Nonlinear effects of stimulated scattering in spherical particles

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The effects of stimulated Raman scattering and stimulated Brillouin scattering in transparent spherical particles are theoretically described. The set of equations for the amplitude coefficients of fields of the incident and Stokes waves is derived and studied. The threshold ratio is obtained for transition from spontaneous scattering to stimulated Raman scattering. The spatial structure of interacting natural oscillational modes of a particle is shown to affect significantly the value of this threshold. It is found that the higher is the Q-factor of resonance modes, the more strongly it is affected by particle surface deformations, which may be spontaneous or connected with the effect of ponderomotive forces in a light field.

Introduction

Nonlinear optical effects of stimulated scattering, such as stimulated Raman scattering (SRS), stimulated Brillouin scattering (SBS), stimulated fluorescence (SF), and stimulated thermal scattering (STS), in gases and condensed extended media have been studied well enough. However, only in the past two decades it was discovered that these effects also manifest themselves in disperse matter (solid or liquid micrometer-size particles). This is because a micrometer-size particle for a long time was not considered as a subject of research in stimulated scattering, since the length of nonlinear interaction of waves needed for the field of stimulated scattering to be formed is at least an order of magnitude longer than the particle diameter. Only in late 70s – early 80s, in Refs. 1–4 and others it was shown that spherical particles, as a laser cavity, can accumulate the light field energy and thus many times increase the time and, consequently, the length of nonlinear interaction between a wave and a medium. Then the following effects were experimentally discovered: SRS and SF,[5,6] BweakerB effects of parametric generation of higher harmonics (coherent anti-Stokes scattering and coherent wave mixing),[7] and then even SBS[8] in droplets of dyed water, ethanol, and fuel.

Some regularities, not typical for extended media, were found in manifestation of stimulated scattering effects in micrometer-size particles. It proved, for example, that the spectral shape of stimulated scattering signals has a characteristic BpeakedB structure within a spontaneous scattering contour,[6] the SRS signal lags behind the pump pulse,[9,10] and the energy threshold of all the above mentioned effects are lower than those in continuous medium.[11]

The main prerequisite of the nonlinear optical effects in micrometer-size particles, as noted above, is the possibility of exciting of the optical field in such particles. The characteristics of these resonances are considered in detail, for example, in Ref. 12. Here we would like only to emphasize that they are observed at certain values of the diffraction parameter of a particle and theoretically can be rather narrow with the Q-factor $Q \sim 10^{10} - 10^{20}$. In practice, the values of Q are, as a rule, below $10^6 - 10^8$ (Ref. 11). This is connected, first, with the radiation absorption in a liquid and, second, with the non-spherical shape of particles because of different physical processes. The capability of transparent particles to focus the light field inside their volume also plays a significant part in appearance of stimulated scattering effects. Two main maxima of the electromagnetic field formed near the shadow and illuminated surfaces of a particle are powerful sources of spontaneous Raman scattering.

The above-mentioned peculiarities of the spatial configuration of the pump and stimulated scattering fields in spherical particles result in the appearance of a time lag between the stimulated scattering signal from particles and the pump pulse.[9,10,13] Within the framework of the existing theoretical model of stimulated scattering process in a transparent particle (see Section 1), the time lag Δt follows from the finiteness of the time of formation of the stimulated scattering signal. In other words, similarly to generation of stimulated radiation in a laser cavity, light wave must travel several times from mirror to mirror (in a micrometer-size particle, along the surface which plays the part of a mirror) to form sufficiently intense radiation at the Raman frequency, which is capable to support the stimulated process. Typical values of the time lag Δt in experiments with the micrometer-size droplets are within the range $1 \le \Delta t \le 10$ ns (Ref. 13) and are practically independent of the particle size. This indicates that resonance modes with the same (by the order of magnitude) Q-factor $Q \sim 10^5 - 10^6$ play the main part in the formation of stimulated scattering signals.

The threshold characteristics of the processes of stimulated scattering, in contrast to the frequency and time characteristics, have received poorer experimental study. As was noted above, the resonance character of

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excitation of stimulated scattering in micrometer-size particles leads to a significantly lower threshold of its manifestation. Moreover, if certain restrictions are imposed on the geometry and time conditions of pumping, anomalously low thresholds of stimulated scattering can be obtained.[14,15]

Note that all the above-listed processes occur inside particles. From this viewpoint, they can be called the processes of volume scattering. The only exception is for the phenomenon connected with Raman scattering of light waves at the surface of a liquid particle, vibrations of which were first excited by high-power laser radiation (this effect was considered in Refs. 16 and 17). As known, such vibrations are caused by ponderomotive forces of light field.[18,19] The amplitude of these forces in transparent optically large particles can be high enough to cause not only significant deformations, but also destruction of particles.[20]

As was noted above, the effects of stimulated scattering in transparent particles have some peculiarities in comparison with those in continuous medium. The existence of these effects in the disperse media is connected with the possibility of exciting the resonance vibrational modes of the electromagnetic field in particles, what results, first of all, in appearance of a resonance structure in the spectra of stimulated scattering in particles. The main prerequisite for practical application of the effects of stimulated scattering is quite a well-defined correspondence between this resonance structure and particle morphology. This actually indicates that the spectral position of resonance peaks in the spectra of stimulated scattering is unambiguously determined by the particle shape, its diffraction parameter $x_a = 2\pi a_0/\lambda$ (a_0 is the particle radius; λ is the radiation wavelength), and the refractive index. Therefore, any changes in the spectral position of resonances at unchanged chemical composition of a particle unambiguously correspond to changes in the above-listed parameters. This idea provides the basis for the techniques of contactless diagnostics of microphysical parameters of aerosol particles.[21,22]

The lower, as compared to the continuous medium, energy threshold of stimulated scattering processes in particles and narrow spectral width of the Raman scattering line (~0.01 cm^{-1}) also open new fields for the practical use of these effects in the Raman spectroscopy of aerosols.[22–24] In contrast to traditional spectroscopic methods of determination of the spectral composition from Raman spectra of samples, Raman spectroscopy, being essentially a sort of intracavity spectroscopy, does not impose restrictions on the minimum size of the medium under study. So, it allows *in situ* diagnostics of aerosol formations to be performed. Transparent particles act as high-Q optical cavities, amplifying the wave of spontaneous Raman scattering by many times and providing the positive feedback for appearance of stimulated radiation. High values of the coefficient of nonlinear conversion of the incident wave into the Raman component significantly increase the sensitivity of micro-Raman spectroscopy, thus allowing detection of even minor admixtures (< 10%) in the basic substance.[22] Moreover, this threshold can be even further decreased by an order of magnitude, if a small amount of a lasing dye is added to the particle under study.[24] The dye, due to its active fluorescence, provides faster growth of the Raman scattering signal.

