INVESTIGATION OF STIMULATED RAMAN SCATTERING OF RADIATION FROM A NEODYMIUM LASER IN LIQUID NITROGEN IN THE PRESENCE OF A RESONATOR

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Stimulated Raman scattering (SRS) in a resonator is studied theoretically and experimentally in the case of so-called nonaxial pumping, when the radiation propagates at a small angle to the resonator axis. The pumping source is a Q-switched neodymium laser (wavelength $\lambda = 1.06\ \mu$). Generation on the first two Stokes SRS components with respective wavelengths $\lambda_1 = 1.41$ and $\lambda_2 = 2.1\ \mu$ is observed. Stationary conditions for generation of two Stokes SRS components in a resonator are considered within the framework of the rate equations. It is shown that the intensity of the first Stokes component at first increases with increase of the pumping rate, reaches a peak at a pumping rate close to the threshold for production of the second Stokes component, and then begins to decrease. The results of the theoretical calculation are in satisfactory agreement with the experiments. The effect of the total line width of the pumping and its pulse duration on the SRS generation conditions in the resonator is investigated experimentally. It is found that under generation conditions the efficiency for transformation into the first Stokes component increases from 3 to 20% when the pumping line width decreases from 30 cm^{-1} to a value $\lesssim 10^{-2}$ cm^{-1}. When the duration of the pumping pulse is increased from 25 to 45 nsec the threshold value of the exciting radiation intensity (with a line width $\lesssim 10^{-2}$ cm^{-1}) correspondingly decreases from 25 to 9 MW/cm^2.

1. INTRODUCTION

It is shown in[1] that with the aid of stimulated Raman scattering (SRS) in a resonator it is possible to realize effective conversion of the laser radiation in the case when only one Stokes component is generated. In practice, if no special measures are adopted, effective generation at only one Stokes component is impossible, for when the intensity of the first Stokes component is sufficiently large, it becomes itself a pumping source for the second Stokes component, etc. When the second Stokes component appears, the first component initially remains approximately constant in a small interval of pump intensities, and then begins to decrease. This behavior of the first two Stokes components was observed by the authors in a Raman laser using liquid nitrogen and excited with a ruby laser[2].

The purpose of the present paper was a theoretical and experimental investigation of SRS in liquid nitrogen in the presence of a resonator, and also a study of the influence of the integral width of the spectral pump line and the duration of its pulse on the generation regime of the first two Stokes components. The pump source was a Q-switched neodymium laser. The Q switching was with the aid of a saturable filter or a Kerr cell, just as in[3], where the authors investigated the efficiency of conversion of SRS into Stokes components without a resonator.

2. THEORETICAL CALCULATION OF THE STATIONARY REGIME OF SIMULTANEOUS GENERATION OF TWO SRS STOKES COMPONENTS IN A RESONATOR WITH "LONGITUDINAL" PUMPING

We shall assume that the absorption of radiation by an active medium filling the resonator can be neglected, and also that the resonator mirrors are fully transparent to the pump radiation propagating parallel to the axis of the Fabry-Perot resonator. Neglecting the "spontaneous" scattering, we write the system of equations and boundary conditions in the following form[1)]:

$$\frac{dS}{dx} = -R_1 S(n_1^+ + n_1^-), \quad (1)$$

$$\frac{dn_1^\pm}{dx} = \pm\frac{R_1 S}{v\alpha_1} n_1^\pm \mp R_2(n_2^+ + n_2^-) n_1^\pm, \quad (2)$$

$$\frac{dn_2^\pm}{dx} = \pm R_2 \frac{\alpha_1}{\alpha_2}(n_1^+ + n_1^-) n_2^\pm; \quad (3)$$

$$S(0) = S_0, \quad n_i^+(0) = r_{0i} n_i^-(0), \quad n_i^-(L) = r_{Li} n_i^+(L), \quad i = 1, 2. \quad (4)$$

Here S(x) is the pump-photon flux density inside the resonator; n_i^+ (n_i^-) is the number of quanta of the i-th Stokes component per field oscillator for the wave-vector direction along the positive (negative) x axis:

$$R_i = N\frac{d\sigma_i}{dO}\Delta O_i,$$

N is the difference of the populations of the initial and final levels of the molecule, $d\sigma_i/dO$ is the differential effective cross section of the Raman scattering, ΔO_i is the solid angle of generation of the i-th SRS Stokes component,

$$\alpha_i = \frac{\omega_i^2 \Delta\omega_i}{(2\pi v)^3}\Delta O_i,$$

v is the velocity of light in the medium, $\Delta\omega_1 = \delta\omega_0 + \Gamma$, $\Delta\omega_2 = \delta\omega_1 + \Gamma$, $\delta\omega_0$ is the spectral width of the pump line, $\delta\omega_1$ is the generation line width of the first Stokes component, and Γ is the structural width of the spon-

1)We use here the notation of [1], where the regime of generation of one SRS Stokes component was investigated in detail.

taneous Raman scattering line; L is the resonator length; r_{0i} and r_{Li} are the reflection coefficients of the front and rear mirrors for the i-th Stokes component.

