Nonlinear synchronization of the pump and Stokes-wave pulses in coherent resonant stimulated Raman scattering

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

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A new effect has been discovered: a nonlinear locking of the Stokes wave by the pump pulse in the coherent regime of stimulated Raman scattering in a resonant three-level medium. The effect causes a complete transfer of energy from the pump pulse to the Stokes signal.

1. Stimulated Raman scattering in resonant media has been attracting increased theoretical and experimental interest because of the possibility of achieving efficient frequency conversion and of substantially shortening light pulses under these conditions. Burnham and Djeu,[1,2] for example, have achieved frequency conversion and have altered the length of the output pulses from excimer lasers using the vapor of several metals at an efficiency $\sim 50\%$. Nazarkin *et al.* have pointed out that it may be possible to substantially reduced the pulse length in a resonant three-level medium with a large ratio of oscillator strengths in a coherent interaction with a Stokes wave. Since the propagation velocities of light pulses are greatly different in a three-level medium, the conversion efficiency, which is determined by the total gain over the pulse-interaction distance (see Ref. 3, for example), should be low. In this letter we wish to point out a new effect: a nonlinear locking of a Stokes wave in the coherent regime of resonant stimulated Raman scattering, which may raise the conversion efficiency to 100%.

2. At a pulse length shorter than all the relaxation times of the medium, the equations for the filling amplitudes of the levels, a_1, a_2, and a_3, and for the smooth envelopes of the pump field (E_1) and the Stokes wave (E_2) are (Fig. 1)

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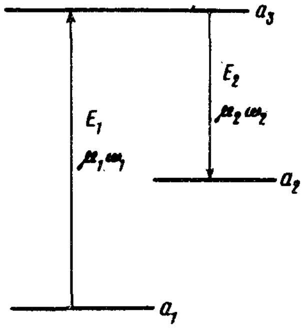

FIG. 1. Level scheme.

$$\frac{\partial a_1}{\partial t} = \frac{i}{2\hbar}\mu_1 E_1 a_3, \qquad \frac{\partial a_2}{\partial t} = \frac{i}{2\hbar}\mu_2 E_2 a_3, \qquad \frac{\partial a_3}{\partial t} = \frac{i}{2\hbar}(\mu_1 E_1^* a_1 + \mu_2 E_2^* a_2),$$
$$i\left(\frac{\partial E_1}{\partial t} + \frac{c}{n_1}\frac{\partial E_1}{\partial x}\right) = 4\pi N \mu_1 \omega_1 a_1 a_3^*, \qquad i\left(\frac{\partial E_2}{\partial t} + \frac{c}{n_2}\frac{\partial E_2}{\partial x}\right) = 4\pi N \mu_2 \omega_2 a_2 a_3^*. \tag{1}$$

Here N is the density of resonant particles, and $\mu_{1,2}$, $\omega_{1,2}$, and $n_{1,2}$ are the dipole moments, frequencies, and nonresonant refractive indices of the 1–3 and 2–3 transitions. We consider the gain of a weak Stokes signal on the 1–3 transition during the propagation of a 2π pulse. The amplitude of the upper level is $a_3 = \mathrm{ch}^{-1}[(t - x/v_0)/\tau_0]$, where $c/v_0 = 1 + 2\pi N\mu_1^2\omega_1\tau_0^2/\hbar = 1 + \Omega^2\tau_0^2$; from (1) we find the following equation for the Stokes wave:

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

Equation (2) describes an intensification of the field E_2 in a layer of thickness $v_0\tau_0$ with a population inversion. It is important to note that the increment in E_2 in the region of the pump pulse is relatively great if it varies relatively slowly over time. We can easily estimate $\mu_2^2\omega_2/\mu_1^2\omega_1 \gtrsim 1$ as the condition under which the removal of the field E_2 from the interaction region becomes less pronounced than the amplification in this region. In this case, even if the propagation velocity of E_2 outside the interaction region is much larger than v_0, the Stokes wave will grow in this region. These arguments can be proved by transforming to an equation for the quantity $\mu = \int_{-\infty}^{t} a_3(x,t')E_2(x,t')dt'$ and to the independent variables

$$z = n_2 x/c\tau_0, \qquad \xi = \frac{t - x/v_0}{\tau_0} \quad \text{and} \quad y = \frac{1}{2}\left(1 + \mathrm{th}\,\xi\right).$$

