Statistical phenomena in Raman scattering stimulated by a broad-band pump

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The results of a theoretical and experimental investigation of stimulated scattering in the field of a noise pump are presented. A complete theoretical picture of the phenomenon is given in the approximation of a prescribed Gaussian noise-pump field. The coherent and noncoherent scattering regimes and the region where transitions between them occur are investigated in detail. The increments, correlation functions, and Stokes-radiation and optical-phonon spectra are computed for arbitrary relations between the pump-correlation time τ_{cor}, the dephasing time T_2, and the characteristic group-delay time T_3. It is shown that in many cases of practical interest a noise pump can be at least as effective as a harmonic pump of the same mean intensity. The feasibility of generation of highly monochromatic optical phonons [spectral linewidth $\Delta\nu_Q \ll \Delta\nu_0 = (\pi c T_2)^{-1}$] in an optical-noise field is noted. The theoretical results have been experimentally verified by investigating SRS in liquid nitrogen excited by a broad-band optical-noise source. The possibility of realizing effective scattering in an essentially nonquasistatic scattering regime (for $\tau_{cor} \ll T_2$ and $\tau_{cor} \ll T_3$) was demonstrated; spectral and energy measurements were performed and are compared with the theory. The obtained results can be used to analyze different types of scattering and such problems as decay instabilities in a plasma, etc.

1. INTRODUCTION

An important section of the theory of nonlinear wave phenomena in dispersive media is the study of the dynamics of the parametric effects and stimulated scattering (decay interactions) in a modulated-pump field. The investigations carried out in recent years and pertaining primarily to nonlinear optics have shown that allowance for the modulation of the pump leads not only to quantitative, but also to qualitative, changes in the results of the theory developed back in 1962—1964 of the parametric interactions of unmodulated waves [1].

Undoubtedly, the most interesting of the new effects are the ones connected with the temporal modulation of the pump. The nonstationary broadening of the spectrum of stimulated scattering in the field of short—compared to the transverse relaxation time—pump pulses, the appearance of pulsed-pump excited stationary modes in stimulated scattering and in parametric amplification, the suppression of the parametric effects owing to the phase modulation of the pump—these are a few of the new effects predicted here theoretically and experimentally observed [2].

The majority of the enumerated time effects have, generally speaking, space analogs (see [3]); it should be noted, moreover, that in practical situations (especially in isotropic media) allowance for the spatial modulation of the pump often can be performed quasistatically, averaging the results of the theory for plane waves over the profile of a real pump beam [4].

The next natural step in the study of the dynamics of decay interactions is the generalization of the theory to the case of the stochastic pump [5]. As applied to stimulated scattering (to which the present paper is primarily devoted), the study of the statistical phenomena due to the stochastic modulation of the pump is of interest from several standpoints.

1. By studying the statistics of the scattered radiation, we can obtain information about the statistics of the pump—in particular, about its higher-order correlation functions.

2. Of fundamental importance is the question of the effectiveness of the noise pump in a nonlinear process. There are two aspects here. On the one hand, of interest is the realization of conditions under which the effectiveness of the noise pump approximates that of a harmonic pump of the same power [2,6,7,24].

Such a formulation of the problem is typical for nonlinear optics, in particular, for the optics of the ultraviolet and x-ray bands, where the construction of highly monochromatic sources is at present meeting with considerable difficulties. On the other hand, as applied to such problems as the laser heating of plasmas, the propagation of radio waves in the ionosphere, etc., parametric processes and stimulated scattering are the causes of the harmful (decay) instabilities of a high-intensity wave [8]. In this connection, it is of interest to find ways of modulating a high-intensity wave in such a manner as to facilitate its stabilization; as will be shown below, noise modulation proves to be one of the promising methods. The present paper contains the results of a theoretical and experimental investigation of the statistics of stimulated Raman scattering (SRS) excited by a noise pump.

In the theoretical part, we attempt to give in the approximation of a prescribed pump field a complete picture of the phenomenon. It should be noted that the nonexistence of the general solutions of the SRS equations for the most interesting case when there develops a lag in the molecular vibrations at the same time as the medium becomes dispersive compels us to resort to such methods of deriving equations for mean quantities as the Fokker-Planck approximation and the Dyson-equation technique. In the experimental part we describe the results of experiments on SRS excitation in liquid nitrogen by optical noise produced by a dye-based superradiant source. The elimination of such competing processes as self-focusing and stimulated Mandel'shtam-Brillouin scattering allows us to carry out a quantitative comparison of the experimental data with the results of the theory developed.

2. THE THEORY OF SRS IN THE FIELD OF A NOISE PUMP

In the given-field approximation (we also neglect population movements), SRS is described by two equations for the off-diagonal element of the density matrix Q and the complex amplitude A_s of the Stokes wave:

$$e_s\frac{\partial A_s}{\partial z}+\frac{1}{u_s}\frac{\partial A_s}{\partial t}+\delta_s A_s=\sigma_1 A_p(\theta)Q^*, \tag{1}$$

$$\frac{\partial Q}{\partial t}+\frac{Q}{T_2}=\sigma_2 A_p(\theta)A_s^*+N(\theta,z). \tag{2}$$

Here u_s is the group velocity, δ_s is the damping constant of the Stokes wave, σ_1 and σ_2 are coupling constants, $\theta = t - z/u_p$ is the running time connected with the pump, and T_2 is the transverse relaxation time. In Eq. (1), $e_s = +1$ if the Stokes wave and the pumping wave are codirectional, and $e_s = -1$ if they propagate in opposite directions. In Eq. (2), $N(\theta, z)$ is the stochastic force describing the intrinsic noise of the medium.

The nature of SRS in the field of a noise pump in a medium of characteristic dimension l is determined by the relation between the correlation time τ_{cor} of the pump, on the one hand, and the transverse relaxation time T_2 and the characteristic group-lag time T_3, on the other. For co-moving waves

$$T_3=l\left(\frac{1}{u_p}-\frac{1}{u_s}\right),$$

while for opposing waves

$$T_3=l\left(\frac{1}{u_p}+\frac{1}{u_s}\right)\approx\frac{2l}{c}.$$

Comparing the indicated characteristic times, we are able to distinguish four characteristic stimulated-scattering regimes, which are all of practical interest.

