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# MODEL OF EXPLOSIVE PULSED VAPORIZATION OF DIELECTRIC LIQUIDS BY INTENSE LASER IRRADIATION OF THEIR SURFACES

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*The mechanism of explosive vaporization interaction of laser radiation with matter is studied theoretically. It is shown that, in dielectric liquids with a free surface, periodic explosive boiling is possible if the laser radiation intensity exceeds the rate of heat transfer from the region of laser radiation absorption. Analytical expressions are obtained to estimate the pulsating boiling period and the thickness of the surface liquid layer dispersed by fluctuation vapor bubbles during each boiling. The degree of absorption of laser radiation by the aerosol formed above the liquid surface is estimated.*

**Key words:** *dielectric liquid, laser radiation, liquid vaporization, spinodal decomposition of the liquid.*

Laser irradiation of a liquid gives rise to the optoacoustic effect consisting of sound generation in the liquid. The mechanisms of optical generation of sound and their efficiency depend on the laser radiation intensity. At low intensities, the thermal mechanism of sound generation occurs, in which radiation absorption leads to thermal expansion of the medium, which is a sound source. At considerable intensities of laser radiation, a vaporization mechanism occurs consisting of the phase transition of the liquid to vapor and sound generation in the liquid due to the recoil of the vapor jet. At even higher radiation intensities, a plasma regime of interaction is observed, in which breakdown occurs above the liquid surface and the plasma region is expanded, leading to sound generation in the liquid.

The thermal mechanism has been extensively studied both theoretically and experimentally [1, 2]. The efficiency of the thermal mechanism of transformation of laser energy to acoustic energy is about 0.001%. The theoretical papers dealing with the thermal mechanism consider the nonlinear effects related to heating-induced variation in the thermodynamic properties of the medium and accounting for the nonlinear terms in the hydrodynamic equations, the influence of inhomogeneity of the medium, etc.

In the vaporization regime of interaction, the efficiency of transformation of laser radiation energy to acoustic energy is two to three orders of magnitude higher than that in the thermal regime. For the theoretical description of the vaporization regime of interaction of radiation with matter, it is necessary to consider nonlinear phenomena such as phase transition of the liquid to vapor and its expansion into the atmosphere. For metallic liquids, a thermal model of surface molecular vaporization has been developed which is valid up to the regime of hydrodynamic failure of material [3], whereas for dielectric liquids having low thermal conductivity and laser radiation absorption coefficient, there is a wide range of radiation intensities $10^3$–$10^7$ W/cm$^2$ in which explosive boiling [4] is observed.

A physical model for explosive boiling was proposed in [5, 6] and it consists of the following. During surface vaporization, the pressure on the liquid surface $p(T_0)$ is always lower than the saturated vapor pressure $p_s(T_0)$. Since $p < p_s(T_0)$ and the maximum liquid temperature $T_{\max} > T_0$, the near-surface layer of the liquid phase is in a superheated metastable state. This state can exist only at temperatures not exceeding the spinodal temperature $T_{\rm sp}(p)$, which determines the boundary of the region of absolute instability of the metastable liquid phase at the specified external pressure $p$. For this reason, a steady-state temperature distribution in the liquid at $T_{\max} > T_{\rm sp}$ appears

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impossible. At $T_{\max} \approx T_{\rm sp}$, in the neighborhood of the temperature maximum there is an explosive transition of the metastable liquid phase to the gaseous state. The excess pressure $\Delta p = p_s(T_{\max}) - p(T_0)$ leads to ejection of the surface layer of the liquid phase and the occurrence of a new interface on which the temperature first decreases due to vaporization and then increases because of heating by radiation. The process of breaking of the surface is then repeated.

The fullest mathematical model for explosive boiling was proposed by Andreev et al., [7], who determined the heating of a liquid surface to the temperature of absolute instability of the metastable liquid for an arbitrary time dependence of the radiation intensity and studied the formation of a vapor cavity in the region of the maximum temperature and its expansion due to vapor inflow from both the bulk of the liquid and the detaching liquid film. The equations of heat conduction and the dynamics of motion of the detaching film were solved using numerical methods.

