INFLUENCE OF RESONANCE PROPERTIES OF TRANSPARENT PARTICLES ON THE STIMULATED BRILLOUIN SCATTERING THRESHOLD

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The effect of stimulated Brillouin scattering (SBS) in transparent spherical microparticles is considered theoretically. The resonance properties of particles, namely, Q-factor and the resonance contour width, are found to influence significantly the threshold characteristics of the SBS process. We have made numerical calculations of the overlapping coefficient of interacting optical pumping fields and the SBS inside a particle determining the magnitude of the SBS threshold intensity. The magnitude of the coefficient is shown to be dependent on the accuracy of correspondence of interacting field space profiles.

1. INTRODUCTION

The processes of stimulated light scattering in dielectric microparticles, such as stimulated Raman scattering (SRS), stimulated Brillouin scattering (SBS), and stimulated fluorescence have recently been the subject of an intensive study throughout the world.[1–16,19,21,22] These phenomena are of particular interest because a spherical particle is an optical microresonator, whose quality is sufficiently high and may be as much as 10^8 (Ref. 2). In this case a spherical particle can be used as a unique optical instrument suitable for solving many problems traditional both for the intracavity spectroscopy[4] and for the physics of aerosols.[1]

Among the nonlinear effects of stimulated light scattering by particles the SRS effect is of a primary practical interest because of a significant spectral separation of the generation lines. A large number of experimental and theoretical studies have been devoted to this phenomenon (a brief overview of the papers can be found in Ref. 22). At the same time it has been found experimentally that SRS in particles is frequently accompanied by other nonlinear process, which, as a rule, occurs well before the SRS and may result in a decrease of the SRS power threshold.[2,9,13,14] The SRS threshold intensity in the case of such a "cascadeB excitation decreases by a factor of 3 to 5. The observed effect is associated with the fact that the SBS threshold intensity is lower than the SRS threshold intensity due to higher amplification factor of the process. Therefore in some cases the SBS wave occurs well before the SRS wave and then it is a more effective source for pumping the SRS process than the incident radiation.

In this connection it is important to investigate the effect of resonance properties of transparent particles, namely, their Q-factor and the resonance contour spectral width, on the efficiency of spatial power redistribution of interacting optical fields affecting the SBS and SRS threshold intensity that is the goal of this work. Besides, the paper describes the basic aspects of the theory of SBS in spherical particles as well as the necessity is demonstrated to take into account the spatial structure of interacting acoustic and optical fields when studying the SBS in microresonators.

2. BASIC RELATIONS

Theoretical investigations along with a numerous experimental papers have shown that the initiation of stimulated light scattering in a spherical particle is conditioned by the existence of high-Q resonance electromagnetic modes capable of essentially increasing spontaneous scattering wave under conditions of phase synchronism. At present the following theoretical model of the SBS in a spherical particle has been formulated.[14]

As a result of electrostriction effect due to interaction of radiation with the particle matter throughout the particle volume there appear spontaneous inelastic light scattering, which is most intense in the focusing areas of the internal optical field (in Fig. 1).

Some waves from spontaneous scattering spectrum escape from a drop, while some waves propagate along its surface due to total internal reflection. On their way along wave propagation path these waves may experience both amplification and attenuation due to the radiation absorption and radiation escape through the surface. When the frequency of a wave from the scattering spectrum coincides with the frequency of a natural high-Q electromagnetic mode of a particle (several closely adjacent modes in the case of

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multimode process) the amplification of a spontaneous Stokes wave exceeds its net losses and the process of stimulated scattering starts in the particle.

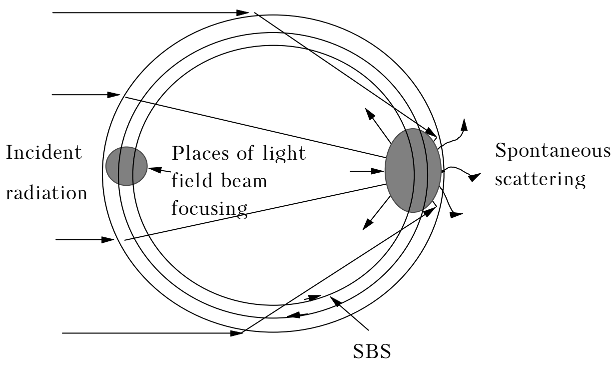

FIG. 1. A schematic diagram illustrating a model of SBS process in a spherical particle.

From the viewpoint of field formation in a resonator the field of stimulated scattering may be considered as a standing wave formed by superposition of electromagnetic waves propagating along opposing directions along a drop spherical surface provided that condition of phase synchronism holds

$\omega_s = \omega_L - \omega_{ac}$; $\mathbf{k}_s = \mathbf{k}_L - \mathbf{k}_{ac}$,

where ω_L, $\mathbf{k}_L$, ω_s, $\mathbf{k}_s$ are the frequencies and wave vectors of the incident and scattered electromagnetic waves, respectively; ω_{ac}, $\mathbf{k}_{ac}$ are the frequency and the wave vector of striction acoustic oscillations.

The initial equations for theoretical analysis of the SBS process in a particle are nonhomogeneous Maxwell equations where the nonlinear part $\mathbf{P}_N(\mathbf{r}, t)$ of the medium polarization $\mathbf{P}(\mathbf{r}, t)$, induced by the pumping field and closely related to a nonlinear scattering process being studied, is a field source for Raman-scattered waves

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

$$\mathrm{rot}\mathbf{H}(\mathbf{r}, t) = \frac{1}{c}\frac{\partial \mathbf{D}(\mathbf{r}, t)}{\partial t} + \frac{4\pi\sigma}{c}\mathbf{J}(\mathbf{r}, t);$$

$$\mathrm{div}\mathbf{H}(\mathbf{r}, t) = 0; \quad (1)$$

$$\mathbf{D}(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}, t) + 4\pi\,\mathbf{P}(\mathbf{r}, t); \ \mathbf{J}(\mathbf{r}, t) = \sigma\mathbf{E}(\mathbf{r}, t).$$

Here $\mathbf{E}$ and $\mathbf{H}$ are the electric and magnetic field vectors inside a particle, respectively; $\mathbf{D}$ is the electric bias vector; $\mathbf{J}$ is the polarization current density; c is the speed of light in vacuum; σ is the particle matter specific conductance. At the sphere boundary it is necessary that the condition holds of the field tangential components continuity.

