Coherent Raman light conversion in a two-level medium

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The inverse scattering problem method is used to investigate Raman scattering in time intervals much shorter than the relaxation times of the medium. Cases are considered in which the energy transfer to the Stokes field is initiated by polarization fluctuations and by a Stokes-field pulse of small area. The fluctuations of the pump field are taken into account. It is shown that the joint action of the pump-intensity and polarization fluctuations produces a spike in the solution that describes the shape of the transmitted pump-field pulse. The compression of the Stokes-field fluctuations during the strongly nonlinear stage of Raman scattering is explained on the basis of the obtained quasi-self-similar solution.

INTRODUCTION

Nonresonant Raman scattering (RS) of light in interaction with a two-level medium over times much shorter than the relaxation time is widely used in various branches of physics and chemistry. In experiments aimed at observing RS it is customary to apply to the end face of the sample a high-power pump pulse, and the transport of the energy into the Stokes field is initiated either by quantum fluctuations of the system or by a small priming Stokes pulse. In the former case, a collective effect can be observed. This effect, cooperative Raman scattering (CRS),¹⁻⁴ was observed in RS experiments in hydrogen.^{2,3} CRS is the analog of a well known collective effect, viz., one-photon superradiance.^{4,5} CRS was described theoretically in a number of papers¹⁻³ on the basis of the McGillivray-Feld equivalent superradiance model.⁶ It was shown that the form of a Stokes pulse in the constantpump approximation is described by a self-similar solution of the sine-Gordon equation (nondegenerate transition 1,2) or its generalization (degenerate transition²). The second case-initiation of RS by a small Stokes pulse-corresponds to coherent Raman amplification (CRA). The theory of CRA generalizes the theory of the one-photon laser amplifier, the properties of which were investigated by Manakov⁷ by the inverse scattering problem methods (ISPM).⁸ Manakov has shown that the waveform of a light pulse after passing through a long enough active medium is described by a quasi-self-similar solution. The ISPM is used in the present paper to describe the evolution of the fields in coherent RS. This method was first used by Kaup⁹ and Steudel¹⁰ to investigate the Maxwell-Bloch equations that describe RS. In the latter paper, and also in Ref. 11, N-soliton solutions were obtained. A non-soliton behavior of the solutions of these equations is investigated here for the first time.

In many papers, quantum effects that determine the initial stage of superradiance^{6,12,13} and of CRS¹⁻³ are simulated quite adequately by a random distribution of the polarization of the medium at the instant of time t = 0. The random nature of the onset of CRS causes the energy of the Stokes pulses to fluctuate greatly. The statistical properties of these fluctuations are diligently investigated at present (see the bibliographies in Refs. 14 and 15). For strong fluctuations of the initial polarization $R_{\pm}(z,0)$ the profile of the Stokes pulse is generally speaking not defined. Only a statistical description is possible in this case. We estimate the role of the statistical and dynamic effects in the following manner. Let Γ be the rate of dephasing due to the stochastic processes, and η_u the nondimensional halfwidth of the Stokes pulse. The condition

$$\eta_u > \Gamma l,$$
 (1)

where l is the length of the sample, means then that the dynamic character of the Stokes pulse prevails over the stochastic one. This is in fact the case considered in the present paper, although some of the results that follow remain in force also in the statistical domain.

A number of experiments have revealed a "compression" of the statistical distribution of the energy of the Stokes pulse with increase of the energy conversion coefficient κ_{ϵ} (Ref. 15). Thus, when κ_{ϵ} changes from 1 to 40% the width of this distribution is decreased by a factor 5-8 (Ref. 15). An attempt was made¹⁶ to explain this effect theoretically, but the model used to describe the nonlinear stage had no connection with RS. A numerical description of the compression of the fluctuations was presented by Lewenstein.¹⁷ We found in the literature no complete and consistent description of the compression of the fluctuations during the nonlinear stage of RS. Some results of the present paper are devoted to a partial filling of this gap. Together with the fluctuations of the initial polarization, account is taken of the stochastic changes of the amplitude and phase of the pump field. It is shown that the joint action of the amplitudes of the polarization and amplitude of the pump field leads to singularities in the solution that describes the profiles of the pulses at the exit from the system. These singularities can be used, in particular, to explain the experiments of Druhl et al.18 and also of Ref. 19.

The formulation of the problem is described in the next section. In the second section is described the procedure of applying the ISPM to CRS and CRA. The modification of the scattering data to allow for the fluctuations is found in Sec. 3. The asymptotic quasi-self-similar solution used to explain the experimental data is obtained in the last section.

