Combined Effects of Molecular Relaxation and Medium Dispersion in Stimulated Raman Scattering of Ultrashort Light Pulses

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A theory is developed for stimulated Raman scattering (SRS) of radiation experiencing rapid (with respect to the transverse relaxation time of the scattered oscillations T_2) amplitude and phase modulation. In distinction to previous work, dispersion (normal or anomalous) of the medium is taken into account. It is found that qualitatively new effects arise under conditions when both molecular relaxation and dispersion are operative. The most important of these are: 1) exponential growth of Stokes components excited by ultrashort light pulses of duration $\tau_p < T_2$ in media with normal dispersion; 2) the appearance of stationary Stokes pulses in a medium with normal dispersion, the duration of which is independent of the distance traversed in the nonlinear medium; 3) the suppression of SRS due to rapid pumping phase modulation; this occurs in media with either normal or anomalous dispersion; 4) competition between processes of stationary mode formation and suppression of amplification due to pumping phase modulation. Numerical estimates show that the effects mentioned above play an important role in stimulated Raman scattering of picosecond pulses in liquids and crystals. The suppression of SRS in the pumping phase modulation field is apparently the main cause of the low efficiency of stimulated scattering in weakly focused beams.

1. INTRODUCTION

PROGRESS in the technique of ultrashort light pulses has greatly stimulated interest in the theory of nonstationary stimulated scattering (see, for example,[1-4]). Principal attention has been paid in the cited papers to nonstationary effects connected with the inertia of the mechanism that effects the energy exchange between the interacting waves (the inertia of the nonlinear polarization of the medium). In fields that are not too strong, nonstationary phenomena connected with the finite transverse-relaxation time T_2 are most significant. The gain in the field of pump pulses of duration $\tau_p < T_2$ decreases; instead of a stationary increase of the intensity of the Stokes wave in accordance with the law

$$I_S(z) = K_0 \exp(\Gamma_0 z), \tag{1a}$$

where Γ_0 is the static gain, there is a much slower increase of I_S; the intensity of the Stokes wave at a length z assumes at the end of the pump pulse the value[1)]

$$I_S(z) \sim K_1 \exp[2(2\Gamma_0\tau_p T_2^{-1}z)^{1/2}] = K_1 e^{G(z)}. \tag{1b}$$

In such an essentially nonstationary case, the gain is accompanied by the effect of the so-called nonstationary spectrum broadening[2-6]. Another class of nonstationary phenomena in stimulated Raman scattering (SRS) is connected with the propagation effects. These effects are due in fact to linear dispersion of the medium and become manifest primarily in the group delay of the pump and Stokes-component waves[2)].

[1)]The values of the pre-exponential factors depend on the scattering regime (scattering from intrinsic noise, amplification of external signal[2]).

[2)]In fields of pulses that are quite intense, a definite role can also be played by nonlinear distortions of the dispersions curves (in particular, distortions due to the real part of the combination susceptibility), but this is usually not manifest in the effects considered below.

If $\tau_p \gg T_2$, then the analysis of the nonstationary wave phenomena in SRS turns out to be particularly simple in the given-pump-field approximation (see[2]). The most significant result of the "wave" nonstationarity is in this case the sharp decrease of the rate of gain (saturation) after a characteristic length is reached, equal to the length of the group delay of the Stokes-wave and pump pulses

$$L_\nu = \tau_p/|\nu|, \qquad \nu = u_p^{-1} - u_S^{-1},$$

where u_p and u_S are the group velocities of the pump and Stokes waves. The saturation of the gain is accompanied by a broadening of the Stokes pulse in accordance with the law $\tau_S \sim z$. We note also that a number of rather general results can be obtained in this case also by taking into account the reaction of the Stokes wave on the pump (see[7-9]). For many cases of practical interest, a separate examination of the nonstationarity connected with the inertia of the nonlinear polarization and of the nonstationarity due to the propagation effects is not justified. Thus, for picosecond pulses ($\tau_p \approx 10^{-12}$ sec), the duration of which is close to or much less than the reciprocal width of the spontaneous Raman-scattering line $\Delta\Omega_{sp}^{-1} = T_2$ in typical liquids and crystals, the group delay length does not exceed 1–1.5 cm.

Certain effects which are possible when both nonstationarity mechanisms act simultaneously have been discussed qualitatively earlier[2,3]. The purpose of the present paper is the development of a consistent nonstationary SRS theory, in which account is taken simultaneously of the action of the molecular relaxation and of the dispersion. The theory is constructed under rather general assumptions concerning the modulation of the pump (short rectangular and bell-shaped pulses without phase modulation, continuous phase-modulated pumping, short pulses experiencing rapid phase modulation). We consider here different relations between the group[3)] velocities of the interacting waves (forward scattering in the case of normal and anomalous dispersion, backward scattering).

The most interesting result of our analysis is the fact that under conditions when the inertia of the molecular oscillations and the dispersion of the medium appear simultaneously, qualitatively new effects arise. We must point out here first the appearance of exponential amplification of the Stokes radiation excited by the pulses $\tau_p > T_2$ at $z < L_\nu$ and accompanied by stabilization of the shape and width of the Stokes pulse. Another important effect is the appearance of SRS as a result of rapid PM pumping (we recall that at $\nu = 0$, according to[2], the PM has practically no effect on the Stokes gain). Estimates show that the indicated effects play an important role in SRS of picosecond pulses in liquids and crystals and in SRS in self-focusing beams. They can become manifest also in other types of stimulated scattering.

