Amplification during stimulated Raman scattering in a nonmonochromatic pump field

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A detailed experimental study has been carried out of the interaction between a narrow-band Stokes signal and a nonmonochromatic pump during stimulated Raman scattering (SRS). Results obtained suggest that, when the pump intensity is less than a critical value ($I_p < I_{crit}$), the spatial growth rate of the signal gain is much smaller than the static growth rate and is determined by the intensity of the corresponding resonance mode of the exciting radiation. For $I_p > I_{crit}$, the growth rate increases sharply and tends to the static value which depends on the resultant intensity of all the modes. The spectrum of the outgoing Stokes signal for $I_p > I_{crit}$ is broadened and, when the gain is large, completely repeats the pump spectrum. The experimental results are interpreted theoretically in terms of a model in which the continuous pump spectrum is replaced by a set of equidistant modes with intermode separation $\Omega \gg 1/T_2$. This model can describe the experimental situation for an arbitrary ratio of the length of the active region to the range of the coherent interaction.

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INTRODUCTION

Stimulated scattering of nonmonochromatic waves has now been extensively investigated and an understanding of the processes accompanying it has been achieved. However, experimental difficulties have, until quite recently, prevented experiments on the amplification of a Stokes signal differing from the pump by the degree of monochromaticity, but it is precisely this situation that has been examined theoretically in the greatest detail (although it is true that this has been done mainly for limiting cases).

In a previous paper,[1] we reported the results of experiments on the amplification of a narrow-band Stokes signal in the field of a broad-band pump. We demonstrated experimentally the existence of a critical intensity and the dependence on the pump linewidth, and found that, when the gain was large, the spectrum of the Stokes signal was broadened and began to reproduce the spectrum of the pump.

The aim of the present work was to examine the broadening of the spectrum of a narrow-band Stokes signal ($\Delta\nu_s \ll \Delta\nu_t$) during amplification in the field of a broad-band pump ($\Delta\nu_p \gg \Delta\nu_t$, where $\Delta\nu_t$ is the spontaneous Raman-scattering linewidth). We have carried out a detailed experimental study of the interaction between the Stokes signal and the pump for different values of the detuning of the signal frequency from the central pump mode, and have compared the experimental results with the theory developed for the particular experimental situation.

THEORY

The complex amplitudes of the multimode pump field and the Stokes wave

$$\mathscr{E}_p = A_p \exp\{i(\omega_p t - k_p z)\} + \text{c.c.}$$
$$\mathscr{E}_s = A_s \exp\{i(\omega_s t - k_s z)\} + \text{c.c.}$$

($\omega_s = \omega_p - \omega_0$, ω_0 is the molecular vibration frequency), will be written in the form

$$A_p = \sum_n A_{pn}(z) \exp\{i\Omega_p(t - z/u_p)\},$$
$$A_s = \sum_m a_{sm}(z) \exp\{i\Omega_m(t - z/u_s)\},$$

where u_p, u_s are the group velocities. For simplicity, we shall assume that the modes are equidistant, i.e., $\Omega_n = n\Omega$.

The SRS theory given below is valid for the case where the frequency separation between neighboring modes is much greater than the spontaneous Raman-scattering linewidth, i.e., $\Omega T_2 \gg 1$, where T_2 is the transverse relaxation time of the molecular vibrations. It then follows from physical considerations that the interaction between the pump modes and the Stokes waves with different mode numbers is nonresonant, and its contribution to the excitation of molecular vibrations can be neglected in comparison with the contribution of resonantly interacting modes with the same numbers. It follows that for $\Omega T_2 \gg 1$, the molecular vibrations may be approximately replaced by a single-mode wave:

$$Q = \sum_n Q_p \exp(i\Omega n t) \approx Q_0.$$

This approximation substantially simplifies the analysis and can be used to consider both linear scattering and nonlinear saturation of stimulated Raman scattering, as well as to obtain a generalization of the Bloembergen nonlinear theory of stimulated Raman scattering to the case of multimode fields. In the case of stimulated Raman scattering, the complex amplitudes satisfy the following equations[2]:

$$\frac{\partial A_s}{\partial z} + \frac{1}{u_s}\frac{\partial A_s}{\partial t} = \frac{1}{2} g A_p Q^*, \quad T_2 \frac{\partial Q}{\partial t} + Q = A_p A_s^*,$$

$$\frac{\partial A_p}{\partial z} + \frac{1}{u_p}\frac{\partial A_p}{\partial t} = -\frac{1}{2}\frac{\omega_p}{\omega_s} g A_s Q,$$

