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Efficiency of anti-Stokes generation in nonstationary stimulated Raman scattering

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***Abstract*. Nonstationary SRS equations are solved analytically for short samples in the approximation of a prescribed exciting field, and numerically for a longer medium taking into account the pumping depletion. The dependence of the energy of Stokes and anti-Stokes waves on their spatial mismatching is studied. The optimal angle of anti-Stokes generation is determined as a function of the length of the system, pump pulse energy, and the rate of transverse relaxation.**

Keywords: *nonstationary stimulated Raman scattering, spatial mismatching, intensities of Stokes and anti-Stokes waves, optimal angle, transverse relaxation.*

1. Introduction

Anti-Stokes emission is usually produced upon the action of a light wave on excited molecules. Apart from this, it may also appear due to the parametric interaction between the laser and Stokes waves [1 – 4]. In the general case, the SRS contains both these effects and the difference between them is arbitrary to a certain extent [4]. For a low excitation energy of molecules, energy transfer from the laser and Stokes waves to an anti-Stokes wave is the dominating process supplying energy to the anti-Stokes wave. The efficiency of this process depends on the phase difference Δkz (see expression (2) below) between the interacting waves [1 – 12].

It has been established theoretically that for stationary SRS in the absence of a phase shift ($\Delta k = 0$), both Stokes and anti-Stokes emission is suppressed [1 – 3]. Experimentally, this effect is frequently observed as a dark region in the Stokes emission pattern of single-pass Raman amplifiers [5 – 6].

In the nonstationary case, when the pump pulse duration is shorter than the transverse relaxation time of molecules or is comparable with it, the effect of the spatial mismatching Δk on the efficiency of anti-Stokes generation is found to be somewhat different [7 – 12]. An analytic solution of the corresponding equations in the approximation of a prescribed pumping field shows that in the asymptotic approximation, the intensity of the anti-Stokes component away from the input boundary is maximum for a certain Δk, and is equal to zero for $\Delta k = 0$ and $\Delta kz \gg 1$, as in the stationary case [7, 8].

The numerical solution of a more general problem shows that the intensity of the initial Stokes radiation additionally affects the efficiency of parametric interaction [9, 10]. When this intensity is low, a situation analogous to the stationary SRS takes place [1 – 4], which was also confirmed experimentally [11]. If, however, the intensity of the input Stokes signal is significant and is only an order of magnitude lower than the pump intensity, the efficiency of conversion of the energy of Stokes and laser waves into the anti-Stokes wave energy is maximal for $\Delta k = 0$ and decreases with increasing Δk [9, 10, 12].

SRS was studied in the amplification regime in Refs [7 – 9, 12], when frequency-shifted weak signals were supplied along with the pump pulse at the input face of the sample. In this work, we analyse the situation which arises, for example, in a single-pass Raman oscillator in which the Stokes emission is produced due to spontaneous Raman scattering. This problem is analogous to the one considered earlier in Ref. [8], but in contrast to that problem, we consider the nonstationary SRS both in relatively short samples (in which the exciting wave intensity remains virtually unchanged) and in longer samples with a significant pumping depletion. The stochastic nature of spon-taneous Raman scattering and the finite value of the transverse relaxation time in the Raman transition are taken into account.

2. Model equations

Consider a medium represented by identical molecules with frequencies ω_{rq} and dipole transition moments $\boldsymbol{d}_{rq}$, $q = 1, 2$ corresponding to the initial and final levels of the Raman transition, and $r = 3, 4, 5, \ldots$ corresponding to the intermediate states ($d_{12} = 0$). We assume that a plane linearly polarised wave is incident on the medium, the intensity of the wave being moderate and its frequency lying far from the resonance with any of the molecule transitions: $|d_{rq}E_L| \ll \hbar(\omega_L - \omega_{rq}), \hbar\omega_{rq}$.

