INFLUENCE OF DROPLET SURFACE DEFORMATIONS ON STIMULATED RAMAN SCATTERING OF LIGHT

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The influence of surface deformations of a liquid particle on the Q-factor of resonant electromagnetic mode of the droplet is analyzed. It is demonstrated that high-order modes are more susceptible to deformation effects as compared with low-order modes. The pondermotive and thermocapillary mechanisms of droplet surface deformations are compared by their influence on the energy threshold of stimulated Raman scattering of light. It is established that the amplitude of pondermotive deformations is considerably lower than the amplitude of thermocapillary deformations; therefore, pondermotive deformations do not significantly influence the energy threshold of stimulated Raman scattering (SRS) of light.

INTRODUCTION

Nonlinear optical effects in microparticles of a weakly absorbing liquid lead to stimulated Raman scattering (SRS), stimulated Mandel'shtam–Brillouin scattering, stimulated fluorescence, etc. In recent years, they attract much attention due to prospects of using these effects as a physical basis for methods of remote laser diagnostics of disperse media. An overview of the main papers on the problem may be found in Ref. 1.

The presence of resonances in the internal optical field in transparent micron particles is the main cause of the appearance of stimulated scattering processes in the particles. The resonances are high Q-factor natural oscillation modes of a dielectric sphere. The modes are similar to the acoustic whispering gallery modes in their structure.[2] The resonances are observed at certain values of the dimensionless diffraction parameter of a particle $x = 2\pi a_0/\lambda$ ($x \gg 1$, where a_0 is the droplet radius; λ is the wavelength of radiation), and their positions and resonance characteristics can be determined directly from the Mie theory.[3]

The spherical surface's disturbances caused by both spontaneous deformations and forced displacements of its parts can change resonance conditions. Under natural conditions, the geometric shape of a droplet always differs from sphere due to thermocapillary deformations of its surface. The deformations are caused by inhomogeneous temperature distribution over the droplet's surface. The pondermotive forces arising in a dielectric in the presence of the electric field of light wave can be the other cause of deformations of the particle's surface.[4]

This paper is devoted to the theoretical study of the influence of surface deformations of a spherical particle on stimulated scattering, and to a comparative estimation for the roles of thermocapillary and pondermotive mechanisms of droplet deformation.

ESTIMATION OF THE INFLUENCE OF DROPLET SURFACE DEFORMATIONS ON THE Q-FACTOR OF RESONANCE MODES

The expression for threshold radiation intensity of pumping I_s leading to the appearance of stimulated scattering in spherical microparticles was obtained in Ref. 2 and has the form

$$I_s = n_a\, x_{nl}/(g_s\, Q_{nl}\, a_0\, B_c), \quad (1)$$

where n_a is the refractive index of the droplet's substance; x_{nl} is the resonance diffraction parameter; g_s is the coefficient of scattering wave amplification due to nonlinear processes; Q_{nl} is the Q-factor of the droplet's resonance mode sustaining the process; B_c is the integral coefficient taking into account the spatial overlapping of the pumping field and stimulated scattering inside the particle. The indices n, l denote the order and the number of the resonance mode, respectively.

As an example, the threshold SRS intensity in water droplets is presented in Fig. 1 as a function of the diffraction parameter for resonances of high and low orders, which can sustain the SRS process with equal probability.[2]

The influence of deformations of particle's surface will, first of all, have an effect on the Q-factor of the mode. The expression for the Q-factor of a natural resonance mode of a droplet at the frequency ω, as known, can be written in the following form:

$$Q = \omega W/P, \quad (2)$$

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where W is the electromagnetic field energy stored, on the average, in a droplet during the oscillation period of a wave. It is calculated by the expression

$$W = \frac{1}{8\pi} \int_V (\varepsilon \mathbf{E}^2 + \mathbf{H}^2)\, \mathrm{d}V.$$

Here **H** and **E** are vectors of the electric and magnetic field strength inside the particle, ε is the dielectric permeability of liquid, and the integral is taken over the whole resonance mode volume V occupied by radiation.

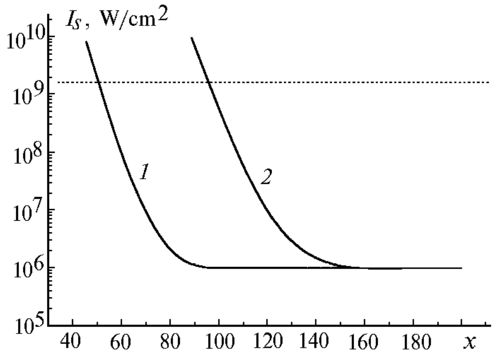

FIG. 1. SRS threshold as a function of the diffraction parameter. Lines 1 and 2 correspond to modes of low and high orders, respectively. The dotted line means the breakdown threshold.