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where g is the gain. Bearing in mind the foregoing discussion, it follows that the mode amplitudes are described by the following set of equations[3]:

$$\frac{d}{dz}a_{sn} = -\frac{1}{2}gA_{pn}\sum_m A_{pm}^* a_{sm}\exp(i\nu\Omega(n-m)z),$$
$$\frac{d}{dz}A_{pn} = -\frac{1}{2}\frac{\omega_p}{\omega_s}ga_{sn}\sum_m A_{pm}a_{sm}^*\exp(-i\nu\Omega(n-m)z), \quad (1)$$
$$n, m = 0, \pm 1, \pm 2, \ldots, \pm N,$$

where $\nu = |1/u_s - 1/u_p|$ describes the dispersion of the group velocity. The pump spectrum is assumed symmetric and consisting of $M = 2N+1$ modes. The solution of (1) is sought for $z \geq 0$ subject to the boundary conditions $a_{sn}(z=0) = a_{sn}^0$, $A_{pn}(z=0) = A_{pn}^0$.

Let $\Delta\omega_p$ and $I_p = \sum_n |A_{pn}|^2$ be the width of the spectrum and the pump intensity, respectively. If the amplification of the Stokes wave over the coherence length $l_{coh} \sim 1/\nu\Delta\omega_p$ is large, i.e., $gI_p \gg \nu\Delta\omega_p$ or $I_p \gg I_{crit}$ ($I_{crit} = \nu\Delta\omega_p/g$), then the medium may be looked upon as nondispersive, and we may substitute $\nu = 0$ in (1).[4] This is referred to as coherent scattering. For this case, (1) yields the following expression for the intensity $I_s = \sum_n |a_{sn}(z)|^2$ of the Stokes wave in an arbitrary cross section z of the scattering region:

$$\frac{I_s}{I_{s0}} = \frac{1+\alpha}{2}\frac{(1+\beta)\gamma + (1-\beta)e^{-\beta G}}{\gamma + e^{-\beta G}}, \quad (2)$$

where

$$\alpha = \frac{\omega_s}{\omega_p}\frac{I_{p0}}{I_{s0}}, \quad \beta = \left[\left(I_{s0} - \frac{\omega_s}{\omega_p}I_{p0}\right)^2 + 4\frac{\omega_s}{\omega_p}I_{p0}I_{s0}K\right]^{1/2}\Big/\left(I_{s0} + \frac{\omega_s}{\omega_p}I_{p0}\right),$$
$$\gamma = \frac{(1+\beta) - \alpha(1-\beta)}{\alpha(1+\beta) - (1-\beta)}, \quad G = \left(I_{p0} + \frac{\omega_p}{\omega_s}I_{s0}\right)gz, \quad (3)$$
$$K = \left|\sum_n a_{sn}^{(0)}A_{pn}^{(0)*}\right|^2\Big/I_{s0}I_{p0}, \quad I_{s0,p0} = I_{s,p}(z=0).$$

Formula (2) is a generalization to the case of a multimode pump of the well-known expression for the intensity of a Stokes wave during stimulated Raman scattering of a monochromatic pump (see, for example Bloembergen[5]). The two expressions become identical in the case of a single mode ($M = 1$) and for complete phase locking of the Stokes mode with the pump at entry into the medium (in both cases, $K = \beta = 1$).

