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ANALYSIS OF PHOTOACOUSTIC MONITORING OF LASER ABLATION IN THE CASE OF LASER PULSES WITH PERIODICALLY MODULATED INTENSITY

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Summary. Liquid - vapor phase transitions is investigated during laser irradiation of water in cases of free and covered surfaces. Intensity of laser pulse (duration ~200ns, wavelength λ=2,94μm) is periodically modulated with period 7ns due to mode beating. This modulation enables one to monitor the irradiated surface movement during laser action. It is shown that energy estimation give lower values for the surface displacement compared with the experimental ones and the effect of pressure recoil on the surface movement should be taken into account. Some peculiar features of the photoacoustic signals and the registered displacement behavior observed in the cases of free and covered surfaces are also discussed together with important details of signal processing.

1. INTRODUCTION

Photoacoustic (PA) effect is widely used in various applications as well as in fundamental research including investigation of nonequilibrium phase transitions during laser action on absorbing condensed substances and related topics (see e.g. [1-11] and references therein). For this purpose in [7,8] nanosecond (submicrosecond) laser pulses with periodically modulated intensity were used.

During action of such Erbium laser pulses with low intensity on absorbing liquids it was found that PA modulation amplitude evolution resembles that of the laser intensity. At higher intensities this evolution becomes strongly non-monotonous with one or two deep minima. This behavior was qualitatively explained as destructive interference effect from two PA mechanisms of pressure generation which are due to thermal expansion and vaporization processes [7].

The method used enables one to obtain information not only on pressure behavior but also on simultaneous displacement of absorbing surface during laser pulse action. However, the displacement signal behavior in the vicinity of PA modulation amplitude minima was not considered before. In this paper new results are presented on nonequilibrium phase transitions in the cases of free and closed irradiated surfaces.

2. EXPERIMENTAL METHOD

Experimental scheme was mainly the same as in [7-11]. Free or covered with transparent plate water surface was irradiated by erbium laser pulses (duration *300ns*, wavelength *2,94μm*, energy up to 10mJ) with periodically modulated intensity (modulation period was *7ns*) due to mode beating. Irradiation spot was varied in interval R_o=*0,5-0,8mm* with laser fluencies *0,2-1J/cm*^{2},

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and spatial energy distribution $\sim exp[-(2r/R_o)^2]$. Absorption length of the laser light in water is about one micron in accordance with absorption coefficient value $\alpha=10^4 cm^{-1}$.

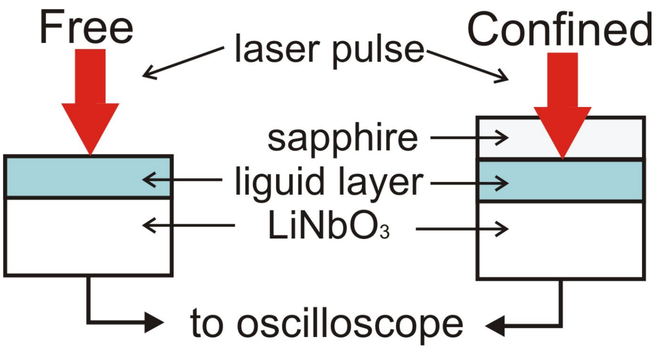

Fig1. Experimental scheme.

Pressure pulses generated in water layers were registered with piezoelectric gauge ShAPR-13M ($LiNbO_3$, diameter = *20mm*, thickness = *7mm*) like that used in [7-11] and oscilloscope Tektronix DPO7254 (Fig.1). Figures 2,3 show pressure pulse and its repetition with opposite sign due to reflections in the water layer from $LiNbO_3$ and irradiated water surfaces. Time interval between the reflections $t = 2h/c = 1500ns$ depends on the layer thickness $h\sim1mm$ and water sound speed $c = 1{,}45km/s$. In the case of covered surface when the liquid layer is confined between two solids with greater acoustic impedance the reflected signal, as it is well known, retains its sign (see, e. g. [9,10])

