STIMULATED RAMAN SCATTERING IN A FIELD OF ULTRASHORT LIGHT PULSES

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We discuss the results of the latest experiment on stimulated Raman scattering (SRS) in a field of ultrashort laser pulses and under conditions of anomalous broadening of the spectral line as a result of the nonlinearity of the refractive index of the scattering medium. We show that to interpret the experimental data it is necessary to analyze the SRS under rather broad assumptions concerning the type of modulation of the pump (amplitude (AM) and phase (PM) modulated pumping, pulsed pumping with phase modulation within the pulse) and the character of the "priming" Stokes signal (noise, narrow-band signal, a superposition of the two, etc.). A nonstationary theory of SRS is developed, in which account is taken of the foregoing factors. Besides a calculation of the intensity of the Stokes components, carried out not only for AM pumping but also for PM pumping and noise and narrow-band signals, appreciable attention is paid to the indicatrix and to the spectrum of the nonstationary SRS. A new mechanism of anomalous broadening of the spectrum of Stokes radiation, connected with the finite time of establishment of the molecular oscillations is considered.

1. INTRODUCTION

THE presently most complete stationary theory of stimulated Raman scattering (SRS), in which the interaction between harmonic pump waves and Stokes and anti-Stokes components is considered (see[1-3]), is valid only for a limited number of experimental situations. This theory is in satisfactory agreement with experiments on linear amplification of RS components in gases in the field of a single-mode nanosecond pump[4] and with the results of measurements of the distribution of the energy over the Stokes components in the essentially nonlinear regime for SRS in liquids, in the case of pumping by pulses of duration 10^{-9}–10^{-10} sec[5] [1)].

The simplest example of a nonstationary problem in which time modulation of the Stokes component is important ("nonstationarity with respect to the signal") is the problem of the SRS line width in a given pump field; closely related to it in scope is the problem of the passage of modulated signals through an amplifier.

An important class of nonstationary problems where the principal role is played by "the nonstationarity with respect to the signal" is connected with the investigation of the generation of giant ultrashort Stokes pulses—an effect first observed by Maier, Kaiser, and Giordmaine[6] in the investigation of SRS at 180°. The theory developed in[8-9] shows that for a satisfactory explanation of the phenomena it is necessary to consider the nonstationary effects in the nonlinear amplification regime.

The appearance of mode-locking lasers and the observation of SRS in a nanosecond pumping field and under conditions of anomalous broadening of the spectral lines in a scattering medium has stimulated the study of processes connected with "nonstationarity with respect to the pump," i.e., with effects due to the amplitude modulation (AM) and phase modulation (PM) of the pumping[2)].

For sufficiently rapidly modulated pumping, an important role is played by the transient processes due to the fact that the characteristic modulation times become comparable with the time of transverse relaxation T_2 (in typical cases, $T_2 \sim 10^{-11}$ sec for liquids and $T_2 \sim 10^{-8}$–10^{-9} sec for gases), and also by processes connected with the difference between the group velocities of the pump wave and the scattered components[14-17].

It should be noted, however, that the presently developed theory of SRS in a modulated pump field is insufficient for the interpretation of the already accumulated various experimental data. Thus, in most published papers the data on the nonstationary SRS and on the nonstationary scattering of other types are analyzed on the basis essentially of the results of Kroll's theory[16], which was developed for stimulated Mandel'shtam-Brillouin scattering (see, for example,[15,18]). The picture obtained in this case is far from complete; it usually deals only with the intensity of the harmonic Stokes signal, amplified in a pulsed pump field under group-synchronism conditions.

At the same time, in order to interpret correctly the new experiments (see, for example,[17-22]), it is necessary to consider the features of SRS under rather broad assumptions concerning the type of pumping (Am and PM pumping; in some cases the most satisfactory model is noise pumping) and the "signal" (a harmonic signal for the interpretation of amplification experiments, and a random-modulated signal for the interpretation of SRS experiments). In the most interesting cases the nonstationarity "with respect to the signal" and "with

[1)]The "amplification regime" is defined as one in which the intrinsic fluctuations of the scattering medium can be neglected, and the signal at the Stokes frequency is fed from an external source; the "scattering regime" is defined as one in which the waves of the scattered components result from the intrinsic fluctuations in the medium.

[2)]We note that certain effects due to pumping modulation were discussed already in 1964–1966 in connection with the investigations of SRS in a multimode pumping field [10-13]. Although effects agreeing with the predictions of [11-13] were observed at approximately the same time in SRS, they were not investigated in detail; for a long time the researchers paid attention to the much stronger self-action effects.

respect to the pump" must be considered simultaneously.

We developed below a theory in which the foregoing factors are taken into account.[3] For simplicity we confine ourselves to examination of nonresonant SRS of first order by fully-symmetrical oscillations for not too strong fields. Such an analysis makes it possible to obtain useful information on many features of the nonstationary SRS in liquids, crystals, and gases. A number of conclusions of such a theory turned out to be applicable to nonstationary SMBS and to nonstationary stimulated Rayleigh-wing scattering (SRWS).

