Modelling of Pulsed Laser Ablation of a Solid Target in a Vacuum in the Explosive Boiling Mode

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Abstract. The model describing the heat and mass transfer processes at transition from the normal vaporization mode to the mode of explosive boiling of a near-surface layer of a solid material with a great absorption factor under the pulsed laser influence in the nanosecond range on a target in a vacuum is proposed. The model includes the description of processes of laser radiation absorption, material heating and evaporation, a lift-off of a liquid film at explosive boiling and vapor expansion. The calculation results of ablation of a flat silicon surface are presented.

Keywords: Rarefied Plumes, Monte Carlo method, Pulsed Laser Ablation, Explosive Boiling.
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INTRODUCTION

Depending on duration and intensity of laser influence on solid materials with a great absorption factor (metals, semiconductors) the different modes of pulsed laser ablation (PLA) are observed [1]. In case of nanosecond duration of laser pulses the increase of laser fluence leads to sudden transition from the normal vaporization mode to the explosive boiling one with the lift-off of a vapor-liquid mass from the irradiated target surface [2, 3]. The model of explosive boiling under pulsed laser influence on solid materials was proposed in paper [3]. This model takes into account that at ablation of condensed substances with significant absorption of radiation the temperature maximum T_m of an evaporating material is at some depth of a target and not at its surface. The model supposes that on reaching the condition $T_m=T_{th}$ (T_{th} is the threshold temperature) during the heating process the thermodynamic instability of a near-surface layer and its lift-off from the target take place. The model [3] describes processes of volume absorption of radiation, heating and evaporation of material as well as (approximately) the lift-offed film movement but it doesn't take into account the gasdynamic processes in the regions filled with vapor. In our model the gasdynamic processes are included in the general model. The features of a vapor-liquid mass outflow into a vacuum under condition of strong absorption of radiation by the lift-offed liquid film are considered.

MODELS AND SIMULATION METHODS

The model describes two successive stages of PLA of a flat target: (1) the stage corresponding to the mode of normal vaporization of a material from a target surface (Figure 1a); (2) the stage corresponding to the mode of explosive boiling of a near-surface layer with its lift-off from a target (Figure 1b). The end of the first stage corresponds to the moment when the condition $T_m=T_{th}$ ($T_{th}=0.9T_c$, T_c is the critical temperature) first takes place. The second stage starts from the instantaneous formation in the section $T=T_m$ the vapor gap with a thickness $H=10^{-9}$m and the vapor parameters in equilibrium at $T_1=(T_{th}+T_w(t_{1K}))/2$ in this gap, where t_{1K} denotes the end of the first stage. The lift-offed film temperature is assumed to be constant and equal to T_1.

Within the framework of our description PLA includes three groups of spatially divided (but interconnected) processes occurring in the volume of a solid body (laser radiation absorption and heating), at the surface (evaporation and condensation) and in the regions filled with vapor (vapor motion). Accordingly the general mathematical model of PLA includes the heat models of a target and a lift-offed layer and the gasdynamic model of a vapor motion. The boundary conditions at the surfaces are coordinated for the heat and gasdynamic models.

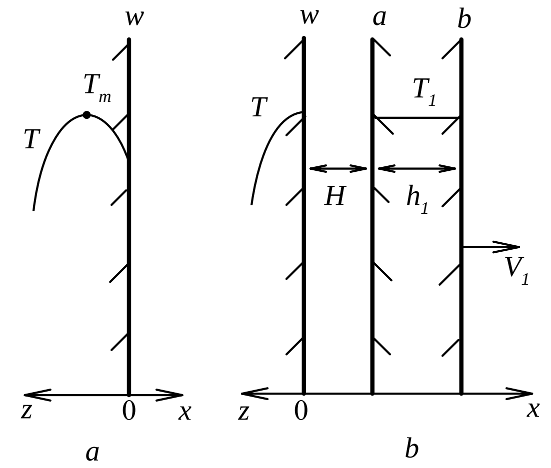


FIGURE 1. The scheme of transition from the normal vaporization mode (a) to the explosive boiling one (b).

