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#### ABSTRACT

A mathematical model of optical breakdown at the droplets of dielectric liquid exposed to pulsed laser radiation is developed. The following results are obtained by calculation methods: the distribution of pressure, density and temperature in the vapour aureole of the particle; the temperature field around the droplet of liquid. It has been found that at high energies in the gas bubble, the conditions for thermal ionization of the gas and for the start of electron avalanche, leading to the formation of plasma are created. Due to the volumetric heat generation the droplet overheats and is in a meta-stable state, the plasma formation is almost opaque to radiation, which leads to a sharp rise in temperature. As a result, an explosion occurs inside the droplet, forming a shock wave that propagates outward. The results can be used to assess the performance of high-power scanning lasers (LIDAR) when there are liquid droplets and other suspensions present in the atmosphere. Lasers can be used in fire and explosion safety systems of aerospace machinery. Another area of application is systems of laser ignition and detonation initiation.

KEYWORDS Fire safety, laser radiation, mathematical modeling, optical breakdown, droplet. ARTICLE HISTORY Received 19 May 2016 Revised 8 August 2016 Accepted 27 August 2016

## Introduction

LOOK

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In this study a mathematical model of optical breakdown at the dielectric liquid droplets exposed to pulsed laser radiation is being developed. The following individual stages of the process are considered: the heating and evaporation of the particles, the forming of a vapour aureole and its ionization by thermal ionization at the shock wave front, the propagation of shock waves in surrounding space around the particle. Simulation of shock wave processes in the vapour aureole of particles is carried out based on the equations of nonstationary flow of an ideal gas. Threshold characteristics of the laser beam parameters, sufficient for the initiation of optical breakdown in the condensed inclusions are determined. On the basis of the developed mathematical model and the numerical method, the processes occurring in a droplet of liquid are researched.

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The widespread use of lasers in everyday life, in devices such as the optical communication equipment, navigation, monitoring system of natural and manmade environments causes the relevance of researches aimed to study the physical processes of interaction of intense laser radiation with the dispersion medium. For optical laser-based systems that use the phenomenon of light reflection and scattering in the transparent and translucent environments, it is important to have an understanding of laser beam characteristics.

A systematic presentation of problems related to the optical breakdown at the condensed inclusions is given in monographs (Zuev, Kopitin & Kuzikovsky, 1980; Kopitin et all., 1990). The focus is on solids inclusions, such as corundum, aluminium, and other refractory materials.

During propagation of laser radiation in the atmosphere it will inevitably interact not only with solids particles, but also with liquid droplets, which are able to concentrate the radiation energy in their volumes, thus lowering the energy threshold for various nonlinear effects (Pogodaev & Rozhdestvenskiy, 1984; Koroteev & Shumay, 1991; Armstrong & Zardecki, 1990). In such cases, it is important to have the understanding of the characteristics of laser beam, sufficient to initiate the optical breakdown (Zemlyanov, Kuzikovsky & Chistyakov, 1981; Emelyanov & Volkov, 2003). These characteristics are of interest in relation to problems of fire detection, transporting radiation through explosive mixtures (Emelyanov & Volkov, 2006; Volkov, Emelyanov & Li, 2003), as well as to the problems of laser initiation of various processes, e.g. ignition or detonation (Bulat & Volkov, 2015b; Kopecek et all., 2003; Frost & Zhang, 2004; Zhang, 2009).

At the present time two basic interaction models are used to study the interaction of laser radiation with a liquid droplet: thermal and explosive (Emelyanov & Volkov, 2006; Loskutov & Strelkov, 1982). The thermal model uses detailed description of such stages as heating, vaporization and ionization of the vapour cloud (Astafieva & Prishivalko, 1998). It is assumed that the heating does not occur quickly, and the size of the vapour cloud has time to adapt to the particle radius, which changes in the process of liquid evaporation. In explosive model the particle heating is assumed to be so rapid that the substance of condensed phase explosively transfers into vapour (Emelyanov & Volkov, 2004). The mechanism of this phenomenon lies in the formation of vapour bubbles inside the droplet when the explosive boiling temperature is reached (the temperature of water at normal pressure is 578 K).

#### **Method**

# **Stages of process**

The viewed interaction process of laser radiation with a droplet (Figure 1) has a pronounced nonlinear optical activity, due to the droplet morphology, namely its quasi-spherical shape of surface. An optically transparent spherical particle acts as a focusing optical system, increasing the intensity of incident light radiation in the inner zones, which are situated near its illuminated and shadowed surfaces.