1. $\tau_{cor} > T_2, T_3$: the quasistatic regime. The molecular vibrations are able to follow the fluctuations in the pump; the dispersiveness of the medium is not manifested.

2. $T_2 < \tau_{cor} < T_3$. SRS in a dispersive medium with broad Raman lines. Such a situation is realized in, for example, experimental investigations of SRS in certain liquids.

3. $T_2 > \tau_{cor} > T_3$. Nonstationary SRS in a nondispersive medium. A fairly typical case of such scattering is forward scattering in gases. In condensed media this regime is usually realized when the scattering is observed in focused beams ($l \approx L_f$ is the focal length of the lens).

4. $\tau_{cor} < T_2, T_3$: there appears a lag in the molecular vibrations and the medium becomes dispersive at the same time. This is the most important regime in the investigation of SRS in large volumes.

Let us proceed to consider the above-enumerated cases.

1. The Quasistatic SRS Regime in the Field of a Noise Pump. Stochastic Instability

Under conditions when the pump can be regarded as a slow (in the time scales T_2 and T_3) function, we have from (1) and (2) for the instantaneous intensity of the Stokes wave the expression

$$I_s(z)=I_{s0}\exp(gI_pz),\quad g=2T_2\sigma_1\sigma_2. \tag{3}$$

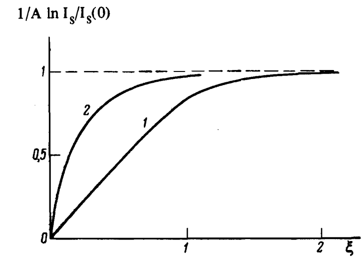

FIG. 1. The mean Stokes intensity as a function of the normalized amplification factor ξ: the curve 1 is for the harmonic pump, while the curve 2 is for a noise pump with $\tau_{cor} > T_2$, T_3 and $A_1 = 27$.

For a Gaussian pump $W(I_p) = \bar{I}_p^{-1}\exp(I_p/\bar{I}_p)$, the mean intensity of the Stokes wave is given by

$$\bar{I}_s=\int_0^\infty I_s W(I_p)\,dI_p=\frac{I_{s0}}{1-g\bar{I}_p z}. \tag{4}$$

It is evident from (4) that in the quasistatic regime the Raman amplification in the field of a noise pump can significantly exceed the amplification in the field of a harmonic pump of the same mean power. Moreover, it follows from (4) that $\bar{I}_s \to \infty$ as $g\bar{I}_p z \to 1$. This implies that the prescribed-field approximation becomes inapplicable at $g\bar{I}_p z = 1$: the Gaussian-pump excursions lead to the divergence of the moments of the Stokes intensity. To eliminate the divergences, we must take into account the back reaction of the Stokes wave on the pump; to the Eqs. (1) and (2) is added an equation for the pump. Then, instead of (3), we have

$$I_s(z)=I_{s0}\frac{e^{A\xi}}{1+e^{A(\xi-1)}},\quad A=\ln(I_{p0}/I_{s0}),\ x=gI_{p0}z,\ \xi=x/A. \tag{4a}$$

and for the Gaussian pump we obtain

$$\bar{I}_s(z)=\int_0^\infty I_s(z)W(I_p)\,dI_p=\frac{I_{s0}}{x_1}\exp\left(A_1-\frac{1}{\xi_1}\right)\times\left\{\frac{\alpha^{1-1/x_1}-1}{1-1/x_1}+\frac{\pi}{\sin\pi(1-1/x_1)}\right\}; \tag{5}$$

$$A_1=\ln\frac{\bar{I}_{p0}}{I_{s0}},\quad x_1=g\bar{I}_{p0}z,\quad \xi_1=\frac{x_1}{A_1},\quad \alpha=\frac{I_{s0}}{\bar{I}_{p0}}. \tag{6}$$

In Fig. 1 we present graphs characterizing (on a logarithmic scale) the growth of the mean intensity of the Stokes component for a noise pump and a harmonic pump of the same power. It can be seen that the growth for the noise pump is considerably more rapid. The experimentally realized excess of the Stokes-component growth rate over the rate computed for the harmonic pump is usually attributed to a positive feed back. In the present case the distinctive "stochastic instability" is, of course, not connected with an absolute instability; it is due to the high sensitivity of the exponentially growing process to the excursions of the Gaussian pump. Of course, for the observation of the mean quantities described by the formula (5) the observation time T_{obs} must be much longer than τ_{cor} (i.e., we must have $T_{obs} \gg \tau_{cor}$). Using (3), we can also easily compute the Stokes-wave and phonon spectral widths.

2. SRS in a Dispersive Medium with Broad Raman Lines

Since in this case $\tau_{cor} > T_2$, the derivative $\partial Q/\partial t$ in Eq. (2) can be neglected. Being primarily interested in the growth rate of the Stokes wave, we set $N(\theta, z) = 0$ and $A_s(0, t) = A_{s0}(t)$. Then from (1), for the co-moving waves, we have

$$A_s(t,z)=A_{s0}\left(t-\frac{z}{u_s}\right)\exp\left\{-\delta_s z+\frac{1}{2}g\int_0^z I_p\left(t-\frac{z}{u_s}-\mu z'\right)dz'\right\}, \tag{7}$$

where $I_p = |A_p|^2$ and $\mu = 1/\mu_p - 1/u_s$ is the detuning of the group velocities.

Representing I_p in the form

$$I_p = \bar{I}_p + \tilde{I}_p, \quad \langle \tilde{I}_p \rangle = 0, \tag{8}$$

we can easily verify that the amplification of the Stokes component in the field of the noise pump exceeds the amplification in the field of a harmonic pump of intensity $\bar{I}_p$ by the factor F(t, z), where

$$F(t,z) = \frac{1}{2} g \int_0^z \tilde{I}_p \left(t - \frac{z}{u_s} - \mu z' \right) dz'. \tag{9}$$

In virtue of (9), the fluctuations in the pump average out when $\tau_{cor} \ll T_3 = \mu z$; therefore, as the pump spectrum broadens (under conditions when $\tau_{cor} > T_2$), the increment tends to the static increment, which is determined by the mean pump intensity $\Gamma_0 = g\bar{I}_p$.