1. THE MAXWELL-BLOCH EQUATION AND FORMULATION OF THE PROBLEM

Two wave packets, propagating along the z axis:

$$\mathscr{E}(z,t) = \sum_{i=1,2} \left(\frac{\hbar \omega_i}{2c} \right)^{\frac{1}{2}} [E_i(z,t) \exp[i(k_i z - \omega_i t)] + \text{c.c.}],$$

interact with a two-level medium that has a transition frequency $\omega = \omega_2 - \omega_1$. The Maxwell-Bloch equation for the elements of the density matrix \hat{R} and for the slow envelopes $E_1(z,t)$ and $E_2(z,t)$ are of the form

$$\partial_{t}R_{+} = 2i(b_{1}|E_{1}|^{2} + b_{2}|E_{2}|^{2})R_{+} + i\varkappa E_{2}E_{1}^{*}R_{0}, \partial_{t}R_{0} = i\varkappa E_{1}E_{2}^{*}R_{+} + \kappa. c.,$$
(2)

$$(\partial_{z} + \partial_{t})E_{1} = -b_{1}(R_{0} - R_{0}^{(0)})E_{1} + i\varkappa R_{-}E_{2}, (\partial_{z} + \partial_{t})E_{2} = -b_{2}(R_{0} - R_{0}^{(0)})E_{2} + i\varkappa R_{+}E_{1}.$$
(3)

Here R_+ is the polarization and R_0 is the difference between the level populations, $R_- = R_+^*$, $R_0^{(0)} = R_0(z,0)$, and the coupling constants $b_{1,2}$ and \varkappa characterize the quadratic Stark effect (QSE) and the Raman interaction, respectively (for details see the review²⁰ and Ref. 10).

We change to a new notation⁹

$$g = (b_1 - b_2) \varkappa^{-1}, \quad x = \varkappa N_0 z, \quad N_0^2 = R_0^2 + R_+ R_-, \quad (4)$$

$$T = \varkappa \int_{-\infty}^{\tau} A(\tau') d\tau', \quad \tau = t - z c^{-1}, \quad A = |E_1|^2 + |E_2|^2, \quad S_+ = 2E_1 E_2^* \exp[i(b_1 + b_2) T \varkappa^{-1}], \quad S_- = S_+^*, \quad S = (|E_2|^2 - |E_1|^2) A^{-1}, \quad r_+ = R_+ \exp[i(b_1 + b_2) T \varkappa^{-1}], \quad r_- = r_+^*, \quad r = R_0 N_0^{-1}.$$

The following conditions correspond to the formulation of the Cauchy problem for the QCS: at the instant t = 0, with the medium polarization having a distribution $r_+(x,0)$ (along the medium, a high power pump-field pulse $E_2(0, T)$ is fed into the end face of the sample. The polarization fluctuations are assumed to be quite slow, since fast fluctuations can hinder the energy transfer to the Stokes field. We shall return to this question in Sec. 3.

The formulation of the Cauchy problem for the CRA is the following; a high-power pump-field pulse is applied to the end face of the sample simultaneously with a Stokes pulse $E_1(0,T)$ of small area, i.e., such that

$$Q = \left| \int_{-\infty}^{\infty} E_2(0,T) E_1(0,T) dT \right| \ll 1.$$

We assume, however, that the area under the pulse is large enough to be able to neglect quantum fluctuations. Note that the competition of the spontaneous RS primer can be decreased by varying the frequency range, since the cross section for spontaneous RS is proportional to ω_1^4 , whereas that of the spontaneous RS is proportional to ω_1 .

The descriptions of the evolution of the fields in CRS and CRA by the ISPM do not differ greatly. We shall therefore investigate hereafter mainly the CRS and indicate, where necessary, the differences that appear when CRA is studied.

2. GENERAL STRUCTURE OF THE SOLUTION BY THE ISPM

ISPM can be used because it is possible to represent the investigated system of equations (2)-(4) as a condition for compatability of two systems of linear equations:

$$\partial_x \varphi = i \left(\begin{array}{cc} -\lambda r & (f - i\lambda)r_+ \\ (f + i\lambda)r_- & \lambda r \end{array} \right) \varphi, \tag{5}$$

$$\partial_{\tau} \varphi = i (2\lambda + g)^{-1} \begin{pmatrix} \frac{1}{2} (2\lambda g + g^2 - 1)S & (f - i\lambda)S_+ \\ (f + i\lambda)S_- & \frac{1}{2} (1 - 2\lambda g - g^2)S \end{pmatrix} \varphi,$$
(6)

here $f^2 - \frac{1}{4}(1 - g^2)$.

The Lax representation (due to the formulation of the problem) is obtained for CRS from (5) to (6) after making the replacements $T \leftrightarrow x, r \leftrightarrow S, r_{\pm} \leftrightarrow S_{\pm}$.

We formulate for (5) the scattering problem, introducing sets of Jost functions, which are solutions φ^{\pm} of (5), defined by the asymptotic relations

$$\varphi^{\pm} \to \exp[-i\lambda\sigma_3 r(\pm\infty, 0)x], \quad x \to \pm\infty, \tag{7}$$

with Eqs. (5) considered in the class of coefficients that decrease rapidly with $x r_{\pm} \rightarrow 0$, $|x| \rightarrow \infty$ meaning that there is no polarization or a Stokes field at $\pm \infty$. We define the scattering matrix in the following manner:

$$\varphi^{-}=\varphi^{+}\hat{T}, \quad \det \hat{T}=1, \quad T_{11}=T_{22}=a, \quad T_{12}=-\overline{T}_{21}=b.$$
 (8)

