## MODEL OF DEFORMATION AND FRAGMENTATION OF LARGE WATER DROPS UPON EXPOSURE TO CO,-LASER RADIATION

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A model of the process of light-induced deformations of large absorbing water drops has been constructed. The role of hydrodynamical mechanism as well as of the effect of phase explosion of overheated liquid in fragmentation of the particles has been analyzed. An experimental data on the dependence of the size of fragments of the drops on the rate of the light energy influx into the material of the particle has been interpreted.

The liquid absorbing particles exposed to the intense laser radiation rapidly change states in their volumes, namely, undergo explosive effervescence.<sup>1</sup> The distribution of the absorbed light energy inside the particles significantly affects the effervescence. When the radiation is absorbed quasiuniformly, which is realized in drops with radii  $a_0 (2\alpha_a)^{-1}$ , where  $\alpha_a$  is the absorption coefficient of a liquid at the laser radiation wavelength, a volume effervescence of the drops with their subsequent fragmentation is observed. This regime, which is referred to as the uniform phase explosion,<sup>1</sup> was studied in sufficient detail in Refs. 1-3. For the absorbing particles with large radii  $(a_0 > (2\alpha_2)^{-1})$  the field of release of the internal energy is strongly nonuniform and is characterized by the existence of the maxima near the illuminated and shadow surfaces of the drop.<sup>4</sup> The experiments described in Refs. 5-10 showed that the explosion of such particles, as a rule, is a multistage process. At first, ejection of the vapor-condensate from the droplet surface occurs followed by a deformation of the particle and its fragmentation into many small drops, which in the case of a prolonged influx of the laser energy are also fragmented.

In this paper, we construct some theoretical models of deformation and fragmentation of large water drops by  $\rm CO_2-$  laser radiation under conditions of high rates of heating when the heat release occurs without equalizing of temperature gradients throughout the volumes of the particles. An employment of the experimental data is a significant point of this approach.

1. The analysis of the above-mentioned experimental data makes it possible to construct the following pattern of the deformation of a liquid particle. As a result of heating of the near-surface layers of the drop (where the heat release is maximum), the conditions for the explosive effervescence of the liquid are realized. The ejections of the vapor-droplet mixture exert a reactive pressure at the near-surface layers of the drop thereby resulting in deformation and motion of the drop. In what follows, the fragmentation of the deformed particle into many small drops can occur under certain conditions.

Let us consider the formulation of the problem of deformation of a liquid particle in the integral form

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(K+K_{\mathrm{M}}+\Pi\right)+N=\int_{S}p(\mathbf{vn})\,\mathrm{d}S'\,;\tag{1}$$

and

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$$M'_0 \frac{\mathrm{d}\mathbf{v}_{\mathrm{M}}}{\mathrm{d}t} = \mathbf{n} \int_{S} p(\mathbf{r}) \mathrm{d}S' + \mathbf{F}_{\mathrm{a}} \,. \tag{2}$$

Equation (1) is the energy balance equation for the deformed particle and Eq. (2) is the equation of motion of its center of mass. In Eqs. (1) and (2),  $K = \frac{1}{2} \int_{V} \rho_1 v^2 dV'$  is the kinetic energy of the liquid contained in the drop,  $\Pi = \sigma S$  is the surface energy,  $N = \frac{1}{2} \eta \int_{V} \left( \frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i} \right)^2 dV'$  is

the rate of viscous dissipation,  $K_{\rm M} = \frac{1}{2} \left( M'_0 v_{\rm M}^2 \right)$  is the kinetic energy of motion of the center of mass,  $M'_0$  and  $\mathbf{v}_{\rm M}$  are the mass and the velocity of the drop after ejection of the vapor-condensate, V and S are the volume and surface area of the deformed drop,  $\mathbf{v}$  is the velocity of flow,  $\mathbf{n}$  is the external normal to the droplet surface,  $\sigma$  is the coefficient of surface tension, p is the field of external pressure at the surface of the particle,  $\eta$  is the dynamic viscosity,  $F_{\rm a}$  is the aerodynamic droplet drag,  $\rho_1$  is the density of liquid, and  $\mathbf{r}$  is the radial distance in the drop.<sup>11</sup>

Since the Reynolds number for this problem  $\operatorname{Re} = va_0/v \gg 1$ , where v is the kinematic viscosity of liquid, then except for the region of boundary layer with the thickness  $\delta \simeq a_0/\sqrt{\operatorname{Re}} \ll a_0$  the flow inside the droplet can be considered to be irrotational.<sup>11</sup>

