STIMULATED RAMAN SCATTERING IN SPHERICAL PARTICLES

Yu.E. Geintz 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 consider the effects of stimulated light scattering in transparent spherical particles (stimulated Raman scattering, stimulated Mandel'shtam–Brillouin scattering, stimulated fluorescence) connected with the amplification of spontaneous scattering wave by resonant modes of the internal optical field of a particle. These phenomena are described theoretically from a unified physical viewpoint, their common regularities are established, the threshold and angular behavior of stimulated scattering are discussed. The effects of Raman light scattering on surface waves caused by ponderomotive light field forces on liquid particles are studied theoretically. We also consider the angular structure of the field of a scattered wave.

INTRODUCTION

Nonlinear optical effects of stimulated light scattering (SS) in a weakly absorbing continuous media (stimulated Raman scattering (SRS), stimulated Mandel'shtam–Brillouin scattering (SMBS)) are well described in scientific literature. Only recently it was discovered that the manifestation of these effects has specific features for a disperse substance.[1–5] For instance, the spectral form of the SRS signal has the "spiky" structure within the Raman contour; the SRS signal is delayed relative to the pump pulse,[1,4] the energy SRS and SMBS thresholds decrease as compared with a continuous medium.[3] The presence of internal optical field resonances in micron particles is the main cause of resonant nonlinear optical effects in the particles (SRS, SMBS, and stimulated fluorescence (SF)).[1] The resonances are observed at certain values of the diffraction parameter of a particle $x_a = 2\pi a_0/\lambda$, $x_a > > 1$ (where a_0 is the droplet radius, λ is the wavelength of laser radiation) and are characterized by the mode order and number of the partial electromagnetic wave yielding the resonance.

The property of a transparent particle to focus light side it also plays an important role in the appearance of the SS effects. Near the shaded and illuminated surfaces, two maxima of the electromagnetic field are formed inside the particle. The maxima are powerful sources of spontaneous Stokes radiation (the Stokes noises).

The ponderomotive forces of the light field generate deformations and oscillations of the liquid particles.[6,17–19] These oscillations of the particle shape cause Raman scattering of light incident onto the particle.[21]

All nonlinear effects have different physical nature, and their similarity is in the fact that they are essentially determined by the spatial structure of the internal optical field near the surface of a particle and, consequently, they are effects of surface nonlinear scattering.

The overwhelming majority of publications on this problem deal with experimental studies. There are only few theoretical papers where the models of some SS effects in particles are presented.[1,4,11,13,20]

The aim of this paper is the theoretical study of a wide class of nonlinear effects of Raman light scattering by transparent particles. We make an attempt to fill a gap in the theory of nonlinear optical interactions in disperse media. The gap is connected with the problem of stimulated light scattering important for atmospheric optical investigations.

1. RESONANCES OF THE OPTICAL FIELD IN WEAKLY ABSORBING PARTICLES

As known from the theory of diffraction of an electromagnetic wave on a dielectric sphere (Mie theory), the internal optical field at $x_a >> 1$, in it is characterized by the presence of many spikes with the intensity difference of about 10–100 times.[7] The maximal values of the internal optical field are reached near the surface of the sphere. However, the investigations showed that, the internal optical field can multiply increase at quite certain; fixed values of the particle radius, especially in the ranges of maxima (10^4–10^6 times).[1] Such an effect was called resonances of the internal optical field and it is considered as natural oscillation (resonant) modes of the spherical particle. The presence of the internal optical field resonances deduced immediately from the Mie theory was also discovered experimentally by the presence of spikes in the scattering spectrum.[8]

The solution of the problem on the diffraction of a plane electromagnetic wave on a sphere has the form defining the field inside it as a series over spherical harmonics[9]:

$$\mathbf{E}(r, \theta, \varphi) = \frac{E_0}{2kr} \sum_{n=1}^{\infty} \sum_{l=-n}^{n} (-i)^{n+1} [b_n(x_a) \mathbf{M}_{nl}(\theta, \varphi) \psi_n(kr) + \frac{1}{k} c_n(x_a) \nabla \times [\mathbf{M}_{nl}(\theta, \varphi) \psi_n(kr)]] + \text{compl.conjug.} \quad (1)$$

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The amplitudes of the partial harmonics (Mie coefficients) are defined by the following expressions:

$$b_n = i^n \frac{2n+1}{n(n+1)} \frac{-m}{\xi_n'(x_a)\,\psi_n(mx_a) - m\,\xi_n(x_a)\,\psi_n'(mx_a)},$$

$$c_n = i^n \frac{2n+1}{n(n+1)} \frac{m}{\xi_n(x_a)\,\psi_n'(mx_a) - m\,\xi_n'(x_a)\,\psi_n(mx_a)},$$

where m is the complex relative refractive index; k is the wave number inside the particle; r, θ, φ are the spherical coordinates; ψ_n, ξ_n are the spherical Bessel functions; $\mathbf{M}_{nl}(\theta, \varphi) = -\frac{i}{[n(n+1)]^{-1/2}} \mathbf{L}\, Y_{nl}(\theta, \varphi)$ are the spherical vector-harmonics; $Y_{nl}(\theta, \varphi)$ are the spherical functions; $\mathbf{L} = -i\mathbf{r}\times\nabla = -i\left[\theta \frac{1}{\sin\theta}\frac{\partial}{\partial\theta} - \varphi\frac{\partial}{\partial\varphi}\right]$ is the angular momentum operator; $E_0 = |\mathbf{E_0}|$; $\mathbf{E}_0$ is the amplitude of the electric field of a light wave incident onto the particle. The primes mean the derivatives of the functions with respect to their arguments.