The elements of the matrix \hat{T} and the set of constants corresponding to the discrete spectrum of the problem (5) constitute the scattering data. Analysis of (5) shows that under the condition $Q_0 = \int |r_{\pm}(x,0)| dx \ll 1$ the problem (5) has no discrete spectrum. This can be easily shown, in particular, at $g \neq 1$, for at small r_{\pm} the problem (5) is equivalent [accurate to $\sim O(|r_{\pm}|^2)$] to the Zakharov-Shabat spectral problem.⁸ In experiments on observation of superradiance (e.g., Refs. 4 and 21) and CRS (Refs. 2 and 3) the quantity Q_0 is always small ($\sim 10^{-8}$). Under these conditions the reflection coefficient $R = ba^{-1}$ is simply the Fourier transform of the polarization:

$$R(\lambda,0) = i(f+i\lambda) \int_{-\infty}^{\infty} r_{-}(x,0) \exp(2i\lambda x) dx.$$
(9)

The next step in the application of the ISPM is to solve singular integral equations, which can be done directly for the spectral problem (5). For comparison with the results of other theories,¹⁻³ however, it is more convenient, following Kaup,⁹ to transform to the Zakharov-Shabat problem. This is effected by changing to a new function $\psi = D^{-1}\varphi$,

$$D = [I \cos (\gamma/2) + i\sigma_{\mathfrak{s}} \sin (\gamma/2)] [I \cos(\beta/2) + i\sigma_{\mathfrak{s}} \sin(\beta/2)] [I \cos(\theta/2) + i\sigma_{\mathfrak{s}} \sin(\theta/2)], \quad (10)$$

where σ_i are Pauli matrices,

$$r_{\pm} = e^{\pm i\theta} \sin\beta, \quad r = \cos\beta, \quad \gamma = \int_{-\infty}^{\infty} \theta_x \cos\beta \, dx,$$
$$\theta_x = \partial_x \theta; \quad \gamma \to 0, \quad x \to -\infty; \quad \beta, \; \theta_x \to 0, \quad |x| \to \infty.$$

The function ψ satisfies the Zakharov-Shabat equation:

$$\partial_x \psi_1 + i\lambda \psi_1 = q_1 \psi_2, \quad \partial_x \psi_2 - i\lambda \psi_2 = q_2 \psi_1,$$
 (11)

where

$$q_{i} = [(if - i/_{2}\theta_{x})\sin\beta + i\beta_{x}/2]e^{i\gamma},$$

$$q_{2} = [(if + i/_{2}\theta_{x})\sin\beta + i\beta_{x}/2]e^{-i\gamma}.$$
(12)

The inverse-problem method for (11) has been well investigated.⁸ For the functions

$$\chi(\lambda, x, T) = (\chi_1, \chi_2)^T = \psi e^{-i\lambda x}, \quad \tilde{\chi}(\lambda) = (-\bar{\chi}_2, \bar{\chi}_1)^T$$

we have the singular integral equations:

$$\chi(\lambda) = \begin{pmatrix} 1 \\ 0 \end{pmatrix} - \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{R(\lambda', T)}{\lambda' - \lambda + i0} e^{-2i\lambda' x} \tilde{\chi}(\lambda') d\lambda', \quad (13)$$

$$\tilde{\chi}(\lambda) = \begin{pmatrix} 0 \\ 1 \end{pmatrix} - \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\overline{R}(\lambda', T)}{\lambda' - \lambda - i0} e^{2i\lambda' x}$$

$$\times \chi(\lambda') \begin{pmatrix} 1, & g \leq 1 \\ (\lambda' - |f|)/(\lambda' + |f|), & g > 1 \end{pmatrix} d\lambda'.$$
(14)

Equation (14) takes into account the singularities of the asymptotic forms of the problem (5) at g > 1 (for details see Studel's paper¹⁰). The evolution of R with T is determined from Eqs. (6) and (10). The potential q_2 is obtained from the equation

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$$q_2 = 2i\lambda\chi_2, \quad \lambda \to \infty. \tag{15}$$

3. ALLOWANCE FOR THE POLARIZATION FLUCTUATIONS AND FOR THE PUMP FIELD

The effect of polarization fluctuations on the RS process in its linear stage has been the subject of many investigations (see the bibliography in Ref. 14). Actually, as will be shown below, contributing to the evolution of the fields are only fluctuations localized in the vicinity of the origin, i.e., at the end face of the sample. We assume that the time of variation typical of polarization fluctuations (in a coordinate frame connected with the pump pulse) is large enough. This condition is necessary for the evolution of the RS. In fact, we shall show with a simple model as an example that fast fluctuation lead to vanishing of the average scattering coefficient, $\langle R_{av} \rangle = 0$. Let $r_{\pm} = |r_{\pm}| \cdot \exp(\pm i\varphi_0)$, where $|r_{\pm}|$ is a definite nonfluctuating amplitude, φ_0 a rapidly fluctuating one, and δ the correlated phase:

$$\partial_x \varphi_0 = F(x), \quad \langle F(x) \rangle = 0, \quad \langle F(x) F(x') \rangle = 2G\delta(x - x').$$
 (16)