2. EQUATIONS OF NONSTATIONARY SRS

A. Fundamental Equations

Let us assume for simplicity that nonresonant SRS takes place for one pair of alternatively-forbidden quantum levels of the scattering medium. We consider the interaction between quasi-monochromatic pump waves E_p and the Stokes component E_S in the form

$$\mathbf{E}_p = \mathbf{a}A_p(t, z)\exp[(i\omega_p t - k_p z)] + \text{c.c.}, \tag{1a}$$

$$\mathbf{E}_S = \mathbf{b}A_S(t, z)\exp[i(\omega_S t \mp k_S z)] + \text{c.c.} \tag{1b}$$

(the $\mp$ signs in (1b) correspond to the most interesting cases when the pump waves and the Stokes radiation have the same or opposite directions). The average frequencies of the pump and of the Stokes component and the transition frequency ω_{21} are connected by the relation

$$\omega_p - \omega_S = \omega_{21} + \Delta, \quad |\Delta| / \omega_{21} \ll 1.$$

It is assumed further that the relative widths of the pump spectra and of the Stokes component are small compared with ω_{21}, i.e., $\Delta\omega_{p,S} \ll \omega_{21}$[4]. Then the behavior of the isotropic medium (we shall deal henceforth mainly with liquids and gases) in the SRS process can be described by the equations for the slow amplitude of the nondiagonal element of the density matrix σ, and the difference between the level populations n[23]:

$$\frac{\partial\sigma}{\partial t} + \left(\frac{1}{T_2} + i\Delta\right)\sigma = i\gamma_\sigma A_p^* A_S n, \tag{2}$$

$$\frac{\partial n}{\partial t} + \frac{1}{T_1}(n - n_0) = 4\,\mathrm{Im}(\gamma_\sigma \sigma^* A_p^* A_S), \tag{3}$$

where n_0 is the equilibrium value of n, T_1 is the lifetime of the particles at the level 2, T_2 is the transverse relaxation time, $\gamma_\sigma = r^*/H^2$, and r is defined as

$$r = \sum_m \left(\frac{p_{a1m}p_{bm2}}{\omega_{m2} - \omega_S} + \frac{p_{am2}p_{b1m}}{\omega_{m2} + \omega_p}\right).$$

Here p_{anm} and p_{bmn} are the projections of the matrix elements of the dipole-moment operator on the vectors a and b. It is necessary to add to Eqs. (2) and (3) the equations for the slowly-varying field amplitudes. In the first approximation of dispersion theory, which is valid for pulses to 10^{-14} sec (see[14]), they are first-order equations:

$$\frac{1}{u_p}\frac{\partial A_p}{\partial t} + \frac{\partial A_p}{\partial z} = -i\gamma_p \sigma^* A_S - \delta_p A_p, \tag{4a}$$

$$\frac{1}{u_c}\frac{\partial A_S}{\partial t} \pm \frac{\partial A_S}{\partial z} = -i\gamma_S \sigma A_p - \delta_S A_S; \tag{4b}$$

here

$$\gamma_p = \frac{2\pi N_0 \omega_p r^*}{k_p c^2 \hbar}, \quad \gamma_S = \frac{2\pi N_0 \omega_S r}{k_S c^2 \hbar},$$

N_0 is the density of the scattering particles, $\delta_{p,S}$ are the damping decrements of the pump and of the Stokes wave. The plus sign in (4b) corresponds to scattering in the pump direction, and the minus sign to scattering at 180°; $u = \partial\omega/\partial k$ is the group velocity.

B. Quasistatic Approximation

For sufficiently slowly modulated waves and relatively short interaction lengths, Eqs. (2)–(4) can be greatly simplified. Let the characteristic times of variation of the amplitudes (τ_a) and of the phases (τ_{ph}) of the pump and of the Stokes component satisfy the condition

$$\tau_{ap}, \tau_{php}, \tau_{aS}, \tau_{phS} \gg T_2. \tag{5}$$

Then we can neglect in (2) the derivative $\partial\sigma/\partial t$ and obtain in the usual manner a system of nonstationary rate equations for $J_{p,S} = |A_{p,S}|^2$ and the population difference n:

$$\frac{1}{u_p}\frac{\partial J_p}{\partial t} + \frac{\partial J_p}{\partial z} = -\frac{2\gamma_\sigma^* \gamma_p T_2}{1+\Delta^2 T_2^2} J_p J_S n - 2\delta_p J_p, \tag{6}$$

$$\frac{1}{u_S}\frac{\partial J_S}{\partial t} \pm \frac{\partial J_S}{\partial z} = \frac{2\gamma_\sigma \gamma_S T_2}{1+\Delta^2 T_2^2} J_p J_S n - 2\delta_S J_S, \tag{7}$$

$$\frac{\partial n}{\partial t} + \frac{1}{T_1}(n - n_0) = -\frac{4\gamma_\sigma^2 T_2}{1+\Delta^2 T_2^2} J_p J_S n. \tag{8}$$

If $\tau_{ap}, \tau_{aS} \gg T_1$, then the derivative $\partial n/\partial t$ in (8) can be neglected, and the problem reduces to a solution of the two equations (6) and (7). For the essentially nonstationary cases, when $\tau_{ap}, \tau_{aS} \ll T_1$, we can disregard the term $(n - n_0)/T_1$ in (8). For many cases of practical interest, the motion of the populations can be neglected in general. For example, for pump pulses of $\sim 10^{-11}$ sec duration, the populations remain relatively unchanged up to fields $E_p \approx 10^7$ V/cm. This can be easily verified by starting from Eq. (8) and recognizing that for typical media $\gamma_\sigma^2 \sim 10^3 - 10^4$ cgs esu, $T_2 = 10^{-11}$ sec, and the coefficients of conversion of picosecond pumping into Stokes radiation does not exceed 0.2 as a rule[19].