In the theory of deformation and fragmentation of particles, the perturbed surface shape is often represented in the form of ellipsoids.<sup>12</sup> Following this approximation we can write for the velocity potential  $\Phi$  (Ref. 11)

$$\Phi = \frac{1}{2} \left( \frac{\mathbf{x}^2}{a_1} \frac{\mathrm{d}a_1}{\mathrm{d}t} + \frac{y^2}{a_2} \frac{\mathrm{d}a_2}{\mathrm{d}t} + \frac{\mathbf{z}^2}{a_3} \frac{\mathrm{d}a_3}{\mathrm{d}t} \right) \,,$$

where  $a_i$  are the semi-axes. The velocity  $\mathbf{v} = \nabla_{\mathbf{r}} \Phi$  and, consequently, we succeed in determining all terms, which enter into Eq. (1).

When the droplet deformation has the form of spheroid  $(a_1 = a_3)$ , using new variables the degree of deformation  $\gamma = a_2 / a_1$  and the dimensionless time  $\tau = \Omega_0 t$ ,  $(\Omega_0 = (8\sigma/\rho_1 a_0^3)^{1/2}$  is the basic frequency of natural

oscillations of the drop), neglecting the viscous dissipation Eq. (1) is reduced to a form

$$\frac{d^{2}\gamma}{d\tau^{2}} - \frac{(\gamma^{2}+2)}{3\gamma(\gamma^{2}+1/2)}\frac{d\gamma}{d\tau} + \frac{135\gamma^{8/3}}{32(\gamma^{2}+1/2)}\frac{dS}{d\tau} = = \frac{\gamma^{8/3}}{2(d\gamma/d\tau)(\gamma^{2}+1/2)4\pi\sigma a_{0}^{2}} \left[\int_{S} p(\mathbf{vn}) \, dS' - \Omega_{0} \, \frac{d}{d\tau} \left(K_{M}\right)\right],(3)$$

where

$$S = \left(2\pi \ a_1^2 + \frac{\pi \ a_2^2}{\sqrt{1 - \gamma^2}} \ln \frac{1 + \sqrt{1 - \gamma^2}}{1 - \sqrt{1 - \gamma^2}}\right) \frac{1}{a_0^2} \ , \ \text{ at } \gamma < 1$$

and

$$S = \left(2\pi \ a_1^2 + 2\pi \ \frac{a_2^2 \ \gamma^2}{\sqrt{\gamma^2 - 1}} \ \mathrm{arcsin} \ \frac{\sqrt{\gamma^2 - 1}}{\gamma} \right) \frac{1}{a_0^2} \ , \quad \mathrm{at} \ \gamma \geq 1.$$

It is not difficult to show that as  $|\gamma - 1| \rightarrow 0$  and with the null right side Eq. (3) goes over to an equation for small oscillations.

2. Let us consider the case, in which the droplet deformation occurs as a result of the reactive pressure of the explosive surface layers. It can be assumed that  $p(\tau) \sim \delta(\tau - \tau_{ex})$ , where  $\delta(\tau)$  is the delta function of Dirac and  $\tau_{ex}$  is the dimensionless time of the start of the explosive effervescence,<sup>3</sup> because the time of formation and growth of the vapor phase in the region of energy release is much less than the time of the particle deformation.

By integrating Eq. (3) over the time and taking into account the fact that according to the principle of conservation of momentum

$$\int_{0}^{\infty} \mathrm{d}\tau' \int_{S} p(\mathbf{vn}) \, \mathrm{d}S' = \frac{I_{\mathrm{p}}^{2}}{2\mathrm{M}_{0}'} \,,$$

where  $I_{\rm p} = \int_{0}^{M_{\rm II}} (\mathbf{vn}) d\mathbf{M} = v_1 M_1 + v_2 M_2$  is the reactive recoil

momentum of the ejected vapor and  $M_1$ ,  $v_1$ ,  $M_2$ , and  $v_2$  are the masses and velocities of the vapor—condensate ejected from the illuminated and shadow sides of the droplet surface, respectively ( $M_{\rm II} = M_1 + M_2$ ), we obtain

$$\frac{16(\gamma^2 + 1/2)}{135\gamma^{8/3}} \left(\frac{d\gamma}{d\tau}\right)^2 + \frac{S(\tau_1)}{2\gamma^{2/3}} = \\ = \frac{1}{2} S(\tau_1 = 0) + \frac{I_p^2}{8\pi a_0^2 \sigma M'_0} - \frac{K_M}{4\pi \sigma a_0^2}, \qquad (4)$$

Here  $\tau_1 = \tau - \tau_{ex}$ ,  $(\tau_{ex} = \Omega_0 t_{ex})$ .