Because the efficiency of the backward Stokes scattering is insignificant in the nonstationary case [13], and the intensities of SRS components of an order higher than the first are relatively low [14], the field in the medium can be represented in the form

$$\boldsymbol{E}(\boldsymbol{r}, t) = \sum_{f=\mathrm{L,s,a}} \boldsymbol{e}_f E_f(z, t) \exp(-\mathrm{i}\omega_f t + \mathrm{i}\boldsymbol{k}_f \boldsymbol{r}) + \mathrm{c.c.} \qquad (1)$$

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Here, E_f are slowly varying amplitudes of laser radiation ($f = \mathrm{L}$) and copropagating Stokes ($f = \mathrm{s}$) and anti-Stokes (f=a) waves; $\boldsymbol{e}_f$ is the corresponding unit vector of polarisation; $\omega_\mathrm{s} = \omega_\mathrm{L} - \omega_{21}$; $\omega_\mathrm{a} = \omega_\mathrm{L} + \omega_{21}$; and ω_{21} is the Raman transition frequency. The degree of phase mismatching of the waves along the z axis is

$$\Delta k = k_{\mathrm{s}z} + k_{\mathrm{a}z} - 2k_{\mathrm{L}z},\ k_{\mathrm{s}x,\mathrm{s}y} + k_{\mathrm{a}x,\mathrm{a}y} - 2k_{\mathrm{L}x,\mathrm{L},y} = 0, \quad (2)$$

where $k_f = \omega_f/v_f = \eta_f\omega_f/c$; v_f is the velocity of the corresponding wave; η_f is the linear part of the refractive index at the frequency ω_f; z and x, y are the axes along and perpendicular to the direction of propagation of the wave E_L (Fig. 1).

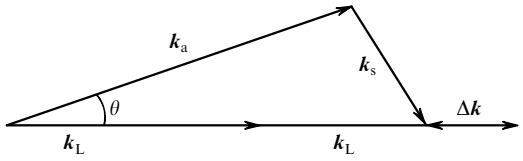

Figure 1. Vector diagram for anti-Stokes parametric generation.

Under typical experimental conditions [11], the pump intensity is $\sim 5 \times 10^9$ W cm^{-2} and below, hence the average number of photons per molecule of a gas under normal conditions and for visible radiation (at ~ 0.5 μm) does not exceed 1.5×10^{-2}; i.e., the level population can be treated as constant in this case. For copropagating waves in gases for samples of length $L \sim 1$ m and below, the quantity $L|v_f^{-1} - v_{f'}^{-1}|$ ($f, f' = \mathrm{L}$, s, a) defining the time of their group delay does not exceed 10^{-14} s. Thus, the effect of the difference in the velocities of interacting waves on the SRS kinetics can be neglected in the case of nanosecond pulses. However, the dispersion of the Raman medium may play a significant role in the estimates of the anti-Stokes generation angle.

Under these conditions, the nonstationary SRS is described by the system of equations [15]:

$$\frac{\partial \mathscr{E}_\mathrm{L}}{\partial \xi} = 2\mathrm{i}[\alpha Q \mathscr{E}_\mathrm{s} + \beta Q^* \mathscr{E}_\mathrm{a} \exp(\mathrm{i}q\xi)], \quad (3)$$

$$\frac{\partial \mathscr{E}_\mathrm{s}}{\partial \xi} = 2\mathrm{i}r_\mathrm{s}\alpha^* \mathscr{E}_\mathrm{L} Q^*, \quad (4)$$

$$\frac{\partial \mathscr{E}_\mathrm{a}}{\partial \xi} = 2\mathrm{i}r_\mathrm{a}\beta^* \mathscr{E}_\mathrm{L} Q \exp(-\mathrm{i}q\xi), \quad (5)$$

$$\frac{\partial Q}{\partial \tau} = \mathrm{i}[\alpha^* \mathscr{E}_\mathrm{L} \mathscr{E}_\mathrm{s}^* + \beta \mathscr{E}_\mathrm{L}^* \mathscr{E}_\mathrm{a} \exp(\mathrm{i}q\xi)] - \gamma Q. \quad (6)$$

Here, $\mathscr{E}_f = \eta E_f/(\pi n_0 \hbar \omega_\mathrm{L})^{1/2}$ are dimensionless complex amplitudes of the Stokes ($f = \mathrm{s}$), anti-Stokes ($f = \mathrm{a}$), and laser fields ($f = \mathrm{L}$); n_0 is the concentration of molecules; $\eta = \eta_f$ ($f = \mathrm{L}, \mathrm{s}, \mathrm{a}$); Q is the nondiagonal element of the collective density matrix, which describes the polarisation induced in the system; $\xi = z/v\Omega^{-1}$ is the dimensionless coordinate; $v = v_f$ ($f = \mathrm{L}, \mathrm{s}, \mathrm{a}$); $\tau = (t - z/v)/\Omega^{-1}$ is the dimensionless delay time; $\gamma = \Gamma/\Omega$; $q = \Delta k v/\Omega$; $r_{\mathrm{s,a}} = \omega_{\mathrm{s,a}}/\omega_\mathrm{L}$; $\alpha = \mu_\mathrm{s}/|\mu_\mathrm{s}|$; $\beta = \mu_\mathrm{a}/|\mu_\mathrm{s}|$; Γ is the transverse relaxation rate for the $|1\rangle - |2\rangle$ transition; $\mu_\mathrm{s} = \mu_{12}^\mathrm{s}$, $\mu_\mathrm{a} = \mu_{21}^\mathrm{a}$ are the Raman polarisabilities defined by the expression