This system of equations is known to be transformed to a wave equation for the electric field strength vector in a particle $\mathbf{E}(\mathbf{r}, t)$

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

It should be noted here that in the right-hand side of this equation there is the value of nonlinear polarization of a medium since we consider only the SBS process connected with a certain dipole transition.

Since electric field in a particle is a sum of fields at the fundamental frequency ω_L (pumping frequency) and at the scattered wave frequency ω_s:

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_L(\mathbf{r}, t) + \mathbf{E}_s(\mathbf{r}, t), \quad (3)$$

where

$$\mathbf{E}_L(\mathbf{r}, t) = \mathrm{Re}\{\tilde{\mathbf{E}}_L\, e^{-i\omega_L t}\} = \frac{1}{2}\tilde{\mathbf{E}}_L\, e^{-i\omega_L t} + \text{complex conjugate (c.c.)},$$

$$\mathbf{E}_s(\mathbf{r}, t) = \mathrm{Re}\{\tilde{\mathbf{E}}_s\, e^{-i\omega_s t}\} = \frac{1}{2}\tilde{\mathrm{E}}_s\, e^{-i\omega_s t} + \text{complex conjugate (c.c.)},$$

then the wave equation (2) is split into two related equations for the fundamental and scattered waves

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

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

Here

$$\mathbf{P}_s = \frac{1}{2}\tilde{\mathbf{P}}_s\, e^{-i\omega_s t} + \text{c.c.} = \chi^{(3)}(\omega_s)\,(\tilde{\mathbf{E}}_L\,\tilde{\mathbf{E}}_L^*)\,\tilde{\mathbf{E}}_s\, e^{-i\omega_s t} + \text{c.c.},$$

$$\mathbf{P}_L = \frac{1}{2}\tilde{\mathbf{P}}_L\, e^{-i\omega_L t} + \text{c.c.} = \chi^{(3)}(\omega_L)\,(\tilde{\mathbf{E}}_s\,\tilde{\mathbf{E}}_s^*)\,\tilde{\mathbf{E}}_L\, e^{-i\omega_L t} + \text{c.c};$$

$\chi^{(3)}$ is the nonlinear dielectric susceptibility of the 3rd order; $\tilde{\mathbf{E}}_L$ and $\tilde{\mathbf{E}}_s$ are the amplitudes of the incident field and the field of SBS wave, respectively; $\mathbf{P}_N = \mathbf{P}_L + \mathbf{P}_s$. Note that formally there may appear infinitely many scattering waves with the frequencies $\omega = \omega_L \pm n\omega_{ac}$, $n = 1 \ldots \infty$. However, we shall consider only the first Stokes wave with $\omega_s = \omega_L - \omega_{ac}$ as being the most intense one. In this case the permutation symmetry of $\chi^{(3)}(\omega_s) = \{\chi^{(3)}(\omega_L)\}^*$ is valid.[18]

Equations for the field (4) are supplemented by an equation for the pressure increase in the medium $p(\mathbf{r}, t)$

$$\nabla^2 p(\mathbf{r}, t) - \frac{1}{c_s^2}\frac{\partial^2 p(\mathbf{r}, t)}{\partial t^2} + \frac{2\,\Gamma_B}{c_s^2}\frac{\partial p(\mathbf{r}, t)}{\partial t} =$$

$$= \frac{\gamma}{8\pi}\nabla^2 |\mathbf{E}(\mathbf{r}, t)|^2, \qquad (5)$$

$$p(\mathbf{r}, t = 0) = 0$$

and by the relationship between the pressure and nonlinear polarization $\mathbf{P}_N$ of the medium

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

Here c_s and Γ_B are the velocity and the attenuation factor of hypersound (the line halfwidth of spontaneous Brillouin scattering); γ is the electrostriction constant; ρ_a is the particle matter density.

The boundary condition for (5) is the generalized form of the Laplace equation[19]:

$$\Big\{ p(\mathbf{r}, t)|_a - \frac{\gamma}{8\pi}\Big|\mathbf{E}(\mathbf{r}, t)|_a\Big|^2 - \beta_s\Big(\frac{1}{R_1(\mathbf{r})} +$$

$$+ \frac{1}{R_2(\mathbf{r})} - \frac{2}{a_0}\Big) + f\Big\}\, n_i = \eta\,\Big(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\Big)\, n_k,$$

where $R_1(\mathbf{r})$ and $R_2(\mathbf{r})$ are the main radii of the particle surface curvature at a point with the radius-vector $\mathbf{r}$; η and v denote the dynamic viscosity and the liquid velocity; $x_{i,k}$ denotes the coordinates; $n_{i,k}$ denotes the vector components of the outward normal to the particle surface $\mathbf{n}_r$; β_s is the tension coefficient of liquid surface; a_0 is the particle radius;

$$f(\mathbf{r}, t) = (\varepsilon_a - 1)/(8\pi)\;[(\varepsilon_a - 1)\;(\mathbf{E}(\mathbf{r}, t)\bullet\mathbf{n}_r)^2 +$$

$$+\; |\mathbf{E}(\mathbf{r}, t)|^2]$$

is the jump in the normal component of electromagnetic field tension at the liquid surface[20]; ε_a is the linear part of the dielectric constant of a particle substance. The symbol $|_a$ denotes that the values of corresponding quantities are taken at the particle boundary.

