RESPONSE OF THE LIQUID DROP SURFACE TO THE PONDERMOTIVE FORCE INDUCED BY A LASER BEAM

Yu.E. Geints and A.A. Zemlyanov

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk
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In this paper we present numerical investigation into the problem on deformations and oscillations of the drop surface under the action of pondermotive force induced by an intense light field. The simulations have been done for a variety of drop size. As a result, it is shown that in small droplets the deformations are induced across a laser beam while being induced along the beam in big drops. We also show in this paper that inhomogeneities of the optical field in big drops may produce strong local perturbations of the surface.

INTRODUCTION

It is known that the volume and surface pondermotive forces act on a dielectric placed in the electromagnetic field.[1] This phenomenon can lead to some nonlinear effects at the interaction of an intense laser radiation and weakly absorbing particles. The effect of stimulated Mandelshtam-Brillouin scattering (SMBS) occurring due to excitation of acoustic waves in a droplet volume is the most important among them. Excitation of the surface (capillary) waves in droplets was investigated theoretically and experimentally in a number of papers.[4,7,8] The aforementioned surface effect can initiate the destruction of aerosols[4,8] or cause the surface Raman scattering of light.[5,6] It is necessary to take it into account when analyzing the threshold conditions of excitation of the stimulated Raman scattering (SRS) and SMBS connected with the resonance properties of transparent spheres.[13,14] The study of laser excitation of disturbances of the droplet surface is of practical interest for the problems of atmospheric optics.[12] However, it was solved in the theoretical papers cited above only for particles of a narrow size range, namely, for small particles. This fact does not allow one to apply the results of such investigations to the particles of arbitrary size.

The main goal of this paper is to solve the problem on excitation of capillary waves on the surface of liquid droplets of arbitrary size under the action of laser radiation of different intensity.

BASIC RELATIONSHIPS

General statement of the problem on deformations of a transparent droplet in the light field includes the hydrodynamical equations of the viscous incompressible liquid[2] written taking into account the pondermotive forces

$$\frac{\partial \boldsymbol{v}}{\partial t} + (\boldsymbol{v}\nabla)\boldsymbol{v} = \nu\Delta\boldsymbol{v} - (1/\rho)\,(\nabla p - \mathbf{f}_E), \quad \mathrm{div}\,\boldsymbol{v} = 0, \quad (1)$$

where t is time; $\boldsymbol{v}$, p, ρ, η and $\nu = \eta/\rho$ are the velocity, pressure, density, dynamical viscosity and kinematic viscosity of the liquid, respectively. Volume density of the pondermotive forces is determined by the relationship[1]:

$$\mathbf{f}_E = \frac{1}{8\pi}\,\rho\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T \nabla E^2, \quad (2)$$

ε is the dielectric constant of the liquid, E is the strength of the electric field inside the particle, and T is temperature.

The kinematic boundary condition on the free surface of a droplet, relating the deformation to velocity, is expressed as follows[2]:

$$\frac{\mathrm{d}F}{\mathrm{d}t} = \frac{\partial F}{\partial t} + \boldsymbol{v}\nabla F = 0. \quad (3)$$

Boundary condition for tensions on the surface, i.e. the dynamic boundary condition, is given in the form[2]

$$\left\{p - \frac{\rho}{8\pi}\frac{\partial\varepsilon}{\partial\rho}\boldsymbol{E}^2 - p_1 - \alpha\left(\frac{1}{R_1} + \frac{1}{R_2}\right) + f\right\} n_i = \\ = \eta\left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\right) n_k. \quad (4)$$

Here $F(\mathbf{r}_1, t) = 0$ is the equation of the deformed liquid surface; $\mathbf{r}_1$ is the radius-vector of a point of the perturbed surface; p_1 is the external (atmospheric) pressure; α is the surface tension coefficient of the liquid; R_1 and R_2 are the main radii of the surface curvature; $\mathbf{n}$ is the external normal to the droplet surface; x_i are the coordinates; and

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$$f = \frac{\varepsilon - 1}{8\pi}\,[(\varepsilon - 1)\,(\mathbf{En})^2 + E^2]$$

is the jump of the normal component of the electromagnetic field strength on the liquid surface.[1] One should take into account, in Eqs. (1) and (4), only low frequency components relative to the frequency of the incident light field.

