# MODELLING OF NANOSECOND LASER ABLATION. CONTINUAL APPROACH

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**Summary.** Modeling of nanosecond laser ablation is considered in the framework of continual approach. Results of recoil pressure calculations are presented for the case of harmonically modulated laser intensity. It is shown that the modulated part of pressure response depends on relative values of thermo-acoustic and surface vaporization contributions. This dependence should be taken into account in acoustical monitoring of irradiated surface movement with the method where intensity modulated laser pulses are used. A brief review of some previous theoretical and experimental works on nanosecond laser ablation is also given.

## 1 INTRODUCTION

Theoretical and experimental investigations of laser ablation continue for more than a half a century (see e.g. [1-93] and references therein), though some problems of intense heating of condensed matter had been considered even before such investigation began [1-3]. For description of laser ablation processes various theoretical methods are used, e.g., molecular dynamics modeling [4-14], kinetic equations [15] or a continual approach, which includes heat conduction equation or full set of hydrodynamic equations with boundary conditions at phase transition fronts [16-18]. A brief review of some previous work and analysis of the continual approach for description of nanosecond laser ablation is given in the present paper (sec. 2) together with some new results on laser ablation modeling in the case of modulated laser intensity (sec. 3, 4). Concluding remarks are given in the final section 5.

## 2 LASER ABLATION MODELING: A BRIEF REVIEW

First-order phase transitions (melting and solidification) were considered in the framework of continual approach as early as in [19-21]. This so-called Stefan problem includes heat conduction equation for melted and solid states with Stefan boundary conditions formulated on the surface dividing the two different phases.

$$æ\frac{\partial T}{\partial z}\Big|_{z_2}^{z_1} = \rho L_m v \qquad (1)$$

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$$T_1 = T_2 \tag{2}$$

here ρ – density, $T$ – temperature, æ – thermal conductivity, $L_m$ – heat of fusion, $v$ – evaporation front velocity.

With some modification the Stefan approach was used much later to describe vaporization process induced by intense heating of a condensed matter [3,11,16-18,22-29]. It should be mentioned that the boundary conditions between vapor and liquid cannot be obtained in the framework of continual approach as well as generalization of (1), which takes into account non-equilibrium properties of the phase transition [30,31].

Boundary conditions used in the case of surface evaporation together with the set of hydrodynamical equations are described in section 3. In the simplified approach (the so-called "heat model" only energy (heat conduction) equation from the set is used [23,32,33]. If no phase transition is taken into account explicitly and there is no sharp phase boundaries then the ablation process can be described with the help of hydrodynamical equations without additional boundary conditions. Such an approach termed as "hydrodynamical model" is applicable for intense liquid heating when generated pressure exceeds critical pressure for liquid-vapor phase transition [34]. Phase transitions description with the help of "phase field model", which sometimes is also termed as "continual approach" [35-37], is not considered here.

In continual approach applications there are several problems, which include, in particular, a lack of information on metastable liquid properties, superheating limit temperature value, and superheated liquid behavior (explosive boiling) after this limit is attained. For example, a question about critical and metal-dielectric (MD) transition temperatures, first posed in ref. [38], and discussed later in many papers [38-46], cannot yet be considered as sufficiently resolved theoretically and experimentally. It is useful to mention that in ref. [39] (footnote [15)]), as well as in [40,45], relation between MD transition and explosive boiling is discussed. In strongly-absorbing dielectric liquids, absorption coefficient can also significantly be changed during laser pulse action [47-60].

Explosive boiling is discussed in many papers [12,13,50,61-71] because it occurs in various processes of practical interest. Importance of explosive boiling investigations is determined also by the fact that critical parameters for many materials including metals are poorly known as it is seen e.g. from [12,13,69-71].

Applicability limit of surface vaporization model depends strongly on absorption coefficient value, which is large for metals and considerably smaller for dielectrics. For this reason, in dielectrics the subsurface temperature maximum $T_M$ can exceed significantly the irradiated surface temperature $T_1$ while for the metals the difference between $T_M$ and $T_1$ is rather small [24,40,62,72]. It means, that surface vaporization model for metals permits to consider relatively higher values of $T_1$, which are closer to $T_M$, than it is for dielectrics. It should be reminded also that, even at temperatures, which are somewhat lower than $T_1$, the subsurface liquid layer is superheated because the surface vaporization pressure is lower, than saturation pressure $P_S(T_1)$.