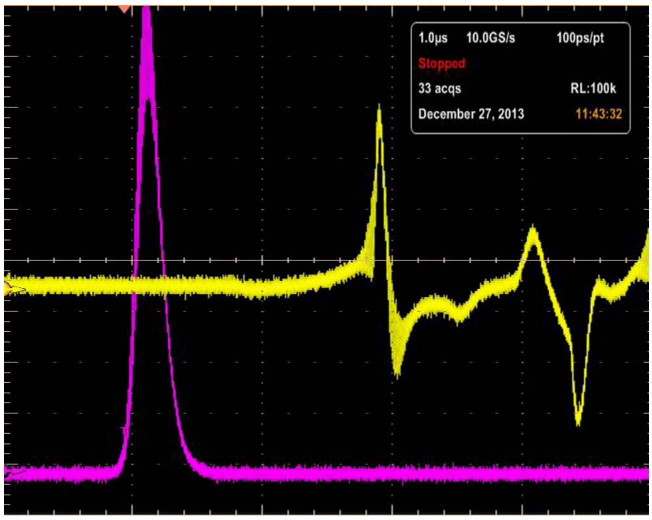

Fig.2 Laser pulse (pink curve) and acoustic signal with its first reflection in water layer (yellow curve).

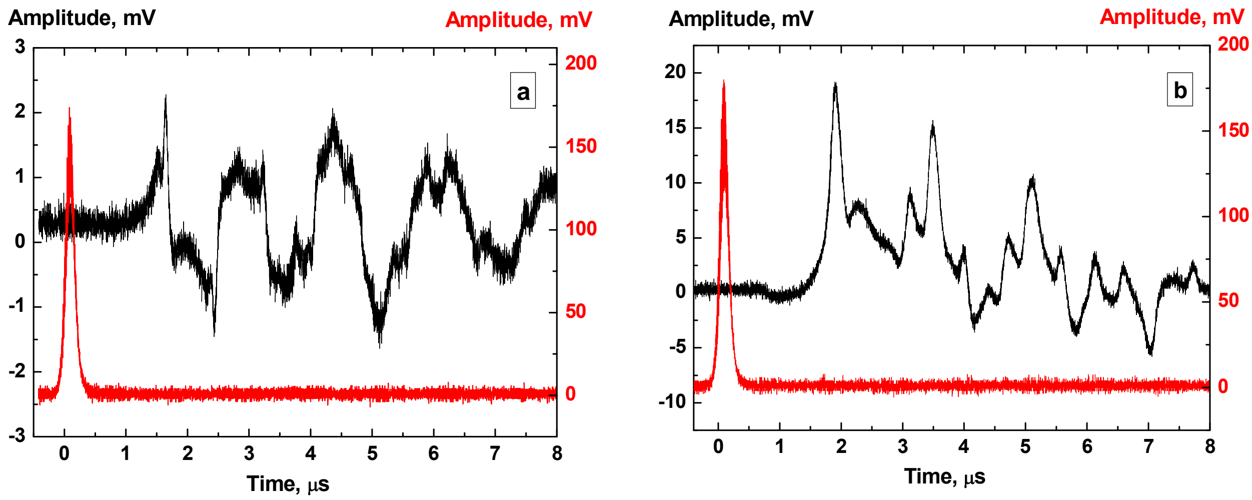

Fig.3 Laser pulse (red, unmodulated) and pressure signals (black) with multiply reflections in the case of free (**a**) and closed (**b**) irradiated water surfaces. Pressure signals change its sign after free surface reflection (a) and remain positive in the closed surface case (b).

Displacement $l=c\Delta t$ of irradiated surface or, more precisely, subsurface absorption zone is determined with the help of time delay Δt which manifests itself in phase (or modulation period) variations of modulated part of photoacoustic pressure signal due to the surface movement. To calculate variation of the acoustic signal modulation period, we used, as before [8, 10, 11], the expression for the time delay $\Delta t = t_n - n\Delta\tau$, which is the difference between the real-time position t_n of the n-th zero point of the signal modulation component and its extrapolated value $n\Delta\tau$ with some fixed half period $\Delta\tau$ determined, e.g., in the acoustic pulse beginning where the laser heating effect is small. Instead of $n\Delta\tau$, one can use the real positions of zero values in the laser pulse modulation component.

This procedure of time delay valuation is valid regardless the delay magnitude *e.g.* Δt may be grater than the time difference between two neighbor zeroes. However it is not the case if the modulation amplitude greatly diminishes as it was observed previously [7] and in the present paper (Fig 4-5). This situation is discussed in Appendix.

3. RESULTS AND DISCUSSION

Time behavior of photoacoustic signal (2), its slow (3) and modulated (4) parts together with time delay (1) are shown in Fig 4-5 at different values of laser fluency $E= 0{,}44\text{-}\ 0{,}95\ J/cm^2$ for the case of free water surface. At this regime one can distinguish between vaporization and thermoacoustical pressure signals (especially in Fig.4). The two signals in absorbing liquid were observed in many papers [2-11].