And, finally, one more field of practical application of the effects of stimulated scattering in micrometer-size particles is associated with the optical technologies, rather than physical optics. Resonance properties of micrometer-size particles open up fresh opportunities for designing high-efficiency laser sources capable of operating not only in the optical range, but also in the SHF region.[25] From the literature it is also known that microcavities were actually applied as high-Q frequency filters in optical communication devices.[26]

In this paper we present a theoretical description of the main characteristics of nonlinear effects of stimulated scattering in weakly absorbing particles. In this study we have mainly focused on a spatial structure of the light field inside a spherical particle under conditions of their resonance excitation. The degree of interaction of these vibrational modes is the factor which determines the efficiency of excitation of nonlinear scattering processes in a particle.

1. Theoretical model of the processes of stimulated scattering in spherical particles

Let us consider the main points of the theoretical description of stimulated scattering in transparent particles. Qualitatively, the appearance of stimulated scattering in a spherical particle can be described as follows.[10,16]

At nonlinear interaction of radiation with the substance of a transparent particle, spontaneous inelastic scattering occurs in the whole particle volume, and it is most intense in the areas of focusing of the optical (pump) field. Raman frequencies in the scattered signal may appear due to interaction of the incident wave with the molecular vibrations of the medium (SRS), scattering on the acoustic phonons resulting from the electrostriction effect (SBS), emission of fluorescing molecules (SF), as well as scattering on thermal fluctuations of the medium density (STS). Some waves of the Raman spectrum leave the droplet, while others propagate along its surface because of the total internal reflection (more precisely, almost total internal reflection) (see Fig. 1). In traveling, these waves attenuate due to absorption and escape through the droplet surface, but they also can be intensified due to medium nonlinearity. If the resonance condition is fulfilled for one or several frequencies of the Raman spectrum, that is, the wave frequency ω_s coincides with the frequency of some natural resonance mode ω_n of a droplet, the amplification of spontaneous wave may exceed the total loss, and thus the stimulated Raman scattering occurs in the particle. From the viewpoint of field formation in a cavity, the field of stimulated scattering can be treated as a standing wave formed by superposition of electromagnetic waves propagating towards each other along the spherical surface of a droplet as the condition of phase matching is fulfilled.

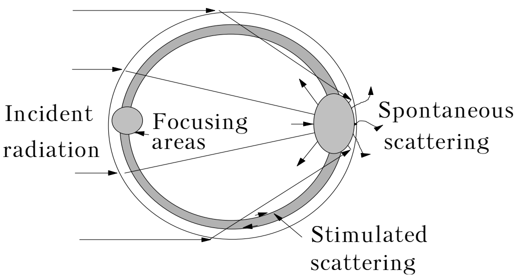

Fig. 1. Evolution of the stimulated scattering process in a spherical particle. Shaded areas show the main sources of spontaneous Raman scattering, which is then intensified by the field of the particle resonance mode (ring zone near the surface).

Analysis shows that, in spite of different physical nature of the phenomena resulting in the occurrence of that or another process of stimulated scattering (SRS, SBS, or SF), they are described similarly.[16] This is explained, first of all, by the fact that all the considered nonlinear optical effects are essentially resonance processes in the sense that their occurrence in a particle is connected with its resonance properties. Therefore, the threshold, angular, and spectral characteristics of all the types of stimulated scattering in the particle are Bfingerprints B of the resonance structure of internal fields.

The initial equations for theoretical analysis of the processes of nonlinear light scattering in a particle are the Maxwell equations, in which the nonlinear medium polarization $\mathbf{P}_N(\mathbf{r}, t)$ induced by the pump field serves as a source of the Raman scattering wave field.[27] As known, upon excluding the magnetic field strength $\mathbf{H}(\mathbf{r}, t)$, this set of equations transforms into the wave equation for the electric field strength vector $\mathbf{E}(\mathbf{r}, t)$ in the particle:

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

where c is the speed of light in vacuum; ε_a and σ are, respectively, the permittivity and conductivity of the particulate matter; the medium is considered homogeneous. At the boundary of spherical region, the conditions of continuity of the tangent field components:

$$(\mathbf{E}_{\mathrm{i}}(\mathbf{r}, t) + \mathbf{E}_{\mathrm{sc}}(\mathbf{r}, t) - \mathbf{E}(\mathbf{r}, t)) \times \mathbf{n}_r = (\mathbf{H}_{\mathrm{i}}(\mathbf{r}, t) + \mathbf{H}_{\mathrm{sc}}(\mathbf{r}, t) - \mathbf{H}(\mathbf{r}, t)) \times \mathbf{n}_r = 0 \qquad (2)$$

are fulfilled. Here $\mathbf{n}_r$ is the outer normal to the particle surface, and subscripts “iB and “scB correspond respectively to the incident and scattered waves outside of the particle.

The electric field $\mathbf{E}(\mathbf{r}, t)$ in a particle is the sum of fields at the frequency of the incident wave ω_{L} (pump frequency) and at the Raman shifted frequencies. From here we restrict our consideration to only the first Stokes wave with the frequency $\omega_{\mathrm{s}} = \omega_{\mathrm{L}} - \omega_{\mathrm{vib}}$ (ω_{vib} is the frequency of the dipole transition of the particulate matter) because it is most intense. Thus, $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_{\mathrm{L}}(\mathbf{r}, t) + \mathbf{E}_{\mathrm{s}}(\mathbf{r}, t)$, where the time dependence of the vector $\mathbf{E}$ can be presented by a harmonic $e^{-i\omega t}$ with a slowly varying amplitude $\tilde{\mathbf{E}}(\mathbf{r}, t)$:

$$\mathbf{E}(\mathbf{r}, t) = \frac{1}{2}\tilde{\mathbf{E}}_{\mathrm{L}}(\mathbf{r}, t)\, e^{-i\omega_{\mathrm{L}} t} + \frac{1}{2}\tilde{\mathbf{E}}_{\mathrm{s}}(\mathbf{r}, t)\, e^{-i\omega_{\mathrm{s}} t} + \text{complex conjugate.}$$