The statement of the problem in the integral form on hydrodynamical effects in nonabsorbing liquid particle in the intense light field is based on the energy conservation law. It is known that the change of kinetic energy in the field of mass forces is given by the expression[2]

$$\frac{\partial}{\partial t}\int\limits_V \frac{\rho v^2}{2}\,\mathrm{d}V = -\int\limits_S \left[\rho\boldsymbol{v}\left(\frac{v^2}{2} + \frac{p}{\rho}\right) - (\boldsymbol{v}\sigma)\right]\mathrm{d}\mathbf{S} - \\ - \int\limits_V \sigma_{ik}\,\frac{\partial v_i}{\partial x_k}\,\mathrm{d}V - \int\limits_V \mathbf{f}\boldsymbol{v}\,\mathrm{d}V. \qquad (5)$$

Here $\boldsymbol{\sigma}$ is the viscous tension tensor; V is the volume, S is the deformed liquid surface; $\mathrm{d}S = \mathbf{n}\mathrm{d}S$.

Subsequent study will be performed under conditions of small deformations of the surface of weakly viscous incompressible liquid. The condition of small deformations means that $|\xi| = |\mathbf{r}_1 - \mathbf{r}_0| \ll 1$, where $\mathbf{r}_0$ is the vector of the point on nonperturbed droplet surface; ξ is the vector of the surface displacement. The flow inside the droplet can be considered as potential in a low viscosity approximation ($\nabla\times\boldsymbol{v} = 0$). The exception is the boundary layer area with the thickness

$$l_\mathrm{b} = r_0(\mathrm{Re})^{-1/2},$$

where $r_0 = |\mathbf{r}_0|$ is the droplet radius and Re is the Reynolds number.[2] The velocity of liquid for the potential flow has the form

$$\boldsymbol{v} = \nabla\Phi,$$

and the vector of the displacement of the droplet surface is

$$\xi(\theta, \varphi) = \int\limits_0^t \nabla\Phi\,\mathrm{d}t',$$

where Φ is the velocity potential, satisfying the Laplace equation in the case of incompressible liquid

$$\Delta\Phi = 0.$$

Under the conditions when $l_b << r_0$, all terms in Eq. (5) can be determined based on the ideal liquid flow approximation. This is possible, because the velocity and its derivatives for the flow of liquid with free surface do not vary significantly in the boundary layer.[2] Thus, when calculating the corresponding volume integrals in Eq. (5) one can ignore the difference between velocities and their derivatives in the vortex flow area and in the area of the ideal liquid flow in the case of the small size area. The correct use of the potential flow approximation for calculating the surface integral in Eq. (5) is provided by the condition of small deformations of the droplet surface.

The sum of the inverse radii of the droplet surface curvature for small displacements can be presented in spherical coordinates as follows[2,10]:

$$\frac{1}{R_1} + \frac{1}{R_2} = \frac{2}{r_0} + \frac{1}{r_0^2}\,(L^2 - 2)\,\xi(\theta, \varphi),$$

where $\mathbf{L} = -\,i\mathbf{r}\times\nabla = -i\left[\boldsymbol{\theta}\,\frac{1}{\sin\theta}\frac{\partial}{\partial\theta} - \boldsymbol{\phi}\,\frac{\partial}{\partial\phi}\right]$ is the angular momentum operator; $\mathbf{L}^2 = \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\times\left(\sin\theta\,\frac{\partial}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\phi^2}$; $\xi = |\xi|$; r, θ and φ are the spherical coordinates.