Neglecting the motion of the populations ($n = n_0 \approx 1$) and assuming (5) to be satisfied, the SRS is described by the system (6) and (7). In this case the influence of the time modulation of the pumping and of the Stokes component on the development of the process is determined entirely by the ratio of the group velocities and by the interaction length. For waves moving together at $u_p = u_S$, introducing new variables

$$\eta = t - z/u_S, \quad z = z \tag{9}$$

we get from (6)–(7) the characteristic system of ordin-

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[3]Preliminary results were reported at the International Conference on Nonlinear Optics (Belfast, September, 1969).

[4]The latter condition is certainly satisfied for the overwhelming majority of nonstationary problems connected with SRS in liquids and gases and for many cases of SRS in crystals. It should be noted, however, some investigations (see, for example, [22]) of CS_2 revealed broadening of the pump spectrum $\Delta\omega_p \sim \omega_{21}$. The mathematical formation of the problem of nonstationary SRS differs in this case from that given in the present section.

ary differential equations of the stationary theory.

The requirement $u_p = u_S$ is obviously the condition for the applicability of the stationary equations for an unbounded medium; on the other hand, if the SRS is considered over a finite length l, then the quasistatic approximation is convenient for the description of the forward scattering (we assume $\tau_{ap} \approx \tau_{aS} \approx \tau_a$) when

$$\tau_a \gg \tau^{(-)} = l|u_p^{-1} - u_S^{-1}| \tag{10a}$$

and for backward scattering when

$$\tau_a \gg \tau^{(+)} = l|u_p^{-1} + u_S^{-1}|, \tag{10b}$$

i.e., so long as the effects of group delay of the interacting waves do not come into play over the length l.

3. NONSTATIONARY STOKES SCATTERING UNDER CONDITIONS OF GROUP SYNCHRONISM

A. Energy Characteristics of Stokes Radiation

The bare radiation at the Stokes frequency in the "scattering regime" is the result of spontaneous Raman scattering. In order to take into account this fact, we introduce in (2) the random force N(z, t) and assume zero initial and boundary conditions for the Stokes waves. Then, in the given-pump-field approximation, the SRS equations in the coordinate system (9) take the form

$$\partial\sigma/\partial\eta + \beta\sigma = i\gamma_\sigma A_p^*(\eta) A_S(\eta, z) + N(\eta, z), \tag{11}$$

$$\partial A_S/\partial z = -i\gamma_S A_p(\eta)\sigma(\eta, z), \tag{12}$$

where $\beta = 1/T_2 + i\Delta$.

The solution of the system (11)—(12) under zero boundary and initial conditions, $A_S(\eta = 0, z = 0) = 0$, takes the form[5)]

$$A_S(\eta, z) = -i\gamma_S A_p(\eta)\int_0^\eta dt e^{-\beta t}\int_0^z N(\eta - t, z - \xi) I_0(\psi_\eta(\xi, t))\,d\xi, \tag{13}$$

where $I_0(x)$ is the modified Bessel function

$$\psi_\eta(\xi, t) = \left[2\frac{\xi}{T_2}\int_{\eta-t}^{\eta}\Gamma(y)\,dy\right]^{1/2}, \quad \Gamma(y) = 2\gamma_S\gamma_\sigma T_2 J_p(y).$$

Introducing the natural assumption that

$$\overline{N(t, z)N^*(t', z')} = g\delta(t - t')\delta(z - z'), \tag{14}$$

we obtain an expression for the average intensity of the Stokes radiation:

$$\bar{J}_S = g\gamma_S^2 J_p\int_0^\eta dt\,\exp\left(-\frac{2t}{T_2}\right)\int_0^z d\xi\, I_0^2(\psi_\eta(\xi, t)). \tag{15}$$

Before we proceed to consider different particular cases, let us note one important conclusion that follows directly from (15). According to (15) the intensity of the Stokes component in the scattering regime is not sensitive to phase modulation of the pumping, regardless of the ratio of the transverse relaxation time T_2 to the modulation period (it is of interest that in the "amplification regime" the situation is different, see Sec. 4). At the same time, according to (13), phase modulation of the pump greatly influences the spectrum of the Stokes wave. In particular, if the structure of the pump spectrum is determined mainly by its phase modulation during the propagation process, then exactly the same structure should be observed also in the spectrum of the Stokes component if the conversion coefficients are not too large. We now turn to a more detailed analysis of relation (15).

Let us consider first the energy characteristics of the Stokes radiation. When $\Gamma \to 0$ formulas (13) and (15) describe the spontaneous Raman scattering (SpRS), the intensity of which, according to (15), is equal to

$$\overline{J_S^{sp}} = {}^1\!/_2 g\gamma_S^2 T_2 J_p(\eta) z(1 - e^{-2\eta/T_2}). \tag{16}$$

It follows therefore directly that the intensity of the spontaneous Raman scattering in the field of short pulses of duration $\tau_p \lesssim T_2$ is greatly reduced compared with the intensity of the spontaneous Raman scattering in a field of pulses with $\tau_p \gg T_2$. For the latter case we obtain from (16) the usual formula for the intensity of spontaneous Raman scattering:

$$\overline{J_S^{sp}} = {}^1\!/_2 g\gamma_S^2 T_2 J_p z. \tag{17}$$

Easily-interpreted formulas are obtained for nonstationary SRS under conditions of large gain, when

$$\psi_\eta(\xi, t) \gg 1. \tag{18}$$

In this case the Bessel function in (13) and (15) can be replaced by its asymptotic expression.

