SURFACE EFFECT OF THE LASER RADIATION PONDEROMOTIVE ACTION ON LIQUID PARTICLES. PART 2. RESONANCE BUILD-UP OF OSCILLATIONS. SURFACE RAMAN SCATTERING

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The problem of the resonance build-up of surface oscillations in transparent liquid weakly viscous particles of an arbitrary size under the action of modulated laser radiation is investigated theoretically. A relation is found between the amplitude of the drop surface deformation and the modulation frequency of acting radiation for different values of a particle radius. The characteristics of forced oscillations at resonance excitation are studied. The width of the resonance characteristical curve of surface oscillations was found to increase as the particle size decreases and the liquid viscosity increases. The problem of light scattering on small surface oscillations of liquid particles is numerically investigated. It is shown that the maximum modulation of scattered light at the frequency of the fundamental harmonic of surface waves occurs in the direction normal to the incident radiation, as well as in the direction of the primary rainbow.

INTRODUCTION

An intense light field acting on a liquid dielectric particle can give rise to deformation of the particle surface.[1] The ponderomotive forces are responsible for this phenomenon. The magnitude of the ponderomotive forces is proportional to the squared electric field intensity of a light wave inside droplets.[6] Variation of the intensity of radiation acting on a liquid particle produces relaxation oscillations of the particle surface. When using the amplitude-modulated laser radiation, modulation frequency of which coincides with one of the natural vibrational frequencies of a droplet, the resonance build-up of these oscillations is possible.[2,5]

The occurrence of the resonance oscillation mode of the surface of liquid particles, the size of which is small as compared to the radiation wavelength ($a_0 << \lambda$, where a_0 is the droplet radius, λ is the wavelength of incident radiation), was reported in the literature.[5] Reference 2 derives the analytical expression for the amplitude of resonance oscillations and gives the estimates for the case of homogeneous distribution of the light field inside droplets. However, for the case of radiation interaction with optically large particles ($a_0 >> \lambda$) under the conditions, when the essentially inhomogeneous distribution of an electromagnetic field takes place, the quantitative results were not obtained.

As known, the natural frequency of mechanical oscillations of a spherical droplet is uniquely connected with the droplet size and physical properties of a liquid.[7] Acting by laser radiation upon a polydisperse drop aerosol and varying the modulation frequency of the acting radiation, we can, in principle, provoke a resonance response of one or another group of particles. Thus, when measuring the natural frequency of droplets in an experiment, it becomes possible to determine the size distribution function of aerosol particles, as well as the viscosity and the surface tension coefficient of a liquid.

This procedure was approved experimentally in Ref. 4, and it has demonstrated the real possibility to obtain the information about the drop aerosol microstructure by irradiating the aerosol with a modulated laser radiation.

However, the direct interpretation of the results obtained in Ref. 4 is problematic due to the lack of *a priori* information on characteristics of resonance ponderomotive oscillations of droplets in an intense light field. The goal of this investigation is to obtain the dependence of the amplitude of resonance surface oscillations of a liquid particle of an arbitrary size on the temporal parameters of acting radiation. The other important problem is to derive and study numerically the expression for the intensity of a light wave scattered on an oscillating droplet, what can serve as a basis for simulating the spectroscopy effect of aerosol particle sizes, which is based on phenomenon of the Raman scattering of light on droplet surface oscillations.

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RESONANCE EXCITATION OF SURFACE OSCILLATIONS OF A LIQUID TRANSPARENT PARTICLE BY LASER RADIATION

The general statement of the problem on deformation of a liquid transparent droplet in a light field is given in Refs. 1–3. It includes the dynamics equations of incompressible liquid written with regard for the ponderomotive forces.