Thus, taking into account the boundary conditions (4), within the framework of the assumptions performed, one obtains the following boundary condition from Eq. (5):

$$\frac{\partial}{\partial t}\int\limits_S \frac{\rho\Phi\boldsymbol{v}}{2}\,\mathrm{d}\mathbf{S} + \int\limits_S \left(\frac{2\alpha}{r_0} + p_1\right)\boldsymbol{v}\mathrm{d}\mathbf{S} + \\ + \alpha\int\limits_S \frac{(\boldsymbol{L}^2 - 2)}{r_0^2}\,\xi\boldsymbol{v}\mathrm{d}\mathbf{S} + \eta/2\int\limits_S \nabla v^2\,\mathrm{d}\mathbf{S} = \int\limits_S f\,\boldsymbol{v}\mathrm{d}\mathbf{S}. \qquad (6)$$

By writing the velocity potential (as harmonic function) in the form of expansion in terms of spherical functions $\left(\frac{r}{r_0}\right)^l Y_{ln}(\theta, \varphi)$:

$$\Phi(r, \theta, \varphi) = \sum_{ln} \Phi_{ln}\left(\frac{r}{r_0}\right)^l Y_{ln}(\theta, \varphi),$$

where $Y_{ln}(\theta, \varphi)$ is the spherical harmonic, one obtains the system for stimulated oscillations of the droplet surface from Eq. (6) by differentiation with respect to time:

$$\frac{\mathrm{d}^2\Phi_{ln}}{\mathrm{d}t^2} + \frac{2}{t_l}\frac{\mathrm{d}\Phi_{ln}}{\mathrm{d}t} + \Omega_l^2\,\Phi_{ln} = \frac{1}{\rho}\frac{\mathrm{d}f_{ln}}{\mathrm{d}t}, \qquad (7)$$

where $f_{ln} = \int\limits_0^{2\pi}\mathrm{d}\varphi\int\limits_0^{2} f(t,\boldsymbol{r}_0)\,Y^*_{ln}(\theta,\varphi)\sin\theta\,\mathrm{d}\theta$;

$t_l = \dfrac{r_0^2}{2\nu\,(2l + 1)\,(l - 1)}$ is the characteristic time of damping of the oscillations due to viscosity forces; $\Omega_l = \left[\dfrac{l\,(l - 1)\,(l + 2)\alpha}{\rho r_0^3}\right]^{1/2}$ is the natural (Rayleigh) frequency of the droplet oscillations. The asterisk means the complex conjugation. Equation (7) is completed by the initial condition $\Phi_{ln}(0) = \mathrm{d}\Phi_{ln}(0)/\mathrm{d}t = 0$.

The following equation for coefficients of the expansion of the surface displacement into a series over spherical functions $\xi(t, \theta, \varphi) = \sum_{ln} \xi_{ln}(t)\, Y_{ln}(\theta,\varphi)$ follows from Eq. (7)

$$\frac{d^2\xi_{ln}}{dt^2} + \frac{2}{t_l}\frac{d\xi_{ln}}{dt} + \Omega_l^2\, \xi_{ln} = \frac{l\, f_{ln}}{\rho r_0}, \tag{8}$$

It differs from Eq. (7) in its right-hand side. Initial conditions for Eq. (8) have the form $\xi(0) = \frac{d\xi(0)}{dt} = 0$.

It should be noted that the equations (7) and (8) of analogous form can be obtained only from the boundary condition (4) without the use of integral equation (5), even if one assumes that the liquid flow is the potential one. The pressure in the liquid is related to the potential Φ:

$$p = \frac{\rho}{8\pi}\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T E^2 - \rho\,\frac{\partial\Phi}{\partial t}.$$

However, the coefficient at the first derivative $(2/t_l)$, characterizing the damping in the system, is determined inaccurately. Therefore, in Ref. 7, where the equation of oscillation was derived based on Eq. (4), the corresponding coefficient was introduced based on phenomenological considerations allowing for the law of mechanic energy dissipation at weak oscillations of the liquid ball surface.[3]

