Raman scattering of light by stationary nonlinear polarization waves

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It is shown that new light-scattering regimes should arise in the case of Raman scattering of light in a medium in which optical solitons are propagating. Investigation of the formation and evolution of Stokes pulses has revealed the dependences of their principal characteristics on the scattering length and on the density of the molecules. The feasibility of the transformation of ultra-short pulses by utilizing optical solitons is considered for single-photon absorption solitons and stimulated Raman scattering solitons.

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1. INTRODUCTION

The following regimes of Raman scattering (RS) have been widely studied to date: spontaneous RS, stimulated RS (SRS), and nonstationary RS in a field of ultra-short pulses (USP). These are accompanied by a square-root dependence of the total growth rate in the Stokes intensity on the scattering length.[1,2] These regimes appear in the propagation of light in an equilibrium, unperturbed medium. We are referring here to solitons of the SRS regime of ultra-short pulses,[3,4] which are not accompanied by amplification at all.

However, if the medium is previously excited with the help of coherent fields, the RS can be of a qualitatively different character.[5,6] In particular, a consequence of the presence of coherent polarization in the medium, induced by the excitation fields, the intensity of the spontaneous RS can turn out to be proportional to the square of the total number of scattering molecules.[6] [1)] Various features of light RS on an excited medium are used in a large number of methods of active spectroscopy.[9]

The general setup of the problem of RS on an excited medium refers to a situation in which the scattered probing pulse is fed to a medium that is excited by a set L of light fields with frequencies ω_L such that

$$\sum_{l=1}^{L} n_l\omega_l=\omega_v, \tag{1}$$

where ω_v is the frequency of the considered molecular transition, L determines the order of the nonlinear process of excitation of the medium ($L=1$, $n_1=1$ corresponds to single photon absorption; $L=2$, $n_1=1$, $n_2=-1$ to Stokes RS, and so on). In Ref. 6, a number of regimes of scattering were analyzed, both in the case of long exciting pulses and in the case of ultra-short excitation pulses. However, the examples considered previously referred to standard cases of excitation of the medium and did not include the important region of effects of soliton propagation of the excited fields.

At the present time it has been reliably established that in resonance interactions of powerful ultra-short light pulses with a medium, stationary light pulses can arise in the material (solitons of the surrounding light field), and their amplitude depends on the wave variable $t'=t-z/V$ (V is the group velocity of the stationary wave packet). Optical solitons are now attracting great attention both from the general physical[10] and from the applied points of view.[11–15]

In each of the known cases of existence of optical solitons (in the case of single photon interaction—the effect of self-induced transparency,[11] in two-photon absorption,[12] in stimulated RS in molecular media[3] and in crystals,[13] in processes of higher order[14,15]), stationary waves of nonlinear polarization (SWNP) connected with the light fields arise in the medium. In the present study, light RS by polarization waves accompanying the optical solitons have been investigated. As is shown below, this form of scattering possesses a number of interesting properties due to the peculiarities of the shape of the solitons and to the possibility of their propagation to a large distance without significant distortion. In particular, the dependence of transformation of the intensity of the scattered light on the density of the medium and on the scattering length turns out to be nontrivial. Thus, the problem considered is of interest not only from the point of view of the optics of ultra-short pulses (the transformation of their frequency, shape and duration, identification of solitons, and so on) but also in connection with the general problem of the possible regimes of light scattering under physically realizable conditions.

2. FUNDAMENTAL RELATIONS

We shall assume for simplicity that the probing field and the SWNP propagate in the same direction, and that the frequencies ω_i and ω_s of the probing field $E_i=\mathscr{E}_i\cos(\omega_i t-k_i z)$ and of the Stokes field $E_s=\mathscr{E}_s\cos(\omega_s t-k_s z)$ that is generated in the probing do not coincide with any of the frequencies of the excitation fields. Then the abbreviated equations for the amplitudes $\mathscr{E}_i$ and $\mathscr{E}_s$ have the form

$$\begin{gathered}\left(\frac{\partial}{\partial z}+\frac{1}{c_i}\frac{\partial}{\partial t}\right)\mathscr{E}_i=\varkappa_i\mathscr{E}_s\int_{-\infty}^{+\infty}v(\Delta\omega)g(\Delta\omega)d(\Delta\omega),\\ \left(\frac{\partial}{\partial z}+\frac{1}{c_s}\frac{\partial}{\partial t}\right)\mathscr{E}_s=-\varkappa_s\mathscr{E}_i\int_{-\infty}^{+\infty}v(\Delta\omega)g(\Delta\omega)d(\Delta\omega),\end{gathered} \tag{2}$$

where $\varkappa_1=\lambda\mu_0 c_i\omega_i N_V/4$, $\varkappa_s=\lambda\mu_0 c_s\omega_s N_V/4$; c_i, c_s are the group velocities of the pump wave and the Stokes wave; λ

is the scattering matrix element; $\mu_0 = 4\pi/c^2$; N_V is the density of the molecules; $g(\Delta\omega)$ is the shape of the line of inhomogeneous broadening, $\Delta\omega = \omega_v - \omega_i + \omega_s$, ω_v is the frequency of the working transition of the molecules; the material equations describe the interaction of the medium both with the exciting fields and with the probing field:

$$\frac{du}{dt} = -\frac{u}{T_2} - v\Delta\omega,$$
$$\frac{dv}{dt} = -\frac{v}{T_2} + u\Delta\omega + \frac{\lambda}{\hbar}\mathscr{E}_i\mathscr{E}_s W + \frac{\lambda_L}{\hbar} Q_L W, \tag{3}$$
$$\frac{dW}{dt} = -\frac{W - W^{eq}}{T_1} - \frac{\lambda}{\hbar}\mathscr{E}_i\mathscr{E}_s v - \frac{\lambda_L}{\hbar} Q_L v,$$

where T_1 and T_2 are the longitudinal and transverse relaxation times, W is the half-difference of the populations of the upper and lower levels of the molecular transition considered; W^{eq} is the equilibrium value of W; λ_L is the matrix element of the nonlinear optical process of order L, represented by the relation (1): $Q_L = \mathscr{E}_1'\mathscr{E}_2'...\mathscr{E}_L'$, $\mathscr{E}_i'$ $(l = 1,2,...,L)$ are the amplitudes of the exciting fields. The nonlinear polarizations of the medium at any frequency are expressed in terms of the functions u and v. In particular, for polarization at the Stokes frequency we have

$$P_s = {}^1/_2\lambda N_V\mathscr{E}_i[u\cos(\omega_s t - k_s z) + v\sin(\omega_s t - k_s z)].$$

In addition, we shall consider the case of exact resonance ($g(\Delta\omega) = \delta(\Delta\omega)$) in the approximation of a specified step-like probing pulse of duration τ_i ($\mathscr{E}_i(t - z/c_i) = \mathscr{E}_i$ with $|t - z/c_i| \leqslant \tau_i/2$). The field $\mathscr{E}_i$ is assumed to be constantly present in the medium over the entire time interval of propagation of the optical soliton through the medium.

Under the assumptions made, it is convenient to represent the polarization of the medium and the population difference in the form

$$v = v_0 + v_1, \quad W = W_0 + W_1,$$

where $v_0 = v_0(t - z/V)$ and $W_0 = W_0(t - z/V)$ are stationary waves due to the propagation of an optical soliton corresponding to some process of excitation of arbitrary order L through the medium. Since, at not too strong probing fields (see the estimates in Refs. 2, 5, and 6), motion of the populations in the scattering process can be neglected, ($W_1 = 0$), it follows from (2) and (3) that

$$\frac{dv_1}{dt} = -\frac{v_1}{T_2} + \frac{\lambda}{\hbar}\mathscr{E}_i\mathscr{E}_s W_0,$$
$$\left(\frac{\partial}{\partial z} + \frac{1}{c_s}\frac{\partial}{\partial t}\right)\mathscr{E}_s = -\varkappa_s\mathscr{E}_i(v_0 + v_1). \tag{4}$$

Carrying out the change of variables $z \to z$, $t - z/c_s \to \tau$ in (4), we obtain for the Stokes amplitude an equation:

$$\frac{\partial^2\mathscr{E}_s}{\partial z\,\partial\tau} + \frac{1}{T_2}\frac{\partial\mathscr{E}_s}{\partial z} + \varkappa_s\frac{\lambda}{\hbar}\mathscr{E}_i^2\mathscr{E}_s W_0 + \varkappa_s\mathscr{E}_i\left(\frac{v_0}{T_2} + \frac{dv_0}{\partial\tau}\right) = 0, \tag{5}$$

which can be solved by the Riemann method if we take it into account that W_0, v_0 and $dv_0/d\tau$ are known functions of argument $(\tau + z/\Delta)$ where $\Delta = (1/c_s - 1/V)^{-1}$ is the group detuning between the SWNP and the Stokes wave.

We carry out the solution of Eqs. (4) and (5) for an illustrative case of practical interest, in which the polarization v_1 induced in the probing is much less than the polarization $v_0(t - z/V)$. This assumption implies neglect of stimulated processes in the scattering of the probing field $\mathscr{E}_i$ both by the SWNP and in those portions of the medium where the SWNP is absent at the given instant. It is not difficult to show that in both cases of a long ($\tau_i \gg T_2$) and a short ($\tau_i \ll T_2$) probing pulse the requirement $\Gamma_0 l \ll 1$ serves as a sufficient condition. Here Γ_0 is the static amplification coefficient of SRS at a give pump power $I_i = c_i|\mathscr{E}_i|^2/8\pi$, and l is the total scattering length. The estimate for the maximum power of the probing field then follows:

$$I_i \ll I_{cr} = \hbar c_i(2\pi\lambda^2\mu_0 c_s\omega_s N_V|W^{eq}|l)^{-1}. \tag{6}$$

Under condition (6) the total amplitude $\mathscr{E}_s$ is equal to the sum of the coherent and noise components of the spontaneous scattering while the noise scattering makes a constant contribution to the intensity, which can be ignored in comparison with the intensity of the scattering from the coherent part of the polarization, arising in the passage through the medium of the soliton. For this reason, the noise source of the unrenormalized Stokes radiation was not taken into account in the initial equations (3) from the very beginning.