The purpose of the present work is to perform a theoretical study of the main processes of explosive boiling and to obtain analytical expressions to estimate the key parameters of explosive boiling — the explosive boiling period, the thickness of the surface film dispersed by vapor bubbles in each boiling, and the degree of radiation absorption by the aerosol formed above the liquid surface.

**Formulation of the Problem.** In the thermal model of surface vaporization, the steady-state liquid-temperature distribution in a coordinate system moving together with the vaporization surface is written as [8]

$$T = T_\infty + (T_0 - T_\infty)\exp\left(-D/(x\chi)\right)$$

$$+ (T_0 - T_\infty + L/c_p)(1-y)^{-1}[\exp\left(-\mu x\right) - \exp\left(-D/(x\chi)\right)], \tag{1}$$

where $y = \mu\chi/D$, $L$ is the latent heat of vapor formation, $c_p$ is the heat capacity, $\mu$ is the laser radiation absorption coefficient, $\chi$ is the thermal diffusivity of the liquid, $T_0$ and $T_\infty$ are the liquid temperatures on the surface and at infinity, respectively, and $x$ is the coordinate reckoned from the surface into the depth of the liquid.

The velocity of motion of the vaporization surface $D$ is related to the radiation intensity $I_\infty$ and the surface temperature $T_0$ by the formula

$$D = J_\infty/[\rho_0(L + (T_0 - T_\infty)c_p)], \qquad T \to T_\infty \quad \text{at} \quad x \to \infty. \tag{2}$$

This temperature distribution in the liquid has a maximum due to the influence of two competing processes. On the one hand, the liquid is heated by the absorbed laser radiation, and on the other hand, it is cooled as heat moves both into the depth of the liquid and to the vaporization surface.

From Eq. (1) it follows that the coordinate and the maximum temperature are given by the formulas

$$x_{\max} \approx 1/\mu, \qquad T_{\max} \approx T_0 + L/(c_p y).$$

For dielectric liquids, the ratio $L/c_p$ is comparable to their critical temperature $T_*$; therefore, the critical temperature is reached for $y \approx 1$. Substituting the value $D = \mu\chi$ into (2), we obtain an estimate for the laser radiation intensity at which the dielectric liquid reaches the temperature $T_*$:

$$J_* = \rho_0 c_p \mu\chi(T_* - T_\infty).$$

For dielectric liquids, we have $J_* = 10^3$–$10^4$ W/cm$^2$, and for metallic liquids, $J_* = 10^8$–$10^9$ W/cm$^2$. In particular, for water, $J_* = 2 \cdot 10^3$ W/cm$^2$ at $\mu = 10^5$ m$^{-1}$ ($\lambda = 10.6$ $\mu$m) and $\chi = 10^{-7}$ m$^2$/sec.

At intensities higher than $J_*$ in the vicinity of the temperature maximum, the liquid enters a metastable state and bubble vapor appears in it in an explosive manner as a result of fluctuation nucleation. The rate of formation of bubble nuclei increases exponentially as the state of the liquid approaches the boundary of stability determined by the spinodal curve. In particular, for water, which is a typical dielectric liquid and is used for the subsequent numerical estimation, the approximate spinodal equation is written as [9]

$$T_{\rm sp} = T_*(0.9 + 0.1p/p_*), \tag{3}$$

where $T_* = 647$ K and $p_* = 221 \cdot 10^5$ Pa are the critical temperature and pressure of water.

From (3), it follows that the spinodal temperature varies in the range $T_* \cdot 10\%$ with the pressure varying over a wide range.

The critical size of a nucleus and the frequency of its formation are given by the expressions

$$r_* = 2\sigma/(p_s - p_0), \qquad \ln N \simeq 88 - 4\pi r_*^2\sigma/(3k_{\rm B}T), \tag{4}$$

where $p_s$ is the saturated vapor pressure in the bubble, $p_0$ is the liquid pressure, $\sigma$ is the surface-tension coefficient of the liquid, and $k_{\rm B}$ is Boltzmann's constant.