The computation of the increment for $\tau_{cor} < T_3$ can be performed under the assumption that the fluctuations in the pump in this case are δ-correlated:

$$\langle \tilde{I}_p(t) \tilde{I}_p(t+\tau) \rangle = \frac{\bar{I}_p^2}{\pi c \Delta\nu_p} \delta(\tau), \tag{10}$$

where $\Delta\nu_p$ is the spectral width of the pump (in cm^{-1}), while the function F is a standard random process (the standardization, when $\tau_p < T_3$, occurs owing to the integration, so that F is a Gaussian process independent of the pump distribution). Using the above-indicated circumstance, we can determine the mean intensity and the correlation function (and, consequently, the spectrum) of the Stokes wave.

If $A(t) = A_0 e^{i\Omega t}$ when z = 0, then

$$\bar{I}_s(z) = |A_0|^2 \exp[(\Gamma_n - 2\delta) z], \tag{11a}$$

where the increment in the field of the noise pump

$$\Gamma_n = \Gamma_0 (1 + \Gamma_0 L_{coh}/2\pi) = \Gamma_0 + 2\Gamma', \quad \Gamma_0 = g\bar{I}_p, \tag{11b}$$

$L_{coh} = (c|\mu|\Delta\nu_p)^{-1}$ being the coherence length. It follows from (11b) that the increment in the noise-pump field exceeds the static value Γ_0, which is determined by the mean intensity of the pump; the excess is determined by the value of the amplification over the coherence length. Since the coherence lengths in the forward and backward directions are different, the latter circumstance leads to an asymmetry in the scattering indicatrix. Moreover, $L_{coh} \to 0$ and $\Gamma_n \to \Gamma_0$ as $\Delta\nu_p \to \infty$ independent of the scattering direction. This is the fundamental difference between the noise pump and the regular pulsed pump, where the shortening of the pulse duration (the broadening of the spectrum) leads to a sharp increase in the asymmetry of the scattering indicatrix (see[3]). This is one of the examples of situations in which the properties of SRS can be used to draw conclusions about the statistics of the envelope of the pump.

For the spectral width of the Stokes component we obtain

$$\Delta\nu_s(z) = \frac{e^{\Gamma' z} - 1}{e^{\Gamma' z} - 1 - \Gamma' z} \frac{\Gamma'}{\pi c |\mu|}. \tag{12}$$

It follows from (12) that the spectrum of the Stokes wave narrows down with increasing z: $\Delta\nu_s/\Delta\nu_p \to \Gamma_0 L_{coh}/\pi$ as $z \to \infty$. Since $Q \approx T_2 \sigma_2 A_p A_s^*$, the spectrum of the phonon wave is broader than the spectrum of the scattered light.

3. A Noise Pump in a Nondispersive Medium with Slowly Relaxing Molecular Vibrations

If the group velocities of the pump and the Stokes wave coincide (i.e., if $u_p = u_s$), then $T_3 = 0$, the corresponding "dispersion" band $\Delta\nu_d = (\pi c T_3)^{-1} \to \infty$, and the solution to Eqs. (1)—(2) can be obtained in the form[2,7]

$$A_s(\theta, z) = \sigma_1 A_p(\theta) \int_0^\theta dt \int_0^z dz' \exp\left(-\frac{t}{T_2}\right) I_0 \left[\frac{2z'}{T_2} \int_{\theta-t}^{\theta} \Gamma_0(y)\, dy \right]^{1/2} N(\theta - t, z - z'), \tag{13}$$

where $\Gamma_0(\theta) = gI_p(\theta)$ and I_0 is the modified Bessel function. Introducing the natural assumption that

$$\langle N^*(t', z') N(t, z) \rangle = G\delta(t - t')\delta(z - z'),$$

we obtain from (13)

$$\bar{I}_s(t,z) = G\sigma_1 \bar{I}_p(\theta) \int_0^\theta \exp\left(-\frac{t}{T_2}\right) dt \int_0^z I_0^2 \left[\frac{2z'}{T_2} \int_{\theta-t}^{\theta} \Gamma_0(y)\, dy \right]^{1/2} dz'. \tag{13a}$$

The increment in the field of the noise pump is evidently determined by the argument of the Bessel function in (13a). By writing the intensity of the pump in the form (8), we can verify that the influence of the fluctuations in the pump is described by the integral:

$$Y(t, \theta) = \frac{1}{t\bar{I}_p} \int_0^t \tilde{I}_p(\theta' - \theta)\, d\theta'. \tag{14}$$

The upper limit of the interval of integration in (14) is equal to $g\bar{I}_p z T_2$. In the case ($\tau_{cor} < T_2$) under consideration, as a result of the averaging due to the integration, the quantity Y is small and

$$\bar{I}_s = \bar{I}_{s0} \exp(g\bar{I}_p z) = I_{s0} \exp(\Gamma_0 z).$$

Thus, a broad-band noise pump turns out to be as effective as a harmonic pump of intensity equal to $\bar{I}_p$ [1)].

The spectrum of the Stokes wave has, according to (13), the same width as the spectrum of the pump. In fact, for $\tau_{cor} < T_2$ the quantity $A_s(t)$ can be represented in the form $A_s(t) = A_p(t)\Phi(t)$, where $\Phi(t)$ is a slowly (in comparison with $A_p(t)$) varying function. Therefore, to a high degree of accuracy

$$\Delta\nu_s \approx \Delta\nu_p. \tag{15}$$

Moreover, the spectral width of the phonon wave is considerably narrower. Since

$$\frac{dQ}{dt} + \frac{Q}{T_2} = \sigma_2 [\bar{I}_p + \tilde{I}_p] \Phi(t),$$

then

$$\Delta\nu_Q \cong \Delta\nu_0 (g\bar{I}_p z)^{-1/2}, \quad \Delta\nu_0 = (\pi c T_2)^{-1}.$$

4. The Noise Pump under Conditions of Simultaneous Manifestation of Molecular Relaxation and Medium Dispersiveness. Noncoherent Scattering

It is not possible in the case under consideration to solve the dynamical equations (1) and (2) exactly. Therefore, the data presented below are based on a different—stochastic—approach in which we seek at once the equations for the mean amplitudes or intensities, the correlation functions, etc.