In the case of condition (6), the Stokes field has the form

$$\mathscr{E}_s(l,\tau) = -\varkappa_s\mathscr{E}_i\Delta\int_\tau^{\tau + l/\Delta} v_0(t')\,dt'. \tag{7}$$

In what follows, in carrying out estimates, we shall find useful an expression for $\mathscr{E}_s(l,\tau)$ with account of inhomogeneous broadening. It is not difficult to show that in this case the right side of (7) must be multiplied by the factor

$$\int_{-\infty}^{\infty} g(\Delta\omega)[1 + (\tau_i'\Delta\omega)^2]^{-1}d(\Delta\omega),$$

where τ_i' is the characteristic time scale of the soliton.

The relations obtained above describe the spatial-temporal structure of the Stokes field which arises in Raman light scattering in a medium through which optical solitons are propagating. The formula (7), which was obtained under the restriction (6) on the intensity of the probing field, shows that the structure of the Stokes pulse depends on the type of the soliton, which determines the shape of the amplitude of the polarization wave $v_0(t - z/V)$, on the scattering length l and on the group detuning Δ. It follows from (7) that the character of the connection of the scattered field with $v_0(t - z/V)$ is typical for cases of coherent interaction of light with matter, when effects of phase correlation between the separate radiators are significant. We proceed to the analysis of the general relations for concrete cases of excitation of the medium by optical solitons.

3. RAMAN SCATTERING FROM A POLARIZATION WAVE OF LIMITED EXTENT

We shall initially trace the features of RS from a SWNP, for the case in which $v_0(t')$ is a step of height $\tilde{v}$ and breadth δ ($v_0(t') = \tilde{v}$ at $0 \leqslant t' \leqslant \delta$). Let $V > c_s$. If $l \leqslant \delta\Delta$ then, in accord

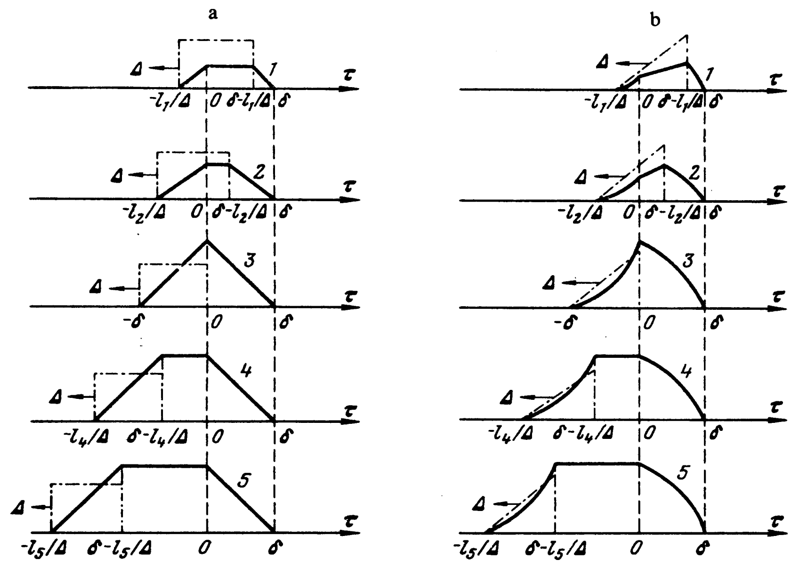

FIG. 1. Stokes pulse in the case of RS from SWNP having limited duration; a—case of step polarization at $V>c_s$; b—case of linearly increasing polarization ($V>c_s$). Curves 1 correspond to scattering length l_1 less than l_{cr}; 2—$l_2(l_{cr}>l_2>l_1)$; 3—$l_3=l_{cr}$; 4—$(l_4>l_3)$; 5—$l_5(l_5>l_4)$. The dashed curve shows the SWNP; the velocity and direction of its propagation are indicated.

with (7),

$$\begin{aligned}&\mathscr{E}_s(l,\tau)=-\varkappa_s\tilde{v}\mathscr{E}_i\Delta(\tau+l/\Delta)\quad\text{at}\quad -l/\Delta\leqslant\tau\leqslant 0;\\ &\mathscr{E}_s(l,\tau)=-\varkappa_s\tilde{v}\mathscr{E}_i l\quad\text{at}\quad 0\leqslant\tau\leqslant\delta-l/\Delta;\\ &\mathscr{E}_s(l,\tau)=-\varkappa_s\tilde{v}\mathscr{E}_i\Delta(\delta-\tau)\quad\text{at}\quad \delta-l/\Delta\leqslant\tau\leqslant\delta;\\ &\mathscr{E}_s(l,\tau)=0\quad\text{at}\quad \tau\leqslant-l/\Delta,\quad\tau\geqslant\delta.\end{aligned}\tag{8}$$