From (1)—(3) we can obtain four integrals:

$$n_i^+ n_i^- = b_i^2 = \text{const}, \quad i = 1, 2; \tag{5}$$

$$S_0 - S(x) = v\alpha_1[n_1^+(x) - n_1^-(x) - n_1^+(0) + n_1^-(0)] + v\alpha_2[n_2^+(x) - n_2^-(x) - n_2^+(0) + n_2^-(0)], \tag{6}$$

$$n_2^+(x) = n_2^+(0)\left[\frac{S(x)}{S_0}\right]^{-1/\gamma_2}; \quad \gamma_2 = \frac{\alpha_2 R_1}{\alpha_1 R_2}. \tag{7}$$

Taking the boundary conditions (4) into account, we obtain from (5)

$$n_i^+(0) = b_i\sqrt{r_{0i}}, \quad n_i^+(L) = b_i/\sqrt{r_{Li}},$$
$$n_i^-(0) = b_i/\sqrt{r_{0i}}, \quad n_i^-(L) = b_i\sqrt{r_{Li}}. \tag{8}$$

It follows further from (7) and (8) that

$$S(L)/S_0 = (\sqrt{r_{02}r_{L2}})^{\gamma_2}. \tag{9}$$

We introduce the following dimensionless quantities: $1 - S(L)/S_0 = \eta$ is the coefficient of pump conversion with respect to the number of photons,

$$\varepsilon = \frac{v_2\alpha_2 b_2}{v_1\alpha_1 b_1}, \quad z = \frac{S_0 R_1}{v\alpha_1}x, \quad I_0 = \frac{S_0 R_1}{v\alpha_1}L, \quad y(z) = \frac{S(z)}{S_0}. \tag{9a}$$

From the definition of ϵ and from (8) we can readily see that when $r_{01} = r_{02}$ and $r_{Li} = R_{L2}$ the quantity $\epsilon\omega_2/\omega_1$ is the ratio of the generation power of the second Stokes component to the generation power of the first Stokes component.

After certain transformations, we obtain for the dimensionless pump intensity y(z) a first-order differential equation

$$\frac{d}{dz}y(z) = -y\sqrt{4\varphi^2(\varepsilon) + \psi^2(y, \varepsilon)}, \tag{10}$$

where

$$\varphi(\varepsilon) = \frac{1 - (\sqrt{r_{02}r_{L2}})^{\gamma_2}}{[(1-r_{01})/\sqrt{r_{01}} + (1-r_{L1})/\sqrt{r_{L1}}] + \varepsilon[(1-r_{02})/\sqrt{r_{02}} + (1-r_{L2})/\sqrt{r_{L2}}]}, \tag{11}$$

$$\psi(y, \varepsilon) = 1 - y - \varphi(\varepsilon)\frac{1-r_{01}}{\sqrt{r_{01}}} - \varepsilon\varphi(\varepsilon)\frac{1-r_{02}}{\sqrt{r_{02}}} + \varepsilon\varphi(\varepsilon)[y^{1/\gamma_2}/\sqrt{r_{02}} - \sqrt{r_{02}}\,y^{-1/\gamma_2}]. \tag{12}$$

The parameter ϵ is determined from the condition $y(I_0) = (\sqrt{r_{02}r_{L2}})^{\gamma_2}$. Further, solving (10), we can calculate the intensities of the first and second Stokes components with the aid of (5)—(8). However, Eq. (10) can be integrated analytically only if $\epsilon = 0$. As can be readily seen from the definition (9a) of ϵ, this case corresponds to the threshold of simultaneous generation of two Stokes components. As a result we obtain for the threshold value of the dimensionless quantity I_0, at which simultaneous generation of two Stokes components begins,