One of the variants of the stochastic approach to the SRS equations (the so-called Fokker-Planck approximation) is based on the fact that for $L > L_{coh}$ the fluctuations in the prescribed noise pump can be assumed to be δ-correlated[9]. For $L > L_{coh}$ the correlation time of the pump is clearly much shorter than the correlation times of the Stokes wave and the phonons; there occurs a smoothing out of the waves being amplified as they move relative to the pump wave. The phase correlations between the interacting waves are largely lost: the scattering becomes noncoherent.

Another variant of the stochastic approach to the analysis of the system (1)—(2) is connected with the use of the Dyson-equation technique [10]. Here the equations for the mean quantities can be derived for an arbitrary correlation of the pump; as a result, passage to both the δ-correlated pump and the harmonic pump limits proves to be possible.

A. The characteristics of noncoherent scattering (the Fökker-Planck approximation). We shall assume, in accordance with the foregoing, that A_p is a δ-correlated Gaussian noise[2]:

$$\langle A_p(\theta)\rangle=0,\quad \langle A_p(\theta_1)A_p(\theta_2)\rangle=0,$$
$$\langle A_p(\theta_1)A_p^*(\theta_2)\rangle=\frac{S(\theta)}{c}\delta(\theta_1-\theta_2), \tag{16}$$
$$S(\theta)=\bar{I}_p(\theta)/\Delta\nu_p.$$

Assuming that the A_s—Q correlation times will be $\tau_{s,Q} \gg \tau_p$, and using (1)—(2), we can separate out those small corrections to the amplitudes that correlate with the amplitude of the pump:

$$\tilde{A}_s=\sigma_1 Q^*\int_0^\infty A_p(\theta+\mu z')dz',\quad \tilde{Q}=\sigma_2 A_s^*\int_0^\infty A_p(\theta-\tau')d\tau'. \tag{17}$$

Using (17), we can express the mixed moments of the amplitudes A_s, Q, and A_p in terms of only A and Q. In consequence, we can derive the equations for the mean intensities of the Stokes wave $\bar{I}_s = \langle A_s A_s^*\rangle$ and the molecular vibrations $\bar{W} = \langle QQ^*\rangle$:

$$\frac{\partial\bar{I}_s}{\partial z}+\frac{1}{u_c}\frac{\partial\bar{I}_s}{\partial t}+\left[2\delta_s-\frac{gS(\theta)}{2T_2c}\right]\bar{I}_s=\frac{g\omega_s S(\theta)}{\omega_Q T_2\mu'}\bar{W}, \tag{18}$$
$$\frac{\partial\bar{W}}{\partial t}+\left[\frac{2}{T_2}-\frac{gS(\theta)}{2T_2\mu'}\right]\bar{W}=\frac{\omega_Q gS(\theta)}{4\omega_s T_2 c}\bar{I}_s; \tag{19}$$

where $\omega_Q = \omega_p - \omega_s$ and $\mu' = \mu c$ is the relative dispersion of the group velocities.

In the stationary case it follows from (18)—(19) that

$$\bar{I}_s(z)=I_{s0}e^{\Gamma z},\quad \bar{W}(z)=\frac{\omega_0 gS}{8\omega_s(1-S/S_{cr})}\bar{I}_s(z), \tag{20}$$
$$\Gamma=g\bar{I}_p\frac{\Delta\nu_0}{\Delta\nu_p}\frac{\pi/2}{1-S/S_{cr}},\quad S_{cr}=\frac{4\mu'}{g}. \tag{21}$$

Thus, the increment in the noise-pump field is a nonlinear function of the intensity of the pump. The key parameter in this case turns out to be the critical value of the spectral density of the pump defined by the formula (21). For $S \ll S_{cr}$, $\Gamma = \Gamma_0\Delta\nu_0/\Delta\nu_p \ll 1$; the increment increases sharply when $S \approx S_{cr}$. The jump in the increment at $S = S_{cr}$ is connected with a corresponding decrease in the damping of the optical phonons.

According to (19), in the field of the noise pump the effective transverse relaxation time increases:

$$T_{2\,\text{eff}}=\frac{T_2}{1-S/S_{cr}}. \tag{22}$$

For the spectral width of the optical phonons we obtain, in accordance with (22), the expression $\Delta\nu_Q = \Delta\nu_0(1 - S/S_{cr})$, i.e., the quantity $\Delta\nu_Q \ll \Delta\nu_0$ for $S \to S_{cr}$.

The spectral width of the Stokes component [11]

$$\Delta\nu_c=\frac{(e^x-1)x}{e^x-1-x}\frac{1}{\pi z\mu'},$$
$$x=2\pi\Delta\nu_0 z\mu'(S/S_{cr})^2[1-S/S_{cr}]^{-1}. \tag{23}$$

It follows from (23) that as $S \to S_{cr}$ the spectrum of the Stokes component rapidly broadens. The theory developed in this section is valid so long as $\Delta\nu_s < \Delta\nu_p$, i.e., so long as $(1 - S/S_{cr})^{-1} < \Delta\nu_p/2\Delta\nu_0$. In the case when $S > S_{cr}$ (here the noncoherent scattering becomes coherent) stimulated Raman scattering cannot be considered in the framework of the Fokker-Planck approximation; this can, however, be done, using the Dyson-equation technique.