2. NONSTATIONARY SRS IN A PULSE-PUMP FIELD

2.1. Fundamental Equations. Role of Sign of the Mismatch of the Group Velocities

We start with a consideration of nonstationary phenomena in a pulsed-pumping field. In the given-field approximation, in the first approximation of dispersion theory, the abbreviated equations describing the Stokes scattering by transitions obeying the alternative forbiddenness are of the following form (see [2])

$$\frac{\partial A_S}{\partial z} - \nu \frac{\partial A_S}{\partial \eta} = \gamma_1 A_p(\eta)\sigma,$$
$$\frac{\partial \sigma}{\partial \eta} = \gamma_2 A_p^*(\eta) A_S - \frac{\sigma}{T_2} + N(\eta, z). \tag{2}$$

Here A_S, $A_p(\eta)$, and σ are the amplitudes of the Stokes wave, laser wave, and molecular oscillations, respectively; T_2 is the transverse relaxation time; the function $N(\eta, z)$ is an extraneous random force characterizing the natural oscillations of the medium, and $\eta = t - zu_p^{-1}$ is the running time (the proper pump time).

Before we proceed to analyze the system (2), we note that the gist of the effects in which the inertia of the molecular oscillations and the group detuning become manifest simultaneously can be understood from qualitative considerations (see also[2,3]). The plots of Fig. 1 characterize the spatial variation of the amplitudes of the Stokes pumping and of the molecular oscillations. Under conditions of normal dispersion ($u_s > u_p$), the Stokes pulse overtakes the pump pulse, and consequently the latter is constantly in a region where the molecular oscillations are strongly excited. As a result, one should expect to obtain an intense Stokes scattering from a large volume of the medium, accompanied by the appearance of a stationary Stokes pulse. To the contrary, in anomalous dispersion ($u_S < u_p$), the pump pulse at $z > L_\nu$ propagates in a practically unexcited medium,

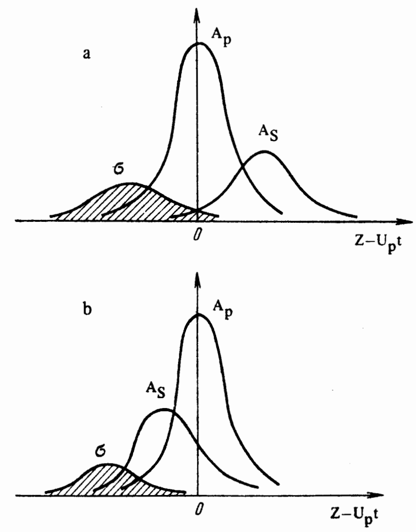

FIG. 1. Spatial variation of the pump intensity, the Stokes wave, and the molecular oscillations when a short (compared with the transverse relaxation time) pump pulse passes through the combination-active medium with normal (a) and anomalous (b) dispersion.

so that at a length $z \approx L_\nu$ the gain becomes saturated. The foregoing is confirmed by a concrete calculation performed in the succeeding subsections of this section. Particular interest attaches here to the case of normal dispersion; besides the complete solutions describing the dynamics of the gain, which are valid for arbitrary z, we give also solutions which are valid at $z > L_\nu$ and take the form of steady-state modes having a stationary amplitude profile.

2.2. Forward Scattering. Stationary Stokes Modes

For the purpose of elucidating the physical features of nonstationary forward scattering in media with normal dispersion, we propose the existence of an amplification regime such that a stationary amplitude profile of the wave is formed:

$$A_S(\eta, z) = A_{S,\text{м}}(\eta) \exp(\Gamma_\text{м} z). \tag{3}$$

Substituting the sought form of the solution in (2) and confining ourselves for the time being to the dynamic analysis ($N = 0$), we arrive at an ordinary second-order differential equation

$$\frac{d^2 B_S}{d\eta^2} + \left(\frac{1}{T_2} + \frac{\Gamma_\text{м}}{\nu} - \frac{1}{A_p(\eta)}\frac{dA_p(\eta)}{d\eta}\right)\frac{dB_S}{d\eta} + \frac{\gamma_1\gamma_2 |A_p(\eta)|^2}{\nu} B_c = 0,$$
$$B_S = A_{S,\text{м}} \exp(-\Gamma_\text{м}\eta/\nu). \tag{4}$$

We determine with the aid of (4) the envelope of the stationary Stokes pulses for two types of pump modulation.

1. Rectangular pulse: $A_p(\eta) \equiv A_p$ at $|\eta| \leq \tau_p/2$ and $A_p(\eta) \equiv 0$ at $|\eta| > \tau_p/2$. In this case the stationary Stokes pulse takes the form (see Fig. 2)

$$A_{S,\text{м}} \sim \begin{cases} \exp\left[\left(\frac{\Gamma_\text{м}}{\nu} - \frac{1}{T_2}\right)\frac{\eta}{2}\right] \sin\left(\frac{\eta}{\tau_p} - \frac{1}{2}\right) b, & |\eta| < \frac{\tau_p}{2}, \\ \exp\left[\frac{\Gamma_\text{м}\eta}{2\nu} - \frac{\eta}{2T_2}\right], & \eta < -\frac{\tau_p}{2}, \\ 0 \quad , & \eta > \frac{\tau_p}{2}. \end{cases} \tag{5}$$

[3)]As shown in[2], allowance for dispersion in first approximation, which reduces to taking the group delay into account, is quite adequate for typical problems in picosecond-pulse optics.

The gain of the stationary pulse is

$$\Gamma_M = 2(\gamma_1\gamma_2 A_p^2 \nu)^{1/2} |\cos b| - \nu / T_2. \quad (6)$$

The parameter b, which determines the shape of the Stokes-wave pulse and its gain, is given by the transcendental equation

$$|\sin b| = b / n \quad (7)$$

and depends on the quantity

$$n = (\Gamma_0 \tau_p T_2^{-1} L_\nu / 2)^{1/2} = (\gamma_1\gamma_2 A_p^2 \tau_p^2 / \nu)^{1/2}, \quad (8)$$

which is determined by the pump intensity and by the properties of the medium. We note that the quantity n is equal, apart from a numerical factor, to the growth increment of the Stokes-wave intensity over the group length in the nonstationary scattering regime, $n \approx G(L_\nu)$ (compare with (1b)). Under real conditions (scattering of powerful picosecond pulses) usually $n \gg 1$ and $b \approx \pi$. Then the mode increment $\Gamma_M \approx (2\Gamma_0 \nu T_2^{-1})^{1/2} - \nu/T^2$ does not depend on the duration of the principal radiation. The vertex of the Stokes pulse is located at the trailing edge of the pump pulse $\eta_M = \tau_p/2$ (see Fig. 2), and the pulse duration $\Delta\eta \approx 3\nu/\Gamma_M$ decreases with increasing gain. In strongly dispersive media the Stokes pulse energy leaves rapidly the pump region and the gain of the mode vanishes at $n \approx 1$.