Radiation penetration length $l = \alpha^{-1}$ and surface evaporation cooling effects on temperature distribution in irradiated samples are known since long ago (see e.g. [24], and ref. therein). However, influence of temperature distribution form on explosion boiling process in some earlier papers was not considered in sufficient details and some previous conclusions need to be corrected. For example, in ref. [24] the explosive boiling process is described as follows:

"If the temperature reached is great enough to cause vaporization of the material at depths below the surface, tremendous pressures arising from the vaporized materials will develop that would explosively remove the intervening material. Material removal based upon such an explosive technique would be considerably more efficient than conventional means of laser drilling that depend upon the vaporization of all material removed."

In the same paper the ref. [23] is mentioned with such quotation from it:

"Material at some depth below the surface reaches its vaporization temperature before the material at the surface has absorbed its latent heat of vaporization. This leads to a pulse of high pressure and subsequent superheating of the underlying material until the temperature rises above the critical point. There is then no longer any distinction between the superheated solid and a highly condensed gas. The emission of the vaporized material, which is delayed relative to the peak of the temperature pulse at the surface, then proceeds like a thermal explosion."

It is clear from these quotations that more detailed information is necessary for adequate description of pressure behavior during explosive boiling which should include, in particular, quantitative estimations of pressure magnitudes and its evolution during explosive boiling. Because of different values $T_M$-$T_1$ for metals and dielectrics pressure jump for later is greater when vaporization regime changes from surface mechanism to explosive (volume) boiling.

Despite the relatively smaller pressure jump for metals in ref. [40] it was supposed that some features of these explosive boiling can be used for experimental determination of critical pressure values during laser ablation. Somewhat different description of explosive boiling in irradiated metals was given later in ref. [25] where additional subsurface superheating due to small difference $T_M$-$T_1$ was considered irrelevant:

"The basic question has thus been answered. The subsurface temperature maximum is found to be a very minor effect provided it is calculated correctly. The mechanism based on explosion due to subsurface superheating, which was discussed above in (d) is therefore wrong. By contrast, explosive boiling, as discussed in (c), remains the only physically sound thermal mechanism able to explain laser sputtering at high fluences."

Analogous conclusion is repeated after twenty years in ref. [73]:

"This indicates that subsurface superheating is impossible in the present case. Our result is consistent with the observation that subsurface heating in metal targets is negligibly small compared with nonmetallic targets.[4,7]" (This two citations [4,7] correspond to ref. [25, 62])

However recent publications [7-11] of MD calculations results for metals show importance of the subsurface superheating for development of explosive boiling process where pressure value increases by factor of about 1.5 in sub-nanosecond time interval when vaporization regime changes from surface one to explosive (volume) boiling.

In dielectrics the jump ratio between possible maximum of explosive boiling pressure and surface vaporization pressure just before the explosion begins should be greater than 1.5 because the difference $T_M$-$T_1$ is greater than it is in metals. Simplified theoretical consideration was presented in [17] for repeating explosive boiling pressure during nanosecond laser irradiation of ablation liquid.

Up to our knowledge, no such sub-nanosecond pressure jumps during nanosecond laser ablation has been detected yet probably because of averaging effect due to laser intensity variation across the irradiation spot and lack of time resolution in pressure measurement. In ref [63,74-88], *e.g.* there is no experimental information on such short pressure peaks during

nanosecond laser-ablation explosive boiling of absorbing liquids or transparent liquids on absorbing solid targets heated with nanosecond laser pulses.

Pressure perturbations in air due to periodic explosive boiling of absorbing liquids heated with $CO_2$ were detected in ref [22,89] on millisecond time scale. Origin of sub-nanosecond pressure peaks observed in [26] was not investigated sufficiently and needs further clarifications.

In the experiment [90,91] approximately twofold shock wave pressure rise above the irradiated Al target was detected and interpreted as transition from normal vaporization to phase explosion (explosive boiling). As it was already mentioned [10], however, in this case one should also take into account the effect of the plasma formation on pressure behavior in laser ablation plume.

Up to now amplitude and duration of explosive boiling pressure peaks are calculated only in ref. [6] while in theoretical considerations [10] of explosive boiling process (induced by nanosecond laser pulses) no information is given on time resolved pressure behavior during the explosion.

From this brief review it follows that additional theoretical and experimental investigations are necessary for obtaining new time-resolved information on explosive boiling processes during nanosecond laser ablation.

## 3 STATEMENT OF THE PROBLEM

In refs [8,93] it was shown how the acoustical pressure response in absorbing dielectric liquids irradiated with harmonically modulated nanosecond laser pulses can be used to determine the irradiated surface displacement. In this case the pressure response is mainly due to thermoacoustical effect while the vaporization pressure effect is small. For metals the situation is different because both thermoacoustical and surface vaporization effects can be relevant in generation of modulated pressure response to nanosecond laser pulses with harmonically modulated intensity.