In Eq. (2), P is the value of light wave energy losses connected with the energy escape from the particle's volume

$$P = \frac{c}{4\pi} \int_s [\boldsymbol{E} \times \boldsymbol{H}]\, \mathbf{n}_r\, \mathrm{d}s,$$

where **n** is the vector normal to the droplet's surface; c is the light speed in vacuum; the integral is taken over a closed surface including the particle. As seen from Eq. (2), a decrease in the resonance Q-factor can be connected both with a decrease in the stored energy and with an increase of the radiation losses.

Estimates of the additional radiation losses show that the effect is insignificant for the case of small deformations. Let us consider the influence of deformations of a droplet's surface on the value of the stored energy of a mode. For this purpose, let us apply the qualitative analysis based on geometrical optics. The resonance modes are standing waves formed by interference of light waves propagating along the surface at angles close to angles of total internal reflection off of the boundary between two media (in our case, this is a spherical boundary between a dielectric and air). Figure 2 presents the behavior of rays forming different resonance modes inside the particle. A standing wave arises when light rays propagating along the surface and underwent reflection at an angle equal or greater than the angle of total internal reflection fall to the initial point in the phase multiple of 2π, i.e., the condition of phase synchronizm is fulfilled.

The two cases considered (see Fig. 2) differ by the depths at which the rays travel with respect to the droplet surface $\Delta r = a_0 - b$, where b is the position of the boundary of rays' localization with respect to the center of the particle, and by the number of reflections necessary to form a standing wave. One can see that, if a ray is localized nearer to the particle's surface, it undergoes more reflections off of the surface.

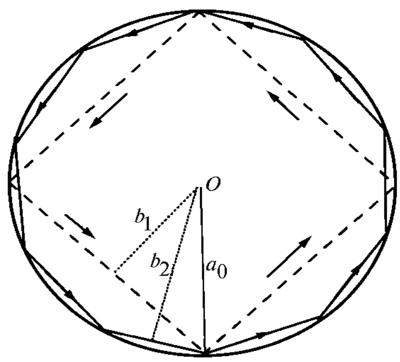

FIG. 2. Geometric scheme of the resonance modes formation. The rays correspond to two cases (solid and dashed lines) differing in the position $\Delta r_{1,2} = a_0 - b_{1,2}$ and the number of reflections necessary for the total rotation.

Figure 3 presents the distribution of the internal particle field for resonance modes of different orders. The graph presents the function $B_i = I(\mathbf{r})/I_0$, where $I(\mathbf{r})$ is the intensity of light field inside the droplet; I_0 is the intensity of the incident field. One can see that the electric field of natural modes is concentrated near the particle surface. The energy of a given mode n and its concentration near the surface of the droplet increase with decreasing order of the mode (Fig. 4).

Let us estimate the influence of the particle surface deformations on the Q-factor of resonance modes. Let us express the distance from the center of a particle to its surface as $r = a_0 + \xi(\theta, \varphi)$ where $\xi(\theta, \varphi)$ are arbitrary deformations of the surface; then, in every reflection, a ray passes an additional distance Δs which is proportional to the amplitude of the deformations. A deformed particle can be replaced by a sphere with a certain efficient radius $r_{\mathrm{ef}} = a_0 + q\xi$ so that the optical path of a ray in it is equal to the path in a deformed particle; q is some coefficient, $0 < q < 1$. Then the deformed particle can be studied theoretically as a sphere with $r = r_{\mathrm{ef}}$.

It follows from the Mie solution that the stored energy of a mode

$$W \sim \begin{cases} |b_n|^2 - \text{for TE waves,} \\ |c_n|^2 - \text{for TN waves,} \end{cases}$$