The boundary-value problem for $\mu \sim e^{\gamma z}$ corresponds to no increase in E_2 in the limits $\xi \to \pm\infty$. In this case Eq. (2) is the hypergeometric equation depending on the parameters[4] $a^2 = [(\mu_2^2\omega_2)/(\mu_1^2\omega_1)][(c/n_1v_0 - 1)/(c/n_2v_0 - 1)]$, $A = (\gamma/\Omega^2\tau_0^2)$

$\times [(c/n_1 v_0 - 1)/(c/n_2 v_0 - 1)]$. If $a < 1/2$, there is a continuous spectrum of solutions ($A = iB$, where B is some arbitrary real number) describing a gain of a continuous signal with a frequency shift $\Delta = B/\tau_0$. The total gain is

$$G(B) = \left| \frac{E_2(\xi \to -\infty)}{E_2(\xi \to +\infty)} \right|^2 = \frac{\mathrm{ch}^2 \dfrac{\pi B}{2}}{\mathrm{ch}^2 \dfrac{\pi B}{2} - \sin^2 \pi a} \tag{3}$$

It follows from (3) that the gain is generally on the order of unity, in agreement with the usual estimates. If the length of the Stokes signal satisfies $\tau_s \ll \tau_0$, the gain is even smaller:

$$G = 1 + \frac{\tau_s}{\tau_0} \pi a \,\mathrm{tg} \pi a . \tag{4}$$

If $a = 1/2, \mu_2^2 \omega_2 / \mu_1^2 \omega_1 = 1/4$, however, the gain in (3) and (4) becomes infinite. A first discrete level appears at this value of a; the solution E_2 localizes near E_1, and it falls off in the limits $\xi \to \pm \infty$. The number of discrete levels is the greatest integer $n = [2a]$, and the corresponding eigenvalues are $A_k = 2a - k$, $k = 1, 2, \cdots, n$.

Asymptotically localized solutions for the field E_2 in the limits $\xi \to \pm \infty$ are

$$E_2 \sim \exp(-\xi + \gamma_k z), \quad \xi \to +\infty; \quad E_2 \sim \exp(A_k \xi + \gamma_k z), \quad \xi \to -\infty .$$

The appearance of a discrete level (localized solutions) corresponds to a nonlinear synchronization of the Stokes wave and the pump wave, i.e., to an unbounded growth of the Stokes pulse in an unbounded medium.

3. In the linear approximation, of course, we cannot determine the maximum conversion efficiency. With $a = 1$ (i.e., in the locking region) the nonlinear regime of the stimulated Raman scattering can be analyzed completely by the method of the inverse scattering problem. Equations (1) are equivalent to the compatibility condition for the matrix equations ($A'_\xi + B'_\tau = [A, B]$)

$$-i\varphi_\tau = A\varphi ; \quad i\varphi_\xi = B\varphi ,$$

if we choose

$$A = \begin{pmatrix} -\lambda & 0 & E_1 \\ 0 & -\lambda & E_2 \\ E_1^* & E_2^* & \lambda \end{pmatrix}, \qquad B = \frac{1}{2\lambda} (a_i a_j^*) . \tag{6}$$

This problem is similar to a problem which we have analyzed previously,[5] that of simultons in a three-level medium. We proceed immediately to the results found from the inverse scattering problem, postponing the intermediate calculations for publication in a detailed paper. The simplest solution which describes a complete transfer of energy to the Stokes wave is