The intensity of the n-th Stokes mode is given by (1):

$$|a_{sn}|^2 = \frac{1}{2(1+\gamma_1 e^{-\beta G})}\left[1 + \frac{1-\alpha}{\beta(1+\alpha)}\right]\Big[|a_{sn}^{(0)}|^2(1+\gamma_1 e^{-\beta G/2})^2 + \frac{\omega_s}{\omega_p}|A_{pn}^{(0)}|^2\gamma_1(1-e^{-\beta G/2})^2 + 2\,\mathrm{Re}\left[a_{sn}^{(0)*}A_{pn}^{(0)}\left(\frac{c}{c^*}\right)^{1/2} \times \left(\frac{\omega_s}{\omega_p}\gamma_1\right)^{1/2}(1-e^{-\beta G/2})(1+\gamma_1 e^{-\beta G/2})\right]\Big]; \quad c = \sum_m a_{sm}^{(0)}A_{pm}^{(0)*}, \quad (4)$$

where $\gamma_1 = 1/\gamma$ and the parameters α, β, G, γ are given by (3).

It follows from (4) that even if there is no "primer" for one of the Stokes modes at entry to the medium, this mode can, nevertheless, be excited if the amplitude of the pump mode corresponding to it is nonzero. In point of fact, substituting $a_{sn}^{(0)} = 0$ ($a_{s(m\neq n)} \neq 0$) in (4), we obtain

$$|a_{sn}|^2 = \frac{\gamma_1}{2(1+\gamma_1 e^{-\beta G})}\left[1 + \frac{1-\alpha}{\beta(1+\alpha)}\right]\frac{\omega_s}{\omega_p}|A_{pn}^{(0)}|^2(1-e^{-\beta G/2})^2.$$

This excitation of Stokes modes that are absent at entry to the medium may lead to the broadening of the spectrum of a narrow-band Stokes wave up to the width of the pump spectrum over sufficiently long interaction lengths. This effect is based on the fact that a given pump mode will, even in the absence of the corresponding Stokes "primer," be scattered by molecular vibrations excited by beats between other modes of the Stokes "primer" and the pump. We may therefore conclude that the spectrum of the narrow-band Stokes signal amplified by coherent SRS will be broadened up to the width of the spectrum of the broad-band pump. Moreover, when the gain is sufficiently large, the shape of the spectrum of the Stokes signal will repeat the pump spectrum, and will cease to depend on the shape of the spectrum of the Stokes "primer" wave [for $I_{p0} \gg I_{s0}$, the only important terms in (4) are $\sim |A_{pn}|^2$].

EXPERIMENTAL METHOD

A block diagram of the apparatus is shown in Fig. 1. The two synchronized neodymium-glass lasers L-I and L-II were Q-switched with a Kerr cell and were used as the pump sources. They could be independently locked by the radiation from the low-power master laser 1 operated under free-running conditions. The optical system incorporating the pump sources and the master laser was equivalent to that used previously.[1] We recall that, in the absence of locking, the pump sources produced monopulses with energy $E \approx 1$ J, length $\tau \approx 30$ nsec, spectrum width $\Delta\nu_p \approx 25$ cm^{-1}, and Lorentz line shape. When locking was used, the spectrum width was $\Delta\nu_p < 0.1$ cm^{-1}, other pulse parameters remaining unaltered. The region of stable frequency locking extended over approximately ± 20 cm^{-1} from the center of the luminescence line of the neodymium silicate glass. The locking region could be expanded to ± 50 cm^{-1} by inserting a Lyot filter with a dispersion band of approximately 120 cm^{-1} into the resonators of

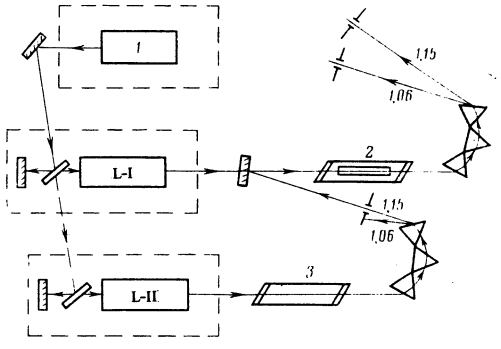
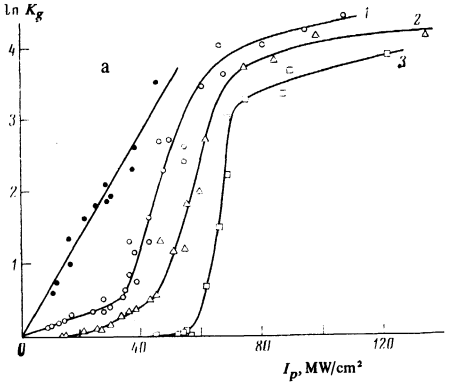