The further growth of the bubbles is determined by the counterpressure from the surrounding liquid and the rate of heat transfer to the surface of the growing bubbles. In the Rayeigh stage, where the bubble pressure is limited by inertial forces, the radius increases linearly with time and is equal to $r = t(2(p - p_0)/\rho_0)^{1/2}$. In the thermal stage, where the bubble growth is limited by heat supply, its radius depends on time as follows:

$$r = t^{1/2}(2k/(\rho_{\rm v}c_0))^{1/2}(T_1/(1 - T_1))^{1/2}, \qquad T_1 = c_{\rm v}(T - T(p))/L.$$

Here $T(p)$ is the saturated vapor temperature, $c_0$ and $k$ are heat capacity and thermal conductivity of the liquid, and $c_{\rm v}$ and $\rho_{\rm v}$ are heat capacity and density of the vapor, respectively.

The growing vapor bubbles break up the liquid surface layer into fine droplets, which, together with the vapor, scatter into the atmosphere. In this case, the liquid surface, on the one hand, is cooled due to surface vaporization, and, on the other hand, it is heated by the laser radiation that has reached the surface. The influence of these two competing processes results in that the liquid again reaches the spinodal state and breakup of the surface layer is repeated.

The periodic explosive boiling above the liquid surface results in the formation of an aerosol which vaporizes in the laser radiation field.

The main processes described by the proposed model of explosive boiling are considered below.

**Periodic Explosive Boiling.** Let us consider the vaporization process from the moment of detachment of the liquid surface layer. In this case, the liquid has an initial temperature with the profile $T = (T_{\rm sp} - T_\infty)\exp(-\mu x) + T_\infty$ obtained from the heat-conduction equation assuming that the energy heat transfer is small compared to the energy release during laser irradiation. The liquid vaporizes rapidly from the surface, resulting in a rapid decrease in the temperature on it.

Under the assumption that the temperature of the outer surface is $T_0$ and the velocity of motion of the liquid boundary is negligibly small compared to the velocity of temperature wave propagation into the depth of the liquid, the temperature profile of the cooled liquid is given by [10]

$$T = T_0 + \Big(a - T_0 + \frac{\chi A t}{k} + \frac{Ax^2}{2k}\Big)\Phi\Big(\frac{x}{2(\chi t)^{1/2}}\Big) + \frac{Ax}{k}\Big(\frac{\chi t}{\pi}\Big)^{1/2}\exp\Big(-\frac{x^2}{4\mu\chi t}\Big) + bx - \frac{Ax^2}{2k},$$

$$\Phi(z) = \frac{2}{\pi^{1/2}}\int\limits_0^z \exp(-\xi^2)\,d\xi, \tag{5}$$

where $A = \mu J$ is the energy-release density under laser radiation and $a = T_{\rm sp}$ and $b = -\mu(T_{\rm sp} - T_\infty)$ are the first coefficient in the Taylor series expansion at the point of maximum of the initial temperature $T = (T_{\rm sp} - T_\infty)\exp(-\mu x) + T_\infty$. From (5), it follows that, at $\mu(\chi t)^{1/2} \ll 1$, the maximum temperature and its coordinate are given by

$$T_{\max} = T_{\rm sp}(1 + \mu J t/(\rho_0 c_p T_{\rm sp}) - m\mu(\chi t)^{1/2}(T_{\rm sp} - T_\infty)/T_{\rm sp}), \qquad x_{\max} = m(\chi t)^{1/2}, \tag{6}$$

where $m \simeq 2[\ln(\mu(\pi\chi t)^{-1/2})(T_{\rm sp} - T_\infty)/(T_{\rm sp} - T_{\rm v})]^{1/2}$ is a time-dependent parameter (for water, $m = 2$–$4$ at $J = 10^3$–$10^6$ W/cm$^2$).