This leads to the fact that a number of nonlinear optical effects, such as stimulated scattering effect and optical breakdown effect (with a lower threshold of its manifestations than in a liquid) occur in micron droplets. To build an interaction model of laser radiation with a particle the thermal approach will be used. This approach allows distinguishing such stages of the process as heating, evaporation, and ionization of vapour cloud.

The interaction of directional radiation flux with the vapour particle leads to the flux focus (Figure 1a) in the area of shadow hemisphere of a droplet (Azarov et all., 2004).

Under overheat conditions, because the liquid intense heat generation, a droplet transfers from the stable state into a meta-stable state, in which the liquid temperature exceeds the temperature of saturated vapour at a given pressure. Due to internal evaporation of overheated liquid (Figure 1b) the pressure in vapour bubble increases, which leads to conditions of internal breakdown occurrence and the formation of opaque micro-plasma spot (Figure 1a), which absorbs radiation.



**Figure 1.** Development of interaction process of laser radiation with a droplet: a) radiation focusing; b) the formation of vapour bubble; c) the formation of plasma cloud; g) the emergence of a shock wave as a result of the explosion of plasma formation; d) the arrival of a shock wave to the droplet border, ionization of gas surrounding the droplet, ejection of small droplets into the environment; f) the occurrence of free electrons flow

Further increase of pressure in the vapour bubble initiates processes of explosive boiling of surface layer (Figure 1d). Increasing the pressure in the vapour bubble leads to formation of a shock wave inside the droplet. For dense media such waves, despite the fact that there is a significant pressure droplet at them, are of acoustic type. The arrival of a shock wave to the border of stages separation causes the rarefaction wave moving inwards the droplet, and a significant increase of fluid particles velocity at the boundary of the droplet. As a result, the emission of substance in the direction of radiation from the rear hemisphere of the particle occurs, in the form of small droplets (Figure 1e). The 3012 🕥 P. V. BULAT ET ALL.

emerged free electrons (Figure 1f) trigger an avalanche breakdown mechanism of external gas surrounding the droplet.

# Model of the laser pulse

The intensity of laser pulse can be represented as

$$I(t, x, y, z) = I_{m0} f_1(t) f_2(x, y) f_3(z) , \qquad (1)$$

where  $I_{m0}$  is the maximum pulse rate. The function  $f_{1in}$  (1) represents a change in pulse rate over time, the function  $f_2$  takes into account the spatial distribution of the pulse intensity and the function  $f_3$  describes pulse attenuation when passing through the medium.

The function  $f_1$  can be written as a continuous piecewise linear dependence of the laser pulse intensity on the time [10]

$$f_1(t) = \sum_{n=1}^{N} \left[ F_n + \left( F_{n+1} - F_n \right) \frac{t - t_n}{t_{n+1} - t_n} \right] \varphi(t_n, t_{n+1}) , \qquad (2)$$

where  $\phi(t_i, t_j)$  describes a single step

$$\varphi(t_i, t_j) = \frac{t - t_i + |t - t_i|}{2|t - t_i| + \delta} - \frac{t - t_j + |t - t_j|}{2|t - t_j| + \delta}.$$
(3)

The spatial distribution of pulse intensity, function  $f_2$  in (1), is subject to Gaussian law

$$f_2(x, y) = \exp\left[-2\left(x^2 + y^2\right)/R^2\right],$$
 (4)

where R is the characteristic radius of laser spot.

It is well known that, when passing through the substance, the light wave expends its energy on the excitation of atoms. During this the intensity of radiation decreases and is calculated according to the law of Bouguer-Lambert-Beer law (Nigmatulin, 1987)

$$f_3(z) = \exp(-\mu z), \tag{5}$$

where z is the thickness of layer, through which the light passes,  $\mu$  is the absorption coefficient, which depends on the nature, condition and concentration of particles as well as on the wavelength of transmitted light.

Thus, obtaining an integral characteristic, taking into account (1)–(5), the total energy of the laser pulse can be calculated

$$Q = \int_{0}^{\infty} \int_{0}^{2\pi} \int_{0}^{\infty} I_{m0} f_{1}(t) \exp\left(-2r^{2} / R^{2}\right) r dr d\phi dt .$$
(6)

Integrating, we obtain a formula for estimating the initial impulse

$$Q = \frac{\pi}{2} I_{\rm m0} \Theta R^2$$

# Modelling of laser heating process

In the problem of modelling the process of laser heating the significant role is played by the energy of heating pulse, at which the processes of developed evaporation of particles are launched and the conditions for ionization of the vapour aureole are created (Zuev, Kopitin & Kuzikovsky, 1980; Kopitin et all., 1990).