$$I_{0\,(\text{порог})} = \frac{R_1 S_0 L}{v\alpha_1} = \frac{1}{[4\varphi^2(0) + p^2]^{1/2}} \times \ln\left|\frac{(\sqrt{r_{02}r_{L_2}})^{-\gamma_2}\sqrt{4\varphi^2(0) + [p - (r_{02}r_{L_2})^{\gamma_2/2}]^2} + \sqrt{4\varphi^2(0) + p^2}\,(r_{02}r_{L_2})^{-\gamma_2/2} - A}{\sqrt{4\varphi^2(0) + (p-1)^2} + \sqrt{4\varphi^2(0) + p^2} - A}\right|, \tag{13}$$

where

$$p = 1 - \varphi(0)\frac{1 - r_{01}}{\sqrt{r_{01}}}, \quad A = \frac{p}{\sqrt{4\varphi^2(0) + p^2}}. \tag{14}$$

Let us estimate the ratio of the generation intensity of the second Stokes component to the density of the first Stokes component. To this end we calculate the average photon density of the first Stokes component in the resonator:

$$\langle N_1\rangle = \frac{\alpha_1}{L}\int_0^L (n_1^+ + n_1^-)\,dx = \frac{\alpha_2}{R_2 L}\ln\frac{n_2^+(L)}{n_2^+(0)} = -\frac{\alpha_1}{R_1 L}\ln|(\sqrt{r_{02}r_{L2}})^{\gamma_2}|;$$

but since[2)] $n_1^+ + n_1^- \geq 2b_1$, it follows that

$$\alpha_1 b_1 < -\frac{\alpha_1}{2R_1 L}\ln|(\sqrt{r_{02}r_{L2}})^{\gamma_2}|. \tag{15}$$

It follows from (15) and (8) that the occurrence of generation at the second Stokes component limits the intensity of generation of the first Stokes component. For the photon flux of the first Stokes component emerging from the resonator we obtain the following upper estimate:

$$S_1 = v\alpha_1 b_1\left[\frac{1-r_{01}}{\sqrt{r_{01}}} + \frac{1-r_{L1}}{\sqrt{r_{L1}}}\right] < -\frac{v\alpha_1}{2R_1 L}\left[\frac{1-r_{01}}{\sqrt{r_{01}}} + \frac{1-r_{L1}}{\sqrt{r_{L1}}}\right]\ln|(\sqrt{r_{02}r_{L2}})^{\gamma_2}|. \tag{16}$$

It can be readily seen that if[3)]

$$1 - \frac{S(L)}{S_0} = 1 - (\sqrt{r_{02}r_{L2}})^{\gamma_2} \gg -\frac{v\alpha_1}{2R_1 S_0 L}\left[\frac{1-r_{01}}{\sqrt{r_{01}}} + \frac{1-r_{L1}}{\sqrt{r_{L1}}}\right]\ln(\sqrt{r_{02}r_{L2}})^{\gamma_2} \tag{17}$$

the number of photons of the second SRS Stokes component in the resonator is much larger than the number of photons of the first Stokes component. Then the efficiency of conversion into the first Stokes component decreases, and the efficiency of conversion into the second Stokes component increases, approaching its limiting value

$$\frac{\omega_2}{\omega_0}[1 - (\sqrt{r_{02}r_{L2}})^{\gamma_2}].$$

The relations obtained here describe the stationary regime of simultaneous generation of two SRS Stokes components in a resonator with "longitudinal" pumping, and make it possible to calculate the coefficient of conversion of the pump into Stokes components and the generation threshold of these components. The results of a numerical integration of (10) at the values of the parameters characterizing the experimental setup are given in Sec. 2. The system of equations (1)—(3) was obtained under the assumption that the Raman-scattering line has a rectangular form, but actually it is Lorentzian. This was taken into account in the calculation of the threshold values of the pump intensity. It is also assumed that the pump line width is much smaller than the line width of the spontaneous Raman scattering. The differential cross section of the Raman scattering in liquid nitrogen, measured by Stoicheff[4] for light with wavelength $\lambda = 0.6943\ \mu$, has been recalculated for the wavelength $\lambda = 1.06\ \mu$ and equals $d\sigma/dO = 3.0$

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2)Since $(n_i^+ - n_i^-)^2 \geqslant 0$ and $(n_i^+ - n_i^-)^2 + 4n_i^+ n_i^- \geqslant 4n_i^+ n_i^- = 4b_i^2$, as follows from (5), we find that the relation $n_i^+ + n_i^- \geqslant 2b_i$ holds for any Stokes component.

3)Condition (17) actually means that the pump intensity greatly exceeds the threshold of simultaneous generation of two Stokes components.