2. Let us consider a bell-shaped pump pulse $A_p(\eta) = A_p \cosh^{-1}(\eta/\tau_p)$. We obtain the stationary Stokes-radiation pulse localized near the main pulse from Eq. (4):

$$A_{S,M}(\eta) = \exp\left[\left(n - 1 - \frac{\tau_p}{T_2}\right)\frac{\eta}{\tau_p}\right] \mathrm{ch}^{-n}\frac{\eta}{\tau_p}. \quad (9)$$

The mode increment is (compare with (6))

$$\Gamma_M = (2n - 1 - \tau_p / T_2) L_\nu^{-1}. \quad (10)$$

The vertex of the Stokes pulse leads the maximum of the main pulse

$$\eta_M = \frac{\tau_p}{2} \ln \frac{2n - 1 - \tau_p/T_2}{1 + \tau_p/T_2}, \quad (11)$$

and its duration is

$$\Delta\eta_M \approx \tau_p[2n(1 + \tau_p/T_2)^{-1}(2n - 1 - \tau_p/T_2)^{-1}]^{1/2}.$$

The mode gain vanishes when $n = 1/2 + \tau_p/2T_2$, and the pulse is then strongly broadened ($\Delta\eta_M \to \infty$)[4]. With increasing pump field, the increment tends to the value $\Gamma_M = 2(\gamma_1\gamma_2 A_p^2 \nu)^{1/2}$; the vertex of the Stokes pulse is located at the trailing edge of the pump pulse, while the duration remains constant at $\Delta\eta_M \approx \tau_p(1 + \tau_p/2)^{1/2}$.

It is seen from the foregoing examples that the waveform of the amplitude modulation of the pump (the rise time of the fronts) influences strongly the stationary amplitude profile of the Stokes pulse, and to a lesser degree the character of the gain itself. Thus, the calculated growth increments can be used in the discussion of the experimental results for pulses which do not coincide exactly in shape with those considered above (including Gaussian pulses). If the Stokes pulse lags the pump, $\nu < 0$ (forward SRS in the case of anomalous dispersion and backward SRS), then there are no exponentially growing modes. The saturation of the gain occurs within the quasistatic length $z \approx L_\nu$.

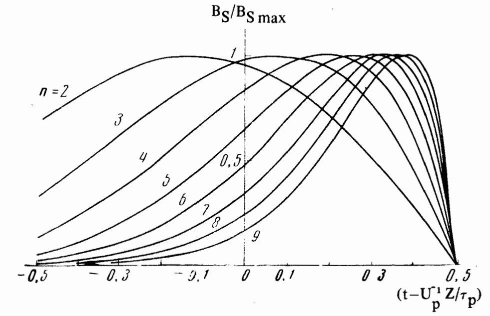

FIG. 2. Waveform of stationary Stokes pulse at $z > L_\nu$ in nonstationary SRS in a medium with normal dispersion, for a rectangular pump pulse at different values of n. We see that the waveform of the stationary pulse differs from the pump pulse, and the Stokes energy is localized in the case of strong pumping near the trailing edge.

2.3. Dynamics of Formation of Stationary Stokes Pulse in Forward Scattering

Let us consider the forward SRS process under ordinary conditions of normal dispersion of the scattering medium ($u_S > u_p$, $\nu > 0$). We assume that the function $N(\eta, z)$ in (2), which describes the natural fluctuations of the medium, is delta-correlated in space and in time

$$\langle N(\eta, z) N(\eta', z')\rangle = g\delta(\eta - \eta')\delta(z - z') \quad (12)$$

(The intensity of the random force g can easily be expressed in terms of the parameters of the spontaneous Raman scattering (see[2]). Let the pump pulse have a bell-shaped form $A_p(\eta) = A_p \cosh^{-1}(\eta/\tau_p)$. We solve Eqs. (2) by the Riemann method. As a result of the calculations we obtain the following expression for the averaged Stokes-pulse intensity:

$$I_S = g\gamma_1^2 A_p^2 \,\mathrm{ch}^{-2}\frac{\eta}{\tau_p}\int_0^\infty \exp\left(-\frac{2\xi}{T_2}\right) d\xi \times \int_0^z \mathrm{ch}^2\frac{\eta - \xi}{\tau_p}\,\mathrm{ch}^{-2}\frac{\eta - \xi + \nu s}{\tau_p} F^2(1 + n, 1 - n; 1; y)\, ds. \quad (13)$$

Here F is a hypergeometric function with argument

$$y = -\mathrm{sh}\frac{\nu s}{\tau_p}\,\mathrm{sh}\frac{\xi}{\tau_p}\,\mathrm{ch}^{-1}\frac{\eta}{\tau_p}\,\mathrm{ch}^{-1}\frac{\eta - \xi + \nu s}{\tau_p}.$$

We note first that under the conditions of group synchronism ($\nu \to 0$, $n \to \infty$) the hypergeometric function goes over into a modified Bessel function of zero order, I_0, and the solution (13) itself goes over into a well known previously obtained solution[2]. In the case of integer values of n, the function F is expressed by a finite series, and in particular $F \equiv 1$ when $n = 1$. In the latter case

$$I_S = \frac{g\gamma_1^2 A_p^2 \tau_p}{\nu}\,\mathrm{ch}^{-2}\frac{\eta}{\tau_p}\,\mathrm{sh}\frac{\nu z}{\tau_p} \times \int_0^\infty \exp\left(-\frac{2\xi}{T_2}\right)\mathrm{ch}^2\frac{\eta - \xi}{\tau_p} \times \mathrm{ch}^{-2}\frac{\eta - \xi + \nu z}{\tau_p}\, d\xi. \quad (14)$$

In Fig. 3 we traced the dynamics of the amplification of the Stokes pulse at $n = 1$ and $\tau_p = T_2/2$. We see that a stationary pulse that increases exponentially with the coordinate is formed at $z \gtrsim 4L_\nu$.