The heating of a droplet of liquid dielectric will be described by using the equation of non-stationary heat conduction taking into account the internal heat sources. Let us write the equation in the conservative form for the gas (k=1) and the particle (k=2) in the spherical coordinate system

$$\rho_k c_k \frac{\partial T_k}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \chi_k \frac{\partial T_k}{\partial r} \right) + G_k(r, t) , \qquad (7)$$

where  $\chi$  is the heat conductivity, *c* is the heat capacity, and the term *G* characterizes the chemical reactions associated with volumetric heat generation.

Let us introduce the boundary condition of conjugation of temperature fields, given the lack of phase transition processes on the surface

$$\left(\chi \frac{\partial T}{\partial r}\right)_{r=r_w} - \left(\chi_p \frac{\partial T_p}{\partial r}\right)_{r=r_w} = \mu I(t).$$
(8)

If the evaporation processes are present, then the equation (8) will have the following form

$$\left(\chi_{\rm p}\frac{\partial T_{\rm p}}{\partial r}\right)_{r=r_{\rm w}} = \rho H_{\rm v} u_{\rm v} - \mu I .$$
<sup>(9)</sup>

Here,  $u_v$  is the velocity of evaporation front,  $H_v$  is the specific heat of phase transition.

Let us introduce a new coordinate associated with the surface of the particle to describe the processes of evaporation and changes in particle's size over time

$$\eta = \frac{r}{r_w(t)} \,. \tag{10}$$

At a constant rate of heat conduction, the equation (7) takes the form

$$\rho_k c_k \left( \frac{\partial T_k}{\partial t} - \frac{\eta}{r_w^2} \frac{dr_w}{dt} \frac{\partial T_k}{\partial \eta} \right) = \frac{\chi_k}{r_w^2} \left( \frac{\partial^2 T_k}{\partial \eta^2} + \frac{2}{\eta} \frac{\partial T_k}{\partial \eta} \right) + G_k(\eta, t) .$$
(11)

With the further conduction of heat, a vapour cavity is formed inside the droplet. The fluid boiling processes lead to increase of pressure and to further expansion of the cavity, which causes a vapour explosion of the droplets.

We will assume that the motion of the fluid inside the droplet is potential and spherically symmetric. In this case, the dynamics of the droplet with gas cavity can be represented by the generalized Rayleigh equation (Armstrong & Zardecki, 1990)

$$\left(1+\frac{r_{\rm b}}{r_{\rm w}}\right)\left(r_{\rm b}\ddot{r}_{\rm b}+2\ddot{r}_{\rm b}\right)-\frac{1}{2}\left(1+\frac{r_{\rm b}^4}{r_{\rm w}^4}\right)r_{\rm b}^2=\frac{p_{\rm b}-p_{\rm w}}{\rho/r_{\rm w}},\tag{12}$$

where  $p_w$  is the pressure at the droplet's external border,  $p_b$  is the pressure on the surface of gas bubble.

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Energy balance relation for the energy absorbed by the particle and spent on phase transition processes during the equilibrium evaporation regime in the dimensionless coordinate associated with a variable radius of the particle, can be represented as

$$\frac{dr_w^2}{dt} = -\frac{2\chi_p}{\rho_p H_v} \left(\frac{\partial T_p}{\partial \eta}\right)_{\eta=1}.$$
(13)

This ratio (13) allows to calculate  $r_w$  in (12) and will allow to determine the current size of the evaporating droplet.

### Ionization of vapour aureole

Mathematical model of plasma formation includes the consideration of the process of pumping the electron component by radiation due to the inverse effect

$$\frac{dT_{a}}{dt} = \frac{6m_{e}}{5m_{a}} (T_{e} - T_{a}) \alpha \nu$$

$$\frac{dT_{e}}{dt} = -\left(T_{e} + \frac{2E}{5k}\right) \frac{1}{\alpha} \frac{d\alpha}{dt} - \frac{6m_{e}}{5m_{a}} (T_{e} - T_{a}) \nu + \frac{2}{5k} \frac{\mu I}{\alpha n}$$

$$\frac{d\alpha}{dt} = \frac{A}{T_{e}^{9/2}} n \left[ \alpha (1 - \alpha) \frac{\beta^{2} n}{1 - \beta} - \alpha^{3} n \right]$$

$$\frac{dm_{p}}{dt} = -\frac{K_{p} I W_{p}}{H_{v}}$$
(14)

Here, *m* is the mass, *n* is the concentration of heavy particles,  $\alpha$  is the degree of ionization,  $\beta$  is the equilibrium degree of ionization, *v* is the frequency of collisions of electrons with atoms and ions, *E* is the ionization potential,  $A=1.05\cdot10^{-8}$  cm<sup>6</sup>·K<sup>9/2</sup>. The indices *a* and *e* in (14) belong to the atoms and electrons, respectively. The value of equilibrium degree of ionization is calculated by ionization equation of Saha at the evaporation temperature.