The natural resonances of the electromagnetic field in the particle are connected with the zeroes of the denominator in the expressions for c_n and b_n, and the position of the zeroes is determined only by the values $m = m_a/m_0$ ($m_a = n_a + i\,\kappa_a$ is the complex refractive index of the drop substance; m_0 is introduced for a non-absorbing medium) and x_a. Thus, in order to find resonances, one should determine the position of zeroes with respect to the scale x_a in the denominators of c_n and b_n (Ref. 10). The deviation of the particle shape from the ideal sphere leads to decomposition of each resonant mode into a multiplet consisting of $(2n + 1)$ lines and, consequently, to a sharp deterioration of resonant properties.[11]

In the general case, the Q-factor of the nth resonant mode the order of l can be written as follows:

$$Q_{nl} = 2\pi\omega_S/(c\,D_{nl}),$$

where c is the speed of light; D_{ln} is the coefficient of total energy losses in the resonator; ω_S is the frequency of the electromagnetic field inside the drop. In its turn,

$$D_{nl} = D_{nl}^r + D_a,$$

where D_{nl}^r are radiation losses (due to tangent escape of radiation through the surface of the drop); D_a are the radiation losses due to absorption in the drop substance. It should be noted that D_{nl}^r is minimal for spherical particles; its value increases with the deviation of the particle shape from the ideal sphere.

Then, the expression for the Q-factor of the spherical resonator is as follows

$$(Q_{nl})^{-1} = (Q_{nl}^r)^{-1} + (Q_a)^{-1},$$

where $Q_a = n_a^2\,\omega_S/(\alpha c)$ ($\alpha = 4\pi\sigma/c$ is the volume absorption coefficient; σ is the specific conductivity of the particle substance); Q_{nl}^r is the radiative Q-factor.

Figure 1 presents the total Q-factor Q_{nl} as a function of the diffraction parameter x_a for resonant modes of different numbers and orders (the points within a curve are resonances of different orders). As one can see from the figure, the values of Q_{nl} are limited from the above due to absorption (the calculations were performed for $\kappa_a = 10^{-8}$).

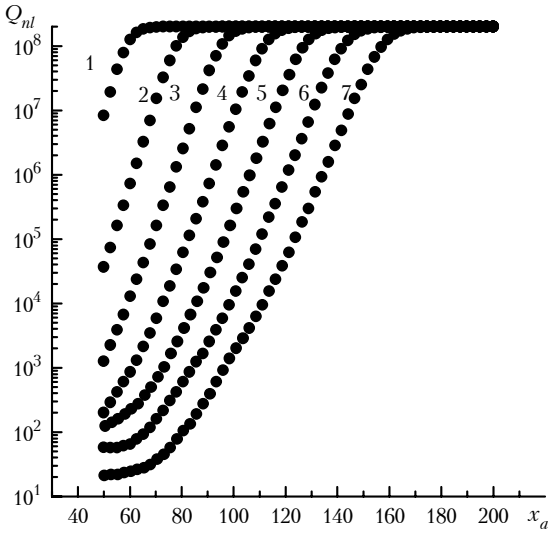

FIG. 1. The Q-factor Q_{ln} of different resonances of the internal optical field as a function of the diffraction parameter of water drops x_a. The figures at the curves are the numbers of the resonance order.

2. THE THEORETICAL MODEL OF STIMULATED LIGHT SCATTERING IN SPHERICAL PARTICLES

Let us briefly consider the basic aspects of the theoretical description of stimulated light scattering in transparent particles. The scattering is caused either by spontaneous scattering at thermal fluctuations of a medium or by emission of fluorescing molecules in the particles.

By now, the following physical model of SS origin in a spherical particle has established in the literature.[1]

When the radiation interacts with the substance of a transparent particle spontaneous in elastic scattering arises in the whole volume of the particle due to thermal motion of molecules of the medium or fluorescence of molecules, that is most intense in the domains of the internal optical field focusing. A portion of waves from the spontaneous Stokes radiation spectrum leaves the drop, and a portion propagates along its surface due to the total internal reflection. The waves are attenuated on their path due to absorption and penetration through the surface; they can also be amplified due to nonlinearities of the medium. In the case when the resonance condition is

satisfied for a frequency from the Stokes spectrum (or for some frequencies), i.e., when the frequency of the Stokes wave coincides with that of one of the natural resonance modes of the drop, the amplification of the spontaneous wave exceeds its total losses, and stimulated scattering arises in the particle. From the viewpoint of formation of fields in a resonator, the SS field can be treated as a standing wave formed by superposition of electromagnetic waves propagating toward each other along the spherical surface of the drop when the condition of phase synchronism is satisfied.

The analysis demonstrates that, in spite of different physical nature of the effects responsible for one or another process of simulated scattering (SRS, SMBS, SF), their theory is similar in general. It is explained, first of all, by the fact that all non-linear optical effects considered here are resonance processes that means that the fact of their occurrence in a particle is connected with its resonance properties. So the threshold, angular, and spectral characteristics of all types of stimulated scattering bring a "print" of the resonant structure of internal fields.

The initial equations for the theoretical analysis of nonlinear scattering in a particle (magnetic permeability $\mu_a = 1$) are nonhomogeneous Maxwell equations in which the nonlinear part the medium polarizations $\mathbf{P}(\mathbf{r}, t)$ induced by the pumping field serves as a source of the field of Raman scattered wave

$$\text{rot}\ \mathbf{E}(\mathbf{r}, t) = -\frac{1}{c}\frac{\partial \mathbf{H}(\mathbf{r}, t)}{\partial t}, \quad \text{div}\ \mathbf{E}(\mathbf{r}, t) = 0,$$

$$\text{rot}\ \mathbf{H}(\mathbf{r}, t) = \frac{1}{c}\frac{\partial}{\partial t}[\mathbf{E}(\mathbf{r}, t) + 4\pi\, \mathbf{P}(\mathbf{r}, t)] + \frac{4\pi\sigma}{c}\mathbf{E}(\mathbf{r}, t),$$

$$\text{div}\ \mathbf{H}(\mathbf{r}, t) = 0.$$

It is well known this the system can be transformed into the wave equations for the electric field strength vector in the particle $\mathbf{E}(\mathbf{r}, t)$ after eliminating the magnetic field strength $\mathbf{H}(\mathbf{r}, t)$

$$\text{rot rot}\,\mathbf{E}(\mathbf{r}, t) + \frac{1}{c^2}\frac{\partial^2 \mathbf{E}(\mathbf{r}, t)}{\partial t^2} + \frac{4\pi\sigma}{c^2}\frac{\partial \mathbf{E}(\mathbf{r}, t)}{\partial t} = -\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}(\mathbf{r}, t). \quad (2)$$