Negative time delay means that the distance between transducer and acoustical radiation source diminishes. Such behavior can be attributed to vaporization process including surface deepening due to vaporization pressure recoil. Simple energy estimation gives $l= E/L$, where L denotes heat of vaporization. At $E=0{,}44J/cm^2$, $0{,}95J/cm^2$ and $L{\sim}2{\cdot}10^3 J/cm^3$ one obtains for $l{\sim}2$ and 5 microns respectively. These values are somewhat lower then displacements 3,5 and 10 microns in figures 4 and 5 respectively, the difference being probably due to recoil pressure effect.

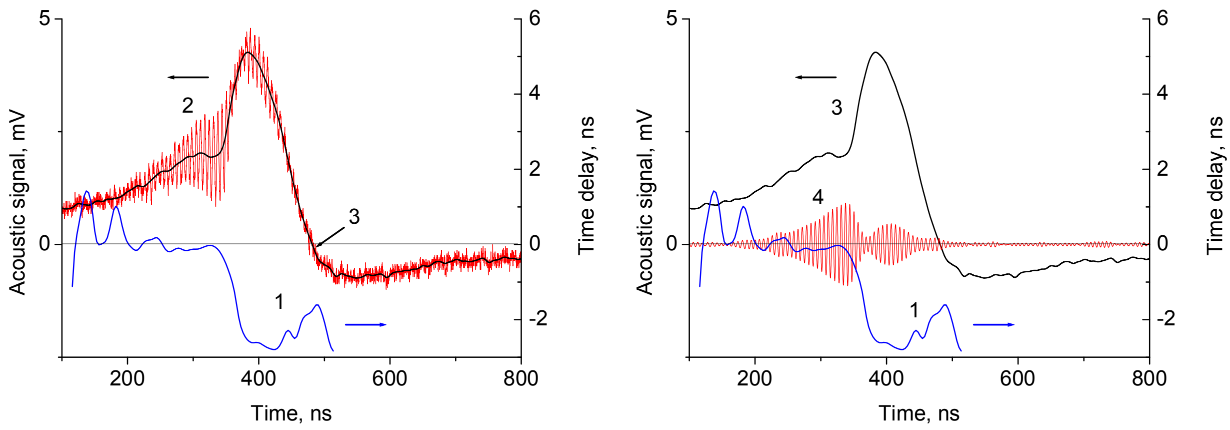

Fig 4 Time behavior of time delay (1), total photoacoustic signal (2), its slow (3) and modulated (4) parts at $E=0{,}44 J/cm^2$ and $R_0=0{,}76mm$

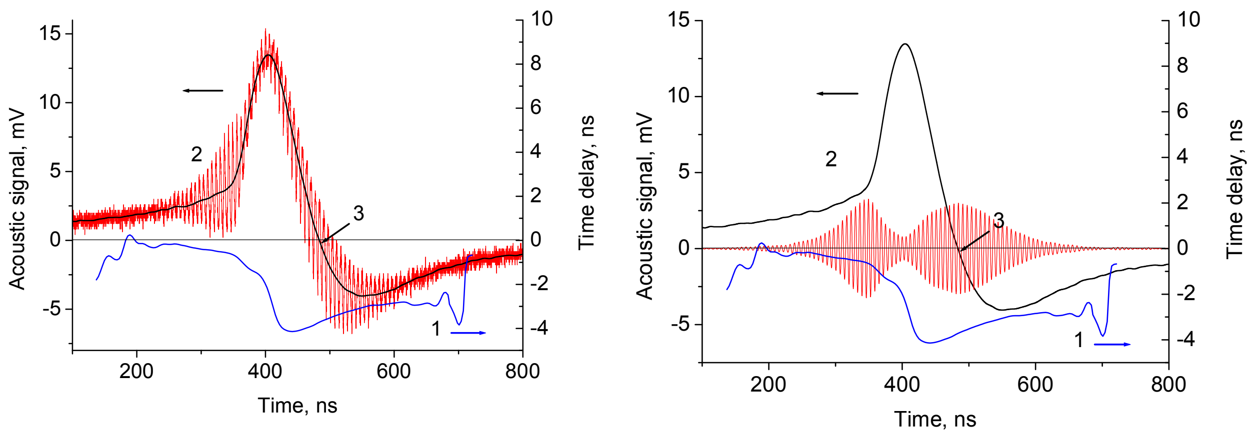

Fig.5 Time behavior of time delay (1), total photoacoustic signal (2), its slow (3) and modulated (4) parts at $E=0{,}95J/cm^2$, $R_0=0{,}76mm$

In the case of closed surface behavior of irradiated liquid demonstrates some not very expected aspects. First of all, despite the rigid upper boundary the thermoacoustical signal has bipolar form even at low intensity (Figs. 6, 7, while the reflected signals retain its sign in the same manner as in Fig. 3.