For pump pulses of rectangular form, i.e.,

$$|A_p(\eta)| = \begin{cases} |A_{p0}| & \text{if } 0 \leqslant \eta \leqslant \tau_p \\ 0 & \text{if } \eta < 0 \text{ and } \eta > \tau_p \end{cases}, \tag{19}$$

we can obtain convenient formulas describing the amplification of the Stokes radiation.

In the essentially nonstationary regime

$$\tau_p \ll T_2. \tag{20}$$

the following formula holds for the average intensity of the Stokes wave:

$$\bar{J}_S = \gamma_S g[8\pi\gamma_\sigma\psi_0(z, \eta)]^{-1}\exp\{2\psi_0(z, \eta)\},$$
$$\psi_0(z, \eta) = (2\Gamma_0 z\eta/T_2)^{1/2}, \quad \Gamma_0 = 2\gamma_S\gamma_\sigma T_2 J_{p0}. \tag{21}$$

We see that in this case the gain is much lower than in the quasistatic case, and depends on the time (Fig. 1); this causes the shape of Stokes pulses to differ strongly from the pump pulse (see Fig. 2).

Integration of (21) with respect to time yields an expression for the energy of the Stokes pulse

$$W_S \approx \frac{\gamma_S g T_2}{16\pi\gamma_\sigma\Gamma_0 z}\exp\{2\psi_0(z, \tau_p)\}. \tag{22}$$

In the more general case, when the condition (20) may also not be satisfied, calculation of the integral in (15) yields for the pulse (19)

$$\bar{J}_S \approx \frac{\gamma_S g e^{\Gamma_0 z}}{8\gamma_\sigma\sqrt{\pi\alpha}}\left[1 + \Phi\left(\sqrt{\frac{2\eta}{T_2}} - \sqrt{\Gamma_0 z}\right)\right], \tag{23}$$

where

$$\alpha = 2\eta/T_2 \text{ if } \Gamma_0 z \geqslant 2\eta/T_2, \quad \alpha = \Gamma_0 z \text{ if } \Gamma_0 z \leqslant 2\eta/T_2.$$

Formula (23) describes the amplification of Stokes

5) Strictly speaking, the solution (13) should consist of two terms [24]. It is easy to show, however, that the second term [24] reflects the specific effects of "turning on the interaction," which are missing under the experimental conditions. We therefore did not take into account this term in this solution. A solution of the type (13) is discussed also in [26].

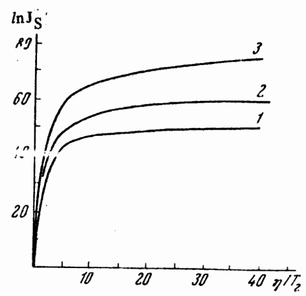

FIG. 1

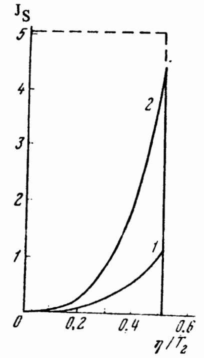

FIG. 2

FIG. 1. Time dependence of the intensity of the first Stokes component in different cross sections of a nonlinear medium: $1-\Gamma_0 z = 50$, $2-\Gamma_0 z = 60$, $3-\Gamma_0 z = 75$.

FIG. 2. Shape of Stokes SRS pulse excited by a rectangular pump pulse in the essentially nonstationary regime, $\Gamma_0 z \gg 2\tau_p/T_2$. Dashed—pump pulse; the solid curves correspond to Stokes pulses in different sections of the medium: $1-\Gamma_0 z = 10$, $2-\Gamma_0 z = 15$. The pump and Stokes-radiation intensities are plotted in different scales. The measurement units are arbitrary.

radiation for arbitrary ratios of τ_p and T_2. When

$$\eta / T_2 \gg 1, \ \sqrt{2\eta / T_2} - \sqrt{\Gamma_0 z} \gg 1 \tag{24}$$

we get from (23)

$$\bar{J}_S = \frac{\gamma_S g}{4\gamma_\sigma \sqrt{\pi \Gamma_0 z}} e^{\Gamma_0 z}. \tag{25}$$

Formula (25) describes the SRS intensity under conditions when the problem is quasistatic "with respect to the pump." It must be emphasized at the same time that the growth of the intensity with increasing coordinates is slower than $\exp(\Gamma_0 z)$, since the gain is accompanied[6)] by a narrowing of the line. From (25) it is easy to obtain also an expression for the width of the spectral line in a field of "almost harmonic" pumping; we note that according to (24) the condition for quasistationary behavior with respect to the pump is of the form $\tau_p/T_2 \gg \Gamma_0 z$, and not simply $\tau_p/T_2 \gg 1$.

We note finally that a formula such as (25) can be written also for a pump pulse of more complicated form, provided its duration is $\tau_p \gg T_2, \Gamma z T_2$. In this case it is necessary to substitute in (25) $\Gamma(\eta)$ in place of Γ_0, where the function $\Gamma(\eta)$ describes the pump envelope. The quasistatic amplification of the Stokes wave in a field of a dome-like pump pulse is accompanied by a narrowing of the Stokes pulse in accordance with the law

$$\tau_S = \tau_p / \sqrt{\Gamma_0 z} \tag{26}$$

a circumstance noted earlier as applied to parametric amplification[7]. For an exponential pulse pump the duration of the Stokes pulse τ_S varies like

$$\tau_S \approx \tau_p / \Gamma_0 z.$$

B. Spectra of Stokes Radiation

Using (13), we can calculate also the spectrum of nonstationary scattering. Let us consider first the spectrum of spontaneous scattering. The calculations are particularly simple if the pump can be regarded as a stationary random process. In this case the scattered field is also stationary. Specifying the correlation function of the complex pump envelope in the form[7)]

$$B_p(\tau) = \langle A_p(\eta) A_p^*(\eta + \tau) \rangle = A_{p0}^2 \exp(-\tau / \tau_0), \tag{27}$$

we obtain for the correlation function of the scattered field, taking (14) into account (to change over to spontaneous scattering it is necessary to let Γ in (13) go to zero)

$$B_S(\tau) = \frac{1}{2} g \gamma_S^2 A_{p0}^2 T_2 \exp\left[-\left(\frac{1}{T_2} + \frac{1}{\tau_0}\right)\tau\right]. \tag{28}$$