Since we consider the field formation in a volume optical resonator, that is in a transparent spherical particle, then in what follows we represent the solution of wave equations for the fields (4) in the form of its expansion over normal electromagnetic vibrational modes of the particle-resonator $\mathbf{E}_{nl}(\mathbf{r})$ having natural frequencies ω_{nl}. For example, for the Stokes wave

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

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

$$\frac{d^2 A_{nl}(t)}{dt^2} + 4\pi\sigma\,\frac{d\, A_{nl}(t)}{dt} + \omega_{nl}^2\, A_{nl}(t) =$$

$$= -\,4\pi \int_V \mathbf{E}_{nl}(\mathbf{r}')\,\frac{\partial^2\, \mathbf{P}_s(\mathbf{r}', t)}{\partial t^2}\, d\mathbf{r}'. \qquad (7)$$

Integration in Eq. (7) is made over the particle volume.

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

$$p(\mathbf{r}, t) = \sum_{n;l;m} \wp_{nlm}(t)\; o_{\,nlm}(\mathbf{r}). \qquad (8)$$

Solution of the set of equations (7) coupled with equations (4) and corresponding initial and boundary conditions makes it possible to comprehensively describe the process of stimulated scattering in a particle.

Note that in (6) the system of partial TE and TH waves (or their linear combination) is used as eigenfunctions, whose type follows from the Mie solution to the problem of diffraction of a plane electromagnetic wave on a sphere[15]:

$$\mathbf{E}_{nl}(\mathbf{r}) =$$

$$= \begin{cases} b_n(x_a)\,\mathbf{M}_{nl}(\mathbf{r})\,\psi_n(k_a\, r) & \text{for TE-wave;} \\ 1/k_a\, c_n(x_a)\,\nabla\,[\mathbf{M}_{nl}(\mathbf{r})\,\psi_n(k_a\, r)] & \text{for TM-wave,} \end{cases}$$

where k_a is the wave number inside a particle; $b_n(x_a)$, $c_n(x_a)$ are the amplitudes of partial waves; $\mathbf{M}_{ln}(\mathbf{r})$ are the spherical vector-harmonics; $x_a = \dfrac{2\pi a_0}{\lambda_L}$ is the particle diffraction parameter.

Under conditions when no pressure increase occurs at the particle surface, $p(\mathbf{r}, t)|_a \approx 0$, that is a good approximation when studying the SBS,[15] the functions $o_{\,nlm}(\mathbf{r})$, have the view

$$o_{\,nlm}(\mathbf{r}) = C_{nm}\;\psi_n\;(\alpha_{nm}\,\mathbf{r}/a_0)\;Y_{nl}(\theta, \varphi),$$

where $C_{nm} = 2/[a_0^3\,\psi_{n+1}\,(\alpha_{nm})^2)$ are the normalization constants; α_{nm} is the mth zero of the spherical Bessel function ψ_n; θ and φ are the spherical coordinates.

The eigenfunctions obey the orthogonality conditions

$$\int_V \mathbf{E}_{nl}(\mathbf{r})\, \mathbf{E}^*_{km}(\mathbf{r})\, \mathrm{d}\mathbf{r}' = \delta_{nk}\, \delta_{lm};$$

$$\int_V o_{nlm}(\mathbf{r})\, o^*_{kij}(\mathbf{r})\, \mathrm{d}\mathbf{r}' = \delta_{nk}\, \delta_{li}\, \delta_{mj}. \quad (9)$$

3. SBS THRESHOLD IN A SPHERICAL PARTICLE

Let us write the energy balance equation in a particle at the scattered wave[10] frequency:

$$\frac{\mathrm{d}W_\mathrm{s}}{\mathrm{d}t} = P_g - (P_a + P_\mathrm{r}). \quad (10)$$

In Eq. (10) the following designations are introduced; W_s is the electromagnetic field energy averaged over the oscillation period accumulated in the particle volume; P_r is the mean power of radiation losses (the radiation escaped through the particle surface); P_a is the mean power of thermal losses inside the particle; $P_g = -\frac{1}{2}\int_V \mathrm{Re}\left\{\tilde{\mathbf{E}}^*_\mathrm{s}\, \frac{\partial\, \tilde{\mathbf{P}}_\mathrm{s}}{\partial t}\right\} \mathrm{d}\mathbf{r}'$ is the mean power of the Stokes wave sources; V is the particle volume.

In what follows we introduce the concept of Q-factor of the droplet resonator at the resonance mode frequency ω_{nl} supporting the SBS process ($\omega_{nl} = \omega_\mathrm{s}$):

$$Q = \omega_\mathrm{s}\, W_\mathrm{m}/(P_a + P_\mathrm{r}),$$

where W_m is the electromagnetic field energy averaged over the oscillation period of a mode. Then the law of energy conservation for the Stokes wave in the particle may be written as

$$\frac{\mathrm{d}W_\mathrm{s}}{\mathrm{d}t} = P_g - \omega_\mathrm{s}\, \frac{y_\mathrm{m}\, W_\mathrm{s}}{Q}\,, \quad (11)$$

where the ratio $W_\mathrm{m}/W_\mathrm{s}$ is denoted as y_m.

It should be noted that in open resonators, such as a transparent spherical particle, the fields of their eigenmodes are not limited by the resonator surface and extend over it. For this reason we always have $y_\mathrm{m} > 1$. However, as calculations[21] showed in the majority of cases the coefficient y_m is approximately equal to unity for high-Q eigenmodes of spherical particles and its deviation from unit is 2–5%. In the subsequent calculations it is taken to be $y_\mathrm{m} = 1$.