where c_n and b_n are amplitudes of the partial waves

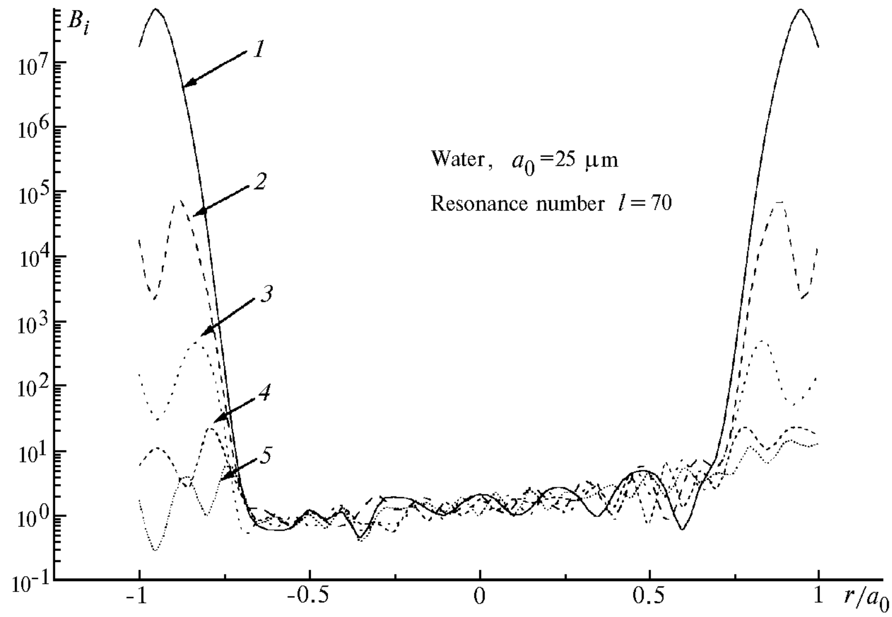

FIG. 3. Field distribution inside the particle over the main section. The figures at curves denote the resonance order. Pumping radiation is incident from the left.

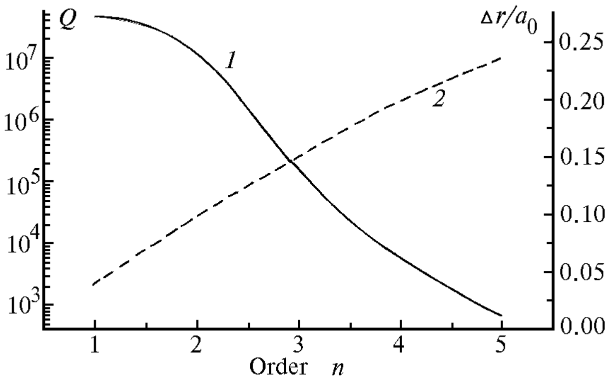

FIG. 4. The Q-factor (1) and relative position of the field maximum inside the particle with respect to the particle's surface (2) as functions of the resonance order.

(Mie coefficients). From the expressions for b_n,[3] let us write the real part for $b_n(x)$ in the form

$$\mathrm{Re}\, b_n(x) = b_n^r / [1 + \beta_n(x)^2],$$

where $\beta_n(x) = [\chi_n(x)\, \psi_n'(mx) - m\psi_n(mx)\chi_n'(x)] / /[\psi_n(x)\psi_n'(mx) - m\psi_n(mx)\psi_n'(x)]$; χ_n, ψ_n are modified spherical Riccati–Bessel functions; b_n^r are the values of the real part of the coefficient b_n at the resonance, i.e., for $x = x_{nl}$.

Let us expand the function $\beta_n(x)$ into the Taylor series near the resonance value of the diffraction parameter x_{nl}: $\beta_n(x) = \beta_n(x_{nl}) + \beta_n'(x)\,(x - x_{nl}) + \dots$; and restrict ourselves only by the linear part of the expansion, for the estimation. Taking into account that $\beta_n(x_{nl}) = 0$, we obtain

$$\mathrm{Re}\, b_n(x) \approx b_n^r / [1 + \beta_n'(x_{nl})^2\, (x - x_{nl})^2].$$

Then, considering that the displacement amplitude of the droplet's surface along the normal at oscillations by the value $q\xi = (r_{\mathrm{ef}} - a_0)/a_0$ corresponds to the relative variation of its diffraction parameter $(x - x_{nl})/x_{nl}$, i.e., drift from resonance, and, since $\beta_n'(x) = (\Gamma_n)^{-1}$ for $x = x_{nl}$, where Γ_n is the half width of the resonance curve we obtain, using the expression for the Q-factor in terms of the half width, $Q_{nl} = x_{nl}/\Gamma_n$, of the resonance curve that

$$\mathrm{Re}\, b_n(x) \approx 1/[1 + (q\, \xi\, Q_{nl}^0)^2],$$

where Q_{nl}^0 is the Q-factor of the corresponding mode for an ideal sphere. As a result, for the Q-factor of a deformed particle, we have

$$Q_{nl}(\xi) \approx Q_{nl}^0 / [1 + (q\, \xi\, Q_{nl}^0)^2]. \quad (3)$$

It follows from this expression that the Q-factor is influenced by a given displacement of the particle's surface stronger for higher Q_{nl}^0. The estimates performed by Eq. (3) for the Q-factor of the resonant modes for two values q are presented in the Table.