We considered here 1D continual model of pressure generation in liquid metal (Hg) under the action of nanosecond laser pulses with harmonically modulated intensity $I = I_0(t)\cdot(1 + h \sin(\omega t))$, where $I_0(t)$ is smooth component of absorbed laser intensity, $h$ – modulation depth, $\omega$ – modulation frequency.

$$\frac{\partial \rho}{\partial t} + u\frac{\partial \rho}{\partial z} + \rho\frac{\partial u}{\partial z} = 0 \tag{3}$$

$$\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial z} + \frac{1}{\rho}\frac{\partial P}{\partial z} = 0 \tag{4}$$

$$\rho C_P\left(\frac{\partial T}{\partial t} + u\frac{\partial T}{\partial z}\right) = \frac{\partial}{\partial z}\left(æ\frac{\partial T}{\partial z}\right) + Q \tag{5}$$

$$Q = I(t)\,\alpha\, exp\big(-\alpha\,(z - z_1)\big) \tag{6}$$

$$I(t) = I_0(t)\,\big(1 + h\,cos(\omega t)\big) \tag{7}$$

$$I_0(t) = \begin{cases} I_m \sin(1.14\, t/\Delta t)^4, & 0 \le t \le \Delta t\ \ \pi/1.14 \\ 0, & otherwise \end{cases} \tag{8}$$

here ρ – density, $u$ – velocity, $P$ – pressure, $T$ – temperature, $C_P$ – heat capacity, æ – thermal conductivity, $Q$ – power density, α – absorption coefficient, $z_1$ – irradiated front position ($z_1(t = 0) = 0$), $\Delta t$ – FWHM of laser pulse. Negative and positive displacements are due to heat expansion and surface vaporization mechanisms. Additional relation $F(\rho,P,T) = 0$, called equation of state, is necessary to solve the system.

Vaporization boundary conditions at $z = z_1$ can be written as follows [30,31]

$$æ\left(\frac{\partial T}{\partial z}\right)_{z_1} = \rho L v \tag{9}$$

$$v = 0{,}82\sqrt{\frac{m}{2\pi kT}}\frac{1}{\rho}P_1(T_1) \tag{10}$$

$$P_1 = 0{,}56\, P_B\, exp\left(A\left(1 - \frac{T_B}{T_1}\right)\right) \tag{11}$$

$$z_1 = \int_0^t (v + u_1)dt \tag{12}$$

From (1-5) in linear approximation it follows the well-known equation (see e.g. [22]).

$$\frac{1}{c^2}\frac{\partial^2 P}{\partial t^2} - \frac{\partial^2 P}{\partial z^2} = \frac{\partial \rho}{\partial T}\frac{\partial^2 T}{\partial t^2} \tag{13}$$

If the heating process is slow enough and pressure dependence of ρ can be neglected than one obtains from equation (13) the expression for thermoacoustic pressure response, which is proportional heat expansion coefficient β

$$P(t) = P_1 + \frac{\beta}{C_P}\left(æ\frac{\partial T_1}{\partial t} + \frac{1}{\alpha}\frac{\partial I}{\partial t}\right) = P_1 + P_{ta}\,, \qquad \beta = -\frac{1}{\rho}\frac{\partial \rho}{\partial T} \tag{14}$$

This expression denote the pressure value at the distance $z^*$ which is lager than the effective heating length ($1/\alpha$ or $\sqrt{(\chi \cdot t^*)}$, $\chi = æ/\rho C_P$) and smaller than effective acoustic wavelength $c_s \cdot t^*$, where $t^*$ denotes characteristic time of laser intensity variation. This approximation corresponds to that used in [27].

Some nonlinear effects can also be taken into account in the framework in such slow approximation approach. In this case first continuum (3) and heat conduction (5) equations are solved while the pressure is determined than from the Euler equation (4). Such approximation is used e.g. [28, 29] and the present paper. For simplicity α, χ, $C_P$ are supposed here to be constant.

In numerical calculation procedure were used standard finite difference schemes (forward difference, midpoint method, 2nd order central difference). Numerical results are in agreement with available analytical formulas for linear thermoacoustical pressure response and the steady-state vaporization regime.

