

Coherent amplification of radiation under forward resonant stimulated Raman scattering

L. A. Bol'shov, N. N. Elkin, V. V. Likhanskiĭ, and M. I. Persiantsev

(Submitted 11 May 1984)

Zh. Eksp. Teor. Fiz. **88**, 47–59 (January 1985)

Stimulated Raman scattering (SRS) of light pulses interacting coherently in a resonant three-level medium is studied numerically and analytically. The Stokes wave is trapped nonlinearly in the pulse interaction region for a wide range of parameters, which results in a high coherent SRS efficiency. Soliton solutions describing 100% conversion of energy into the Stokes wave are derived by the inverse scattering method.

1. INTRODUCTION

Interest has increased recently in stimulated Raman scattering (SRS) in resonant media, because such scattering may be accompanied by effective frequency conversion and substantial shortening of light pulses. For example, frequency conversion and pulse shortening were achieved experimentally in Refs. 1, 2, where excimer lasers with $\approx 50\%$ efficiency and various metal vapors were used. There are two reasons for the special interest in coherent transformation of resonance radiation. First, intense short pulses can propagate without losses in resonant absorbing materials^{3,4}; second, the resonant interaction of light pulses under these conditions is highly nonlinear. According to Ref. 5, the efficiency of three-frequency parametric interaction among ultrashort radiation pulses can reach 100% in resonant media if the oscillator strengths for the resonant transitions satisfy certain conditions. Frequency conversion occurs over a distance of a few light pulse lengths in the medium.

It was pointed out in Ref. 6 that coherent interaction with the Stokes wave can substantially shorten radiation pulses in resonant three-level media when the ratio of the oscillator strengths is large. Because the interacting light pulses travel at widely different velocities in the three-level medium, the conversion efficiency (defined as usual⁶ as the total amplification along the pulse interaction length) must be low if the oscillator strengths are comparable.

In the paper we discuss a new effect which involves nonlinear trapping of a Stokes wave during coherent SRS in a resonant medium. We show that in addition to the amplification, the dispersion of the Stokes wave also plays an important role and causes the pulses to be coupled. The nonlinear trapping can increase the efficiency to nearly 100%.

2. FUNDAMENTAL EQUATIONS AND LINEAR ANALYSIS

We will analyze the interaction among coherent light pulses in a resonant three-level medium for the Λ -configuration discussed in Ref. 6. For pulse lengths less than the relaxation times of the medium, the equations

$$\begin{aligned} \frac{\partial a_1}{\partial t} &= \frac{i}{2\hbar} \mu_1 E_1 a_3, & \frac{\partial a_2}{\partial t} &= \frac{i}{2\hbar} \mu_2 E_2 a_3, \\ \frac{\partial a_3}{\partial t} &= \frac{i}{2\hbar} (\mu_1 E_1^* a_1 + \mu_2 E_2^* a_2), \end{aligned} \quad (1a)$$

$$\begin{aligned} i \left(\frac{\partial E_1}{\partial t} + \frac{c}{n_1} \frac{\partial E_1}{\partial x} \right) &= \frac{4\pi N \mu_1 \omega_1}{n_1} a_1 a_3^*, \\ i \left(\frac{\partial E_2}{\partial t} + \frac{c}{n_2} \frac{\partial E_2}{\partial x} \right) &= \frac{4\pi N \mu_2 \omega_2}{n_2} a_2 a_3^*. \end{aligned} \quad (1b)$$

govern the amplitudes $a_{1,2,3}$ for the level population and the smooth envelopes E_1, E_2 of the pump and Stokes waves, respectively.

Here N is the number density of the resonant particles; $\mu_{1,2}, \omega_{1,2}$, and $n_{1,2}$ are the dipole moments, frequencies, and nonresonant refractive indices for transitions 1–3 and 2–3. We will assume throughout that only level one is populated initially: $|a_1| = 1, |a_2| = |a_3| = 0$. Two pulses are input to the medium—an intense pumping pulse, and a weak radiation pulse resonant with the 2–3 transition.

We consider the initial stage of the amplification of the weak Stokes signal during interaction with a pump pulse whose propagation velocity is independent of time. Although a linear analysis of the signal gain at the frequency ω_2 will not enable us to find how much of the energy is transferred to the Stokes wave, it will permit us to determine the principal features of pulse interaction during coherent resonant SRS. In the absence of the Stokes wave, the pump pulse travels in a resonantly absorbing two-level medium. As the input signal evolves in time, it splits into separate components which travel at different velocities without attenuation. In the limit of large separation, these component pulses have area equal to 2π (i.e., they are 2π -pulses^{3,4}).

$$E_1 = \frac{2\hbar}{\mu_1 \tau} \operatorname{sech} \frac{t-x/v}{\tau},$$

whose velocity v and duration τ are related by

$$\frac{c}{n_1 v} - 1 = \frac{2\pi N \mu_1^2 \omega_1}{n_1 \hbar} \tau^2 \equiv \Omega_1^2 \tau^2. \quad (2)$$

In the linear approximation, Eqs. (1a), (1b) imply the equation

$$\frac{\partial E_2}{\partial t} + \frac{c}{n_2} \frac{\partial E_2}{\partial x} = \frac{2\pi N \mu_2^2 \omega_2}{n_2 \hbar} a_3(x, t) \int_{-\infty}^t a_3(x, t') E_2(x, t') dt' \quad (3)$$

for the amplification of a weak Stokes signal pumped by a 2π -pulse. In this case, $a_3 = \operatorname{sech}[(t-x/v)\tau^{-1}]$ gives the dependence of a_3 on x and t .

It would appear that the amplification of the Stokes wave during interaction with the 2π pumping pulse could readily be found by the following simple reasoning. The region where the population a_3 is high acts as an amplifying medium for the field E_2 ; the diameter of the region where $|a_3|^2 \sim 1$ is comparable in order of magnitude to the length $L = v\tau$ of the 2π -pulse in the medium. If the duration τ_s of the Stokes pulse is much less than τ , we can roughly approximate the right-hand side of (3), which describes the amplification, by the expression $2\pi N\mu_2^2\omega_2\tau_s E_2/n_2\hbar$. The interaction length L_{int} is equal to $v\tau c/(c-v)$ and the gain parameter in the exponential is given by

$$\frac{2\pi N\mu_2^2\omega_2\tau_s}{n_2\hbar c} L_{\text{int}} \sim \frac{\mu_2^2\omega_2\tau_s}{\mu_1^2\omega_1\tau}.$$

This rough analysis leads to the result

$$E_{2\text{ out}} = E_{2\text{ in}} \exp\left(\frac{\mu_2^2\omega_2\tau_s}{\mu_1^2\omega_1\tau}\right)$$

(compare with Ref. 6). In this approach we completely neglect the influence of the pumping pulse on the velocity of the Stokes signal; the above estimate is certainly incorrect if the Stokes pulse is retarded or trapped.