The equation for the field (2) is completed by equations describing a particular physical mechanism governing the formation of a non-linearly scattered wave. For instance, they are the wave equation for non-linear polarization and the kinetic equation for the populations' difference of the "operating" transition levels of a molecule of the particle's substance

$$\frac{\partial^2 \mathbf{P}(\mathbf{r}, t)}{\partial t^2} + \frac{2}{T_2}\frac{\partial \mathbf{P}(\mathbf{r}, t)}{\partial t} + \omega_{\text{vib}}^2\, \mathbf{P}(\mathbf{r}, t) = \frac{2 N_0\, \omega_{\text{vib}} |\mu_{12}|}{3\hbar}\Delta N\, \mathbf{E}(\mathbf{r}, t),$$

$$\frac{\partial (\mathrm{D}N)}{\partial t} + \frac{\mathrm{D}N - \mathrm{D}N_e}{T_1} = \frac{2}{N_0\, \hbar\, w_{\text{vib}}}\frac{\partial \mathbf{P}(\mathbf{r}, t)}{\partial t}\mathbf{E}(\mathbf{r}, t). \quad (3)$$

Here N_0 is the concentration of molecules in the substance, ω_{vib} is the frequency of the vibrational transition of a molecule, $|\mu_{12}|$ is the dipole matrix element, T_1 and T_2 are the longitudinal and transversal relaxation times respectively, ΔN_e is the equilibrium value of the difference of populations, $\hbar$ is the Planck constant.

For the process of stimulated fluorescence the corresponding equations have the form[20]

$$\frac{\partial^2 \mathbf{P}(\mathbf{r}, t)}{\partial t^2} + \frac{2}{T_2}\frac{\partial \mathbf{P}(\mathbf{r}, t)}{\partial t} + \left(\frac{1}{T_2^2} + \omega_f^2\right)\mathbf{P}(\mathbf{r}, t) = -\frac{2 N_M\, \omega_f |\mu_{12}|}{3\hbar}\Delta N\, \mathbf{E}(\mathbf{r}, t),$$

$$\frac{\partial \mathrm{D}N}{\partial t} = \frac{2\, \mathbf{E}(\mathbf{r}, t)}{\hbar\, \omega_{\text{vib}}\, N_M}\left(\frac{\partial \mathbf{P}(\mathbf{r}, t)}{\partial t} + \frac{1}{T_2}\mathbf{P}(\mathbf{r}, t)\right) - D(\Delta N - \Delta N_e), \quad (4)$$

where N_M is the concentration of "active" molecules in the substance of a particle, ω_f is the frequency of fluorescence, D is the factor proportional to the probability of stimulated transitions in the molecule.

To describe the SMBS process, it is necessary to use the wave equation for the light field (2) jointly with the equation for the pressure in a medium $p(\mathbf{r}, t)$

$$\nabla^2 p(\mathbf{r}, t) - \frac{1}{c_s^2}\frac{\partial^2 p(\mathbf{r}, t)}{\partial t^2} + \frac{2\Gamma_B}{c_s^2}\frac{\partial p(\mathbf{r}, t)}{\partial t} = \frac{\gamma}{8\pi}\nabla^2 |\mathbf{E}(\mathbf{r}, t)|^2 \quad (5)$$

and the expression connecting the pressure and non-linear polarization of the medium $\mathbf{P}_N$

$$\mathbf{P}_N(\mathbf{r}, t) = [\gamma/(4\pi\, c_s^2\, \rho_a)]\, p(\mathbf{r}, t)\, \mathbf{E}(\mathbf{r}, t).$$

Here c_s, Γ_B are the velocity and attenuation coefficient of hypersound in a liquid, γ is the electrostriction constant, ρ_a is the density of the particle's substance.

Then, the solution of the wave equation for the field (2) is represented as an expansion over natural electromagnetic vibrational modes of the resonator $\mathbf{E}_{nl}(\mathbf{r})$ with natural frequencies ω_{nl}

$$\mathbf{E}(\mathbf{r}, t) = \sum_{n,l} A_{nl}(t)\, \mathbf{E}_{nl}(\mathbf{r}), \quad (6)$$

where the coefficients $A_{nl}(t)$ satisfy the equations

$$\frac{\mathrm{d}^2 A_{nl}(t)}{\mathrm{d}t^2} + 4\pi\sigma_{ef}\frac{\mathrm{d}\, A_{nl}(t)}{\mathrm{d}t} + \omega_{nl}^2\, A_{nl}(t) = -4\pi \int_{V_a} \mathbf{E}_{nl}(\mathbf{r}')\frac{\partial^2 \mathbf{P}(\mathbf{r}', t)}{\partial t^2}\,\mathrm{d}\mathbf{r}'. \quad (7)$$

Integration in Eq. (7) is performed over the volume of the particle. Here we introduce the coefficient σ_{ef} which has the meaning of the effective absorption taking into account the losses of light wave caused not only by the absorption in the substance but also due to escape of radiation from the resonator-particle.

The expansions over eigenfunctions similar to Eq. (6) are performed also for nonlinear polarization $\mathbf{P}_N(\mathbf{r}, t)$ and pressure $p(\mathbf{r}, t)$. In the latter case,

$$p(\mathbf{r}, t) = \sum_{n,l,m} \wp_{nlm}(t)\, \Pi_{nlm}(\mathbf{r}).$$

The solution of Eq. (7) jointly with the Eqs. (3)–(5) and corresponding initial and boundary conditions enable one to comprehensively describe the stimulated scattering process in a particle.

Let us note that the system of partial TE and TM waves is usually taken as eigenfunctions in Eq. (6). Their form is defined by Mie solution to the problem of plane electromagnetic wave diffraction on a sphere (1)

$$\mathbf{E}_{nl}(\mathbf{r}) = \begin{cases} b_n(x_a)\,\mathbf{M}_{nl}(\mathbf{r})\,\psi_n(kr) & \text{for TE waves,} \\ 1/k\; c_n(x_a)\,\nabla\times[\mathbf{M}_{nl}(\mathbf{r})\,\psi_n(kr)] & \text{for TM waves.} \end{cases}$$

Under the condition that the change in the pressure is zero on the surface of the particle (what is a sufficiently good approximation in studying SMBS[11]), the functions $\mathbf{\Pi}_{nlm}(\mathbf{r})$ have the form

$$\mathbf{\Pi}_{nlm}(\mathbf{r}) = C_{nm}\, \psi_n(\alpha_{nm}\, r/r_0)\; Y_{nl}(\theta, \varphi),$$

where C_{nm} are normalization factors; α_{nm} is the mth zero of the Bessel spherical function ψ_n.