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[4]This case was discussed in[2].

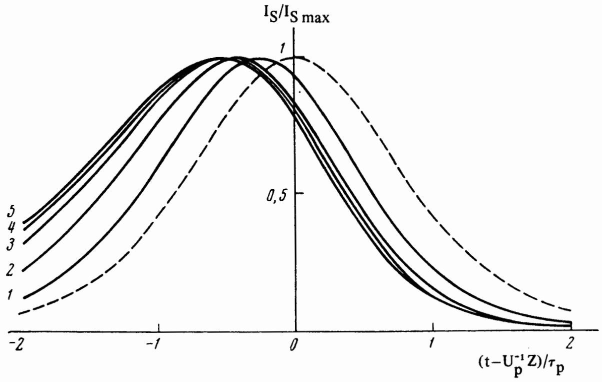

FIG. 3. Dynamics of formation of a stationary Stokes pulse in forward SRS as a function of the length of the scattering medium in the case n = 1 and $\tau_p = T_2/2$. The abscissas represent the time reckoned from the center of the pump pulse. The parameter of the curves is the normalized distance z/L_ν traversed by the pulse in the medium. Plots 1, 2, 3, 4, and 5 correspond to z/L_ν = 1, 2, 3, 4 and 5, respectively. The dashed curve is the pump envelope. The ordinates represent the normalized power $I_S/I_{S.\,max}$.

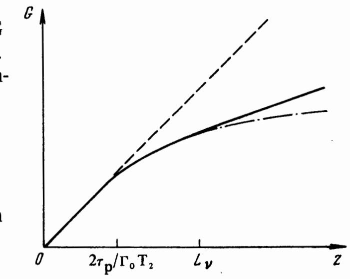

FIG. 4. Growth increment G of the Stokes-wave amplitude vs. the length of the medium z (continuous curve). A stationary exponentially-amplified mode sets in at $z > L_\nu$. The dashed line shows the gain in a nondispersive and nonrelaxing medium. The dash-dot line shows the gain of short pulses in nondispersive medium.

An analysis of the general expression (13) shows that the stationary mode of the Stokes wave is always formed behind the group length $z \geq L_\nu$:

$$I_S = g\frac{\gamma_1}{\gamma_2}\frac{2^{-2n}}{1+\tau_p/T_2}\Gamma(2n)\Gamma\left(\frac{\tau_p}{T_2}\right)\Gamma\left(2n-1-\frac{\tau_p}{T_2}\right)\Gamma^{-4}(n)\exp(2\Gamma_M z)A^2_{S.M}(\eta), \tag{15}$$

where $\Gamma(n)$ is the gamma function, and the mode profile $A_{S.M}$ and the gain Γ_M are given by the previously derived formulas (9) and (10). At large gains ($n \gg 1$) in a field of picosecond pulses ($\tau_p \ll T_2$) we have from (15)

$$I_S = g\frac{\gamma_1}{\gamma_2}\frac{2^{2n-2}}{\pi n}\exp(G_M)A^2_{S.M}(\eta), \quad G_M = 2\Gamma_M z \tag{16}$$

(see Fig. 4).

2.4. Dynamics of Backward Raman Scattering

In backward SRS the Stokes and pump pulses propagate in opposite directions with a relative velocity equal to double the velocity of light. The group mismatch $|\nu| = u_p^{-1} + u_S^{-1} \approx 2/c$ greatly exceeds in magnitude the mismatch $\nu \approx (u_p - u_S)/c^2$ occurring in forward scattering. Accordingly, the group length $L_\nu = \tau_p/|\nu|$ is greatly decreased, and the effects of the group delay become manifest earlier and more sharply than in forward SRS.

The solution (13) obtained for forward SRS is also valid for backward scattering, with due allowance for the reversal of the sign of the group mismatch; the parameter n becomes imaginary in this case, $n = i|n|$. The intensity of the backward Stokes pulse at large gains, $|n| \gg 1$, is given by

$$I_S \approx g\frac{\gamma_1^2 A_p^2}{8\pi n}\int_0^\infty d\xi\exp\left(-\frac{2\xi}{T_2}\right)\left\{\frac{\mathrm{ch}[(\eta-\xi)/\tau_p]}{\mathrm{sh}(\xi/\tau_p)}\right\}^{1/2} \times\int_0^z\left\{\frac{\mathrm{ch}[(\eta-\nu s)/\tau_p]}{\mathrm{sh}(\nu s/\tau_p)}\right\}^{1/2}\frac{\exp(2n\arcsin y_1)}{\mathrm{ch}^2[(\eta-\nu s)\tau_p]}ds. \tag{17}$$

$$y_1 = 2\left[\mathrm{sh}\frac{\nu s}{\tau_p}\mathrm{sh}\frac{\xi}{\tau_p}\mathrm{ch}\frac{\eta-\xi}{\tau_p}\mathrm{ch}\frac{\eta-\nu s}{\tau_p}\right]^{1/2}\left(\mathrm{ch}\frac{\eta}{\tau_p}\mathrm{ch}\frac{\eta-\xi-\nu s}{\tau_p}\right)^{-1}.$$

Unlike the forward SRS, gain saturation sets in at lengths $z \gtrsim L_\nu$ in backward scattering, and the saturation effect depends on the ratio of the group length L_ν to the quasistatic $L_\tau = \tau_p/T_2\Gamma_0$. If $L_\nu < L_\tau$ the gain saturation occurs in a quasistationary regime ($G = \Gamma_0 z$); the saturation level is $G = \Gamma_0 L_\nu$.