$\times 10^{-32}$ cm^2/sr.

The stationary regime of generation of an arbitrary number of Stokes components was analyzed on the basis of a system of equations similar to (1)—(4). Two cases should be distinguished here.

1. An even number of Stokes components is generated in the resonator. The coefficient of pump conversion does not depend on either the length of the resonator or on the pump power, and is determined completely by the reflection coefficients of the resonator mirrors, the parameters of the medium, and the spontaneous Raman scattering at the wavelengths of the Stokes components. If the pump power greatly exceeds the threshold value, then the predominant part of the generation energy at the SRS is distributed over the even Stokes components, and the intensities of all the odd components are limited.

2. An odd number of Stokes components is generated in the resonator. In this case the coefficient of pump conversion can be made as close as possible to unity by increasing the resonator length or the pump power. When the pump power is much larger than the threshold value, the main part of the SRS generation energy is distributed among the odd Stokes components, and the intensities of the even components are limited.

3. EXPERIMENTAL RESULTS

We investigated experimentally the dependence of the output energy of the first ($\lambda_1 = 1.41\ \mu$) and second ($\lambda_2 = 2.1\ \mu$) SRS Stokes components on the pump intensity ($\lambda = 1.06\ \mu$). We investigated the influence of the integral width of the spectral line ($\Delta\nu_p \lesssim 10^{-2}$ cm^{-1} and $\Delta\nu_p = 30$ cm^{-1}) and the pump pulse duration (20—50 nsec) on the generation of SRS in the resonator.

The experimental setup is shown in Fig. 1. We used so-called longitudinal non-axial pumping[2,5], in which the exciting radiation propagates at a small angle to the Fabry-Perot resonator axis. Such a method makes it possible to separate spatially the light beams of the pump and SRS generation.

The neodymium-laser resonator was Q-switched with a Kerr cell or with a saturable filter; the integral width of the spectral line was $\Delta\nu_p = 30$ and $\Delta\nu_p \lesssim 10^{-2}$ cm^{-1}. The neodymium-laser emission spectrum was investigated with the aid of an STÉ-1 spectrograph (Fig. 1) or a Fabry-Perot interferometer with an intermediate 3-cm ring; the divergence was determined from the distribution of the spot in the focal plane of the 30-cm telephoto lens (camera 1 in Fig. 1) and amounted on the average to $\approx 3 \times 10^{-3}$ rad; the lens L_1 with focal length 125 cm focused the neodymium-laser radiation in the center of the cell with liquid nitrogen. The duration and structure of the laser pulse was investigated with the aid of a coaxial photocell and an I-2-7 oscilloscope. The pump energy entering the liquid-nitrogen cell was regulated by means of filters F_1 and measured by calorimeter K_1.

The resonator was made up of a glass window W_4 and a mirror M having the following reflection coefficients:

$$r_0 = \begin{cases} 92\% & \text{at } \lambda_1 = 1.41\ \mu \\ 5\% & \text{at } \lambda_2 = 2.1\ \mu \end{cases}$$

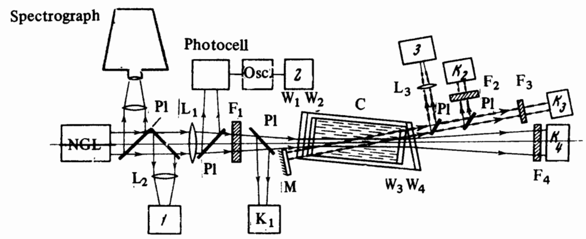

FIG. 1. Block diagram of the experimental setup for the investigation of SRS generation in a resonator with longitudinal non-axial pumping. NGL—neodymium-glass Q-switched laser; Pl—plane-parallel glass plate; L_1—lens with f = 125 cm; L_2, L_3—telephoto lenses with f = 30 cm; 1—3—photographic cameras; M—dielectric mirror with reflection coefficients $r_{01} = 92\%$ at $\lambda_1 = 1.41\ \mu$ and $r_{02} = 5\%$ at $\lambda_2 = 2.1$; C—cell with liquid nitrogen; W_1—W_4—windows of cell, the length of which is L = 20 cm; K_1—K_4—calorimeters, F_1—light filters to attenuate the neodymium-laser radiation; F_2, F_3—selective filter transmitting radiation with wavelengths $\lambda_1 = 1.41$ and $\lambda_2 = 2.1\ \mu$; F_4—filter cutting off the radiation of the neodymium-laser and transmitting the Stokes components.