According to (6), in the initial period of time, the temperature at the point of maximum decreases due to cooling of the boundary, and then, because of heating by the laser radiation, it reaches the initial value. The total time of this process, which characterizes the pulsating vaporization period and the thickness of the broken surface layer, are equal, respectively, to

$$\tau = m^2\chi(\rho_0 c_p(T_{\rm sp} - T_\infty)/J)^2, \qquad \delta = m(\chi\tau)^{1/2}. \tag{7}$$

For water, $\tau \approx 1.2 \cdot 10^4 J^{-2}$ and $\delta \approx 10^{-1}J^{-1}$. The time $\tau$ is in seconds, and thickness $\delta$ in meters.

**Spinodal Decomposition of the Liquid and Breakup of its Surface Layer.** Depending on the relation between the time of formation of the vapor layer $t_{\rm layer}$, which will be called the time of spinodal decomposition of

the liquid, and the time of break of the liquid surface layer by a single bubble $t_b$, two versions of breakup of the surface layer are possible: 1) at $t_{\text{layer}} \ll t_b$, a vapor layer is formed in the neighborhood the temperature maximum and the further breakup of the surface layer is determined by the growth rate of the vapor layer and the development of instability of the thin surface liquid layer to the breakup into droplets; 2) at $t_{\text{layer}} \gg t_b$, the surface liquid layer is broken by single growing bubbles in a time equal to the total time required for the bubbles to fill the entire liquid region at the maximum temperature $t_{\text{layer}}$.

We estimate the time $t_{\text{layer}}$, following to [9], from the expression defining the probability of the occurrence of a vapor layer in the liquid heating region:

$$p(t) = 1 - \exp\Big( - cN(t) \Big/ \Big( h \Big| G_T \frac{dT}{dt} \Big|^3 \Big) \Big). \tag{8}$$

Here $N(t) = N_0 \exp[-G(T(t))]$ is the frequency of formation fluctuation nuclei, $h = |(1/2)(dG/dT)_0 \times (d^2T/dx^2)_0|^{-1/2}$ is the characteristic thickness of the layer in which boiling centers are formed, and $c$ is a constant that depends on the bubble growth dynamics. From (8), substituting the expression $d^2T/dx^2 \approx T_{\text{sp}} - T_{\text{v}}/(\chi\tau)$ from (4) and the expression $dG/dT \approx 16\pi\sigma^3/(3k_{\text{B}}T_{\text{sp}}^2 p_s^2)$ from (4), we obtain the following expression for the thickness of the nucleation layer:

$$h \approx (\chi\tau)^{1/2}[k_{\text{B}}T_{\text{sp}}^2 p_s^2/(8\sigma^3(T_{\text{sp}} - T_{\text{v}}))]^{1/2}.$$

According to estimates, the thickness of the nucleation layer $h$ is a few percent of the thickness of the liquid surface layer $\delta$.

Expression (8) defines the probability of a vapor layer occurring at a certain time $t$ from the beginning of heating of the liquid. However, the quantity of interest is not this time, but rather the interval during which the probability of formation of the vapor layer varies significantly, for example, from 1 to 70%. This time interval determines the time of spinodal decomposition of the liquid $t_{\text{layer}}$. From (8), it follows that the indicated increase in the probability occurs as the nucleation frequency increases by two orders of magnitude. This increase in the frequency corresponds to the superheating of the dielectric liquid by the quantity $\Delta T = 1$–$2^\circ$C [9]. Equation (6) implies the following estimate of the time of liquid superheating: $t_{\text{layer}} = \rho_0 c_p \Delta T/(\mu J)$.

The time of break of the surface layer by a single bubble is estimated from the time of growth of this bubble in the Rayleigh stage to a size comparable to the thickness of the liquid surface layer: $t_b = \delta(\rho_0/p_s)^{1/2}$. Substitution of the quantity $\delta$ from (7) yields

$$t_b/t_{\text{layer}} = m^2\mu\chi(T_{\text{sp}}/\Delta T)(\rho_0/p_s)^{1/2}.$$

For dielectric liquids, this ratio is smaller than unity, which indicates breakup of the surface layer by single bubbles in the total time $t_{\text{layer}}$.

We note that this result is in contradiction to the formulation of the problem in [7], where it is assumed a priori that, in the neighborhood of the temperature maximum, a vapor cavity 0.01 $\mu$m thick is formed in a time equal to 0.1 nsec. It separates the thin surface layer which, remaining stable, vaporizes throughout the action of the laser pulse.

