Stimulated Raman scattering in a spherical microparticle

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Differential equations for time-dependent amplitudes of coupled waves at the Stokes frequency and the frequency of incident radiation in a spherical particle are obtained based on the method of expanding optical fields into a series over eigenfunctions of the stationary linear scattering problem. Solutions of the equations for the start of the SRS process and under steady state conditions are analyzed. The SRS threshold is determined, and the threshold for the steady state SRS at a given intensity is found for the case of double resonance between the fields. It is shown that to excite the SRS, one should compensate for the loss of the Stokes wave due to absorption and emission through the particle surface. To provide for steady state SRS generation, it is necessary to additionally compensate for the energy loss due to pump depletion.

Introduction

The studies of Stimulated Raman Scattering (SRS) generation in microparticles under the exposure to high-power laser radiation are important, first, for Raman spectroscopy of droplets, since they increase the sensitivity and information content of measurements as compared with a linear case.[1] On the other hand, problems of lasing at whispering gallery modes in microcavities are now actively discussed in the literature, that is, in the case that a microparticle plays the role of a microlaser.[2,3] The overview of these problems can be found, for example, in Refs. 1, 4, and 5.

The nature of stimulated emission from a spherical microcavity is connected with generation of radiation being in resonance with the cavity's eigenmodes. Therefore, the experimental and theoretical aspects of these two research areas have much in common. It should be noted that mostly the publications concerning this subject describe experiment. They consider physical grounds and report some findings on the SRS and Stimulated Brillouin Scattering (SBS) processes, third harmonic generation, induced fluorescence, and lasing in microparticles. To make these studies comprehensive, it is necessary to complete them with a consistent quantitative description of the generation process, which would allow one to correctly interpret and explain experimental findings, as well as to predict results anticipated from new experiments planed.

In this paper, we consider theoretical description of the SRS process in a transparent microparticle based on the method of expansion into a series over eigenfunctions of the problem on steady state linear scattering. The SRS effect in a particle is analyzed at the time of its initiation and under conditions of steady state scattering.

Basic equations

Assume that only two waves take part in the process of nonlinear scattering, namely, the pump wave and the Raman scattering (Stokes) wave with the frequencies ω_L and ω_s, respectively, which are related by the phase matching equation $\omega_s = \omega_L - \Omega_R$ (Ω_R is the frequency of molecular oscillations). The wave equations under these conditions have the following form:

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

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

where $\mathbf{E}_L$ and $\mathbf{E}_s$ are the real electric vectors of the pump and Stokes waves, respectively; ε_a and σ are the dielectric constant and conductivity of the particulate matter; c is the speed of light in vacuum; $\mathbf{P}_N^L$ and $\mathbf{P}_N^s$ are the real vectors of nonlinear polarization at the frequencies ω_L and ω_s. The medium is assumed nonmagnetic and isotropic; dispersion effects are ignored. The equations for the fields (1) and (2) are completed with the corresponding boundary conditions[4] consisting in the continuity of tangential spherical field components (θ and φ components) at the particle surface:

$$(\mathbf{E}_L)_{\theta,\varphi} = (\mathbf{E}_L^{sc})_{\theta,\varphi} + (\mathbf{E}_L^{i})_{\theta,\varphi};$$

$$(\mathbf{H}_L)_{\theta,\varphi} = (\mathbf{H}_L^{sc})_{\theta,\varphi} + (\mathbf{H}_L^{i})_{\theta,\varphi};$$

$$(\mathbf{E}_s)_{\theta,\varphi} = (\mathbf{E}_s^{sc})_{\theta,\varphi}, \quad (\mathbf{H}_s)_{\theta,\varphi} = (\mathbf{H}_s^{sc})_{\theta,\varphi},$$

where the superscripts "*sc*" and "*i*" are for the scattered and incident radiation, respectively.

Let us pass from the real field vectors to their complex representation:

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$$2\mathbf{E}(\mathbf{r};\,t) = \tilde{\mathbf{E}}(\mathbf{r};\,t)\,e^{i\omega t} + \tilde{\mathbf{E}}^*(\mathbf{r};\,t)\,e^{-i\omega t};$$

$$2\mathbf{P}_N(\mathbf{r};\,t) = \tilde{\mathbf{P}}_N(\mathbf{r};\,t)\,e^{i\omega t} + \tilde{\mathbf{P}}_N^*(\mathbf{r};\,t)\,e^{-i\omega t},$$

where $\tilde{\mathbf{E}}$, $\tilde{\mathbf{P}}_N$ are slowly varying functions of time, and represent the fields of interacting waves as series over particle's eigenfunctions $\mathbf{E}_{np}^{\mathrm{TE,TH}}(\mathbf{r})$, $\mathbf{H}_{np}^{\mathrm{TE,TH}}(\mathbf{r})$ that describe the spatial profiles of the fields of TE and TH vibrational modes with the frequencies $\omega_{np}^{\mathrm{TE,TH}}$:

$$\mathbf{E}_{\mathrm{L,s}}(\mathbf{r};\,t) = \sum_{n=1}^{\infty}\sum_{p=1}^{\infty}[A_{np}^{\mathrm{L,s}}(t)\,\mathbf{E}_{np}^{\mathrm{TE}}(\mathbf{r}) - iB_{np}^{\mathrm{L,s}}(t)\,\mathbf{E}_{np}^{\mathrm{TH}}(\mathbf{r})];$$

$$\mathbf{H}_{\mathrm{L,s}}(\mathbf{r};\,t) = \sqrt{\varepsilon_a}\sum_{n=1}^{\infty}\sum_{p=1}^{\infty}[iA_{np}^{\mathrm{L,s}}(t)\,\mathbf{H}_{np}^{\mathrm{TE}}(\mathbf{r}) + B_{np}^{\mathrm{L,s}}(t)\,\mathbf{H}_{np}^{\mathrm{TH}}(\mathbf{r})], \qquad (3)$$

where the functions $A_{np}^{\mathrm{L,s}}(t)$ and $B_{np}^{\mathrm{L,s}}(t)$ describe time behavior of the fields. The functions $\mathbf{E}_{np}^{\mathrm{TE,TH}}(\mathbf{r})$, $\mathbf{H}_{np}^{\mathrm{TE,TH}}(\mathbf{r})$ forming the orthogonal system within a sphere satisfy homogeneous Maxwell equations and are expressed through vector spherical harmonics $\mathbf{M}_{np}(r, \theta, \varphi)$, $\mathbf{N}_{np}(r, \theta, \varphi)$.[6]

Substitution of Eq. (3) into Eqs. (1)–(2) after some transformations (see Ref. 7) leads to the system of differential equations for the expansion coefficients of the incident and Stokes waves. Consider waves with TE polarization. The corresponding equations have the form:

$$\frac{d^2}{dt^2}A_{np}^{\mathrm{L,s}}(t) + 2\Gamma_{np}^{\mathrm{L,s}}\frac{d}{dt}A_{np}^{\mathrm{L,s}}(t) + \omega_{np}^2 A_{np}^{\mathrm{L,s}}(t) = J_{np}^{\mathrm{L,s}}(t), \quad (4)$$

where the "stimulating forces" are expressed as:

$$J_{np}^{\mathrm{L}}(t) = F_{np}^{\mathrm{i}}(t) + \frac{4\pi}{\varepsilon_a}\int_{V_a}\mathbf{E}_{np}^*\,\frac{\partial^2\mathbf{P}_{\mathrm{N}}^{\mathrm{L}}}{\partial t^2}\,d\mathbf{r},$$

$$J_{np}^{\mathrm{s}}(t) = -\frac{4\pi}{\varepsilon_a}\int_{V_a}\mathbf{E}_{np}^*\,\frac{\partial^2\mathbf{P}_{\mathrm{N}}^{\mathrm{s}}}{\partial t^2}\,d\mathbf{r}. \qquad (5)$$

Here $\Gamma_{np}^{\mathrm{L,s}} = \omega_{np}/[2Q_{np}(\omega_{\mathrm{L,s}})]$ is the mode attenuation factor; Q_{np} is the particle's total Q-factor that accounts for the total mode loss due to absorption and emission[4]; V_a is the particle volume. The term $F_{np}^{\mathrm{i}}(t)$ is connected with the influx of electromagnetic energy into the particle from the incident radiation; it can be found from the linear elastic scattering problem.[7]

Let a plane wave

$$\mathbf{E}^{\mathrm{i}}(\mathbf{r};\,t) = E_0\,\mathbf{p}_e\,\tilde{f}(t)\,\exp[i(\omega_{\mathrm{L}}t - k_{\mathrm{L}}z)],$$

be incident onto a spherical particle. Here E_0 is the real amplitude, $\mathbf{p}_e$ is the wave polarization vector, $k_{\mathrm{L}} = \omega_{\mathrm{L}}/c$, and $\tilde{f}(t)$ is the function of time (time profile of the radiation). From here on we restrict our consideration to the situation that radiation pulses incident on the particle have such duration that the delay effects of optical fields at scattering can be neglected. Under these conditions, for $F_{np}^{\mathrm{i}}(t)$ we obtain:

$$F_{np}^{\mathrm{i}}(t) = -\frac{ic}{\varepsilon_a}\times\left\{\int_{S_a}\left[\omega_{np}\,(\mathbf{E}^{\mathrm{i}}\times\mathbf{H}_{np}^*) - i\frac{\partial}{\partial t}(\mathbf{H}^{\mathrm{i}}\times\mathbf{E}_{np}^*)\right]\cdot\mathbf{n}_r\,d\mathbf{s}\right\} = E_0\,f(t)\,K_{np}^n. \qquad (6)$$

