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EXPLOSIVE BOILING OF TRANSPARENT LIQUIDS ON ABSORBING TARGETS HEATED BY SHORT LASER PULSES

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Abstract

Explosive boiling induced by sub-nanosecond laser pulses is theoretically analyzed in the case of transparent liquids on metal targets. In this case the variation of boiling start times due to inhomogeneous distribution of laser intensity through the irradiation spot can be reduced up to laser pulse duration and registered explosive boiling pressure signals become less distorted. The results are compared with experimental investigations of photoacoustic (PA) signals induced in metal target under transparent liquid layer irradiated by laser pulses of about 100 ps duration and wavelength 532 nm.

Introduction

Explosive boiling process including the case of transparent liquids on absorbing targets heated by laser pulses was studied for many years. Nevertheless there are some unsolved important questions in this problem, in particular concerning time dynamics of the process and its peculiarities in near-critical region. Explosive boiling can only occur when the pressure in the heated zone is lower than critical pressure and this fact can be used to experimentally determine critical pressure value. During the process of laser ablation in sub-critical region explosive boiling can manifest itself as multiple pressure peaks in PA signal from the irradiated zone [1].

Earlier no such peculiarities in pressure behavior were observed for nanosecond laser ablation, with the exception of works [2, 3], where short (sub-nanosecond) pressure pulses were registered during irradiation of water with erbium laser pulses (τ = 200 ns, λ = 2.94). The pressure increase due to explosive boiling was observed e.g. in [4] as increase of shock-wave velocity in ambient atmosphere near the irradiated Al target. Explosive boiling also took place in the experiments [5, 6] where the process of ejection of thin transparent liquid film from the laser heated target was observed. However in this case the pressure behavior also was not directly registered.

Vaporization process of transparent liquid (water) on pulsed-laser-heated metal surface (KrF excimer laser with pulse duration 24 ns and λ = 248 nm) was investigated in [7]. These authors wrote that when the laser fluence exceeds the bubble nucleation threshold ~ 49.8 mJ/cm^2, the pressure pulse width jumps to 70 ns, which is comparable to the bubble growth time ~100 ns. They concluded that the rapid bubble growth rather than collapse is a source of enhanced pressure generation.

It is clear that the registered pressure rise time in explosive boiling can significantly exceed the real rise time of the process if the radiation intensity is not constant over the radiation spot due to variation of explosive boiling start time in different points. While in ref. [7] it was mentioned that good spatial uniformity of the excimer laser beam is attained by using a tunnel-type beam homogenizer no quantitative estimations of inhomogeneity are given.

It is possible to reduce the inhomogeneity effect by diminishing of space variation of radiation intensity or using sufficiently short laser pulses. In the later case the variation of boiling start times reduces to laser pulse duration provided it is not too short compared with the nucleation time.

It should be mentioned further that laser pulse duration determines the pressure value in the boundary region between liquid and target. If this pressure exceeds the saturation pressure of liquid at achieved temperature then no explosive boiling occurs during laser pulse action.

In this paper the explosive boiling process of transparent liquid on absorbing target heated by sub-nanosecond laser pulse is investigated.

First, some theoretical analysis of the PA and vaporization pressure signals is presented. Then the theoretical estimations are compared with experimental results obtained for the case of transparent liquid (water) on metal (gold) targets heated by laser radiation with wavelength 532 nm and pulse duration τ ~ 0.1 ns. Concluding remarks are given in the final section.

Theoretical estimations of photoacoustic and vaporization pressure signals

Photoacoustic signal in the halfspace $z>0$ of the absorbing irradiated matter due to its thermal expansion can be written in linear approximation as follows (see e.g. [3, 8]):

$$P(z^*,t)=P_0(t)+\int_0^{z^*}dz'\int_{z'}^{z^*}\left(\frac{\partial^2 P}{\partial z^2}\right)dz= \quad (1)$$
$$=P_0(t)+\frac{\beta}{c_p}\left\{\kappa\frac{\partial T}{\partial t}+\frac{1}{\alpha}\frac{\partial I_0}{\partial t}\right\}$$

where $P_0(t)$ and $T(t)$ are the pressure and the temperature at the metal surface $z=0$, β - thermal expansion coefficient, c_p - heat capacity, κ - thermal conductivity coefficient, α - light absorption coefficient, I_0 - absorbed laser intensity. Equation *(1)* is valid if $\sqrt{\chi\tau}<z^*<c\tau$, where c - sound velocity of metal (for gold c = $3.2\cdot10^3$ m/s), $\chi=\kappa/\rho c_p$ - thermal diffusivity coefficient.