#### Numerical method

Simulation of gas-dynamic processes in the vapour aureole of particle is reduced to integration of the equations of unsteady flow of an ideal gas. In the vector form the conservation laws are as follows

$$\frac{\partial \mathbf{U}}{\partial t} + \frac{\partial \mathbf{F}(\mathbf{U})}{\partial x} = \mathbf{G} \ . \tag{15}$$

The vector of gas-dynamic variables, vector of the flow and the source term are defined as

$$\mathbf{U} = \begin{pmatrix} \rho \\ \rho u \\ \rho e \\ \rho Y_j \end{pmatrix}, \quad \mathbf{F} = \begin{pmatrix} \rho u \\ \rho u^2 + p \\ (e + p)u \\ \rho u Y_j \end{pmatrix}, \quad \mathbf{H} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \\ \omega_j \end{pmatrix}.$$
(16)

Here,  $\rho$  is the density, *u* is velocity, *p* is pressure, *e* is the total energy per mass unit,  $Y_j$  is mass concentration of *j*-th component of the mixture.

The system of equations (15)-(16) is closed by equations of thermal and caloric state. The mass concentration change rate of *j*-th component of the

mixture is calculated depending on burning behaviour of the specific chemical composition.

For equation sampling the methods of control volume and splitting by physical processes are used. Flows are calculated using the Godunov scheme of 2nd order (Bulat & Bulat, 2015; Volkov & Bulat, 2015; Bulat & Volkov, 2015a; Bulat et all., 2015).

## **Results and discussion**

The calculations were performed for a pulsed chemical HF-laser with the following characteristics  $t_i=2.6 \text{ µs}$ ,  $\lambda=4.2 \text{ µm}$ , R=5 mm,  $\Theta=1.5 \text{ µs}$ . The following tasks were performed consequently: heating of a droplet up to explosive transfer temperature, formation of a shock wave from explosion products, registration of initial electron concentration in a gas due to thermal ionization behind a shock wave front, the development of an electron avalanche in a vapour aureole.

### The conditions of optical breakdown

Numerical modelling results for fixed-size particles with different pulse power are shown in Tables 1–3.

ruble in conditions of optical breakdown for ip of microns								
Q, J	t <sub>e</sub> , ms	α	T>Te	Breakdown				
5	-	-	-	-				
10	1.83	10 <sup>-2</sup>	+	+				
15	1.38	10 <sup>-2</sup>	+	+				
20	1.13	10 <sup>-2</sup>	+	+				
30	0.93	10 <sup>-2</sup>	+	+				
50	0.72	10 <sup>-2</sup>	+	+				

Table 1. Conditions of optical breakdown for  $r_p$ =5 microns

Table 2. C	Conditions	of optical	breakdown	for $r_p=10$ microns
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Q, J	t <sub>e</sub> , ms	α	T>Te	Breakdown
5	-	-	-	-
10	-	-	-	-
15	1.41	10 <sup>-5</sup>	+	+
20	1.16	10 <sup>-2</sup>	+	+
30	0.96	10 <sup>-2</sup>	+	+
50	0.77	10 <sup>-2</sup>	+	+

Table	3.	Conditions	of	optical	breakdown	for	r <sub>p</sub> =20	microns
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Q, J	t <sub>e</sub> , ms	α	T>T <sub>e</sub>	Breakdown
5	-	-	-	-
10	-	-	-	-
15	1.48	10 <sup>-11</sup>	+	-
20	1.23	10 <sup>-11</sup>	+	-
30	1.04	10 <sup>-11</sup>	+	+
50	0.83	10 <sup>-11</sup>	+	+

The first column corresponds to the energy of laser pulse. The second column specifies the time required for heating of a particle and for formation of a shock wave from the explosive transformation products. The third column shows the value of ionization degree behind a shock wave. The fourth column marks the fulfilment of explosive transformation conditions. The fifth column contains