The spatiotemporal evolution of droplet surface deformations is given by the dynamic boundary condition, which is in essence a modified analog of the Laplace formula[1]:

$$\left\{p - \frac{\rho_a}{8\pi}\left(\frac{\partial\varepsilon_a}{\partial\rho_a}\right)_T E^2 - p_1 - \sigma\left(\frac{1}{R_1} + \frac{1}{R_2}\right) + f\right\}\mathbf{n}_i =$$

$$= \eta\left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\right)\mathbf{n}_k. \tag{1}$$

Here p, T, σ, ρ_a, and η are pressure, temperature, the surface tension coefficient, the density, and the dynamic viscosity of a liquid, respectively; p_1 is the outer (atmospheric) pressure; R_1 and R_2 are the major radii of surface curvature; $v_{i,k}$ are the liquid velocity components; ε_a is the absolute dielectric constant of a particle matter; $E(\theta, \varphi)$ is the electric field intensity on the particle surface; $x_{i,k}$ are the coordinates; $\mathbf{n}$ is the vector of the external normal to the particle surface. The value of a step of the normal component of electric field tension on the particle surface, determining the surface density of the ponderomotive forces, is given by the following expression[6]:

$$f(\theta,\varphi) = \frac{\varepsilon_a - 1}{8\pi}\left[(\varepsilon_a - 1)\,(\mathbf{E}(\theta,\varphi)\,\mathbf{n})^2 + E^2(\theta,\varphi)\right],$$

where θ, φ are the spherical coordinates.

From Eq. (1) or from the equations of energy balance of a deformed particle[7] we can derive the equation of small oscillations of weakly viscous liquid.[2,3] This equation serves as a base for theoretical analysis of droplet behavior in the laser radiation field.

The value of the particle surface shift is represented as the expansion in terms of the spherical functions:

$$a(t,\theta,\varphi) - a_0 = \xi(t,\theta,\varphi) = \mathrm{Re}\left\{\sum_{l;\,n} \xi_{ln}(t)\; Y_{ln}(\theta,\varphi)\right\}, \tag{2}$$

where ξ_{ln} are the expansion coefficients of the particle surface shift; a_0 is the radius of an unperturbed droplet; $Y_{ln}(\theta,\varphi)$ are the spherical functions.

Now we consider the oscillations symmetric by a spherical angle φ (this assumption is justified since the field intensity inside a droplet irradiated by a plane wave is also symmetric about the angle φ). In this case, for the expansion coefficients of the surface shift the following set of equations is valid

$$\frac{\mathrm{d}^2\xi_l}{\mathrm{d}t^2} + \frac{2}{t_l}\frac{\mathrm{d}\xi_l}{\mathrm{d}t} + \Omega_l^2\,\xi_l = \frac{l\,f_l(t)}{a_0\,\rho_a}, \quad l = 2, 3, \ldots, \tag{3}$$

where

$$f_l(t) = \int_0^\pi f(t, \theta)\,Y^*_{l0}(\theta)\sin\theta\,\mathrm{d}\theta$$

are the coefficients of expansion of the function $f(t, \theta)$ in terms of spherical harmonics;

$$t_l = a_0^2/[2(2l + 1)(l - 1)\nu] \tag{4}$$

is the characteristic damping time of oscillations due to viscous forces; $\nu = \eta/\rho_a$ is the kinematic viscosity of a liquid;

$$\Omega_l = \sqrt{l\,(l + 2)\,(l - 1)\,\sigma/\rho_a\,a_0^3} \tag{5}$$

are the natural oscillation frequencies of a droplet.

The set of equations (3) is supplemented with the initial conditions at $t = 0$: $\xi_l = 0$, $\frac{\mathrm{d}\xi_l}{\mathrm{d}t} = 0$.

Now we consider the problem of excitation of ponderomotive oscillations in a droplet by harmonic modulated radiation. The time dependence $f_l(t)$ is given in the following form: $f_l(t) = f_l^0(a_0, \lambda)\times(1 - \cos\Omega t)$, $t \geq 0$, where $f_l^0(a_0, \lambda)$ is the time-independent coefficient determined by a specific form of the electric field on the particle surface; Ω is the modulation frequency of acting radiation. For the steady oscillations ($t \gg t_l$) from Eq. (3) we obtain the solution well-known in the theory of oscillations[8]:

$$\xi_l(\Omega) = \frac{f_l^0 l\cos\left[\Omega t + \arctan\dfrac{2t_l^{-1}\,\Omega}{\Omega^2 - \Omega_l^2}\right]}{\rho_a\,a_0[(\Omega^2 - \Omega_l^2)^2 + 4t_l^{-2}\,\Omega^2]^{1/2}}. \tag{6}$$

At resonance ($\Omega = \Omega_l$)

$$\xi_l(\Omega) = \frac{f_l^0\,l\sin\Omega t}{2\rho_a\,a_0\,\Omega_l\,t_l^{-1}}. \tag{7}$$

As noted above, values of the coefficients f_l^0 are determined by the electromagnetic field distribution on the droplet surface. For large particles, this distribution has a sharply inhomogeneous structure with a great number of peaks and a large scatter in values. That is why f_l^0 can be found only from numerical calculations.