On the other hand the bipolar form is in accordance with observed negative time delay which means cavity formation between the liquid surface and the plate surface. The corresponding liquid surface displacements (cavity depth ~1,4 and 3 microns respectively) are in qualitative agreement with the estimations *l~E/L* provided pressure effect is taking into account.

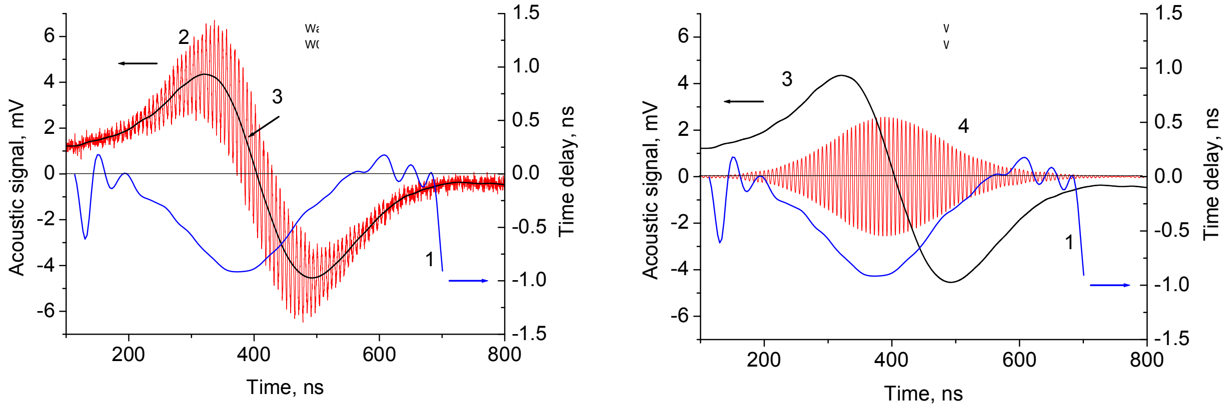

Fig.6 Time behavior of time delay (1), total photoacoustic signal (2), its slow (3) and modulated (4) parts at E=0,22J/cm2, R0=0,76mm

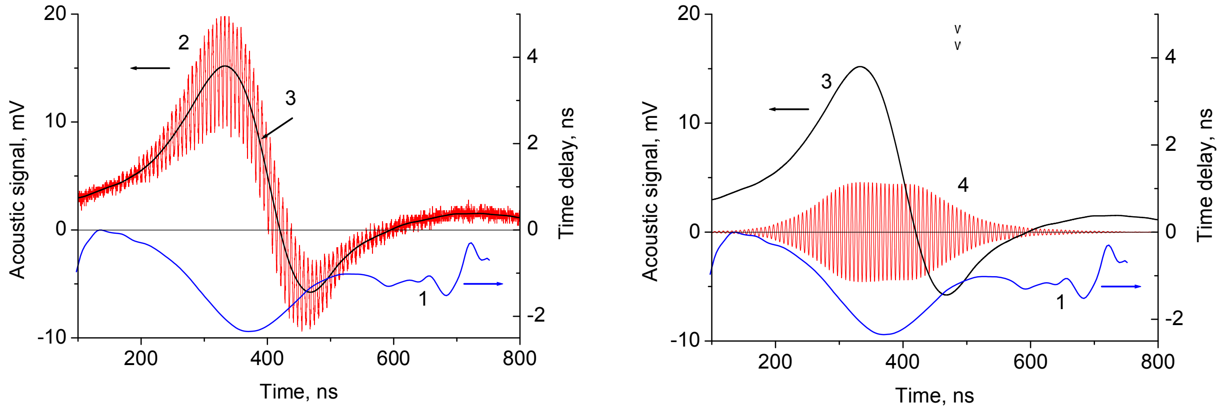

Fig.7 Time behavior of time delay (1), total photoacoustic signal (2), its slow (3) and modulated (4) parts at E=0,44J/cm2, R0=0,76mm

In figures 8 and 9 irradiation spot $R_0=0,52mm$ is smaller than in Figures 6 and 7 ($R_0=0,76mm$). Laser fluencies in figures 6 and 8 are approximately the same ($E\sim0,5J/cm^2$) and the overall behavior of acoustic signal and displacement are also similar. In particular, the negative and positive amplitude ratio of acoustic signal is lower then in figure 6.