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the conclusion about the development of electron avalanche under these conditions. In particular, Table 1 shows that at the droplet with  $r_p=5 \mu m$  the breakdown occurs at the energy  $Q\sim10$  J. By increasing the droplet size the time of its heating increases and ionization degree behind a shock wave decreases. However, as can be seen from Table 2, the value of  $Q\sim15$  J is sufficient to initiate the laser breakdown, as estimated by calculation. If  $r_p=20 \mu m$  a reduction in value of initial ionization degree behind a shock wave occurs. This leads to the fact that the electron avalanche is not developed for low-energy laser pulse and the limit of plasma formation increases to  $Q\sim30$  J. Changing the size of the focal spot has a significant impact on the threshold value of optical breakdown. Thus, for droplets with  $r_p=10 \mu m$  the threshold power is 12 J, when R=3mm, 30 J when R=5 mm and 100 J when R=10 mm.

## Transformation of scattering indicatrices

Mie scattering theory, for certain cases (when refracting or reflecting particles are circular or elliptical in shape), predicts the transformation of scattering indicatrix. Indicatrices lose their symmetry (forward scattering may exceed the backward scattering and vice versa), and gradually become multi-pedalled. The mathematical basis of Mie theory is the expansion of equations for reradiated electromagnetic wave in terms of small parameter x=ka, where k is the wave number, a is the particle radius. With the increase of this parameter more and more expansion members in terms of x powers have to be taken into account. The frequency dependence of scattering intensity I also changes and becomes slower than the law of Rayleigh would suggest. The distributions of radiation intensity in a spherical droplet for various values of Mie parameter (parameter x) is shown in Figure 2.



Figure 2. Distributions of radiation intensity when x=1 (a) and x=2 (b)

## The results of calculation of the droplet heating process

The values of temperature inside the droplet and in the environment (Figure 2) at different time points: 1 - t=0.05, 2 - 0.30, 3 - 0.55, 4 - 0.80, 5 - 1.05, 6 - 1.30, 7 - 1.55 µs are obtained.



Figure 2. Temperature field around the water droplet at Q=20 J

Figure 2 shows that there is a local increase in temperature at the boundary of particle due to supply energy flow. A thin layer of high temperature, where ionization processes occur, is formed near the boundaries of a droplet.

At high energies the so-called process of electron avalanche begins inside the droplet after the thermal ionization and the droplet transitions into a metastable state. Line 7 describes the process of explosion beginning with a temperature of 698 K, and with time of process development of 54  $\mu$ s.

Depending on particle location f in relation to the beam axis different heating takes place. The particles, located on the periphery, do not enter the regime of advanced evaporation and do not create conditions for plasma forming regime. Different time of breakdown occurrence is associated with different radiation intensity, which depends on the distance from the beam axis to the particles. Thus, for the most distant particles breakdown conditions do not occur.

## Calculation results of the shock wave formation

Expansion of plasma formation generates a strong shock wave, the intensity of which decreases with increase of distance from the particle centre (Figure 3). The lines correspond to different points in time after the end of laser pulse:  $1 - 2.60, 2 - 2.75, 3 - 2.90, 4 - 3.05, 5 - 3.20, 6 - 3.35 \,\mu\text{m}$ .

Plasma formation becomes almost opaque to radiation, which results in a rise in temperature and pressure of evaporation products, as well as to the formation of a non-stationary gas-dynamic flow in the vicinity of micro-plasma formation. The limiting factors of plasma formation process are the droplet heating time to an explosive transformation temperature (at low pulse energies large droplet do not have enough time to heat up, and small ones intensively exchange heat with the environment), the intensity of the shock wave, which promotes the thermal ionization of gas (for massive droplets the intensity of this wave is small) and the development of an electron avalanche. The occurrence of breakdown is the result of competition of these factors.

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Figure 3. The distribution of pressure (a), density (b), velocity (c) and temperature (d) in the aureole vapour of the particle

# **Conclusion**

A mathematical model of the interaction of laser radiation with transparent droplet is developed, basing on thermal approach. The following process stages are considered: heating, evaporation, formation of vapour aureole, thermal ionization. The characteristics of the laser pulse required to create optical breakdown are calculated. The created model can be used to estimate the thresholds of optical breakdown. These given data show that the proposed mathematical model is sufficiently informative for assessing the characteristics of the optical breakdown threshold. The adopted scheme of the electron avalanche occurrence on seed electrons, which are generated by thermal ionization in the shock wave, explains the qualitative picture of the process and provides the basis for quantitative estimates. Using experimental data allows to adjust the model and to improve the reliability of the forecasts based on it.

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#### **Disclosure statement**

No potential conflict of interest was reported by the authors.

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