At the free surface $P_0(t)=0$ and the signal temporal form is bipolar. If the surface is not free, for example due to contact with liquid layer, then in linear approximation instead of (1) one has:

$$P(t)=K\frac{\beta}{c_p}\left(\kappa\frac{\partial T}{\partial t}+\frac{1}{\alpha}\frac{\partial I_0}{\partial t}\right)+(1-K)\frac{c\beta}{c_p}I_0(t) \quad (2)$$

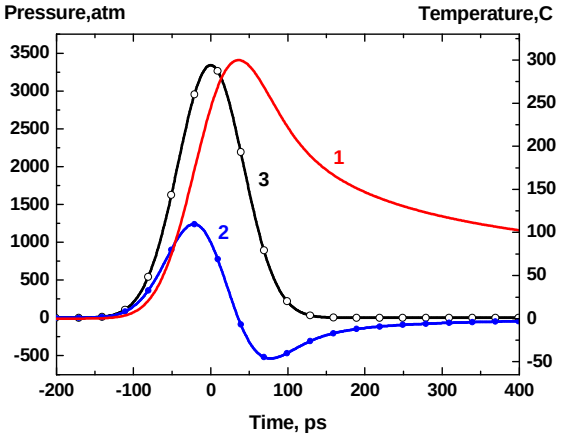
where $K=\rho c/(\rho c+\rho_l c_l)$, ρ - metal density, ρ_l - liquid density, c_l - liquid sound velocity. In (2) the contact heating of liquid layer is neglected. In this approximation water pressure effect on photoacoustic signal is small because $\rho_l c_l < \rho c$ and K~1. If instead of water we have a matter with acoustical impedance $\rho_l c_l$ much greater than that of gold, then in equation (2) K<<1 and only the second term remains. In the considered approximation, where $\sqrt{\chi\tau}<<c\tau$, this term should be always greater than the first term at K~1.

For metals with high absorption coefficient the term with $1/\alpha$ in (1)-(2) can be omitted. In this approximation the surface temperature $T(t)$ is determined from the expression:

$$T(t)=\int_{-\infty}^{t}\frac{I_0(t-t_1)}{\rho c_p\sqrt{\pi\chi t_1}}dt_1 \quad (3)$$

Surface temperature evolution (curve 1) and PA signals generated in gold target with free K = 1 (curve 2) and confined K = 0 (curve 3) surfaces irradiated with laser pulses are presented at Fig. 1 for pulse durations 100 ps (a) and 400 ps (b). The laser pulse temporal form corresponds to curve (3) in accordance with equation (2) for K = 0.

(a)



(b)

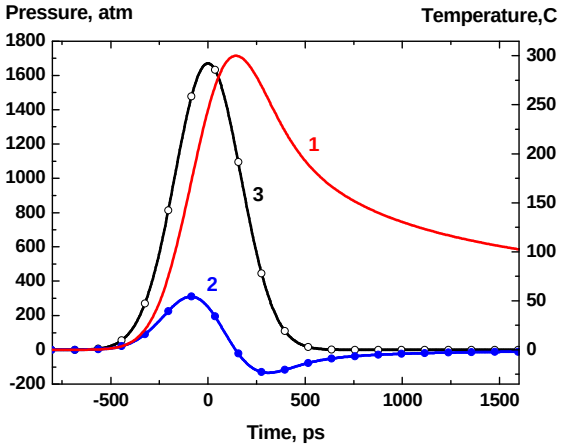


Fig. 1 Surface temperature (1) and PA pressure signals at the free (2) and confined (3) gold surface for different laser pulse durations τ and fluences E: (a) τ = 100 ps, E = 10 mJ/cm^2, (b) τ = 400 ps, E = 20 mJ/cm^2

However the linear approximation is not sufficient to quantitatively describe the water pressure effect in our case while it is sufficient for description of the PA signal generated in the gold target. When water is heated up to $0.95T_c$ ~ 340 ºC its density reduces more than 1.6 times which is far beyond the linear approximation (for water values of critical temperature and density are T_c = 647 K, ρ_c = 0.32 g/cm^3 correspondingly).