At $(t_l\,\Omega_l)^{-2} \ll 1$, the relationship in the form of Eq. (6) determines the resonance curve with the halfwidth

$$\Delta_l \approx 2\sqrt{3}/t_l. \tag{8}$$

Figure 1 shows the relative amplitude $\bar{\xi} = \xi/a_0$ of steady oscillations of the droplet surface versus the modulation frequency of acting laser radiation Ω. The resonance peaks in the function $\bar{\xi}(\Omega)$ correspond to the cases of coincidence of the modulation frequency and the droplet natural frequencies. In this case, the shift amplitude is maximal for the fundamental vibrational mode $l = 2$.

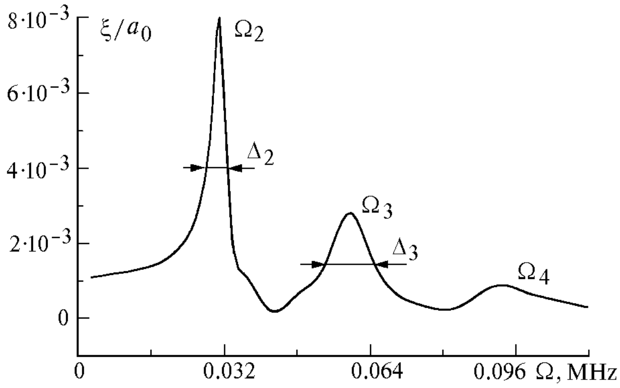

FIG. 1. The relative amplitude of steady oscillations for a water droplet with $a_0 = 25$ μm *vs. the modulation frequency of the continuous laser radiation with* $I_0 = 10^5$ W/cm^2. *The maximum values correspond to the droplet resonance frequencies of different orders.*

For small droplets ($a_0 \ll \lambda$) the internal light field can be considered practically homogeneous. In this case,

$$\mathbf{E}(a_0) = \frac{3}{2 + \varepsilon_a}\,\mathbf{E}_0;$$

$$f(\theta) = \frac{\varepsilon_a - 1}{8\pi}\Big[(\varepsilon_a - 1)E_0^2 \sin^2\theta + E_0^2\Big],$$

where $\mathbf{E}_0$ is intensity of the light field incident on a particle.

Thus, for the coefficients f_l^0 we obtain[2]

$$f_l^0 = -\frac{3E_0^2}{2\sqrt{5\pi}}\,\frac{(\varepsilon_a - 1)^2}{(\varepsilon_a + 2)^2}\,. \tag{9}$$

Substituting Eqs. (4), (5), and (9) into Eq. (7) we obtain

$$\xi_2 = \frac{3E_0^2}{40\sqrt{5\pi}}\,\frac{(\varepsilon_a - 1)^2}{(\varepsilon_a + 2)^2}\,\frac{a_0^{5/2}}{\sqrt{\sigma}\,\sqrt{\rho_a}\nu},\quad (\Omega = \Omega_2)\,. \tag{10}$$

Now we consider the situation, when a liquid particle is subject to a succession of short laser pulses with the repetition rate equal to one of the resonance frequencies of droplet oscillations. The typical form of the amplitude-frequency characteristics close to the fundamental oscillation mode Ω_2 for the particles of a different radius is given in Fig. 2. Calculations of the function $\xi(\Omega)$ were made numerically using the set of equations (3) with the Runge-Kutta numerical differentiation method of the fourth order.