We next estimate the parameters of the medium for which the Stokes wave can be trapped in the pulse interaction region. With respect to a coordinate system moving with the pumping pulse, the escape of amplified radiation from the interaction region is described by the expression

$$(c/n-v)(\partial E_2/\partial x) \sim (c/n-v)(E_2/v\tau),$$

which by (2) is of order $(2\pi N\mu_2^2\omega_1/n_1\hbar)\tau E_2$, and the amplification is $\sim (2\pi N\mu_2^2\omega_2/n_2\hbar)\tau E_2$. The Stokes wave will be trapped if the gain in the interaction region exceeds the field losses in the moving coordinate system, i.e., if $\mu_2^2\omega_2 > \mu_1^2\omega_1$. We will show below that this estimate agrees in order of magnitude with the result found by solving (3) exactly.

If we use the dimensionless variables $\xi = (t-x/v)/\tau$ and $z = xn_1/c\tau$ and make the change of variable

$$u = \int_{-\infty}^{\xi} a_3(\xi') E_2(\xi', z) d\xi'$$

we can rewrite Eq. (3) in a coordinate system moving at velocity v in the form

$$\frac{\partial^2 u}{\partial \xi^2} + \text{th } \xi \frac{\partial u}{\partial \xi} - \frac{1}{\Omega_1^2 \tau^2} \frac{c/n_1 v - 1}{c/n_2 v - 1} \frac{\partial^2 u}{\partial \xi \partial z} + \frac{\mu_2^2 \omega_2 (c/n_1 v - 1)}{\mu_1^2 \omega_1 (c/n_2 v - 1)} \frac{u}{\text{ch}^2 \xi} = 0. \quad (4)$$

If we assume a solution of (4) of the form $u \sim \exp(\gamma z)$, the standard change of variables $Y = (1 + \tanh \xi)/2$ reduces (4) to the hypergeometric equation

$$Y(1-Y) \frac{d^2 u}{dY^2} + \left(\frac{1-v}{2} - Y\right) \frac{du}{dY} + \kappa^2 u = 0. \quad (5)$$

Here we have written

$$\nu = \frac{\gamma}{\Omega_1^2 \tau^2} \frac{c/n_1 v - 1}{c/n_2 v - 1}, \quad \kappa^2 = \frac{\mu_2^2 \omega_2}{\mu_1^2 \omega_1} \frac{c/n_1 v - 1}{c/n_2 v - 1}.$$

The solution of (5) satisfying the boundary condition $u \rightarrow 0$ for $\xi \rightarrow -\infty$ is^{7,8}

$$u = Y^{(1+\nu)/2} F\left(-\kappa + \frac{1+\nu}{2}, \kappa + \frac{1+\nu}{2}, \frac{3+\nu}{2}; Y\right). \quad (6)$$

In this case the condition that the field be bounded for $\xi \rightarrow \pm \infty$ is equivalent to

$$[Y(1-Y)]^{1/2} (du/dY) < \text{const for } Y \rightarrow 0; 1.$$

If we use the connection formula for the hypergeometric function when the argument Y is replaced by $1-Y$ (cf. Refs. 7, 8), we find the asymptotic expressions

$$E_2(\xi) = D \left\{ \frac{1+\nu}{2} \exp \nu \xi + \frac{2}{3+\nu} \left[\left(\frac{1+\nu}{2} \right)^2 - \kappa^2 \right] \times \exp[(\nu+2)\xi] \right\}, \quad \xi \rightarrow -\infty, \quad (7)$$

$$E_2(\xi) = D \left\{ 2\Gamma\left(\frac{3+\nu}{2}\right) \Gamma\left(\frac{1-\nu}{2}\right) \times [\Gamma(1+\kappa)\Gamma(1-\kappa)]^{-1} \frac{\kappa^2}{1+\nu} \exp(-\xi) \right.$$

$$+ \Gamma\left(\frac{3+\nu}{2}\right) \Gamma\left(\frac{\nu-1}{2}\right) \left[\Gamma\left(\frac{1+\nu}{2} + \kappa\right) \Gamma\left(\frac{1+\nu}{2} - \kappa\right) \right]^{-1} \times \left[\frac{2}{3-\nu} \left[\left(\frac{1+\nu}{2} \right) \left(\frac{3-\nu}{2} \right) - 1 + \kappa^2 \right] \times \exp(-2\xi + \nu\xi) + \frac{1-\nu}{2} \exp \nu \xi \right] \left. \right\}, \quad \xi \rightarrow +\infty$$

for the fields. Here D is an arbitrary constant.

Equation (7) and the conditions that E_2 be bounded for $\xi \rightarrow \pm \infty$ imply that for $\kappa < 1/2$ there exist two solutions in the continuous spectrum ($\nu = i\Delta\tau$, $\text{Im}\Delta = 0$, where Δ is the displacement of the Stokes signal frequency from resonance for the 2-3 transition). These solutions describe the amplification of a continuous signal and behave asymptotically as $E_2 \sim \exp[\Delta(t-x/c)]$ for $\xi \rightarrow \pm \infty$, which corresponds to radiation of frequency $\omega_2 + \Delta$. The gain of the weak wave can be calculated from the ratio of the signal amplitudes for $\xi \rightarrow \pm \infty$:

$$g(\Delta) = \frac{|E_{2\text{ out}}|^2}{|E_{2\text{ in}}|^2} = \frac{\text{ch}^2(\pi\Delta\tau/2)}{\text{ch}^2(\pi\Delta\tau/2) - \sin^2 \pi\kappa}. \quad (8)$$