The beam divergence of the SRS laser at the first Stokes component was investigated with the aid of camera 3 and a telephoto lens (f = 30 cm). The photograph of the spot and the results of its interpretation are shown in Fig. 2. The calorimeter K_2 and filter F_2 were used to measure the radiation energy at the second Stokes component ($\lambda_2 = 2.1\ \mu$), while calorimeter K_3 and filter F_3 were used to measure the generation energy at the first Stokes component ($\lambda_1 = 1.41\ \mu$). At the same time, the total energy at both Stokes components in the direct pump beam passing through the cell was measured with calorimeter K_4 with filter F_4, which cut off the neodymium-laser radiation completely.

The integral width of the spectral line of the neodymium laser Q-switched with a Kerr cell ($\Delta\nu_p = 30$ cm^{-1} remained practically constant from experiment to experiment. At the same time, when a saturable filter was used for the Q switching, the pump radiation consisted of narrow spectral lines, the number of which

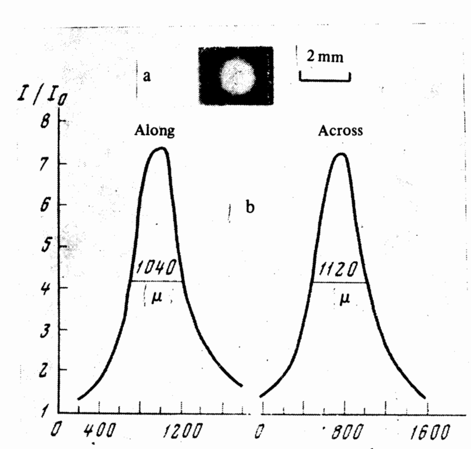

FIG. 2. Photograph of the angular distribution of the radiation of the first SRS Stokes component ($\lambda_1 = 1.41\ \mu$), obtained at the focus of a 30-cm lens (a), and the result of its photometry (b).

changed from experiment to experiment. The duration and structure of the neodymium-laser pulse also varied from experiment to experiment. In the case of Q-switching with a saturable filter, the number of the pulses also changed. Therefore, the only results used in the experimental plots were those in which one smooth bell-shaped pump pulse was obtained and one spectral component (in the case of a saturable filter) was obtained with the STE-1 spectrograph with resolution 1 Å. In spite of this, the experimental points have a large scatter. Subdivision of the points by pump-pulse duration intervals (20–30 nsec, 30–40 nsec, 40–50 nsec) greatly decreases their scatter. By way of an example, Fig. 3a shows a plot of the generation energy of the first and second SRS Stokes components in the case of narrow-band pumping[4] ($\Delta\nu_p \lesssim 10^{-2}$ cm^{-1}) for an interval of exciting-radiation pulse duration τ amounting to 30–40 nsec; Fig. 3b shows the corresponding theoretical dependence of the intensities of the first and second Stokes components in the stationary regime on the pump intensity. This plot was obtained by numerically integrating Eq. (10), with allowance for (9), for the concrete parameters of the setup used in the experiments: length of active part of the cell L = 20 cm; $\lambda = 1.06\ \mu$; $\lambda_1 = 1.41\ \mu$; $\lambda_2 = 2.1\ \mu$; $\Delta\nu_p < 0.01$ cm^{-1}; $\Delta\nu_l = 0.067$ cm^{-1}[6]; $r_{01} = 0.92$; $r_{02} = 0.05$; $r_L = 0.08$; $d\sigma/dO = 3 \times 10^{-32}$ cm^2/sr. We calculated at the same time the intensity of the Stokes components emerging from the resonator through W_4 (see Fig. 1) in the same direction in which the experimental measurements were made. Since the mirror reflection coefficient at the wavelength $\lambda_1 = 1.41\ \mu$ was 92%, practically the entire radiation at the first Stokes component was emitted to one side, but only 42% of the second component was emitted in the same direction, and the remainder passed through the mirror, the reflection coefficient of which was 5% at $\lambda_2 = 2.1\ \mu$. Thus, the maximum theoretical value of the efficiency (in both directions) is 75% at $\lambda_1 = 1.41\ \mu$ and 50% at $\lambda_2 = 2.1\ \mu$.