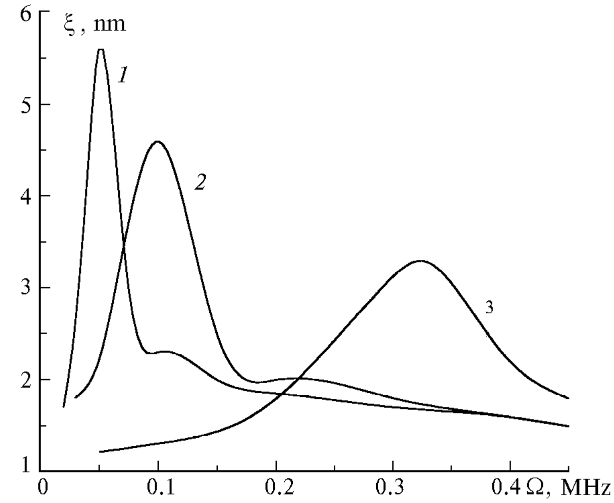

FIG. 2. The amplitude of steady oscillations vs. the pulse repetition rate of the laser radiation at $I_0 = 10^7$ W/cm^2 and $t_\mathrm{p} = 10$ ns *for different size of a droplet:* $a_0 = 15$ *(1)*, 11 *(2), and* 5 μm *(3).*

Figure 2 shows that as the particle size increases, the resonance curve becomes narrower with simultaneous increase in its Q-factor. This is connected with the growth of the characteristic damping time of oscillations due to viscous strengths. This result well agrees with Eq. (8).

The analytical solution of the problem under study for the sequence of laser pulses is complicated. Therefore, for qualitative study of the dependence of the droplet oscillation amplitude on the power and time parameters of pulse action, let us consider one pulse with the time profile given in the form

$$I(t) = I_0\,\frac{t}{t_\mathrm{p}}\,\exp\left\{-\frac{t}{t_\mathrm{p}}\right\}. \tag{11}$$

Here I_0 and t_p are the pulse peak intensity and the characteristic duration, respectively. Having substituted Eq. (11) into the right-hand side of Eq. (3) and having solved this equation by the method of indefinite coefficients, we obtain

$$\xi_l(t) = \frac{8\pi\, l\, I_0 f_l^0}{c\, n_a\, \rho_a\, a_0\, t_\mathrm{p}}\,\frac{1}{((1/t_l - 1/t_\mathrm{p})^2 + \Omega_l^2)^2}\times$$
$$\times\Bigg[t\,\exp(-t/t_\mathrm{p})\left(\left(\frac{1}{t_l} - \frac{1}{t_\mathrm{p}}\right)^2 + \Omega_l^2\right) -$$
$$-\,2\exp(-t/t_\mathrm{p})\left(\frac{1}{t_l} - \frac{1}{t_\mathrm{p}}\right) +$$
$$+\,\exp(-t/t_l)\left(\frac{1}{\Omega_l}\left(\frac{1}{t_l} - \frac{1}{t_\mathrm{p}}\right)^2 \sin\Omega_l t\ +\right.$$
$$\left.+\,2\left(\frac{1}{t_l} - \frac{1}{t_\mathrm{p}}\right)\cos\Omega_l t - \Omega_l \sin\Omega_l t\right)\Bigg], \tag{12}$$

where c is the light speed, n_a is the refractive index of a liquid.

In the limit of "short" acting pulses ($t_p \ll \Omega_l^{-1}$, t_l) we have

$$\xi_l(t) \approx \frac{8\pi\, l\, I_0\, f_l^0\, t_p}{c n_a\, \rho_a\, a_0\, \Omega_l} \exp(-t/t_l)\sin(\Omega_l\, t),$$

$$t > t_p\,. \qquad (13)$$

It follows from Eq. (13) that the surface of an oscillating droplet shifts by the harmonic law with the natural frequency Ω_l damping exponentially in time with the constant t_l^{-1}.

In the opposite situation, i.e., in the case of "long" pulses ($t_p \gg \Omega_l^{-1}$, t_l) the solution of Eq. (12) transforms to the form

$$\xi_l(t) \approx \frac{8\pi\, l\, I_0\, f_l^0}{c n_a\, \rho_a\, a_0\, (1/t_l^2+\Omega_l^2)} \frac{t}{t_p} \exp(-t/t_p),$$

$$t \le t_p\,. \qquad (14)$$

It is clearly seen that in this case, the droplet oscillations are lacking, and the time dependence of the droplet surface shift is determined by the shape of a laser pulse.