FIG. 1. Block diagram of apparatus.

the pump sources. The active material was SF_6 liquefied at 30 atm with a vibrational shift of $\Omega = 775$ cm^{-1} and SRS linewidth $\Delta\nu_l \approx 1$ cm^{-1}.[1] Radiation from L-I was used as the pump in the amplifying cell 2. Radiation from L-II was used as the pump for cell 3 to produce the Stokes signal. The Stokes radiation was then directed into the amplifying cell along the same path as the pump radiation. To prevent a reduction in the interaction length between the Stokes signal and the pump, due to misalignment of the optical elements, a metal lightguide of square cross section (4×4 mm) and 90 cm long was introduced into cell 2 (length of active region 100 cm). We measured the input and output energy of the Stokes signals, E^s_{in} and E^s_{out}, and the corresponding quantities for the pump pulses. The STÉ-I spectrograph was used at the second-harmonic frequency for the simultaneous detection of the spectra of input and output Stokes signals ($\lambda_s = 1.15$ μ) and the pump spectrum ($\lambda_p = 1.06$ μ). The simultaneous arrival of the interacting pulses in the amplifying cell 2 was monitored with the IPL-2M two-beam oscillograph.

EXPERIMENTAL RESULTS

Figure 2 shows graphs of $\ln K_g = \ln(E^s_{out}/E^s_{in})$ against the pump intensity. L-I was used without locking and the pump linewidth was $\Delta\nu_p \approx 25$ cm^{-1}, thus satisfying the conditions for broad-band scattering ($\Delta\nu_p \approx 25$ cm^{-1} $\gg \Delta\nu_l \approx 1$ cm^{-1}), whereas L-II was used with locking and the linewidth of the output Stokes signal was $\Delta\nu_s \sim \Delta\nu_p^{L\text{-}II} < 0.1$ cm^{-1}. The experiments were performed for the following cases: 1) Stokes signal in resonance with central pump modes; 2) Stokes signal in resonance with pump modes at $\Delta\Omega \approx 13$ cm^{-1} from the central modes,



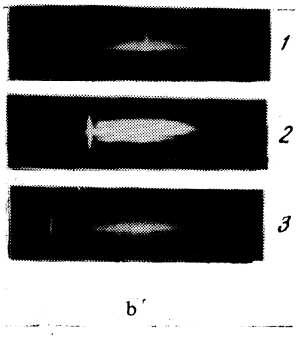


FIG. 2. a) $\ln K_g$ as a function of pump intensity: ○—$\Delta\Omega = 0$; △—$\Delta\Omega = 13$ cm^{-1}; □—$\Delta\Omega = 24$ cm^{-1}; ●—$\Delta\nu_s \approx \Delta\nu_p < 0.1$ cm^{-1}; 1, 2, 3—amplification of output Stokes signal for $I_p \gg I_{crit}$ and the corresponding spectrograms (b).

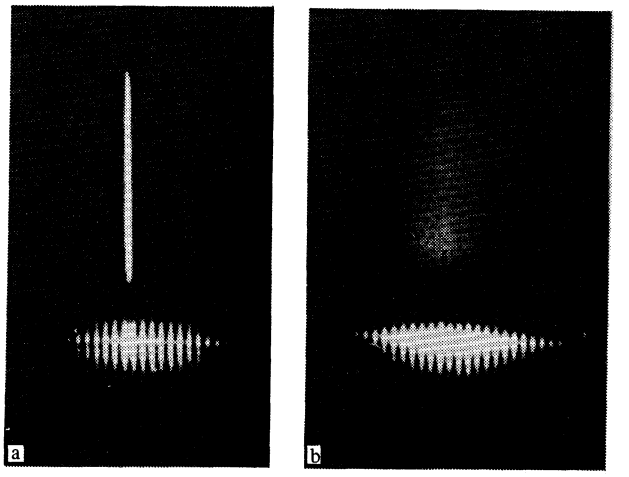


FIG. 3. Spectrograms of the input (top) and output (bottom) Stokes signals for $I_p \gg I_{crit}$: a—$\Delta\nu_s^{in} < 0.1$ cm^{-1}, b—$\Delta\nu_s^{in} \approx 25$ cm^{-1}.