The kinetic energy of motion of the center of mass of the drop can be determined with the help of Eq. (2) and has the form

$$K_{\rm M} = \frac{M_0' v_{\rm M}^2}{2} = \frac{2 I_{\rm p}^2}{9M_0'} \,,$$

where  $M'_0 = M_0 - (M_1 + M_2)$  and  $M_0$  is the starting mass of the drop with radius  $a_0$ . The initial conditions for Eq. (4) are as follows:

$$\gamma \left(\tau_{1} = 0\right) = \gamma_{0}; \quad \left(\frac{d\gamma}{d\tau}\right) \bigg|_{\tau_{1}=0} = -\frac{1}{a_{0}} \times \left[\frac{43\left(5\left(M_{1}^{2} v_{1}^{2} + M_{2}^{2} v_{2}^{2}\right) + 26M_{1}v_{1}M_{2}v_{2}\right)}{288\pi\sigma M_{0}'}\right]^{1/2}, \quad (5)$$

where  $\gamma_0 = a_2^0/a_1^0$ ,  $a_2^1 = a_0$  and  $a_0^1 = 3M'_0/(4\pi\rho_1 a_0^2)$ . The masses of the ejected vapor–condensate  $M_1$  and  $M_2$  can be determined from the solution of the problem of phase explosion of the drop<sup>1,3</sup> and from the given form of the temperature field inside the particle at  $t = t_{\rm ex}$ . The values  $v_1$  and  $v_2$ , as shown experimentally in Refs. 5–10 vary in the range ~ 400–600 m/s at high rates of droplet heating. The theoretical calculation of the products of explosive fragmentation of the water drops gives analogous values.<sup>1,3</sup>

In the present paper we have made a numerical calculation of the dependence of  $\gamma(\tau)$  based on Eqs. (4) and (5) with different initial conditions. We have considered the situations with high rates of heating  $((M_1 + M_2)/M'_0 = 0.5, 0.25, \text{ and } 0.15 \text{ for } a_0 = 15, 30, \text{ and } 50 \,\mu\text{m})$  and deformation. The calculated results are shown in Fig. 1.



FIG. 1 The time dependence of the reciprocal degree of deformation of the water drops with different starting radii:  $a_0 = 15 (1 \text{ and } 1'), 30 (2 \text{ and } 2'), \text{ and } 50 \,\mu\text{m} (3 \text{ and } 3') \text{ at}$  $J_{\rm p} = 2 \cdot 10^9 (1-3) \text{ and } 5 \cdot 10^8 \text{ K/s} (1'-3').$ 

3. When the density of the radiation energy w was close in value to the threshold of effervescence of the liquid  $w_{\rm ex}$ , we have experimentally found another deformation pattern of large drops,<sup>14</sup> when at first the particle extended in the direction of propagation of radiation owing to a formation and growth of vapor regions near the surface, which was followed by the relaxation oscillations of the particle.

Let us consider the model of such deformations of the droplet with its subsequent relaxation based on the following ideas. During the time, which is called deformation time  $t_d$ , after the start of the laser action, vapor bubbles arise near the illuminated and shadow sides which distort the droplet surface. The drop acquires the shape of a spheroid with the principal axis, which coincides

Mathematically, a formulation of the problem, which corresponds to the model, is reduced to Eq. (3) with the null right side and the initial conditions prescribed at time  $\tau = \tau_d \ (\tau_d = \Omega_0 t_d)$ :

$$\gamma(\tau_{\rm d}) = \gamma_0; \left( \frac{d\gamma}{d\tau} \right) \Big|_{\tau = \tau_{\rm d}} = 0$$

The values of the initial deformation can be determined from Eq. (4), where  $\tau_1 = \tau - \tau_d$ ,  $K_M = 0$ , and  $v_1$  and  $v_2$  are the velocities of motion of the surfaces of the illuminated and shadow hemispheres of the drop. For large drops at low rates of energy influx when a small number of bubbles is formed, the velocity of motion of the liquid surface in the case, in which the bubble rise to the surface, can be determined from the solution of the problem of the dynamics of the bubble, which grows in the droplet center. The solution of this problem is well known.<sup>15</sup> Thus, for determination of  $\gamma_0$ , we succeed in writing down the following equation:

$$\left(1 + \frac{\gamma_0^2}{\sqrt{\gamma_0^2 - 1}} \arcsin \frac{\sqrt{\gamma_0^2 - 1}}{\gamma_0}\right) \gamma_0^{-2/3} (\gamma_0 - 1)^{1/3} = \frac{a_0 \rho_2}{3\sigma} (v_1^2 + v_2^2)$$

where  $\rho_2$  is the vapor density.