$$\mu_{mn}^f = \frac{1}{\hbar}\sum_r \frac{(\boldsymbol{d}_{mr}\boldsymbol{e}_f)(\boldsymbol{d}_{rn}\boldsymbol{e}_\mathrm{L})}{\omega_{rn} + \omega_\mathrm{L}} + \frac{(\boldsymbol{d}_{mr}\boldsymbol{e}_\mathrm{L})(\boldsymbol{d}_{rn}\boldsymbol{e}_f)}{\omega_{rm} - \omega_\mathrm{L}}$$

($f = \mathrm{s}, \mathrm{a}$; $m \neq n = 1, 2$). The quantities $\Omega^{-1} = \eta^2/\pi n_0 \omega_\mathrm{L}|\mu_\mathrm{s}|$ and $v\Omega^{-1}$ define the time and spatial scales of the nonstationary SRS, respectively. We take into account the dependence of the refractive index on the frequency of light in expression (2). A similar approach was used in Ref. [14].

We assume that the Stokes emission is generated in the medium due to spontaneous Raman scattering, which can be described by specifying a small initial polarisation

$$\mathscr{E}_\mathrm{s}(0, \tau) = 0, \quad (7)$$

$$Q(\xi, 0) = a, \quad (8)$$

while the anti-Stokes component is generated due to parametric interaction of the laser and Stokes waves, i.e.,

$$\mathscr{E}_\mathrm{a}(0, \tau) = 0. \quad (9)$$

In expression (8), a is a random quantity described by the Gaussian distribution

$$P_0(a) = \frac{2a}{\kappa^2}\exp\left(-\frac{a^2}{\kappa^2}\right), \quad (10)$$

where $\kappa = 1/\sqrt{N}$ is the distribution width [16–18], and N is the total number of molecules in the sample.

It follows from Eqns (3)–(6) that their solutions have a certain symmetry relative to the quantity q:

$$\mathscr{E}_f(-q) = -\mathscr{E}_f^*(q) \quad (f = \mathrm{L}, \mathrm{s}, \mathrm{a}), \quad Q(-q) = Q^*(q). \quad (11)$$

Thus, the intensities of the waves and the polarisation of the medium are independent of the sign of q.

As in previous papers [7–12], we consider the scattering for the case of a weak dependence of Raman polarisabilities on frequency:

$$\omega_\mathrm{s}|\mu_\mathrm{s}|^2 \approx \omega_\mathrm{a}|\mu_\mathrm{a}|^2.$$

3. Solution of Maxwell–Bloch equations

Let us estimate a number of quantities appearing in Eqns (3)–(6) for the example of SRS in hydrogen. According to Ref. [19], the transverse relaxation time T_2 for the $Q_{01}(1)$ vibrational–rotational transition is 7.5×10^{-9} s atm/p_0 (where p_0 is the gas pressure). At low pressures, the polarisation relaxation time associated with the motion of molecules is $T_2^* = 2/(k_\mathrm{L} - k_\mathrm{s})\bar{V}$ ($\bar{V}$ is the average thermal velocity of molecules), which amounts to about 4.3×10^{-10} s for the given transition at room temperature ($T = 300$ K).

Due to a decrease in the mean free path of molecules and its approaching to the length $1/(k_\mathrm{L} - k_\mathrm{s})$ with increasing pressure, the average velocity determining the Doppler broadening becomes lower than the mean thermal velocity $\bar{V}$ and continues to decrease gradually [20]. In this case, the inhomogeneous broadening of the Stokes line decreases, while its homogeneous broadening increases, so that its total

width decreases at first, attains its minimum value $\Delta\omega_{\min} = 4 \times 10^8$ s^{-1} (Dicke effect) in the vicinity of $p_0 = 1.85$ atm, and then increases [21]. Because the time $T_2 \simeq 4.1 \times 10^{-9}$ s in the vicinity of $\Delta\omega_{\min}$, the value of T_2^* is about 6.7 ns.

