pi} \chi_i$ are the absorption coefficient, the thermal diffusivity, and the heat conductivity coefficient, respectively; c_{pi} is the specific heat at constant pressure; and temperature T_i is measured from the constant initial value T_{0i}. Note that the temperature continuity condition can be, generally speaking, violated if the heat exchange at the interface between two media is deteriorated [5].

By assuming that a change in the density $\Delta\rho = \rho_i - \rho_{0i} = -\beta_i \rho_i \Delta T_i$ in the heated region of the substrate is mainly caused by its temperature dependence, we obtain from the linearized continuity and Euler equations (see, for example, [6])

$$\frac{\partial^2 P_2}{\partial z^2} = \frac{\partial^2 \rho_2}{\partial t^2} = -\beta_2 \rho_2 \frac{\partial^2 T_2}{\partial t^2}, \tag{4}$$

where β_i is the thermal expansion coefficient. The approximation used in (4) means, in particular, that the depth $z_{\rm m} = \max(1/\alpha, \sqrt{\chi_2 t})$ of substrate heating during the laser pulse duration τ determined by the depth of absorption ($1/\alpha$) or temperature influence ($\sqrt{\chi_2 t}$), is small compared to the characteristic wavelength $\lambda = v_{\rm s2} t$ of the excited acoustic pulse, where $v_{\rm s2}$ is the sound speed in the substrate. For pressure $P(z^*, t)$ in the region $z_{\rm m} < z^* < \lambda$, we obtain from (4):

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$$P(z^*,t) = P(0,t) + \int_0^{z^*} \mathrm{d}z' \int_z^{z^*} \frac{\partial^2 \rho_2}{\partial t^2} \mathrm{d}z$$
$$= P(0,t) + \frac{\beta_2}{c_{p2}} \left[\kappa_2 \frac{\partial T_2}{\partial t} + \frac{1}{\alpha} \frac{\partial I_0(t)}{\partial t} \right]. \quad (5)$$

Because the radiation intensity absorbed in this region and temperature perturbations can be assumed zero, the pressure $P(z^*,t)$ is virtually independent of z^*, i.e. $P(z^*,t) = P(t)$, which can be used as the boundary condition for an acoustic wave propagating deep inside the substrate [6, 7], which can be detected with a pressure transducer located, for example, on the rear side of the substrate.

A photoacoustic signal detected in this way depends on the pressure $P(0,t) = P_0(t)$ at the interface $z = 0$. If this interface is free, i.e. $P_0 = 0$, then upon the action of a laser pulse symmetric in time with respect to its maximum, a photoacoustic signal will have a bipolar shape, according to (5).

In the case when the substrate surface borders the half-space of another continuous medium, the value of $P_0(t)$ can be determined by solving the complete problem in the left and right half-spaces under the condition of the equality of pressures and velocities at the interface. Taking into account the relation between the pressure and velocity in a sound wave, this condition can be written in the form

$$v_1 - \frac{P_0 - P_1}{a_1} = v_2 + \frac{P_0 + P_2}{a_2}, \quad (6)$$

where

$$a_i = \rho_i v_{si}; \qquad P_1 = \frac{\beta_1 \kappa_1}{c_{p1}} \frac{\partial T_1}{\partial t}\bigg|_{z=0};$$
$$P_2 = \frac{\beta_2}{c_{p2}} \left[\kappa_2 \frac{\partial T_2}{\partial t}\bigg|_{z=0} + \frac{1}{\alpha} \frac{\partial I_0(t)}{\partial t} \right];$$
$$v_1 = \beta_1 \int_{-z^*}^{0} \frac{\partial T_1}{\partial t} \mathrm{d}z; \qquad v_2 = -\beta_2 \int_0^{z^*} \frac{\partial T_2}{\partial t} \mathrm{d}z.$$