In this case the wave equation (1), in fact, breaks into two related equations: for the incident and scattered waves, and nonlinear polarization $\mathbf{P}_{\mathrm{nel}}$ in the approximation of an only slightly nonlinear process also breaks into two terms:

$$\mathbf{P}_{\mathrm{nel}} = \mathbf{P}_{\mathrm{s}} + \mathbf{P}_{\mathrm{L}} = \chi^{(3)}(\omega_{\mathrm{s}})\, (\tilde{\mathbf{E}}_{\mathrm{L}}\, \tilde{\mathbf{E}}_{\mathrm{L}}^{*})\, \tilde{\mathbf{E}}_{\mathrm{s}}\, e^{-i\omega_{\mathrm{s}} t} + \chi^{(3)}(\omega_{\mathrm{L}})\, (\tilde{\mathbf{E}}_{\mathrm{s}}\, \tilde{\mathbf{E}}_{\mathrm{s}}^{*})\, \tilde{\mathbf{E}}_{\mathrm{L}}\, e^{-i\omega_{\mathrm{L}} t} + \text{complex conjugate,}$$

where $\chi^{(3)}$ is the third-order nonlinear dielectric susceptibility. The Eq. (1) for field is supplemented with the equations describing specific physical mechanism of occurrence of a nonlinearly scattered wave.[16]

Because we consider the formation of fields in a volume optical cavity (spherical particle in our case), our next step is to present the solution of the wave equation (1) for the fields $\mathbf{E}_n(\mathbf{r})$ and $\mathbf{H}_n(\mathbf{r})$ as a series expansion over eigenfunctions (normal vibrational modes) of the particle with the natural frequencies ω_n, which are complex for open cavities.[28] These functions form the complete orthogonal system within the framework of the electrodynamics problem (1) for the closed cavity without radiative loss and satisfy the equations:

$$\text{rot } \mathbf{E}_n(\mathbf{r}) = i\,\frac{\omega_n}{c}\,\mathbf{H}_n(\mathbf{r}); \quad \text{rot } \mathbf{H}_n(\mathbf{r}) = -\, i\,\frac{\varepsilon_a\,\omega_n}{c}\,\mathbf{E}_n(\mathbf{r}). \qquad (3)$$

Then, for example, for the Stokes wave field we have

$$\mathbf{E}_{\mathrm{s}}(\mathbf{r}, t) = \sum_n A_n(t, \omega_{\mathrm{s}})\, \mathbf{E}_n(\mathbf{r}),$$

$$\mathbf{H}_{\mathrm{s}}(\mathbf{r}, t) = -i\sqrt{\varepsilon_a}\sum_n B_n(t, \omega_{\mathrm{s}})\, \mathbf{H}_n(\mathbf{r}), \qquad (4)$$

where the time dependence is taken into account through the coefficients A_n and B_n and, besides, their frequency dependence is shown explicitly. The subscript n, in the general case, is used for a set of several subscripts, the number of which depends on the specific geometry of the cavity. For a sphere, for example, there are three subscripts: the mode number n, the azimuth number l, and the resonance order j.

Note that the above-described mathematical approach that uses a series expansion of fields in an open cavity over the eigenfunctions of a closed cavity and taking into account actual boundary conditions at its surface is known in the literature as the Slater method.[29] However, one should keep in mind that the expansions (4) do not converge to actual values at $r = a_0$ because the tangential components of the eigenfunctions of the ideal cavity $\mathbf{E}_n(\mathbf{r})$ and $\mathbf{H}_n(\mathbf{r})$ equal zero at the surface. Therefore, the use of the expansion of the actual fields $\mathbf{E}$ and $\mathbf{H}$ under the rot sign is incorrect, from the general point of view. We should write the expansion of the functions (rot $\mathbf{E}$) and (rot $\mathbf{H}$) over the systems $\mathbf{E}_n(\mathbf{r})$ and $\mathbf{H}_n(\mathbf{r})$. Using Eqs. (3) and (4), we have

$$\int_V \mathrm{rot}\, \mathbf{E}(\mathbf{r}')\, \mathbf{H}_n^*(\mathbf{r}')\, \mathrm{d}\mathbf{r}' = -\, i\, \frac{\varepsilon_a\, \omega_n}{c} A_n(t) +$$

$$+ \int_S [\mathbf{E}(\mathbf{r}') \times \mathbf{H}_n^*(\mathbf{r}')]\, \mathbf{n}_r\, \mathrm{d}\mathbf{r}' = -\, \frac{\omega_n}{c} \frac{\mathrm{d}B_n(t)}{\mathrm{d}t}\,.$$

The expansion for (rot $\mathbf{H}$) has quite similar form.

After some transformations typical for the theory of resonators, we derive the set of equations for the expansion coefficients $A_n(t)$:

$$\frac{\mathrm{d}^2 A_n(t)}{\mathrm{d}t^2} + \frac{4\pi\sigma}{\varepsilon_a} \frac{\mathrm{d}A_n(t)}{\mathrm{d}t} + \omega_n^2\, A_n(t) =$$

$$= -\, \frac{4\pi}{\varepsilon_a} \int_V \mathbf{E}_n^*(\mathbf{r}')\, \frac{\partial^2 \mathbf{P}_\mathrm{s}(\mathbf{r}',\, t)}{\partial t^2}\, \mathrm{d}\mathbf{r}' - \frac{i \omega_n c}{\varepsilon_a}\, \Pi_n(t), \quad (5)$$

where

$$\Pi_n(t) = \int_S [\mathbf{E}(\mathbf{r}') \times \mathbf{H}_n^*(\mathbf{r}')]\, \mathbf{n}_r\, \mathrm{d}S\, -$$

$$- \frac{i}{\omega_n} \frac{\partial}{\partial t} \int_S [\mathbf{H}(\mathbf{r}') \times \mathbf{E}_n^*(\mathbf{r}')]\, \mathbf{n}_r\, \mathrm{d}S.$$

Let us analyze this equation. As seen, it describes the fading stimulated oscillations. These oscillations are excited by an external force acting at the frequency ω_s and proportional to the medium polarization induced by the pump field. The surface integral $\Pi_n(t)$ in the right-hand side of Eq. (5) takes into account the energy loss of the Stokes wave due to nonlinear relation of the natural modes through the particle surface. For the spherical cavity it can be transformed as

$$\Pi_n(t) = 2A_n(t)\, \frac{\omega_n\, \varepsilon_a}{c\, Q_n^\mathrm{rad}} \left[1 + \frac{i}{2Q_n^\mathrm{rad}} \right].$$