## 4 RESULTS AND DISCUSSION

Fig. 1 show pressure behavior and irradiated surface displacement at two different laser fluences $E$ = 20 (a) and 200 (b) mJ/cm$^2$ with no intensity modulation. At lower intensity (a) the thermoacoustic pressure (3,a) contribution is comparable to the total signal (2,a) which at later times after the maximum laser intensity (1,a) is determined mainly by the vaporization mechanisms. The irradiated surface displacement (4,a) is only about fifty nanometers and negative due to heat expansion effect. At higher intensity the displacement (4,b) is more significant (≈450nm) and positive because the vaporization effect is superior except for the initial part of the laser pulse where $\Delta z$ is negative. The total pressure behavior (2,b) is also mainly due to vaporization mechanism because thermoacoustic contribution (3,b) is rather small.

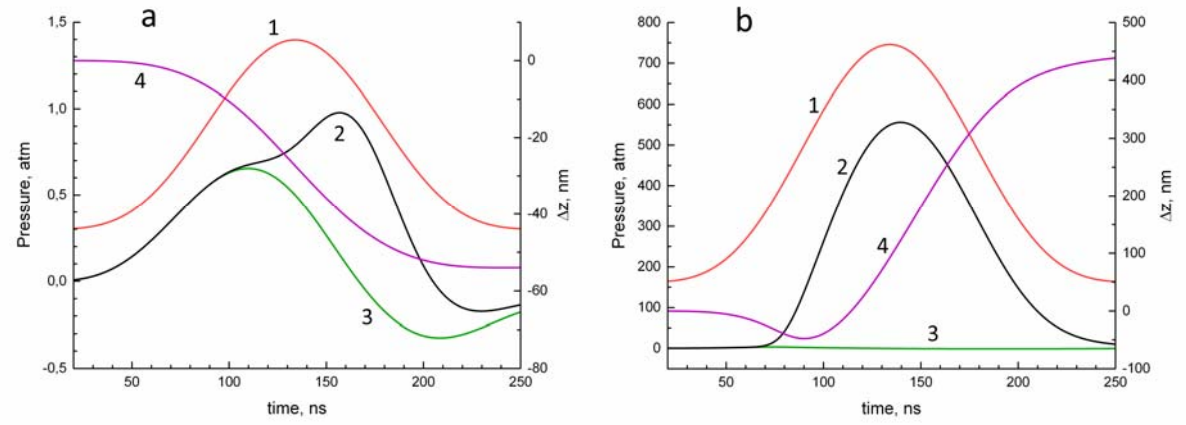


Figure 1: Time dependence of thermoacoustic pressure part (3), total (including vaporization) pressure (2) and irradiated surface displacement $\Delta z$ (4) during laser pulse (1) action at different fluences $E$ = 20 (a) and 200 (b) mJ/cm$^2$.

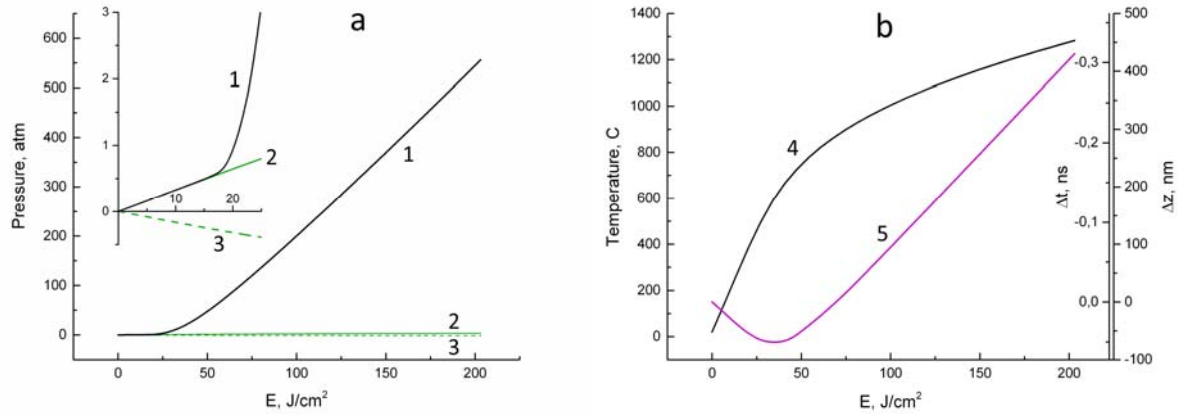


Figure 2: Maximum values of the total pressure (1), its thermoacoustic component (positive 2 and negative 3), surface temperature (4), and surface displacement value $\Delta z$ at $t$ = 200 ns (5) versus laser fluences.