Using Eqs. (5), we obtain by iteration the scattering data $(r(x,0) \equiv -1)$:

$$a(\lambda) \approx 1 - \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \exp[2i\lambda (x-x_1)] \\ \times |f+i\lambda|^2 r_+(x) r_-(x_1) dx dx_1 + \dots,$$

$$b(\lambda) \approx i \int_{-\infty} \exp(2i\lambda x) (f+i\lambda) r_{-}(x) dx + \dots$$

In the phase-diffusion model (see, e.g., Ref. 22), it can be easily shown by using the results of Ref. 14 that $\langle b(\lambda) \rangle = 0$ (the angle brackets denote here and elsewhere averaging over the fluctuations). A similar result was obtained earlier for an attenuator by Elgin and Kaup.²² In our case there is one more time scale—the characteristic time of instability evolution during the linear RS stage: $\tau_{in}^{-1} \sim \varkappa |E_2(0,0)|$. At $\tau_{in} \geq G^{-1}$ the perturbation has time to dissipate by diffusion, and no RS developes. At $\tau_{in} \sim G^{-1}$ the phase-diffusion model is not valid and $\langle b(\lambda) \rangle \neq 0$. The threshold value $E_2(0,0)$ can be roughly estimated from the condition $\tau_{in} \approx G^{-1}$.

Let us determine the time dependence of $R(\lambda, x, T)$. Under initial conditions $|r_{\pm}| \leq 1, r \approx -1$ and $E_1(0,T) = 0$ the matrix D in (10) can be set equal to unity accurate to $\sim O(|r_{\pm}|^2)$. We assume that the pump field is strong enough, in the case of CRS we get

 $R(\lambda, x, T)$

$$=R(\lambda,0)\exp\left[i\left(g-\frac{1}{2\lambda+g}\right)\int_{0}^{1}A^{-1}|E_{2}(0,T')|^{2}dT'\right].$$
(18)

It can be seen from (18) that the evolution of the fields in CRS is determined only by the pump-field amplitude. The situation is different in the case of CRA. The spectral problem should be chosen to be the system (6) and the spectral parameter must be suitably redistributed. For a very weak Stokes pulse and for $f \neq 0$ the problem (6) is equivalent to the Zakharov-Shabat problem. The arguments given above and in the present section can be repeated, but with the substitution $r_{\pm} \rightarrow E_1^*E_2$. Just as above, we are using the phase-diffusion model. Given the Stokes and pump field amplitudes, the fast fluctuations of the pump-field lead to vanishing of the coefficient $\langle b(\lambda) \rangle$.

The characteristics of the pump field, especially its fluctuations, are determined by the laser-radiation source.²³ The dynamics of the field near and above the lasing threshold is described by the van der Pol equations.²⁴ With increasing distance from the threshold, the amplitude of the intensity fluctuations becomes small compared with the mean value. Linearization of the van der Pol equations leads to a Brownian-motion model.^{25,26} The radiation transformed in RS is frequently used as a new radiation source, therefore the study of the influence of the pump fluctuations on the Stokes field is of particular interest. On the other hand, since the initial stage of the RS is characterized by development of instability, even relatively weak fluctuations of the pump field can influence substantially the form of the Stokes-field pulse. Indeed, such an effect was observed in experiments carried out by Safonov.¹⁹ The same experiments have revealed for the first time an abrupt decrease of the Stokespulse shape fluctuations due to fluctuation of the pump field during the strongly nonlinear stage of the RS. This effect can also be explained within the framework of our approach. Note that the Brownian-motion model, which will be used from now on, corresponds to the case of high-frequency fluctuations. This model was used to investigate the linear stage of the RS in Ref. 27. Since the phase of the pump field plays no role for CRS, the Brownian-motion model, which describes the statistical properties of the intensity $AI_p = |E_2(0,T)|^2$ of the pump field, takes the form

$$I_{I_{I}} = I_{0} + I', \quad \partial_{T}I'(T) = -\nu I'(T) + F_{I}(T),$$

$$\langle F_{I}(T_{1})F_{I}(T_{2}) \rangle = 2D\delta(T_{1} - T_{2}), \qquad (19)$$

where I_0 is the nonfluctuating part of the intensity, I' is a small fluctuating increment, ν^{-1} is the correlation time of fluctuations with mean value $\overline{I}' = D\nu^{-1}$. The fluctuating force is δ -correlated, i.e., it is assumed that the fluctuation time interval is smaller than the other characteristic times of the process. We average over the realizations of the random process in the following manner. We break up the time interval from 0 to T into M intervals, the mean value $\langle R \rangle$ can be represented as a path integral:

$$\langle R \rangle = R(\lambda, x, 0) \exp\left[i\xi \int_{0}^{1} I_{0}(T') dT'\right]C,$$

$$C = \lim_{M \to \infty} \int \dots \int dI_{0}' \dots dI_{M}' U(I_{0}') U(I_{0}', I_{1}') \dots U(I_{M-1}', I_{M}')$$

$$\times \prod_{k=0}^{M} \exp\left[i\frac{T}{M}\xi I_{k}'(T)\right],$$
(20)