Here the parameter of radiative Q-factor of the cavity $Q_n^\mathrm{rad} = \omega_n\, W_n / P_\mathrm{rad}$ is introduced, where

$$W_n = \frac{\varepsilon_a}{8\pi} \int_V \mathbf{E}_n(\mathbf{r}')\, \mathbf{E}_n^*(\mathbf{r}')\, \mathrm{d}\mathbf{r}'$$

is the mean energy of the electromagnetic field accumulated in a natural mode during the oscillation period, and

$$P_\mathrm{rad} = \frac{c}{8\pi} \int_S \mathrm{Re}\{[\mathbf{E}_n(\mathbf{r}') \times \mathbf{H}_n^*(\mathbf{r}')]\}\, \mathbf{n}_r\, \mathrm{d}S$$

is the mean power of the radiative loss of the light wave. Finally, equation for the expansion coefficients of the field $\mathbf{E}(\mathbf{r},\, t)$ in a particle takes the form

$$\frac{\mathrm{d}^2 A_n(t)}{\mathrm{d}t^2} + \frac{\omega_n}{Q_n^\mathrm{abs}} \frac{\mathrm{d}A_n(t)}{\mathrm{d}t} + \left[\omega_n \left(1 + \frac{i}{2Q_n^\mathrm{rad}} \right) \right]^2 A_n(t) =$$

$$= -\, \frac{4\pi}{\varepsilon_a} \int_V \mathbf{E}_n^*(\mathbf{r}')\, \frac{\partial^2 \mathbf{P}_\mathrm{s}(\mathbf{r}',t)}{\partial t^2}\, \mathrm{d}\mathbf{r}'. \quad (6)$$

The parameter Q_n^abs in the left-hand side of this equation is essentially the cavity Q-factor as well (at the natural mode frequency ω_n), but in contrast to the radiative Q-factor Q_n^rad, it is caused only by the absorption of light wave by the particulate matter:

$$Q_n^\mathrm{abs} = \frac{\omega_n\, W_n}{P_\mathrm{abs}} = \frac{\omega_n\, c^2}{4\pi\sigma}\,.$$

Here $P_\mathrm{abs} = \dfrac{1}{2} \displaystyle\int_V \sigma(\mathbf{r}')\, \mathbf{E}_n(\mathbf{r}')\, \mathbf{E}_n^*(\mathbf{r}')\, \mathrm{d}\mathbf{r}'$ is the mean power of the light wave lost due to the absorption. The parameter Q_n^abs is a characteristic of the cavities of any type, but the definition of the radiative Q-factor Q_n^rad makes sense only for open systems with diffraction losses. The value of the Q-factor Q_n^rad for transparent spherical particles can be very large[12] (see Fig. 2).

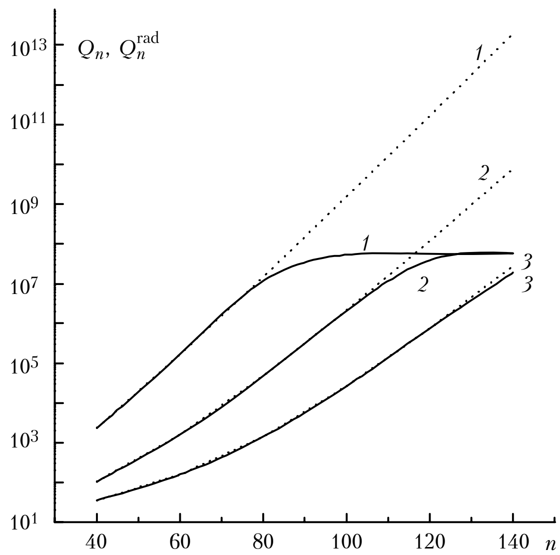

Fig. 2. Radiative Q-factor Q_n^rad (dashed line) and total Q-factor Q_n (solid line) of resonance modes of a spherical particle vs. the mode number n for different orders j (figures at the curves). The absorption coefficient of the particulate matter chosen is of the order of $\kappa_a = 10^{-8}$.

The frequencies of natural oscillations in the open cavity are complex values, rather than real ones as in ideal closed systems, $\omega'_n = \omega_n [1 + i/(2Q_n^{\rm rad})] = \omega_n + i\omega''_n$. The degree of this non-ideality is taken into account by the parameter inversely proportional to the radiative Q-factor:

$$\frac{1}{Q_n^{\rm rad}} = \frac{2\omega''_n}{\omega_n}.$$

This reasoning allows us to define the total (effective) Q-factor Q_n as

$$1/Q_n = 1/Q_n^{\rm abs} + 1/Q_n^{\rm rad}. \qquad (7)$$

It is proportional to time characteristic of the existence of electromagnetic field of a natural mode in the cavity τ_n. Obviously, the allowance for absorption in a dielectric results in a restricted exponential growth of the Q-factor of the particle as its size grows. This is well seen in Fig. 2, which shows the dependence of the total Q-factor Q_n of different resonance modes on the modal index n. The absorption coefficient of the particulate matter was chosen to be at the level of $\kappa_a = 10^{-8}$.

Studies of electromagnetic oscillations in optical cavities with loss often manipulate with the equation for the expansion coefficients of electromagnetic field in the following form[27,30]:

$$\frac{{\rm d}^2 A_n(t)}{{\rm d}t^2} + 2\gamma_e \frac{{\rm d}A_n(t)}{{\rm d}t} + \omega_n^2 A_n(t) = = -\frac{4\pi}{\varepsilon_a} \int\limits_V \mathbf{E}_n^*(\mathbf{r}') \frac{\partial^2 \mathbf{P}_{\rm s}(\mathbf{r}',t)}{\partial t^2} {\rm d}\mathbf{r}', \qquad (8)$$

where the vibration damping coefficient equals $\gamma_e = \dfrac{\omega_n}{2Q_n^{\rm abs}}$ and determines the half-width of the resonance contour of the corresponding natural mode with allowance for the wave loss due to absorption. Note that the structure of a solution to this equation is similar to that of the Eq. (6) solution, if the loss for emission, which is typical of open cavities, is taken into account in the coefficient γ_e, and the parameter $(1/Q_n^{\rm rad})$ is considered small in the case when the terms of the order of $(1/Q_n^{\rm rad})^2$ are negligible.