B. Coherent and noncoherent SRS (the Dyson-equation method). We shall now assume that the pump wave is a Gaussian stochastic process, but with an arbitrary correlation function $K(\tau) = \langle A_p^*(t)A_p(t + \tau)\rangle$ and spectrum

$$G(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}K(\tau)e^{i\omega\tau}d\tau.$$

In (24) we have retained only terms of definite parity with respect to A_p; it follows from the structure of Eqs. (1) and (2) for $N(\theta, z) = 0$ and the boundary condition $A_s(t, z = 0) = A_{s0}$ that A_s is an even, while Q is an odd function of A_p.

$$A_s=\sum_{n=0}^{\infty}A_{s,2n},\quad Q=\sum_{n=0}^{\infty}Q_{2n+1},$$
$$A_m,\ Q_m\sim\langle A_p(t_1z_1)A_p(t_2z_2)\ldots A_p(t_mz_m)\rangle; \tag{24}$$

The approximate representation of the amplitudes in the form of a few leading terms of the series (24) is, in the case of SRS, ineffective: even the steady-state solution $A_s(z) = A_{s0}\exp(gI_pz)$, in which usually $gI_pz \approx 10$—25, cannot be well approximated in this way. Another method of estimating the amplitudes consists in separating out from the series (24) certain infinite subsequences that are exactly summable. Such an approach is analogous to the method employed in, for example, the theory of multiple scattering [12], and is connected with the derivation of the so-called Dyson equations for the mean amplitudes or the Bethe-Salpeter equations for the correlation functions. The Dyson-equation method can, in principle, be developed for application to linear equations of the type (1) and (2) [13], as well as to nonlinear equations that take saturation into account [10,14]. The representation in the form of a finite series in A_p is then used not directly to determine, for example, $\bar{A}_s$ or $\bar{I}_s = \langle A_sA_s^*\rangle$, but to compute approximately the coefficients of those equations which these mean quantities satisfy. In the first approximation—it is sometimes called the Bourret approximation [12]—into the coefficients of the equations for the mean quantities enter only quantities that are of second order in A_p, i.e., certain linear functionals of the correlation function $K(\tau)$.

The equation for the mean amplitude of the Stokes wave in a medium without losses has, in this approximation, the form[3]:

$$\frac{\partial\bar{A}_s}{\partial z}=\bar{A}_s\frac{g}{2}\int_{-\infty}^{\infty}\frac{G(\omega)d\omega}{1+\omega^2T_2^2} \tag{25}$$

and describes both the coherent ($\tau_{cor} > T_2, T_3$) and the noncoherent ($\tau_{cor} < T_2, T_3$) SRS regimes if

$$\frac{\Gamma_0}{2(1+\Delta\nu_p/\Delta\nu_0)}\frac{1-\exp(-2\pi\Delta\nu_p\mu' z)}{2\pi\Delta\nu_p\mu'}\ll 1.$$

The fulfillment of the last condition is necessary if A_p is a Gaussian stochastic process.

If the complex amplitude of the pump contains only one stochastic parameter—a diffusing phase—i.e., if

$$A_p(t)\sim\exp\left\{i\int_{-\infty}^{t}\xi(t')dt'\right\},\quad \langle\xi(t)\xi(t+\tau)\rangle=D\delta(\tau),$$

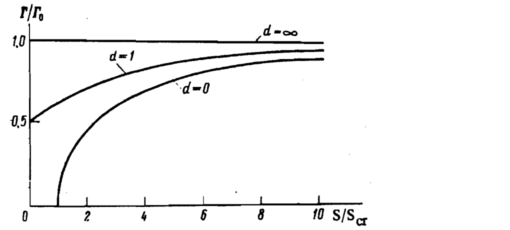

FIG. 2. The dependence of the increment of the noise-pump excited SRS on the reduced spectral density of the pump. The curve parameter $d = \Delta\nu_0/\Delta\nu_p$ is the ratio of the spontaneous Raman line width to the spectral width of the pump.

then it can be shown that Eq. (25) is exact.

The estimation of the increment Γ for the mean intensity $\bar{I}_p(z)$ is more complicated and shows that Γ satisfies the following transcendental equation:

$$\Gamma=g\left(\frac{1}{T_2}+\frac{\Gamma}{2\mu}\right)\int_0^\infty \exp\left[-\left(\frac{1}{T_2}+\frac{\Gamma}{2\mu}\right)\tau\right]K(\tau)\,d\tau.$$

For a Lorentz pump spectrum this equation goes over into a quadratic equation and determines two values for the increment ($d = \Delta\nu_0/\Delta\nu_p$):

$$\frac{\Gamma_{1,2}}{\Gamma_0}=\frac{1}{2}\left[1-(1+d)\frac{S_{cr}}{S}\right]\pm\left\{\frac{1}{4}\left[1-(1+d)\frac{S_{cr}}{S}\right]^2+\frac{S_{cr}}{S}d\right\}^{1/2}. \quad (26)$$

The plot of Γ_1 as a function of the relative spectral intensity of the pump is shown in Fig. 2. Notice that according to (26) for $\Delta\omega_0 \ll \Delta\omega_p$

$$\Gamma_1\approx\Gamma_0-\Gamma_{cr},\qquad \Gamma_{cr}=gS_{cr}\Delta\nu_p=4\mu'\Delta\nu_p. \quad (27)$$

Allowance for saturation leads to the nonlinear equation for $\bar{A}_s$:

$$\frac{\partial\bar{A}_s}{\partial z}=\bar{A}_s\frac{g}{2}\int_{-\infty}^{\infty}\frac{G(\omega)}{1+\omega^2T_2^2}\exp\left[-\frac{\omega_p}{\omega_s}\frac{g}{1+\omega^2T_2^2}\int_0^z|\bar{A}_s|^2dz'\right]d\omega,$$

from which follows, in particular, the possibility of a complete transfer of the energy of the broad pump line to the narrow SRS line in a highly dispersive medium [10,14].