It is easily seen that the width of the spontaneous-scattering spectrum is

$$\Delta\omega_S^{sp} \approx (T_2 + \tau_0) / T_2 \tau_0. \tag{28a}$$

When $\tau_0 \gg T_2$ we have $\Delta\omega_S^{sp} \approx 1/T_2$, just as in a monochromatic pumping field.

The width and the shape of the spectrum of the Stokes component in the stimulated scattering regime are determined not only by the pump spectrum but also by the form of its amplitude modulation. Let us stop to discuss this circumstance, which is of greatest interest for experiment, in greater detail. The correlation function of the Stokes radiation, with allowance for (14), takes the form

$$B_S(\eta, \tau) = \gamma_S^2 g \Big\langle A_p(\eta) A_p^*(\eta + \tau) \cdot \times \int_0^\eta dt \exp\left(-\frac{2t + \tau}{T_2}\right) \int_0^t I_0(\psi_\eta(\xi, t)) I_0(\psi_{\eta+\tau}(\xi, t + \tau)) d\xi \Big\rangle \tag{29}$$

For the subsequent analysis we shall use the condition (18), which enables us to replace the Bessel functions by exponentials.

In the general case, the correlation function $B_S(\eta, \tau)$ depends on the time and corresponds thus to the correlation function of a nonstationary process. For estimates of the width of the spectrum, we shall use a function corresponding to time averaging over an interval exceeding the pump modulation period:

$$B_S(\tau) = \int_0^\infty B_S(\eta, \tau) d\eta. \tag{30}$$

Such an averaging is actually always carried out in experiments on nonstationary scattering.

Let us consider the following cases:

1. The pump radiation has the constant intensity and has a correlation function (27). Then calculation by means of formulas (29) and (30) yields

$$B_S(\tau) \sim \exp\{-\Delta\omega_S^{sp}\tau + [\Gamma_0 z(\Gamma_0 z + 2\tau / T_2)]^{1/2}\}.$$

Finding from this the effective correlation time τ_{eff} and assuming that $\Delta\omega_S \approx 1/\tau_{eff}$, we obtain

$$\Delta\omega_S \approx \frac{\Delta\omega_c}{1 - 2\Gamma_0 z}[m - (m^2 + 2\Gamma_0 z - 1)^{1/2}], \tag{31}$$
$$m = 1 - \Gamma_0 z / \tau_0 \Delta\omega_S^{sp}.$$

Formula (31) shows that at small gains, i.e., as $\Gamma_0 z \to 0$, the width of the spectrum $\Delta\omega_S \to \Delta\omega_S^{sp}$ (28a). With

[6)]A similar relation was obtained for SMBS by Tang [25] by means of a theory that uses equations for the individual spectral components.

[7)]Formula (27) corresponds, for example, to oscillation with a diffusing phase.

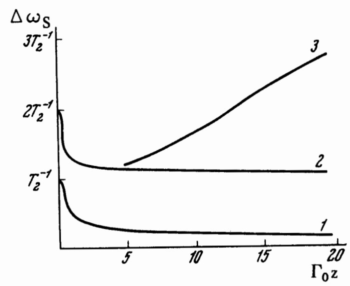

FIG. 3. Width of spectral line of SRS Stokes component as a function of the reduced distance traversed in the medium in the pulsed pumping field. The parameter of the curves is the ratio of the duration of the rectangular pulse pump τ_p to the time of transverse relaxation T_2: $1-\tau_p = \infty$, $\tau_0 = \infty$, $2-\tau_p = \infty$, $\tau_0 = T_2$; $3-\tau_p = T_2$, $\tau_0 = \infty$.

increasing gain, the spectrum becomes narrow, and in the limit as $\Gamma_0 z \to \infty$ its width becomes equal to the width of the pump spectrum, i.e.,

$$\Delta\omega_S \to \Delta\omega_p = 1/\tau_0. \tag{31a}$$

On the other hand if the pump radiation is monochromatic ($\tau_0 \to \infty$, $m \to 1$), then the narrowing of the Stokes spectrum is described by the formula

$$\Delta\omega_S \approx 1/T_2(1+\sqrt{2\Gamma_0 z}). \tag{31b}$$

2. The pump is a rectangular pulse with duration τ_p, having no phase modulation ($\tau_0 \to \infty$). Then from (29) and (30) we can obtain

$$B_S(\tau) \sim \exp\left\{-\frac{\tau}{T_2}+\left(2\Gamma_0 z\frac{\tau_p-\tau}{T_2}\right)^{1/2}\right\} \quad \text{for } \Gamma_0 z \geqslant \frac{2\tau_p}{T_2}.$$

The corresponding width of the Stokes spectrum is

$$\Delta\omega_S \approx T_2^{-1}[(d^2+2\Gamma_0 z)^{1/2}-1-d]^{-1},\ d=\Gamma_0 z[1-(2\tau_p/\Gamma_0 z T_2)^{1/2}]. \tag{32}$$