It is clear that the symmetry of the initial equation (1) about the field vectors allows us to write an equation, similar to Eq. (5), for the expansion coefficients of the electric field of the incident wave: $\mathbf{E}_{\rm L}(\mathbf{r}, t) = \sum\limits_n A_n(t, \omega_{\rm L})\, \mathbf{E}_n(\mathbf{r})$. The only peculiarity of this equation is specific equation for the surface integrals $\Pi_n(t)$, because the boundary conditions for the pump wave field differ from those for the Stokes wave. They are given by Eq. (2), and include, in spite of the scattered fields, the incident wave fields. Therefore, denoting the combination of surface integrals as

$$-\frac{i\omega_n c}{\varepsilon_a}\left[\int\limits_S [\mathbf{E}_{\rm i}(\mathbf{r}') \times \mathbf{H}_n^*(\mathbf{r}')]\, \mathbf{n}_r\, {\rm d}S - -\frac{i}{\omega_n}\frac{\partial}{\partial t}\int\limits_S [\mathbf{H}_{\rm i}(\mathbf{r}') \times \mathbf{E}_n^*(\mathbf{r}')]\, \mathbf{n}_r\, {\rm dS}\right] = F_{\rm i}(t),$$

where $F_{\rm i}(t)$ is the external force providing for the energy influx into the cavity, we derive the differential equation for the expansion coefficients of the incident wave (at the frequency $\omega_{\rm L}$):

$$\frac{{\rm d}^2 A_n(t,\omega_{\rm L})}{{\rm d}t^2} + \frac{\omega_n}{Q_n^{\rm abs}}\frac{{\rm d}A_n(t,\omega_{\rm L})}{{\rm d}t} + \left[\omega_n\left(1 + \frac{i}{2Q_n^{\rm rad}}\right)\right]^2 \times \times A_n(t, \omega_{\rm L}) = F_i(t) - \frac{4\pi}{\varepsilon_a}\int\limits_V \mathbf{E}_n^*(\mathbf{r}')\frac{\partial^2 \mathbf{P}_L(\mathbf{r}', t)}{\partial t^2}{\rm d}\mathbf{r}'. \qquad (9)$$

The nonlinear polarization term in the right-hand side of this equation is responsible for the energy outflow from the pump into the Stokes wave.

Solution of the set of equations (6) and (9) along with the corresponding equations of nonlinear processes, as well as the initial and boundary conditions, allows a complete description to be achieved of the process of stimulated scattering in a particle.

In deriving equations (6) for the expansion coefficients of the electromagnetic fields in a cavity, we did not explicitly mention the fact that the source of scattered wave is initially the spontaneous Raman scattering arising throughout the particle volume. The intensity of spontaneous Raman scattering is proportional to the square amplitude of the pump field. Therefore, the nonlinear polarization $\mathbf{P}_{\rm s}$ in the right-hand size of Eq. (6) can actually be presented as a sum of two terms $\mathbf{P}_{\rm st}$ and $\mathbf{P}_{\rm sp}$, which correspond, respectively, to the induced and spontaneous mechanisms of energy influx to the stimulated scattering wave[29]: $\mathbf{P}_{\rm s} = \mathbf{P}_{\rm st} + \mathbf{P}_{\rm sp}$. As to the induced part of the polarization $\mathbf{P}_{\rm st}$, it has been defined above through the nonlinear susceptibility of the matter and describes the coherent interaction of the pump field and the Stokes wave field.

The specific equation for the second component $\mathbf{P}_{\rm sp}$ responsible for the spontaneous scattering is determined by the physical mechanisms of inelastic scattering in the medium. Thus, for example, for the phenomenon of Raman scattering we use the following representation:

$$\mathbf{P}_{\rm sp}(\mathbf{r},t) = N_0 \frac{\partial\alpha}{\partial q}\tilde{q}_{\rm sp}^*(\mathbf{r},t)\, \tilde{\mathbf{E}}_L(\mathbf{r})\, {\rm e}^{-i\omega_s t} + \text{complex conjugate}, \qquad (10)$$

where $\tilde{q}_{\rm sp}^*(\mathbf{r}, t) = F_{\rm sp}(\mathbf{r}, t)\, T_2\, (1 - {\rm e}^{-t/T_2})$ is the amplitude of the molecular vibrations of the medium due to only the processes of spontaneous scattering; α is the polarizability of the particulate matter; N_0 is the concentration of molecules; and $F_{\rm sp}(\mathbf{r}, t)$ is some external distributed force that induces the molecular vibrations. As to the latter parameter, it is usually assumed to be delta-correlated in time and space: $\langle F_{sp}(\mathbf{r}, t)\, F_{sp}(\mathbf{r}', t')\rangle = F_0^2\, \delta(t - t')\, \delta(\mathbf{r} - \mathbf{r}')$, where the coefficient F_0 is connected with the quantum-mechanical characteristics of the medium molecules.

2. Energy threshold of the stimulated scattering in a micrometer-size particle

To describe the spatial structure of external sources of the Raman wave in a more explicit way, let us transform the right-hand side of Eq. (8) with the help of Eq. (10). Toward this end, let us apply the expansion for the pump field $\mathbf{E}_L(\mathbf{r})$ and the Stokes wave field $\mathbf{E}_s(\mathbf{r})$ over the eigenfunctions of the spherical cavity and use the approximation of slowly varying amplitudes. Besides, the pump field is considered preset $(\mathbf{P}_L \approx 0)$, and the pulse of acting radiation is assumed long enough to assume that $\mathrm{d}\tilde{A}_n(\omega_L)/\mathrm{d}t \ll \mathrm{d}\tilde{A}_n(\omega_s)/\mathrm{d}t$, where $\tilde{A}_n$ is the slowly varying amplitude. Then Eq. (8) for the coefficients $A_n(t, \omega_s)$ takes the form

$$2(i\omega_s - \gamma_e)\frac{\mathrm{d}\tilde{A}_n(t, \omega_s)}{\mathrm{d}t} + (\omega_s^2 - \omega_n^2 + 2i\,\gamma_e\omega_s)\,\tilde{A}_n(t, \omega_s) =$$

$$= -\frac{4\pi}{\varepsilon_a}\chi^{(3)}(\omega_s)\sum_l\sum_{l'}\sum_{n'}\tilde{A}_l(t, \omega_L)\,\tilde{A}_l^*(t, \omega_L)\,\times$$

$$\times\left[2i\omega_s\frac{\mathrm{d}\tilde{A}_n(t, \omega_s)}{\mathrm{d}t} + \omega_s^2\,\tilde{A}_{n'}(t, \omega_s)\right]S_4 +$$

$$+ N_0\,\omega_L^2\,\frac{4\pi}{\varepsilon_a}\frac{\partial\alpha}{\partial q}F_0\,T_2\,\tilde{A}_n(t, \omega_L), \qquad (11)$$

where $S_4 = \int\limits_V \mathbf{E}_n^*(\mathbf{r}')\,\mathbf{E}_{n'}(\mathbf{r}')\,\mathbf{E}_l^*(\mathbf{r}')\,\mathbf{E}_{l'}(\mathbf{r}')\,\mathrm{d}\mathbf{r}')$ are the integral coefficients of spatial overlap of the field of natural modes in the cavity. The subscripts of these coefficients are the numbers of interacting modes. As follows from the equation derived, the overlap coefficients in the cavity are a selector of the natural modes, and obviously the modes, whose spatial structures are closest to that of the pump field, have the advantages in the process of Raman wave evolution.