A comparison of the spinodal decomposition time of the liquid with the pulsating boiling period of the liquid shows that the periodic boiling regime is disrupted at the radiation intensity exceeding the quantity

$$J = 2m^2\mu\chi\rho_0 c_p(T_{\text{sp}} - T_\infty)^2/\Delta T.$$

For water, $J = 2$ MW/cm$^2$. At high intensities, quasiuniform boiling of the surface layer proceeds until the beginning of hydrodynamic expansion of the material at $J \approx 10$ MW/cm$^2$ [11].

We consider the model of periodic liquid boiling in the thermodynamic coordinates of the state of the medium. The optothermodynamic phase trajectory of the liquid in the surface layer is shown in Fig. 1 ($p_s$ and $p_{\text{sp}}$ are binodal and spinodal curves, respectively). Curve 1 corresponds to the initial period of liquid heating to the spinodal temperature $T_{\text{sp}}$. At the temperature $T_{\text{sp}}$ in the vicinity of the temperature maximum, vapor bubbles are formed in an explosive manner; the pressure in these bubbles is equal to the saturation pressure vapor $p_s(T_{\text{sp}})$. The saturation pressure $p_s$ determines the pressure in the liquid surface layer during the small time interval of its spinodal decomposition $t_{\text{layer}}$ (curve 2). The breakup of the surface layer and the scattering of the droplets are followed by rapid vaporization of the material from the surface, resulting in rapid cooling of the liquid and a

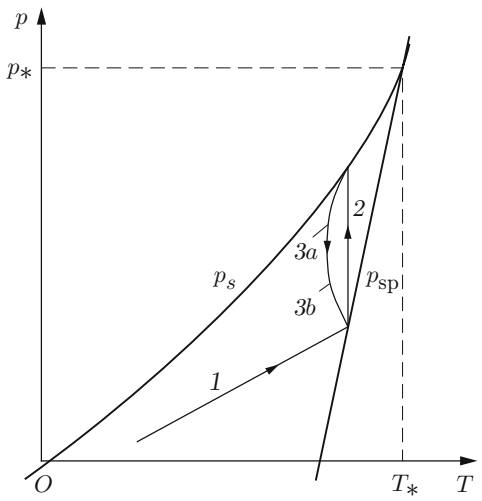


Fig. 1. Optothermodynamic phase trajectory of the liquid: 1) initial period of liquid heating to the spinodal temperature $T_{\rm sp}$; 2) period of spinodal decomposition of the liquid; 3) period of pressure decrease in the liquid (segment 3a refers to rapid cooling of the liquid and segment 3b refers to heating of the surface layer by laser radiation].

decrease in the liquid pressure (segment 3a). However, due to the heating of the surface layer by laser radiation, the spinodal temperature (segment 3b) is reached again and the boiling process is repeated.

The time of circulation around the thermodynamic loop is equal to the period of pulsating boiling of the liquid $\tau$. As the radiation intensity increases, the loop is shifted upward and the boiling period tends to zero.

**Vaporization of Aerosol.** The periodic boiling of the liquid leads to the formation of a vapor–droplet medium (aerosol) above the liquid surface, which scatters into the atmosphere toward the incident radiation. As a result of radiation absorption in the aerosol, the radiation intensity decreases. The energy of the absorbed radiation is consumed in vaporization of the aerosol. To estimate the degree of attenuation of laser radiation near the surface, we consider the following simplified model problem.

Let the duration of laser radiation be much larger than the period of pulsating boiling of the liquid. Then, steady-state aerosol flow of variable density decreasing in the direction of its motion is established above the liquid surface.

Assuming that radiation absorption has an insignificant influence on the rate of aerosol flow, we find the steady-state density distribution of the aerosol in the direction of its motion.