One should give some comments concerning the form of the stimulating force in Eq. (8). The matter is that in some papers devoted to the problem of the liquid flow in droplets at the presence of the light fields (for example, Refs. 6, 9 and 10) the expressions for the forces are given not accurately. Thus, the limits of applicability of the estimates and calculations performed in these papers for the cases, when the effect of pondermotive forces is essential, are not clear,[6,10] or are valid only within certain range of liquid particles size.[9]

The equations for weak oscillations (7) and (8) are obtained in most rigorous way in this paper. The case is a limitation, when weakly viscous liquid approximation is not valid. If one estimates $\mathrm{Re} \sim \Omega_2\, r_0^2/\nu$ the condition of smallness of the boundary layer is broken, for example, for water droplets with the size $r_0 \le 0.03$ μm. To study such oscillations, one should use other methods (see Ref. 11).

To solve Eq. (8), it is necessary to know the form of the function $f(\mathbf{r})$, based on the Mie solution for the light field inside the droplet. It is known that such a solution for the sphere and the linearly polarized wave incident on it has the form enabling the field to be presented as a series over spherical harmonics:

$$\boldsymbol{E}(r, \theta, \varphi) = \frac{E_0}{2kr}\sum_{l=1}^{\infty}(-i)^{l+1}[b_e(x_a)\,\mathbf{M}_{l,1}(\theta, \varphi)\,\psi_l(r) + \frac{1}{k}\,c_m(x_a)\,\nabla\times\mathbf{M}_{l,1}(\theta, \varphi)\,\psi_l(r)] + \mathrm{c.c.}, \tag{9}$$

where b_e and c_m are the amplitudes of the partial harmonics (Mie coefficients[15]); x_a is the diffraction parameter of the particle; k is the wave number inside the particle; $\mathbf{M}_{l,m}(\theta, \varphi) = \frac{i}{[l(l+1)]^{1/2}}\,\mathbf{L}\,Y_{l,m}(\theta, \varphi)$ are the “evenB and “oddB spherical vector-harmonics[15]; $E_0 = |\mathbf{E}_0|$; $\mathbf{E}_0$ is the light field incident on the particle.

It is possible to solve the problem for the case of the uniform electromagnetic field inside the droplet. Then

$$\mathbf{E} = \frac{3}{\varepsilon + 2}\,\mathbf{E}_0;\; f = \frac{\varepsilon - 1}{8\pi}\,[(\varepsilon - 1)\,E^2\cos^2\theta + E^2],$$

and the coefficients of expansion of the function f over spherical functions are

$$f_{ln} = 6\sqrt{\frac{4\pi}{5}}\,\frac{E_0^2}{8\pi}\,\frac{(\varepsilon - 1)^2}{(\varepsilon + 2)^2}\,\delta_{2l}\,\delta_{0n}. \tag{10}$$

Here δ_{mk} is the Kronecker delta-function ($\delta_{mk} = 1$ at $m = k$, and $\delta_{mk} = 0$ at $m \ne k$). As follows from Eq. (10), only elliptical oscillations occur in small droplets.

RESULTS OF NUMERICAL CALCULATIONS

The system of equations (8) was numerically solved in two stages. At the first stage, the values of coefficients $f_{ln}(t, \theta, \varphi)$ were calculated using the Gauss quadrature and representation of the internal field in the form (9). Then the differential equations (8) were solved using the Runge-Kutt difference scheme of the fourth order. The shape of incident laser pulse was selected in the form $I(t) = I_0 t/t_p \exp(-t/t_p)$, where I_0 and t_p are the peak intensity and duration of the pulse, respectively.

The form of the function characterizing the spatial distribution of stimulating force for water droplets of different size is shown in Fig. 1. In computations we considered only azimuthal-symmetric oscillations of a droptet surface (symmetric in an angle φ)

As follows from Fig. 1, the stimulating force has maxima on the droplet poles at Mie parameter $x_a < 1$, and they move to the equator zones, to the spherical coordinates $\theta = \pi/2$, $\varphi = 0$ and $\varphi = 2\pi$, respectively, for $x_a > 1$.