The results of numerical solution of the problem of the relaxation oscillations are shown in Fig. 2, where the values of the relative deviation of the oscillation frequency  $(\Omega - \Omega_0)/\Omega_0$  from the frequency of natural oscillation  $\Omega_0$  are plotted on the Y axis as functions of the initial radius of the particles. The experimental data taken from Ref. 14 are shown as well. As can be seen from the figure, the degree of nonlinearity of the oscillations increases as the size of the particles decreases. The quantitative difference of the experimental data from the calculated results in the region of large particles indicates that the modes of oscillations, whose frequency deviated from the frequency of the fundamental mode, were recorded in the experiments.



FIG. 2. The relative deviation of the frequency of deformation oscillations of the drop vs its size according to the data of Ref. 14. The solid curve denotes the theoretical calculation.

4. The above–considered models of the interaction between the intense laser radiation and the absorbing large drops, indicate that, under certain conditions, significant deformations of particles can occur.

When considering the physical mechanisms of fragmentation of such droplets, one can select two fundamental processes, which cause their fragmentation, i.e., the hydrodynamical instability and phase explosion. In the first case, the drop, whose volume remains unheated by the radiation, is fragmented under the impact on the front surfaces produced by the vapor-condensate separating from the regions of energy release, and the reason for this is the evolution of the surface perturbations at high rates of deformation. According to the experiments of Ref. 16, in the case of hydrodynamical instability, the fragmentation of the drops into many small drops occurs at  $\gamma = \gamma_{cr} \simeq 0.1$ . In the second case, the explosive effervescence of the droplet, which has not yet reached the size of the critical deformation  $\gamma_{cr}$ , is responsible for the fragmentation. Here, the increase in the volume of overheated liquid, when the droplet oblates in the direction perpendicular to the direction of propagation of laser radiation, turns out to be important.

In what follows, let us consider the characteristic times of the process, i.e., the time of effervescence of a homogeneous layer  $t_{\rm ex}$  (see Ref. 3), the time of reaching the critical deformation  $t_{\rm cr}$ , after which the fragmentation due to the hydrodynamical instability occurs ( $\gamma_{\rm cr} = \gamma(t_{\rm cr})$ ), and finally, the time of reaching an absorption uniformity  $t_{\rm un}$  when the transverse size of the particle becomes comparable with  $\alpha_{\rm r}^{-1}$ .

The question about which of these two processes of fragmentation of the deformed particle – the hydrodynamical instability or explosive effervescence – will predominate, is determined by the ratio of the deformation rate and the heating rate, which, in their turn, depend on the particle size and the energy parameters of the radiation. The dependence of  $t_{\rm cr}$  and  $t_{\rm un}$ , calculated based on Eq. (4) on the particle size for different values of the parameter  $J_{\rm p} = \alpha_{\rm n} \omega_{\rm p} / \rho_{\rm 1} C_{\rm p} t_{\rm p}$  characterizing the rate of heating where  $\omega$  is the total energy density in the radiation pulse with the width  $t_{\rm p}$ , and  $C_{\rm p}$  is the isobaric heat capacity of liquid, are shown in Fig. 3. The data analysis shows that, when the rate of energy influx  $J_{\rm p}$  and the droplet size  $a_0$  are different, various fragmentation regimes can occur.



FIG. 3. The characteristic times  $t_{\rm cr}$  (1),  $t_{\rm ex}$  (2), and  $t_{\rm un}$  (3) of the process of fragmentation of the drop vs its size at  $J_p = 5 \cdot 10^9 (1-3)$  and  $2 \cdot 10^9 K/s (1'-3')$ .