The variation in the aerosol density due to vaporization is described by the differential equation

$$\frac{d\varphi}{dx} = -\frac{\varphi\mu J}{\rho_0 V L}, \qquad \varphi\Big|_{x=\infty} = 0, \quad \varphi\Big|_{x=0} = \frac{\delta(J_0)}{V\tau(J_0)}. \tag{9}$$

Here $\varphi = \rho_1/\rho_0$ is the relative density of the aerosol, $\rho_1$ is the density of the aerosol, $\rho_0$ is the density of the liquid, $J_0$ is the radiation intensity near the liquid surface, $V$ is the velocity of the vapor–droplet flow, and $x$ is the coordinate reckoned from the liquid surface in the aerosol flow direction.

It should be noted that the boundary value of the relative density of the aerosol on the liquid surface $\varphi\Big|_{x=0} = \delta(J_0)/(V\tau(J_0))$ is determined by the intensity of the radiation that has reached the surface.

The decrease in the radiation intensity due to absorption in the aerosol is described by the Buger law, which in the adopted notation has the form $J = J_\infty \exp\Big(-\int\limits_x^\infty \mu\varphi\, dx\Big)$. The coefficient of radiation absorption in the aerosol is assumed to be proportional to its density.

Introducing the auxiliary variable $y = \int\limits_x^\infty \mu\varphi\, dx$, we write the integrodifferential equation (9) with the boundary conditions as follows:

$$y'' + Ay' \exp(-y) = 0,$$

$$y\Big|_{x=\infty} = 0, \qquad y'\Big|_{x=0} = -\mu\delta(J_0)/(V\tau(J_0)), \qquad A = \mu J_\infty/(\rho_0 V L).$$

The solution of this equation is the function

$$y(x) = \ln[1 + (\exp y\Big|_{x=0} - 1)\exp(-Ax)],$$

where $y\Big|_{x=0} = -\ln(1 - \mu\varphi\Big|_{x=0}/A)$.

Using the relation $\varphi = -y'/\mu$, we finally express the relative density of the aerosol as

$$\varphi(x) = A/\{\mu[1 - (1 + (\exp y\Big|_{x=0} - 1)\exp(-Ax))^{-1}]\}. \tag{10}$$

Substituting the boundary value $y\Big|_{x=0}$ into (10) after expressing the thickness $\delta$ and the period $\tau$ from relations and (7), we obtain the following equation:

$$y\Big|_{x=0} = -\ln\{1 - [L/(c_p(T_{\text{sp}} - T_\infty))](J_0/J_\infty)\}.$$

Since $J_0/J_\infty = \exp(-y\Big|_{x=0})$, we have

$$J_\infty/J_0 = 1 + L/(c_p(T_{\text{sp}} - T_\infty)).$$

The physical meaning of the relation obtained is that, in the adopted approximations, the radiation attenuation does not depend on the intensity of the incident radiation and is determined only by the thermodynamic properties of the liquid. For example, for water, the radiation intensity near the surface is approximately 40% of the incident-radiation intensity. Thus, 40% of the radiation power reaches the water surface and is consumed in its breaking, and 60% is consumed in vaporization of the aerosol.

The distance at which the density of the aerosol becomes negligibly small compared to its value on the liquid surface is comparable to the quantity $A^{-1}$ [see (10)], whose value for water at intensity $J_\infty = 1$ MW/cm$^2$ is equal to $10^{-3}$ m.

At laser beam diameters greater than the indicated distance, a gas-dynamic explosion can be used to model the entire region of explosive boiling of the liquid, and it can be employed in models of gas-dynamic description of interaction of laser radiation with matter, including arbitrary time dependences of the radiation intensity and arbitrary parameters of the atmosphere [12, 13].

In conclusion, we note that experimental observation of the pulsating vaporization regime is complicated since even insignificant nonuniformity in the radiation intensity distribution on the spot of its focusing on the liquid surface leads to rapid disappearance of the pulsating component of the pressure recorded by an acoustic sensor placed in the liquid. It is possible to show that the pulsating pressure component decreases in inverse proportion to time, and at $t \geqslant \tau(J/\Delta J)$, it becomes negligibly small ($\Delta J/J$ is the degree of nonuniformity in the radiation intensity distribution over the spot).

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