C. Noncoherent scattering in focused beams. The specific distinctive feature that essentially distinguishes noncoherent scattering from scattering in the field of a harmonic pump turns out to be the strong influence that conditions introduced by the focusing have on the scattering threshold. Let us recall that in the first approximation the total amplification (and, consequently, the scattering threshold) produced by a focused harmonic pump in a medium of dimensions $z \gg L_f$, the lengthwise dimension of the focal spot, does not depend on the focal length f of the lens focusing the pump[4)]. In fact, since the focal-spot radius $r_f \approx \lambda_p f/\pi r$, $L_f \approx \lambda_p(f/r)^2$, r being the initial radius of the beam, the total amplification

$$G=gI_pL_f=g\frac{P_p}{\pi r_f^2}L_f=gP_p\pi\lambda_p^{-1}.$$

Let us now estimate the SRS threshold for a focused noise pump. The spectral density of the pump at the focus is equal to $S_f = (r/r_f)^2S$. There is a jump in the amplification at $S_f = S_{cr}$, i.e., at a pump power of

$$P_{cr}=\pi r_f^2S_{cr}\Delta\nu_p=\frac{4\lambda_p^2f^2\Delta\nu_p\mu'}{\pi r^2g}. \quad (28)$$

Since $\Gamma < \Gamma_0$, the threshold P^n_{thr} for the noise pump generally speaking exceeds the threshold P^h_{thr} for the harmonic pump, but they could be close.

If $P_{cr} > P^h_{thr}$, then the value of the threshold in the case of the noise pump depends on both f and f^2.

We can, in accordance with the foregoing, write that

$$P^n_{thr}/P^h_{thr}=\begin{cases}1 & \text{for } f<f',\\ (f/f')^2 & \text{for } f>f',\end{cases} \quad (29)$$

$$f'=\frac{r}{2}\left(\frac{G_{thr}}{\lambda_p\Delta\nu_p\mu'}\right)^{1/2} \quad (30)$$

while G_{thr} is the total amplification produced by the focused harmonic pump and corresponding to the SRS threshold.

3. THE EXPERIMENTAL RESULTS

1. Apparatus. Optical-Noise Generator

In our experiments (a block diagram of the setup is shown in Fig. 3) the broad-band pump source was the radiation of a rhodamine-6G-dye laser operating in the two-pass superradiance regime. The advantages of such an optical-noise generation scheme are a relatively high coefficient of conversion of the pump into superradiance and the simultaneous generation of a broad frequency spectrum (see [16]). The cuvette with the dye was pumped by the second harmonic of a single-mode neodymium glass laser ($\lambda = 0.53\ \mu$). The pump energy was 0.18 J and the pulse duration was 20 nsec. The concentration of the rhodamine 6G was 2×10^{16} cm^{-3}; as the solvent we used ethyl alcohol. The line width $\Delta\nu_p$ of the dye's radiation was then equal to 250 cm^{-1}, and the wavelength at the generation line center was $\lambda_p = 0.5625\ \mu$.

The length of the dye-filled cylindrical cuvette, which was equal to 120 mm, corresponded to the length of the confocal parameter of the lens L_1, which focused the second-harmonic radiation of the neodymium laser. Such coordination allowed the attainment of the minimum possible divergence (1.2×10^{-3} rad) of the dye's radiation. The angle between the directions of the forward and backward superradiant beams was $<3°$. The cuvette with the dye had a Brewster window. The maximum superluminescence energy was 0.03 J. The simultaneity of the generation of the entire frequency spectrum was specially monitored. The superluminescent radiation was led to an ISP-51 spectrograph with a UF-85 cham-

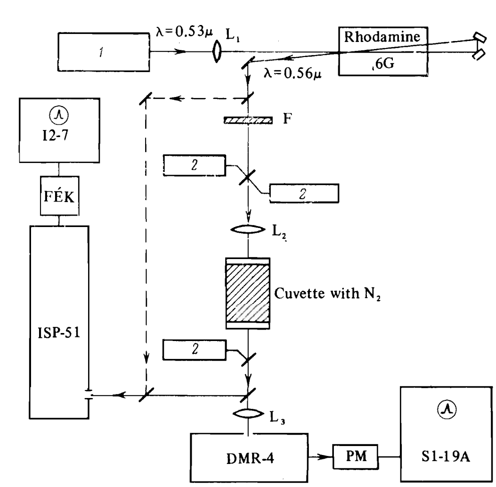

FIG. 3. Block diagram of the experimental setup used to investigate SRS excitation in liquid nitrogen by optical noise: 1) pump generator, 2) calorimeters, ISP-51) spectrograph, FÉK) coaxial photocell, DMR-4) double monochromator, I2-7 and S1-19A) oscillographs, $L_{1,2,3}$) lenses.

ber. At the exit of the chamber was placed a slit that separated out different sections corresponding to different frequency components of the generated spectrum of the dye. After passing through the exit slit, the radiation was recorded by an FÉK-09 coaxial photocell and an I2-7 oscillograph. The measured pulse widths of the individual frequency components were compared with the pulse width of the radiation of the entire superluminescence spectrum. The equality of the measured widths (25 nsec) attested to the fact that the entire spectrum was generated at the same time.

The beam obtained from the dye was focused by the lens L_2 (see Fig. 3) on a spot inside the cuvette with the liquid nitrogen (length 25 cm). The energy of the first Stokes SRS component was measured with the aid of a circuit consisting of a DMR-4 double monochromator, an FÉU-22 photomultiplier, and an S1-19A oscillograph. Energies exceeding 10^{-4} J were recorded directly with the aid of calorimeters. The spectral compositions of the exciting and scattered light beams were monitored on the ISP-51 spectrograph with the UF-85 chamber. The shapes and widths of the pump and Stokes-component pulses were recorded by the FÉK-09 photocell and the I2-7 oscillograph.