The differential equation (11) under these assumptions allows rather a simple solution. Not making our consideration less general, let us simplify the problem under consideration assuming the pump field and the Stokes wave field unimodal ($\omega_s = \omega_n$; $\omega_L = \omega_m$). This allows us to avoid summation in the right-hand side of Eq. (11). At BweakB pump, if the induced part of the nonlinear polarization is negligible, the solution of Eq. (11) describes the process of spontaneous Raman scattering. The wave amplitude of spontaneous Raman scattering is

$$\tilde{A}_n(t, \omega_s) \approx \frac{4\pi}{\varepsilon_a}\frac{\partial\alpha}{\partial q}\frac{N_0\,\omega_L^2\,F_0\,T_2}{2(i\omega_s - \gamma_e)}\exp(-\gamma_e t)\,\times$$

$$\times\int\limits_0^t \exp(\gamma_e t')\,\tilde{A}_n(t', \omega_L)\,\mathrm{d}t'. \qquad (12)$$

(For brevity, the modal indices in the integral interaction coefficients are omitted.)

In the another asymptotic case of a BstrongB pump field, the dominating mechanism in the scattering process is the energy outflow from the pump into the scattered wave induced by the Raman wave itself, what results in the nonlinear growth of its amplitude:

$$\tilde{A}_n(t, \omega_s) \approx \frac{\partial\alpha}{\partial q}\frac{4\varepsilon_a\,N_0\,\omega_L^2\,F_0\,T_2}{c^2\omega_s\,g_s\,\tilde{A}_m^*(t, \omega_L)\,S_4}\,\times$$

$$\times\exp\left\{\frac{c^2\,g_s\,S_4}{16\pi}\int\limits_0^t|\tilde{A}_n(t', \omega_L)|^2\,\mathrm{d}t'\right\}, \qquad (13)$$

where $g_s = -32\pi^2\,\omega_s/(c^2\varepsilon_a)\,\mathrm{Im}\{\chi^{(3)}(\omega_s)\}$ is the stationary gain of the stimulated Raman scattering.[31]

The threshold of transition from spontaneous Raman scattering to stimulated Raman scattering apparently corresponds to the threshold value of the pump wave amplitude $\tilde{A}_m(\omega_L)$, at which the solution of Eq. (11) at $t \to \infty$ becomes unstable. Using the standard definition of the asymptotic stability of the solution of an ordinary differential equation,[32] we obtain the condition of SRS generation in the cavity:

$$|\tilde{A}_m(\omega_L)|^2 > 8\pi\omega_s/(c^2\,g_s\,Q_n\,S_4). \qquad (14)$$

Here we should emphasize two circumstances. First, the Eq. (14) has been derived in the approximation of a preset pump field, that is, the SRS threshold is obviously underestimated. The allowance for exhaustion of the pump wave, as well as the energy outflow from the Stokes wave to higher harmonics leads to an increase of the threshold value $\tilde{A}_m(\omega_L)$. Second, the coefficients in Eq. (14) correspond to the expansion of the pump field inside the particle, and, as follows from the Mie theory, it can be tens or even hundreds times stronger than the field of the incident wave. Therefore, if we define the threshold of stimulated scattering in the particle as some threshold intensity $I_{st}(\omega_L)$ of the exciting light wave before its incidence on the particle at which generation of the stimulated radiation occurs, then, separating the amplitude part in Eq. (14), we have

$$I_{st}(\omega_L) = \frac{n_a\,\omega_s}{c g_s Q_n\,|b_m(x_a)|^2 S_4}, \qquad (15)$$

where the coefficient $b_m(x_a)$ takes into account the degree of modification of the incident wave field by the particle morphology.

3. Coefficient of spatial overlap of light fields in a spherical particle

The equation for the threshold pump intensity resulting in transition from spontaneous scattering to stimulated one has been derived above (Eq. (15)). Similar result can also be obtained within the framework of a different approach based on the law of conservation of the electromagnetic energy in the particle irradiated.[16] In the above designations this law for the vectors of the electromagnetic field of scattered wave can be written as

$$\frac{dW_s}{dt} = P_g - (P_{abs} + P_{rad}), \qquad (16)$$

where $P_g = -\frac{1}{2}\int\limits_V \mathrm{Re}\,\{\tilde{\mathbf{E}}_s^* \, (\partial\tilde{\mathbf{P}}_s/\partial t)\}\, d\mathbf{r}'$ is the average power of the sources of the Stokes wave.

From the condition that $\frac{dW_s}{dt} = 0$ we can obtain the equation for the threshold pump intensity ensuring the stimulated scattering in a transparent particle to occur:

$$I_t(\omega_L) = \frac{n_a\, \omega_s}{g_s\, Q(\omega_s)\, B_c(\omega_L, \omega_s)}, \qquad (17)$$

where $Q(\omega_s)$ is the total Q-factor of the particle at the frequency ω_s, and $B_c(\omega_L, \omega_s)$ is the integral coefficient allowing for the spatial overlap of the interacting fields inside the particle:

$$B_c(\omega_L, \omega_s) = \left(\int\limits_V B_s(\mathbf{r})\, d\mathbf{r}'\right)^{-1} \int\limits_V B_s(\mathbf{r})\, B_L(\mathbf{r})\, d\mathbf{r}'. \qquad (18)$$

In Eq. (18) $B_L(\mathbf{r})$ and $B_s(\mathbf{r})$ are the inner field inhomogeneities, known from the Mie theory, at the frequencies ω_L and ω_s, respectively (see Ref. 33). If the fields $\tilde{\mathbf{E}}_L$ and $\tilde{\mathbf{E}}_s$ are unimodal, then

$$B_s(\mathbf{r}) = |b_n(x_a)|^2[\mathbf{E}_n(\mathbf{r})\, \mathbf{E}_n^*(\mathbf{r})],$$

$$B_L(\mathbf{r}) = |b_m(x_a)|^2[\mathbf{E}_m(\mathbf{r})\, \mathbf{E}_m^*(\mathbf{r})].$$

The field overlap coefficient $B_c(\omega_L, \omega_s)$ in this case takes the following form:

$$B_c(\omega_L, \omega_s) = |b_m(x_a)|^2 S_4,$$

and Eq. (17) for the SRS threshold transforms into Eq. (15).