It follows from (6) that the pressure pulse in the substrate has the form

$$P(t) = P_0 + P_2 = \frac{a_2(P_2 - P_1)}{a_1 + a_2} + \frac{a_1 a_2}{a_1 + a_2}(v_1 - v_2),$$
$$v_1 - v_2 = Q\left(\frac{\beta_1}{\rho_{01} c_{p1}} - \frac{\beta_2}{\rho_{02} c_{p2}} \right) + \frac{\beta_2}{\rho_{02} c_{p2}} I_0(t); \quad (7)$$

where

$$Q = \kappa_1 \frac{\partial T_1}{\partial z}\bigg|_{z=0} = \kappa_2 \frac{\partial T_2}{\partial z}\bigg|_{z=0}$$

is the heat flow through the interface of two media. If the contact heating of a transparent medium can be neglected (for $Q < I_0$), expression (7) is simplified:

$$P(t) = \frac{a_2}{a_1 + a_2} \frac{\beta_2}{c_{p2}} \left[\kappa_2 \frac{\partial T_2}{\partial t}\bigg|_{z=0} + \frac{1}{\alpha} \frac{\partial I_0(t)}{\partial t} \right] + \frac{a_1}{a_1 + a_2} \frac{v_{s2} \beta_2}{c_{p2}} I_0(t). \quad (8)$$

The last term in the right-hand side of (8) is proportional to the absorbed intensity, i.e. unlike the case of a free interface, a photoacoustic signal can have a monopolar shape if this term is the main one in (8).

After the beginning of the explosive boiling up and formation of a vapour film, the boundary conditions for the acoustic signal on the surface being irradiated change. If, for example, upon changing its volume the vapour film behaves as a cavity with saturated vapour contacting with liquid at constant temperature, $P(0,t)$ in (5) can be assumed approximately constant during the expansion of the substrate heated by the second laser pulse. In this case, the second photoacoustic signal will have the bipolar shape against the pressure produced by the first laser pulse. However, this simplest case neglects a number of other possible effects that can considerably affect the shape of the photoacoustic signal produced by the second laser pulse. The relative role of these effects can be qualitatively estimated from the shape of the photoacoustic response to the second laser pulse.

3. Experimental

We used in experiments a 1.064-μm Nd:YAG laser emitting nearly Gaussian 2-ns (FWHM) pulses. Figure 1a shows the scheme of the experimental setup for generating two laser pulses of the same duration separated in time by a few nanoseconds. The laser beam was split with a beamsplitter into two beams with the intensity ratio of 1:6. The first (higher-energy) beam was directed to a sample, and the second one propagated through a delay line

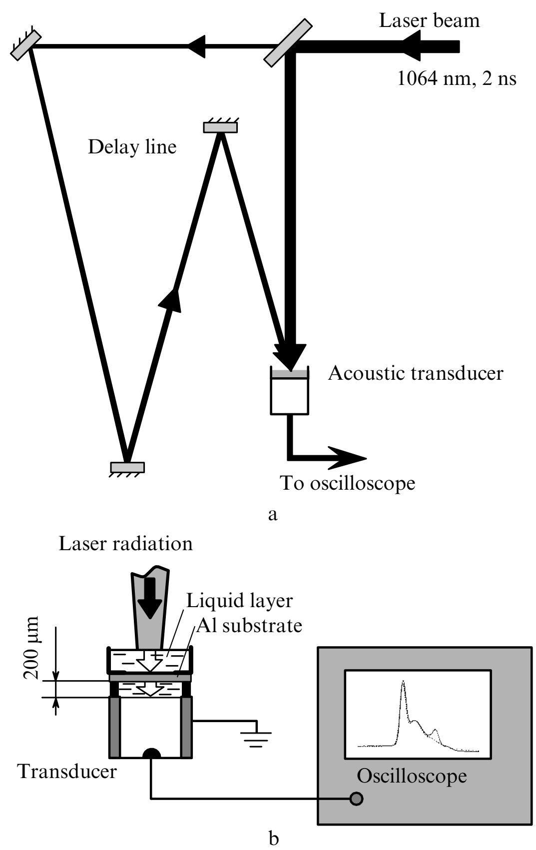

Figure 1. Scheme of the experimental setup: delay line (a) and recording a pressure signal (b).

and then was incident on a sample within 19 ns after the arrival of the first laser pulse. The ratio of the diameter of the main beam to that of the delayed beam was varied in experiments from unity to two, the main beam diameter being constant (6 mm). The main beam energy incident on a sample could be varied with an attenuation system place in the beam. The delayed pulse energy remained constant or was varied proportionally to a change in the main pulse energy.