and 3) $\Delta\Omega \approx 24$ cm^{-1}. For comparison, we also give $\ln K_g$ as a function of the pump intensity when both L-I and L-II were operated under locked conditions ($\Delta\nu_p < 0.1$ cm^{-1}); $\Delta\nu_s < 0.1$ cm^{-1}). It follows from this graph that the static gain was $g_{SF_6} = (7 \pm 1) \times 10^{-4}$ cm/MW.[1)] Analysis of these curves shows that, over the initial part of the curves, the rate of increase in the amplification of the narrow-band Stokes signal is determined by the intensity of the resonance pump modes but, after a certain critical value of the intensity, the curve rises steeply, approaching the static level. The shape of the amplification curves for pump intensities $I_p > 70$ MW/cm^2 corresponds to the saturation of Raman scattering [$I^s_{in} \approx 0.3$ MW/cm^2; $K_g(I_p \approx 70$ MW/cm$^2) \sim 100$]. Figure 2 also shows that, as the detuning $\Delta\Omega$ of the frequency of the Stokes signal from the central pump modes increases, the effectiveness of the interaction is appreciably reduced. Figure 2 also shows spectrograms of the output Stokes signal for different detuning and pump intensities greater than I_{crit}. It is clear that the broadening of the Stokes spectrum occurs in the direction of the strongest central modes. Analysis of spectrograms of the pump and Stokes signals for $I_p > I_{crit}$ shows that the two are identical. There is no broadening of the spectrum for $I_p < I_{crit}$.

The following experiment was carried out to determine the influence of the Stokes line shape at high gain on the spectrum of the output signal. A Lyot filter with a dispersion band $\Delta\nu \approx 2.5$ cm^{-1} was introduced into the resonator of L-I. Without locking, the spectrum of L-I was a set of equidistant lines with an overall spectrum width of about 30 cm^{-1} and linewidths < 1 cm^{-1}. L-II was operated either with locking ($\Delta\nu_s^{in} < 0.1$ cm^{-1}) or without locking ($\Delta\nu_s^{in} \approx 25$ cm^{-1}). The spectrograms of the input and output Stokes signals for both cases are shown in Fig. 3 for $i_p \gg I_{crit}$. It is clear that, when the gain is large, the spectrum of the output Stokes signal repeats the pump spectrum and is independent of the shape of the spectrum of the input signal.

We may now summarize our conclusions drawn from

the above experimental results: 1) the growth rate of the amplification of the narrow-band Stokes wave for $I_p < I_{crit}$ is proportional to the intensity of the corresponding resonance pump modes; 2) for $I_p > I_{crit}$, the growth rate of the amplification begins to increase and approaches the static value; 3) the effectiveness of the interaction decreases with increasing detuning $\Delta\Omega$; and 4) the spectrum of the output Stokes signal at high gain is independent of the position and shape of the spectrum of the input signal, and completely repeats the pump spectrum.

DISCUSSION

1. To compare the experimental results with the predictions of the above theory, let us replace the continuous pump spectrum by a set of M discrete modes separated by $\Omega(\Omega T_2 > 1)$. For the intensity of the n-th pump mode, we then have

$$|A_{pn}^{(0)}|^2 = \int_a^b I_{p0}(\omega)\,d\omega, \quad a = \left(n - \frac{1}{2}\right)\frac{3\Delta\omega_p}{M}, \quad b = \left(n + \frac{1}{2}\right)\frac{3\Delta\omega_p}{M}, \qquad (5)$$