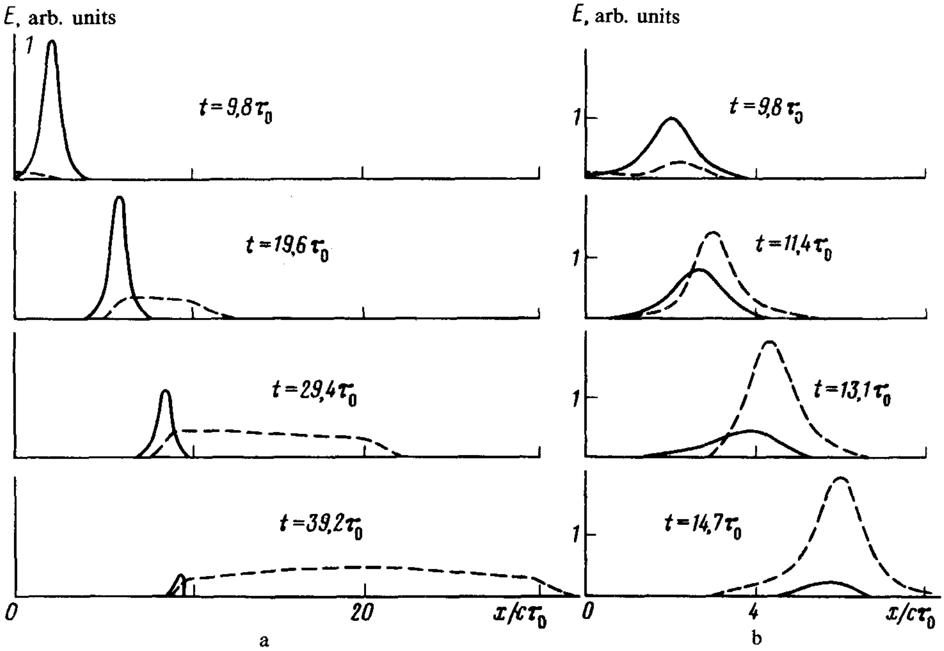

FIG. 2. Evolution of the light pulses during resonant stimulated Raman scattering. Solid curves—Pump pulse; dashed curves—Stokes pulse. The amplitude of the input pulse, E_2, is $5\times10^{-2}E_1$. a: $(\mu_2^2\omega_2/\omega_1^2\omega_1)^{1/2} = 0.55$. b: $(\mu_2^2\omega_2/\mu_1^2\omega_1)^{1/2} = 1.5$.

$$E_{1,2} = \frac{2\hbar}{\mu_{1,2}\tau}\left[\mathrm{ch}\left(\frac{t - x/v_{1,2}}{\tau} - \varphi_{1,2}\right) + \frac{1}{2}\exp\left(\frac{t - x/v_{1,2}}{\tau} - \varphi_{1,2}\right)\exp\left(\pm\frac{x - x_0}{L}\right)\right]^{-1}, \tag{7}$$

where τ is the length of each of the pulses; $v_1 = v$; $v_2 = c$, $L = c/2\Omega^2\tau$; and $\varphi_{1,2}$ and x_0 are determined by the initial conditions. Analysis of the problem with the initial conditions $E_{1,2}(x_0,t)$ shows that a threshold is involved in the onset of solution (2); the threshold condition is the same as the condition for the appearance of a 2π pulse E_1. Consequently, no matter how low the input level E_2, there is a complete transfer of the energy of the 2π pulse E_1 to the 2π pulse E_2. The conversion length increases logarithmically with decreasing input amplitude.

4. Equations (1) have been solved numerically for an arbitrary ratio of oscillator strengths. Figure 2 shows the results for $a = 0.55$ and $1 = 1.5$. At $a = 0.55$ the leading edge characteristically stretches out; this stretching can be explained well by a linear analysis and by a qualitative picture of the effect. We see from this figure that the 100% interaction efficiency is not reached because the case $a = 1$ is a special one. This circumstance is particularly important for experiments on resonant stimulated Raman scattering in the coherent regime.

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