Figure 1b presents the scheme for recording a pressure pulse. A pressure transducer (lithium niobate crystal) was mounted under a 1-mm-thick aluminium substrate and was separated from it by a thin ($\sim$200 μm) liquid layer to provide good acoustic contact. Signals of two types were measured in experiments: a pressure transducer signal in the presence of a few millimetre-thick liquid layer (water or acetone) on the irradiated surface of the aluminium substrate and the so-called dry signal detected upon irradiation of the dry aluminium surface. The pressure pulse signal from the transducer was recorded with an oscilloscope.

4. Results and discussion

Figure 2 presents the oscillograms of pressure pulses recorded by exposing the dry aluminium target to single- or two-pulse laser radiation. According to expression (5), the pulses have a characteristic bipolar shape, the delay between successive pressure pulses corresponds to the 19-ns optical delay, and their amplitude ratio is in good agreement with the energy ratio of laser pulses. Recall that the negative half-wave of such a pulse is produced by a thermal expansion wave reflected from the free boundary.

To determine the absolute value of pressure from (5) and (8), it is necessary to know only the target absorptivity because the rest of the thermal parameters of the target are well known. By assuming now that the absorptivity of the system changes weakly in the presence of the liquid or vapour phase on the target surface, we can analyse the dynamics of the absolute value of pressure on the sample surface by comparing analytic expressions (5) and (8) with oscillograms of photoacoustic pressure. In particular, the pressure corresponding to the explosive boiling up of the overheated liquid can be found in this way.

The oscillograms obtained upon laser heating of a sample under a transparent liquid (water) layer by pairs of laser pulses are presented in Fig. 3 for two energies of the first pulse, the second-pulse energy being 17 % of the first-pulse energy in both cases. The dashed curves show the shapes of signals measured upon irradiation by the first pulse only. The signal amplitude in Fig. 3a exceeds that of the 'dry' signal (Fig. 2a) for the same laser pulse energy due to the additional back pressure from the adjacent liquid phase. In the given case, this excess is not great (20 % – 30 %) and is in satisfactory agreement with estimates made by expressions (5) and (8) predicting a somewhat greater value. According to the same estimates, the absolute pressure value does not exceed 40 atm when the temperature of the substrate surface achieves 300 °C. The presence of the back pressure also explains the monopolar shape of measured signals.

The trailing edge of the first pulse exhibits a distinct feature corresponding to the explosive boiling up. The surface temperature at this moment exceeds the boiling temperature of the liquid at the pressure acting on it,

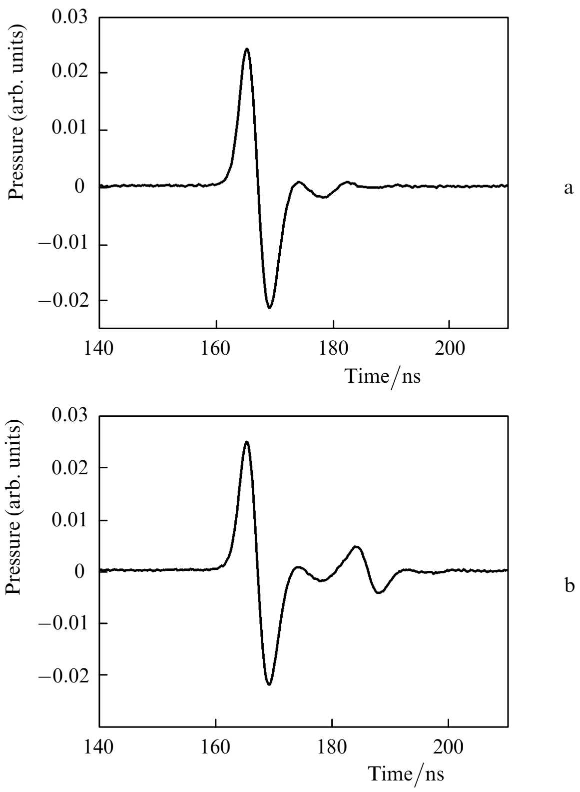

Figure 2. Pressure signals observed upon irradiation of the dry target surface by one (a) and two (b) laser pulses. The bipolar signal shape in both cases corresponds to the free (unloaded) interface.