Let us now pass to the discussion of the results obtained by numerical solution of the set of equations (3). In the numerical experiments, the characteristics of water droplet oscillations were studied under the effect of either continuous (λ = 0.69 μm) radiation modulated with the frequency Ω or a sequence of laser pulses within a wide range of the relative pulse duration $q = 2\pi/(t_p\,\Omega)$.

The time of establishment of stationary oscillations as a function of the particle size under modulated action was studied. This study has shown that as the droplet size increases, the time of establishment of the stationary mode in oscillations t_{st} increases relative to the period of the fundamental oscillation $T_2 = 2\pi/\Omega_2$. Thus, for small-sized particles ($a_0 \le 1$ μm) the natural oscillations become steady for practically a single period: $t_{st} \approx T_2$, while at $a_0 \ge 15$ μm already four or five periods are required. The same increase in the response time of oscillations is also observed for the pulse action. ” esides, in this case with the increase of the droplet size, the type of oscillations varies as well. The shape of oscillations becomes rather complex, and the harmonic components of oscillations at the resonance frequency are not clearly seen. This can be explained by the fact that with increasing the particle size, the frequency difference between adjacent modes decreases.

From Eq. (5) it follows that the frequency interval between the adjacent n and (n + 1) modes is expressed as

$$\Delta\Omega_n = \sqrt{\frac{\sigma\, n}{\rho_a\, a_0^3}} \left(\sqrt{(n+1)(n+3)} - \sqrt{(n-1)(n+2)}\right)$$

and, hence, a great number of normal modes participate in the formation of the particle surface disturbance. Thus, the deformation oscillations of large particles are the superposition of oscillations at different natural resonance frequencies.

As noted above, with the decrease in the droplet radius the resonance curve broadens with simultaneous decrease in its maximum value (see Fig. 2), what is connected with reduction of the relaxation time of oscillations due to the viscosity forces.

Let us now estimate the particle size a_0^* at which a particle in fact loses its resonance characteristics. Toward this end, we determine the conditions when the amplitudes of the particle surface shift at the resonance $\Omega = \Omega_l$ and away from it are equal. This condition is true when the value of the droplet radius $a_0 \le a_0^*$ where

$$a_0^* = 25\, \rho_a \nu^2/(2\sigma).$$

Thus, for water at $\rho_a = 10^3$ kg/m^3, $\nu = 10^{-6}$ m^2/s, $\sigma = 7.42{\cdot}10^2$ H/m we obtain $a_0^* \approx 0.2$ μm.

Figure 3 shows the maximal amplitude of forced oscillations of a water droplet with a_0 = 10 μm as a function of the radiation intensity in the pulse (t_p = 10 ns) and continuous (modulated at the frequency $\Omega = \Omega_2$) radiation modes. From this figure, we see that this dependence is linear, that also follows from the analytical solution (12). Differences in the amplitudes of the droplet surface shift at the same values of I_0 are caused by the difference in the energy transmitted by a light wave for one period in the pulse and harmonic modes.

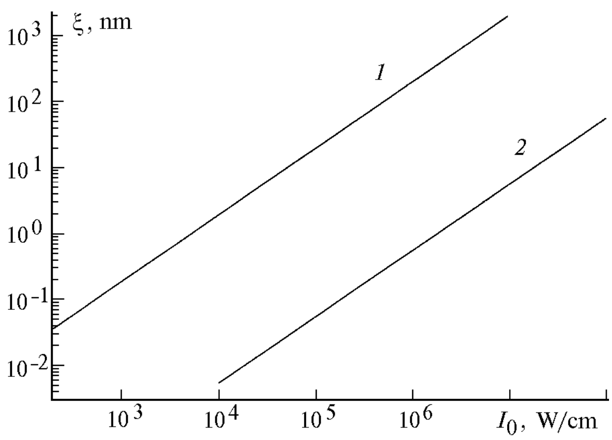

FIG. 3. The amplitude of steady oscillations of a water droplet with a_0 = 10 μm vs. the laser radiation intensity for the cases of modulated (1) with the frequency $\Omega = \Omega_2$ and pulse mode (2).