The heat model of a target

For the description of radiation absorption by the target material the Lambert-Beer law is used

$$I(z,t) = I_0(t)(1-R)\exp(-\alpha z). \quad (1)$$

Here $I_0(t)$, $I(z, t)$ are the radiation intensities at the surface and inside the body at a distance z from the surface at time t correspondingly, R denotes the reflection factor of a material surface, α is the absorption factor. In the general case R and α depend on the body temperature. It is supposed that the time dependence of laser radiation intensity corresponds to the Gaussian profile

$$I_0(t) = \frac{2E_0\sqrt{\ln 2}}{\tau_L\sqrt{\pi}}\exp\left(-4\ln 2\left(\frac{t-\tau_S}{\tau_L}\right)^2\right), \quad (2)$$

where E_0 is the laser fluence, τ_L is the pulse full width at half maximum of the intensity profile (FWHM), τ_S is the time lag from the initial moment of computation till the moment of the maximum $I_0(t)$ achievement (in computations $\tau_S = 1.5\tau_L$).

For the description of the material thermal state the one-dimensional nonstationary heat conduction equation with the volume source of heat written down in a frame of reference connected with the moving evaporation front [4] is used

$$c\rho\left(\frac{\partial T}{\partial t} - \omega(t)\frac{\partial T}{\partial z}\right) = \frac{\partial}{\partial z}\lambda\frac{\partial T}{\partial z} + \alpha I(z,t). \quad (3)$$

The initial condition is

$$T(z,0) = T_0. \quad (4)$$

The boundary conditions are

$$\lambda\left.\frac{\partial T}{\partial z}\right|_{z=0} = \rho\omega(t)L,\ T(\infty,t) = T_0. \quad (5)$$

Here ρ, c, λ are the density, specific heat and heat conduction coefficient of a material correspondingly; L is the specific heat of evaporation (sublimation); T_0 denotes the initial temperature, $\omega(t)$ is the propagation velocity of the evaporation front.

The propagation velocity of the evaporation front is defined by the relation

$$\omega(t) = \frac{mF}{\rho}, \quad (6)$$

where F is the total particles flux through the surface coinciding with the evaporation front, m is the particle mass.

The total flux F consists of the flux F^+ of atoms formed due to the evaporation process and the flux F^- of atoms returned to the surface (back flux of particles)

$$F = F^+ - F^-. \quad (7)$$

The value of F is unknown beforehand. It can be found as a result of a joint solution of the heat and gasdynamic problems. At different time moments the ratio of fluxes F^+ and F^- can be arbitrary. In the considered model all particles returning to the surface are supposed to be condensed.

For the description of evaporation the Hertz-Knudsen law is used. According to it the flux of evaporated atoms is equal to

$$F^+ = \frac{1}{4} n_s u = \frac{p_s(T_w)}{(2\pi m k_B T_w)^{1/2}}. \quad (8)$$

Here n_s and p_s denote the concentration and pressure of saturated vapor at the surface temperature T_w, $u = (8 k_B T_w / \pi m)^{1/2}$ is the average thermal velocity of evaporated particles, k_B is the Boltzmann's constant.

For the pressure $p_s(T_w)$ definition the Clausius-Clapeyron equation is used

$$p_s(T_w) = p_b \exp\left(\frac{L}{R}\left(\frac{1}{T_b} - \frac{1}{T_w}\right)\right), \quad (9)$$

where T_b is the boiling temperature at the normal pressure p_b=10^5 Pa, R=k_B/m is the gas constant.

The velocity distribution function for atoms scattering from the surface is supposed to be semi-Maxwellian

$$f_{w+} = \frac{F^+}{2\pi}\left(\frac{1}{RT_w}\right)^2 \exp\left(-\frac{v_x^2 + v_y^2 + v_z^2}{2RT_w}\right), \; v_x > 0. \quad (10)$$

Here v_x, v_y, v_z are the velocity components of particles.

At the initial time moment there are no particles in the calculation region. At time $\tau=t/\tau_L=0$ the laser radiation pulse is switched on. For each time step processes of heating of a target and scattering of particles have been divided and modelled sequentially. At first equation (3) describing the process of heating of a material have been solved. Then vapor expansion have been simulated.

The heat model of a lift-offed film

Now there are no reliable data on optical characteristics of the lift-offed layer allowing to estimate quantitatively a fraction of laser radiation absorbed by the layer. At these conditions there is one way of the analysis: to consider the possible extreme cases. Let's assume that the lift-offed film absorbs a fraction (1-R_1) of laser radiation falling on it (R_1 is the reflection factor). At that the film shields completely a target from laser radiation. The sharp increase in absorption of radiation at a separation of a layer can be concerned with the development of instability of this layer and transition to a mode of spatial non-uniform heating and evaporation of the lift-offed layer [6].