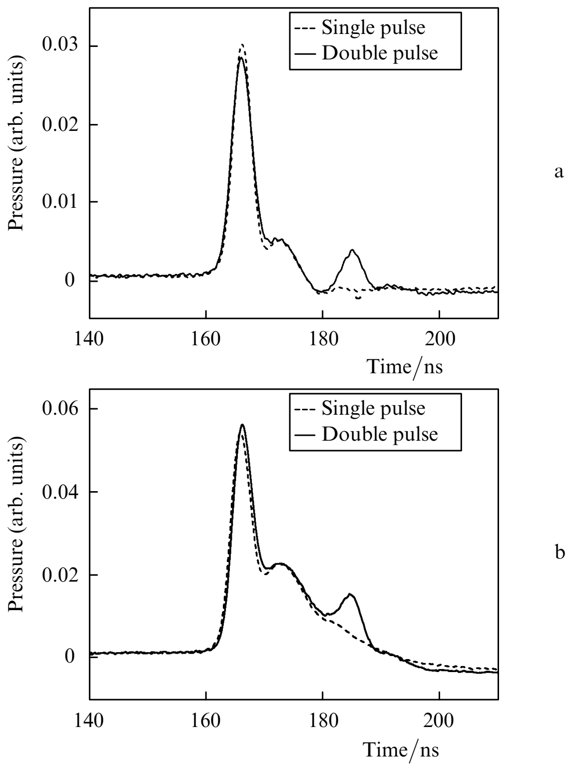

Figure 3. Comparison of pressure signals produced by single and double laser pulses for the first-pulse energies 17 (a) and 30 mJ (b). In the case (b), the probe pulse acts on the target covered by a vapour film, and the pressure response produced by the probe pulse has the monopolar shape.

resulting in the appearance of a vapour cavity and an additional pressure pulse on the target surface. As the laser pulse energy increases (Fig. 3b), this pressure increases faster than the amplitude of the photoacoustic signal. In this case, the width of the additional pressure pulse, corresponding to the lifetime of the vapour cavity, also increases. For the energy corresponding to Fig. 3b, this width exceeds the delay time between two laser pulses, and the second pulse is incident on the substrate at the moment when the vapour cavity exists. Note that the pressure signal produced by the second (probe) laser pulse remains monopolar in this case, as in the absence of the vapour cavity (Fig. 3a).

The invariable shape of the second signal and its proportionality to the incident energy in the presence and absence of a vapour cavity contradict the above-mentioned assumption about a constant pressure P_0 on the substrate surface in contact with a vapour film. This result can be explained by several reasons. In the range of vaporisation pressures P_v under study which do not exceed 10 atm and for the vapour-cavity lifetime ~ 10 ns, the vapour-cavity thickness $h \approx P_v t / a_1$ proves to be quite small (a few nanometres), which enhances the role of local non-one-dimensional effects related to the state of the real surface of the substrate and breaking the continuity of the film. The improvement of the surface quality and increasing the vapour-cavity thickness, for example, by increasing the main-pulse duration will weaken the influence of these effects. In addition, in the case of the one-dimensional approximation, the assumption that P_0 is constant may be not fulfilled due to the additional heat flow to the liquid surface adjacent to the vapour film through this film from the substrate surface heated by the probe laser pulse. Let us emphasise that the mean free path of vapour molecules under conditions under study considerably exceeds the thickness h of the film being formed.

5. Conclusions

Our experimental study and simple linearized one-dimensional estimates have shown that pressure produced by a probe pulse on the substrate surface due to compression of a vapour cavity is not constant (compression is caused by the thermal expansion of the substrate) because otherwise the signal would have the bipolar shape, as observed for a free surface. Thus, the vapour film produced in our experiments cannot be treated as a cavity containing saturated vapour in contact with a liquid at constant temperature.

We have also shown that the two-pulse photoacoustic method can be successfully used for studying heat-and-mass transfer processes during the explosive boiling up of liquids in contact with a substrate heated by laser pulses. In this case, to create conditions close to the one-dimensional ones, which are the most convenient for quantitative analysis, it is necessary to use laser pulses with the uniform intensity distribution over the laser beam spot. The results of such experiments will be presented elsewhere.

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