For a weak signal of arbitrary form, the amplification depends on the ratio of the lengths of the 2π and Stokes pulses. If $\tau_s \gg \tau$, the gain is given by $g = 1 + \tan^2(\pi\kappa)$; if $\tau_s \ll \tau$, g is considerably smaller: $g = 1 + (\tau_s/\tau)\pi\kappa \tan(\pi\kappa)$. The gain for radiation at frequency ω_2 becomes infinite for $\kappa = 1/2$, which corresponds to the first discrete eigenvalue for which the solution for E_2 is localized near the 2π -pulse. In dimensionless quantities, this condition is equivalent to $\mu_2^2\omega_2 = \mu_1^2\omega_1/4$, which is similar to the estimate found above. The number n of discrete states for $\kappa > 1/2$ is equal to

the integral part of 2κ , and the corresponding eigenvalues are $\nu(k) = 2\kappa - k$ for $k = 1, 2, \dots, n$. For large distances, the localized solutions ($\kappa > 1/2$) have the following limiting behavior in terms of dimensional quantities:

$$E_2 \approx \exp\left\{\frac{\nu}{\tau}\left(t - \frac{x}{c}\right)\right\}, \quad x \rightarrow +\infty, \quad (9)$$

$$E_2 \approx \exp\left\{-\frac{1}{\tau}\left[t - x\left(\frac{1+\nu}{v} - \frac{\nu}{c}\right)\right]\right\}, \quad x \rightarrow -\infty.$$

The Stokes wave is compressed if $\nu > 1$, i.e., if $\mu_2^2 \omega_2 > \mu_1^2 \omega_1$. If the ratio of the resonant transition oscillator strengths is large, the rise time of the amplified signal is substantially less than the pump pulse length. The length of the rising and falling edges of the Stokes signal depends on the velocity of the pumping 2π -pulse, which in turn depends on the pulse length (2). The falling edge is shorter than the rising edge if $c/v > 2\nu/(\nu + 1)$. If the ratio of the oscillator strengths is large ($\mu_2^2 \omega_2 \gg \mu_1^2 \omega_1$), the rising edge of the Stokes pulse is shorter than the falling edge if and only if $v < c/2$.

In order to calculate the efficiency of coherent SRS we must analyze the nonlinear stage of the pulse interaction. We did this in the general case by solving system (1) numerically; however, the nonlinear analysis can be carried out analytically if the oscillator strengths are equal.

3. SOME EXACT SOLUTIONS

Inverse scattering methods were used in Ref. 5 to study the interaction of radiation of two frequencies with a three-level system with a common lower level (the V -configuration) and equal oscillator strengths. This method can also be applied to the A -configuration under the same conditions $n_1 = n_2$, $\kappa^2 = \mu_2^2 \omega_2 / \mu_1^2 \omega_1 = 1$. We consider two matrix equations^{9,10}

$$-i\partial\varphi/\partial\tau = A\varphi, \quad (10)$$

$$i\partial\varphi/\partial\zeta = B\varphi \quad (11)$$

with the property that their consistency condition is equivalent to (1):

$$\partial A/\partial\zeta + \partial B/\partial\tau = i[A, B]. \quad (12)$$

In terms of the dimensionless variables

$$\tau = \Omega(t - x/c), \quad \zeta = \Omega x/c,$$

$$\varepsilon_{1,2} = \mu_{1,2} E_{1,2} / 2\hbar\Omega, \quad \Omega^2 = 2\pi N \mu_1^2 \omega_1 / \hbar,$$

we can choose the matrices A and B to be

$$A = \lambda J + U \begin{pmatrix} -\lambda & 0 & \varepsilon_1 \\ 0 & -\lambda & \varepsilon_2 \\ \varepsilon_1^* & \varepsilon_2^* & \lambda \end{pmatrix}, \quad B = \frac{1}{2\lambda} \rho, \quad (13)$$

where λ is the spectral parameter, J is a diagonal matrix with constant elements, and the fields of both of the pulses appear as the elements of the "potential" matrix U ; ρ is the density matrix of the medium: $\rho_{ij} = a_j a_i^*$.

The potential $U(\tau, \zeta)$ is expressible in terms of a matrix function $\psi(\tau, \zeta, \lambda)$ associated with the solution of (10), (11) (Ref. 9):

$$U = \lim_{|\lambda| \rightarrow \infty} \lambda[J, \psi]. \quad (14)$$

The number of zeros of the function $\psi(\lambda)$ in its domain of analyticity is equal to the number of solitons into which the potential $U(\tau, \zeta = 0)$ splits [$U(\tau, \zeta = 0)$ is specified in terms of the pulse fields $\varepsilon_{1,2}$ at the entrance to the medium]. The soliton parameters depend on the location of the zeros in the λ plane, which in turn is determined by $U(\tau, \zeta = 0)$. If there are N zeros $\lambda_1, \lambda_2, \dots, \lambda_N$, we can write $\psi(\lambda)$ in the form

$$\psi = 1 + \sum_{j=1}^N \frac{D_j}{\lambda - \mu_j}, \quad (15)$$

where $\mu_j = \lambda_j^*$ and the elements of the matrices D_j are given by

$$(D_j)_{kl} = (M_j)_{lk}(X_j)_l, \quad M_j(\tau, \zeta) = m_j(\zeta) \exp(i\lambda_j J \tau). \quad (16)$$

Like the zeros λ_j , the vectors $m_j(0)$ are determined by the potential at the input, and the dependence $m_j(\zeta)$ can be calculated using Eq. (11). We can find the vectors X_j by solving the equations

$$M_j + \sum_{k=1}^N \frac{M_j M_k}{\lambda_j - \mu_k} X_k = 0. \quad (17)$$

Equations (14)–(16) yield the expression

$$\varepsilon_1 = -2 \sum_i (M_i)_1 (X_i)_3, \quad \varepsilon_2 = -2 \sum_i (M_i)_2 (X_i)_3 \quad (18)$$

for the N -soliton solution.