At  $t_{\rm cr} \ll t_{\rm ex}$  (10  $\mu{\rm m}~a_0$  25  $\mu{\rm m}$ ), the hydrodynamical fragmentation regime prevails. Here, the rate of deformation of the drop is much higher then the rate of the droplet heating up to the temperature of the explosive effervescence. For the drop with  $a_0$  25 µm, when  $t_{\rm ex} \ll t_{\rm cr}$  and  $t_{\rm un}$ , a fragmentation of the front layers as a result of their explosive effervescence is typical of the process. In the intermediate region of the particle size ( $a_0 \simeq 25 \ \mu m$ ) when the rates of deformation and heating are close in value, the fragmentation mechanisms compete  $(t_{\rm cr} \sim t_{\rm ex})$ . If the condition  $t_{\rm un} \ll t_{\rm cr}$ , and  $t_{\rm ex}$  is satisfied, the transverse size of the droplet by the time of its fragmentation becomes comparable with the absorption length, as a result, the entire volume of the particle already becomes substantially overheated and its explosive effervescence can occur. The small particles "fragments" formed due to the fragmentation have an elevated temperature and can repeatedly effervescence virtually without any additional energy consumption.

Let us consider in more detail the fragmentation of the particle in the case of hydrodynamical instability. In this case it is very important to establish the size of the small drops – "fragments", formed due to hydrodynamical fragmentation as well as the dependence of particle size on the energy of heating radiation and the size of the initial drops. Let us estimate the effective size of the fragments  $\overline{a}_k$  using the balance relation for the energy. It is obvious that the total energy  $W_0$  stored by the deformed particle with an account of the outflow of those fraction of the energy  $K_{\rm m}$  which was spent on light–reactive motion of the center of mass in the course of fragmentation will be transformed into the kinetic energy  $K_{\rm N}$  and surface energy  $\Pi_{\rm N}$  of the fragments  $N_{\rm k}$ 

$$W_0 = \int_0^t dt' \int_S (\mathbf{vn}) p \, dS' + \Pi \, (t=0) - K_M = K_N + \Pi_N.$$

We employ the approximation of monodisperse fragments for the estimates

$$K_N = \frac{2\pi}{3} \rho_1 N_k a_k^{-3} v_k^{-2}; \quad \Pi_N = 4\pi\sigma a_k^2 \cdot N_k ,$$

where  $\overline{v}_k$  is the mean velocity of motion of the fragments. It can be estimated from the relation  $\overline{v}_k \simeq (da/dt) \Big|_{\gamma=}$ . The calculations showed that in the interval of the deformation rates under consideration  $K_N \ll \Pi_N$ . Taking this into account as well as the fact that  $N_k = 3M'_0/4\pi\rho_1 \overline{a}_k^3$ , we obtain for the

degree of fragmentation of the drop  $d = a_0 \sqrt{a_k}$ 

$$d = \frac{a_0 \,\rho_1 W_0}{3M_0' \,\sigma} \,. \tag{6}$$

It follows from Eq. (6) that the degree of fragmentation d is proportional to the impact energy given to the drop, which in its turn depends on the rate of heating  $J_{\rm p}$ . Since both the numerator and denominator of Eq. (6) are proportional to  $a_0^3$ , the dependence  $d(a_0)$  is weak (in contrast to the case of uniform explosion, when  $d \sim a_0^{-3}$ .

The result of calculation of  $d(J_p)$  based on Eq. (6) is shown in Fig. 4 (solid line). It can be seen from the figure that the degree of fragmentation of the drops increases as the rate of energy influx increases. This fact was also established in the experimentally in Ref. 7, the results are shown in Fig. 4. Since a cw CO<sub>2</sub> laser was used in Ref. 7, in this case  $J_{\rm p} = \alpha_0 I_0 / \rho_1 C_{\rm p}$ , where  $I_0$  is the radiation intensity.



FIG. 4. The degree of fragmentation of the drops with  $a_0 = 10$  (1), 15 (2), 20 (3), and 25 µm (4) vs the rate of heating according to Ref. 7. The solid curve denotes the result of theoretical calculation.

The analysis of the data shown in Fig. 4 indicates that at low rates of the influx of the light energy, the fragments have large size, which provides, when they are further exposed to pulse of laser light and the energy density in the pulse is quite high, for their effective heating up to the explosive effervescence. The experimental investigations described in Ref. 14 also indicate such a double explosion of the large drops ( $a_0 \le 23 \mu m$ ).

For the large particles  $a_0 \gg \alpha_{\alpha}^{-1}$  when the unequality  $t_{\rm ex} \ll t_{\rm cr}$  and  $t_{\rm un}$  is valid, the regime of a layer–by–layer explosion can occur. In this case one can neglect the effect of deformation, and the fragmentation can be treated as a sequence of the phases of heating, effervescence, and separation of the absorbing surface layer. An experiment with such a "pulsating" explosion was described in Ref. 5.

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