Here $f(t) = \tilde{f}(t)\,\exp\{i\omega_{\mathrm{L}}t\}$; $\mathbf{E}^{\mathrm{i}}$, $\mathbf{H}^{\mathrm{i}}$ are the vectors of the electric and magnetic fields of the incident wave; $\mathbf{n}_r$ is the external normal to the particle surface limited by the surface S_a. In this equation, the coefficient K_{np}^n accounts for the degree of excitation of the internal field mode (with the index "np") by each external field mode (with the index "n"). At circular polarization of the pump wave ($\mathbf{p}_e = \mathbf{e}_x + i\mathbf{e}_y$, where $\mathbf{e}_x$, $\mathbf{e}_y$ are basis vectors) for the considered TE modes of the internal field, it is equal, in particular, to

$$K_{np}^n = \frac{ic^2 R_n}{\varepsilon_a\,k_{\mathrm{L}}\,z_{np}\,V_a}\left[\psi_n(k_{\mathrm{L}}a_0)\,\psi_n'^*(n_a k_{np}a_0) - \frac{1}{n_a}\frac{\omega_{\mathrm{L}}}{\omega_{np}}\psi_n'(k_{\mathrm{L}}a_0)\,\psi_n^*(n_a k_{np}a_0)\right], \qquad (7)$$

where $k_{np} = \omega_{np}/c$; z_{np} is the normalization coefficient for the eigenfunctions:

$$z_{np}^{-2} = \int_0^{2\pi}d\varphi\int_0^{\pi}\sin\theta\,d\theta\int_0^{a_0}r^2\,dr\,|\mathbf{M}_{np}|^2;$$

ψ_n are Riccati–Bessel spherical functions; $R_n = i^n(2n+1)/[n(n+1)]$. Primes stand for derivatives with respect to the full argument of the function.

It should be noted that the equations for the coefficients of TH modes $B_{np}(t)$ are fully similar to the system (4). The only difference is the equation for the coefficients K_{np}^n.

Quasistationary approximation

The differential equation (4) is solved in the approximation of slowly varying amplitudes, that is, it is assumed that $A_{np}^{\mathrm{s}}(t) = \tilde{A}_{np}^{\mathrm{s}}(t)\,e^{i\omega_{\mathrm{s}}t}$, where $\tilde{A}_{np}^{\mathrm{s}}(t)$ is the slowly varying amplitude, or, what is the same, in the quasistationary approximation. The vectors of nonlinear polarization responsible for Raman scattering in the isotropic medium can be presented as follows for the waves with the frequencies ω_{L} and ω_{s}:

$$\mathbf{P}_{\mathrm{N}}^{\mathrm{L}}(\mathbf{r},\,t) = \chi_{\mathrm{R}}^{(3)}(\omega_{\mathrm{L}})\,[\mathbf{E}_{\mathrm{s}}(\mathbf{r},\,t)\cdot\mathbf{E}_{\mathrm{s}}^*(\mathbf{r},\,t)]\,\mathbf{E}_{\mathrm{L}}(\mathbf{r},\,t),$$

$$\mathbf{P}_{\mathrm{N}}^{\mathrm{s}}(\mathbf{r},\,t) = \chi_{\mathrm{R}}^{(3)}(\omega_{\mathrm{s}})\,[\mathbf{E}_{\mathrm{L}}(\mathbf{r},\,t)\cdot\mathbf{E}_{\mathrm{L}}^*(\mathbf{r},\,t)]\times\mathbf{E}_{\mathrm{s}}(\mathbf{r},\,t) + \mathbf{P}_{\mathrm{N}}^{\mathrm{sp}}(\mathbf{r},\,t), \qquad (8)$$

where $\chi_R^{(3)}$ is the nonlinear (Raman) third-order dielectric susceptibility of the medium, and $\mathrm{Im}\chi_R^{(3)}(\omega_L) = -\mathrm{Im}\chi_R^{(3)}(\omega_s)$; $\mathbf{P}_N^{sp}(\mathbf{r}, t)$ is the nonlinear polarization responsible for spontaneous Raman scattering.

At Raman resonance, the Raman susceptibility becomes purely imaginary

$$\chi_R^{(3)}(\omega_s) = -i\,\frac{N_0 T_2}{16 m \Omega_R}\left(\frac{\partial\alpha}{\partial \mathsf{q}_k}\right)^2$$

and it is usually related to the steady state SRS gain factor g_s:

$$g_s = -\frac{32\pi^2\,\omega_s}{\varepsilon_a\, c^2}\,\mathrm{Im}(\chi_R^{(3)}). \qquad (9)$$

Here q_k is the coordinate of nuclei displacement in the molecule; α is the medium polarizability; m is the reduced mass of the molecule; T_2 is the cross-relaxation time; N_0 is the concentration of medium molecules.