Here $U(I_0')$ is the initial Gaussian distribution of the fluctuating intensity increment I', $\xi = g - 1/(2\lambda + g)$, $U(I'_k, I'_{k+1})$ is the probability of the $I'_k \rightarrow I'_{k+1}$ transition within a time interval T/M. In the Brownian-motion model

$$U(I_0') = \exp\left[-(I_0'/\bar{I}')^2\right],$$
 (21)

$$U(I_{k}'I_{k+1}') = \exp\left\{-\frac{[I_{k+1}'-I_{k}'\exp(-\nu T/M)]^{2}}{\bar{I}'^{2}[1-\exp(-2\nu T/M)]}\right\} \\ \times \left\{\pi\bar{I}'^{2}\left[1-\exp\left(-\frac{2\nu T}{M}\right)\right]^{\frac{1}{2}}\right\}.$$

The Gaussian integrals in (20) can be evaluated exactly (see Ref. 28):

$$C = \exp\{-(\xi^2 \bar{I}'^2 / \nu^2) [\nu T - 1 + \exp(-\nu T)]\}.$$
 (22)

Note that on going from the regime $\nu T \ge 1$ to $\nu T \le 1 < T \simeq \varkappa A \tau$, see Eq. (4), but it is known that $\tau \to 0$ with increase of time, see Ref. 7 for details, so that such a transition is possible), the fluctuation spectrum, i.e., the Fourier transform of (22), broadens greatly, roughly by a \overline{I}'^2/ν times. This effect is known in molecular optics as the Dicke effect.²⁹ A consequence of this effect is the compression of the temporal fluctuations. We confine ourselves hereafter to the limit $\nu T \ge 1$, i.e., to a short fluctuation-damping time; then Eq. (22) reduces to

$$C = \exp\left(-\xi^2 \bar{I}'^2 T/2\nu\right). \tag{23}$$

Note that within the framework of perturbation theory all the results reported in the next section can be generalized to the case of an arbitrary ratio $1/\nu T$, but to avoid unwieldy equations we confine ourselves to the limit $\nu T \ge 1$.

It remains for us to substitute the expression obtained for R in the right-hand sides of (13) and (14) and find the solution of this set of equations.

4. QUASI-SELF-SIMILAR SOLUTION

It is impossible to obtain an exact solution of the set of integral equations (13) and (14). We seek an approximate asymptotic solution. It is easy to show within the framework of the linear theory that for a very small initial polarization, such that $\ln Q_0^{-1} \ge 1$ (recall that $Q_0 \sim 10^{-8}$ in the experiments of Refs. 2, 3, and 21), the solution that describes the Stokes field is centered in the region of large values of the self-similar variable

$$\eta = 2(xW)^{\frac{1}{2}} \geqslant \eta_0 \gg 1, \quad W = \int_0^1 I_0(T') dT'.$$

This fact permits the use of the stationary phase method to calculate the integrals in the right-hand sides of (13) and (14). This approximation was used earlier by Manakov,⁷ who investigated a similar system of equations. Let us estimate the integral in the right-hand side of (13):

$$\Xi(\zeta, x, T) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\exp[-2i\lambda(x-y) + i\xi W - \sigma\xi^2 T]}{\zeta - \lambda + i0}$$
$$\times \tilde{\chi}(\lambda) \rho(y) \theta(x-y) dy d\lambda.$$
(24)

We have put here

 $\rho(y) = (f+i\lambda)r_{-}(y, 0), \quad \sigma = \overline{I}^{\prime 2}/2\nu.$

We change to a new integration variable $\Lambda = (2\lambda + g)[(x - y)/W]^{1/2}$:

$$\Xi(\zeta, x, T) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\exp\{-\frac{i}{2}i\eta'(\Lambda - \Lambda^{-i}) - \sigma\xi^2 T + ig[-W - y + x]\}}{\zeta - \lambda + i0}$$
$$\times \tilde{\chi}(\lambda)\rho(y)\theta(x - y)[W/4(x - y)]^{\frac{1}{2}}dy d\Lambda, \qquad (25)$$

where $\eta' = 2[W(x - y)]^{1/2}$.

The integrand in (25) has a saddle point λ_0^{\pm} :

$$(2\lambda_0^{\pm}+g)(x-y)^{1/2}=\pm iW^{1/2}$$

Integration with respect to Λ at $\eta_0 \ge 1$ reduces to an estimate of the integral in the vicinity of the points λ_0^+ , and expression (25) takes the form

$$\Xi(\zeta, x, T) = \left(\frac{W}{8\pi}\right)^{\frac{1}{2}} \times \int_{-\infty}^{\infty} \frac{\exp\{\eta' + ig(x-y-W) - \sigma T[g^2 + (y-x)/W + 2ig((x-y)W^{-4})^{\frac{1}{2}}]\}}{[\eta'(x-y)]^{\frac{1}{2}}(\zeta-\lambda_0^+)} \tilde{\chi}(\lambda_0^+)\rho(y)\theta(x-y)dy.$$
(26)