In what follows we will need the scattering matrix $S(\lambda)$, which is defined by the expression

$$\varphi^-(\tau, \lambda) = \varphi^+(\tau, \lambda) S(\lambda), \quad (19)$$

where φ^\pm are two distinct fundamental matrices of solutions of Eq. (10); they are uniquely determined if we impose the asymptotic conditions $\varphi^\pm \rightarrow \exp(i\lambda J \tau)$ as $\tau \rightarrow \pm \infty$. $S(\lambda)$ can be factored as

$$S^+ = S S^-, \quad (20)$$

where S^+ and S^- are upper and lower triangular matrices whose diagonal elements are the principal minors of $S(\lambda)$:

$$S^+ = \begin{vmatrix} 1 & S_{12} S_{33} - S_{13} S_{32} & S_{13} \\ 0 & S_{22} S_{33} - S_{32} S_{23} & S_{23} \\ 0 & 0 & S_{33} \end{vmatrix}, \quad (21)$$

$$S^- = \begin{vmatrix} S_{22} S_{33} - S_{32} S_{23} & 0 & 0 \\ S_{23} S_{31} - S_{21} S_{33} & S_{33} & 0 \\ S_{21} S_{32} - S_{31} S_{22} & -S_{32} & 1 \end{vmatrix}.$$

The zeros of S^\pm determine the soliton parameters.^{9,10} Moreover, the vectors $m_j(0)$ span the kernel of the matrices $S^\pm(\lambda_j)$, which are singular for $\lambda = \lambda_j$:

$$m_j(0) = \ker S^\pm(\lambda_j) \quad (22)$$

[by the definition of the operator kernel, $S^\pm(\lambda_j) m_j = 0$]. Because of (22), Eq. (11) and the limiting behavior of the matrix B as $\tau \rightarrow \infty$ determine the dependence of m_j on ζ :

$$m_i(\xi) = \exp[-iB_\infty(\lambda_i)\xi] m_i(0), \quad B_\infty = B(\tau \rightarrow \infty). \quad (23)$$

We first consider the one-soliton solutions. Let the zero of the matrices S^\pm lie on the imaginary axis in the λ plane: $\lambda_0 = i\eta$, where η is real. (The case of arbitrary λ_0 , $\text{Re } \lambda_0 = \Delta\omega \neq 0$, corresponds to off-resonance coherent SRS. The solutions in this case are similar to the ones derived below, except that the dependence of the pump pulse velocity on the pulse length is different.) We will assume that the medium was in the ground state for $t \rightarrow -\infty$ and that all the particles were in level 1. Then in the general case we have

$$M(\tau, \xi) = \begin{pmatrix} \alpha_1 \exp(\eta\tau - \xi/2\eta) \\ \alpha_2 \exp \eta\tau \\ \alpha_3 \exp(-\eta\tau) \end{pmatrix}, \quad (24)$$

where the α_j are the components of the vector $m(0)$. We find from Eqs. (17), (18) that

$$\begin{aligned} \varepsilon_1 &= 4i\eta\alpha_1\alpha_3 \exp(-\xi/2\eta) D_1^{-1}, \quad \varepsilon_2 = 4i\eta\alpha_2\alpha_3 D_1^{-1}, \\ D_1 &= |\alpha_1|^2 \exp(2\eta\tau - \xi/\eta) + |\alpha_2|^2 \exp(2\eta\tau) + |\alpha_3|^2 \exp(-2\eta\tau). \end{aligned} \quad (25)$$

In terms of dimensional variables,

$$E_{1,2} = \frac{2\hbar}{\mu_{1,2}\tau} \left[\text{ch}\left(\frac{t-x/v_{1,2}}{\tau} - \varphi_{1,2}\right) + \frac{1}{2} \exp\left(\frac{t-x/v_{1,2}}{\tau} - \varphi_{1,2}\right) \times \exp\left(\pm \frac{x-x_0}{L}\right) \right]^{-1}. \quad (26)$$

Here both pulses are of length $\tau = (2\eta\Omega)^{-1}$, the velocity v_1 of pulse E_1 is related to τ by $c/v_1 = 1 + \Omega^2\tau^2$, $v_2 = c$, and the constants $\varphi_{1,2}$ determine the soliton coordinate and depend on which fields enter the medium: $\varphi_{1,2} = \ln(\alpha_3/\alpha_{1,2})$. Solution (26) shows that for $x \rightarrow -\infty$ all of the soliton energy is concentrated in the pumping 2π -pulse,

$$E_1 \rightarrow \frac{2\hbar}{\mu_1\tau} \text{sech}\left(\frac{t-x/v_1}{\tau} - \varphi_1\right), \quad E_2 \rightarrow 0, \quad x \rightarrow -\infty. \quad (27)$$

When $x \rightarrow +\infty$, the energy of pulse E_1 is completely transferred to the pulse E_2 , which travels at $v_2 = c$ without attenuation and is also of area 2π , i.e.,

$$E_1 \rightarrow 0, \quad E_2 \rightarrow \frac{2\hbar}{\mu\tau} \text{sech}\left(\frac{t-x/v_2}{\tau} - \varphi_2\right), \quad x \rightarrow +\infty. \quad (28)$$

The characteristic energy transfer length

$$L = c/2\Omega^2\tau \sim v\tau/2 \quad (29)$$

is comparable to the length of pulse E_1 and 100% conversion occurs over a distance $x_0 = 2L \ln(\alpha_1/\alpha_2)$. For $\alpha_1 \neq 0$, $\alpha_2 = 0$ and $\alpha_2 \neq 0$, $\alpha_1 = 0$, (25) specializes to solutions which describe the propagation of the single-frequency 2π -pulses E_1 and E_2 , respectively.

To construct the two-soliton solutions, we must specify the two zeros λ_1, λ_2 or S^\pm in the complex λ plane (we take them to lie on the imaginary λ axis, so that the fields are in exact resonance with the corresponding transitions) and the two vectors

$$m_1 = \ker S^+(\lambda_1) = \begin{pmatrix} \alpha_1 \\ \alpha_2 \\ \alpha_3 \end{pmatrix}, \quad m_2 = \ker S^+(\lambda_2) = \begin{pmatrix} \beta_1 \\ \beta_2 \\ \beta_3 \end{pmatrix} \quad (30)$$

The solution is readily found from (17), (18), and (30). If all the components of m_1, m_2 are nonzero, the solution describes 100% transfer of energy from the two pumping 2π -pulses to the Stokes wave.

We will omit the elaborate expression for the exact solution, which is the two-soliton analog of (26). The behavior of the two-soliton solutions is very different if some of the components $\alpha_{1,2}, \beta_{1,2}$ of the vectors (30) vanish ($\alpha_3 \neq 0, \beta_3 = 0$). For example, if $\alpha_2 = \beta_2 = 0$ then the two-soliton solution describes a collision between two 2π pumping pulses in a two-level medium and is given by the expression in Ref. 9. If $\alpha_1 = \beta_1 = 0$, the solution describes two Stokes signal pulses traveling at the speed of light in a transparent medium (i.e., without attenuation). If one of α_2 or β_2 vanishes, the two-soliton solution describes partial energy transfer to the Stokes signal and is a superposition of the two 2π pumping pulses for $t \rightarrow -\infty$ and a superposition of the Stokes signal and pumping pulse (both of area 2π for $t \rightarrow +\infty$).