Figure 4 shows the experimental dependence of the efficiency of energy conversion into Stokes components on the intensity of the exciting radiation at two values of the integral width of the spectral pump line $\Delta\nu_p \lesssim 10^{-2}$ cm^{-1} and $\Delta\nu_p = 30$ cm^{-1}. It should be noted that the threshold intensity of the exciting radiation needed to produce generation at the first SRS Stokes component, is essentially different for different intervals of pump-pulse durations. The corresponding dependence is shown in Fig. 5. As already noted above, these measurements were performed simultaneously with measurements of the total energy of the Stokes components in the direct pump beam. Although qualitatively a "transfer" of the photons from the direct beam into the SRS generation beam was observed, the exact quantitative relations for this process could not be established, owing to the large scatter of the experimental points.

[4]The terms narrow band ($\Delta\nu_p \ll \Delta\nu_l$) and broad band ($\Delta\nu_p \ll \Delta\nu_l$) pumping characterize the ratio of the line with $\Delta\nu_p$ of the exciting radiation to the line width $\Delta\nu_l$ of the spontaneous Raman scattering, which amounts to 0.06 and cm^{-1} for liquid nitrogen [6].

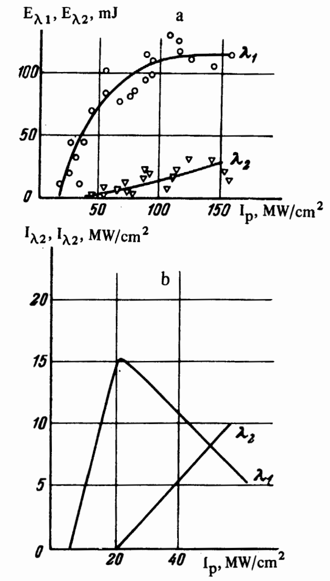

FIG. 3

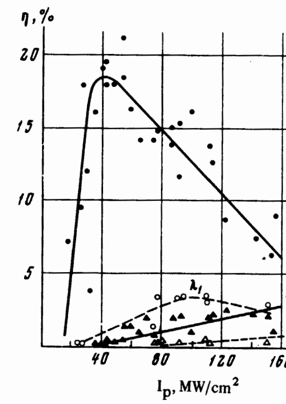

FIG. 4

FIG. 3. Yield of the emission of the Stokes SRS components in the resonator (in one direction) against the pump intensity. a) Experimental plot; E_{λ_1} and E_{λ_2}—generation energy of the first and second Stokes component, respectively; $\Delta\nu_p \leqslant 10^{-2}$ cm^{-1}; $30 \leqslant \tau \leqslant 40$ nsec. b)—Theoretical plot, I_{λ_1} and I_{λ_2}—stationary intensities of the first and second Stokes components.

FIG. 4. Experimental dependence of the efficiency of energy conversion into Stokes components on the intensity of the longitudinal pumping by a neodymium laser ($\lambda = 1.06\mu$) for an exciting-pulse duration interval 30–40 nsec. Narrow-band pumping, $\Delta\nu_p \lesssim 10^{-2}$ cm^{-1}; ●—$\lambda_1 = 1.41\mu$, ▲—$\lambda_2 = 2.1\mu$; broad-band pumping, $\Delta\nu_p = 30$ cm$^{-1} \gg \Delta\nu_e$; O—$\lambda_1 = 1.41\mu$, Δ—$\lambda_2 = 2.1\mu$.

4. DISCUSSION OF EXPERIMENTAL RESULTS

The results of the experiments show that in the presence of a resonator the efficiency of pump conversion into Stokes components of SRS is strongly dependent on the integral line width $\Delta\nu_p$ of the exciting radiation in the generation regime. It should be noted that such a dependence is characteristic precisely of SRS in a resonator. As established by the authors of[3], the SRS yield without a resonator (in the "direct" beam) is practically independent of the integral width of the spectral line of the pumping, and the conversion efficiency in this case amounts to 20–25%. The efficiency of conversion into generation in the presence of a resonator is also ~20% in the case of narrow-band pumping ($\Delta\nu_p \lesssim 10^{-2}$ cm^{-1} Fig. 4), whereas in the case of broad-band pumping ($\Delta\nu_p = 30$ cm^{-1}), the efficiency drops to ~ 3%. The reason for such a difference may lie in the dynamics of the spectral line of the neodymium laser when the latter is Q-switched with a Kerr cell. Apparently, at each instant of time the neodymium laser emits a relatively small number of narrow-band spectral components, each of which has a short lifetime. These components appear successively in different sections of the luminescence line, in analogy with the case of the free-generation regime[7]; this is