If the second harmonic radiation from a neodymium laser ($\lambda_L = 5.32 \times 10^{-7}$ m) is used for pumping, the Stokes wavelength is $\lambda_s = 6.83 \cdot 10^{-7}$ because the width $\Delta\nu$ of the $Q_{01}(1)$ transition is 4158.55 cm^{-1} [22]. Since the differential Raman cross section for hydrogen molecules is $d\sigma/d\Omega \simeq 5.28 \times 10^{-31}$ cm^2 mole^{-1} sr^{-1} [23], the quantity $|\mu_s|^2 = k_s^4 d\sigma/d\Omega$ will be approximately equal to 5×10^{-51} cm^6.

For the pressure $p_0 = 3$ atm at temperature $T = 300$ K, the concentration of molecules is $n_0 = 7.2 \times 10^{19}$ cm^{-3}. Because only 66.7 % molecules are in the ground state in this case, we obtain $\Omega = 3.7 \times 10^{10}$ s^{-1}. Therefore, the temporal scale is $t_0 = \Omega^{-1} \simeq 2.7 \times 10^{-11}$ s and the spatial scale is $L_0 = v\Omega^{-1} \simeq 8.2$ mm. Thus, choosing the pump pulse duration $t_L < T_2^* \simeq 7$ ns (several hundred t_0), we can neglect the inhomogeneous broadening, whereas the transverse relaxation should be taken into account.

3.1 Short sample

Consider first a medium of a relatively small size, i.e., the case $L \simeq (1-2)L_0$ ($L \sim 1$ cm for the experimental conditions considered above). In this case, the change in the pump field during SRS can be neglected and treated as specified, i.e., $\mathscr{E}_L = \mathscr{E}_L(0,\tau)$ [24].

Let us introduce the new amplitudes of fields and polarisation

$$A_s = \alpha \mathscr{E}_s \frac{\exp(\gamma\tau)}{\mathscr{E}_L},$$

$$A_a = \beta \frac{\exp(iq\xi)\mathscr{E}_a \exp(\gamma\tau)}{\mathscr{E}_L}, \qquad (12)$$

$$B = Q \exp(\gamma\tau).$$

Then, Eqns (4)–(6) take the form

$$\frac{\partial A_s}{\partial \xi} = ipB^*, \qquad (13)$$

$$\frac{\partial A_a}{\partial \xi} = iqA_a + ipB, \qquad (14)$$

$$\frac{\partial B}{\partial \tau} = i|\mathscr{E}_L|^2 A_s^* + i|\mathscr{E}_L|^2 A_a,$$

where $p/2 = r_s = r_a|\beta|^2$. Let us introduce a new variable

$$u(\tau) = \int_0^\tau |\mathscr{E}_L|^2 d\tau,$$

which is proportional to the pump pulse energy at the instant τ. In this case, the equation for $\partial B/\partial\tau$ is considerably simplified:

$$\frac{\partial B}{\partial u} = iA_s^* + iA_a. \qquad (15)$$

Let us differentiate Eqn (15) with respect to ξ and take into account Eqns (13) and (14). As a result, we obtain a new equation for polarisation, which does not contain the amplitudes of the Stokes field:

$$\frac{\partial^2 B}{\partial u \partial \xi} = -qA_a. \qquad (16)$$

Thus, the problem is reduced at the first stage to the solution of a system of two equations (14) and (16). Their solution can be obtained by the method of successive approximations in the parameter q, and has the form

$$A_a = -\frac{a}{u} \exp(iy) \sum_{k=0}^{\infty} j_k(y) \frac{(-ix)^{k+1}}{(k!)^2}, \qquad (17)$$

$$B = a\left\{1 + y\exp(iy) \sum_{k=0}^{\infty} [j_k(y) - ij_{k+1}(y)] \frac{(-ix)^{k+1}}{[(k+1)!]^2}\right\}. \qquad (18)$$