If $l\geqslant\delta\Delta$,

$$\begin{aligned}&\mathscr{E}_s(l,\tau)=-\varkappa_s\tilde{v}\mathscr{E}_i\Delta(\tau+l/\Delta)\quad\text{at}\quad -l/\Delta\leqslant\tau\leqslant-l/\Delta+\delta;\\ &\mathscr{E}_s(l,\tau)=-\varkappa_s\tilde{v}\mathscr{E}_i\delta\Delta\quad\text{at}\quad -l/\Delta+\delta\leqslant\tau\leqslant 0;\\ &\mathscr{E}_s(l,\tau)=-\varkappa_s\tilde{v}\mathscr{E}_i\Delta(\delta-\tau)\quad\text{at}\quad 0\leqslant\tau\leqslant\delta;\\ &\mathscr{E}_s(l,\tau)=0\quad\text{at}\quad \tau\leqslant-l/\Delta,\quad\tau\geqslant\delta.\end{aligned}\tag{9}$$

For the case $V>c_s$ the picture of the evolution of the Stokes pulse in its propagation through the medium, obtained in accord with (8) and (9), is shown in Fig. 1a. The case $V<c_s$ is similar to the case $V>c_s$, the sole difference being that the step of the SWNP, $v_0(t')$, will shift relative to $\mathscr{E}_s$ not to the left but to the right. In both cases, the character of the dependence of the scattering amplitude on l is determined by the relation between the duration of the SWNP induced by the soliton, and the quantity l/Δ—the time of group detuning of the Stokes wave and the SWNP over the distance l.

We shall now clarify qualitatively the picture of the evolution for the case $V>c_s$. An arbitrary point z of the medium seres as the source of the Stokes waves in the interval of time between z/V and $\delta+z/V$, when the SWNP passes through this point (as the time origin we take the instant of entry of the leading edge of $v_0(t')$ into the medium). These waves reach the end of the medium ($z=l$) after a time interval

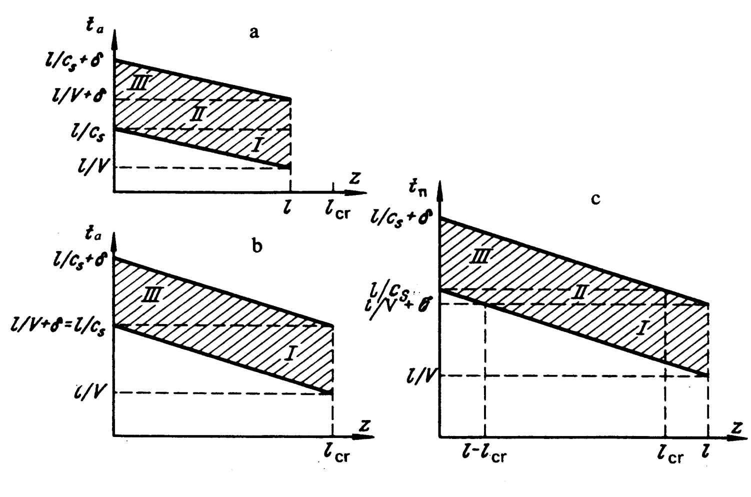

FIG. 2. Intervals of time of arrival of Stokes waves from various points of the sample; a—$l<l_{cr}$, b—$l=l_{cr}$, c—$l>l_{cr}$. The possible values of t_a lie in the shaded region. Regions I and III denote the spatial-temporal regions of formation of the leading and trailing edges of the Stokes pulse, region II is the central region of the pulse.

$(l-z)/c_s$. Therefore, waves from the point z arrive at the exit from the medium in an arrival time interval t_a which satisfies the condition

$$z/V+(l-z)/c_s \leqslant t_a \leqslant z/V+\delta+(l-z)/c_s. \quad (10)$$

Possible values of t_a as a function of the location of the point z are shown in Fig. 2. We introduce the critical length $l_{cr}=\delta\Delta$, passing through which the polarization wave overtakes the Stokes by a value equal to its extent $V\delta$, and these waves pass out of the region of interaction.

At $l<l_{cr}$ (Fig. 2a), the leading edge of the Stokes pulse ($l/V \leqslant t_a \leqslant l/c_s$ or $-l/\Delta \leqslant \tau \leqslant 0$) is formed by waves arriving from points in the medium with the following coordinates z: $(l/c_s - t_a)\Delta \leqslant z \leqslant l$. At an arbitrary instant t_a, the total Stokes amplitude at the point $z=l$ is proportional to the "working" length of the medium, from which Stokes photons arrive, since in the case of a step SWNP the separate Stokes waves increase linearly from $t_a=l/V$ to $t_a=l/c_s$. Thus, the leading edge of the pulse is formed principally from the scattering in the back end of the medium. The back end of the pulse is formed by the following portions of the medium: $0 \leqslant z \leqslant \Delta\,(l/c_s+\delta-t_a)$, where $l/V+\delta \leqslant t_a \leqslant l/c_s+\delta$. The total amplitude decreases linearly from $t_a=l/V+\delta$ to $t_a=l/c_s+\delta$ (i.e., from $\tau=\delta-l/\Delta$ to $\tau=\delta$). At the moment t_a of the interval $l/c_s \leqslant t_a \leqslant l/V+\delta (0 \leqslant \tau \leqslant \delta - l/\Delta)$ Stokes photons from all points of the medium arrive at the point $z=l$. Therefore, the middle part of the pulse is formed by the total length of the sample. The ampliltude $\mathscr{E}_s$ is maximal here and does not depend on τ. It is clear from (10) and Fig. 2a that the widths of the leading and trailing edges will increase with incrase in l, with simultaneous increase in the maximum instantaneous intensity of the pulse and decrease in the extent of its middle part.