If it is supposed that the water density depends only on temperature then one can obtain from the continuity equation the convective velocity v_l in water. This velocity can be used further to estimate the pressure at the liquid-metal boundary with formulae $P=\rho_l c_l v_l$. As it has already been mentioned in introduction the explosive boiling can occur only if this pressure is lower than critical value P_c.= 220 atm.

It should be minded that the boiling process can begin after the laser pulse because the generated

pressure relaxes faster than the surface temperature (see Fig. 1). Explosive boiling begins when the pressure at the boundary drops below saturation pressure and the liquid temperature T attains its superheating limit The maximum pressure jump during this process corresponds approximately to the saturation pressure value $P_s(T)$ at the superheating limit in this condition. For water $P_s(T)$ = 98.7 atm at $T = 0.9T_c$ and $P_s(T)$ = 146 atm at T = 0.95 T_c. These pressure values are considerably lower than shown at Fig. 1 PA pressure generated in gold target. This fact gives rise to certain problems in registration of explosive boiling in the case of short laser pulses.

Experimental results

PA pressure signals from irradiated zone were measured with piezoelectric transducer ($LiNbO_3$) working in current source regime. In this case electric signals are proportional to the pressure signals if the time constant τ_0 of the transducer circuit is smaller than laser and acoustic pulse durations. It follows from the equation for voltage signal U in the transducer circuit (see, e.g., [9]):

$$U + \tau_0 \frac{dU}{dt} = \frac{\gamma S_0 R}{d} \int_0^d \frac{\partial p(t,z)}{\partial t} dz \qquad (4)$$

where $p(t,z)$ - pressure wave in the piezotransducer, $\tau_0 = RC_0$, R - resistive load, γ - piezoelectric constant, S_0 - area of the gauge, d - its thickness, $C_0 = \frac{\varepsilon S_0}{4\pi d}$ - capacity of the gauge.

Experimental setup is shown at Fig. 2.

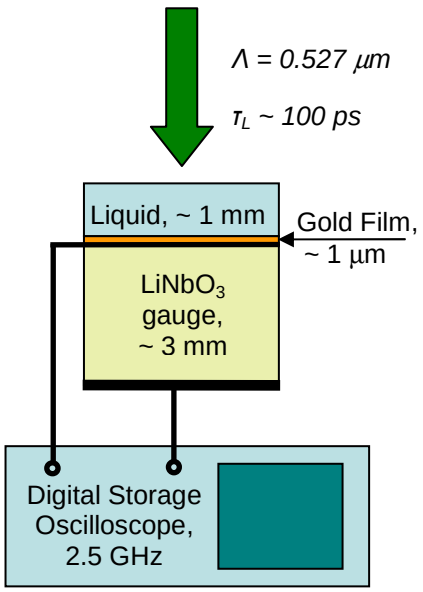


Fig. 2 Scheme of experimental setup

Multiply signals from $LiNbO_3$ piezotransducer with period $\Delta t = d/c_t$ = 420 ns corresponding to its thickness d = 3 mm and sound velocity $c_t = 7.1 \cdot 10^3$ m/s are shown at Fig. 3. The first positive signal comes from upper surface of the gauge and the first negative signal – from the bottom surface. Other signals are caused by multiple reflections from these boundaries. Approximately two-fold increase of the first negative signal amplitude compared with the first positive signal is due to gauge properties with almost free bottom surface [9] and can be obtained from equation (4).

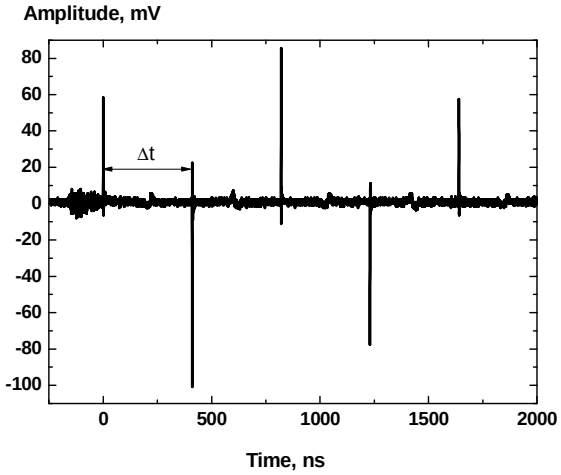


Fig. 3 Multiple reflection of PA signal in piezotransducer with period Δt = 420 ns

PA signals in the case of dry (1) and water-covered (2) gold surface are shown at Fig. 4. As it is expected the signal (2) is greater than (1) and the difference is larger than described by expression (2) in linear approximation. Simple estimations of mentioned above nonlinear water contribution to PA signal show that it is comparable with relative difference between signals (2) and (1) in Fig (4).