In a field of powerful short pulses ($L_\nu > L_\tau$) a nonstationary scattering regime develops over the group length; according to (17) we have

$$G_{-\nu,\tau} = 2n\arcsin\left\{\left[\mathrm{sh}\frac{2\nu z}{\tau_p}\mathrm{sh}\frac{2(\eta-\nu z)}{\tau_p}\right]^{1/2}\mathrm{ch}^{-1}\frac{\eta}{\tau_p}\right\}. \tag{18}$$

The nonstationary gain ($G \sim z^{1/2}$) saturates here at the level $G_{-\nu,T} = \pi|n|$, which is lower, owing to the inertia of the molecular oscillations, than in the case of the quasistatic gain.

3. SRS OF PHASE-MODULATED PUMP RADIATION

Under conditions of group synchronism ($\nu = 0$), phase modulation (PM) of the pump, $A_p(\eta) = A_{p0}(\eta)e^{i\varphi(\eta)}$ has practically no influence on the amplification of the Stokes component (see[1,3]). Group-delay effects, do, however, change appreciably the picture of SRS of phase-modulated pumping. Indeed, in this case the PM of the pump extends to the Stokes component

$$A_S = \tilde{A}_S(\eta, z)e^{i\varphi(\eta+\nu z)}. \tag{19}$$

The phase of the molecular oscillations strives to follow the phase difference between the pump and the Stokes component:

$$\sigma = \tilde{\sigma}e^{i\varphi(\eta)-i\varphi(\eta+\nu z)}. \tag{20}$$

However, owing to the finite relaxation time T_2, the molecular oscillations cannot follow the rapid beats of the driving force, and this leads in final analysis to a decrease of the gain, and also to its saturation under certain conditions. For the amplitudes $\tilde{A}_S$ and $\tilde{\sigma}$ defined in accordance with (19) and (20), the equations in (2) take the form

$$\frac{\partial\tilde{A}_c}{\partial z} - \nu\frac{\partial\tilde{A}_S}{\partial\eta} = -i\gamma_1 A_{p0}(\eta)\tilde{\sigma}, \tag{21a}$$

$$\frac{\partial\tilde{\sigma}}{\partial\eta} + \left\{\frac{1}{T_2} + i[\Omega(\eta) - \Omega(\eta+\nu z)]\right\}\tilde{\sigma} = i\gamma_2 A_{p0}(\eta)\tilde{A}_S + Ne^{i\varphi(\eta)-i\varphi(\eta+\nu z)}. \tag{21b}$$

In (21) the instantaneous frequency is

$$\Omega(t) = \partial\varphi/\partial t. \tag{22}$$

We note, first, that certain general conclusions concerning the influence of PM of the pump on the gain of the Stokes waves can be drawn without solving the equations in (21) exactly. We see that in a nondispersive medium

($\nu = 0$) the PM of the pump does not influence the SRS (in the scattering regime). On the other hand, in a dispersive medium, the SRS becomes sensitive to changes in the pump frequency (the derivative of the phase). Neglecting envelope group-delay effects ($z < L_\nu = \tau_p/|\nu|$), it is easy to estimate the characteristic spatial scales within which phase modulation can exert an influence on the gain of the Stokes wave. For $\tau_p \gg T_2$ this influence comes into play, obviously, if the frequency deviation exceeds the width of the spontaneous scattering line, $\Delta\Omega_{max} \gg 2/T_2$. For pulses with a linear time variation of the frequency (quadratic variation of the phase)

$$\Omega = \Omega_0 t / \tau_p \tag{23}$$

the beat frequency in (21) increases in proportion to the traversed distance:

$$\Delta\Omega = \Omega(\eta + \nu z) - \Omega(\eta) = \Omega_0 z / L_\nu. \tag{24}$$

PM of the pump greatly limits the growth of the Stokes component when $|\Delta\Omega| \gtrsim 2/T_2$, i.e., over distances $z \gtrsim l_{ph_1}$, where

$$l_{ph_1} = 2L_\nu / \Omega_0 T_2. \tag{25}$$

We can verify further that when $z > l_{ph_1}$ the growth increment of the Stokes waves is practically independent of z. For pulses that are short compared with the transverse relaxation time ($\tau_p < T_2$), the frequency deviation should be compared with the nonstationary width of the Stokes line (compare with[2]). For a rectangular pump pulse

$$\Delta\omega_S = (\Gamma_0 z / 2\tau_p T_2)^{1/2},$$

this gives a characteristic space scale

$$l_{ph_2} = \Gamma_0 l_{ph_1} / \nu\Omega_0, \tag{26}$$

Usually $l_{ph_2} > l_{ph_1}$. For a bell-shaped pump pulse $\Delta\omega_S \approx \Delta\omega_p$ and we get $l_{ph_2} \approx (\Omega_0\nu)^{-1}$ in place of (26). A more complicated picture arises when $z > L_\nu$; in this case competition sets in between the effect of occurrence of exponentially growing modes and the suppression of the SRS via the PM. This regime will be analyzed in Sec. 4; in the remaining subsections of this section we shall confine ourselves to the case $z < L_\nu$.