The amplitude A_s is determined from Eqn (15):

$$A_s = \frac{a}{u} \exp(-iy) \sum_{k=0}^{\infty} [(k+1)j_k(y) - yj_{k+1}(y) + iyj_k(y)] \frac{(ix)^{k+1}}{k!(k+1)!}, \qquad (19)$$

where $y = q\xi/2$; $j_k(y)$ is a spherical Bessel function of the first kind of the order k; $x = g\xi$; and $g = pu$. The solution (17)–(19) is valid for any x and y, i.e., for any g, q and ξ. The quantity $2y = q\xi = \Delta kz$ is the difference between the phases of interacting waves. The meaning of the quantity x will be explained below. Taking into account the transformation (12), we finally obtain

$$\mathscr{E}_a(\xi,\tau) = -a\frac{p\xi}{\beta}\mathscr{E}_L(0,\tau)\exp\left(-i\frac{q\xi}{2} - \gamma\tau\right) \times \sum_{k=0}^{\infty} j_k\left(\frac{q\xi}{2}\right) \frac{[-ip\xi u(\tau)]^k}{(k!)^2}, \qquad (20)$$

$$\mathscr{E}_s(\xi,\tau) = a\frac{p\xi}{\alpha}\mathscr{E}_L(0,\tau)\exp\left(-i\frac{q\xi}{2} - \gamma\tau\right) \sum_{k=0}^{\infty} \frac{[ip\xi u(\tau)]^k}{k!(k+1)!} \times \left[(k+1)j_k\left(\frac{q\xi}{2}\right) - \frac{q\xi}{2} j_{k+1}\left(\frac{q\xi}{2}\right) + i\frac{q\xi}{2} j_k\left(\frac{q\xi}{2}\right)\right], \qquad (21)$$

$$Q(\xi,\tau) = a\frac{q\xi}{2}\exp\left(-i\frac{q\xi}{2} - \gamma\tau\right) \times \sum_{k=0}^{\infty} \left[j_{k-1}\left(\frac{q\xi}{2}\right) - j_k\left(\frac{q\xi}{2}\right)\right] \frac{[-ip\xi u(\tau)]^k}{(k!)^2}. \qquad (22)$$

For further analysis, it should be interesting to consider two particular cases of the solutions (20)–(22): $q = 0$ and $|q|\xi \gg 1$. Because $j_k(y) \approx \sqrt{\pi}(y/2)^{k+1/2}[2\Gamma(k+3/2)]^{-1}$ for a small argument [$\Gamma(z)$ is the gamma function], we obtain for $q = 0$

$$\mathscr{E}_a(\xi,\tau) = -ia\frac{p}{\beta}\xi\mathscr{E}_L(0,\tau)\exp(-\gamma\tau), \qquad (23)$$

$$\mathscr{E}_s(\xi,\tau) = ia\frac{p}{\alpha}\xi\mathscr{E}_L(0,\tau)\exp(-\gamma\tau), \qquad (24)$$

$$Q(\xi,\tau) = a\exp(-\gamma\tau). \tag{25}$$

For $|q|\xi \gg 1$, we use the asymptotic representation $j_k(y) \approx \sin(y - k\pi/2)/y$ for $y \gg 1$. This gives

$$\mathscr{E}_a = a\frac{p}{\beta q}\mathscr{E}_L(0,\tau)\exp(-\gamma\tau)\Big\{J_0\big([4p\xi u(\tau)]^{1/2}\big) - \exp(-iq\xi)I_0\big([4p\xi u(\tau)]^{1/2}\big)\Big\}, \tag{26}$$

$$\mathscr{E}_s = a\frac{i}{\alpha}\mathscr{E}_L(0,\tau)\exp(-\gamma\tau)\left(\frac{p\xi}{u(\tau)}\right)^{1/2} I_1\big([4p\xi u(\tau)]^{1/2}\big), \tag{27}$$

$$Q = a\exp(-\gamma\tau)I_0\big([4p\xi u(\tau)]^{1/2}\big), \tag{28}$$

where $J_n(z)$ and $I_n(z)$ are the Bessel function and the modified Bessel function of the order n. One can see that in this case the anti-Stokes component can be neglected and we arrive at a solution analogous to the one obtained earlier by Carman et al. [25].

Solutions (20), (21) or (23), (24), (26), (27) allow us to find the intensity of the corresponding wave

$$I_f = \frac{c\eta|E_f|^2/2\pi}{\hbar\omega_f n_0 SL}\,S = \frac{\Omega}{2r_f l}|\mathscr{E}_f|^2, \tag{29}$$

which is equal to the number of photons with the frequency ω_f ($f = $ s, a) flying through the transverse cross section S of the sample per unit time (per molecule). The energy of a Stokes (anti-Stokes) pulse of duration t_L is

$$W_f = \int_0^{t_L} I_f(L,t)\mathrm{d}t = \frac{1}{2r_f l}\int_0^{\tau_L}|\mathscr{E}_f(l,\tau)|^2\mathrm{d}\tau \;(f = \mathrm{s},\mathrm{a}), \tag{30}$$

where $\tau_L = t_L/t_0$; $l = L/L_0$.