Q_{nl}^0	Q_{nl}, for q = 1	Q_{nl}, for q = 0.1
10^{10}	$1.46 \cdot 10^1$	$1.46 \cdot 10^3$
10^8	$1.46 \cdot 10^3$	$3.2 \cdot 10^5$
10^6	$3.2 \cdot 10^5$	$1 \cdot 10^6$
10^4	$1 \cdot 10^4$	$1 \cdot 10^4$

Thus, one can conclude that low order modes are more susceptible to surface deformations as compared with high order modes, and, consequently, there may occur such cases for which the resonance conditions are violated for the former ones while the latter one will still remain capable of sustaining the process of stimulated scattering.

COMPARATIVE ANALYSIS OF THERMOCAPILLARY AND PONDERMOTIVE DEFORMATIONS OF THE DROPLET'S SURFACE

As was noted above, thermocapillary and pondermotive mechanisms are the main physical mechanisms able to cause deformations of a droplet's surface. The amplitude of the deformations can be a criterion for comparing them. The expression for thermocapillary deformations of a droplet is well-known[5]

$$|\xi_{\text{t.Г.}}| = \sqrt{k_\text{B}\,T/4\pi\gamma}, \qquad (4)$$

where k_B is the Boltzmann constant; T is the particle's surface temperature; γ is the surface tension coefficient. As seen from Eq. (4), the amplitude of these deformations does not depend on the radius of a particle is only determined by the values T and γ.

Pondermotive forces can cause significant deformations of the particle's surface at high intensity of incident radiation what is characteristic of the development of stimulating scattering effects.[6,7]

The general formulation of the problem on deformations of a transparent droplet in a light field includes the hydrodynamic equation for incompressible liquid with the allowance for pondermotive forces. The theory of the process can be found, for instance, in Refs. 4 and 8.

The dynamic boundary condition

$$\left\{p - \frac{\rho_a}{8\pi}\left(\frac{\partial\varepsilon}{\partial\rho_a}\right)_{т} E^{т} - p_1 - \gamma\left(\frac{1}{R_1} + \frac{1}{R_{т}}\right) + f\right\} n_i =$$

$$= \eta\left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\right) n_k .$$

is valid on the droplet's surface. Here p is the pressure in the liquid; p_1 is the external (atmospheric) pressure; R_1, R_2 are the radii of principal curvature of the surface; ρ_a is the density of the liquid; η is the kinematic viscosity of the liquid; x_i are the coordinates; $f = \frac{\varepsilon - 1}{8\pi}\cdot[(\varepsilon - 1)\,(\mathbf{E}\mathbf{n}_r)^{т} + E^{т}]$ is the jump of the normal component of electric field tension on the surface of the droplet.

Further study is performed under conditions of small viscosity and small deformation amplitude. One can write the following equation for coefficients of the expansion of the surface displacement into a series over spherical functions:

$$\frac{d^{т}\,\xi_{nl}}{dt^{т}} + \frac{т}{t_l}\frac{d\,\xi_{nl}}{dt} + \Omega_l^{т}\,\xi_{nl} = \frac{l\,f_{nl}(t)}{a_0\,\rho_a}\,, \qquad (5)$$

where

$$t_l = \frac{a_0^{т}}{т(тl + 1)\,(l - 1)\,\nu}\,;\ \Omega_l = \sqrt{\frac{l(l + т)\,(l - 1)\,\gamma}{\rho_a\,a_0^3}}\,;$$

$$f_{nl}(t) = \iint f(t, a_0)\,Y_{nl}(\theta)\,\sin\theta\,d\theta\,d\varphi\,,$$

t_l is the characteristic time of particle's oscillations decrement due to viscosity; Ω_l are the natural oscillation frequencies of the droplet; ν is the dynamic liquid viscosity.

The equation (5) completed by the initial conditions $\xi_{nl}(t = 0) = \frac{d\xi_{nl}(t = 0)}{dt} = 0$ was solved numerically. The Runge–Kutta scheme of the fourth order was used as a difference scheme.