Because of the stochastic nature of spontaneous scattering, to reveal the form of the last term in Eq. (8), we have to solve the equation for the harmonic oscillator under the effect of a random force for the complex function q_k (see, for example, Ref. 8):

$$\frac{\partial^2 q_k}{\partial t^2} + 2\Gamma_k\,\frac{\partial q_k}{\partial t} + \Omega_R^2\, q_k = f_E(\mathbf{r};\, t) + f_{sp}(\mathbf{r};\, t), \quad (10)$$

where

$$2\mathsf{q}_k = q_k + q_k^*;\; f_E = \frac{1}{2m}\,\frac{\partial\alpha}{\partial q_k}\, n_m(\mathbf{E}_L\cdot\mathbf{E}_s^*)$$

is the stimulating force − the source of stimulated emission; n_m is the difference in population of the energy levels active in the Raman transition (in our approximation it is thought constant); $\Gamma_k = 1/T_2$; $f_{sp}(\mathbf{r};\, t)$ is the random distributed force. As to the latter, it is assumed delta-correlated in space and time

$$\langle f_{sp}(\mathbf{r};\, t)\, f_{sp}(\mathbf{r}';\, t')\rangle = F_0^2\delta(t - t')\,\delta(\mathbf{r} - \mathbf{r}');$$

F_0 is the root-mean-square amplitude of a random perturbation determined through the Raman scattering cross section of the matter.

At Raman resonance, the solution of equation (10) has the form:

$$q_k(t) = \frac{e^{-\Gamma_k t}}{\Omega_R}\int_0^t \sin[\mathring{\Omega}_R(t - t')]\, e^{\Gamma_k t'}\,[f_E(t') + f_{sp}(t')]\,\mathrm{d}t' = q_k^E(t) + q_k^{sp}(t),$$

where

$$\mathring{\Omega}_R = \Omega_R\sqrt{1 - (\Gamma_k^2/\Omega_R^2)}.$$

For the spontaneous component of the Stokes polarization, we obtain:

$$\mathbf{P}_N^{sp}(\mathbf{r},\, t) = N_0\,\frac{\partial\alpha}{\partial q_k}\,(q_k^{sp})^*\,\mathbf{E}_L(\mathbf{r};\, t) + \mathrm{c.c} \approx$$

$$\approx -\frac{N_0}{\Omega_R}\,\frac{\partial\alpha}{\partial q_k}\,\tilde{\mathbf{E}}_L(\mathbf{r};\, t)\, e^{i\omega_s t - \Gamma_k t}\int_0^t \sin[\mathring{\Omega}_R(t - t')]\,\times$$

$$\times\, e^{\Gamma_k t'}\, f_{sp}^*(\mathbf{r};\, t')\,\mathrm{d}t' + \mathrm{c.c}, \qquad (11)$$

where $\tilde{\mathbf{E}}_L$ is the slowly varying function of time.

Transform Eq. (5) for the source of the Stokes wave J_{np}^s. For this purpose, let us expand the fields into a series over particle's eigenmodes and take into account Eq. (8). As a result, we obtain:

$$J_{np}^s(t) = -\frac{4\pi\,\chi_R^{(3)}(\omega_s)}{\varepsilon_a}\sum_{n'}\sum_{p'}\frac{\mathrm{d}^2}{\mathrm{d}t^2}\left[\,|A_{n'p'}^L|^2\sum_m\sum_q A_{mq}^s(t)\right]\times$$

$$\times\int_{V_a}(\mathbf{E}_{n'p'}\cdot\mathbf{E}_{n'p'}^*)\cdot(\mathbf{E}_{mq}\cdot\mathbf{E}_{np}^*)\,\mathrm{d}\mathbf{r} + F_{np}^{sp}(t),$$

where

$$F_{np}^{sp}(t) = -\frac{4\pi}{\varepsilon_a}\int_{V_a}\mathbf{E}_{np}^*\,\frac{\partial^2\,\mathbf{P}_N^{sp}}{\partial t^2}\,\mathrm{d}\mathbf{r}$$

is the source of spontaneous radiation at the frequency ω_s. By definition, the eigenmodes, over which the field is expanded, are believed interacting with each other within the volume of the particle-microcavity.[9] Energy exchange between the modes is possible only in the presence of local inhomogeneities in the dielectric constant or through mode interaction on the particle surface at its deformations. Consequently, in the equation obtained, we can omit summation over modes of the Stokes field. With the allowance for Eq. (11), we obtain:

$$J_{np}^s(t) = -i\,\frac{c^2\, g_s}{8\pi\omega_s}\,\times$$

$$\times\sum_{n'}\sum_{p'}\frac{\mathrm{d}^2}{\mathrm{d}t^2}[\,|A_{n'p'}^L(t)|^2\, A_{np}^s(t)]\, S_{n'p'}^{np} + F_{np}^{sp}(t), \quad (12)$$

where $S_{n'p'}^{np}$ stands for the integral of spatial overlap of optical modes belonging to the incident and the Stokes waves:

$$S_{n'p'}^{np} = \int_{V_a}[(\mathbf{E}_{n'p'}\cdot\mathbf{E}_{n'p'}^*)\cdot(\mathbf{E}_{np}\cdot\mathbf{E}_{np}^*)]\,\mathrm{d}\mathbf{r}, \qquad (13)$$

and summation is taken only over the pump field modes.