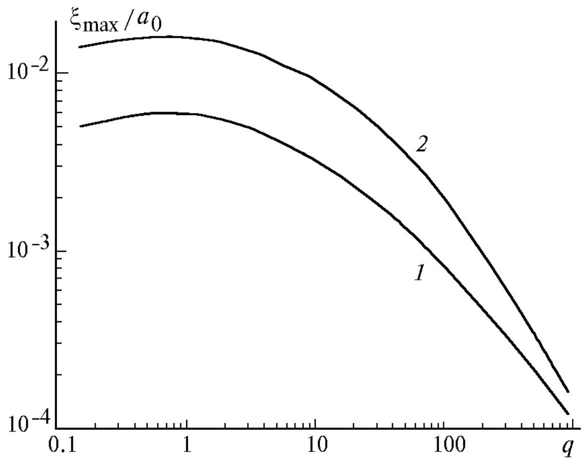

FIG. 4. The maximum shift of a surface of water particle with $a_0 = 3$ μm *(1) and* 15 μm *(2) vs. the relative pulse duration of the acting radiation at the constant peak intensity in pulses* $I_0 = 10^7$ W/cm^2.

Figure 4 shows the maximum relative shift of the surface of droplets with different radius as a function of the relative pulse duration q. The curves in this figure are plotted assuming that the peak intensity of a laser pulse I_0 is constant. With increase of the relative pulse duration, the amplitude of a surface shift decreases linearly for $q > 1$, that also follows from the analytical solutions (13) and (14); and for $q < 1$ it passes into the saturation with a small maximum observed at $q \approx 1$ what corresponds to coincidence of the pulse duration and the period of resonance oscillations at the fundamental frequency.

SURFACE RAMAN SCATTERING AT RESONANCE OSCILLATIONS OF DROPLETS

Oscillating droplets are the scatterers with a dynamically varying surface shape. It is evident that the intensity of radiation scattered on such particles will also vary in time. The question arises: What will be the amplitude of these pulsations and what are the best angles for receiving of this signal, from the viewpoint of separating out its dynamic component.

The problem under analysis may be formulated as the problem of emission of a spherical volume with the given distribution of electromagnetic field into the surrounding volume.[3] Figure 5 shows the geometry of the problem.

Let us assume that the plane electromagnetic wave $\tilde{\mathbf{E}}_0 = \mathbf{E}_0 e^{i\omega t - ik_0 z}$ is incident (in the positive direction of the axis z) on a drop, surface oscillations of which are excited by intense radiation also directed along the axis z. It is necessary to find the field at the point with the radius vector $\mathbf{r}$. We proceed from the Helmholtz equation for the vector potential $\mathbf{A}(\mathbf{r}, t)$ of an electromagnetic field

$$\nabla^2 \mathbf{A}(\mathbf{r}, t) + \frac{\omega^2}{\varepsilon_a} \mathbf{A}(\mathbf{r}, t) = - \mathbf{J}_a(\mathbf{r}, t) , \qquad (15)$$

under the condition: $\mathrm{div}\, \mathbf{A}(\mathbf{r}, t) = 0$. Here $\mathbf{J}_a = \varepsilon_a \dfrac{\partial \mathbf{E}(\mathbf{r},t)}{\partial t}$ is the density of the polarization current induced by the particle internal field.

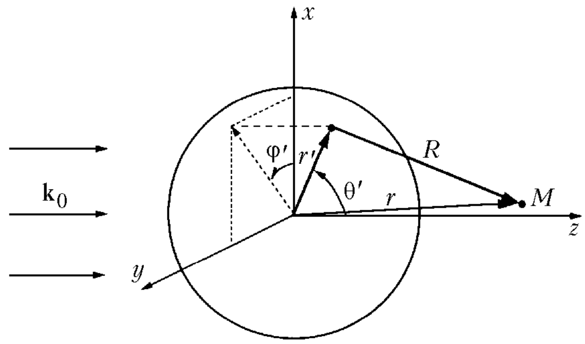

FIG. 5. Geometry of the problem. A spherical particle is at the origin of the Cartesian coordinate system. The spherical coordinates (r', θ', φ') *are also shown; M is the observation point.*