Change of the thickness of the layer is defined by the equation

$$\frac{dh_1}{dt} = -\omega_a(t) - \omega_b(t), \quad (11)$$

where $\omega_a(t)$ and $\omega_b(t)$ are the propagation velocities of the evaporation (condensation) fronts at the surfaces a and b (Figure 1b) determined according to formula (6). At definition of particles fluxes to the surfaces a and b the movement of the layer in the laboratory system of coordinates with the velocity v_1 is taken into account.

The film motion is defined by the equation

$$\frac{dm_1 v_1}{dt} = p_a - p_b, \quad (12)$$

where m_1=$\rho_{Si} h_1(t)$ is the layer mass per unit area, p_a and p_b are the pressures at the surfaces a and b.

The film temperature is defined by the equation

$$\frac{dQ}{dt} = I_0(t)(1 - R_1) + (F_a + F_b)Lm + E_{ak} + E_{bk}, \quad (13)$$

where E_{ak} and E_{bk} denote the fluxes of kinetic energy of particles for surfaces a and b; $Q=m_1cT_1$.

The model of vapor motion

For modelling of expansion of a material evaporated from a surface the Direct Simulation Monte Carlo method (DSMC) was used. In the computer code the collision scheme without time counter (NTC - scheme) was realized. Mechanics of collisions corresponded to the hard spheres model. All particles returning to the surface were supposed to be absorbed by the material. The detailed description of the method and algorithm of DSMC corresponding to a normal evaporation mode is given in paper [4]. For re-indexation of particles and sampling of macroparameters the nonuniform grid with retraction to a surface, one-zone stationary grid for a mode without superheated liquid layer lift-off and moving two-zone one in the case of presence of explosive boiling, was used. The number of the modeling particles was $2*10^6$.

Matching of the heat and gasdynamic problems was made using the boundary conditions at the surface. The modeling atoms were put into the calculation region according to the relation (8) with the velocity distribution function (10). The surface temperature was calculated at the stage of the solution of the heat conduction equation. During the direct modelling the particles back flux F^- towards the surface at a given time step was calculated. This value was used at the next time step to determine the total flux F and the propagation velocity of the evaporation front ω.

CALCULATION RESULTS AND ANALYSIS

The calculations have been performed for a flat silicon target. The optical and thermophysical parameters for the crystalline silicon have been taken from [3, 5]. The calculation results for $\tau_L=1.3*10^{-8}$s, $\alpha=10^7$ m^{-1}, $R=R_1=0.6$ and two values of $E_0=4*10^4$ and $4.75*10^4$ J/m^2 are presented below. At $E_0=4*10^4$ J/m^2 the normal evaporation mode is realized, at $E_0=4.75*10^4$ J/m^2 the explosive boiling mode with a lift-off of a single layer is taken place. In the general case the number of lift-offed liquid layers can be greater.

The following dimensionless parameters are used: T is referred to the boiling temperature T_b=3504K, ρ - to the vapor density in equilibrium at T_b and p_b (ρ_b=0.1kg/m^3), velocity – to the most probable velocity of vapor particles at T_b ($\sqrt{2RT_b}$ =1.44 km/s), time – to the pulse width $\tau_L=1.3*10^{-8}$s.

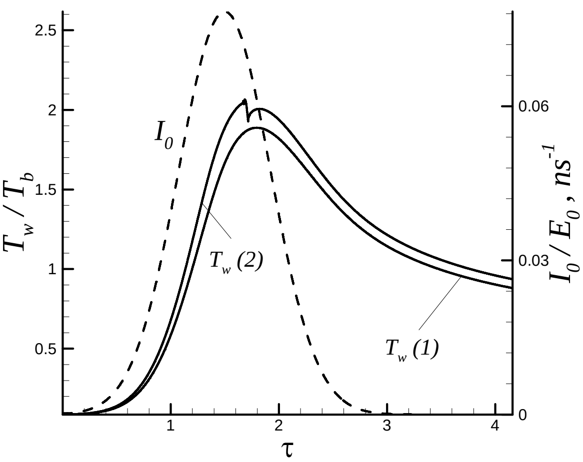


FIGURE 2. $I_0(\tau)$ and $T_w(\tau)$ dependencies:
1 – $E_0=4*10^4$J/m^2; 2 – $E_0=4.75*10^4$J/m^2.