3.1. Dynamics of SRS for a Linear Variation of the Pump Frequency

In accordance with the foregoing, we can put in (21) $A_{p0}(\eta) \approx A_{p0}(\eta + \nu z)$. Introducing a new variable $\eta_S = \eta + \nu z$ and changing over to the normalized amplitude of the Stokes pulse

$$A_S = A_{p0}(\eta_S) f(z, \eta_S), \tag{27}$$

we obtain from (21) the equation

$$\frac{\partial^2 f}{\partial\eta_S \partial z} + \left\{\frac{1}{T_2} + i[\Omega(\eta_S - \nu z) - \Omega(\eta_S)]\right\}\frac{\partial f}{\partial z} - \gamma_1\gamma_2 A_{p0}^2(\eta_S) f = N e^{i\varphi(\eta_S - \nu z) - i\varphi(\eta_S)}. \tag{28}$$

We seek the solution of (28) by the Riemann method. The Riemann function R satisfies the equation

$$\frac{\partial^2 R}{\partial\eta_S \partial z} + \left\{\frac{1}{T_2} + i[\Omega(\eta_S - \nu z) - \Omega(\eta_S)]\right\}\frac{\partial R}{\partial z} - \gamma_1\gamma_2 A_{p0}^2(\eta_S) R = 0 \tag{29}$$

with boundary conditions R = 1 at z = 0 and $\eta_S = 0$. For a linear variation of the pump frequency we can obtain an exact solution of (29):

$$R = \exp(-\eta_S / T_2)\Phi(-{}^1\!/\!_4 i\Gamma_0 l_{ph_1};\ 1;\ \xi), \tag{30}$$

(where Φ is the confluent hypergeometric function). In the absence of PM of the pump ($\Omega_0 \to 0$), and also in the presence of PM but at $\nu = 0$ ($l_{ph_1} \to \infty$), the function Φ is expressed in terms of a Bessel function, and consequently

$$R = \exp(-\eta_S / T_2) I_0[(2\Gamma_0 z\eta_S / T_2)^{1/2}], \tag{31}$$

i.e., (31) contains as a particular case the previously obtained[1-3] solutions of the equations of nonstationary SRS. Using (30), we obtain for the average Stokes-wave intensity

$$I_S = g\gamma_1^2 |A_{p0}|^2 \int_0^z dz_1 \int_0^{\eta_S} d\eta_1 \exp\left(-\frac{2\eta_1}{T_2}\right)\left|\Phi\left(-\frac{i\Gamma_0 l_{ph_1}}{4},\ 1;\ \frac{2iz_1\eta_1}{l_{ph_1}T_2}\right)\right|^2. \tag{32}$$

3.2. Quasistationary Amplification Regime

We consider first SRS of long ($\tau_p \gg T_2$) PM pump pulses. The upper limit in the internal integral of (32) can then be allowed to go to infinity ($\eta_S \to \infty$). This yields

$$I_C = \frac{1}{2} g\gamma_1^2 |A_{p0}|^2 T_2 \int_0^z \exp\left[\frac{\Gamma_0 l_{ph_1}}{2}\operatorname{arctg}\frac{z_1}{l_{ph_1}}\right] \times F\left(-\frac{i\Gamma_0 l_{ph_1}}{4}\ \frac{i\Gamma_0 l_{ph_1}}{4}\cdot 1;\ \frac{1}{1 + (l_{ph_1}/z_1)^2}\right) dz_1, \tag{33}$$

where F is the hypergeometric function. For the calculations that follow we can use the asymptotic representation of the function F (see[10], p. 88). Calculation yields

$$I_C \approx \frac{g\gamma_1 (1 + z^2/l_{ph_1}^2)}{4\gamma_2(\pi\Gamma_0 z)^{1/2}} \exp\left(\Gamma_0 l_{ph_1}\operatorname{arctg}\frac{z}{l_{ph_1}}\right),$$

$$1/\Gamma_0 \ll z \ll \Gamma_0 l_{ph_1}^2/4. \tag{34}$$

From (34) we see that at $z \ll l_{ph_1}$ we have

$$I_S \approx \frac{g\gamma_1}{4\gamma_2(\pi\Gamma_0 z)^{1/2}} \exp(\Gamma_0 z)$$

(the usual exponential growth corresponding to group synchronism). With increasing z, the growth of I_S slows down and saturation sets in at distances $z > l_{ph_1}$ (see Fig. 5). The growth increment tends in this case to $\pi\Gamma_0 l_{ph_1}/2$. At $z \gg l_{ph_1}$ it follows from (34) that

$$I_C \approx \frac{g\gamma_1}{2\pi\gamma_2}\frac{z}{l_{ph_1}} \exp\left(\frac{\pi}{2}\Gamma_0 l_{ph_1}\right), \tag{35}$$

i.e., at large distances there remains only weak linear growth of the Stokes component.

3.3. Short Phase-modulated Pump Pulses

For short pump pulses it is necessary to take into account the finite limits of the internal integral in (32). We confine ourselves to the case of greatest practical interest, that of large gain $2\Gamma_0 z\eta_S/T_2 \gg 1$. This allows us to use an asymptotic formula for the confluent hypergeometric function Φ (see[10], p. 269):

$$\Phi(ia, 1; -ix) = \frac{1}{4\sqrt{\pi}} \frac{ix^{1/2} + 2[(a+i/2) - x/2]^{1/2}}{(a+i/2)^{1/2}[(a+i/2)x - x^2/4]^{1/4}}$$
$$\times \exp\left\{\frac{ix}{2} + [(a+i/2)x - x^2/4]^{1/2} - 2ia \ln \frac{ix^{1/2} + [4(a+i/2) - x]^{1/2}}{2(a+i/2)^{1/2}}\right\} \quad (36)$$

where $x = 2\eta_S z/T_2 l_{ph_1}$, $a = \Gamma_0 l_{ph_1}/4$, and $ax \gg 1$ by definition.