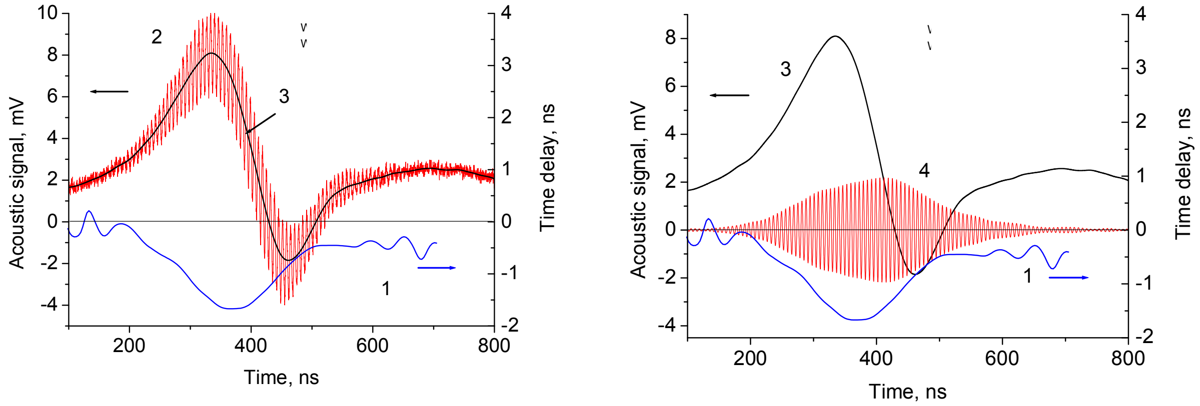

Fig.8 Time behavior of time delay (1), total photoacoustic signal (2), its slow (3) and modulated (4) parts at E=0,5J/cm^2, R_0=0,52mm

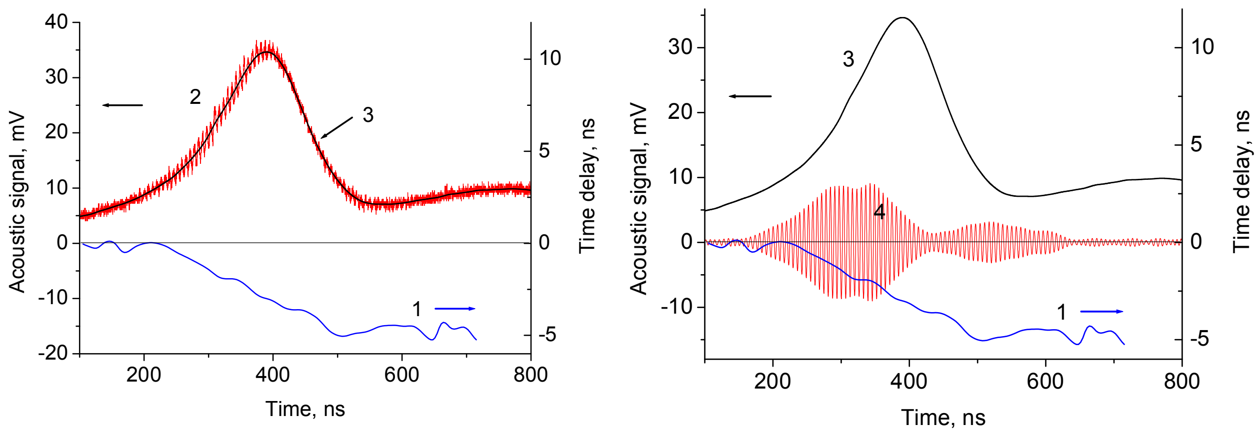

Fig.9 Time behavior of time delay (1), total photoacoustic signal (2), its slow (3) and modulated (4) parts at E=0,95J/cm^2, R_0=0,52mm

In figure 8 pressure signal is practically monopolar due to vaporization pressure as at is also in the case of free surface case at higher intensity. However, no distinct difference between thermoacoustic and vaporization pressure signal is observed in the closed surface case in contrast to free surface case.

Other particular feature of the closed surface case is fast diminishing of cavity depth at low intensity (Fig.5) which can be hardly explained in one dimensional picture of the process. This effect is probably due to the fact the cavity consist of many close packed micron sized bubbles which can quickly collapse after the end of laser pulse. It is interesting to note that from dispersion equation for capillary wave frequency $\omega^2=(\sigma/\rho)\cdot k^3$ where σ and ρ are surface tension and density of water one obtains for $T=2\pi/\omega\sim 100ns$ at wavelength $\lambda=2\pi/k\geq 1,5\mu m$.