It is easily noted that owing to the exponential growth of the integrand the main contribution to the integral is made by the region of small y. Using the expansion

$$\eta' \approx \eta - y (W/x)^{\frac{1}{2}} + \ldots,$$

we represent (26) in the form

 $\Xi(\zeta, x, T)$

$$= \left(\frac{W}{8\pi\eta x}\right)^{\frac{1}{2}} \frac{\exp\left\{\eta + ig\left(x - W\right) - \sigma T\left[g^2 - x/W + 2ig\left(x/W\right)^{\frac{1}{2}}\right]\right\}}{\zeta - \lambda_0^+}$$

$$\times \bar{\chi}(\lambda_0^+) \Phi(\mu, x) (j + i\lambda_0^+), \qquad (27)$$

 $\Phi(\mu, x) = \int_{0}^{x} r_{-}(y, 0) e^{\mu y} dy, \quad \mu = -\frac{\sigma T}{W} - \left(\frac{W}{x}\right)^{\frac{1}{2}} - ig.$

We find similarly the integral in the right-hand side of (14):

$$\bar{\Xi}(\zeta, x, T) = \left(\frac{W}{8\pi\eta x}\right)^{\frac{1}{2}} \frac{\exp\{\eta - ig(x-W) + \sigma T[g^2 + x/W + 2ig(x/W)^{\frac{1}{2}}]\}}{\zeta - \lambda_0^{-1}} \times \chi(\lambda_0^{-1}) \Phi^{\bullet}(\mu, x) (f - i\lambda_0^{-1}) H,$$
(28)

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$$H=1, \quad g \leq 1;$$

$$H=[iW^{1/2}+(2f-g)x^{1/2}][iW^{1/2}-(2f-g)x^{1/2}]^{-1}, \quad g>1.$$

Using (13), (14), (27), and (28) we obtain a simple system of algebraic equations:

$$\chi(\zeta) = \begin{pmatrix} 1 \\ 0 \end{pmatrix} - \Xi(\zeta), \qquad (29)$$

$$\tilde{\chi}(\zeta) = \begin{pmatrix} 0\\1 \end{pmatrix} + \tilde{\Xi}(\zeta), \qquad (30)$$

Putting $\zeta = \lambda_0^-$ in (29) and $\zeta = \lambda_0^+$ in (30) we get, using (15),

$$q_{2} = \frac{2\eta}{x} \frac{HPe^{\eta}}{(2\pi\eta)^{\frac{1}{2}}} \left(1 + \frac{H|P|^{2}e^{2\eta}}{2\pi\eta}\right)^{-1}, \qquad (31)$$

where

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$$H = 1, \ g \leq 1;$$

$$H = [iW'^{h} + x'^{h}(2f + g)][iW'^{h} - x'^{h}(2f - g)]^{-1}, \ g > 1,$$

$$P = -\frac{1}{2}i\Phi^{*} \exp \left[-ig(W + x) - \sigma T(g + x/W)\right](f - i\lambda_{0}^{-}).$$

Equations (12) and (31) describe the form of the first peak of an infinite sequence of peaks whose amplitude decreases at infinity (as $\eta \to \infty$). At $\theta_x = \sigma = 0$, g = 0 and $r_{-}(y,0) = \delta(y)$ the solution is simplest. In this case q_2 reduces to a Painleve transcendental of type III. The same conclusion can be reached by comparing the solution of Eqs. (13) and (14) with a self-similar solution of the sine-Gordon equation.⁷ It is obviously impossible to obtain the function $E_{1,2}(x,T)$ in explicit form. Figure 1 shows the results of a numerical calculation. We change in (12) to the self-similar variable

$$i\partial_{\eta}\beta + \partial_{\eta}\theta\sin\beta + 2if\frac{x}{\eta}\sin\beta = 2q_{2}\frac{x}{\eta}e^{i\gamma}.$$
 (32)

It is seen from the figure and from an analysis of (32) that the pulses have different shapes at $\vartheta \ll 1$ and $\vartheta \approx 1$, where $\vartheta = 2f l/\eta_0$. In fact, if $\vartheta \ll 1$, $\theta_x = 0$ and g = 0, the solution reduces to that of the sine-Gordon equation; this is just the case considered in Refs. 1 and 2. Experimental observation of CRS in hydrogen^{2,3} yielded $\vartheta \sim 10^{-2}$. To reach the most nonlinear regime ($\vartheta \sim 1$) in a sample 10 cm long the required active-atom density is $10^{19}-10^{20}$ cm⁻³. The $\vartheta \sim 1$ regime is easier to reach for cesium atoms ($\varkappa \sim 10^{-14}$, Ref. 20); an atom density $\sim 10^{14}$ cm⁻³ suffices at the same atom density.