At $l=l_{cr}$ (Fig. 2b) the Stokes wave from the point $z=0$, which arises when the leading edge of the polarization passes through the beginning of the sample, reaches the end of the sample $z=l$ just at the instant when the trailing edge of the polarization appears at the point $z=l$, and the last Stokes wave arises at this point, i.e., $l/c_s=l/V+\delta$. Therefore, the intensity turns out to be maximal only at one instant of time, $t_a=l/c_s=l/V+\delta$, when the Stokes waves from all points of the sample arrive at the point $z=l$. At $l=l_{cr}$ the maximal instantaneous amplitude of the pulse achieves its largest possible value: $\mathscr{E}_s = -\varkappa_s\tilde{v}\mathscr{E}_i\, l_{cr}$ (at $\tau=0$).

At $l>l_{cr}$ (Fig. 2c), the leading edge of the pulse is formed by the points $l/c_s - t_a)\Delta \leqslant z \leqslant l$, and the temporal region of its formation is determined by the inequalities $l/V \leqslant t_a \leqslant \delta + l/V$. Correspondingly, for the trailing edge, $0 \leqslant z \leqslant \Delta\,(l/c_s+\delta-t_a)$, where $l/c_s \leqslant t_a \leqslant \delta + l/c_s$. It is clear from Fig. 2c that in these regions $\mathscr{E}_s$ depends linearly on τ. In the case $l>l_{cr}$ there again exists an interval of arrival times $\delta+l/V \leqslant t_a \leqslant l/c_s$ at which the amplitude is maximal and is equal to the largest possible value $\overline{\mathscr{E}}_s$, since the spatial region of formation of the pulse $[(l/c_s-t_a)\Delta \leqslant z \leqslant (l/c_s+\delta-t_a)\Delta\,]$ has a length equal to l_{cr}. Upon further increase in l the maximal value of the amplitude does not increase, since the Stokes waves going from points of the medium separated by a distance greater than $\delta\Delta$ will reach the point $z=l$ at different instants of time.

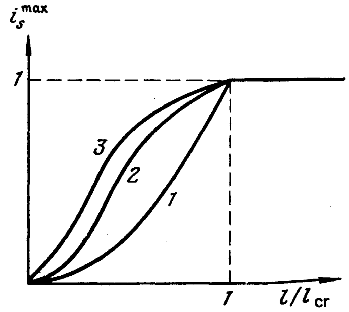

FIG. 3. Intensity of RS from bounded SWNP as a function of the scattering length ($i_s^{max}=(8\pi/l_{cr}{}^2c_s)[(\beta+1)/\alpha\delta^\beta\varkappa_s\,\mathscr{E}_i]^2 I_s^{max}$): 1—$\beta=0$; 2—$\beta=1$; 3—$\beta>1$.

The dependence of the maximal instantaneous scattering intensity $I_s^{max}=c_s|\mathscr{E}_s^{max}|^2/8\pi$ on l is shown in Fig. 3 (curve 1). At $l<l_{cr}$, the soliton and the Stokes wave do not emerge from the region of interaction; therefore the central part of the pulse ($0 \leqslant \tau \leqslant \delta - l/\Delta$) is formed as a result of scattering by the entire volume of the medium and I_s^{max} turns out to be proportional to $(N_V\, l)^2$—the square of the total number of scattering particles (per unit cross section of the active volume). Thus, at $l \leqslant l_{cr}$ the case of coherent (cooperative) RS is realized.[5,7] At $l>l_{cr}$ the RS also has a cooperative character; however, the duration of the SWNP is now less than the characteristic time of the group detuning. Since the interacting waves manage to "diverge" over the length l, not all the molecules of the active volume partake in the scattering, but only that part $N_V\, l_{cr}$. Therefore, $I_s^{max} \sim (N_V\, l_{cr})^2$ and does not depend on the length of the scattering volume. Only the duration of the Stokes pulse increases with increase in l.

The basic qualitative features of formation and evolution of the Stokes pulse are preserved also for SWNP of arbitrary type, which has a restricted duration, for example, for waves of the form $v_0(t')=\alpha(t')^\beta$, where $\alpha, \beta>0$, $0 \leqslant t' \leqslant \delta$. Only the form of the Stokes pulse changes. Thus, at $\beta=1$, $V>c_s$ (Fig. 1b), the pulse is nonsymmetric; the constant-intensity section appears only at $l>l_{cr}=\delta\Delta$, while at $l<l_{cr}$ the maximal amplitude corresponds to the point $\tau=\delta-l/\Delta$.