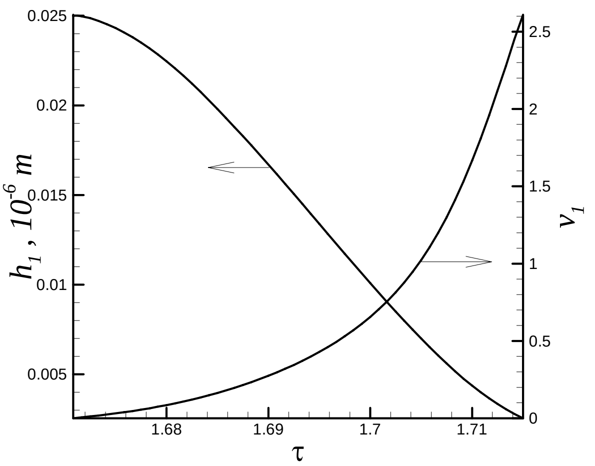
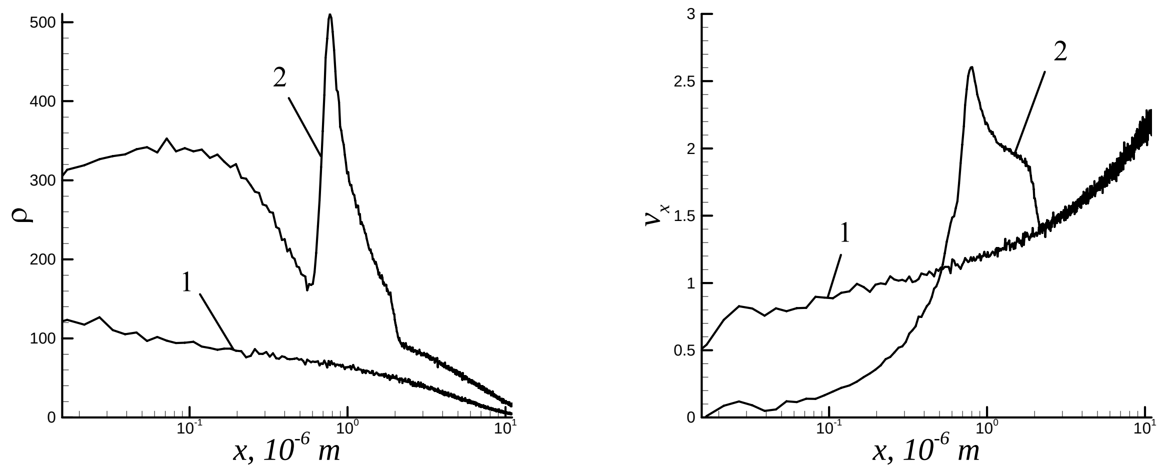


FIGURE 3. $h_1(\tau)$ and $v_1(\tau)$ dependencies.

In Figure 2 the calculation results of time evolution of T_w are presented. The same figure shows the $I_0(\tau)$ dependence. Maximums of T_w are shifted slightly towards the great τ in comparison with the $I_0(\tau)$ maximum. The near-surface layer is in a liquid state at τ>1 (melting temperature is T_{melt}=1685K). At E_0=4.75*10^4 J/m^2 explosive boiling of the near-surface layer occurs at τ=1.67. The lift-offed layer shields the target from laser radiation, therefore T_w of the target is decreasing. After film evaporation and stopping of shielding T_w is rapidly relaxing to the temperature profile typical of the normal evaporation mode.

In Figure 3 the time evolutions of the thickness and the velocity of the lift-offed film are shown. The initial film thickness is equal to 0.025 μm. Total evaporation of the layer occurs during 0.575 ns. For this time its velocity achieves 3.76 km/s. The film acceleration is great. By the moment of total evaporation the layer flies approximately 0.6 μm.

Figures 4 a and b present the profiles of ρ and v_x of the vapor after total layer evaporation (τ=1.715). Explosive boiling results in essential change of parameters of a vapor plume. The three-zone gasdynamic structure is formed in the flow field. There is the thin vapor layer with the high pressure and density and small velocity near the target surface. Till the moment of the end of evaporation of lift-offed liquid film expansion of this vapor determined the acceleration of the liquid film. Downstream there is the narrow high-speed vapor layer with the rather high density. This layer was generated as a result of compression by a moving liquid film and evaporation of this liquid film. The layer moves with the supersonic velocity (Mach number is in the range 1.6 – 1.8) and has the high dynamic pressure. In the section where ρ and v_x are maximal the vapor dynamic pressure ρv_x^2 achieves the value about 7*10^{3}atm. To the right of the high-speed layer there is a zone of free non-stationary vapor expansion in a vacuum with the structure typical of a mode of normal evaporation [4].



a b

FIGURE 4. ρ(*x*) (a) and $v_x(x)$ (b) profiles at τ=1.715: 1 - E_0=4*10^{4}J/m^2; 2 – E_0=4.75*10^{4}J/m^2.