The components of the unknown electromagnetic field are expressed in terms of the vector potential as

$$\mathbf{H}(\mathbf{r}, t) = \mathrm{rot}\, \mathbf{A}(\mathbf{r}, t) \ ; \quad \mathbf{E}(\mathbf{r}, t) = - \frac{\partial \mathbf{A}(\mathbf{r}, t)}{\partial t}.$$

The solution of Eq. (15) is known from Ref. 9. Then the complete electric field $\mathbf{E}(\mathbf{r}, t)$ can be found from the ratio

$$\varepsilon_a \mathbf{E}(\boldsymbol{r},t) = \mathbf{E}_0\, e^{i\omega t - i k_0 z} +$$

$$+ \mathrm{rot\, rot} \int\limits_{V_a} \frac{(\varepsilon_a - 1)\, \mathbf{E}(\mathbf{r}',t)\, e^{ikR}}{4\pi R}\, d\mathbf{r}',$$

where k_0 is the wave number outside the particle; $k = \sqrt{\varepsilon_a} k_0$, $R = |\mathbf{r} - \mathbf{r}'|$ is the distance between the observation point and the elementary source in the particle volume. The first term in the right-hand side of the equation is the incident field, and the second term is the electric field induced by the polarized elements of the particle volume. The integration is made over the droplet volume V_a.

The further consideration is only for the scattered field. In this case in the far zone ($kr \gg 1$), we have

$$\mathbf{E}_s(\boldsymbol{r}, t) \approx \frac{k_0^2(\varepsilon_a - 1) e^{i\omega t}}{4\pi r} \int\limits_{V_a} \mathbf{E}(\mathbf{r}', t)\, e^{ikr' \cos\gamma} d\mathbf{r}'. \qquad (16)$$

Here γ is the angle between the vectors $\mathbf{r}$ and $\mathbf{r}'$, $r = |\mathbf{r}|$.

For weak perturbations of the droplet surface $\xi \ll a_0$ the integral over the volume of a deformed particle can be represented as a sum of integrals

$$\int\limits_{V_a}\mathbf{E}(\mathbf{r}', t)\, e^{ikr'\cos\gamma}d\mathbf{r}' = \int\limits_{V_{a_0}}\mathbf{E}(\mathbf{r}', t)\, e^{ikr'\cos\gamma}d\mathbf{r}' + \int\mathbf{E}(\mathbf{r}', t)\, e^{ikr'\cos\gamma}do' \int\limits_{a_0}^{a(\theta,\varphi)}\mathbf{r}'^2\, d\mathbf{r}' , \qquad (17)$$

where V_{a_0} is the volume of an unperturbed sphere; $do' = \sin\theta'\, d\theta'\, d\varphi'$. $\mathbf{E}(\mathbf{r}', t)$ can be set equal to its value in the absence of perturbations of the spherical surface $\mathbf{E}_a(\mathbf{r}', t)$. Then the expression (16) is transformed to the form

$$\mathbf{E}_s(\boldsymbol{r}, t) \approx \frac{k_0^2(\varepsilon_a - 1)}{4\pi r}e^{i\omega t}\Big[\int\limits_{V_{a_0}}\mathbf{E}(\mathbf{r}', t)\, e^{ikr'\cos\gamma}d\mathbf{r}' + a_0^2\int\mathbf{E}_a(a_0, \theta', \varphi', t)\, e^{ikr'\cos\gamma} \times \mathrm{Re}\Big\{\sum_{l;\, n}\xi_{ln}(t)\, Y_{ln}(\theta', \varphi')\, e^{i\Omega_l t}\, do'\Big\}\Big] . \qquad (18)$$

The first term in the right-hand side of Eq. (18) describes the conventional elastic scattering at the frequency of the incident radiation ω. The second term is the Raman scattering with frequencies $\omega \pm \Omega_l$ at particle surface waves.