However, the behavior of I_s^{max} on the scattering length appears to be a new point. At $l \leqslant l_{cr}$ (Fig. 3)

$$I_s^{max}=\frac{c_s}{c_i}l_{cr}{}^2\left(\frac{\alpha\delta^\beta\varkappa_s}{\beta+1}\right)^2 I_i\left[1-\left(1-\frac{l}{l_{cr}}\right)^{\beta+1}\right]^2. \quad (11)$$

At $\beta>0$ the intensity increases with increase in l not according to a quadratic law, as in the case of coherent RS,[5,7] but more rapidly. According to Eq. (11), I_s^{max} increases in the general case according to a polynomial law with the power of the polynomial dependent on the specific form of the SWNP. If the SWNP has a portion of a power-law growth with exponent β, then I_s^{max} turns out to be a polynomial of degree $2(\beta+1)$ over l/l_{cr}. Moreover, although I_s^{max} is proportional to the sqaure of the density of molecules, it depends—in contrast to other RS regimes—on N_V and l, but separately and not in the combination ($N_V\, l$) that gives the total number of particles.

4. RAMAN SCATTERING FROM McCALL AND HAHN SOLITONS

We now consider RS from SWNP corresponding to real optical solitons. In the propagation of a McCall-Hahn soliton in the medium,[11] the amplitude of the resonance excitation field is equal to

$$\mathscr{E}_i{}' = \frac{\hbar}{\mu\tau_i{}'}\,\mathrm{sech}\,\frac{t'}{\tau_i{}'},$$

where μ is the dipole moment of the transition and $v_0(t')$ has an exponentially decaying wing:

$$v_0(t') = -2\,\mathrm{sh}\,(t'/\tau_i{}')\,\mathrm{ch}^{-2}(t'/\tau_i{}'). \qquad (12)$$

Considering the case of a nonspherically symmetric medium (there is no alternate prohibition) and after substitution of (12) in (7), we find

$$\mathscr{E}_s(l,\tau) = A_1\left[\mathrm{ch}^{-1}\left(\frac{\tau + l/\Delta}{\tau_i{}'}\right) - \mathrm{ch}^{-1}\frac{\tau}{\tau_i{}'}\right]; \quad A_1 = -2\varkappa_s\mathscr{E}_i\tau_i{}'\Delta.$$

Because of the presence of two maxima (shifted in phase by π) in the polarization, the Stokes pulse has a two-peak form (Fig. 4). The extremal points $\tau_\pm = \tau_0 \pm \tau_i'$ arsinh (cosh τ_0/τ_i') are symmetric relative to points of zero amplitude $\tau_0 = -l/2\Delta$. In the case $V > c_s$ (Fig. 4), the left peak propagates with a velocity exceeding the group velocity c_s of the Stokes wave, while the right peak is group-retarded. With increase in l, the group advance of the left peak increases and the retardation of the right side decreases, since τ_- tends monotonically to $-\infty$, while τ_+ goes to zero. Both at $V > c_s$ and at $V < c_s$, the intensity at the maxima of the pulse increases according to the square law $[I^m{}_s{}^a \sim (N_V\, l)^2]$ at $l/\tau_i'|\Delta| \ll 1$, and tends toward a final value $J_s = 4c_s(\varkappa_s\tau_i'\Delta)^2 I_i/c_i$ at $l/\tau_i'|\Delta| \gg 1$. Saturation of the maximal intensity is achieved at distances of the order of several $\tau_i'|\Delta|$. Thus, the behavior of the intensity at the maxima of the pulse in the case of a polarization wave with wings exponentially decaying at infinity is similar to the model case of bounded SWNP considered above. At other points of the pulse, the growth of the intensity is described by more complicated dependences. For example, at $2\tau|\Delta| \ll l \ll 2\tau_i'|\Delta|$, we have $I_s = c_s(\varkappa_s/\tau_i'\Delta)^2 I_i\, l^4/c_i$. Since $\varkappa_s \sim N_V, (\tau_i')^2 \sim N_V^{-1}$, the intensity increases like $N_V^3\, l^4$.

We carry out an estimate for the extreme value J_s of the maximum intensity and for the characteristic length $\bar{l} \equiv \tau_i'|\Delta|$. We shall consider the case of a broad line of inhomogeneous broadening ($T_2^* \ll \tau_i'$, where T_2^* is the time of inhomogeneous broadening), corresponding to conditions of experiments on the observation of self-induced transparency.[16,17] Assuming that $c_i \approx c_s \approx c/\eta$ (η is the nonresonant part of the refractive index of the excitation field) and using the relation[11]

$$\eta/c - 1/V = \alpha_a\tau_i{}'/2,$$

where α_a is the absorption coefficient, we obtain, with the account taken of inhomogeneous broadening,