3. THE ENERGY THRESHOLD OF STIMULATED SCATTERING AT RESONANCES OF THE INTERNAL OPTICAL FIELD OF PARTICLES

As in the case of any nonlinear process, there is a threshold intensity of incident radiation for the stimulated scattering in particles to occur. The effect is not observed at lower intensity. The value of the threshold intensity can be obtained from the integral form of the electromagnetic energy conservation principle in a particle (at the frequency of scattered wave ω_S). It follows from the Maxwell equations

$$\mathrm{d}W_S/\mathrm{d}t = P_g - (P_a + P_r). \qquad (8)$$

Here we introduce the following designations:

$W_S = \frac{1}{16\pi}\int_{V_a} (\varepsilon_a \mathbf{E}_S(\mathbf{r},t)\mathbf{E}_S^*(\mathbf{r},t) + \mathbf{H}_S(\mathbf{r},t)\mathbf{H}_S^*(\mathbf{r},t))\mathrm{d}\mathbf{r}'$ is the electromagnetic field energy accumulated in the volume of the drop during the oscillation period $T = 2\pi/\omega_0$, ω_0 is the frequency of light incident onto the particle,

$P_r = \frac{c}{8\pi}\int_{S_a} [\mathbf{E}_S(\mathbf{r}, t) \times \mathbf{H}_S(\mathbf{r}, t)]\, \mathbf{n}_r\, \mathrm{d}o'$ is the average power of radiative losses through the surface of the droplet during the period T,

$P_a = \frac{\sigma}{2}\int_{V_a} \mathbf{E}_S(\mathbf{r}, t)\, \mathbf{E}_S^*(\mathbf{r}, t)\, \mathrm{d}\mathbf{r}'$ is the average power of heat losses inside the particle,

$P_g = -\overline{\left(\int_{V_a} \mathbf{E}_S(\mathbf{r}, t)\, \frac{\partial \mathbf{P}}{\partial t}\, \mathrm{d}\mathbf{r}'\right)_T}$ is the average power of sources of the Stokes wave, $\mathbf{n}_r$ is the external normal to the surface of the particle, ε_a is the dielectric permeability of the particle substance.

The electric field inside the particle is represented as a sum of quasimonochromatic fields

$$2\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_L(\mathbf{r}, t)\, e^{i\omega_0 t} + \mathbf{E}_S(\mathbf{r}, t)\, e^{i\omega_S t} + \text{compl.conjug.},$$

where $\mathbf{E}_L$, $\mathbf{E}_S$ are the complex slowly varying field amplitudes at the pump frequency and Stokes radiation, respectively.

Let us introduce the concept of Q-factor of the resonator by the expression

$$Q = \omega_S\, W_S/(P_a + P_r).$$

The condition of SS appearance is defined as

$$\mathrm{d}W_S/\mathrm{d}t = 0.$$

For the threshold intensity of pumping radiation above which the stimulated scattering takes place, we have

$$I_S = 2\pi\, n_a/(g_S\, Q_{nl}\, \lambda_S\, B_c). \qquad (9)$$

Here λ_S is the wavelength of scattered light, g_S is the gain coefficient of the corresponding stimulated scattering process, B_c is the integral coefficient taking into account the spatial overlapping of interacting fields inside the particle.[12] The threshold values I_S are as lower for better overlapping.

Besides, the value B_c depends also on whether or not the pumping field is in resonance. If the incident field frequency corresponds to the frequency of a resonance mode of the particle; the so-called "double resonance" occurs.[3] It means that there exist both the resonance for the Stokes wave λ_S and for the pumping wavelength λ_L. Let us note that this situation is rather hard to be realized due to small width of the resonance lines of the particles (for water drops, this width is of the order of 3 cm^{-1}). In the most frequent situation of a single resonance, the value B_c only insignificantly differs from unity in the whole range of particles' dimensions studied. However, the values B_c sharply increase at a "double resonance". They reach the value ~10^3 for high Q-factor resonances.

The threshold values I_S for SRS and SMBS processes in water drops of different radius (the absorption index $\kappa_a = 10^{-8}$) are presented in Fig. 2. The threshold of optical breakdown for water drops[3] is also presented here. As to the SMBS, three cases are considered. They are "single" resonance (the resonance for the Stokes wave only), "double" resonance (for the pumping wave and the scattered wave) and "triple" resonance (the pumping, scattered, and acoustic wave). Note that, in the latter case, the value of the threshold SMBS intensity has anomalously small value because the acoustic wave induced by pumping radiation falls into one of the acoustic resonances of the particle that leads to a sharp increase of the disturbance amplitude of the dielectric constant of the medium and, consequently, non-linear polarization (according to Eq. (5)). However, this "triple" resonance is even more rare and seems to be obtained only by simultaneous laser and acoustic excitation of a drop with corresponding frequencies.[13]

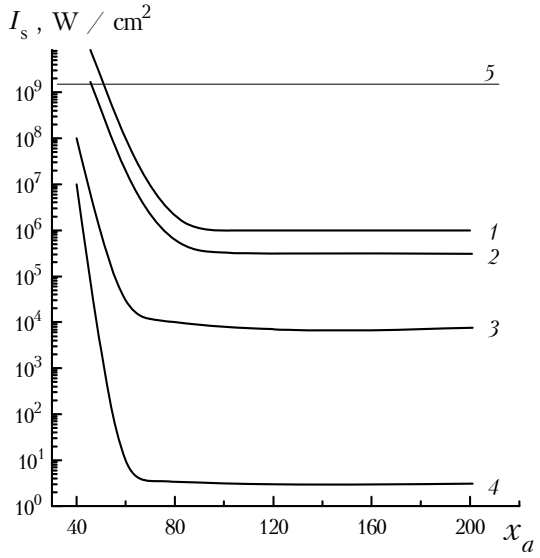

FIG. 2. The threshold of SRS(1) and SMBS(2–4) in water drops of different dimensions: "single" resonance (1,2), "double" resonance (3), and "triple" resonance (4). The curve (5) is the threshold of optical break-down of particles.