Coming back to Eq. (12), note that every pump field mode contributes to the development of the selected mode of Raman scattering wave, and its contribution is proportional to the parameter of overlap of these modes.

Within the quasistationary approximation, the initial equation takes the form:

$$\left\{2i\omega_s + 2\Gamma_{np}^s + \frac{2i}{\omega_s}\left[\frac{\mathrm{d}G_{np}^s(t)}{\mathrm{d}t} + i\omega_s\, G_{np}^s(t)\right]\right\}\frac{\mathrm{d}\,\tilde{A}_{np}^s(t)}{\mathrm{d}t} +$$

$$+\,\omega_s^2\left[\Delta_{np_s}^2 + \frac{2i\,\Gamma_{np}^s}{\omega_s} - \frac{2}{\omega_s^2}\,\frac{\mathrm{d}\,G_{np}^s(t)}{\mathrm{d}t} + \frac{i}{\omega_s}\,G_{np}^s(t)\right]\times$$

$$\times\,\tilde{A}_{np}^s(t) = \tilde{F}_{np}^{sp}(t), \qquad (14)$$

where $\Delta_{np_s} = (\omega_{np} - \omega_s)/\omega_s$ is the relative frequency detuning of the Stokes field mode;

$$G^{\rm s}_{np}(t) = \frac{c^2\, g_{\rm s}}{8\pi} \sum_{n'}\sum_{p'} |A^{\rm L}_{n'p'}(t)|^2\, S^{np}_{n'p'}.$$

Express the amplitude coefficient of the Stokes mode $A^{\rm s}_{np}(t)$ through the integral equation. Under conditions favorable for the development of stimulated emission at the long-living resonance modes ($\omega_{\rm L,s} \gg \Gamma^{\rm s}_{np}$, $G^{\rm s}_{np}$), this equation can be written as:

$$A^{\rm s}_{np}(t) = \tilde{A}^0_{np_{\rm s}}(t)\, e^{D^{\rm s}_{np}(t)} \exp\,[i\mathring{\omega}^{\rm s}_{np}\, t + i\varphi^{\rm s}_{np}(t)]. \quad (15)$$

In Eq. (15), the following designations are introduced:

$$\mathring{\omega}^{\rm s}_{np} = \omega_{\rm s}\,(1 - \Delta^2_{np_{\rm s}}/2)$$

is the mode frequency of the Stokes wave field generated;

$$\varphi^{\rm s}_{np}(t) = \frac{1}{2\omega_{\rm s}} \int_0^t [G^{\rm s}_{np}(t') - \Gamma^{\rm s}_{np}]^2\, {\rm d}t';$$

$$D^{\rm s}_{np}(t) = \frac{1}{2}\,(1 - \Delta^2_{np_{\rm s}}) \int_0^t [G^{\rm s}_{np}(t') - 2\Gamma^{\rm s}_{np}]\, {\rm d}t'$$

is the function accounting for the mode gain and attenuation;

$$\tilde{A}^0_{np_{\rm s}}(t) = \frac{1}{2i\omega_{\rm s}} \int_0^t e^{-D^{\rm s}_{np}(t')}\, \tilde{F}^{\rm sp}_{np}(t'){\rm d}t'$$

is the amplitude factor characterizing spontaneous Raman scattering.

The equation for the intensity of stimulated scattering wave averaged over the particle volume

$$\bar{I}_{\rm s}(t) = \frac{1}{V_a} \int_{V_a} I_{\rm s}(\mathbf{r};\, t)\, {\rm d}\mathbf{r} = \frac{c\, n_a}{8\pi V_a} \sum_n \sum_p |A^{\rm s}_{np}(t)|^2$$

with the allowance for Eqs. (9)–(15) under condition of resonance excitation ($\Delta_{np_{\rm s}} = 0$) can be written as (mode indices are omitted):

$$\bar{I}_{\rm s}(t) = \bar{I}_{\rm sp}(t)\, e^{2D^{\rm s}(t)}. \quad (16)$$

Here

$$\bar{I}_{\rm sp}(t) = 2\left[\frac{8\pi\, N_0 F_0}{\Gamma_k\, n_a\, \Omega_{\rm R}} \frac{\partial\alpha}{\partial q_k}\right]^2 \int_0^t e^{-2D^{\rm s}(t')} \times$$

$$\times \sum_{n'}\sum_{p'} |A^{\rm L}_{n'p'}(t')|^2\, S^{np}_{n'p'}\, {\rm d}t'$$

characterizes the intensity of spontaneous Raman scattering. The intensity $\bar{I}_{\rm sp}$ tends to saturation as the factor $D^{\rm s}$ increases.