Let us now consider the component determining the power of a Stokes wave source. In the framework of the theoretical model accepted of the process for nonlinear polarization at the frequency ω_s the following expression is used:

$$\tilde{\mathbf{P}}_\mathrm{s} = \chi^{(3)}\, (\tilde{\mathbf{E}}_\mathrm{L}\, \tilde{\mathbf{E}}^*_\mathrm{L})\tilde{\mathbf{E}}_\mathrm{s}. \quad (12)$$

By assuming that time behavior of complex amplitudes is slower than that of the exponential factor, i.e., according to the so-called approximation of slowly varying amplitudes, this expression is modified for the source power of inelastic scattering wave as follows:

$$P_g = -\frac{1}{2}\int_V \mathrm{Re}\left\{\tilde{\mathbf{E}}^*_\mathrm{s}\, \frac{\partial\, \tilde{\mathbf{P}}_\mathrm{s}}{\partial t}\right\} \mathrm{d}\mathbf{r}' =$$

$$= -\frac{\omega_\mathrm{s}}{2}\int_V \mathrm{Im}\,\{\chi^{(3)}\}\, (\tilde{\mathbf{E}}_\mathrm{s}\, \tilde{\mathbf{E}}^*_\mathrm{s})\, (\tilde{\mathbf{E}}_\mathrm{L}\, \tilde{\mathbf{E}}^*_\mathrm{L})\, \mathrm{d}\mathbf{r}'. \quad (13)$$

Taking into account the fact that the Stokes wave amplification factor g_s at stimulated scattering is related to the nonlinear medium susceptibility $\chi^{(3)}$ as follows:

$$g_\mathrm{s} = -\, 32\pi^2\, \omega_\mathrm{s}/(c^2\varepsilon_a)\, \mathrm{Im}\,\{\chi^{(3)}\},$$

the expression (13) reduces to the form

$$P_g = \frac{c^2\, \varepsilon_a}{64\pi^2}\int_V g_\mathrm{s}\, (\tilde{\mathbf{E}}_\mathrm{s}\, \tilde{\mathbf{E}}^*_\mathrm{s})\, (\tilde{\mathbf{E}}_\mathrm{L}\, \tilde{\mathbf{E}}^*_\mathrm{L})\, \mathrm{d}\mathbf{r}' =$$

$$= g_\mathrm{s}\int_V I_\mathrm{s}(\mathbf{r})\, I_\mathrm{L}(\mathbf{r})\, \mathrm{d}\mathbf{r}',$$

where the radiation intensity

$$I(\mathbf{r}) = cn_a/(8\pi)(\tilde{\mathbf{E}}\tilde{\mathbf{E}}^*)$$

and $n_a = \sqrt{\varepsilon_a}$ is the medium refractive index.

The condition for the SBS to occur is determined as $\frac{\mathrm{d}W_\mathrm{s}}{\mathrm{d}t} = 0$, and, consequently, from Eq. (11) we have

$$\omega_\mathrm{s} n_a/(cQ)\int_V I_\mathrm{s}(\mathbf{r})\, \mathrm{d}\mathbf{r}' = g_\mathrm{s}\int_V I_\mathrm{s}(\mathbf{r})\, I_\mathrm{L}(\mathbf{r})\, \mathrm{d}\mathbf{r}'\,.$$

The Stokes wave intensity I_s and the pumping wave intensity I_L inside a particle may be represented as products, where the amplitude independent of space coordinates is separated out in the form of a factor

$I_L = I_{L0}\,B_L(\mathbf{r});\; I_s = I_{s0}\,B_s(\mathbf{r}),$

where B_L and B_s are dimensionless factors of inhomogeneity in the internal optical fields. Since I_{L0} is actually the intensity of a wave incident on the particle, the threshold pumping radiation intensity, above which the stimulated scattering occurs is

$$I_t = 2\pi\, n_a/[g_s\, Q(\omega_s)\, \lambda_s\, B_c(\omega_L, \omega_s)], \qquad (14)$$

where λ_s is the wavelength of scattered light; $B_c(\omega_L, \omega_s)$ is the integral coefficient that allows for the spatial overlap of interacting optical fields inside the particle[22]:

$$B_c(\omega_L, \omega_s) = \left(\int_V B_s(\mathbf{r})\, d\mathbf{r}'\right)^{-1} \int_V B_s(\mathbf{r})\, B_L(\mathbf{r})\, d\mathbf{r}'.$$

As follows from this expression, the coefficient B_c or, in fact, the efficiency of the optical field interaction in the particle is directly proportional to the pumping field intensity (through the coefficient B_L). The closer is coincidence of the interacting fields the higher is the value of the coefficient B_c. It is evident that the maximum value of B_c should be expected in the cases when both the pumping field and the SBS field are simultaneously in resonance with a natural electromagnetic mode of a particle, i.e., in the so-called double resonance. Moreover, since the frequency shift of a SBS wave, $\Delta\omega = \omega_L - \omega_s$, is rather small both spectral lines may be within one and the same resonance contour $\omega_L \approx \omega_s = \omega_{nl}$. This situation is schematically shown in Fig. 2*a*, where the halfwidth of the resonance curve of a particle natural mode is denoted as $\Gamma = x_a/Q$.

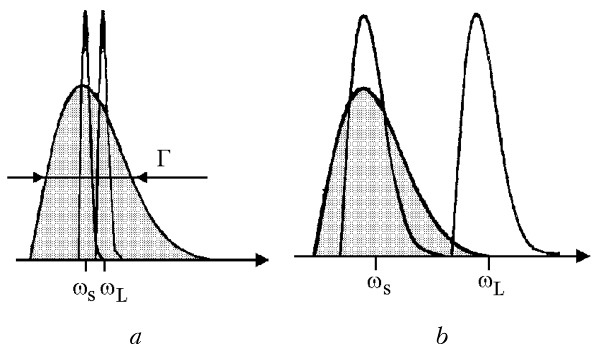

FIG. 2. A schematic diagram of relative position of the pumping line and SBS contours in the case of double (a) and single (b) resonances. A shaded contour is a resonance curve of one of normal modes of a particle.