In general, Fig. 2 shows that the SMBS threshold intensity is always lower than the SRS threshold intensity. It is connected with higher value of the SMBS gain ($g_S \approx 5 \cdot 10^{-3}$ cm/MW, see Ref. 2) as compared with the corresponding gain coefficient for SRS ($g_S \approx 10^{-3}$ cm/MW, see Ref. 1). As to the dependence of I_S on the drop radius, the figure demonstrates that the threshold intensity sharply increases with the decrease of the drop size, due to similar decay of the radiative Q-factor for small particles (see Fig. 1). For $x_a \geq 100$, I_S in fact does not depend on the radius of liquid particles since the Q-factor is limited by the losses due to absorption in liquid. For $x_a \leq 20$–40, the SRS and SMBS effects can be suppressed by the optical breakdown inside the particle.[1]

4. ANGULAR STRUCTURE OF STIMULATED LIGHT SCATTERING BY A SPHERICAL PARTICLE

In experiments, SS there are observed luminous arcs on the surface of a spherical particle, depending on pump intensity, forward and backward along its main diameter.[14] These luminous regions are sources of the SS signal outside the particle and make the angular distribution of scattering which, according to experiments,[15,16] significantly differs from the diagram of elastic scattering.

Let us consider the angular structure of the SS field far from a spherical particle and study its dependence on the spatial structure of the internal Stokes field.

The problem considered can be formulated as a problem of emission from a spherical volume with a given spatial distribution of the electromagnetic field into the space surrounding the particle. The mathematical formulation of the problem is based on the Helmholtz equation for the vector potential of the electromagnetic field $\mathbf{A}(\mathbf{r}, t)$

$$\nabla^2 \mathbf{A}(\mathbf{r}, t) + (\omega_S^2/\varepsilon_a)\, \mathbf{A}(\mathbf{r}, t) = -\, \mathbf{J}_a(\mathbf{r}, t) \qquad (10)$$

provided that $\mathrm{div}\mathbf{A}(\mathbf{r}, t) = 0$. Here $\mathbf{J}_a(\mathbf{r}, t) = \varepsilon_a \dfrac{\partial \mathbf{E}_S(\mathbf{r}, t)}{\partial t}$ is the density of polarization currents induced by the internal field of the particle $\mathbf{E}_S(\mathbf{r}, t)$. The field components ($\mathbf{E}_S(\mathbf{r}, t) \equiv \mathbf{E}(\mathbf{r}, t, \omega_S)$) can be expressed in terms of the vector potential as follows:

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

The solution of the equation (10) is known

$$\mathbf{A}(\mathbf{r}, t) = i\, \omega_S\, \varepsilon_a \int_{V_a} \frac{\mathbf{E}_S(\mathbf{r}', t)}{4\pi R} \exp(-i\, \mathbf{k}_S \mathbf{R})\, d\mathbf{r}'.$$

Here $\mathbf{r}$ is the radius vector of the observation point at which the value of the field is sought, $\mathbf{r}'$ is the radius vector of points inside the drop, $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, $\mathbf{k}_S$ is the wave vector of the Stokes wave. The integral is taken over the volume of the particle V_a.

At the observation point with the radius vector $\mathbf{r}$, the SS field is written as follows:

$$\mathbf{E}_S(\mathbf{r}, t) = \mathrm{rot}\,\mathrm{rot} \int_{V_a} \frac{(\varepsilon_a - 1)\, \mathbf{E}_S(\mathbf{r}', t)}{4\pi \varepsilon_a R} \exp(-i \mathbf{k}_S \mathbf{R})\, d\mathbf{r}'. \qquad (11)$$

As one can see, the intergand in Eq. (11) contains the intensity of the electric Stokes field $\mathbf{E}_S(\mathbf{r}', t)$ in a particle. Its spatial configuration corresponds to one of the resonance modes $\mathrm{TE}_n(\mathrm{TM}_n)$. As it was mentioned above, it can be represented as a standing wave localized near the surface of the drop. So, using the well known Mie solution (1), for instance, for the TE_n-resonant mode of the spherical resonator, we have the expression

$$\mathbf{E}_S(\mathbf{r}', t) = A_E(t)\, b_n(x_a)\, \psi_n(k_S r')[\mathbf{M}_{nm}(\theta, \varphi) + \mathbf{M}^*_{nm}(\theta, \varphi)]/2, \qquad (12)$$

where A_E is the amplitude of the SRS wave. A similar expression can be written for TM_n-modes.

By substituting Eq. (12) into Eq. (11) and expanding the integrand in a series over spherical harmonics, we finally obtain

$$\mathbf{E}_S(\mathbf{r}, t) \cong \frac{k_S^2}{4\pi r} A_E(t)\, b_n(x_a) \int_{V_a} \sum_{l,m} \frac{2l+1}{l(l+1)} \psi_n(k_S r') \times$$

$$\times \psi_l(k_S r)\, Y_{l1}(\theta, \varphi)\, [\mathbf{M}_{nm}(\theta', \varphi') + \mathbf{M}^*_{nm}(\theta', \varphi')]\, d\mathbf{r}'. \quad (13)$$

One of the results of numerical calculations by Eq. (13) is presented in Fig. 3 where the results on the angular SRS structure from an ethanol drop with the radius $a_0 = 15.1$ μm irradiated by the second harmonic of a Nd-YAG laser ($\lambda = 0.53$ μm) are compared with the corresponding experimental data from in Ref. 15. Experimental data are shown by dots. The Stokes radiation has the wavelength $\lambda_S = 0.65$ μm. The calculated angular dependence of the elastic scattering intensity is also shown here. Typical *V*-shaped angular dependence of the SRS signal is clearly seen in the figure. It was also observed in Ref. 16.