For pulses with duration $\tau_p \ll T_2$ at distances $z < l_{ph_2}$, the amplification of the Stokes wave is given by

$$I_S \approx \frac{g\gamma_1 a}{2\pi\gamma_2 \sqrt{x}(4a - x)^{3/2}} \exp\left\{[x(4a-x)]^{1/2} + 4a \arcsin \frac{1}{2}\sqrt{ax}\right\}. \quad (37)$$

At $z \ll l_{ph_2}$ it follows from (37) that

$$I_S \approx \frac{g\gamma_1 a}{8\pi\gamma_2 (2\Gamma_0 z\eta_S/T_2)^{1/2}} \exp[2(2\Gamma_0 z\eta_S/T_2)^{1/2}], \quad (38)$$

i.e., an expression coinciding with (1b) is obtained. With increasing z, the growth of I_S slows down and saturation of the growth increment sets in at $z \to l_{ph_2}$ (see Fig. 5). At $z \gg l_{ph_2}$ we obtain from (32)

$$I_S \approx \frac{g\gamma_1}{2\pi\gamma_2} \frac{z\tau_p}{l_{ph_1} T_2} \exp\left(\frac{\pi}{2}\Gamma_0 l_{ph_1}\right). \quad (39)$$

Thus, just as for long pulses, the limiting value of the growth increment is $\pi\Gamma_0 l_{ph_1}/2$. This value is attained, however, at large distances, since $l_{ph_2} \gg l_{ph_1}$.

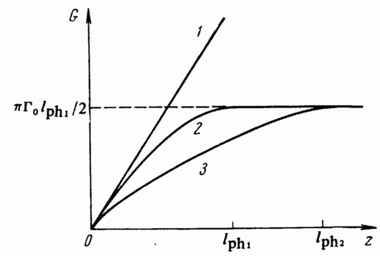

FIG. 5. Growth increment G of the first Stokes component in a phase-modulated pump field ($A_p(\eta) = A_p \exp(i\eta^2/\tau_{ph}^2)$) for the cases $\nu = 0$ (curve 1) and $\nu \neq 0$ (2, 3). Curve 2 is constructed for $\tau_p/T_2 \gg 1$ and curve 3 for $\tau_p/T_2 \gg 1$. $z < L_\nu$ in all cases.

4. GENERAL CASE. PUMP WITH FAST AMPLITUDE MODULATION AND PHASE MODULATION IN A STRONGLY DISPERSIVE MEDIUM

Under conditions when effects of envelope group delay are significant ($z > L_\nu$), the interaction of nonstationary phenomena caused by the amplitude and phase modulation of the pump has a complicated character. Of greatest interest, just as in the absence of PM, is the case of normal dispersion. Competition sets in here between the processes of formation of exponentially growing stationary modes and the suppression of the scattering by the PM of the pump. It is interesting that in a sufficiently powerful pumping field (or in the case of not too rapid PM), stationary phase-modulated modes that increase exponentially with distance are possible for all values of z. The conditions for the occurrence of such modes can be determined by using the procedure of Sec. 2.2.[5)] For a Gaussian pulse experiencing quadratic PM

$$A_p(\eta) = A_{p_0} \exp[-(\tau_p^{-2} + i\tau_{ph}^{-2})\eta^2] \quad (40)$$

(τ_{ph} is connected here with the quantity Ω_0 introduced in (23) by the relation $\Omega_0 = 2\tau_p/\tau_{ph}^2$) we can obtain an approximate expression (which is valid near the vertex of the pulse) for an exponentially growing stationary PM mode

$$A_S(\eta, z) = \exp(\Gamma_m z) \exp[(\Gamma_m - \nu T_2^{-1})\eta/2\nu - (\tau_p^{-2} + i\tau_{ph}^{-2})\eta^2/2] H_m(y) e^{-y^2/2} \quad (41)$$

where $H_m(y)$ are Hermite polynomials

$$y \approx (4n^2 - \tau_p^4\tau_{ph}^{-4})^{1/4}\eta/\tau_p + (1 + i\tau_p^2\tau_{ph}^{-2})(4n^2 - \tau_p^4\tau_{ph}^{-4})^{-1/4},$$

and n is determined by formula (8).

The mode increment is

$$\Gamma_m = L_\nu^{-1}(4n^2 - \tau_p^4/\tau_{ph}^4)^{1/2} - \nu/T_2. \quad (42)$$

It follows from (42) that the mode increment decreases with increasing rate of phase modulation ($\tau_{ph} \to 0$); at the same time, if $\tau_{ph} > \tau_p/\sqrt{n}$, the exponential growth is retained. The permissible rate of phase modulation is larger the higher the pump power ($n \sim \sqrt{I_p}$). Exact expressions for the modes can be obtained in a pump field of the form

$$A_p(\eta) = A_{p_0} \frac{\exp[-2id \ln \mathrm{ch}(\eta/\tau_p)]}{\mathrm{ch}(\eta/\tau_p)}. \quad (43)$$

Such a bell-shaped pulse with a phase modulation that slows down towards the edges of the pulse is apparently a good approximation of real PM pulses obtained in solid-state generators of picosecond pulses (see[12]). Near the vertex of the pulse, the PM is given by $\varphi = -id\eta^2/\tau_p^2$, and the quantity τ_{ph}^2 introduced in (40) is connected with d by the relation $d \approx \tau_p^2 - \tau_{ph}^2$. In this case the Stokes-wave mode takes the form

$$A_S = \exp\left[\left(\lambda - \frac{\tau_p}{T_2} - 1 - id\right)\frac{\eta}{\tau_p}\right]\left(\mathrm{ch}\frac{\eta}{\tau_p}\right)^{-\lambda - id} \exp\left[\frac{z}{L_\nu}\left(2\lambda - 1 - \frac{\tau_p}{T_2}\right)\right], \quad (44)$$

where $\lambda = (n^2 - d^2)^{1/2}$.