4. CONCLUSIONS

The results obtained in the present paper demonstrate new possibilities which gives application of nanosecond laser pulses with periodically modulated intensity for investigation of liquid-vapor phase transition. In this method pressure generated in irradiated zone as well as movement of these zone are measured simultaneously with the same piezoelectric transducer. Comparison of measured surface displacement in the case of free and covered water surface irradiated with erbium laser pulses shows that simple energy considerations underestimate this displacement and the effect of pressure recoil should be taken into account. In the cases where modulation amplitude of PA signals approaches zero value during the laser pulse action a special care should be taken about possible jump appearance in Δt calculations. This problem can be avoided if laser pulse modulation is achieved with the help of series of short pulses. The method is also applicable for monitoring of laser metal ablation processes. To obtain additional information on the phase transition with the help of this method it is necessary to use constant laser intensity distribution across the radiation spot.

APPENDIX

During registration of time delay we observed in some cases unusual behavior of Δt which is shown in fig. A1 where the experimental conditions are practically the same as in fig.4.

However, evolution of time delay Δt in fig.A1 drastically differs from that in fig.4. These differences are due to significant modulation amplitude diminishing which together with experimental noise affect the procedure of Δt evaluation.

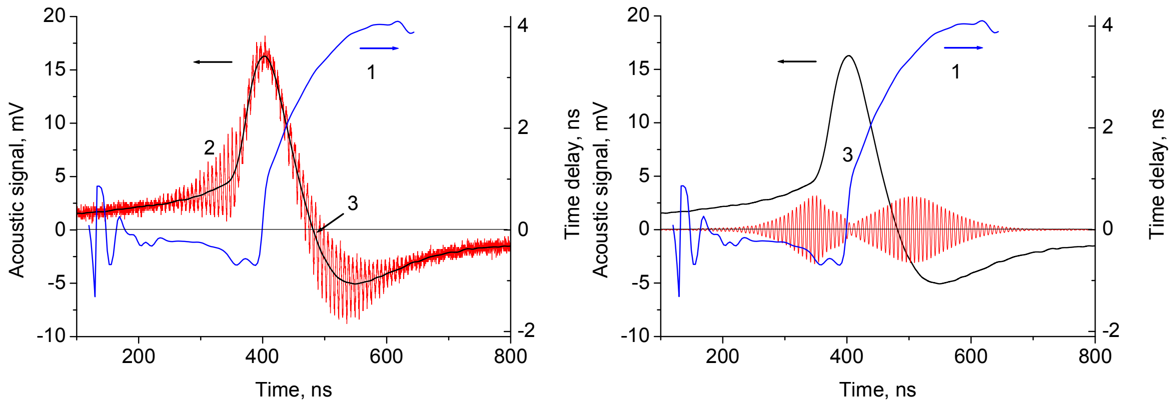

Fig. A1 Time behavior of time delay (1), total photoacoustic signal (2), its slow (3) and modulated (4) parts at $E=0{,}95 J/cm^2$, $R_0=0{,}76mm$. In comparison to fig.4 here more wide bandwidth is used in processing time delay signal.

To clarify this situation we considered some model cases where the effect of noise and modulation amplitude diminishing became more evident.

Fig. A2 shows model PA signal with modulated part and zero time delay Δt (horizontal line). Modulation period is 6,6 ns and modulation amplitude has no deep minimum during the pressure pulse. If such deep minimum occurs (fig.A3) then instead of straight line for Δt one obtains the line with a jump Δt_j=3,3ns (half modulation period). In the case of two such minima in the modulation amplitude behavior two negative jumps with the same amplitude appear in Δt time dependence (fig. A4).