Values of pressure maxima determined by thermoacoustic and surface vaporization

mechanisms at different laser fluences are shown on fig. 2a. It is evident relevant from the insert inlet in fig. 2a that thermoacoustic contribution (positive 2 and negative 3) is relevant only for small $E \leq 20$ mJ/cm$^2$. Dependence of maximum temperature (curve 4) and displacement value at $t = 200$ ns (curve 5) on laser fluence $E$ are shown in fig. 2b. While vaporization and thermoacoustic pressure contributions becomes equal at $E = 20$ mJ/cm$^2$ the irradiated surface displacement becomes positive only at $E = 80$ mJ/cm$^2$. It should be mentioned also that the surface temperature maximum (curve 4) increases more slowly at higher fluences because the vaporization process approaches to the (quasi) steady-state regime.

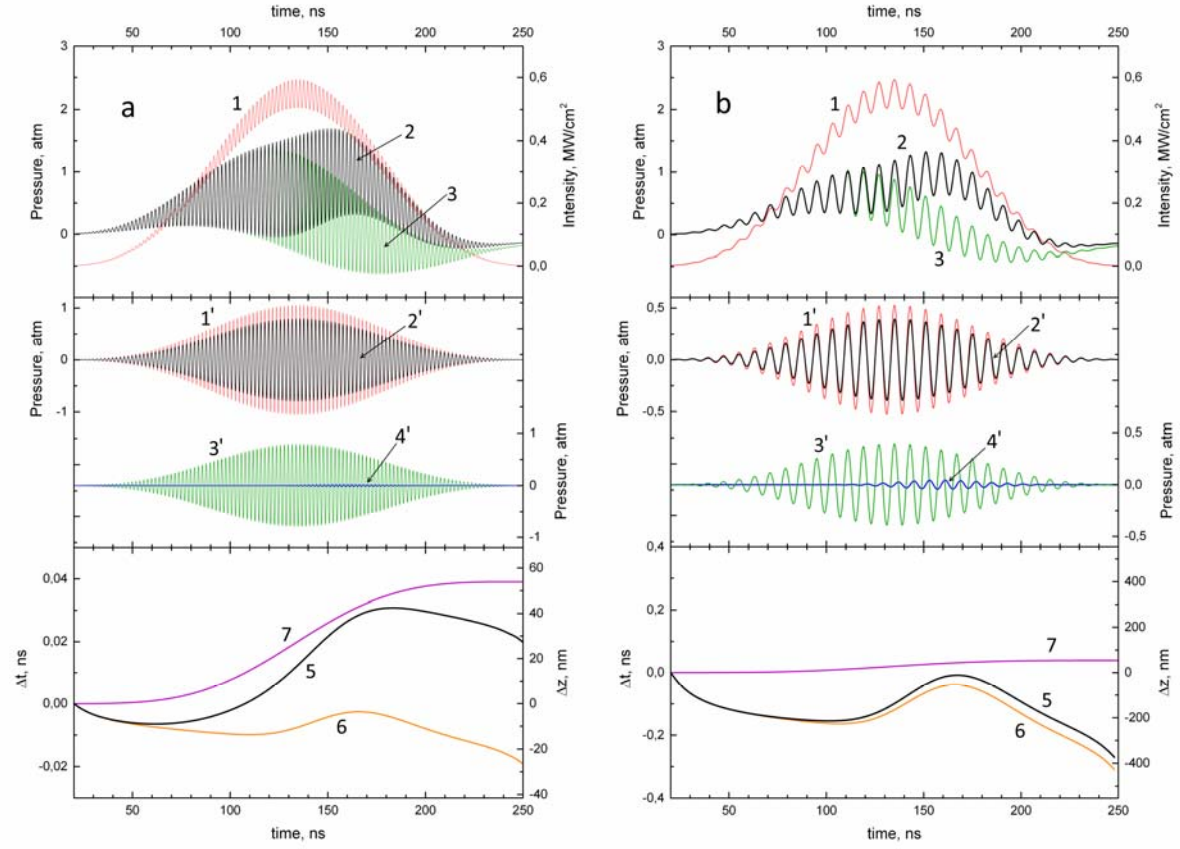


Figure 3: Response of ablation process to modulated laser intensity (1) at $E = 20$ mJ/cm$^2$ with different modulation period $\tau = 2$ ns (a) and 8 ns (b): total pressure signal (2), thermoacoustic component (3); modulated intensity component (1′), modulated total pressure component (2′), modulated thermoacoustic component (3′), modulated vaporization component (4′); time delays in laboratory (5) and moving (6) frames, the irradiated front displacement (7), $\Delta z = -c_s \cdot \Delta t$.