As known the value Γ for the near surface natural modes decreases[6] with increasing particle size and, hence, at $\Delta\omega \geq \Gamma$ the condition of double resonance will be violated (Fig. 2*b*) that results in a decrease of the coefficient B_c.

The results of numerical calculations of the dependence $b_c(x_a)$ are shown in Fig. 3. Different curves in Fig. 3 correspond to different orders of resonance modes supporting the SBS such a separation becomes clear if one allows for the fact that the modes of different numbers and orders (i.e., with different set of indices n and l) can have close values of the diffraction resonance parameter x_a, and in this case the magnitude of Q-factor of these modes, namely, the quantity B_c, are quite different. However, the question on which of these mode is basic for the Stokes wave amplification is still to be addressed, within the theoretical model considered.

Figure 3 shows that the values of the coefficient B_c can either decrease or increase with the growth of the particle diffraction parameter depending on the order of the resonance chosen. This fact points to violation of the double resonance in the first case and to the growth of the efficiency of field interaction in the second case. For a comparison Fig. 3 also presents calculated values of $B_c(x_a)$ for the SRS process in water drops. In these calculations we used the most intense band of stretching vibrations of water, with $\Delta\omega = 3500\ \text{cm}^{-1}$ that provides for the only resonance and only for the SRS field. As is seen from Fig. 3 the values B_c for the SRS process, are much lower than that for SBS. This values, in fact, show the lower level to which the magnitude B_c tends with increasing particle size ($x_a \gg 1$).

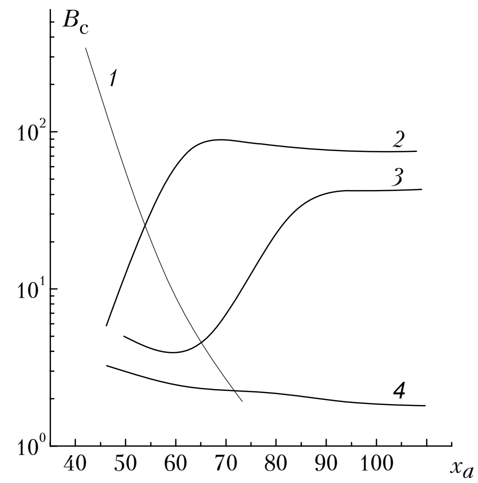

FIG. 3. The coefficient of spatial overlapping of optical fields in water droplets, B_c, as a function of x_a for the case of SBS (curves 1 to 3) and SRS (4). Calculations for SBS have been made using the normal modes TE_{1l} (curve 1), TE_{2l} (curve 2), and TE_{3l} (curve 3) of particles.

As follows from the above the efficiency of interaction between optical fields strongly depends on the ratio between the values $\Delta\omega$ and Γ. Therefore it is worth considering the dependence of coefficient B_c on the parameter $\Delta\omega/c$. This dependence obtained by averaging the results over more than twenty different combinations of mode indices n and l, is depicted in Fig. 4. The ascending portion of the curve in Fig. 4 corresponds to double resonance ($\Delta\omega \ll c$) and shows the increase in Q-factor of normal modes of the particle with increasing particle size (see Fig. 5).

In this case the value B_L grows and, as a consequence, the value of the spatial overlapping coefficient B_c increases. Narrowing of the resonance contour and subsequent break of the double resonance define the descending curve of the dependence $b_c(\Delta\omega/c)$.

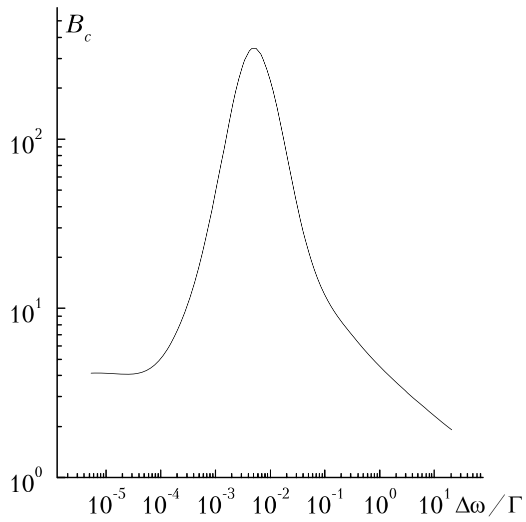

FIG. 4. The dependence of field spatial overlap coefficient B_c on the parameter $\Delta\omega/\Gamma$ in water drops.

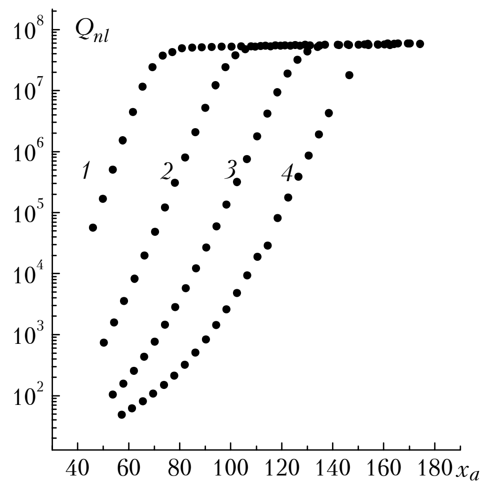

FIG. 5. Q-factor of Q_{nl} resonances of different orders internal optical field depending on the diffraction parameter of water drops x_a (the absorption coefficient $\varkappa_a = 10^{-8}$).