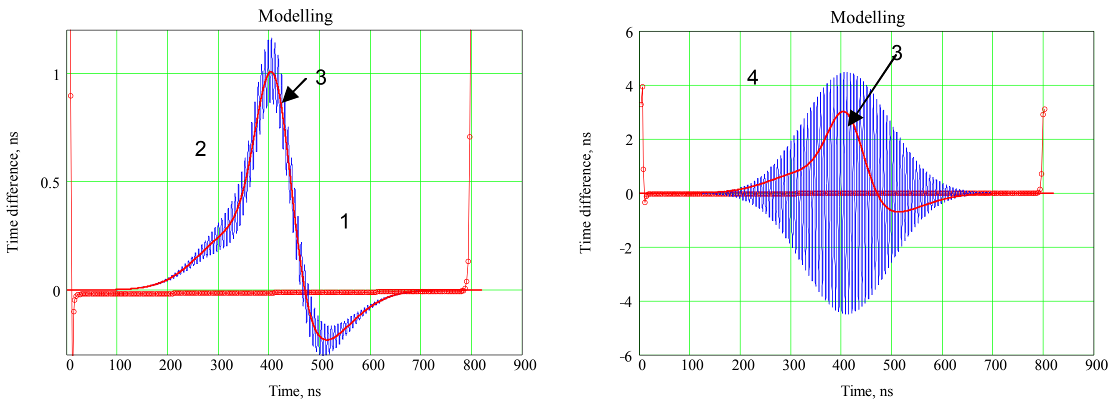

Fig. A2 Model calculation of time delay evolution of Δt (curve 1), slow part of PA signal (curve 3) and modulation part (curve 4). Curve 2 denotes the total PA signal. In this case Δt is constant.

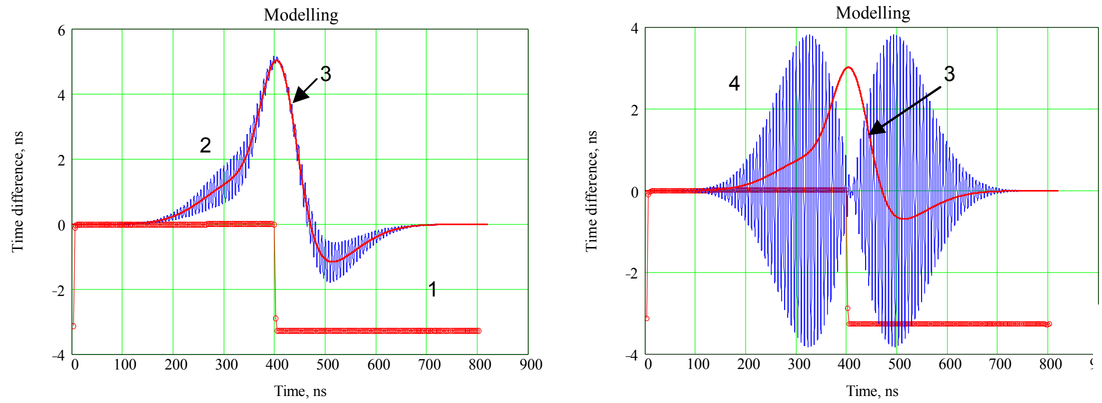

Fig. A3 Model calculation of time delay evolution of Δt (curve 1), slow part of PA signal (curve 3) and modulation part (curve 4). Curve 2 denotes the total PA signal. In this case Δt is piecewise constant function with negative jump corresponding to modulation amplitude zeroing.

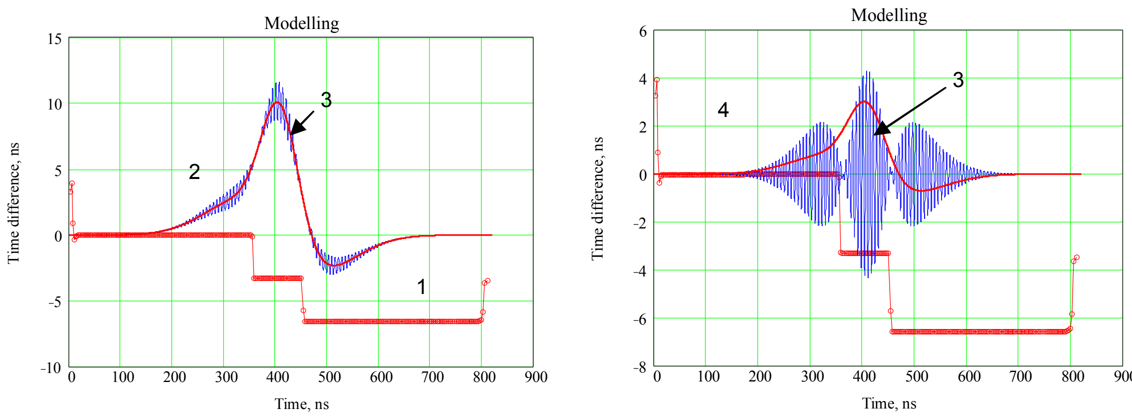

Fig. A4 Model calculation of time delay evolution of Δt (curve 1), slow part of PA signal (curve 3) and modulation part (curve 4). Curve 2 denotes the total PA signal. In this case Δt is piecewise constant function with two negative jumps corresponding to modulation amplitude zeroing.