$$J_s = (\mu\lambda\omega_s c_s N_V/\alpha_a)^2 I_i (T_2{}^*/\tau_i{}')^2$$

and $\bar{l} = 2/\alpha$. For radiation of a CO_2 laser in BCl_3 gas ($\omega_v = 2\times10^{14}\ \mathrm{s}^{-1}$) at a pressure of 100 mTorr and $T = 300$ K, the values $\alpha_a = 0.15\ \mathrm{cm}^{-1}$-$\mathrm{Torr}^{-1}$ and $\tau_i' \approx 80$ ns were obtained in Ref. 17. Thus, $\bar{l} = 1.3$ m. Since $T_2^* = 10$ ns under the stated conditions, at $\lambda = 10^{-23}\ \mathrm{cm}^3$ and $\omega_s = 2\times10^{15}\ \mathrm{s}^{-1}$, we obtain $\gamma \equiv J_s/I_i \approx 5\times10^{-2}$ for the intensity conversion coefficient. The considered RS regime from the McCall-Hahn soliton can be obtained at distances l less than the distance l_r at which the soliton is destroyed because of relaxation processes. According to Ref. 11, at $T_2 \ll T_1$, we have $l_r = (3/2\alpha_a)T_2/\tau_i'$ and, since $\tau_i' \ll T_2$, the condition $l_r \gg \bar{l}$ is satisfied, i.e., saturation of the maximal intensity, is possible. At $l = l_r$, the distance between the peaks of the Stokes pulse, characterizing its duration, becomes of the order of T_2.

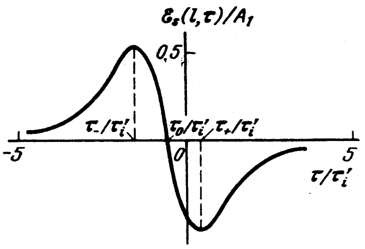

FIG. 4. Stokes pulse in; the excitation of a medium by solitons of one-photon absorption ($V > c_s$, $l/\tau_i'\Delta = 1$).

5. RAMAN SCATTERING FROM SRS SOLITONS

We now consider the case of excitation of the medium by SRS solitons.[3] Let ω_i', η_i', c_i', and ω_s', η_s', c_s' be respectively the frequencies, refractive indices, and group velocities of the pump and Stokes waves the interaction between which leads to the onset of the soliton regime of SRS (λ' is the matrix element of the interaction of these waves). In the region of anomalous dispersion, the amplitudes of the SRS solitons have the form

$$\mathscr{E}'_{1,2} \equiv \mathscr{E}'_{i,s} = [2a_{i,s}/(1+b^2t'^2)]^{1/2},$$

where

$$a_i = \hbar\mu_0 c\omega_i{}' c_i{}' V W^{eq} N_V [2\eta_i{}'(V - c_i{}')]^{-1},$$
$$a_s = \hbar\mu_0 c\omega_s{}' c_s{}' V W^{eq} N_V [2\eta_s{}'(c_s{}' - V)]^{-1},$$

$b^{-1} = \hbar/\lambda'(a_i\, a_s)^{1/2}$ is the characteristic time scale of the solitons, and $c_s' < V < c_i'$. The SWNP corresponding to them is given by the expression

$$v_0(t') = (-2W^{eq}bt')\,[1 + (bt')^2]^{-1}. \qquad (13)$$

According to (7) and (13), the amplitude of the Stokes waves produced by probing with an SWNP field of amplitude $\mathscr{E}_i$ has the form

$$\mathscr{E}_s(l,\tau) = A_2 \ln\frac{1 + b^2(\tau + l/\Delta)^2}{1 + b^2\tau^2};$$

$$A_2 = -\frac{1}{2}\frac{\lambda}{\lambda'}\frac{\omega_s}{(\omega_i{}'\omega_s{}')^{1/2}}\frac{c_s{}^2}{c_i{}'c_s{}'}\frac{[(V - c_s{}')(c_i{}' - V)]^{1/2}}{V - c_s}\mathscr{E}_i. \quad (14)$$

The Stokes pulse has a two-peak shape (see Fig. 5, which corresponds to the case $V > c_s$), and is symmetric relative to

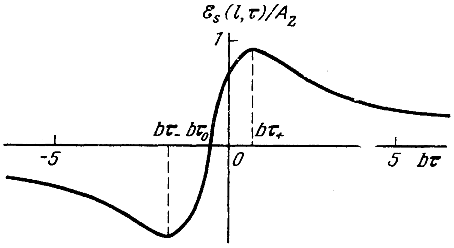

FIG. 5. Stokes pulse for the case of excitation of the medium by SRS solitons ($V > c_s$, $bl/\Delta = 1$).

$\tau_0 = -l/2\Delta$. The group advance of the leading edge of the pulse (the left peak) increases without limit, while the group retardation of the trailing edge (the right peak) approaches zero, since the extremal values of $\mathscr{E}_s$ correspond to the points

$$\tau_\pm = -\frac{l}{2\Delta}\left(-1 \pm \left[1 + \left(\frac{2\Delta}{bl}\right)^2\right]^{1/2}\right).$$

At $l \ll 2|\Delta|/b$, the intensity at the maxima of the pulse increases as $(N_V l)^2$, while its behavior at $l \gg 2|\Delta|/b$ is described by the relation

$$I_s^{max} \sim \ln^2(bl/|\Delta|).$$

The absence of a limiting value for $I_s^{\max}$ is a characteristic feature of excitation of a medium by SRS solitons and is connected with the slow (Lorentzian) damping of the wings of the SWNP. We note an interesting dependence of $I_s^{\max}$ on the total number of particles $N_V l$ at $l \gg 2|\Delta|/b$:

$$I_s^{max} \sim \ln^2(AN_V l),$$

where A is a constant of dimensionality length squared, and also the nonmonotonic character of the dependence of the intensity of the pulse on l at the points $\tau(|\tau - \tau_0| \ll \tau_0)$ located near points of zero intensity: $I_s \sim \{(bl/2\Delta)[1 + (bl/2\Delta)^2]^{-1}\}^2$ (I_s increases quadratically in l at $bl/2|\Delta| \ll 1$, reaches a maximum, and then falls to zero).