Fig. 3-5 demonstrates response of ablation regime to the modulated laser pulses (curve 1) at different fluences $E = 20$ (fig.3), 80 (fig.4), 200 mJ/cm$^2$ (fig.5) and different modulation periods $\tau = 2$ ns (fig. 3a, 4a, 5a), 8 ns (fig. 3b, 4b, 5b). Total pressure response (curve 2) and its modulated parts contain (curve 2′) in general case contributions from thermoacoustic (curves 3,3′) and surface vaporization (curve 4′) mechanisms. Relative value of these contributions changes depending on $\tau$ and $E$. Vaporization pressure contribution is small at

small $E$ and small $\tau$ while thermoacoustic modulated pressure grows at small $\tau$. They are comparable in amplitude in the case of fig. 4a. In fig. 3a,b modulated part thermoacoustic pressure (3′) is lower than vaporization part (4′), while in fig. 4b,5a,b the relation is inversed.

Evolution in time of the two contributions to the modulated pressure response results in effective changes of the time interval $\Delta t_n$ which contains $n$ modulation periods of the response in comparison with the interval $n \cdot \tau$, where $\tau$ is the modulation period of the initial parts of the pulse.

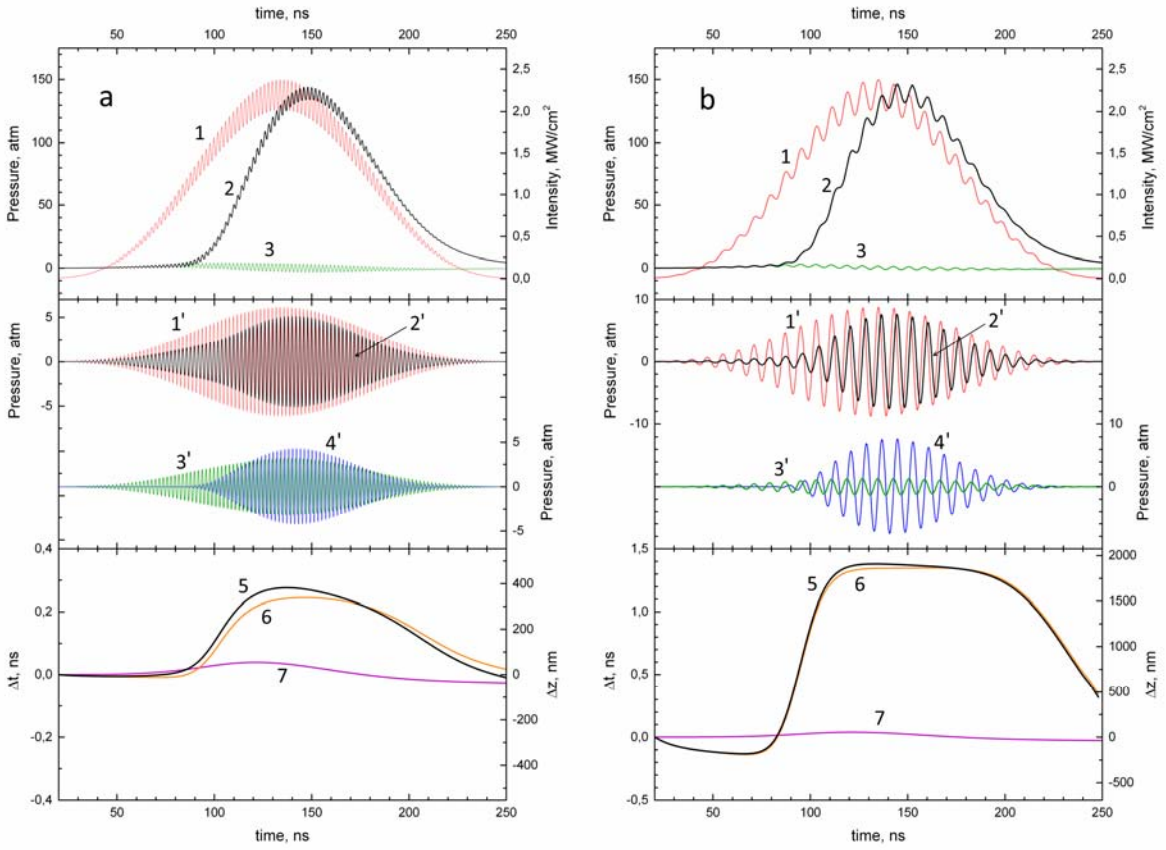


Figure 4: Response of ablation process to modulated laser intensity (1) at $E$ = 80 mJ/cm$^2$ with different modulation period $\tau$ = 2 ns (a) and 8 ns (b): total pressure signal (2), thermoacoustic component (3); modulated intensity component (1′), modulated total pressure component (2′), modulated thermoacoustic component (3′), modulated vaporization component (4′); time delays in laboratory (5) and moving (6) frames, the irradiated front displacement (7), $\Delta z = -c_s \cdot \Delta t$.