Figures A5, A6 show the case when Δt initially is not constant and white noise component is added to modulation signal. The noise effect is visible from the inserted curves in fig.A5b which demonstrate Δt behavior with and without noise addition. As it is expected this effect becomes important in the beginning and in the end of PA signal where its amplitude significantly diminishes.

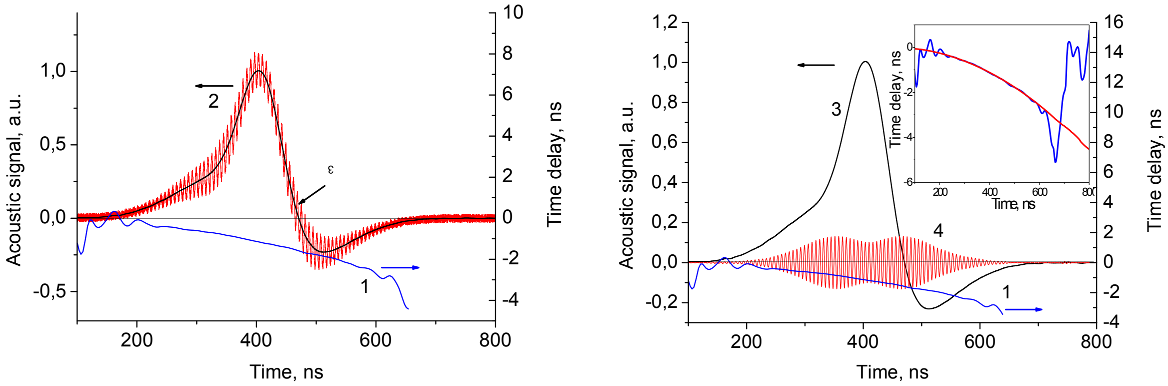

Fig. A5 Model calculation of time delay evolution (curve 1) for slow part of PA signal (curve 3) and modulation part (curve 4). Curve 2 denotes the total PA signal. Magnified view of curve 1 is shown in the inlet.

In fig. A6 it is shown Δt behavior when amplitude diminishes to zero values twice during the PA pulse. In these cases one can observe two jumps in Δt behavior as in fig.A4. The jump amplitude is equal to half of the modulation period. However, in fig. A6 jumps positive and negative sign is visible. The different jump signs are due to the noise effect and the bandwidth used in PA signal processing.

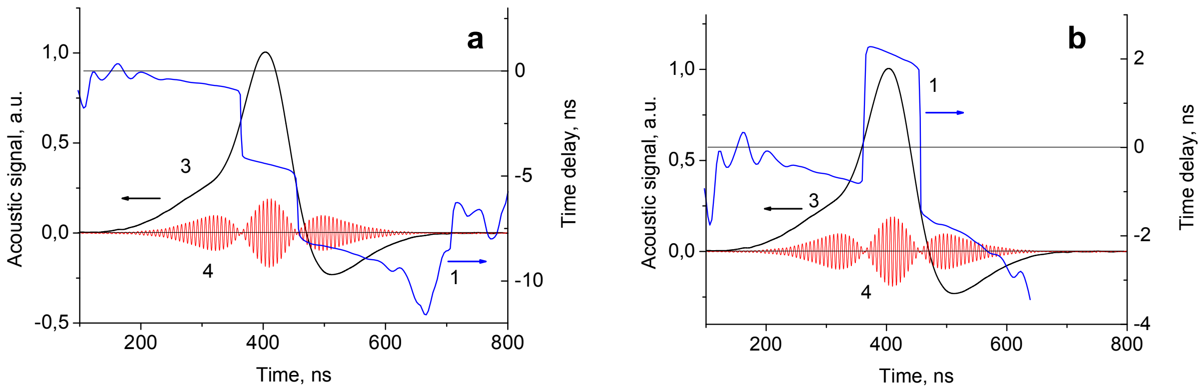

Fig. A6 Model calculation of time delay evolution (curve 1) for slow part of PA signal (curve 3) and modulation part(curve 4) with two amplitude zeros and different bandwidth. In (**a**) case bandwidth is smaller then in (**b**) case.

One should take into account these peculiarities of the signal processing in the cases when modulation amplitude goes to zero during the pulse. We hope that evolution of Δt in fig.5 is not affected with such jump.

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