If $n \le [(1 + \tau_p/T_2)^2 + 4d^2]^{1/2}/2$, the exponential gain vanishes (it is seen that the PM increases the threshold pump intensity at which the exponential gain is retained at $z > L_\nu$). Let us determine the laws of phase and frequency modulation of the Stokes wave. Introducing a real amplitude and a real phase, we have

$$A_S = |A_S| \exp(i\varphi_S), \quad \varphi_S = -\frac{d\eta}{\tau_p} - d \ln \mathrm{ch}\frac{\eta}{\tau_p}$$

and for the frequency we get

$$\omega_S = \frac{d\varphi_S}{d\eta} = -\frac{d}{\tau_p}\left(1 + \mathrm{th}\frac{\eta}{\tau_p}\right); \quad \Delta\omega_S = -\frac{d}{\tau_p}\mathrm{th}\frac{\eta}{\tau_p}.$$

It is of interest to compare the widths of the pump spectra (43) and of the phase-modulated stationary Stokes mode. Calculation yields for the ratio of the pump and Stokes spectrum widths

$$\Delta\omega_S/\Delta\omega_p \approx \lambda/d[4\pi^2 n^2 - (4\pi^2 + 1)d^2]^{1/2}. \quad (45)$$

[5)]Exact solutions for pulses experiencing simultaneous phase and amplitude modulation (of the type of the solution obtained in Sec. 2 for a bell-shaped pulse) can be obtained only for specially chosen types of modulation. One of the examples where such a solution is possible is pump modulation in accordance with the law $A_p(\eta) = A_{p_0}[\cosh(\eta/\tau_p)]^{-1-2id}$. In the present article, however, we confine ourselves to more lucid approximate relations. Some results pertaining to exact nonstationary solutions will be reported by us in subsequent papers (see[11]).

5. CONCLUSION

The results present a sufficiently complete picture of SRS in a pump field with rapid amplitude and phase modulation. The most important effect due to the joint action of molecular relaxation and normal dispersion of the medium is the effect of formation of stationary exponentially growing Stokes modes[6)]. This circumstance must be taken into account in the interpretation of experiments on forward SRS of picosecond pulses in crystals and liquids (at $\tau_p \approx 10^{-12}$ sec, a typical value for condensed media is $L_\nu \approx 1-2$ cm, and the length of the scattering medium is usually noticeably larger). Comparing (1b) and (10), we can easily verify that the growth increment of the intensity of a Stokes wave at $z > L_\nu$, under conditions favoring the appearance of modes, exceeds the nonstationary growth increment attainable over the group length (we are considering the case $n \gg 1$, which is of greatest practical interest):

$$\frac{G(v>0)}{G(v=0)}=\frac{4[\Gamma_0\tau_p L_\nu/2T_2]^{1/2} zL_\nu^{-1}}{[\Gamma_0\tau_p L_\nu/T_2]^{1/2}}\approx\frac{z}{L_\nu}.$$

The use of this circumstance makes it possible to develop effective Raman lasers for picosecond pulses, by making the pump and Stokes pulses pass many times through the scattering medium in a traveling-wave resonator that ensures an optical path length $z \gg L_\nu$. The possibility of exciting exponentially-growing modes in the field of picosecond pulses makes it desirable to obtain laser mode locking in all cases when a high efficiency of the stimulated scattering is desired. Indeed, the ratio η of the growth increments of the first Stokes component in the field of a nanosecond pulse and a train of N picosecond pulses with the same total energy is

$$\eta \approx (2\Gamma_0 L_\nu N T_2/\tau_p)^{1/2}.$$

Here τ is the duration of the nanosecond pulse. Typical values of $\Gamma_0 L_\nu$ do not exceed 10—30; for liquids and crystals, on the other hand, $NT_2/\tau \approx 10^{-3}$ and consequently $\eta \approx 10^{-1}$. The foregoing is apparently the reason for the very high efficiencies of stimulated Raman scattering attained in a field of picosecond pulses[7)][15]. The sharp decrease of the growth increment and its saturation in a dispersive medium, due to rapid PM of the pump are apparently the main causes of the uneven gain of SRS in a train of picosecond pulses (see[3,15]) and of the suppression of SRS in self-focusing beams.

In estimates, when dealing with pulses with specified PM, the value of Ω_0 in (23)—(25) can be set approximately equal to the spectral width caused by the phase modulation. In a medium with a nonlinear retractive index ($n = n_0 + n_2 I$) the phase of the pump varies like $\varphi = 2kn_2 I(t)z$ (see[16]). It can be shown that in this case the characteristic length l_{ph1}, at which saturation of the gain sets in, is expressed in terms of the experimentally-observed width of the pump spectrum at the exit from a sample of length l by the formula $l_{ph} = (2L_\nu l/T_2\Delta\omega_l)^{1/2}$. As shown in[18], the values of l_{ph} calculated from this formula do not exceed several tenths of a centimeter for typical experiments with strongly-focusing liquids.

The competition between the effect of formation of stationary modes and the suppression of the SRS by the phase modulation apparently explains the results of the experiments[19] on excitation of SRS in calcite by picosecond pulses from a neodymium laser and its second harmonic. At double the frequency, the index of the phase modulation decreases, and this leads to a more effective excitation of the SRS in spite of the decrease in the peak power.

The developed theory can be used to analyze the characteristics of higher Stokes and anti-Stokes components in nonstationary scattering. An interesting question is that of nonstationary scattering by polaritons. Application of the calculation procedure described here to this question (see A. A. Golger, Diploma Thesis, Physics Department, Moscow State University, 1970) has made it possible to establish that the width of the spectral line of infrared oscillations in the nonstationary regime is of the order of the corresponding width of the spontaneous line, independently of the width of the pump spectrum. We note, finally, that a natural next step in the development of a nonstationary SRS theory is its generalization to include the case of randomly-modulated pumping. Some results pertaining to Gaussian pumping are given in[20].

[6)]There is a definite analogy between Stokes modes in SRS and stationary modes in three-wave parametric amplification (compare with [13,14]). It is important to emphasize, however, that stationary Stokes modes are formed also in SRS by fully-symmetrical local oscillations (with arbitrary wave number), whereas in parametric amplification the modes are formed for propagating waves. In a certain sense, molecular oscillations are analogous to a wave with a group velocity that differs strongly from the pump velocity.

[7)]We note that stabilization of the width of the Stokes spectrum, due, apparently, to formation of stationary modes, was observed in benzene by Nurminskiĭ[17].

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