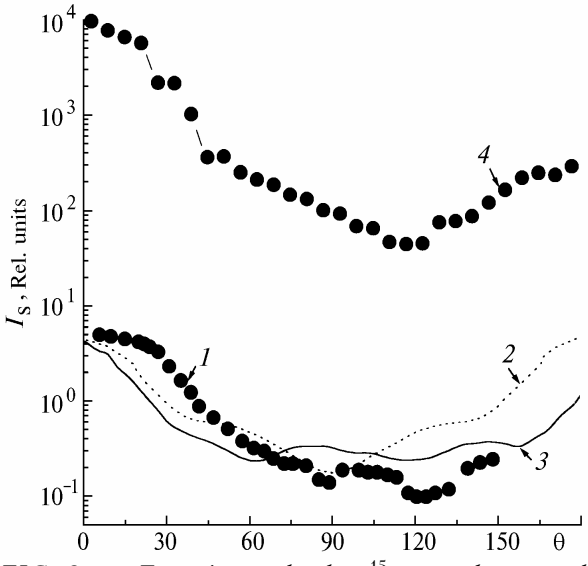

FIG. 3. Experimental data[15] *on the angular SRS(1) and elastic scattering structure (4) from an ethanol drop of the radius* 15.1 μm *in the far zone. Solid lines are theoretical calculations of the angular SRS function at one-mode (2) and multimode (3) regime of SRS generation in a drop.*

The results of the study demonstrate that the directional pattern of SS from a particle is more isotropic on the whole than the diagram of the elastic scattering. The angular structure of a one-mode. SS, i.e., when the SS process is sustained only by a single natural mode of the particle, is symmetric in contrast to the elastic scattering. It is a consequence of angular symmetry of the internal Stokes field at resonance with the natural vibration mode of the particle.

At the same time, the situations when several modes simultaneously have natural frequencies close to the Stokes frequency ω_S are possible due to high spectral density of natural modes in a particle.[1] In this case, they all give a contribution into the process of SS development in a particle. In such a multimode regime of the SS generation the angular distribution on the Stokes field in the far zone becomes asymmetric.

5. RAMAN SCATTERING OF LIGHT ON OSCILLATIONS OF THE DROP SURFACE CAUSED BY PONDEROMOTIVE FORCES OF THE LIGHT FIELD

The pondermotive forces of the electromagnetic field cause deformations and oscillations of transparent drops.[6,17-19] The oscillations of the drop surface lead to Raman scattering of the light wave incident on it.

In Ref. 21, Raman scattering by small drops ($ka_0 \ll 1$) at free oscillations of the drop surface was studied theoretically for the first time. The problem of Raman light scattering on an oscillating transparent drop was not solved for the case of optically large particles ($ka_0 \geq 1$).

Let us consider solution of the problem based on the integral equation for light field, that follows from the Helmholtz equation (10) for the vector potential of the field.

The complex electric field $\mathbf{E}_1(\mathbf{r}, t)$ at the observation point with the radius vector r is written as follows

$$\varepsilon_a\, \mathbf{E}_1(\mathbf{r}, t) = \mathbf{E}_0\, e^{i\omega_0 t - ikr} +$$

$$\text{rot rot} \int_{V_a} \frac{(\varepsilon_a - 1)\, \mathbf{E}(\mathbf{r}', t)\, e^{i\mathbf{kR}}}{4\pi R}\, d\mathbf{r}',$$

where $\mathbf{k}$ is the wave vector of the incident wave.

Further consideration is performed only for a scattered wave. Here

$$\mathbf{E}_{iss}(\mathbf{r}, t) = \text{rot rot} \int_{V_a} \frac{(\varepsilon_a - 1)\, \mathbf{E}(\mathbf{r}', t)\, e^{i\mathbf{kR}}}{4\pi\, e_a\, R}\, d\mathbf{r}'. \quad (14)$$

The deformation of the drop with the radius a arising due to ponderomotive forces at a point with the spherical coordinates (θ, φ) is given by the relation

$$a(t; \theta, \varphi) - a_0 = \xi(t; \theta, \varphi) =$$

$$= \text{Re}\, \{\sum_{l,n} \xi_{ln}(t)\, Y_{ln}(\theta, \varphi)\, e^{i\Omega_l t}\},$$

where the value of deformation ξ is expanded over spherical harmonics. Here $\Omega_l = [l(l-1)(l+2)\beta/(\rho_a a_0^3)]^{1/2}$ are natural (Rayleigh) frequencies of the drop

oscillations, β is the surface tension coefficient of the liquid.

Let us represent the integral over the volume of the deformed particle as a sum of integrals

$$\int_{V_a} \frac{(\varepsilon_a - 1)\,\mathbf{E}(\mathbf{r}', t)\,e^{i\mathbf{kR}}}{4\pi\,\varepsilon_a R}\,d\mathbf{r}' = \int_{V_{a_0}} \frac{(\varepsilon_a - 1)\,\mathbf{E}(\mathbf{r}', t)\,e^{i\mathbf{kR}}}{4\pi\,\varepsilon_a R}\,d\mathbf{r}' +$$

$$+ \int \frac{(e_a - 1)\,\mathbf{E}(\mathbf{r}', t)\,e^{i\mathbf{kR}}}{4\pi\,\varepsilon_a R}\,do' \int_{a_0}^{a(\theta,\varphi)} r^2\,d\mathbf{r}' ,$$

where V_{a_0} is the volume of the undisturbed sphere; $do' = \sin\theta' d\theta' d\varphi'$.

Under the assumption that under conditions far from the resonance ($\xi \ll a_0$), the disturbances of the surface are small the electromagnetic field in the deformed particle $\mathbf{E}(\mathbf{r}', t)$ can be taken equal to its value $\mathbf{E}_L(\mathbf{r}', t)$ in the absence of disturbances of the spherical surface. Then, for the case of a quasiharmonic field $\mathbf{E}_L(\mathbf{r}, t) = \mathbf{E}_L^0(\mathbf{r}, t)e^{i\omega_0 t}$, in the far zone $\mathbf{kr} >> 1$, Eq. (14) can be transformed to the following form

$$\mathbf{E}_{iss}(\mathbf{r}, t) \approx \frac{k^2\,(e_a - 1)}{4\pi\,\varepsilon_a\,\mathbf{r}} e^{i\omega_0 t - i\mathbf{kr}} \left[\int_{V_{a_0}} \mathbf{E}_L^0(\mathbf{r}', t)\,e^{i\mathbf{kR}'\cos\vartheta}\,d\mathbf{r}' + \right.$$

$$\left. + a_0^2 \int \mathbf{E}_L^0(a_0, \theta', \varphi', t)\,\mathrm{Re}\left\{ \sum_{l,n} \xi_{ln}(t)\,Y_{ln}(\theta', \varphi')\,e^{i\Omega_l t} \right\} do' \right], \quad (15)$$

where ϑ is the angle between the vectors $\mathbf{r}$ and $\mathbf{r}'$. The first summand in the right-hand side of Eq. (15) is usual elastic scattering at the frequency of incident radiation wave ω_0; at the same time, the second summand is the Raman scattering on the surface waves of the particle with the frequencies $\omega_0 \pm \Omega_l$.