For a rectangular pulse of amplitude $E_L^{(0)}$ and duration t_L, formula (30) can be written in the explicit form

$$W_f = a^2 M_f^2 \quad (f = \mathrm{s},\mathrm{a}).$$

Here,

$$M_s^2 = \sum_{k,n=0}^{\infty}\{[(k+1)j_k(y) - yj_{k+1}(y)][(n+1)j_n(y) - yj_{n+1}(y)] + y^2 j_k(y)j_n(y)\}\frac{x^{k+n+1}}{[(k+1)!(n+1)!]^2} \times (k+n)!\gamma^*(k+n+1,\,2\gamma\tau_L), \tag{31}$$

$$M_a^2 = \sum_{k,n=0}^{\infty} j_k(y)j_n(y)\frac{x^{k+n+1}}{(k!\,n!)^2}(k+n)!\gamma^*(k+n+1,\,2\gamma\tau_L), \tag{32}$$

where $\gamma^*(m,z)$ is an incomplete gamma function [26]; $y = ql/2 = \Delta kL/2$; $x = p\mathscr{E}_L^{(0)}\tau_L l = GL$; $\gamma\tau_L = \Gamma t_L$; $\mathscr{E}_L^{(0)}$ is the dimensionless amplitude corresponding to $E_L^{(0)}$;

$$G = \frac{g}{v\Omega^{-1}} = \frac{4\pi^2 n_0|\mu_s|^2 P_L t_L}{c\eta^3\hbar}k_s \tag{33}$$

is the nonstationary gain; and P_L is the intensity of laser radiation.

We see that the Stokes and anti-Stokes components behave in different manners for different values of q and Δk. It follows from formulas (23), (24) and (29) that for a complete phase matching ($\Delta k = 0$), i.e., in the direction of the angle

$$\theta_0 = \left(\frac{4k_a k_L - 4k_L^2 - k_a^2 + k_s^2}{2k_a k_L}\right)^{1/2}$$

(Fig. 1), the intensities I_s and I_a are close to each other and do not increase with time even if $\Gamma = 0$. This situation is analogous to stationary SRS [1–4]. If, however, the mismatching of waves becomes significant ($|\Delta k|z \gg 1$), i.e., the angles θ between them are much larger than θ_0, the coupling between the anti-Stokes wave and the Stokes wave is lost completely, and the former is not generated while the second is amplified to the maximum extent.

If $t = (4pu\xi)^{1/2} \gg 1$ in Eqns (27) and (28), we can use the asymptotic representation $I_1(t) \sim (2\pi t)^{-1/2}\exp t$ and write

$$I_s \sim \xi^{1/2}\exp\left[2(4g\xi)^{1/2}\right] \sim z^{1/2}\exp\left[4(Gz)^{1/2}\right].$$

Thus, the quantity x appearing in the solutions (17)–(22) defines the increment of the Stokes emission intensity in the absence of the anti-Stokes component.

Because the parameter a in the formula $W_f = a^2 M_f^2$ is a random quantity, the energies $W_{s,a}$ of the Stokes and anti-Stokes pulses fluctuate, and their distribution functions are analogous to those obtained earlier in Ref. [18]:

$$P(W_f) = \frac{1}{\bar{W}_f}\exp\left(-\frac{W_f}{\bar{W}_f}\right), \tag{34}$$

where the mean pulse energy is given by

$$\bar{W}_f = \int_0^{\infty} W_f P(W_f)\mathrm{d}W_f = \kappa^2 M_f^2, \tag{35}$$

and their relative standard deviations can be written in the form

$$\delta_f = \left[\int_0^1 (W_f - \bar{W}_f)^2 P(W_f)\mathrm{d}W_f\right]^{1/2}\bar{W}_f^{-1} = 1 \;(f = \mathrm{s},\mathrm{a}). \tag{36}$$

Because the mean anti-Stokes pulse energy is close to zero in accordance with formulas (32), (35), (23) and (26) for small and large values of the parameter q, it can be expected that this energy will be maximum for a certain intermediate value q_{opt} (Fig. 2). Calculations based on formulas (32) and (35) show that in the absence of transverse relaxation, the energy $\bar{W}_a$ indeed assumes its largest value for $|y| \approx x/4$ ($|q_{opt}| \approx g/2$) or

$$|\Delta k_{opt}| \approx \frac{G}{2}. \tag{37}$$

The average Stokes pulse energy increases monotonically with $|q|$.