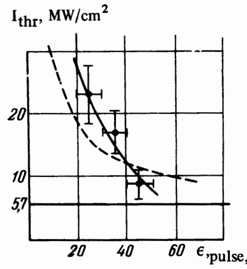

FIG. 5. Threshold pump intensity needed to produce generation at the first SRS Stokes component in the resonator vs. length of the exciting-radiation pulse: solid curve—experimental; dashed—theoretical, calculated from formula (20).

precisely the cause of the large integral width of the spectral line of the generation. In order to obtain effective conversion into SRS in the direct beam, it is sufficient that the spectral pumping line width at each instant of time be smaller than the width of the spontaneous Raman scattering line. As seen from the experimental plot (Fig. 5), reduction of the duration of the exciting pulse leads to the increase of the threshold pump intensity necessary to establish generation, for in this case the generation should develop after a smaller number of passes of light through the resonator. This reasoning is confirmed qualitatively by the experimental results, namely, as seen from Fig. 4, in the case of broad-band pumping, where the presence of short-lived narrow-band spectral components is assumed, the SRS generation threshold in the resonator is higher, and the conversion efficiency is lower.

From the theoretical results (Fig. 3b) and the experimental investigations of a liquid-nitrogen SRS laser excited with a ruby laser[2] it follows that the emission energy of the first component should decrease starting with the instant where generation occurs at the second Stokes component; the rate of growth of the energy of the second component practically coincides with the rate of growth of the first component in the initial section. On the other hand, the character of the variation of the Stokes component differs in this case from that mentioned above. First, the energy of the second Stokes component decreases smoothly to zero with decreasing pump intensity, i.e., there is no clearly pronounced threshold; second, the energy of this component increases much more slowly than the energy of the first component in the initial section; third, starting with the instant of the appearance of the second Stokes component, the energy of the first does not decrease but remains constant. This indicates that, unlike in[2], the generation at the second Stokes component does not have time to become established within the time of action of the pump pulse. In[2], the duration of the excited-radiation pulse was 70 nsec, and the reflection coefficient of the dense resonator mirror was 97% for both Stokes components. In our case the maximum pump pulse duration was ~45 nsec, and the reflection coefficients of the mirror were 92% at $\lambda_1 = 1.41\ \mu$ and 5% at $\lambda_2 = 2.1\ \mu$, i.e., the resonator for the second Stokes component was made up of surfaces with reflecting coefficients 8% and 5%, which increased still further the threshold pumping intensity needed to produce generation in the resonator at this component.

Let us estimate the dependence of the threshold pump intensity on the pulse duration. The power of the amplified spontaneous emission emerging from an active medium of length L is[8,9]

$$p^{sp} = \hbar\omega_1 \frac{S}{\lambda_1^2} dO\, \Delta\nu_l\, (K-1),$$

where $\hbar\omega_1$, λ_1, and $\Delta\nu_l$ are the quantum energy, wavelength, and line width of the spontaneous emission of the first Stokes component; S is the cross section area of the active medium; dO is the generation solid angle; $K = \exp(aIL)$ is the gain per pass, and I is the pump intensity. Inasmuch as the medium is located in a Fabry-Perot resonator with mirror reflection coefficients r_{01} and r_{L1}, it follows that after N passes of the photons in the resonator the power of the spontaneous emission reaches the level

$$P^{sp} = \hbar\omega_1 \frac{S}{\lambda_1^2} dO\, \Delta\nu_l\, (K-1) \times \frac{1-(K^2 r_{01} r_{L1})^{N/2}}{1-(K^2 r_{01} r_{L1})^{1/2}}.$$

We shall assume that the generation process is established when the spontaneous emission increases to a certain power level P_0. Since the time of establishment of the generation is very large if the self-excitation condition $K\sqrt{r_{01} r_{L1}} = 1$ is satisfied, it follows that to obtain the generation in the case of shorter pump pulses it is necessary to produce such an excitation intensity as to make $K\sqrt{r_{01} r_{L1}} \gg 1$. Then, neglecting unity in (18) and setting this expression equal to the power of the spontaneous radiation P_0 at the generation threshold, we obtain the dependence of the threshold pump intensity I_{thr} on the number of passes N:

$$\hbar\omega_1 \frac{S}{\lambda_1^2} dO\Delta\nu_l K (K^2 r_{01} r_{L1})^{(N-1)/2} = P_0.$$