It is easy to see that this formula describes the broadening of the Stokes-radiation spectrum in the nonstationary regime. When $\Gamma_0 z \gg 2\tau_p/T_2$ (strong nonstationary behavior), we get from (32)

$$\Delta\omega_S \approx (\Gamma_0 z/2T_2\tau_p)^{1/2}. \tag{32a}$$

Thus, at not too large gains ($\Gamma_0 z < 2\tau_p/T_2$), the spectrum of the Stokes radiation in a field of a rectangular pumping pulse narrows down in accordance with (31b) and reaches a minimum value at the point $\Gamma_0 z = 2\tau_p/T_2$:

$$\Delta\omega_{S\,min} = 1/(2\sqrt{T_2\tau_p}-T_2).$$

Further increase of $\Gamma_0 z$ leads to the spectrum broadening described by formulas (32) (see Fig. 3). At large $\Gamma_0 z$, the width of the Stokes-radiation spectrum can greatly exceed the width of the pump spectrum $\Delta\omega_p \approx \tau_p^{-1}$. It should be noted that the "anomalous" broadening of the Stokes spectrum in the essentially nonstationary regime, described by formula (32a), is connected with rapid variations of the real amplitude and consequently can be observed by methods of intensity interferometry.

For pump pulses of more complicated form, it is impossible to obtain convenient analytic expressions describing the width of the Stokes spectrum. Numerical calculations, on the other hand, show that if the rear front of the pump pulse decreases sharply practically to zero, then the Stokes spectrum broadens in comparison with $\Delta\omega_p$ if $\Gamma_0 z > 2\tau_p/T_2$, but the rate of expansion is lower than is given by formulas (32).

4. AMPLIFICATION OF STOKES SIGNAL IN A RAMAN AMPLIFIER

We now turn to investigate the singularities of the nonstationary "amplification regime." Assume that a Stokes signal with amplitude $A_{S0}(\eta)$ is applied to the input of the Raman cell. It is now necessary to solve Eqs. (11) and (12) with boundary conditions

$$A_S|_{z=0} = A_{S0}(\eta), \quad A_c|_{\eta=0} = 0; \tag{33}$$

The random force $N(\eta, z)$, by definition, is insignificant in this case. Then, using Riemann's method, we obtain

$$A_S(\eta, z) = A_{S0}(\eta) + 2z\gamma_S\gamma_\sigma A_p(\eta)\cdot \times \int_0^\eta \frac{dt A_p^*(\eta-t)A_{S0}(\eta-t)e^{-\beta t}}{\psi_\eta(z,t)} I_1(\psi_\eta(z,t)). \tag{34}$$

The solution (34) differs in form significantly from (13), which corresponds to the "scattering regime." It should be noted, in particular, that the integrand now contains the complex pulse amplitude raised to the first power; therefore, unlike the results of Sec. 3, we should expect phase modulation of the pump to affect the intensity of the Stokes component.

Let us stop to discuss this in greater detail. In order to separate the effect in the "purest form," let us consider amplification of a harmonic signal in a pump field constituting an oscillation with constant amplitude and diffusing phase[8]. We assume that the pump radiation has a correlation function (27), i.e., the pump spectrum has a Lorentz contour with half-width $\Delta\omega_p = \tau_0^{-1}$. From (34) we obtain for the average intensity of the first Stokes component (for simplicity, we neglect the detuning Δ)

$$\bar{J}_S(\eta, z) = 2(\gamma_S\gamma_\sigma)^2 T_2 z J_p(\eta) \int_0^\eta dt \int_0^\eta dt' B_p(t-t')e^{-\beta(t+t')}F(t,t'), \tag{35}$$

where

$$F(t, t') = [\psi_\eta(z, t)\psi_\eta(z, t')]^{-1} I_1(\psi_\eta(z, t)) I_1(\psi_\eta(z, t')). \tag{36}$$

Let us estimate the value of the double integral in (35) under conditions when the phase diffusion coefficient is sufficiently large:

$$\tau_0 \ll T_2, \quad \tau_0 < T_2/\Gamma_0 z. \tag{37}$$

It is easy to verify that under the foregoing assumptions the phase modulation of the pump does not influence the intensity growth coefficient, and only decreases the preexponential factor in a ratio $\Gamma_0 z T_2/\tau_0$. This circumstance must be taken into account in the interpretation of experiments in which the SRS is excited by broadband signals obtained from lasers operating in the regime of partial mode synchronization.

Let us consider now the amplification of a random Stokes signal in a randomly modulated pump field. Then, using (34), we can write for the average intensity of the Stokes radiation

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[8]For AM pumping (particularly pulsed pumping) the results of calculations by means of formula (34) are similar to the result of Sec. 3. If the pump pulse duration is $\tau_p < T_2$, then it follows from (34) that $J_S = J_{S0} I_0^2(\sqrt{2\Gamma_0 z\eta/T_2})$.

$$J_S(\eta, z) = 2(\gamma_S\gamma_\sigma)^2 T_2 z J_p(\eta) \int_0^\eta dt \int_0^\eta dt' \cdot B_{p0}(t-t') B_{S0}(t-t') e^{-\beta(t+t')} F(t,t'),$$
$$B_{S0}(t-t') = \overline{A_{S0}(\eta - t) A_{S0}^*(\eta - t')}. \quad (38)$$