A. The characteristics of liquid nitrogen. Liquid nitrogen was chosen as the dispersive medium for several reasons. First, the Raman amplification in it is relatively high (for $\lambda_p = 0.56\ \mu$ and a vibrational frequency of $\nu_Q = 2326.5\ \text{cm}^{-1}$, $g = 2.15 \times 10^{-2}$ cm/MW). Moreover, the high thresholds for such other nonlinear processes as self-focusing and stimulated Mandel'shtam-Brillouin scattering allows us to carry out a quantitative investigation of SRS. The line width of the spontaneous scattering in nitrogen is equal to $\Delta\nu_0 = 0.067\ \text{cm}^{-1}$, so that $\Delta\nu_p \gg \Delta\nu_0$ in our experiments. The relative dispersion of the group velocities in the wave band under consideration is $\mu' = c\mu \approx 6 \times 10^{-3}$; therefore, for the optical-noise source used in the experiment the coherence length for codirectional waves was $L_{coh} = 0.67$ cm, while for opposing waves $L_{coh} = 2 \times 10^{-3}$ cm. Consequently, for the critical intensity values $I_{cr} = S_{cr}\Delta\nu_p$ we have

$$I_{cr} = 280\ \text{MW/cm}^2\ \text{(for forward scattering)},$$

$$I_{cr} = 93\ \text{GW/cm}^2\ \text{(for back scattering)}.$$

The latter value was unattainable in our experiment.

2. The Experimental Data

The experiments described below were performed with focused beams (it was not possible to excite SRS under our conditions with an unfocused noise pump). Lenses of focal lengths f = 9–60 cm were used. The dimensions of the focal regions for these lenses were experimentally determined. The spatial distribution of the radiation was photographically recorded, after which microphotometric scanning was carried out. The experimental data were compared with the theoretical results. Since the effective nonlinear-interaction length L in the focused beam for SRS was close to the dimensions L_f of the focal region, the variation of the focal length of the lens was equivalent to the variation of L. The relation between L and L_{coh} was varied in our experiments in precisely this manner.

We measured: a) the dependence of the SRS threshold on the focal length of the lens; b) the energy characteristics of the scattering; c) the asymmetry in the Stokes scattering (at angles of 0 and 180° to the pump beam); d) the spectra of the Stokes components under different conditions. In all the cases analogous measurements were carried out with a normal pump at the same time as the measurements with the noise pump were performed; as a normal pump we used the second harmonic of the neodymium laser ($\lambda_p = 0.53\ \mu$; $\Delta\nu_p \approx 0.01\ \text{cm}^{-1}$).

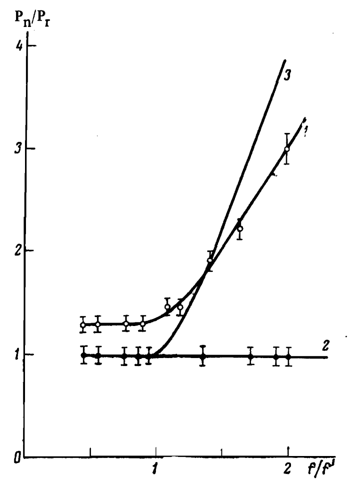

FIG. 4

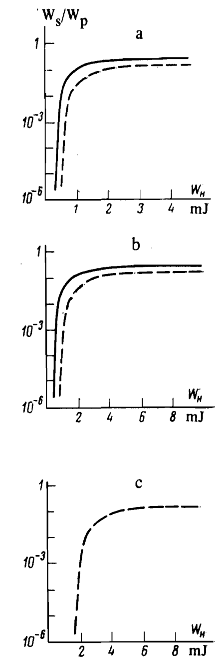

FIG. 5

FIG. 4. Threshold pump powers corresponding to SRS excitation in liquid nitrogen as a function of the focal length of the lens focusing the pump. The curve 1 corresponds to the noise pump, the curve 2 to the normal pump; the curve 3 was computed from the formula (27). The threshold pump powers are normalized to the value P_h of the threshold for the harmonic pump, while the focal length is normalized to the value f′ found from the formula (28).

FIG. 5. Plots of the energy of the first Stokes component as a function of the energy of the noise (dashed curves) and normal (continuous curves) pumps: a) $L \approx L_{coh}$; b) $L \gg L_{coh}$ (L = 11.2 cm); c) L = 25 cm (the scattering region encompasses the whole length of the cuvette).

Let us now turn to the discussion of the experimental data. Figure 4 shows graphs characterizing the dependence of the SRS threshold on the focal length of the lens that focuses the noise pump. Also shown in the same figure are the corresponding data for the harmonic pump. The spread of the threshold-energy values corresponding to a given f did not exceed 10%. It can be seen that the experimental data presented in this section are in good agreement with the theory developed in Sec. 2.

The experimentally determined value of f′ (to it corresponds the beginning of the rise in the curve 1) turned out under the conditions of our experiment to be equal to $f'_{exp} = 20$ cm. The calculated value $f'_{theor} = 30$ cm; the discrepancy should clearly be attributed to the aberrations that arise in the focusing of the optical noise and the errors in the data on the group-velocity dispersion. The effects of the incomplete spatial coherence of the optical noise should, apparently, also explain the appreciable discrepancy between the threshold en-

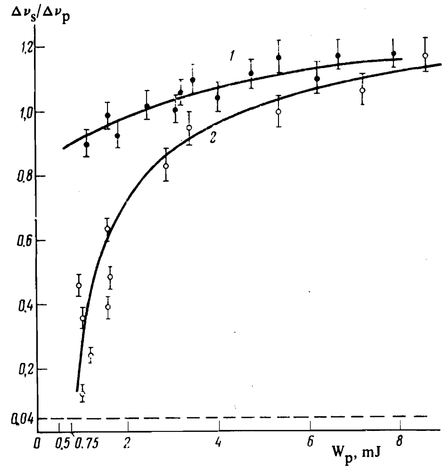

FIG. 6. The relative spectral line width of the first Stokes component excited by the noise pump as a function of the pump energy. The curve 1 corresponds to the case when $L \approx L_{coh}$, the curve 2 to the case when $L \gg L_{coh}$. The dashed line indicates the theoretical value of the minimum band width of the Stokes wave for $L \gg L_{coh}$.

ergies for the noise and harmonic pumps for $f < f'$; it should be emphasized at the same time that this discrepancy does not exceed 20%. The energy characteristics of SRS in the field of the noise pump are summarized in the table; also given in the table for comparison are the corresponding characteristics for the harmonic pump.