For the pump wave, the integral formulation of the problem is as follows:

$$A^{\rm L}_{np}(t) = \tilde{A}^0_{np_{\rm L}}(t)\, e^{-D^{\rm L}_{np}(t)} \exp\,[i\mathring{\omega}^{\rm L}_{np}(t) + i\varphi^{\rm L}_{np}(t)],$$

where

$$\mathring{\omega}^{\rm L}_{np} = \omega_{\rm L}\left(1 - \frac{\Delta^2_{np_{\rm L}}}{2}\right);\quad \Delta_{np_{\rm L}} = \frac{\omega_{np} - \omega_{\rm L}}{\omega_{\rm L}};$$

$$\varphi^{\rm L}_{np}(t) = -\frac{1}{2\omega_{\rm L}} \int_0^t [G^{\rm L}_{np}(t') + \Gamma^{\rm L}_{np}]^2\, {\rm d}t';$$

$$D^{\rm L}_{np}(t) = \frac{1}{2}\,(1 - \Delta^2_{np_{\rm L}}) \int_0^t [G^{\rm L}_{np}(t') + 2\Gamma^{\rm L}_{np}]\, {\rm d}t';$$

$$\tilde{A}^0_{np_{\rm L}}(t) = \frac{1}{2i\omega_{\rm L}} \int_0^t e^{D^{\rm L}_{np}(t')}\, \tilde{F}^{\rm i}_{np}(t'){\rm d}t';$$

$$G^{\rm L}_{np}(t) = \frac{c^2\, g_{\rm s}\, \omega_{\rm L}}{8\pi\omega_{\rm s}} \sum_n\sum_p |A^{\rm s}_{np}(t)|^2\, S^{np}_{n'p'}.$$

SRS thresholds

To find the SRS threshold, we have to express the volume-averaged pump intensity inside the particle $\bar{I}_{\rm L}$ through the incident radiation intensity $I_0 = cE_0^2/8\pi$. Two problem formulations are possible, namely, (1) determination of the threshold for initiation of stimulated emission; (2) determination of the threshold for SRS generation with the intensity exceeding the intensity of spontaneous scattering. Consider the first one. Here we can apparently use the approximation of the given pump field.

The function $G^{\rm s}_{np}$ can be presented as follows:

$$G^{\rm s}_{np}(t) = (cg_{\rm s}/n_a)\, \bar{B}^{np}_c\, \bar{I}_{\rm L}(t), \quad (17)$$

where

$$\bar{I}_{\rm L}(t) = \frac{1}{V_a}\int_{V_a} I_{\rm L}(\mathbf{r};\, t)\, {\rm d}\mathbf{r} = \frac{cn_a}{8\pi V_a}\sum_{n'}\sum_{p'} |A^{\rm L}_{n'p'}(t)|^2; \quad (18)$$

$$\bar{B}^{np}_c(\omega_{\rm L};\, \omega_{np}) = \frac{cn_a}{8\pi \bar{I}_L} \sum_{n'}\sum_{p'} |A^{\rm L}_{n'p'}|^2\, S^{np}_{n'p'} \quad (19)$$

is the normalized coefficient of spatial overlap of interacting fields inside the particle. The coefficient $\bar{B}^{np}_c$ weakly depends on time both at the initial stage and at the stage of steady state SRS. At the initial stage of the process, it can be calculated separately in the linear approximation, that is, within the Mie theory. In this case, $\bar{B}^{np}_c$ is largely determined by the particle morphology and optical properties. In the case that the Stokes wave field can be assumed single-mode ($\omega_{np} = \omega_{\rm s}$), from Eq. (19) with the allowance for Eq. (13) it follows that

$$\bar{B}^{np}_c(\omega_{\rm L};\, \omega_{\rm s}) = V_a\left[\int_{V_a} (\mathbf{E}_{\rm L}\cdot\mathbf{E}^*_{\rm L})\, {\rm d}\mathbf{r}\right]^{-1} \times$$

$$\times \int_{V_a} (\mathbf{E}_{np}\cdot\mathbf{E}^*_{np}) \sum_{n'}\sum_{p'} |A^{\rm L}_{n'p'}|^2\, (\mathbf{E}_{n'p'}\cdot\mathbf{E}^*_{n'p'})\, {\rm d}\mathbf{r}.$$

After convolution of the sum in the right-hand side of this equation and making use of the identity following from the eigenfunctions normalization condition

$$|A^{s}_{np}|^2 = \int_{V_a} (\mathbf{E}_s \cdot \mathbf{E}^*_s)\, d\mathbf{r},$$