4. NONLINEAR SUSCEPTIBILITY OF $\chi^{(3)}$ IN THE SBS PROCESS IN MICRORESONATORS

Equation (14) is used to calculate the threshold of stimulated scattering in particles of different types (SRS, SBS, stimulated fluorescence), however, the characteristics of each specific process are determined by the gain factor g_s for a scattered wave, or more specifically, by a particular type of nonlinear susceptibility of the medium $\chi^{(3)}$. In addition, the resonance characteristics of particles manifest themselves in this parameter. Let us now consider the parameter $\chi^{(3)}$ in a more detail for the process of SBS in transparent particles.

As was noted above, the influence of light on medium results in additional nonlinear polarization of the medium according to Eq. (12). Introducing the complex pressure amplitude as $p(\mathbf{r}, t) = \frac{1}{2}\,\tilde{p}\,e^{-i\omega_{ac}t} +$ + c.c., the nonlinear polarization at the scattered wave frequency $\omega_s = \omega_L - \omega_{ac}$ may be written as

$$\mathbf{P}_s(\mathbf{r}, t) = \gamma\,\tilde{p}^*(\mathbf{r}, t)\,\tilde{\mathbf{E}}_L\,e^{-i\omega_s t}/(8\pi\,c_s^2\rho_a) + \text{c.c.},$$

from where the expression is derived for nonlinear susceptibility at SBS

$$\chi^{(3)}(\omega_s) = \gamma\,\tilde{p}^*\,(\tilde{\mathbf{E}}_L^*\,\tilde{\mathbf{E}}_s)^{-1}/(8\pi\,c_s^2\rho_a)\,. \qquad (15)$$

Let us now return to the pressure equation (5), and let a solution to it be written as a series expansion over the eigenfunctions of the droplet-resonator (8). By substituting Eq. (15) into Eq. (5) and integrating over the droplet volume, taking into account the orthogonality of eigenfunctions according to condition (9), we obtain the equation for the expansion coefficients

$$\frac{\partial^2 \wp_{nlm}}{\partial t^2} - 2c_B\,\frac{\partial \wp_{nlm}}{\partial t} - \Omega_{nlm}^2\,\wp_{nlm} =$$

$$= -\,e^{-i\omega_{ac}t}\,\frac{\gamma\,c_s^2}{8\pi}\int_V \nabla^2\,(\tilde{\mathbf{E}}_s^*\,\tilde{\mathbf{E}}_L)\,o_{nlm}^*\,d\mathbf{r}', \qquad (16)$$

where Ω_{nlm} denotes the natural acoustic frequencies of the resonator.

Because in the investigation of threshold characteristics we deal with the steady state fields, whose amplitudes $\tilde{\mathbf{E}}$ and $\tilde{p}$ do not depend on time, then for each of the fields $\mathbf{E}_L$ and $\mathbf{E}_s$ the wave equations (4) reduce to the Helmholtz equation and solution (16) takes the form

$$\wp_{nlm} = \frac{\gamma\,c_s^2\,k_L^2}{4\pi}\,\frac{e^{-i\omega_{ac}t}}{\omega_{ac}^2 - \Omega_{nlm}^2 + 2i\,c_B\,\omega_{ac}} \times$$

$$\times \int_V (\tilde{\mathbf{E}}_s^* \, \tilde{\mathbf{E}}_L) \, o^*_{nlm}(\mathbf{r}') \, d\mathbf{r}'. \qquad (17)$$

Having multiplied Eq. (18) by Π_{nlm} and summing over all normal acoustic modes of the resonator we obtain the law of pressure variation in a droplet in the case of the SBS process

$$p(\mathbf{r}, t) = \frac{\gamma \, c_s^2 \, k_L^2}{4\pi} \sum_{n;l;m} \frac{e^{-i\omega_{ac}t}}{\omega_{ac}^2 - \Omega_{nlm}^2 + 2i \, c_B \, \omega_{ac}} \times$$

$$\times \, o_{nlm}(\mathbf{r}) \int_V (\tilde{\mathbf{E}}_s^* \, \tilde{\mathbf{E}}_L) \, o^*_{nlm}(\mathbf{r}') \, d\mathbf{r}'. \qquad (18)$$

The quantity characterizing the frequency shift of a generated acoustic wave from the resonance is

$$\omega_{ac}^2 - \Omega_{nlm}^2 + 2i \, c_B \, \omega_{ac} = 2i \, c_B \, \omega_{ac} \, (1 - i \, d_{nlm}) \equiv D_{nlm},$$

where $d_{nlm} = [(\omega_{ac} - \Omega_{nlm})/\omega_{ac}]\,[\Omega_{nlm}/c_B(\Omega_{nlm})]$. Let us denote $Q_{nlm}^{ac} = \Omega_{nlm}/c_B(\Omega_{nlm})$, that is the droplet Q-factor for a normal acoustic mode. Then we have, for the reduced frequency shift

$$d_{nlm} = Q_{nlm}^{ac} \, (\omega_{ac} - \Omega_{nlm})/\omega_{ac} \, .$$

As earlier we present the complex amplitude $\tilde{\mathbf{E}}_L$ of the field as a product of a time-dependent amplitude and a spatial function: $\tilde{\mathbf{E}}_L(\mathbf{r}, t) = E_L(t) \, \mathbf{b}_L(\mathbf{r})$. Similar expression may be written for $\tilde{\mathbf{E}}_s$. Note that earlier introduced function of the internal field inhomogeneity $B_L(\mathbf{r})$ is related to $\mathbf{b}_L(\mathbf{r})$ by an obvious relationship

$$B_L(\mathbf{r}) = \frac{\tilde{\mathbf{E}}_L \, \tilde{\mathbf{E}}_L^*}{E_L \, E_L^*} = \mathbf{b}_L(\mathbf{r}) \, \mathbf{b}_L^*(\mathbf{r}).$$