From this we can readily see that if the Stokes signal has a correlation time τ_S much shorter than the correlation time of the pump radiation τ_0 ($\tau_S \ll \tau_0$), i.e., the signal spectrum width $\Delta\omega_S \gg \Delta\omega_p$, then random PM of the pumping plays practically no role. Indeed, in this case $B_{S_0}(t-t') \approx A_{S_0}^2 \delta(t-t')$, and only the average pump intensity $J_p(\eta)$ is involved in the integrand. Further calculations lead to formulas for $\overline{J}_S$, analogous to those of Sec. 3. For example, if the quasistatic amplification conditions (24) is satisfied we obtain (compare with (25))

$$\overline{J}_S = \frac{J_{S0}}{4\sqrt{\pi\Gamma_0 z}} e^{\Gamma_0 z}. \quad (39)$$

In concluding this section, let us consider the amplification of a Stokes signal in the field of a pumping pulse whose duration satisfies the condition (20), and whose frequency increases linearly with time (such a model is applicable to pulsed solid-state lasers),

$$A_p = A_{p0} \exp(i\eta^2 / \tau_{ph}^2). \quad (40)$$

If we assume that $\tau_p/\tau_{ph} \gg 1$, then the integral in (34) can be estimated by the stationary-phase method. If

$$\eta / \tau_{ph} \gg \psi_0(z, \eta) \quad (41)$$

then an estimate yields

$$A_S \approx A_{S0} \sqrt{\frac{\pi}{4} \frac{\Gamma_0 z \tau_{ph}^2}{T_2 \eta}} I_1(\psi_0(z,\eta)) \exp\left(\frac{i\eta^2}{\tau_{ph}^2}\right). \quad (42)$$

From a comparison of (35) with (42) we see that the presence of a frequency shift within the limits of the pump pulse reduces the amplitude gain by an approximate factor $\sqrt{\Gamma_0 z \tau_{ph}^2 / \tau_p T_2}$.

The Stokes pulse, according to (42), is frequency modulated in accordance with the same law as the pump pulse.

5. EFFECT OF GROUP DELAY

A. SRS Indicatrix

The finite relaxation time of the molecular oscillations, as shown in the preceding sections, decreases the gain of the Stokes components, and consequently also the energy of the Stokes waves. This decrease however, as can be readily seen, does not depend on the observation direction. Therefore the decisive role in the determination of the form of the scattering indicatrix is played by effects of group delay.

In order to reveal their role most clearly, we neglect first the effects of molecular relaxation. Then the calculation can be carried out on the basis of Eq. (7), assuming the pump field to be specified and propagating with velocity u_p in the z direction. The linear loss δ_S will be neglected. We write the solution of this equation in the form

$$J_S^{(\mp)} = J_{S0} \exp\left[\int_0^z \Gamma(\eta_\mp - \nu_\mp x)\, dx\right], \quad (43)$$

where $\nu_\mp = u_p^{-1} \mp u_S^{-1}$ and $\eta_\mp = t \mp z/u_S$. The plus and minus signs preceding J pertains to the intensities of the forward and backward Stokes radiation, respectively.

It is easy to see from (43) that the gain of the Stokes signal reaches saturation at a length (see (10))

$$l_\mp = \tau_p / \nu_\mp. \quad (44)$$

The ratio of the radiation energy scattered forward and backward (the energy asymmetry coefficient) is given by

$$R = \frac{W_S^{(-)}}{W_S^{(+)}} \approx \frac{l_+}{l_-} \exp[\Gamma_0(l_- - l_+)], \quad (45)$$

where $W_S^{(\mp)}$ are the energies of the forward (–) and backward (+) Stokes pulses.

The group-delay effects should become most strongly pronounced in the case of pumping by picosecond pulses. In this case, generally speaking, it is impossible to regard the response of the molecular system as having a steady state. On the other hand, simultaneous allowance for the nonstationary character of the response of the medium and of the detunings of the group velocities introduces great difficulties in the solution of Eqs. (11) and (12), where the amplitude A_p must be regarded as a function not of η but of $\eta_\mp - \nu_\mp z$. It is possible, however, to solve this system of equations for pump pulses of two types, rectangular and exponential[9]. We present here the formulas for the growing part of the Stokes pulse in pumping by a rectangular pulse of duration $\tau_p \ll T_t$, in the case of forward scattering:

$$\overline{J_S^{(-)}} \approx \frac{\gamma_S g \eta_-}{8\pi\gamma_\sigma\psi_0(z,\eta_-)(\nu_- z + \eta_-)} \exp[2\psi_0(z,\eta_-)] \quad \text{if } \eta_- \leqslant \nu_- z; \quad (46a)$$

$$\overline{J_S^{(-)}} \approx \frac{\gamma_S g}{8\pi\gamma_\sigma\psi_0(z,\eta_-)} \left\{\exp[2\psi_0(z,\eta_-)] - \frac{1}{2}\sqrt{\frac{\eta_-}{z\nu_-}} \exp[2\psi_0(z,\nu_- z)]\right\} \quad \text{if } \eta_- \geqslant \nu_- z; \quad (46b)$$

It is seen from these formulas that the intensity in the Stokes pulse reaches a maximum value at a length $l_- = \tau_p/\nu_-$:

$$\overline{J_{S,max}^{(-)}} \approx \frac{\gamma_S g \sqrt{T_2}}{16\pi\gamma_\sigma\sqrt{2\Gamma_0 l_- \tau_p}} \exp\left[2\left(2\Gamma_0 l_- \frac{\tau_p}{T_2}\right)^{1/2}\right]. \quad (47)$$

A similar formula is obtained for the maximum of the backward-scattered radiation intensity, if l_- is replaced by l_+ in (47).