Eq. (19) for the field overlap coefficient takes the following form (mode indices are omitted):

$$\bar{B}_c(\omega_L;\,\omega_s) = V_a \left[\int_{V_a} (\mathbf{E}_L \cdot \mathbf{E}^*_L)\, d\mathbf{r} \cdot \int_{V_a} (\mathbf{E}_s \cdot \mathbf{E}^*_s)\, d\mathbf{r} \right]^{-1} \times$$
$$\times \int_{V_a} (\mathbf{E}_L \cdot \mathbf{E}^*_L)\,(\mathbf{E}_s \cdot \mathbf{E}^*_s)\, d\mathbf{r}. \qquad (20)$$

Within the approximation of the given field, the ratio $\bar{I}_L/I_0$ is constant in time and determined by the following integral equation:

$$\frac{\bar{I}_L}{I_0} = \bar{B}_L = \frac{1}{V_a E_0^2} \int_{V_a} \mathbf{E}_L(\mathbf{r})\, \mathbf{E}^*_L(\mathbf{r})\, d\mathbf{r}. \qquad (21)$$

Note that in the majority of cases the factor $\bar{B}_L$ is equal to unity and significantly differs from it only at resonance excitation of the particle by the pump field.

Then, finally, the energy density of the incident light wave w_0, at which stimulated emission occurs in the particle, is determined by the following equation:

$$w_0 > w_0^{th} = n_a\, \omega_s\, t/(c g_e\, Q_s\, \bar{B}_L). \qquad (22)$$

In Eq. (22), the effective coefficient of SRS amplification in the microcavity is introduced as

$$g_e = g_s\, \bar{B}_c.$$

This coefficient reflects the difference in the rate of the Stokes wave generation in the particle as compared with the extended medium. This leads to a significant decrease of the process thresholds and in some cases allows the continuous-wave radiation to be used to pump the microcavity.[10,11]

Figure 1 shows the dependence of the g_e/g_s ratio on the effective Q-factor of the Stokes field eigenmodes Q_s. The calculation was performed for water droplets of different radius ($n_a = 1.33$, $\lambda_L = 0.53\ \mu m$; $\lambda_s = 0.65\ \mu m$) in two situations of nonlinear interaction between waves: resonance of only the Stokes field ("single" resonance) and resonance of both waves ("double" resonance).

It follows from Fig. 1 that the ratio g_e/g_s is close to unity at nonresonance SRS excitation. A significant growth of the efficiency of nonlinear interaction is observed only in the case of double resonance between the fields. This circumstance was noticed for the first time in the experimental paper[12] when studying the SRS excitation thresholds in droplets of a water solution of glycerin. Detailed theoretical studies of the coefficient of spatial overlap of the fields at different variants of SRS and SBS excitation in spherical particles were performed, for example, in Refs. 4 and 13.

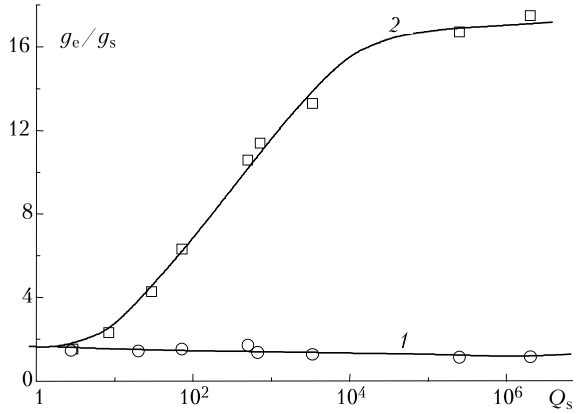

Fig. 1. The dependence of g_e/g_s ratio on the Q-factor for the resonance modes Q_s of the Stokes field at excitation of the SRS due to single (*1*) and double (*2*) field resonance. Solid line is a guide for eye.

If the incident radiation is long enough in time, then we can turn to the radiation intensity in place of the energy density in this equation. Thus, the condition of SRS generation at continuous or quasicontinuous pump takes the following form:

$$I_0 > I_0^{th} = \frac{n_a\, \omega_s}{c g_e\, Q_s\, \bar{B}_L} = \frac{n_a^2\, V_a\, \omega_L\, \omega_s}{c^2\, g_e\, \sigma_{ex}(a_0;\, \omega_L)\, Q_L Q_s}, \qquad (23)$$

where $\sigma_{ex}(a_0;\, \omega_L)$ is the extinction cross section of a particle for the incident radiation. This equation was earlier derived in our papers 4 and 13 when considering the energy balance of the Stokes wave in a particle.

Let us emphasize once more that the considered excitation threshold of the Raman wave corresponds, in fact, to fulfillment of the condition for appearance of positive feedback in the particle-cavity for the Stokes wave, when its total loss due to absorption and emission through the particle surface becomes equal to the gain due to nonlinear interaction with the pump field. The intensity of stimulated scattering under such conditions is low and, as is seen from Eqs. (15)–(16), corresponds to the intensity of spontaneous Raman scattering.