Thus, in the case of a weak depletion of pumping during nonstationary SRS, there exists an optimal difference between the wave vectors of the interacting waves, which

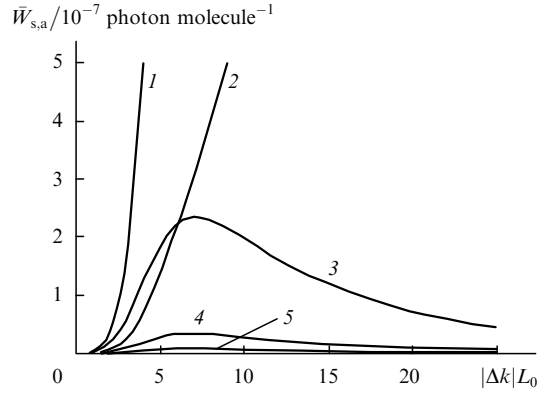

Figure 2. Dependences of the average energies of Stokes (*1*, *2*) and anti-Stokes (*3*–*5*) pulses on spatial mismatching in the case of a weak depletion of pumping for the transverse relaxation rate Γ_0 (*1*, *3*), 0.005Ω (*2*, *4*), 0.010Ω (*5*), $t_L = 200t_0$, $\mathscr{E}_L^{(0)} = 0.2$, $L = 2L_0$, and $\kappa = 10^{-6}\sqrt{2}$.

determines the direction θ of the most intense anti-Stokes generation (Fig. 1). Since $|\Delta k_{opt}|$ amounts to a few inverse centimetres and is therefore much smaller than k_f, it follows from Fig. 1 that angle θ is defined by the expression

$$\theta = \left(\theta_0^2 + \Delta\theta^2\right)^{1/2}, \tag{38}$$

where $\Delta\theta^2 = k_s|\Delta k_{opt}|/k_a k_L$. The angle $\Delta\theta$ is comparable with angle θ_0 and has a value equal to a few milliradian. Thus, under the experimental conditions described above, the quantity $|\Delta k_{opt}| \simeq 5.8\ \text{cm}^{-1}$ for $p_0 = 3$ atm, $t_L = 5 \times 10^{-9}$ s and $P_L = 5 \times 10^9\ \text{W cm}^{-2}$. This gives $\theta_0 = 3.0$ mrad and $\Delta\theta = 3.2$ mrad.

In the case under consideration here ($t_L \sim T_2$), transverse relaxation practically does not change the direction of the anti-Stokes radiation having the highest intensity, but lowers the SRS pulse energy at both frequencies.

3.2 Extended system

The intensity of the Stokes component increases with increasing the sample length. For $L > L_0$, this increase is significant and the intensity of Stokes radiation becomes comparable with that of laser radiation. Therefore, we must reject in this case the prescribed pump field approximation.

Taking into account the depletion of the exciting wave, we obtain from Eqns (3)–(5) the law of conservation of the number of photons:

$$N_L(\xi,\tau) + N_s(\xi,\tau) + N_a(\xi,\tau) = N_L(0,\tau), \tag{39}$$

where

$$N_f(\xi,\tau) = \frac{\eta^2|E_f|^2/2\pi}{\hbar\omega_f n_0} = \frac{|\mathscr{E}_f|^2}{2r_f} \quad (f = L, s, a)$$

is the number of photons with frequency ω_f per molecule. Thus, the increase in the intensity of the Stokes wave is limited by the pump intensity.

One can see from Eqn (5) that the product of the pump field and polarisation determines the intensity of anti-Stokes generation. In turn, it follows from Eqn (6) that the latter depends mainly on the product of the amplitudes of Stokes and laser waves ($I_a \ll I_s$). For $|\Delta k|z \ll 1$, the amplitude $\mathscr{E}_s$ is small (the growth of $\mathscr{E}_s$ is suppressed), while for $|\Delta k|z \gg 1$ the amplitude $\mathscr{E}_L$ is small (depletion of $\mathscr{E}_L$ pumping due to the growth of $\mathscr{E}_s$). The product $\mathscr{E}_s\mathscr{E}_L$ of amplitudes taking Eqn (39) into account will be maximum when they are comparable, i.e., for a certain intermediate value of $|\Delta k|$. Consequently, as in the case of weak pumping depletion, there exists an optimal spatial mismatching of the interacting waves in the present case, when the intensity of the anti-Stokes component is the highest. This is confirmed by a numerical analysis of Eqns (3)–(5) (Fig. 3).