We recall that here $K = \exp(aI_{thr}L)$. After simple transformations we reduce this expression to the form

$$I_{thr} = \frac{1}{aL} \ln (r_{01} r_{L1})^{-1/2} + \frac{1}{N} \frac{\ln(\sqrt{r_{01} r_{L1}}\, P_0/\hbar\omega_1 S\lambda_1^{-2} dO\, \Delta\nu_l)}{aL}. \quad (19)$$

Thus, a larger pump intensity is necessary to exceed the threshold of SRS generation in the resonator when the exciting pulse duration is decreased. In[4], the ratio $P_0/\hbar\omega_1 S\lambda_1^{-2} dO\Delta\nu_l$ was measured for the case of excitation of liquid nitrogen by a ruby laser. In the estimates, we shall use the value $\sim 10^8$ given in[4] for this ratio. The other constants in (19) are as follows: $a = 0.83 \times 10^{-2}$ cm/MW[4] (recalculated to $\lambda = 1.06\ \mu$ and $\lambda_1 = 1.41\ \mu$); $r_{01} = 0.92$, $r_{L1} = 0.08$; $N = \tau v/l$, where l is the resonator length (60 cm); $v = 2.5 \times 10^{10}$ cm/sec—velocity of light in the medium. Substituting these figures in (19) we obtain (τ is in nsec)

$$I_{thr}[\text{MW/cm}^2] = 5.7 + \frac{250}{\tau}. \quad (20)$$

The theoretical plot is shown in Fig. 5 by the dashed line. It gives a qualitatively correct description of the experimental curve; the observed quantitative discrepancy is apparently connected with the large experimental errors in the determination of the threshold pump intensity.

It should be noted that the theoretical and experi-

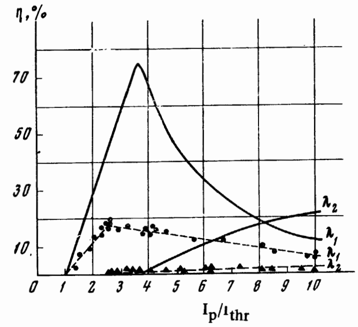

FIG. 6. Experimental ($\Delta\nu_p \lesssim 10^{-2}$ cm^{-1}, dashed curves) and theoretical (solid curves) plots of the conversion efficiency (in one direction) into the first and second SRS Stokes components in a resonator vs. the excess of the pump over threshold. The theoretical value of the threshold is I_{thr} = 5.7 MW/cm²; the experimental value is I_{thr} = 17 MW/cm².

mental plots of the efficiency of conversion into Stokes components on the excess of the pump intensity I_p over its threshold value I_{thr} have practically an identical character (Fig. 6). In the case of an approximately threefold increase over the threshold of SRS generation in the resonator, the second Stokes component appears, and the efficiency of conversion into the first component begins to decrease from that instant on. As follows from Fig. 6, and also from the results of[2], this behavior is typical of SRS in a resonator with longitudinal non-axial pumping in the case of simultaneous generation of two Stokes components. The experimental value of the efficiency of pump conversion into the first Stokes component (at the maximum) is smaller by a factor 3.5 than the theoretical value. This is apparently connected with the fact that no account was taken in the theoretical calculations of the radiation loss inside the resonator (scattering, absorption, for example by the impurities of carbon dioxide in the liquid nitrogen, etc.), and also the presence of many oscillation modes. In addition, the stationary regime of SRS was not reached in the experiment, as is evidenced, particularly, by the appreciable conversion of the pump in the Stokes component in the case of the direct beam (see Sec. 2).

5. CONCLUSIONS

A. The theoretical model developed within the framework of the rate equations for the simultaneous generation of two Stokes components gives a correct qualitative description of SRS in a resonator with longitudinal pumping. The quantitative disparity between the theoretical and experimental results is most probably connected with the nonstationary character of the SRS generation and with the presence of many oscillation modes.

B. The efficiency of conversion in the case of generation decreases with increasing integral width of the spectral pump line. Knowledge of the integral width of the spectral pump line is insufficient for an unambiguous estimate of the gain of the Stokes component of the SRS and of the associated conversion efficiency. It is necessary also to have information on the temporal structure of the spectrum.

C. The threshold pump intensity necessary for SRS generation to develop in the resonator increases with increasing duration of the exciting-radiation pulse. To attain a stationary SRS generation regime at a resonator length l = 60 cm the lengths of the exciting pulses must be not smaller than 250 nsec.

D. In the case of simultaneous generation of two Stokes components and longitudinal excitation of the SRS in the resonator, there is an optimal excess of pump intensity (over its threshold value), at which the efficiency of conversion into the first Stokes components reaches a maximum value.

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187