The expansion coefficients of the full displacement of the drop surface $\xi_{ln}(t)$ satisfy the oscillation equation in the right-hand side of which the coercive force $f(t; a_0)$ is standing

$$\frac{d^2\xi_{ln}}{dt^2} + \frac{2}{t_l}\frac{d\xi_{ln}}{dt} + \Omega_l^2\,\xi_{ln} = \frac{l\,f_{ln}}{\rho_a\,a_0}\,. \quad (16)$$

Here $f_{ln} = \int f(t, a_0)\,Y_{ln}^*(\theta, \varphi)\,do'$, $t_l = \dfrac{a_0^2}{2\nu(2l+1)(l-1)}$

is the characteristic time of oscillation attenuation due to viscous forces, ν is the kinematic viscosity of the liquid. The initial conditions for Eq. (16) have the form $\xi_{ln}(0) = \dfrac{d\xi_{ln}(0)}{dt} = 0$.

The coercive force f is a jump of the normal component of the electric field strength on the drop surface

$$f = \frac{\varepsilon_a - 1}{8\pi}[(\varepsilon_a - 1)(\mathbf{E}(a_0, \theta, \varphi, t)\mathbf{n}_r)^2 + (\mathbf{E}(a_0, \theta, \varphi, t))^2]. \quad (17)$$

In this expression, one should take into account only the low frequency components as compared with the frequency of the exciting light field. As seen from Eq. (17), the form of the function $f(a_0, \theta, \varphi, t)$ depends on the angular structure of the internal electromagnetic field on the drop surface. The structure is given by the Mie solution (1).

Numerical calculations made by Eqs. (16)–(17) for water drops with different diffraction parameters[17] demonstrate that, for small particles ($x_a \ll 1$), the coercive force has maxima at the poles of the drop, in correspondence with the internal optical field distribution. For large drops ($x_a > 1$), they shift to equatorial zones.

This leads to the fact that the initial phase of oscillations of small drops differs from that of large drops by π/2. Small particles are deformed along the direction perpendicular to the direction of incident radiation, and large particles are deformed in parallel to the action. The amplitudes of oscillations are also different. For instance, $\xi_{max}/a_0 = 3 \cdot 10^{-5}$ for water drops with $x_a = 0.3$ (the incident radiation has the wavelength $\lambda_L = 0.53$ μm), and $\xi_{max}/a_0 = 10^{-3}$ for $x_a = 3$ if the radiant flux density is $I_0 = 10^8$ W/cm^2 and its duration is $t_p = 10^{-7}$ sec.

When the drop is irradiated by radiation modulated at the frequency $\Omega_{ex} \sim \Omega_l$, the drop oscillations become stimulated, and their frequency corresponds to the coercive force frequency after the transition to a steady state. This process is demonstrated in Fig. 4 where the value of relative displacement $\xi(t)/a_0$ of the surface of a water drop with radius $a_0 = 5$ μm in the directions $\theta = 180°$ and $\theta = 90°$ is presented as a function of time. The radiant flux density was 10^7 W/cm^2. The figure presents three situations: modulation frequency is lower than the frequency of drop oscillation at the fundamental mode, $\Omega_{ex}/\Omega_2 = 0.1$ (Fig. 4*a*); the modulation frequency is higher than the fundamental mode frequency, $\Omega_{ex}/\Omega_2 = 1$ (Fig. 4*c*); the case of resonant excitation of oscillations, $\Omega_{ex}/\Omega_2 = 2$ (Fig. 4*b*). As follows from the figure, the oscillation amplitude is essentially higher in the latter case, and it continues to grow in time what can cause the destruction of the particle under long action.[19]

An example of the numerical calculations of the intensity of a scattered light wave ($\lambda_L = 0.53$ μm, continuous radiation), namely, at oscillations of the surface of a water drop of radius $a_0 = 20$ μm, is presented in Fig. 5. The parameters of incident radiation pulse exciting the oscillations are as follows: $I_0 = 10^8$ W/cm^2, $t_p = 10^{-7}$ s. Figure 5*a* shows the function $\xi(t)/a_0$ along the direction $\theta = 180°$. Figure 5*b* presents the angular structure of the scattering signal (in the plane of the vectors $\mathbf{k}$ and $\mathbf{E}_L$) for the time moment $t = 13.5$ μs. Figure 5*c* presents the scattered signal along three directions (backwards, transversally, and in the direction of the primary rainbow) as a function of time. Dashed lines denote the signal of elastic scattering on an nondisturbed particle. As follows from Fig. 5*a*, after the end of the

laser pulse action the drop exhibits free damping oscillations at the fundamental (the lowest) natural frequency $\Omega_2 \approx 0.3$ MHz. In accordance with the oscillations, changes the scattered signal. Its temporal behavior takes the form of "flares" mentioned earlier in Ref. 18. The maximal changes relative to the level of nondisturbed scattering are observed along the direction transverse to the incident radiation and the direction of rainbow. This is natural because these are directions that are characterized by the fact that the maximum changes of the elastic scattering intensity with respect to angle occur here. Thus, the contribution of inelastic scattering becomes appreciable in the angles of the minimum of elastic scattering.