The temporal behavior of the displacement of water droplet surface $\xi(t)/\xi_{\max}$, illustrating this effect, is shown in Fig. 2 for two directions $\theta = 0$ and $\theta = \pi/2$.

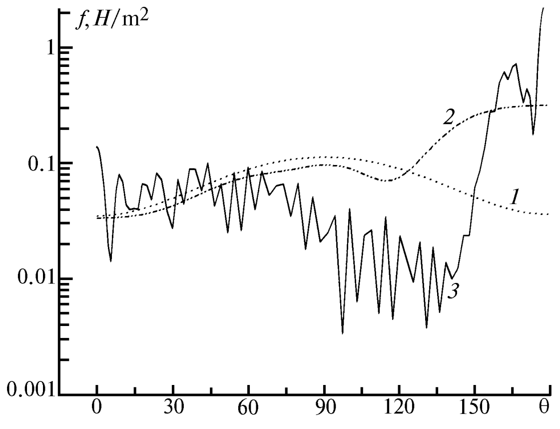

FIG. 1. The density of the normal component of the resultant pondermotive force on the surface of water droplets with the radius r_0 = 0.03 (*1*), 0.3 (*2*), *and* 3 μm (*3*) *as a function of polar angle* θ. *Z-axis is directed along the direction at* θ = 0–180°. *The incident radiation intensity was taken to be* $I_0 = 10^4$ W/cm².

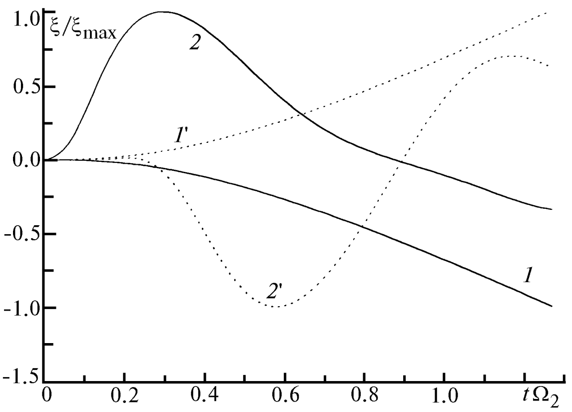

FIG. 2. Relative displacement of the surface of water droplets with r_0 = 0.03 (*1, 1′*) *and* 3 μm (*2, 2′*) *as functions of time. Two directions are considered: along, at* θ = 0°, (*solid curve*) *and across,* θ = 90°, *the beam* (*dashed line*) *with the intensity* $I_0 = 10^8$ W/cm², *and pulse duration* $t_p = 10^{-7}$ sec.

It is well seen from the figure that the initial phase of oscillations of small and big droplets is different by π/2. Small particles are deformed in the direction perpendicular to the incident beam of radiation, while the big ones are deformed along the beam. Amplitudes of the oscillations are also different: $\xi_{max}/r_0 = 3\cdot10^{-5}$ for r_0 = 0.03 μm and $\xi_{max}/r_0 = 10^{-3}$ for r_0 = 3 μm at the intensity of the incident radiation pulse $I_0 = 10^8$ W/cm² with the pulse duration $t_p = 10^{-7}$ sec and wavelength λ = 0.53 μm.

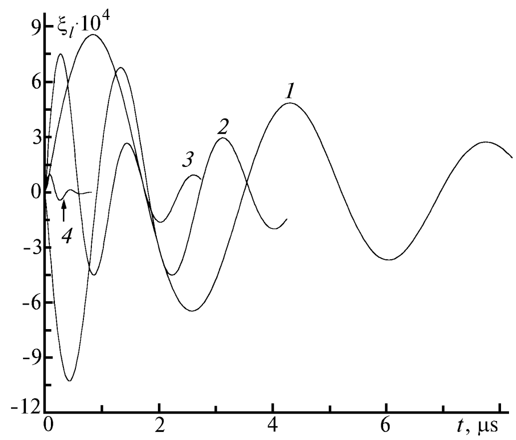

FIG. 3. Temporal behavior of the partial harmonics of a water droplet with r_0 = 3 μm *and indices* l = 2 (*1*), 3 (*2*), 4 (*3*), *and* 9 (*4*).