Time dependence of $\Delta t_n = t_n - n \cdot \tau$ for modulated pressure response in the laboratory reference frame is given in figs 3-5 by the continuous (instead of discrete points sets) curves (6). The same procedure for modulated part of laser intensity gives $\Delta t \approx 0$ with deviations smaller than $10^{-5}$ ns when the position of zeros are used in the calculation. Maximum value of the curves (6) is about 1.3 ns for $E$ = 200 mJ/cm$^2$ and $\tau$ = 8 ns (fig. 5b) while at $\tau$ = 2 ns the maximum is 0.3 ns. Fig. 6 shows how thermoacoustic (a) and vaporization (b) pressure maximum amplitudes for non-modulated (curve 1) and modulated (curves 2, 3) parts depend on laser fluence. Curve 1b on fig. 6b corresponds to curve 1 on fig. 2a for P > 10 atm.

Modulation amplitude for vaporization pressure at $\tau$ = 2 ns is lower than at $\tau$ = 8 ns which means that the period $\tau$ = 2 ns is shorter than the period $\tau_m$ corresponding to the modulation amplitude maximum [22]. Relative values of thermoacoustic amplitude (fig. 6a) can be understood from the linear approximation (formula 14) which contains time derivative of the surface temperature.

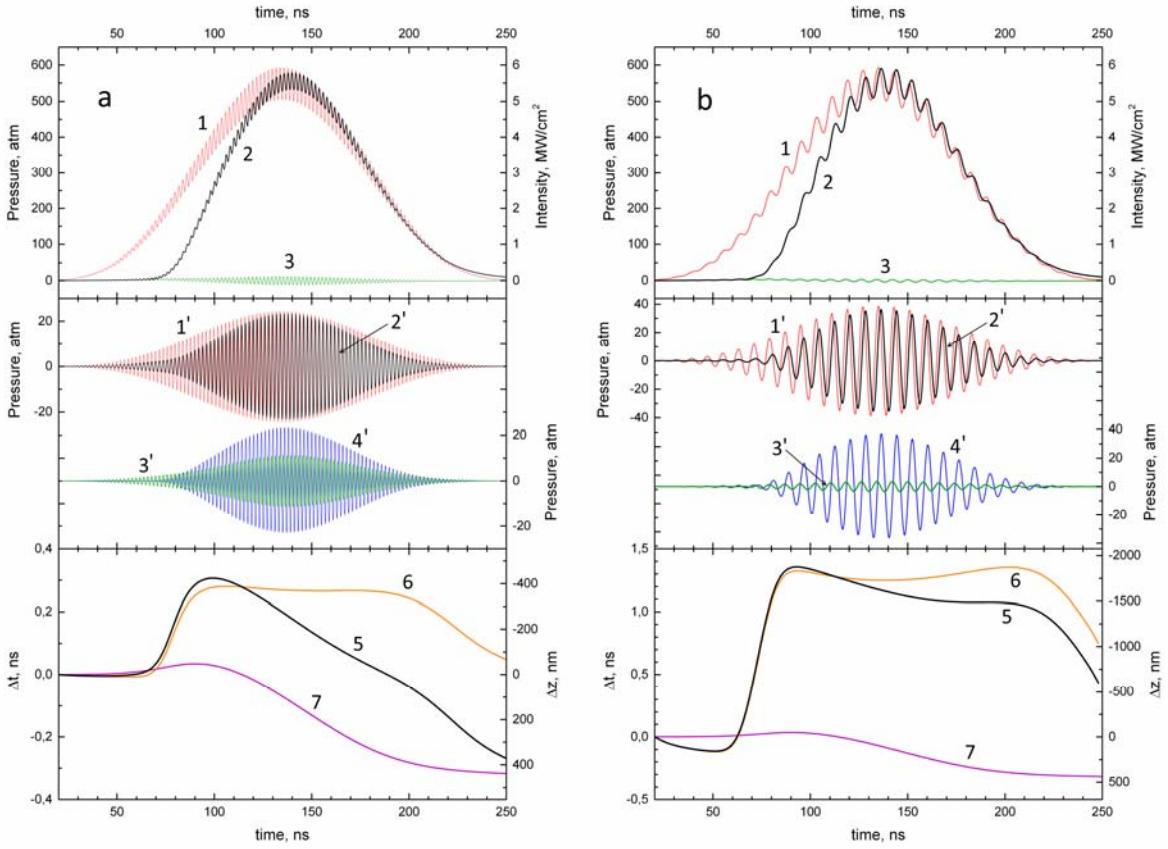


Figure 5: Response of ablation process to modulated laser intensity (1) at $E$ = 200 mJ/cm$^2$ with different modulation period $\tau$ = 2 ns (a) and 8 ns (b): total pressure signal (2), thermoacoustic component (3); modulated intensity component (1′), modulated total pressure component (2′), modulated thermoacoustic component (3′), modulated vaporization component (4′); time delays in laboratory (5) and moving (6) frames, the irradiated front displacement (7), $\Delta z = -c_s \cdot \Delta t$.

It is clear that irradiated surface movement gives rise to changes of modulation period and $\Delta t$ detected in laboratory reference frame as compared with these values determined in reference frame moving with the surface. This form of Doppler effect was used in ref. [8] for monitoring of irradiated surface movement assuming that $\Delta t^{int}$ is small.