The case $\beta_1 = \alpha_2 = 0$ is of interest with regard to the interaction of radiation with a three-level medium in the A -configuration. The two-soliton solution in this case has the form

$$\begin{aligned} E_{1,2} &= \frac{2\hbar}{\mu_{1,2}} \left[\tau_{2,1} \text{ch}\left(\frac{t-x/v_{2,1}}{\tau_{2,1}} - \varphi_{2,1}\right) - \left(\frac{1}{\tau_1} + \frac{1}{\tau_2}\right)^{-1} \right. \\ &\quad \left. \times \exp\left(-\frac{t-x/v_2}{\tau_{2,1}} + \varphi_{2,1}\right) \right] D_2^{-1}, \\ D_2 &= \tau_1\tau_2 \text{ch}\left(\frac{t-x/v_1}{\tau_1} - \varphi_1\right) \text{ch}\left(\frac{t-x/v_2}{\tau_2} - \varphi_2\right) - \left(\frac{1}{\tau_1} + \frac{1}{\tau_2}\right)^{-2} \\ &\quad \times \exp\left(-\frac{t-x/v_1}{\tau_1} + \varphi_1\right) \exp\left(-\frac{t-x/v_2}{\tau_2} + \varphi_2\right), \quad (31) \\ v_2 &= c, \quad c/v_1 = 1 + \Omega^2\tau_1^2, \quad \varphi_1 = \ln(\alpha_3/\alpha_1), \quad \varphi_2 = \ln(\beta_3/\beta_2). \end{aligned}$$

This solution is completely analogous to the two-soliton solution for a V -medium.⁵ It describes the passage of a 2π -pulse E_2 through a 2π -pulse E_1 in such a way that the amplitude, form, and velocity of both pulses are unchanged: the only change is that the pulse phases are shifted by $\phi = \ln[(\tau_1 + \tau_2)/(\tau_1 - \tau_2)]$ relative to one another. The pulse E_2 moves in a transparent medium apart from the region of interaction with pulse E_1 , where it is amplified by the medium. If $\tau_1 \approx \tau_2$, the pulse interaction length and phase shifts increase logarithmically because the competing effects of amplification, dispersion, and drift become comparable. The passage of the Stokes 2π -pulse through a 2π pumping pulse without any transfer of energy has a simple explanation if $\tau_s \ll \tau$. It is known⁴ that when a 2π -pulse crosses a resonant medium (absorbing or amplifying), it leaves the medium in its initial state. When the Stokes pulse travels near the 2π pumping pulse, the leading edge of the Stokes pulse takes the particle from state 3 to state 2, after which the trailing edge returns the particle to state 3. The trailing edge of the pumping pulse puts the entire system into the initial state, for

which $|a_1| = 1$. The net result of this interaction is that there is no change in the upper level populations and hence no energy transfer from one pulse to the other.

In practice, the form of the input pulses generally differs from the soliton solutions for all x and t . From a practical viewpoint it is therefore of interest to consider the time evolution of the Stokes wave and pump pulses when the pulse shapes at the entrance to the medium are specified. The pulse shapes will be described by soliton solutions for sufficiently large distances. Some of these soliton solutions describe 100% transfer from the pumping pulse to the Stokes signal, others describe partial transfer, while still others describe pulse interaction with no energy transfer.

In order to ascertain which case actually occurs, we will systematically examine what happens when two square-wave pulses enter the medium simultaneously,

$$\varepsilon_1 = \begin{cases} 0, & \tau < 0, \tau > \tau_1 \\ u_1, & 0 < \tau < \tau_1 \end{cases}, \quad \varepsilon_2 = \begin{cases} 0, & \tau < 0, \tau > \tau_1 \\ u_2, & 0 < \tau < \tau_1 \end{cases}. \quad (32)$$

We factor the scattering matrix S and note that only the zeros of the element S_{33} in (21) are relevant as far as solitons are concerned. All three components $\alpha_{1,2,3}$ of the vector $m(0)$ spanning $\ker S^+(\lambda_0)$ are in general nonzero, so that we have two nonvanishing fields $E_{1,2}$ [cf. Eq. (25)]. However, if $S_{33} \neq 0$ and $S_{22}S_{33} = S_{23}S_{32}$, the condition $S^+(\lambda_0)m(0) = 0$ automatically yields $\alpha_3 = 0$, i.e., $E_1 = E_2 = 0$. We will therefore consider only the case $S_{33} = 0$. The matrix $S^+(\lambda_0)$ is then given by

$$S^+ = \begin{pmatrix} 1 & \frac{u_1 u_2^*}{(|u_1|^2 + |u_2|^2)} & i \frac{u_1}{\gamma} \exp(i\lambda_0 \tau_1) \sin \gamma \tau_1 \\ 0 & \frac{|u_1|^2}{(|u_1|^2 + |u_2|^2)} & i \frac{u_2}{\gamma} \exp(i\lambda_0 \tau_1) \sin \gamma \tau_1 \\ 0 & 0 & 0 \end{pmatrix}, \quad (33)$$

where $\gamma = (\lambda_0^2 + |u_1|^2 + |u_2|^2)^{1/2}$. The value λ_0 is determined by the equation

$$\cos \gamma \tau_1 + i(\lambda_0/\gamma) \sin \gamma \tau_1 = 0. \quad (34)$$

The threshold condition for a single zero $\lambda_0 = i\eta$ to appear in Eq. (34) is

$$\pi < (\theta_1^2 + \theta_2^2)^{1/2} < 3\pi, \quad \theta_{1,2} = 2 \int_{-\infty}^{\infty} u_{1,2} d\tau.$$

Although this condition coincides with the threshold for formation of simultons (two-frequency pulses which propagate together in a V -medium),^{5,10} the nature of the solution is completely different. Indeed, if we solve $S^+ m = 0$ for $u_2 \neq 0$, we get

$$\alpha_1 = 0, \quad \alpha_2 = -i\alpha_3 \frac{|u_1|^2 + |u_2|^2}{\gamma u_2^*} \exp(-\eta \tau_1) \sin \gamma \tau_1. \quad (35)$$

The asymptotic form of the resulting pulse can be found by substituting $\alpha_{1,2,3}$ from (35) into the solution (25). We find that a single Stokes wave is formed for arbitrarily small input fields u_2 , i.e., all of the energy in the pumping field is transferred to the Stokes signal. The conversion length diverges logarithmically as $u_2 \rightarrow 0$.