At time τ=1.715 the flow in the region presented in Figure 4 is continual. During the time the three-zone structure is rapidly enough relaxing to the flow structure typical of a mode of normal evaporation. Figure 5 shows the profiles of ρ(*x*) and $v_x(x)$ at time τ=4.2 for E_0=4.75*10^{4}J/m^2. Curves 1 correspond to the calculation without the lift-off of the liquid layer, curves 2 – with the layer lift-off. It is well visible that influence of explosive boiling on vapor expansion becomes negligibly small by this time.

The calculation results at E_0=4.75*10^{4}J/m^2 presented above correspond to a mode of explosive boiling with a lift-off of a single liquid layer. At the same laser pulse duration the increase in the value of E_0 results in periodic separations of liquid layers. Calculations show that within the framework of the considered model at E_0=5*10^{4}J/m^2 a lift-off of two layers is taken place. At that the first layer separates earlier than it occurs at E_0=4.75*10^{4}J/m^2. Time between a lift-off of the first and the second layers is about 1 ns. Further increase in E_0 results in a separation of three and more layers. Our data on parameters and dynamics of lift-offed layers correspond well enough to the data [3].

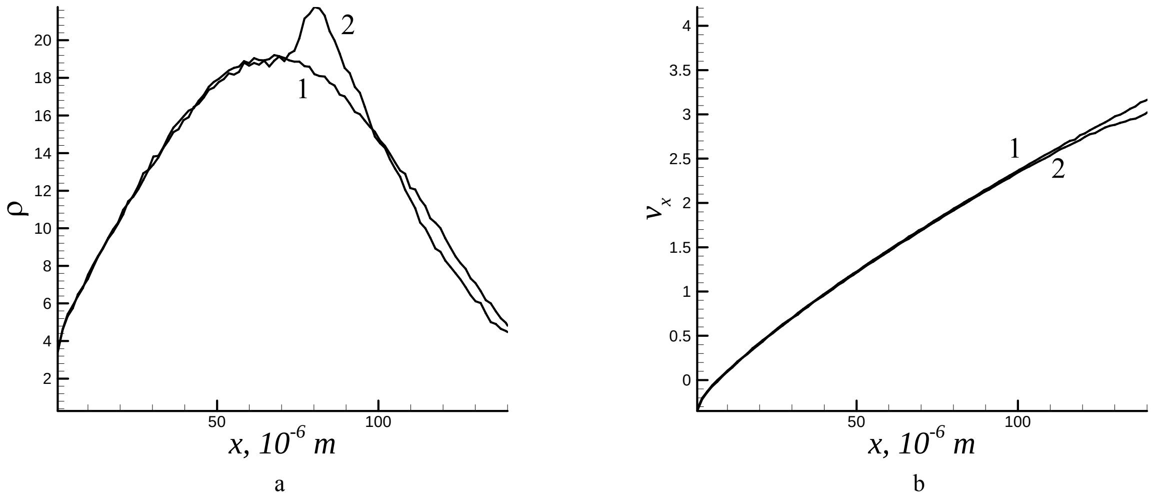


FIGURE 5. $\rho(x)$ (a) and $v_x(x)$ (b) profiles at τ=4.2 for E_0=4.75*10^4J/m^2: 1 – without the layer lift-off; 2 – with the layer lift-off.

CONCLUSION

Explosive boiling of condensed substances under pulsed laser influence in the nanosecond range relates to the very complicated physical phenomena. Meanwhile the detailed simulation of this phenomenon is beyond the bounds of real opportunities of the researchers. At the same time the certain progress in understanding of the explosive boiling mechanism under PLA of solid materials can be achieved within the framework of the set of approximate models allowing finding the limiting estimations of the basic parameters of this phenomenon. In our opinion, the proposed model is a good basis for formation of such model set. The model is open. The structure of our model and used simulation methods allow to change, include and exclude separate elements of the model. To a number of actual problems of the development of the approximate model of explosive boiling under PLA of solid materials it should be related the following ones: (1) extending of the model to a mode with a separation of several layers; (2) including of the processes of the breakdown of the lift-offed liquid films to fine drops and two-phase mixture expansion in the model.

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