The threshold values of I_t for the SRS and SBS processes in water droplets at the varied droplet radius are given in Refs. 16 and 34. In Ref. 34 we also studied the behavior of the overlap coefficient B_c for different cases of stimulated scattering in spherical particles. Figure 3 shows the dependence of the coefficient B_c on the resonance half-width $\Gamma = x_a/Q$ at the frequency of a scattered wave for three processes: SRS, SRS at "double" resonance of optical fields (pump wave and Stokes wave), and SBS. The data shown in the figure were obtained from the numerical calculations of the coefficient B_c for different configurations of the spatial overlap of resonance modes in water droplets. Every group of data is unified by a spline for illustration.

It is the difference in the behaviors of $B_c(\Gamma)$ at SRS and SBS that attracts our attention first of all. If in the first case this dependence is monotonic, then the dependence $B_c(\Gamma)$ for SBS has a pronounced maximum associated with the violation of the resonance conditions for the pump field. Thus, to the right from the maximum of the $B_c(\Gamma)$ curve for SBS, the stimulated process runs at the resonance of both the interacting fields of the pump and scattered waves, and to the left from the maximum, the resonance is observed only for the Stokes wave.

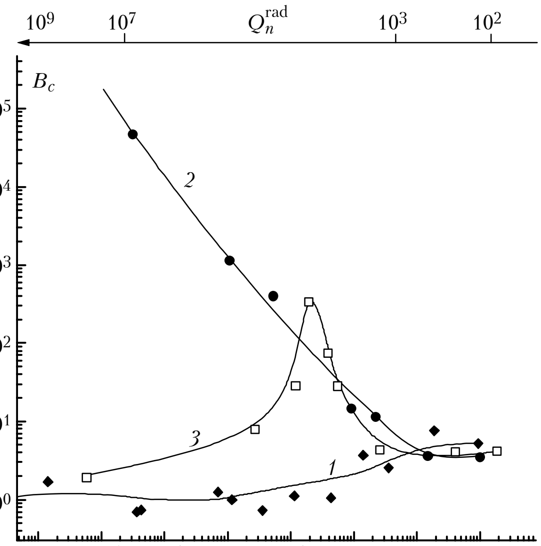

Fig. 3. Field overlap coefficient B_c vs. the halfwidth of resonance modes of a spherical particle Γ at SRS (*1*), "double" resonance SRS (*2*), and SBS (*3*). The radiative Q-factor of the resonance modes Q_n^{rad} is plotted on the upper scale.

There is also a difference between the values of the overlap coefficient for the single- and double-resonance SRS. At resonance pumping of SRS in the particle, the values of B_c are significantly higher and tend to growth with decreasing half-width of the resonance modes due to the increase in their Q-factor. At non-resonance pumping the dependence is quite opposite. In this case a decrease in the B_c coefficient is connected with narrowing of the resonance maximum of the spatial distribution of the scattered wave field at its simultaneous shift along the radius toward the particle surface, where the pump field has a local minimum. In the limit of very narrow resonances ($c \le 10^{-7}$), the spatial configuration of the pump field in the spherical particle, which is taken into account through the coefficient B_c, has no effect on the process of stimulated scattering, and the values of B_c are close to unity, as in the case of an extended medium.

With all the above mentioned differences, the dependences $B_c(c)$ shown in the figure have a common property, namely, at large c they all converge to the same level $B_c \approx 4.5$. In this interval of BwideB natural modes, their resonance nature becomes practically implicit, and the excess of the overlap coefficient over unity is caused only by focusing of the incident field because of a spherical surface of the particle.

4. Influence of surface deformations of spherical particles on the Q-factor of their natural modes

As follows from the above-said, the effective Q-factor Q is an important characteristic affecting processes of nonlinear interaction of light fields in optical cavities. The value of the Q-factor itself significantly depends on the geometry of a microcavity, in spite of the dependence on optical characteristics of the particulate matter. As to spherical particles, any deformations of their surface finally result in worsening of their resonance properties. One of the reasons is that electromagnetic waves forming the field of a resonance mode interact with distortions of the spherical surface and scatter on them thus causing additional emission from the cavity. However, it should be noted that such an approach gives somewhat overestimated values of the Q-factor as compared to the experimental ones and does not explain some facts observed in the experiment (for example, effect of splitting of natural modes over the azimuth index l in spheres,[35] selection of low-Q modes in deformed particles[36]).

Another one and more likely, in our opinion, cause for the decrease in Q-factor is violation of the conditions of phase matching for resonances of optical modes in deformed particles. This changes the spatial structure of natural oscillations and shifts the frequency position of resonances. Below this effect is considered in a more detail.

To estimate the effect of surface deformations on the Q-factor of resonance modes, we proceed from the model of formation of stimulated scattering in a particle. The resonance mode is treated as a standing wave formed by superposition of two waves propagating toward each other along the boundary of the principal cross section of a spherical particle (Fig. 4). The plane of this cross section passes through the center of the sphere and is inclined at an angle θ_{nl} to the z axis. This angle is determined by the ratio of the azimuth subscript of the resonance mode l to its number n: $\theta_{nl} = \arccos(l/n)$. Since the subscript l varies in the interval $(n; -n)$, the plane of the circle, within which the mode field is mostly localized, lies at the polar angle $\theta = 0 - \pi/2$.

To form the standing wave, traveling waves must come at the initial point with the phase multiple of 2π, that is, the condition of the phase matching must be fulfilled: $ka_0 = n$, where k is the wave number inside the particle. Consequently, to preserve the phase matching, any deformations of the spherical surface changing the path length of the traveling waves by δL must be compensated for by the corresponding change of the absolute value of the wave vector δk: $\delta L/L_0 = \delta k/k$, where $L_0 = 2\pi a_0$ is the geometrical path length in an ideal sphere.