Thus, in contrast to the cases of restricted SWNP and SWNP with exponentially damped wings, no saturation of the maximal intensity $I_s^{\max}$ with increase in l takes place. Actually, the quantity $I_s^{\max}$ will be limited to the value reached at a distance of order

$$\tilde{l}_r = 4a_i(T_1^{-1} + T_2^{-1})(\hbar c \mu_0 \omega_i' \eta_i' N_V |W^{eq}|)^{-1},$$

at which destruction of the SRS solitons occurs.[3]

We shall now make estimates. We shall assume that in order of magnitude

$$T_2 \ll T_1,\quad \omega_i' \approx \omega_s' \approx \omega_s,\quad c_i' \approx c_s' \approx c_s,\quad |W^{eq}| \approx 1,\quad \eta_i' \approx 1,$$
$$\lambda' \approx \lambda,\quad V - c_i' \approx c_s' - V \approx V - c_s. \qquad (15)$$

Then $\tilde{l}_r \approx cbT_2(\pi\lambda'\omega_i' N_V)^{-1}$. At $N_V = 5\times10^{16}\ \text{cm}^{-3}$ and $T = 300$ K we can assume $T_2 \approx 100$ ns and $T_2^* \approx 1$ ns. For $\omega_i' = 2\times10^{15}\ \text{s}^{-1}$, $\lambda' = 10^{-23}\ \text{cm}^3$, and $bT_2 = 10$ we obtain $\tilde{l}_r \approx 2$ m, and $\tilde{l} \equiv 2|\Delta|/b \approx 0.3$ m. Since $\tilde{l}_r \gg \tilde{l}$, the greatest value of the conversion coefficient γ, which is obtained at a distance of the order of $\tilde{l}_r$, can be estimated from the formula

$$\gamma = I_s^{max}/I_i = (bT_2^*)^2 \ln^2(2\tilde{l}_r/\tilde{l}),$$

where we have used the conditions (15) and taken into account the inhomogeneous broadening. For the selected parameters, $\gamma \approx 0.3$. At $l \approx \tilde{l}_r$ the distance between the peaks of the Stokes pulse, which characterizes its duration, is equal to $\tau_+ - \tau_- \approx l/|\Delta| \approx 1.3T_2$. We note that since this value is linear in l, while $I_s^{\max}$ depends on l logarithmically, at distances less than $\tilde{l}_r$ we can obtain a Stokes pulse of ultra-short duration with sufficiently large γ. For example, at $l = 2\tilde{l}$ we have $\tau_+ - \tau_- \approx 0.4T_2 \approx 40$ ns. At the same time, according to (14), $\gamma \approx 0.08$.

It should be noted that the results obtained in Secs. 2–5 for Stokes scattering are easily transformed to the anti-Stokes RS.

6. CONCLUSIONS

The following conclusions can be drawn from the discussion just given.

1. In light RS by optical solitons, the coherent part of the Stokes scattering field is described by the relations (5)–(7), which establish the connection of its spatial-temporal structure with the wave of nonlinear polarization that is due to the soliton propagation, with the scattering length l, and with the group detuning Δ.

2. The character of the dependence of the Stokes amplitude on l is determined by the relation between the characteristic time scale of the SWNP and the time of group detuning l/Δ between the SWNP and the Stokes wave produced in the course of probing. If l is less than the critical value l_{cr}, which is determined by the equality of the indicated times, then specific scattering regimes can exist that differ from those known previously. These regimes are described by different power dependences of the instantaneous intensity on l and on the density of scatterers N_V of the form $l^p N_V^q$, in which the indices p and q are not identical. Thus, the so-called coherent or cooperative RS[6,7]—the analog of Dicke superradiance—is only one of the possible particular manifestations of the considered effects.

3. In propagation of bounded SWNP and SWNP with exponentially decaying wings through the medium (the McCall-Hahn soliton), the maximum instantaneous intensity of the scattered light at $l > l_{cr}$ reaches saturation. Solitons with slowly damped Lorentzian wings (SRS solitons) have typically no saturation; with increase in l the intensity increases logarithmically, and this increase is limited only by relaxation processes which lead to destruction of the solitons.

4. The given estimates show that the considered forms of scattering can be observed under the conditions of earlier experiments on self-induced transparency. In their observation, the characteristic two-peak form of the Stokes pulse should appear. Estimates also indicate the possibility of use of new RS regimes for transformation and control of ultrashort pulses.

[1)]Such a regime, called coherent or cooperative RS, can also be observed in an initially incoherent system of molecules, thanks to the spontaneous induction of intermolecular correlations in the course of the scattering process itself.[7,8]

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