It was observed in experiment³ that the first peak of a Stokes field is not always the largest, whereas the theory based on the sine-Gordon equation leads to a Stokes-field pulse shape similar to that shown in Fig. 1 for $\vartheta \leq 1$. The cause of the discrepancy may be that the CRS theory used in Refs. 1-3 does not take the quadratic Stark effect into account. For two-photon interactions, neglect of this effect is generally speaking wrong, since the reduced coupling constant g that represents it can be of the order of unity (see, e.g., Ref. 10). For hydrogen, g = 0.18. A numerical analysis of Eq. (32) with a right-hand side a sequence of 2π pulses of alternating sign and decreasing amplitude, shows that the quadratic Stark effect gives rise to modulation of the spike amplitudes, with a periodic approximately five times the half-width of these peaks. This result agrees qualitatively with experiment.^{2,3}

The solution obtained can explain the fluctuation com-



FIG. 1. Dependence of $G = |E_2 E_1^*|^2$ on the self-similar variable η . Solid line— $\vartheta = 0.03$, dashed— $\vartheta = 1.2$.

pression observed in a number of experiments (see the Introduction). This is easiest to do in an approximation in which the levels have constant populations. The Maxwell-Bloch equations reduce then (g = 0) to the equations used to investigated a one-photon laser amplifier.⁷ The respective intensities, $I_p = |E_2|^2$, and $\tilde{I}_c = |E_1|^2$, of the pump and Stokes fields are of the form (it suffices to consider the form of the first spike)

$$\begin{split} & I_{\rho} \approx A \left[\operatorname{th} \left(\eta - \frac{1}{2} \ln \eta - \eta_{0} + \delta \right) \right]^{2}, \\ & I_{c} \approx A \operatorname{ch}^{-2} \left(\eta - \frac{1}{2} \ln \eta - \eta_{0} + \delta \right), \end{split} \tag{33}$$

where δ denotes the contribution of the fluctuations. The condition (1), with $\delta = \Gamma l$, permits the use of the expansion

ch
$$(\tilde{\eta}+\delta) \approx ch \, \tilde{\eta}+\delta sh \, \tilde{\eta}+\delta^2 ch \, \tilde{\eta}+\dots,$$
 (34)

where $\tilde{\eta} = \eta - \frac{1}{2} \ln \eta - \eta_0$. It is easily noted from (33) and (34) that the contribution of the fluctuations in the shape of the Stokes pulse is of the order of δ at $\varkappa_{\varepsilon} \ll 1$ and of the order of σ^2 at $\varkappa_{\varepsilon} \approx 1$. In the case of strong fluctuations one cannot speak of a definite shape of the Stokes pulse and a statistical description of the RS is needed. The Dicke effect described above, however, should take place even in this case, too.

The behavior of the solution (31) is determined by the form of the dependence of r_{-} on x at t = 0. In particular, a singularity occurs at the point $\mu = 0$ if g = 0 and $r_{-}(x,0) \sim x^2$. The Stokes field reverses sign on passing through this point. Druhl et al.¹⁸ observed in their experiments a sharp spike in shape of the transmitted pump field. To analyze this phenomenon, they introduced in the numerical calculation¹⁸ a jump π in the phase of the field at a specified instant of time. This spike was identified in Refs. 18 and 30 with a soliton (in the sense used in Ref. 8) of the Maxwell-Bloch equations, which are formally equivalent to the equations of the one-photon self-induced transparency. It is known, on the other hand, that at small initial polarization and at $E_1(0,\tau) \equiv 0$ (or $\int_{-\infty}^{\infty} E_2 E_1^*(0,\tau) d\tau \ll 1$ these equations have no soliton solution. Kaup obtained recently³⁰ an unexpected result; he has shown that a soliton solution (a pole in the upper half-plane, in the language of the spectral problem⁸) can appear as a result of dissipation. This result was obtained by a perturbation theory based on the ISPM equations for the linear stage of RS. It is important to note that in the experiments in which the "soliton" was observed the dissipation is not small. Thus, the relaxation time is $T_2 = 3$ ns whereas the length of the Stokes pulse is $\sim 10^2$ nm. It is known also that a soliton was observed also in Ref. 18 randomly, approximately once every 20 "shots" (Ref. 31). Yet according to Kaup's theory³⁰ it is natural to expect this soliton to appear practically in each shot, since the relaxation is always large. Note also that a soliton was always observed upon total depletion of the pump and on the trailing edge of the Stokes pulse, i.e., in the strongly nonlinear stage of the RS. The solution (31) of the present paper also contains a dip in the Stokes-pulse profile (or a spike in the shape of the passing pump field), for example at $r_{-}(x,0) \sim x^2$. The appearance of this dip (spike) is possible on the trailing edge of a Stokes pulse. Since the distribution $r_{-}(x,0)$ can vary randomly from shot to shot, one should appear a singularity to appear in (31) at some instant of time. The initial condition for the soliton is complete depletion of the pump, i.e., $E_2 \approx 0$. No explanation is given in Refs. 18 and 30 for the fact that the soliton covers a larger distance at a high initial field intensity. Moreover, the time of the spike is chosen in Ref. 18 phenomenologically. Using the solution obtained in the present paper, one can estimate the position of the spike. Assume for simplicity $\eta = -\sigma T/W + (W/x)^{1/2} - ig$ for the trailing edge of a Stokes pulse) that

$$g=0, \quad \theta_x=0, \quad W\approx AI_0\tau, \quad \bar{I}'^2=\alpha I_0^2.$$

The condition $\mu = 0$ yields

$$t_{sp} = cl(1 + A\alpha^2 \sigma^{-2} I_0)^{-1}.$$
(35)

Comparing with the experimental results,¹⁸ we find that Eq. (35) describes qualitatively correctly the location of the spike. It is also easy to show that the spike is narrower for larger I_0 . It appears that the second peak in the shape of the passing pump field, observed in experiments¹⁹ with paraand ortho-hydrogen, is of similar nature.