As a result we obtain from Eq. (18) the following formula:

$$p(\mathbf{r}, t) = e^{-i\omega_{ac}t} \, \frac{\gamma \, c_s^2 \, k_L^2}{4\pi} \, E_L \, E_s^* \times$$

$$\times \sum_{n;l;m} \frac{o_{nlm}(\mathbf{r})}{D_{nlm}(\mathbf{r})} \int_V \mathbf{b}_L(\mathbf{r}') \, \mathbf{b}_s^*(\mathbf{r}') \, o^*_{nlm}(\mathbf{r}') \, d\mathbf{r}'.$$

By denoting the integral in the right-hand side of the equation as

$$K_{nlm} = \int_V \mathbf{b}_s(\mathbf{r}') \, \mathbf{b}_L^*(\mathbf{r}') \, o_{nlm}(\mathbf{r}') \, d\mathbf{r}', \qquad (19)$$

we obtain the final expression for pressure inside a particle

$$p(\mathbf{r}, t) = e^{-i\omega_{ac}t} \, \frac{\gamma \, c_s^2 \, k_L^2}{4\pi} \, E_L \, E_s^* \sum_{n;l;m} \frac{o_{nlm}(\mathbf{r}) \, K_{nlm}^*}{D_{nlm}(\mathbf{r})} \, . \qquad (20)$$

Taking into account this result one obtains, from (15), the expression for the medium nonlinear susceptibility characteristic of the SBS process at the scattered wave frequency

$$\chi^{(3)}(\omega_s) = \frac{\gamma^2 \, \omega_L^2 \, n_a^2}{16\pi^2 \, c^2 \rho_a \, \mathbf{b}_s(\mathbf{r}) \, \mathbf{b}_L^*(\mathbf{r})} \sum_{n;l;m} \frac{o_{nlm}(\mathbf{r}) \, K_{nlm}^*}{D_{nlm}(\mathbf{r})} \, . \qquad (21)$$

In the case of an acoustic resonance $\omega_{ac} = \Omega_{nlm}$, when the frequency difference d_{nlm} equals zero, the summation in the right-hand side of Eq. (23) vanishes, and we have

$$\chi^{(3)}(\omega_s) = -\frac{i \, \gamma^2 \, \omega_L^2 \, n_a^2}{32\pi^2 \, c^2 \rho_a \, \omega_{ac} \, \Gamma(\omega_{ac})} \times$$

$$\times \frac{o\,(\mathbf{r}; \Omega_{nlm} = \omega_{ac}) \, K^*(\omega_{ac})}{\mathbf{b}_s(\mathbf{r}) \, \mathbf{b}_L^*(\mathbf{r})} =$$

$$= \chi_\infty^{(3)} \, \frac{o\,(\mathbf{r}; \Omega_{nlm} = \omega_{ac}) \, K^*(\omega_{ac})}{\mathbf{b}_s(\mathbf{r}) \, \mathbf{b}_L^*(\mathbf{r})},$$

where $\chi_\infty^{(3)}$ denotes the nonlinear susceptibility of an extended medium in the SBS process (optical cell).[17]

As can be seen from the expression derived, specific features of stimulated scattering formation on the acoustic waves in a spherical particle is the interference among spatial structures of the interacting pumping fields, acoustic, and scattered waves. The integral coefficient K_{nlm}, introduced by Eq. (19), allows for this mutual influence, and its value strongly depends on the resonance characteristics of a particle. At stimulated scattering in an extended medium the set of eigenfunctions $\mathbf{E}_{nl}(\mathbf{r})$ and $o_{nlm}(\mathbf{r})$ is replaced by plane waves. In this case no summation over indices is needed[15]

$$\mathbf{E}_{nl}(\mathbf{r})\big|_L \sim e^{-i(\mathbf{k}_L\mathbf{r})}; \; \mathbf{E}_{nl}(\mathbf{r})\big|_s \sim e^{-i(\mathbf{k}_s\mathbf{r})};$$

$$o_{nlm}(\mathbf{r}) \sim e^{-i(\mathbf{k}_{ac}\mathbf{r})}.$$

In the case of SBS effect in a counter wave

$$K_{nlm} \sim e^{-i(\mathbf{k}_L - \mathbf{k}_s - \mathbf{k}_{ac}) \, \mathbf{r}} = 1.$$

The numerical calculations of the coefficient K_{nlm} are beyond the scope of this paper, and will make the subject of other studies.

The threshold values of I_t for the SRS and SBS processes in water droplets of different radii (the absorption index $\varkappa_a = 10^{-8}$), calculated using (14), are shown in Fig. 6.

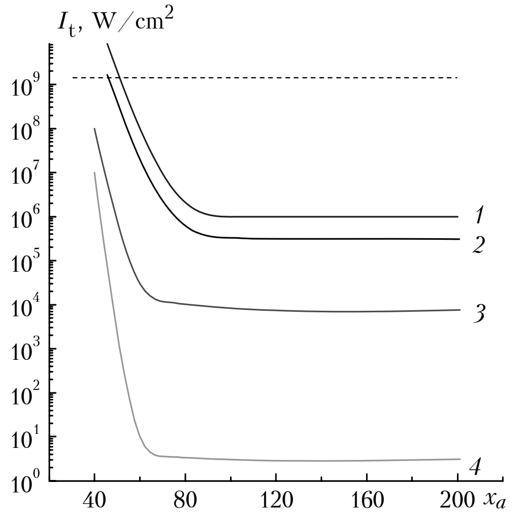

FIG. 6. Theoretical dependence of the SRS intensity threshold (1) and of SBS (2–4) in water drops of different size[22]*: single (1, 2), double (3), and triple resonances of SBS (4).*[12] *The dashed curve denotes the optical breakdown threshold of water drops.*[9]