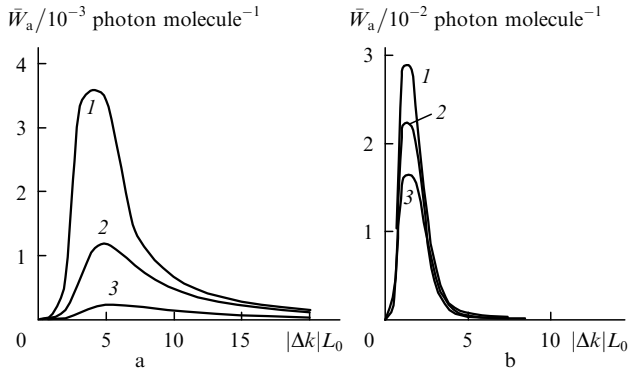

Figure 3. Dependences of the average energies of anti-Stokes pulses on spatial mismatching in the case of a strong depletion of pumping for a sample of length (a) $L = 5L_0$ and (b) $L = 10L_0$ for different transverse relaxation rates. The remaining parameters are the same as in Fig. 2.

The larger the value of L, the higher the increment of the Stokes radiation intensity and the pumping intensity decrement, i.e., the smaller the difference $|\Delta k|$ for which the product $\mathscr{E}_s\mathscr{E}_L$ assumes its highest value. Consequently, the difference $|\Delta k_{opt}|$ decreases with increasing L ($|\Delta k_{opt}| \sim L^{-1}$) and approaches zero for large values of L. Apart from $|\Delta k_{opt}|$, i.e., the angle of optimal anti-Stokes generation, the angular dimensions of the anti-Stokes ring (Fig. 3) also decrease upon an increase in the sample length.

In contrast to Sec. 3.1, the pump pulse duration has an insignificant effect on the difference $|\Delta k_{opt}|$ (and hence on the angle $\Delta\theta$) in the case of a strong depletion of pumping: these quantities increase only slightly upon an increase in t_L. Transverse relaxation also has a similar effect on these quantities in the case $t_L \sim T_2$ considered here. However, an increase in the transverse relaxation rate leads to a rapid decrease in the energy of anti-Stokes pulses, although the sensitivity $\bar{W}_a$ to the variation of this parameter decreases with increasing L.

4. Conclusions

In the regime of generation of SRS pulses having a duration comparable with the transverse relaxation time in the Raman transition, a suppression of the amplification of Stokes and anti-Stokes components is observed in the direction of a complete spatial matching of laser, Stokes and anti-Stokes waves defined by the angle θ_0. For an angle of emission much larger than θ_0, the connection between the anti-Stokes and Stokes waves is completely lost and the former wave is not generated, while the amplification of the latter is maximum. Consequently, there exists an optimal spatial mismatching of waves participating in the para-

metric process in which the anti-Stokes radiation intensity is the highest.

For relatively short samples ($L \leqslant L_0$) in which the attenuation of the laser radiation can be neglected, the angle θ of the most efficient anti-Stokes generation is about twice as large as the angle θ_0 of complete phase matching. The optimal angle θ for such samples does not depend on their length and is directly proportional to the square root of the exciting pulse energy. The direction of the most efficient anti-Stokes radiation is practically stable while the energy of the corresponding pulses experiences large-scale (about 100 %) fluctuations.

For extended systems ($L > L_0$) with a significant depletion of the pump pulses, the optimal angle of anti-Stokes generation decreases with increasing L approximately in inverse proportion to $L^{1/2}$ and approaches the angle of complete phase matching for large values of L. The pump pulse duration in long media does not affect the angle θ significantly. As in short samples, the spread in the optimal angles of anti-Stokes generation is quite small. The energy dispersion of Stokes and anti-Stokes pulses decreases with increasing L, and amounts to 10 % – 15 % in the direction of angle θ for $L \simeq 10L_0$ in the absence of phase relaxation.

Under conditions of conservation of phase memory of molecules, the angular width of the anti-Stokes ring (at half-height) is close to the difference $\theta - \theta_0$. Enhancement of the collision dephasing considerably broadens the anti-Stokes rings and slightly increases their mean angles. In this case also, no significant increase is observed in the spread of energies of Stokes and anti-Stokes pulses.

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