The number of solutions of Eq. (34) increases with the pumping field amplitude (energy per pumping pulse). For $3\pi < (\theta_1^2 + \theta_2^2)^{1/2} < 5\pi$ the matrix $S^+(\lambda)$ has two zeros, and the pulses which are produced subject to the initial conditions (32) are given asymptotically by

$$\varepsilon_2 = 2 \frac{t_1 - t_2}{t_1 + t_2} \frac{t_1 \Phi_1 - t_2 \Phi_2}{t_1 t_2 \Phi_1 \Phi_2 - 2t_{12}^2 (1 + \Phi_{12})},$$

$$\Phi_{1,2} = \text{ch} \left(\frac{t-x/c}{t_{1,2}} - \varphi_{1,2} \right), \quad \Phi_{12} = \text{ch} \left(\frac{t-x/c}{t_{12}} - \varphi_{12} \right),$$

$$\frac{1}{t_{12}} = \frac{1}{t_1} + \frac{1}{t_2}, \quad (36)$$

$$\varphi_{12} = \varphi_1 + \varphi_2,$$

$$\varepsilon_1 = 0, \quad \varphi_{1,2} = \ln \left[\frac{u_2}{\gamma_{1,2}} \exp \frac{\tau_1}{t_{1,2}} \sin \gamma_{1,2} \tau_1 \right],$$

where $\lambda_{1,2}$ are the roots of Eq. (34) and $t_{1,2} = \tau_1^{-1} \text{Im} \lambda_{1,2}$. The solution (36) describes energy transfer from the pump field to the Stokes pulse, which has two peaks and travels at the speed of light. The first peak is shorter than the second, and in each case the energies are proportional to the durations. The form of the solutions is analogous for high pump pulse energies at the input—all the energy is transferred to the Stokes signal, which has N peaks, where N is the number of roots of Eq. (36).

Another case which can be solved completely occurs when the propagating pulses are nonoverlapping square waves at the input:

$$\varepsilon_1 = \begin{cases} 0, & \tau < 0, \tau > \tau_1 \\ u_1, & 0 < \tau < \tau_1 \end{cases}, \quad \varepsilon_2 = \begin{cases} 0, & \tau < \tau_2, \tau > \tau_3 \\ u_2, & \tau_2 < \tau < \tau_3 \end{cases}, \quad (37)$$

$$0 < \tau_1 < \tau_2 < \tau_3.$$

The matrix S^+ is then

$$S^+ = \begin{pmatrix} S_{11} & S_{12} & S_{13} \\ S_{21} & S_{22} & S_{23} \\ S_{31} & S_{32} & S_{33} \end{pmatrix}, \quad (38)$$

where

$$S_{11} = 1, \quad S_{21} = S_{31} = S_{32} = 0,$$

$$S_{12} = \frac{u_1 u_2^*}{\gamma_1 \gamma_2} \exp[i\lambda_0 (\tau_1 - \tau_2 - \tau_3)] \sin \Gamma_1 \sin \Gamma_2,$$

$$S_{13} = \frac{i u_1}{\gamma_1} \sin \Gamma_1 \exp(i\lambda_0 \tau_1),$$

$$S_{22} = \left(\cos \Gamma_1 + i \frac{\lambda_0}{\gamma_1} \sin \Gamma_1 \right) \exp(-i\lambda_0 \tau_1),$$

$$S_{23} = \frac{i u_2}{\gamma_2} \exp[-i\lambda_0 (\tau_1 - \tau_2 - \tau_3)]$$

$$\times \sin \Gamma_2 \left(\cos \Gamma_2 + \frac{i\lambda_0}{\gamma_2} \sin \Gamma_2 \right),$$

$$S_{33} = \exp[-i\lambda_0 (\tau_1 + \tau_3 - \tau_2)] \left(\cos \Gamma_1 + \frac{i\lambda_0}{\gamma_1} \sin \Gamma_1 \right)$$

$$\times \left(\cos \Gamma_2 + \frac{i\lambda_0}{\gamma_2} \sin \Gamma_2 \right).$$

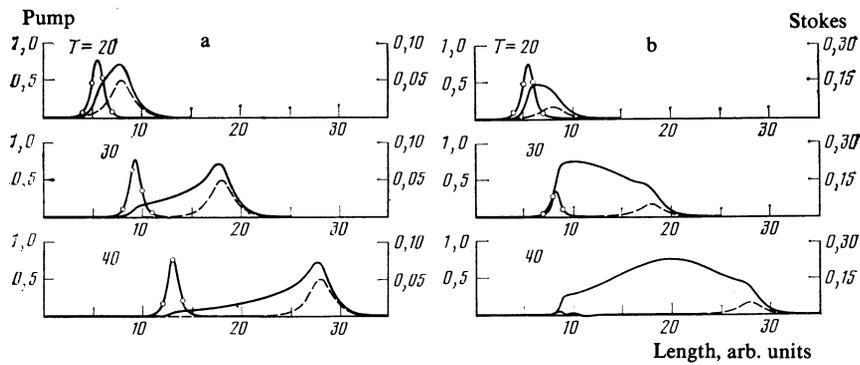


FIG. 1. Pulse interaction near the critical point $\kappa = 0.5$: a) $\kappa = 0.4$; b) $\kappa = 0.6$. In both cases, the 2π pumping pulse (of duration $\tau_1 = 1.2247$ and velocity $v_1 = 0.4$) enters the medium after a delay $T_1 = 6$ (cf. the curve with the open circles). The weak Stokes seed wave ($\tau_2 = 1.0$, $v_2 = 1.0$, amplitude $\alpha_2 = 0.05$) enters the medium after a delay $T_2 = 12$ (solid curve). For clarity, the propagation of the Stokes wave in the absence of interaction is also shown (dashed curve).