The dashed line in the figure shows the optical breakdown threshold of water droplets.[9] For SBS three situations were considered: single resonance (only for the Stokes wave $\Delta\omega/c \geq 0.5$), double resonance (simultaneously for the pumping wave and scattered wave $\Delta\omega/c \sim 10^{-2}$) and triple resonance (for pumping, scattered and acoustic waves). In this case the value of the SBS intensity threshold is anomalously low because the acoustic wave induced by the pumping radiation falls into one of the acoustic resonances of the particle thus resulting in a sharp rise of the amplitude of the dielectric constant perturbation in the medium and, consequently, in a nonlinear polarization. Similarly triple resonance occurs very rarely and, evidently, it can be obtained only at simultaneous excitation with laser and acoustic waves of corresponding frequencies.[12] Figure 6 shows that the SBS threshold intensity is lower than the SRS threshold intensity. As noted above, this is connected with a larger value of the SBS gain factor ($g_s \approx 5\cdot10^{-3}$ cm/MW) as compared with the corresponding value for the SRS process ($g_s \approx 10^{-3}$ cm/MW[18]). As to the dependence of I_t on the droplet radius, one can see from the figure that the threshold intensity sharply increases with the droplet size decrease due to similar drop in the Q factor of small particles for radiation. At $x_a \geq 100$ the value of I_t is practically independent of the radius of liquid particles because of a limited rise of the resonance Q-factor due to the absorption losses in liquid. For the particles of moderate size ($x_a \leq 40$) the optical breakdown inside the particles may prevent the initiation of the effects of stimulated light scattering.

5. CONCLUSION

Let us now formulate the main results of the study presented. We have established that the resonance characteristics of transparent microparticles, i.e., their Q factor and the resonance contour width, may have a noticeable effect on the threshold characteristics of the stimulated light scattering processes initiated in transparent microparticles.

The investigations made revealed that the value of the overlap coefficient B_c for interacting optical fields of the pump and SBS inside the particle. Its value determining the value of the SBS threshold intensity, depends on the accuracy with which their spatial profiles coincide. In this case the values of B_c essentially increase when the frequency shift between the pumping waves and SBS is less than the halfwidth of the line of a natural particle resonance, i.e., $\Delta\omega < \Gamma$ (double resonance). The increase in particle size leads to excitation of higher quality surface modes and, hence, to a decrease in Γ. The condition of double resonance is violated ($\Delta\omega > \Gamma$), and the values of the coefficient B_c decrease.

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REFERENCES

1. A.S. Serpenguzer, J.C. Swindal, R.K. Chang, and W.P. Acker, Appl. Opt. **31**, No. 18, 3543–3551 (1992).
2. J.-Z. Zhang, G. Chen, and R.K. Chang, J. Opt. Soc. Am. B. **7**, No. 1, 108–115 (1990).
3. S.-H. Qian, J.B. Snow, and R.K. Chang, Opt. Lett. **10**, No. 10, 499–501 (1985).
4. R.K. Chang, S.-H. Qian, and J. Eickmans, in: *Proceedings of Symposium on the Laser Spectroscopy Methods*, Israel (1985), pp. 1–10.
5. J.B. Snow, S.-H. Qian, and R.K. Chang, Opt. News **12**, No. 5, 5–7 (1986).
6. J.-Z. Zhang, D.H. Leach, and R.K. Chang, Opt. Lett. **13**, No. 4, 270–272 (1988).
7. H.-M. Tzeng, M.B. Long, R.K. Chang, and P.W. Barber, Proc. SPIE **573**, 80–83 (1985).
8. S.-H. Qian and R.K. Chang, Phys. Rev. Lett. **56**, No. 9, 926–929 (1986).
9. R.G. Pinnick, A. Biswas, J. Pendleton, and R.S. Armstrong, Appl. Opt. **31**, No. 3, 311–317 (1992).
10. Yu.E. Geints, A.A. Zemlyanov, and E.K. Chistyakova, Dep. v VINITI, No. 76–B97, Jan. 10, Moscow (1997).
11. G.V. Belokopytov and N.P. Pushechkin, Pis'ma Zh. Tekh. Fiz. **17**, Is. 22, 71–75 (1991).

12. G.V. Belokopytov and N.P. Pushechkin, Izv. Vyssh. Uchebn. Zaved., Ser. Radiofiz. **35**, No. 6–7, 498–509 (1992).
13. J.-Z. Zhang and R.K. Chang, J. Opt. Soc. Am. B. **6**, No. 27, 151–153 (1989).
14. A.S. Serpenguzer, G. Chen, and R.K. Chang, J. Opt. Soc. Am. B. **9**, No. 6, 871–883 (1992).
15. C.D. Cantrell, J. Opt. Soc. Am. B. **8**, No. 10, 2158–2180 (1991).
16. E.M. Khaled and S.C. Hill, IEEE Transactions on Antennas and Propagation **41**, No. 3, 295–303 (1993).
17. V.V. Kuz'min, *Nonlinear Optics and Molecular Light Scattering.* Transactions of FIAN (Nauka, Moscow, 1991), 152 pp.
18. J.P. Shen, *Principles of Nonlinear Optics* (Nauka, Moscow, 1989), 557 pp.
19. Yu.E. Geints and A.A. Zemlyanov, Atmos. Oceanic Opt. **9**, No. 10, 854–858 (1996).
20. L.D. Landau and E.M. Lifshits, *Electrodynamics of Continuous Media* (Gostekhizdat, Moscow, 1957), 266 pp.
21. T. Kaiser, S. Lange, and G. Schweiger, Appl. Opt. **33**, 7789–7797 (1994).
22. Yu.E. Geints and A.A. Zemlyanov, Atmos. Oceanic Opt. **10**, No. 4–5, 313–321 (1997).
23. C. Bohren and D. Huffman, *Absorption and Scattering of Light by Small Particles* (Willey, New York–London, 1983), 662 pp.