where $I_{p0}(\omega)$ is the spectral intensity of the ingoing pump. The spectrum of the latter can be assumed to have the Lorentz form, which corresponds to the experimental situation. Moreover under our conditions, $I_{p0} = 10-130$ MW/cm^2 and $I_{p0} \sim 0.3$ MW/cm^2 so that $I_{p0} \gg \omega_p I_{s0}/\omega_s$. From (2), we then have the following expression for the case of a monochromatic Stokes "primer":

$$\frac{I_s(z)}{I_s(0)} \approx \left[\frac{\omega_p}{\omega_s}\frac{I_{s0}}{I_{p0}} + \frac{I_{p0}}{|A_{pn}^{(0)}|^2}e^{-\sigma}\right]^{-1}, \qquad (6)$$

where $A_{pn}^{(0)}$ is the amplitude of the pump mode with which the monochromatic "primer" resonates. It follows from (6) that, as the intensity of the pump mode with which the Stokes signal resonates decreases, SRS becomes less effective.

Figure 4 shows theoretical curves for the gain as a function of pump intensity for three different shifts of the frequency of the monochromatic "primer" relative to the spectrum of the multimode pump. These curves were calculated from the experimental parameters of the Stokes signal and the pump (the pump spectrum of width 25 cm^{-1} was replaced by 11 equidistant modes with frequency separation ~ 7 cm^{-1}). Comparison of the theoretical curves (Fig. 4) with experimental results (Fig. 2) shows good quantitative agreement for pump intensities greater than I_{crit}. We recall that we can neglect group delay for $I_p > I_{crit}$, and it is precisely for this case that the theory was developed.

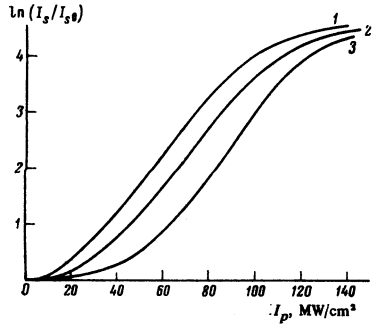


FIG. 4. Theoretical dependence of ln (I_s/I_{s0}) on the pump intensity for $\nu = 0$: 1—$\Delta\Omega = 0$; 2—$\Delta\Omega = 13$ cm^{-1}; 3—$\Delta\Omega = 24$ cm^{-1}.

We can now use (6) to determine the pump intensity difference ΔI_p^{mn} corresponding to equal values of the gain in the case of resonance between the Stokes "primer" and the n-th and m-th pump modes, i.e., the mean shift of the curves in Fig. 4 along the abscissa axis

$$\Delta I_p^{mn} = \frac{1}{zg}\ln\left(\frac{|A_{pm}^{(0)}|^2}{|A_{pn}^{(0)}|^2}\right). \qquad (7)$$

Since $z = 100$ cm, $g_{SF_6} \sim 7\times10^{-4}$ cm/MW, and $\Delta\Omega$ is 13 and 24 cm^{-1}, we find from (7) that $I_p^{013} \sim 11$ MW/cm^2 and $\Delta I_p^{024} \approx 22$ MW/cm^2. These values are in good agreement with the mean shifts of the corresponding experimental curves (Fig. 2).

2. To explain the experimental amplification curves for $I_p < I_{crit}$, we must obviously take into account the dispersion of the medium. The solution of (1) for $\nu \neq 0$ is rather laborious and inconvenient for analysis so that, for simplicity, we confine our attention to the approximation of a given pump field in the case of SRS of two-mode radiation.[2)] We also assume that the pump modes have equal intensities and the Stokes "primer" is monochromatic:

$$|A_{p1}(\omega_p - \Omega)|^2 = |A_{p2}(\omega_p + \Omega)|^2 = 1/2\, I_{p0},$$
$$I_{s0} = |a_{s2}^{(0)}(\omega_s + \Omega)|^2.$$

The total intensity of the Stokes wave is then given by (1) as follows:

$$\frac{I_s}{I_{s0}} = \frac{\exp(1/2 gI_{p0}z)}{[(gI_{p0}/4)^2 - (\nu\Omega)^2]}\left\{\left(\frac{gI_{p0}}{4}\right)\mathrm{ch}\left[2z\sqrt{\left(\frac{gI_{p0}}{4}\right)^2 - (\nu\Omega)^2}\right] - (\nu\Omega)^2\right\}. \qquad (8)$$

It is clear from this expression that, for $I_{p0} \ll 4\nu\Omega/g$, the gain growth rate is $\Gamma \approx \frac{1}{2}gI_{p0}$ and, for $I_{p0} \gg 4\nu\Omega/g$, this figure increases by a factor of two: $\Gamma \approx gI_{p0}$.