Two circumstances should be noted: the possibility, which follows from the table, of obtaining a high scattering efficiency in the field of a noise pump and a sharp asymmetry in the scattering indicatrix R (cf. the data given in Sec. 2). Of course, the noise-pump energies required to achieve high efficiencies turn out, generally speaking, to be higher than for the harmonic pump. A quantitative idea about this is given by the plots in Fig. 5, which show the dependence of the energy of the first Stokes component on the pump energy for the noise (for different relations between L and L_{coh}) and normal pumps.

From the experimental graphs of Fig. 5 we can find the difference between the threshold energies for the noise and harmonic pumps: $\Delta W = W_n - W_h$. The theory (see also Fig. 2) gives (see formula (27))[5)]

$$\Delta W = S_{cr} \Delta\nu_p \pi r^2_f \tau_p,$$

where τ_p is the pump-pulse duration. Using the last formula, we obtain from the graphs of Fig. 5b for the critical intensity the value $I_{cr} = 100$ MW/cm^2.

Finally, in Fig. 6 we show the plots of the dependence of the spectral width of the first Stokes component on the pump energy. It can be seen that the behavior of the curves are determined, in complete accord with the theory (cf. the data cited in Sec. 2), by the relation between L and L_{coh}. For $L \sim L_{coh}$ the quantity $\Delta\nu_s \approx \Delta\nu_p$ = const. In contrast, the sharp growth of the Stokes-line width with increasing pump energy when $L \gg L_{coh}$ is distinctly visible.

4. CONCLUSION

The experimental data indicates that the theory developed in Sec. 2 satisfactorily describes the distinctive features of SRS excited by a noise pump in the case when $L > L_{coh}$ (experimental data pertaining to the case

The energy characteristics of SRS excited in liquid nitrogen by noise and normal pumps

Spectral width of pump	Efficiency (max.)	Indicatrix asymmetry
$\Delta\nu_p = 0.01$ cm^{-1} (harmonic pump)	20 %	1
$\Delta\nu_p = 250$ cm^{-1} (noise pump)	12 %	∞

$L \leq L_{coh}$ can be found in the recent paper by Grasyuk et al.[17]). The theory expounded in Sec. 2 was developed for a Gaussian noise pump. This model is quite adequate not only for the superradiant pump source used in our experiments, but also, as analysis shows, for a multimode laser pump if the phases of the modes are not correlated and the amplification produced by an individual mode is small (if $\Gamma_{0n} z \ll 1$, where $\Gamma_{0n} = g I_{p,n}$, $I_{p,n}$ being the intensity of the individual mode).

The formulas obtained are directly applicable to SMBS (under conditions when the lifetimes of the free phonons are not long) and, with some modifications, to stimulated temperature scattering. However, speaking of SMBS with a noise pump, we should point out one important circumstance. If $\tau_{cor} < T_{ph}$, T_3 (here T_{ph} is the lifetime of a phonon), then the equations describing SMBS have the same form as (18) and (19). This means that the effective damping constant of the hypersound

$$\delta^h_{eff} = (1 - S/S_{cr})\delta^h,$$

decreases with increasing pump power. Thus, the damping of the acoustic phonons in the field of the noise pump may turn out to be so weak that it will be necessary to take the effects of the propagation into account. The noted effect may turn out to be useful in hypersound-generation technology.

Considering the obtained results in the aspects noted in the Introduction, we can assert the following.

1. The characteristic that is very sensitive to the pump statistics turns out to be the scattering indicatrix. Useful information can be obtained from the investigation of the spectrum of the Stokes component.

2. In all the regimes of practical interest the increment for the Stokes component in the case of the noise pump (at any rate for forward scattering) can be made to be equal at least to $\approx \Gamma_0$, i.e., the noise pump can be made as effective as the harmonic pump. In this case, however, the spectrum of the Stokes component usually turns out to be quite broad: $\Delta\nu_s \approx \Delta\nu_p$. To narrow down the Stokes spectrum, we can use the optical resonator. The self-excitation threshold for a Raman laser of length L with a noise pump turns out in this case to be equal to

$$I_{thr} = \frac{2(1-R)}{gL}\left(1 + \frac{\Delta\nu_p}{\Delta\nu_0}\right).$$

where R is the reflection factor. At the same time, it must be emphasized again that it is possible to generate in the field of the noise pump extremely narrow ($\Delta\nu_Q \ll \Delta\nu_0$) optical or acoustic phonon lines (see also[11]).

3. If stimulated scattering is a harmful effect, then S_{cr} should be increased. In particular, in optical fibers, where SRS and SMBS prove to be the cause of harmful "nonlinear" reflections (see[18]), S_{cr} can be increased by influencing the magnitude of the group-velocity de-

tuning μ. The developed theory can also be used to investigate the decay processes in a plasma in the field of a noise pump. It should be noted that by approximating the pump by a process with a diffusing phase, we can extend the developed approach to the case of arbitrary $\tau_{cor} \gtrless T_2, T_3$ [19] (see also [20,21]).

Of course, the statistical theory of stimulated scattering cannot yet be regarded as a complete theory. It would be interesting to allow for the effects of the spatial coherence of the pump [6]; in certain cases allowance for the diffraction of the Stokes radiation may prove to be essential[6]. Another interesting problem is connected with allowance for the back reaction of the Stokes wave on the pump and with the fluctuations in the population difference; some results have already been obtained here with the aid of the Dyson-equation method; they are expounded in [10,14] (see also the review article [23]).

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[1]For $\tau_{cor} > T_2$ the results obtained with the aid of the formulas (13) go over into the results obtained in sec. 1.

[2]The time dependence of S and $\bar{I}_p$ in (16) describes the averaged envelope of the laser pulse.

[3]The Dyson equations for the correlation functions are considered in [15].

[4]Actually, there is a weak dependence: it is due to abberations and the effects of the spatial noncoherence of the Stokes radiation and the pump.

[5]We neglect the small difference (less than 20%) between the durations of the pump pulse and the SRS.

[6]Interesting results in this direction were recently obtained by Bespalov and Pasmanik [22] for the monochromatic pump.

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