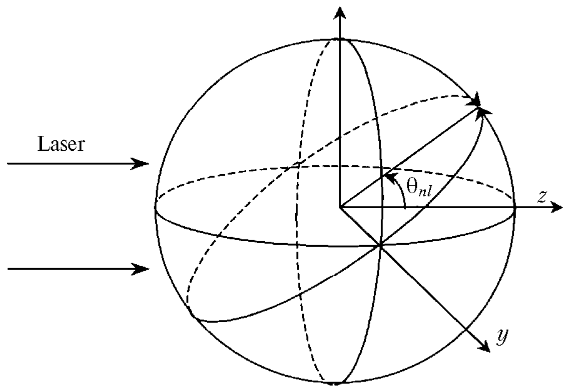

Fig. 4. Scheme illustrating the distribution of the field of a resonance mode with the azimuth subscript l. The electromagnetic field is localized in the ring zone inclined at the angle θ_{nl} to the equator.

Assuming $\xi(\theta) = a(\theta) - a_0$, where $a(\theta)$ is the radius of the deformed particle (we consider deformations symmetrical over the spherical angle φ), we obtain for the length increment δL:

$$\delta L \cong \int\limits_{\theta_{nl}}^{\pi-\theta_{nl}} \xi(\theta)\mathrm{d}\theta \cdot \frac{2\pi a_0}{\pi - 2\theta_{nl}} = \xi_A \int\limits_{\theta_{nm}}^{\pi-\theta_{nl}} \bar{\xi}(\theta)\mathrm{d}\theta \cdot \frac{2\pi a_0}{\pi - 2\theta_{nl}},$$

where ξ_A is the amplitude of surface deformations; $\bar{\xi} = \xi(\theta)/\xi_A$.

In the approximation of small surface deformations ($\xi_A \ll 1$), the cross section of the deformed particle can yet be considered as a circle with some effective radius a_{eff} depending on the amplitude and angular structure of deformations:

$$a_{\mathrm{eff}} = 1/2\pi\ (L_0 + \delta L) = a_0\ (1 + \xi_A\ q_{nl}).$$

Here $q_{nl} = \dfrac{1}{\pi - 2\theta_{nl}} \int\limits_{\theta_{nl}}^{\pi-\theta_{nl}} \bar{\xi}(\theta)\mathrm{d}\theta$ is the conversion coefficient (obviously, $|q_{nl}| \le 1$). Then for the increment of the diffraction parameter of the effective sphere δx for the TE(TH)$_{nl}$-mode we have

$$\delta x = x_{\mathrm{eff}} - x_0 = \delta k/n_a\ a_0 = x_0\ \xi_A\ q_{nl}, \qquad (19)$$

where x_0 is the resonance value of the diffraction parameter of the unperturbed sphere; $x_{\rm eff} = k_0\, a_{\rm eff}$.

As an example, Fig. 5 shows the dependence of the coefficients q_{nl} on the ratio (l/n) at deformation of liquid droplets under exposure to wave train of picosecond laser pulses (the data on deformations have been borrowed from Ref. 36). One can see that q_{nl} varies from 10^{-3} to 10^{-2} and approaches maximum for the modes lying in the plane of the droplet equator $(l = n)$.

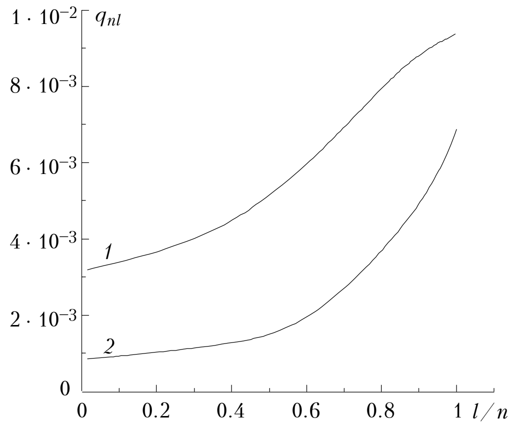

Fig. 5. Conversion coefficients q_{nl} vs. the ratio (l/n) for the droplets of water (*1*) and CS_2 (*2*).

As calculations show, the shape of the resonance curve of the particle natural modes corresponds to the Lorentz profile, therefore in the close vicinity of some resonance we can introduce the so-called Q-function:

$$Q_{\rm q}(x) = \frac{Q_0}{1 + (x - x_0)^2/\Gamma^2}\,,$$

where Q_0 is the Q-factor of some resonance mode of an unperturbed sphere (modal subscripts are omitted for simplicity) with the resonance curve half-width Γ. The value of this function at $x = x_0$ apparently coincides with the resonance Q-factor. Using Eq. (19) and taking into account that $Q_0 = x_0/\Gamma$, we obtain

$$Q_{\rm q}(x) = \frac{Q_0}{1 + (q_{nl}\,\xi_A\, Q_0)^2}\,.$$

As follows from this equation, the higher is the initial Q-factor of the resonance mode (higher Q_0 values), the stronger is the effect of deformations on it. In this case the modes with lower Q-factor, but more stable to particle deformations has an advantage in the development, because the electromagnetic field of these modes is concentrated farther from the surface.[12]

Conclusion

Let us formulate briefly the main results of this work. The theoretical consideration of the basic effects of stimulated scattering (SRS, SBS) in transparent spherical particles has shown that the main parameters determining the character of manifestation of different nonlinear wave processes are the effective Q-factor Q and the coefficient B_c of spatial overlap of the optical fields of interacting modes. The set of equations was derived for the amplitude coefficients of the fields of the Stokes and incident waves. The approximated solutions were obtained for two asymptotic cases of spontaneous Raman scattering and developed SRS, and the threshold of transition between these two cases was found. The numerical estimates showed the influence of the spatial structure of the interacting natural oscillation modes of a particle on the value of this threshold. It was found that the coefficient of overlap of the optical fields of the pump radiation and stimulated scattering inside the particle depends on how close are their spatial profiles. The value of B_c was shown to increase markedly if the frequency shift between the pump wave and the wave of stimulated scattering is less than the natural resonance line half-width of a particle (SBS), or the pump is also under resonance conditions (BdoubleB resonance SRS).

Significant influence of the shape deformations on the resonance properties of spherical particles was established. The analytical equation was derived for the change in the Q-factor of natural resonance modes at small deviations of the particle shape from sphere. It was shown that the higher the Q-factor of the resonance modes, the stronger the effect of surface deformations on it.

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