We note in conclusion that results similar to those given above can be obtained by studying other physical systems, e.g., when it comes to describing the characteristics of a twophoton laser amplifier or of two-photon superradiance (the sum of the carrier frequencies, which are not equal to each other, is equal to the frequency of the atomic transition). The formulation of the problem should include, as a required element specified at the point x = 0, a high-power pulse of one of the fields. The generation of the second pulse is initiated either by a priming pulse (amplifier) or by quantum fluctuations (superradiance).

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- ¹B. M. Chernoborod, Opt. Spectrosk. **49**, 692 (1980).
- ²V. S. Pivtsov, S. G. Rautian, V. P. Safonov, et al., Zh. Eksp. Teor Fiz. 81, 468 (1981) [Sov. Phys. JETP 54, 250 (1981)].
- ³A. A. Zabolotskiĭ, S. G. Rautian, V. P. Safonov, and B. M. Chernoborod, ibid. 86, 193 (1984) [59, 696 (1984)]
- ⁴A. V. Andreev, V. I. Emel'yanov, and Yu. A. Il'inskii, Usp. Fiz. Nauk 131, 653 (1980) [Sov. Phys. Usp. 23, 493 (1980)].
- ⁵R. H. Dicke, Phys. Rev. 93, 99 (1954).
- ⁶J. C. McGillivray and M. S. Feld, Phys. Rev. A 14, 1169 (1976).
- ⁷S. V. Manakov, Zh. Eksp. Teor. Fiz. 83, 68 (1982) [Sov. Phys. JETP 56, 37 (1982)].
- ⁸V. E. Zakharov, S. V. Manakov, S. P. Novikov, and L. P. Pitaevskii, Soliton Theory, Plenum, 1980.
- ⁹D. J. Kaup, Physica (Utrecht) D6, 143 (1983).
- ¹⁰H. Steudel, *ibid.* p. 155.
- ¹¹R. Meinel, Opt. Comm. 49, 224 (1984).
- ¹²I. R. Gabitov, V. E. Zakharov, and A. V. Mikhailov, Zh. Eksp. Teor. Fiz. 86, 1204 (1984) [Sov. Phys. JETP 59, 703 (1984)]
- ¹³F. Haaka, J. W. Haus, H. King, et al., Phys. Rev. A 23, 1322 (1981).
- ¹⁴J. A. Walmsley and M. G. Raymer, *ibid* A33, 382 (1986).
- ¹⁵A. S. Grabchikov, S. Ya. Kilin, V. P. Kozich, and N. M. Iodo, Pis'ma Zh. Eksp. Teor. Fiz. 43, 118 (1986) [JETP Lett. 43, 151 (1986)].
- ¹⁶M. Trippenbach and K. Rzazewski, Phys. Rev. A 31, 1932 (1985).
- ¹⁷M. Lewenstein, Z. Phys. B 56, 69 (1984).
- ¹⁸K. Druhl, R. G. Wenzel, and J. L. Carlsten, Phys. Rev. Lett. 51, 1171 (1983).
- ¹⁹S. G. Rautian, V. P. Safonov, and B. M. Chernobrod, Izv. AN SSSR, ser. fiz., 50, 1513 (1986).
- ²⁰A. Steudel, Ann. d. Phys. B 34, 188 (1977).
- ²¹N. Skribanowitz, I. P. Herman, J. C. McGilliway, and M. S. Feld, Phys. Rev. Let. 30, 309 (1973).
- ²²J. N. Elgin and D. J. Kaup, Opt. Comm. 43, 233 (1982).
- ²³S. Chandrasekhar, Rev. Mod. Phys. 15, 1 (1943).
- ²⁴H. Haken, in: Quantum Optics, S. M. Kay and A. Maitland, eds., Academic, 1970, p. 201. ²⁵P. Avan and C. Cohen-Tannoudji, J. Phys. B **10**, 155 (1977).
- ²⁶P. Zoller, *ibid.* **11**, 2825 (1978).
- ²⁷M. G. Raymer, J. Mostowski, and J. L. Carlstein, Phys. Rev. A1, 2304 (1979).
- ²⁸M. A. Abramowitz and I. A. Stegon, eds., Handbook of Math. Functions, Dover, 1964.
- ²⁹F. F. Baryshnikov and V. S. Lisitsa, Zh. Eksp. Teor. Fiz. 82, 1058 (1982) [Sov. Phys. JETP 55, 618 (1982)].
- ³⁰D. J. Kaup, Physica (Utrecht) D19, 125 (1986).
- ³¹J. R. Ackerhalt and P. W. Milonni, Phys. Rev. A 33, 3183 (1986).

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