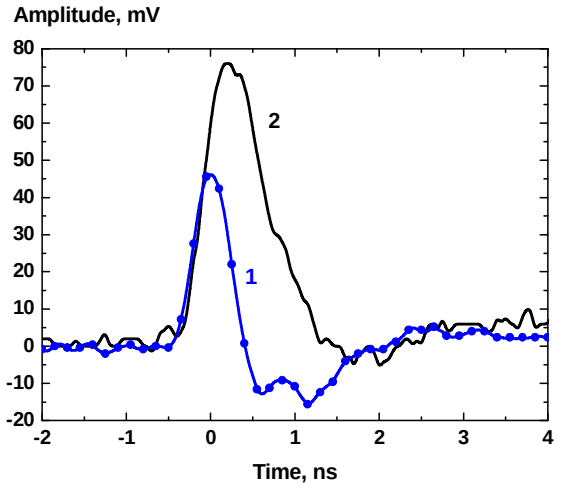


Fig. 4 Experimental PA signals from free (1) and confined (2) gold surface

At Fig. 5 several pressure signals with different amplitude are shown which have some shoulder at the falling part of the pulse. For normalized signals (Fig. 5b) these shoulders have the same form and position. It is supposed that these peculiarities visible also in Fig. 4 are due to acoustic pulse reflections in gold film because its time delay approximately corresponds to sound round trip time in the film $\Delta t_g = 2h/c = 0.6$ ns, where $h = 1$ μm is gold film thickness.

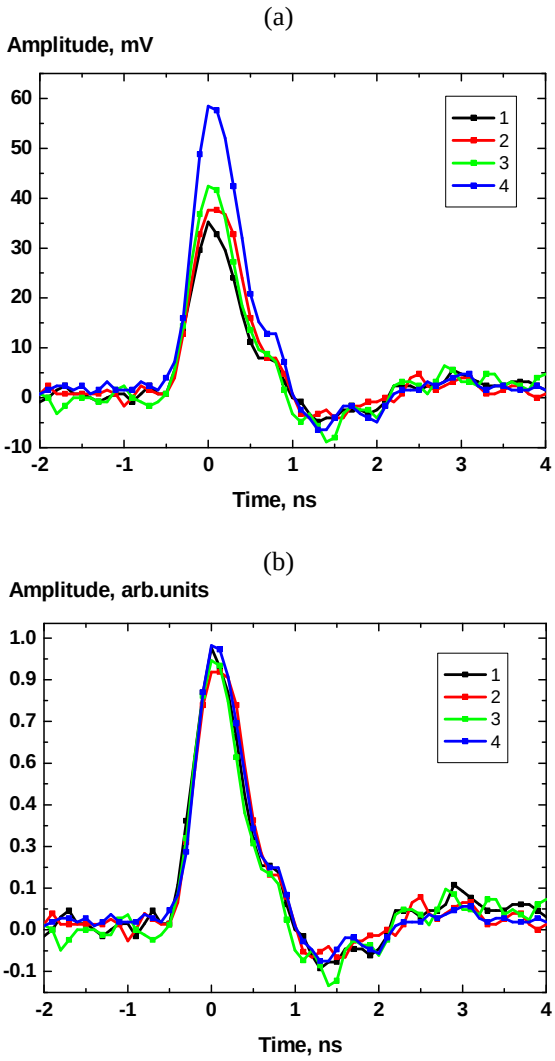


Fig. 5 Experimental PA signals (absolute values (a) and normalized curves (b)): 1 – E = 32 mJ/cm^2, 2 – E = 33 mJ/cm^2, 3 – E = 44 mJ/cm^2, 4 – E = 44 mJ/cm^2

The PA pulses width in presented signals is greater than it should be for laser pulses duration $\tau = 100$ ps. This discrepancy is not fully understood now. It may be, among other reasons, due to thickness inhomogeneity of gold film or to the fact that characteristic gauge circuit time τ_0 is not small compared with laser pulse duration τ.