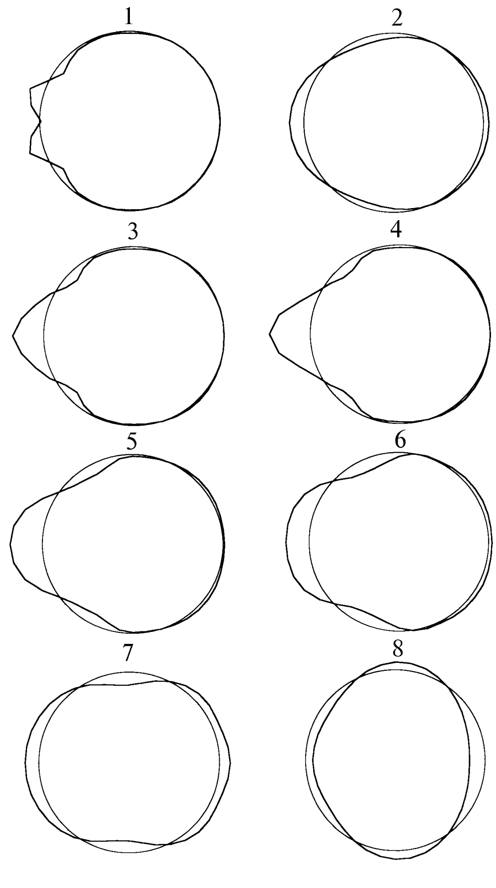

FIG. 4. The shape of the water droplet surface (main cross section) with r_0 = 25 μm *at different time moments since the beginning of the action of radiation* ($I_0 = 10^9$ W/cm²; $t_p = 10^{-7}$ sec, *direction of incidence from left to right):* t = 50 (*1*), 80 nsec (*2*), 0.2 (*3*), 0.5 (*4*), 5 (*5*), 15 (*6*), 30 (*7*) *and* 40 μsec (*8*).

Temporal behavior of the amplitudes of partial oscillations of a water droplet with $r_0 = 3$ μm, $I_0 = 10^9$ W/cm^2, and $t_p = 10^{-7}$ sec is shown in Fig. 3. As follows from Eq. (7), damping of the high frequency harmonics occurs quite quickly, and their influence on the entire pattern of the droplet deformation is noticeable only at the initial stage of oscillations. The calculated shape of the droplet surface with $r_0 = 25$ μm (main cross section) is shown in Fig. 4 at different time moments. The radiation pulse had the same parameters as in the previous figure. As follows from the figure, local deformations of the water droplet surface, initially caused by the high frequency oscillation modes, are subsequently developed to the elliptic oscillations at the fundamental frequency $\Omega_2 = \left[\frac{8\alpha}{\rho r_0^3}\right]^{1/2}$. In addition, quite strong deformations ($\xi_{max}/r_0 \sim 0.3$) of the droplet surface are observed. In some cases they can lead to its destruction.[8]

CONCLUSION

Thus, we have formulated the equations of small deformations of the surface of a weakly viscous transparent liquid particle under the effect of pondermotive forces of the light field. Essentially different deformations of small and large particles (from the viewpoint of the Mie theory) were revealed from the numerical solutions of the equations. It is shown that the deformations of small particles are initiated perpendicularly to the incident beam, while the deformations of large particles are along the laser beam. The possibility of quite strong deformations to occur under certain conditions is established theoretically. They can lead to destruction of a particle as well as to significant scattering of incident light on the particle surface.

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