The problem of excitation of the SRS wave with a preset intensity level involves determination of a certain level of the SRS gain coefficient G^s in the particle, which, in its turn, depends on the pump wave intensity. The solution to this problem can be obtained only numerically, and we plan to present it in our following papers. Here we restrict the consideration to analysis of an important issue – the steady state of the SRS generation in a particle.

At $I_0 \geq I_0^{th}$ the generation of stimulated radiation occurs in the particle; this generation is, in the general case, nonstationary, and the steady state SRS can be established at a rather long irradiation. Such a steady state SRS generation in a microparticle was observed experimentally in Refs. 10 and 11.

The condition for achieving a steady state nonlinear scattering is the zero time derivative of the right-hand side of Eq. (16). This is achieved, when the volume-average intensity of the incident wave inside the particle also reaches some stationary level

$$\bar{I}_{\rm L}^{\rm st} = 2n_a\,\Gamma^{\rm s}/(cg_{\rm e}). \quad (24)$$

Let us find the relation of the intensity of incident radiation I_0 to the intensity of the steady state SRS, $I_{\rm s}^{\rm st}$. Write the integral equation for the intensity of pump wave field inside the particle under conditions of double resonance between the fields, when the laser radiation incident on the particle is in resonance with one particle's mode, and SRS generation occurs at other particle's mode (mode indices are omitted):

$$\bar{I}_{\rm L}(t) = \bar{I}_{\rm L}^0(t)\; e^{-2D^{\rm L}(t)}, \quad (25)$$

where

$$\bar{I}_{\rm L}^0(t) = (cn_a/8\pi)\;\tilde{A}_{\rm L}^0(t)\;[\tilde{A}_{\rm L}^0(t)]^*.$$

From Eqs. (24)–(25) under condition of quasicontinuous excitation, we have the sought relationship between the steady state intensities of the interacting waves in the particle and the pump intensity:

$$\bar{I}_{\rm L}^{\rm st} = 4\pi\,|K_{np}^n|^2\; I_0/[(G_{\rm st}^{\rm L} + 2\Gamma^{\rm L})^2\; cn_a\;\omega_{\rm L}^2]. \quad (26)$$

Here $G_{\rm st}^{\rm L}$ is the value of the factor $G^{\rm L}$ in the steady state case.

For a low-intensity Stokes radiation, Eq. (26) transforms into the above equation for the threshold intensity of the incident field leading to SRS generation under conditions of a steady state pump. For generation of the Stokes wave of higher intensity, the corresponding threshold value increases by $(1 + G_{\rm st}^{\rm L}/2\Gamma^{\rm L})^2$ times.

Thus, at a steady state generation of the Stokes radiation, the energy of the incident light field is additionally lost in the particle-microcavity, and this is equivalent to a decrease in the cavity Q-factor at the frequency of incident radiation. In this treatment of the processes, the relation for the threshold intensity of incident radiation leading to the SRS in a particle keeps true for the case of excitation of the SRS wave having a finite amplitude with the only difference that $Q_{\rm L}$ in Eq. (23) is replaced with $Q_{\rm L}(1 + \eta)^{-2}$, where η is the pump depletion factor.

This factor can be determined from numerical solution of the stationary SRS problem. However, for tentative estimates we can use its approximate value obtained from the linear theory:

$$\eta \approx \frac{cg_{\rm e}\,\omega_{\rm L}}{2\Gamma^{\rm L}\,n_a\,\omega_{\rm s}}\,\bar{I}_{\rm s}^{\rm st} = \frac{Q_{\rm L}}{Q_{\rm s}}\,\frac{\bar{I}_{\rm s}^{\rm st}}{\bar{I}_{\rm L}^{\rm st}}\,.$$

The level $\eta = 1$ corresponds to the condition that pump depletion should be taken into account. Thus, the pump depletion conditions occur at

$$\bar{I}_{\rm s}^{\rm st} = (Q_{\rm s}/Q_{\rm L})\;\bar{I}_{\rm L}^{\rm st}$$

in the stationary SRS regime.

Conclusion

In this paper, we have considered theoretically the process of stimulated Raman scattering in a transparent microparticle using the approach based on expansion of optical fields of coupled waves into series over eigenfunctions of the stationary linear scattering problem. Differential equations are obtained for time-dependent amplitudes of the waves at the Stokes frequency and the frequency of the incident radiation in a spherical microparticle. Analysis of solutions of the equations for the initial stage of the SRS process and the conditions for occurrence of the steady state showed that the threshold of SRS excitation is determined by the loss of the Stokes wave due to absorption and emission through the particle surface. To provide for steady state SRS generation, one should additionally compensate for the energy loss due to pump depletion.

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