Here $\Gamma_1 = \gamma_1 \tau_1$, $\Gamma_2 = \gamma_2 (\tau_3 - \tau_2)$, $\gamma_{1,2} = (\lambda^2 + |u_{1,2}|^2)^{1/2}$. The threshold condition (26) for a soliton solution to appear is seen to coincide with the condition for formation of the 2π -pulse E_1 . The soliton parameters are determined by the constants

$$\begin{aligned} \alpha_1 &= -i \frac{u_1}{\gamma_1} \exp(i\lambda_0 \tau_1) \sin \Gamma_1 \left(1 - \frac{|u_2|^2}{\gamma_2^2} \sin^2 \Gamma_2 \right), \\ \alpha_2 &= -i \frac{u_2}{\gamma_2} \exp[i\lambda_0 (\tau_2 + \tau_3)] \sin \Gamma_2, \\ \alpha_3 &= 1, \quad \lambda_0 = i\eta, \quad \tau = \frac{1}{2\eta\Omega}. \end{aligned} \quad (39)$$

Thus for arbitrarily small input fields $u_2 \neq 0$, all three components $\alpha_{1,2,3}$ are nonzero, which implies that all of the energy in E_1 is transferred to E_2 . The conversion length diverges logarithmically as the amplitude u_2 of E_2 tends to zero:

$$\begin{aligned} x_0 &= 2L \ln \left[\frac{u_1/\gamma_1}{u_2/\gamma_2} \exp \frac{2(\tau_3 + \tau_2 - \tau_1)}{\tau} \frac{\sin \Gamma_1}{\sin \Gamma_2} \right. \\ &\quad \left. \times \left(1 - \frac{|u_2|^2}{\gamma_2^2} \sin^2 \Gamma_2 \right) \right]. \end{aligned} \quad (40)$$

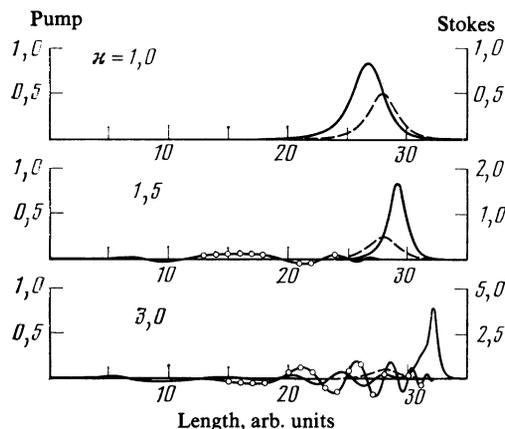


FIG. 2. Evolution of the pulses in media with $\kappa = 1, 1.5$, and 3 at equal times, for the same input pulse parameters as in Fig. 1. The amplification, pulse shortening, and shift ahead of the Stokes signal are seen to increase with κ . In addition, for large κ there is a prolonged transfer of excitation on the trailing edge of the pulses, which results in an elongated weakly damped oscillating "tail." The Stokes seed (dashed curve) is shown to 1:10 scale (its amplitude α_2 is equal to 0.05).

The above examples of evolving square-wave input pulses show that in general, the electromagnetic field energy is concentrated in the Stokes signal at asymptotically large distances. This occurs because the soliton solutions which do not describe energy transfer to the Stokes signal are unstable with respect to decay of the pumping pulses. This assertion was proved above for the special case of a 2π pumping pulse and has been confirmed in various typical cases by solving (1) numerically.

4. NUMERICAL CALCULATIONS

The exact nonlinear calculations show that in all of the cases considered, a significant amount of energy is transferred to the Stokes frequency when pulses interact during coherent SRS. However, the following question arises (it is

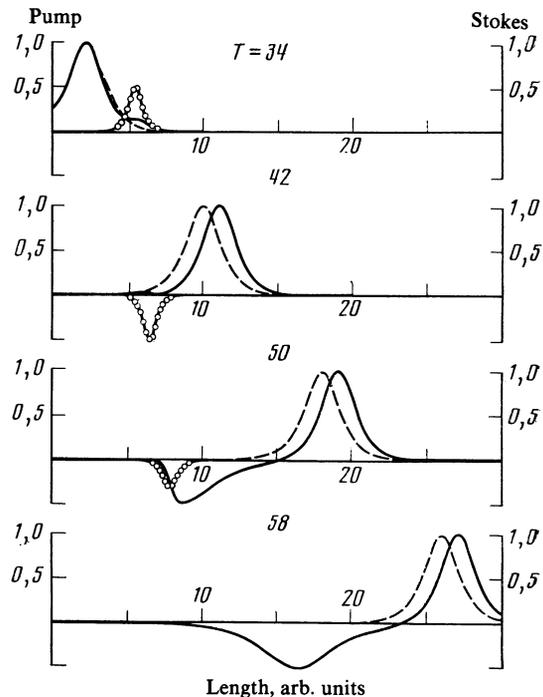


FIG. 3. Collision of two 2π -pulses in a medium with $\kappa = 1.0$. The Stokes pulse ($\tau_2 = 1.0$) enters the medium after a delay $T_2 = 32$ and overtakes the pumping 2π -pulse ($\tau_1 = 2.0$, $v_1 = 0.2$, $T_1 = 7$). At time $T = 42$ the field pattern corresponds to passage of the Stokes pulse through the pumping pulse [cf. Eq. (31)]; however, this solution is unstable, so that all of the energy is transferred to the second Stokes pulse.

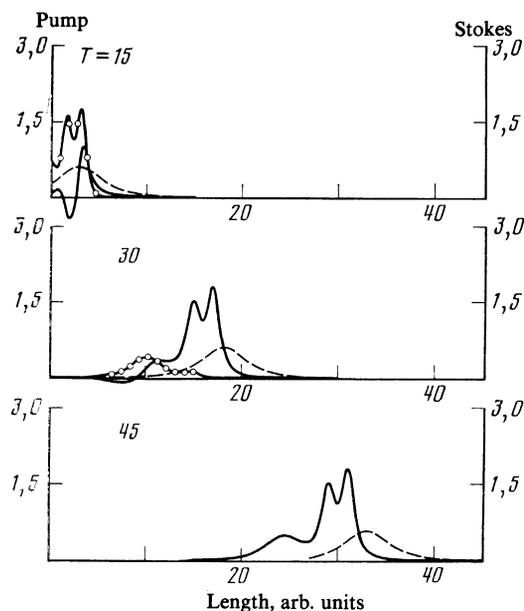


FIG. 4. Situation for a pump pulse of large area (6π); $\tau_1 = \tau_2 = 2.0$, $T_1 = T_2 = 12$; $\alpha_1 = 3.0$, $\alpha_2 = 0.05$. The competition between pumping of the Stokes pulse and decomposition of the pump pulse into separate 2π -pulses is evident. The final form of the Stokes pulse repeats the form of the pumping pulse at the instant the pumping starts to dominate over decomposition. The Stokes seed (dashed curves) is not shown to scale.