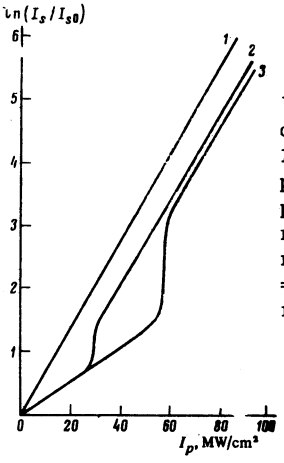


FIG. 5. Theoretical dependence of ln (I_s/I_{s0}) on the pump intensity: 1—resonance of single-mode input Stokes signal with single-mode pump; 2, 3—resonance of single-mode input Stokes signal with one mode of a two-mode pump for $\nu\Omega = 5\times10^{-3}$ cm^{-1} and $\nu\Omega = 10^{-2}$ cm^{-1}, respectively.

Figure 5 shows the gain as a function of the pump intensity. It is clear that, for $I_{p0} \ll 4\nu\Omega/g$, the only mode to be scattered is that which resonates with the Stokes "primer" mode. As the pump intensity approaches $I_p \sim I_{crit} = 4\nu\Omega/g$, the effectiveness of SRS rises sharply and the entire pump spectrum is efficiently scattered for $I_p > 4\nu\Omega/g$. Inclusion of the dispersion of the medium thus leads to the characteristic jump in the gain between noncoherent ($gI_p \ll \nu\Delta\omega_p$) and coherent ($gI_p \gg \nu\Delta\omega_p$) SRS, and this has, in fact, been observed experimentally. It follows that the critical intensity is determined exclusively by the medium and pump parameters, and is independent of the length of the wave-interaction region, so that the jump in the gain can be used to determine the dispersion of group velocity for Raman-active media. For $\Delta\nu_p = 25$ cm^{-1} and liquefied SF_6, we have (Fig. 2) $I_{crit} \approx 40$ MW/cm^2 and hence the relative dispersion is $|\nu| = \nu c = I_{crit} g/2\pi\Delta\nu_p \approx 2\times10^{-4}$. We note that, to determine $|\nu|$, we use the expression for I_{crit} given by D'yakov.[8] We also note, for comparison, that recalculation of the data reported by Carman and Mack[9] yields $|\nu| = 3\times10^{-4}$ for liquefied SF_6.

3. An important feature of the output Stokes signal is its spectrum. The prediction that the spectrum of the output Stokes signal repeats the pump spectrum at high gain [see (4) and the discussion thereafter] is in complete agreement with experiment (Fig. 3).

CONCLUSIONS

Our results have thus enabled us to explain the physical mechanism involved in the interaction between noncoherent beams during stimulated scattering. The input Stokes beam is used to form a signal whose characteristics are identical with those of the pump, and this part of it is amplified best of all. When the intensity of the exciting radiation and the length of the interaction region are large enough, the amplified Stokes signal is then found to repeat practically completely the characteristics of the pump.

The physical model used here to calculate the signal spectra provides a relatively simple and graphic description of the interaction between multimode waves, and good agreement between theory and experiment is achieved at high gain and $I_p > I_{crit}$.

[1)] In our previous work[1] we used uncollimated pump and Stokes beams for which we obtained $g_{SF_6} = (5 \pm 1)\times10^{-4}$ cm/MW because of the spatial noncoherence of the interacting waves (see, for example, Betin and Pasmanik[6]).

[2)] This case was also considered by Koroteev[7] but the dependence of the growth rate on the pump intensity was not investigated.

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