Let us now analyze the results obtained. Figures 5 and 6 present the temporal behavior of the maximum amplitudes of pondermotive deformations of water droplets of different radii. The droplets were affected by laser pulses with the wavelength $\lambda = 0.53т$ μm, of 10^{-8} s duration. The dependence of the exciting force on time was determined by the shape of the pumping pulse $I(t) = I_0\,t/t_\text{p}\,\exp\{-\,t/t_\text{p}\}$ where I_0, t_p are the peak intensity and characteristic pulse duration, respectively. For a comparison, root-mean-square surface displacements due to thermocapillary deformations are also presented here. The shaded area means typical time of SRS generation. They are taken from experimental works.[6,7]

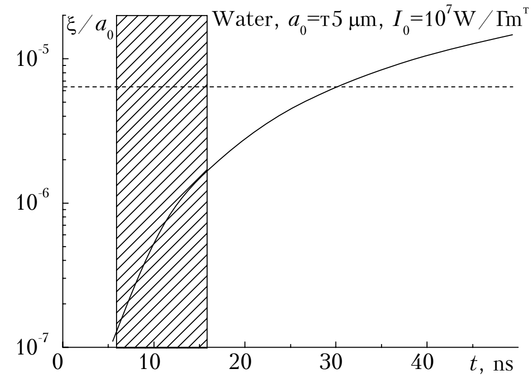

FIG. 5. The amplitude of pondermotive (solid line) and thermocapillary (dashed line) deformations as functions of time. The shaded area means the time of SRS generation.

The choice of the parameters for calculation (droplet radius a_0, intensity of the acting radiation I_0) corresponded to two limiting situations at which SRS is possible (see Fig. 1).

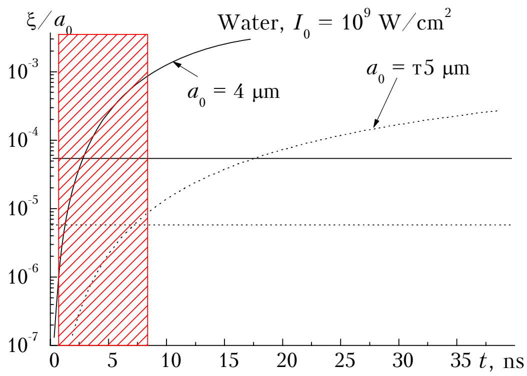

FIG. 6. The amplitude of pondermotive deformations as a function of time for particles of two radii. The straight lines show the amplitude of thermocapillary deformations. The shaded area means the time of SRS generation.

As seen from Fig. 5, for large particles (a_0 = 25 μm), the amplitude of pondermotive deformations is much lower than the thermocapillary ones, during the time of SRS existence. So one can neglect the influence of pondermotive forces on the value of Q-factor and, consequently, on the SRS threshold. One could expect that the deformations would influence the SRS generation with the increase of pulse duration but the study shows that the rise of the velocity of the displacement amplitude decreases. Although the maximum amplitude value becomes higher, the pondermotive deformations are weaker than thermocapillary ones during all the time of the SRS generation.

For small droplets (a_0 = 4 μm, see Fig. 6), the situation is different. The amplitude of pondermotive surface deformations increases much more rapidly and, after certain time, it becomes equal to the amplitude of thermocapillary deformations and then exceeds it. However, like in the previous case, the influence of pondermotive deformations on the SRS threshold is insignificant since their amplitude is considerably less than the amplitude of thermocapillary deformations at the moment of SRS appearance in both cases.

CONCLUSION

Thus, from the results of the work performed, one can come to the following conclusions.

Surface deformations of a droplet lead to a disturbance of the internal field structure in the particle, i.e., resonance conditions for the natural modes of a spherical particle are broken. High order modes are less susceptible to disturbances and they remain able to sustain the process of stimulated scattering.

The amplitude of droplet's surface deformations caused by pondermotive forces at the moment of stimulated scattering in the entire considered range of particles' size is considerably less than the amplitude of thermocapillary deformations. Therefore, it makes no any significant effect on the energy threshold of the SRS.

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REFERENCES

1. Yu.E. Geints, A.A. Zemlyanov, and E.K. Chistyakova, Atmos. Oceanic Opt. **7**, No. 7, 488–495 (1994).
2. Yu.E. Geintz, A.A. Zemlyanov, and E.K. Chistyakova, Atmos. Oceanic. Opt. **8**, No. 10, 803–807 (1995).
3. A.P. Prishivalko, *Optical and Thermal Fields Inside Light-Scattering Particles* (Minsk, 1983), 190 pp.
4. A.A. Zemlyanov, Kvant. Elektron. **1**, 2085–2088 (1974).
5. H.M. Lai, P.T. Leung, and K. Young, Phys. Rev. **A41**, No. 9, 5199–5204 (1990).
6. P. Chylek, A. Biswas, M.A. Jarzembski, V. Srivastava, and R.G. Pinnick, Appl. Phys. Lett. **52**, No. 19, 1642–1644 (1988).
7. J.–Z. Zhang, D.H. Leach, R.K. Chang, Opt. Letters **13**, No. 4, 270–272 (1988).
8. Yu.E. Geints, A.A. Zemlyanov, Atmos. Oceanic Opt. **9**, No. 10, 854–858 (1996).