Applicability limits of such approximations depend on variation of relative values of different pressure generation mechanisms contributions and nonlinear effects. From fig. 3-5 it follows that effective time delay (curve 6) in moving reference frame can be comparable or even greater than the corresponding value due to irradiated surface movement (curve 7).

In experiments [26] it was observed that modulation amplitude of pressure response in the case of water irradiated with modulated nanosecond erbium laser pulses demonstrates

strongly non-monotonous behavior which was probably due to interference effect between thermoacoustic and vaporization pressure contributions. Our calculations show, however, that modulation amplitude due to surface vaporization mechanism is small compared with thermoacoustic contribution at the applicability limit of the surface evaporation model. This ablation regime needs further experimental and theoretical investigations.

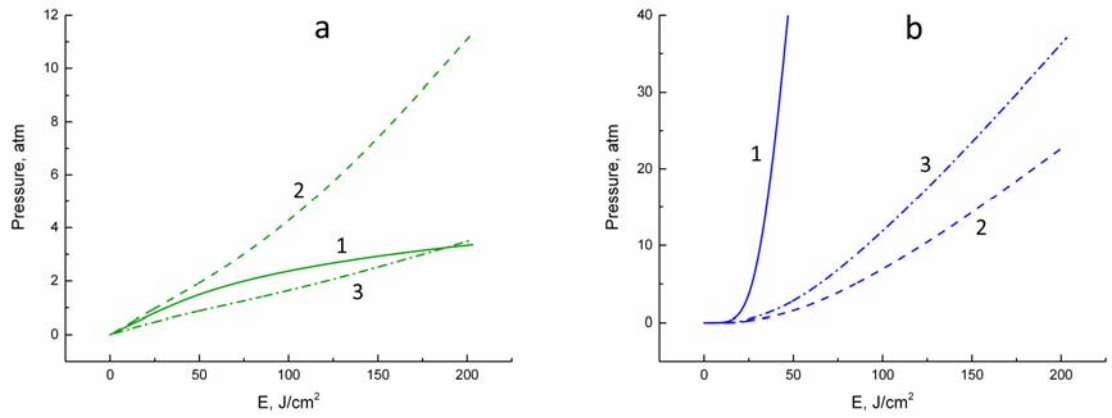


Figure 6: Maximum values of the pressure component: smooth (1) and modulated (2, 3), thermoacoustic (a) and vaporisation (b) versus laser fluence *E*.

## 5 CONCLUSIONS

From the calculated results and discussion presented above it follows that time depended evolution of modulated recoil pressure part generated in the target depends not only on irradiated surface movement but also on effects of non-linearity in pressure generation mechanisms and interplay between thermoacoustic and surface vaporization contributions. In general case these additionally effects are not small compared to that due to surface movement and should be correctly taken into account in the procedure of surface movement determination based on comparison of modulated recoil pressure response with modulated part of laser pulse.

Our consideration also permits to suggest that instead of harmonic modulation of laser intensity it is probably more convenient to use intensity modulation in the form of short peaks repetition e.g. due to mode-locking. Recoil pressure calculation in such regime will be presented elsewhere.

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