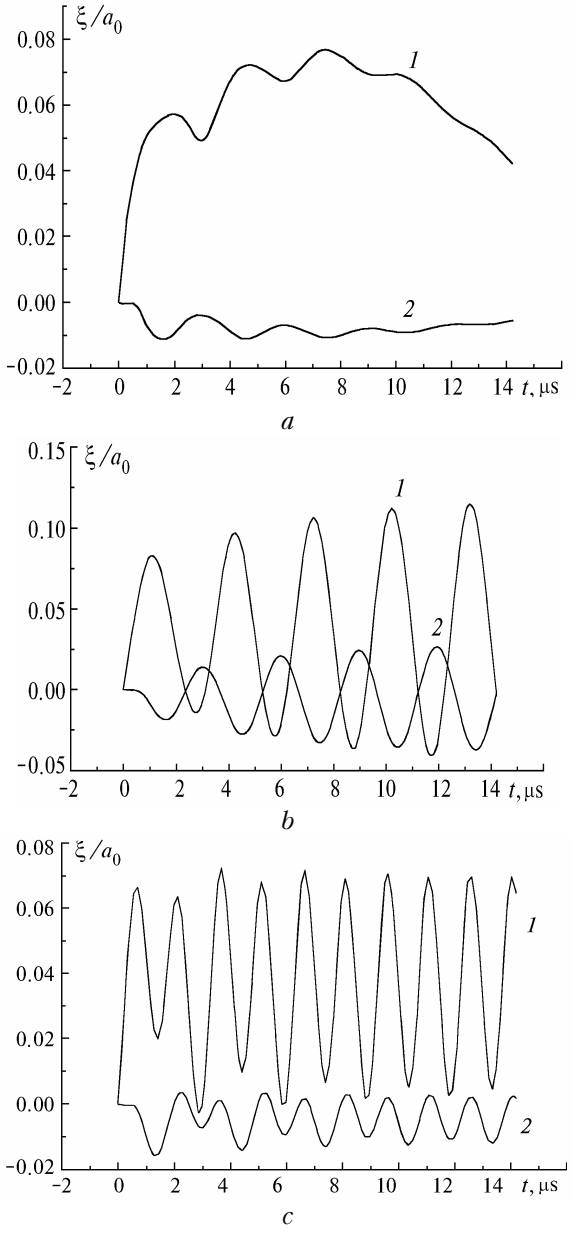

FIG. 4. Relative displacement of a water drop of radius $a_0 = 5$ μm for $\theta = 180°$ (1) and $\theta = 90°$ (2) as a function of time irradiated by modulated laser radiation ($\lambda_L = 0.53$ μm). The relative modulation frequency Ω_{ex}/Ω_2 is 0.1 (a), 0.1 (b), 2 (c).

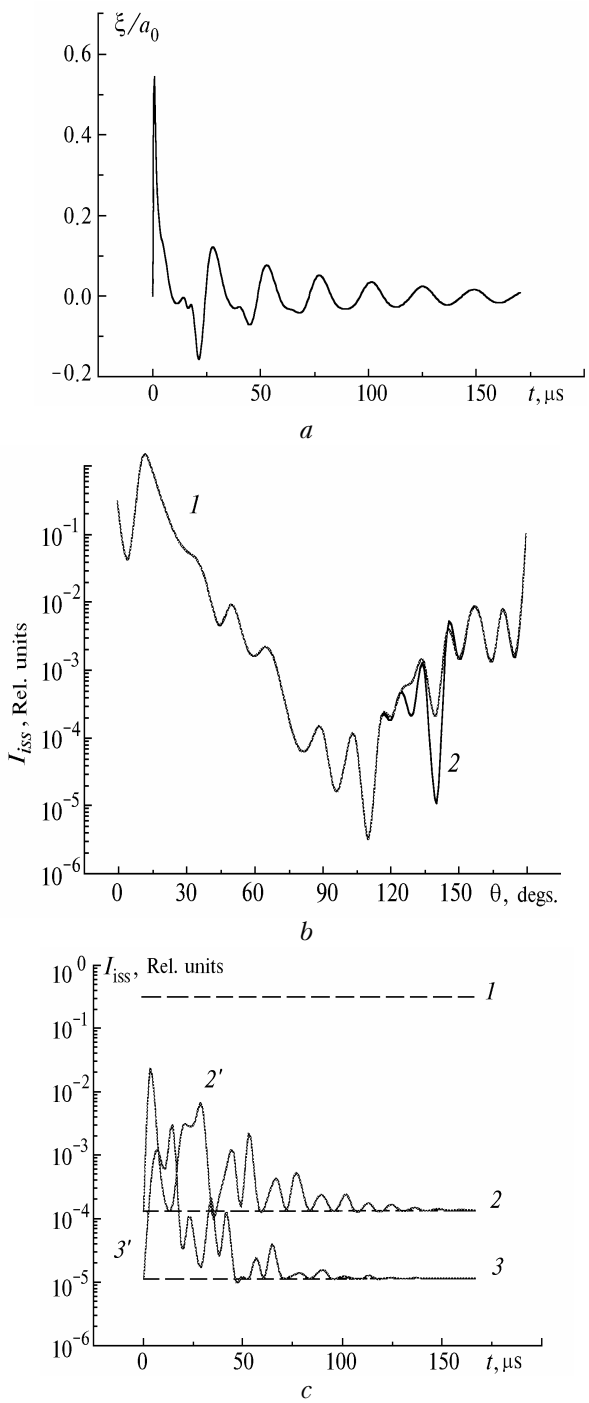

FIG. 5. Numerical calculations of the intensity of a scattered light wave: a) the relative displacement of a water drop of radius $a_0 = 20$ μm as a function of time along the direction $\theta = 180°$ due to ponderomotive forces at a pulse irradiation; b) angular function of radiation scattered on a water drop at the excitation of oscillations on its surface (1). The curve (2) is the elastic scattering on a nondisturbed drop; c) temporal function of radiation scattered on an oscillating water drop for different observation angles: $\theta = 0°$ (1, 1′), $\theta = 90°$ (2, 2′), $\theta = 137°$ (3, 3′). The curves 1, 2, 3 are levels of the elastic scattering signal in the absence of oscillations.

CONCLUSION

Let us briefly formulate the major results of the paper.

Nonlinear optical SRS, SMBS, SF effects in transparent particles as microresonators are described theoretically from unified physical viewpoints.

A relation taking into account resonance conditions of parametrically interacting fields for the energy threshold of stimulated scattering in a particle is established.

The SS angular parameters are studied. The SS directional pattern is shown to be less asymmetric along forward and backward directions.

The effect of Raman light scattering is theoretically studied for oscillations of the surface of liquid particles of arbitrary dimensions. The oscillations were induced by ponderomotive forces of intense light irradiation with different regimes of time modulation. It is established that the effect manifest itself along the direction close to the rainbow scattering angle, and along the direction transverse to the incident radiation.

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