Equation (18) results in the expression for the intensity written to the square terms

$$I_s(\mathbf{r},t) = \frac{c\sqrt{\varepsilon_a}}{8\pi}\mathbf{E}_s(\mathbf{r}, t)\, \mathbf{E}_s^*(\mathbf{r}, t) \approx \left(\frac{k_0^2(\varepsilon_a - 1)}{4\pi r}\right)^2\Big[S(\mathbf{r}, t)\, S^*(\mathbf{r}, t) + 2S^*(\mathbf{r}, t)\, a_0^2\int\mathbf{E}_a(a_0, \theta', \varphi', t)\, e^{ikr'\cos\gamma} \times \mathrm{Re}\Big\{\sum_{l;\, n}\xi_{ln}(t)\, Y_{ln}(\theta', \varphi')\, e^{i\Omega_l t}\, do'\Big\}\Big] , \qquad (19)$$

where

$$S(\mathbf{r}, t) = \frac{c\sqrt{\varepsilon_a}}{8\pi}\int\limits_{V_{a_0}}\mathbf{E}(\mathbf{r}', t)\, e^{ikr'\cos\gamma}d\mathbf{r}'.$$

Hence, it follows that the intensity of the scattered electromagnetic field at the Raman frequencies is proportional to the squared radius of a droplet and the amplitude of its surface deformations. The time dependence $I_s(t)$ is determined by the superposition of oscillations at the droplet natural frequencies.

If the intensity of acting radiation is modulated at a certain frequency Ω, then at $\Omega = \Omega_l$ a sharp increase of the Raman component of a scattered signal I_s occurs due to the resonance behavior of $\xi_l(\Omega)$ (see Fig. 2). As the modulation frequency Ω changes, every such peak of the intensity of the scattered radiation corresponds to the resonance build-up of oscillations in a particle of a certain size. The square-law dependence of I_s on a_0 indicates that the increase in the particle size increases its contribution to the intensity of the scattered radiation.

The numerical calculations of the angular dependence of the scattered signal intensity by Eq. (19) have shown that the relative change of the intensity as compared to the level of unperturbed (elastic) scattering at the frequencies of mechanical oscillations of a droplet is maximal in the direction perpendicular to the direction of the action, as well as in the direction of the angle of primary rainbow $\theta \sim 137°$. This determines the angular range for optimum reception of the dynamic component of the scattered signal. The time dependence of intensity of the light scattered for several observation angles is given in Fig. 6.

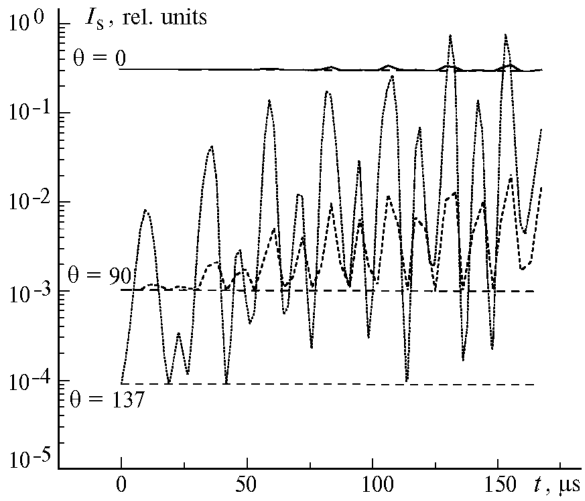

FIG. 6. Time dependence of the intensity of the light scattered at an oscillating droplet with $a_0 = 20$ μm under the action of the modulated radiation ($\Omega = \Omega_2$, with $I_0 = 10^7$ W/cm²) for different observation angles.

The initial radius of a particle is $a_0 = 20$ μm, a particle is subjected to continuous radiation modulated with the frequency $\Omega_2 = 43$ kHz at $\lambda = 0.53$ μm and $I_0 = 10^7$ W/cm².

CONCLUSION

The theoretical investigations of the amplitude of surface oscillations of transparent weakly viscous particles as a function of the modulation parameters of an acting radiation have shown the following.

1. The dependence of the amplitude of surface deformation of transparent liquid weakly viscous particles on the modulation frequency of an acting radiation is of resonance character. The decrease in the droplet radius and the increase in the liquid viscosity result in the growing width of the resonance curve of surface shifts.

2. The intensity of the light scattered at the particle surface oscillations depends resonantly on the

modulation frequency of the acting radiation. With the increasing size of a droplet, its contribution to the total intensity of the scattered radiation grows. The intensity of the scattered radiation is maximal in the direction normal to the direction of the acting radiation, as well as in the direction of the angle of primary rainbow, since the modulation of the particle surface reaches its maximum just in these directions.

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