From theoretical estimations it follows that photoacoustic pressure is rather high for laser intensity which is necessary to obtain the superheating limit temperature. It means that pressure rise due to explosive boiling should be relatively low compared with the total registered pressure signals and should appear after its maximum.

Possible manifestation of such effect is illustrated with Fig. 6 where four signals are presented in the narrow range of laser fluence E. The curve at E = 35 mJ/cm^2 corresponds to the case with preheated target which temperature was higher (ΔT ~ 60-70 °C) than initial target temperature T ~ 20 °C in all other cases.

This signal and the curve at E = 47 mJ/cm^2 are somewhat wider than the other two curves in the range. This change of the width can be probably attributed to the explosive boiling pressure shown at Fig.7 as difference signal which amplitude is about 20 % from the total pressure signal.

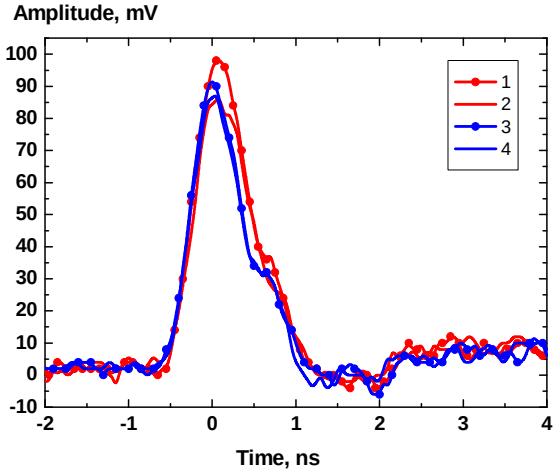


Fig. 6 Experimental photoacoustic signals: 1 – E = 47 mJ/cm^2, 2 – E = 35 mJ/cm^2 (preheated), 3 – E = 42 mJ/cm^2, 4 – E = 43 mJ/cm^2

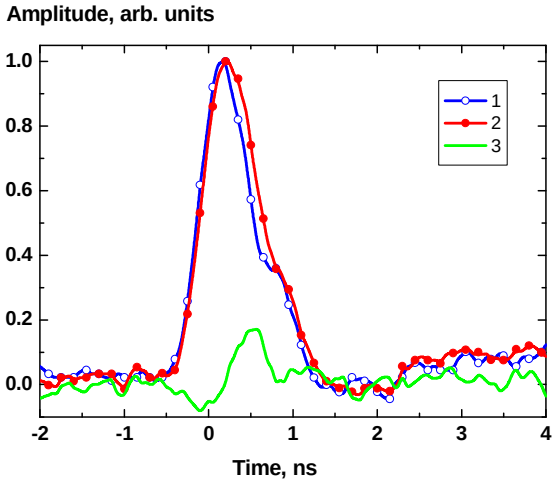


Fig. 7 Normalized photoacoustic signals of different width: 1 – 700 ps, 2 – 800 ps, 3 – difference between (1) and (2)

Conclusions

Sub-nanosecond PA pressure pulses in gold target induced by sub-nanosecond (~ 0.1 ns) laser irradiation are registered in the cases of dry and water-covered surface.

The observed difference between the two cases is greater than predicted in linear approximation because of nonlinear response of heated water layer.

PA pulses widths (~ 400 ps in dry surface case) exceed the estimated value which corresponds to laser pulse width τ probably, among other reasons, due to some uncertainty in τ determination, to thickness inhomogeneity of gold target or to not too small value of piezo-gauge time constant τ_0.

It is supposed that PA signal due to explosive boiling in the considered case can be inferred from comparison of the PA signals with almost equal amplitude and different widths at laser about 47 mJ/cm^2. This fluence is higher than simple theoretical estimations with supposition of temperatures equality at the liquid – metal boundary.

Further studies of the explosive boiling process are needed in particular using targets with small thermal expansion coefficient (e.g. Si) as well as smaller value of τ_0 in gauge circuit. Two subsequent short laser pulses can be used to diminish PA pressure signals which in the present conditions considerably exceed the explosive boiling pressure.

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