To estimate the energy of the Stokes radiation scattered forward and backward, it is necessary to multiply $J_{S\,max}$ by the duration of the corresponding Stokes pulses. The duration of the pulse moving forward is $\tau_S^{(-)} \approx \sqrt{2\tau_p T_2/\Gamma_0 l_-}$. On the other hand, the duration of the pulse moving backward is $\tau_S^{(+)} \approx l_-\nu_+$ at a cell length $l = l_-$. We then obtain for the asymmetry coefficient the expression

$$R \approx \left(\frac{2\tau_p T_2}{\Gamma_0 l_+}\right)^{1/2} \frac{l_+}{l_-^2 \nu_+} \exp\left[2\left(2\Gamma_0 \frac{\tau_p}{T_2}\right)^{1/2} (\sqrt{l_-} - \sqrt{l_+})\right]. \quad (48)$$

We see therefore that the nonstationary character of the response greatly decreases the asymmetry of the radiation, but still leaves it quite appreciable.

[9]The amplification picture is greatly altered by group detuning in the case of phase modulated pumping. Here, unlike the results of Sec. 4, there is an appreciable decrease of the gain. This question is discussed in greater detail in [28].

B. Influence of Group-delay Effects on the Spectrum of the Stokes Components

It is obvious that at appreciable cell lengths the duration of the backward Stokes pulse cannot greatly influence the width of the spectrum. At the same time, the gains attainable in the case of short pulses $(2\Gamma_0 l_+ \tau_p/T_2)^{1/2}$ are small. Consequently, the width of the backward-scattered radiation spectrum should become approximately equal to the width of the spontaneous-radiation spectrum following excitation by a short pulse.

The width of the spectrum of the forward-scattered Stokes radiation at lengths $z \ll l_-$ in the field of pumping pulses having $\tau_p \ll T_2$ is determined by formula (32a). When $z > l_-$, a broadening of the Stokes pulse (a narrowing of the spectrum) begins. The duration of the pulse is then determined by the formula

$$\tau_S^{(-)} \approx (2\tau_p\tau_2/\Gamma_0 l_-)^{1/2} + (z - l_-)\nu_-.$$

6. CONCLUSION

The present results make it possible to consider a much wider group of nonstationary-scattering problems than in the case of the customarily employed Kroll theory[16] and the works is based on it[15,18]. New factors are simultaneous allowance for the "local" (connected with T_2) and "wave" nonstationary effects, the consideration of a wide class of pumping waves, and allowance for spontaneous transitions. Of course, the relations obtained in the present paper include also Kroll's results; in particular, they follow from formulas (35) with $\Delta = 0$, $\tau_0 \to \infty$ and $\Gamma_0 z - 2\eta/T_2 > 1$. Although the analysis presented in the present paper is limited mainly to cases of pulsed and phase-modulated pumping, the general expressions make it possible to analyze also other problems of practical importance.

Interesting conclusions can be obtained, in particular, for a Gaussian noise pump; under conditions when the dispersion effects are negligible and the noise correlation time $\tau_c \ll T_2$, the gain of the spectral Stokes component of the wave is determined by the average pump intensity:

$$S(\omega_S) \sim \exp[2\gamma_S\gamma_\sigma T_2 \bar{J}_p z], \qquad \bar{J}_p = \frac{1}{T}\int_0^T J_p(\eta)\,d\eta.$$

A similar result can be obtained also for multimode pumping with unsynchronized modes.

Of definite practical interest may be the possibilities, considered above, of quasistatic (see formula (26)) or nonquasistatic (32a) compression of the pulse. Under conditions of small group delays, it is possible to realize a threefold compression of the pulse; experiments in which such effects were observed in liquids excited by picosecond pumping are described in our paper[27] 10) (see also[28]). The formulas (21)–(23) for the intensities of the scattered components also agree well with the experimental data obtained in the study of the scattering of ultrashort light pulses in liquids[18,27]. Although, insofar as we know, there are still no direct experiments on the amplification of narrow-band Stokes signals in the field of broadband pumping, the results of Sec. 4 of the present paper can be used to interpret experiments on SRS under conditions of anomalous broadenings of the pump spectrum as a result of self-action.

One of the most interesting aspects of SRS of picosecond pulses are the strong-field effects, which can lead, in particular, to noticeable changes of the population difference. This question is discussed in a number of papers[7,8,29,30]; the effects arising here have features in common with resonant self-bleaching[31,32]. We present a general result that follows from the system (2)–(12). If the pump pulse duration is $\tau_p \ll T_2$, then in the case of exact resonance ($\Delta = 0$) it follows from this system that the behavior of the population difference is described by the formula

$$n(\eta) = \cos\left\{2\gamma_\sigma \int_{-\infty} \left[J_S(t)J_{p0}(t) - \frac{\omega_p}{\omega_S}J_S^2(t)\right]^{1/2} dt\right\},$$

where $J_{p0}(t)$ is the input pump pulse. We see that n, as a function of the time and field intensities, can assume values from 1 to −1. Equations (4) show that the first to occur is the process of conversion of the pump radiation in the Stokes radiation. When n becomes negative, the Stoke wave is transformed into a pump wave. Since the population difference can reverse sign many times during the interaction time (provided, of course, that the pump-pulse energy is sufficiently large), we should expect the pump and Stokes waves to break up into individual pulses.

We note also that when $n < 0$ favorable conditions are produced for two-photon antistokes scattering, having no specific angular structure.

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10)Effects connected with nonlinearity of the refractive index were excluded in this case.

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57