associated with the special nature of the case $\kappa = 1$, which can be solved by the inverse scattering method): Is the pumping of the Stokes signal merely a consequence of a hidden symmetry of system (1) for $\kappa = 1$, or is it a general property of coherent SRS valid whenever the trapping condition $\kappa > 1/2$ is satisfied? In order to answer this question we must study the nonlinear stage of scattering for $\kappa \neq 1$. To this end we carried out numerical experiments covering the interval $0.3 \leq \kappa \leq 3.0$. The difference scheme was a modification of the one in Ref. 11; it was second-order in space and time and was fully conservative in the sense that the integrals

$$|a_1|^2 + |a_2|^2 + |a_3|^2 = 1,$$

$$\int_0^l \frac{E_1^2 dx}{8\pi\hbar\omega_1} \Big|_{\tau=t} + N \left(l - \int_0^l |a_1|^2 dx \right) \Big|_{\tau=t} + \int_0^l \frac{cE_1^2}{8\pi\hbar\omega_1} d\tau \Big|_{x=0}^l = 0,$$

$$\int_0^l \frac{E_2^2 dx}{8\pi\hbar\omega_2} \Big|_{\tau=t} - N \int_0^l |a_2|^2 dx \Big|_{\tau=t} + \int_0^l \frac{cE_2^2}{8\pi\hbar\omega_2} d\tau \Big|_{x=0}^l = 0,$$

were left invariant (here l is the length of the medium). These integrals express conservation of the number of quanta in the field and in the resonant medium participating in the interaction. We used two methods to test the accuracy of the difference scheme. First, we compared the solution found numerically for $\kappa = 1$ with the exact analytic solution; second, we monitored the accuracy for $\kappa \neq 1$ by performing test calculations with a smaller mesh size. We found that the solutions were accurate when a grid spacing equal to one-tenth the characteristic pulse length was employed. The error in the solution ranged from 10^{-2} to 10^{-3} , depending on the problem.

Figure 1 illustrates the markedly different character of the interaction for $\kappa < 1/2$ and $\kappa > 1/2$. For example, when $\kappa = 0.4$ (Fig. 1a) the Stokes pulse is not significantly amplified; the pumping pulse gives up only a small fraction of its energy, and its area remains equal to 2π . The duration of the Stokes signal also increases appreciably. By contrast, for $\kappa = 0.6$ (Fig. 1b), all of the energy of the 2π pumping pulse is transferred to the Stokes wave. The trapping of the trailing edge of the Stokes pulse in the interaction region is apparent in this case, and the SRS quantum efficiency is 100%. The conversion length and the relations between the leading and trailing edges of the Stokes signal are in complete agreement with the results of the linear analysis. For $\kappa > 1$, most of the energy is converted into the Stokes signal over short distances in the medium, and considerable modulation of the trailing pulse edges is observed. Figure 2 shows some pulse-forms calculated for identical times for three oscillator strength ratios $\kappa = 1, 1.5, 3$. Figure 3 illustrates a collision between two 2π -pulses (a Stokes pulse and a pumping pulse). The two pulse shapes at the input were specified so that the pulse lengths were much less than the time interval between them. To within an exponentially small term, this initial condition coincides with the asymptotic behavior of the two-soliton solution (31) for $t \rightarrow -\infty$, which describes a collision of pulses without any energy transfer. During the initial stage of the collision, the pulses pass through one another as described by solution (31); however, after the Stokes 2π -pulse overtakes the pumping 2π -pulse, the latter transfers energy to the Stokes wave. This occurs because the two-soliton solution (31) becomes unstable, with the result that all of the energy is transferred to the Stokes signal if the initial conditions differ even very slightly from the exact solution. Figure 4 illustrates the coherent SRS process for an input pulse of large area (6π). In this case, all of the energy is transferred to the Stokes signal, which has three peaks.

We thank A. P. Napartovich and A. M. Dykhne for helpful discussions of the above results.

¹R. Burnham and N. Djeu, *Opt. Lett.* **3**, 215 (1978).

²N. Djeu, *Appl. Phys.* **35**, 663 (1979).

³S. L. McCall and E. L. Hahn, *Phys. Rev.* **183**, 457 (1969).

⁴I. E. Poluëktov, Yu. M. Popov, and V. S. Roitberg, *Kvantovaya Élektron. (Moscow)* **4**, 557 (1974) [*sic*]; *Usp. Fiz. Nauk* **114**, 97 (1974) [*Sov. Phys. Usp.* **17**, 673 (1974)].

⁵L. A. Bol'shov, V. V. Likhanskii, and M. I. Persiantsev, *Zh. Eksp. Teor. Fiz.* **84**, 903 (1983) [*Sov. Phys. JETP* **57**, 524 (1983)].

⁶A. V. Nazarkin, I. A. Poluektov, and I. I. Sobel'man, *Pis'ma Zh. Eksp. Teor. Fiz.* **37**, 313 (1983) [*JETP Lett.* **37**, 371 (1983)].

⁷A. Erdelyi (ed.), *Higher Transcendental Functions*, Vol. 1, McGraw-Hill, New York (1953).

⁸L. D. Landau and A. M. Lifshits, *Quantum Mechanics: Non-Relativistic Theory*, Pergamon Press, Oxford (1965).

⁹V. E. Zakharov, S. V. Manakov, S. P. Novikov, and L. P. Pitaevskii, *Theory of Solitons: Inverse Scattering Method* [in Russian], Nauka, Moscow (1980).

¹⁰L. A. Bol'shov, V. V. Likhanskii, and M. I. Persiantsev, *Inst. At. Energ. Preprint No. 3685*, Moscow (1982).

¹¹L. A. Bol'shov, N. N. Elkin, T. K. Kirichenko, V. V. Likhanskii, and M. I. Persiantsev, *Inst. At. Energ. Preprint No. 3732*, Moscow (1983).

Translated by A. Mason