Coherent transient effect in Raman pulse propagation

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In this paper we develop a theoretical analysis which describes a coherent transient effect of two entering pulses—one laser and one Stokes—in a Raman-active medium. The field (Maxwell) and atomic (Schrödinger, density-matrix representation) equations are coupled in a self-consistent manner in time and space by virtue of the resultant second-order nonlinear macroscopic polarizations at the two frequencies. This treatment is accomplished under the circumstances where atomic coherence plays a predominant role. The equations derived here may be referred to as "Raman Bloch-Maxwell equations." The resultant equations yield a new aspect of transient stimulated Raman scattering and thus coherent Raman propagation. The transient-pulse behavior may be understood as self-induced modulation of coherent amplification and absorption. The self-induced modulation for the step-function input pulse is described by a combination of Jacobian elliptic functions depending on the input intensity. Computer calculations of transient-pulse evolution reveal interesting pulse breakup phenomena with peak amplification, energy transmission, pulse advance or delay, and pulse-narrowing effects.

I. INTRODUCTION

The investigation of optical coherent interactions with matter has received considerable attention with the recent development of ultrashort-light-pulse techniques. Coherent propagation of optical pulses on the basis of quantum-mechanical coherent interaction in a time region within a homogeneous relaxation time is primarily revealed as the self-induced transparency effect.[1] This phenomenon has been studied by McCall and Hahn in connection with one-photon resonant interactions with a collection of independent two-level systems.

Particular interest of the problem is that optical waves with different frequencies are concerned with coherent multiphoton transition in a multilevel system. For local coherent effects such as photon echoes, some theoretical approaches of Raman echoes[2] and double resonance echoes[3-5] have been reported. While, for coherent propagation effects, a qualitative prediction of two-photon self-induced transparency, where twice the propagating frequency is resonant with the two-level system, has been discussed in previous works.[6,7] In our previous papers,[8-11] the first observation and theoretical study with respect to a new type of two-photon coherent propagation of two different-frequency optical short pulses in a gaseous three-level system were presented.

We shall now discuss the coherent Raman propagation effect.[12] Recent development of the theory of stimulated Raman scattering as well as experimental studies are gradually confirming transient behavior of the Raman process in the picosecond region.[13-17] In a time region shorter than longitudinal and transverse relaxation times, T_1 and T_2, coherent propagation effects may dominantly appear, yielding large signal amplification and modulation predicted by previous papers.[16-21] However, the predictive analysis did not take proper consideration of the pulse formation in the self-consistency requirement in the circumstances where atomic coherence plays a predominant role. A treatment using rate equations is not sufficient for coherent Stokes scattering for the ultrashort pulse propagation under these circumstances. Analyses of two-photon coherent radiation[22] in Raman transitions have been recently reported by Hopf[23] and Brewer,[24] who dealt with free-induction decay coherently excited by a step-function input pulse. These theoretical treatments will be modified if they involve the coherent propagation.

In the present paper, we shall especially investigate transient effects of the laser and the Stokes pulses undergoing coherent propagation in a Raman-active medium of which spectrum broadening is restricted to a "sharp line" case. Here we take into account the Raman-active response coherently excited by a resonant beating of two externally generated coherent fields. It is emphasized that this process should be analyzed under the following conditions: coherent interaction predominantly acting on the system, and the two propagating fields determined in a self-consistent manner. It has been already clarified by the authors that such coherent propagation of different-frequency optical short pulses interacting with a three-level system results in two-photon self-induced transparency.[10,11] We can use a similar procedure for the present case. First, we can obtain the system of coherent propagation equations from the combined Maxwell and Schrödinger (density-matrix representation) equations in self-consistent form for the Stokes and laser fields. The equations of motion for the medium may be referred to as "Raman Bloch equations," analogous to "two-photon Bloch equations" in Ref. 10. Second, an analytical description is given in connection with a set of Jacobian elliptic functions, which shows a periodic, amplified self-induced modulation for the Stokes wave. Third, a transient pulse behavior simulated by computer calculation reveals anomalous pulse evolutions, pulse-energy transmissions, and pulse velocities which are inherent to a coherent stimulated Raman transition accompanying population inversion. In the present paper we shall deal with only the envelope formation of the laser and the Stokes pulses. The other complementary aspects of the problem, namely the equation of the phase evolution and the frequency chirping of the pulse at different pulse velocities of the Stokes and laser waves, will be considered in a forthcoming, more extended paper.

II. FORMULATION

The basic equations which describe the coherent propagation of both the Stokes and the laser fields $\mathcal{E}_S(z,t)$ and $\mathcal{E}_L(z,t)$ in a three-level system including a representative virtual state are derived by combining Maxwell's equations and the equations of motion by means of a density matrix. We can develop the present formulation in a way similar to that of Ref. 10 by simply altering the level composition. Here, we take into consideration only the first-order Stokes generation, although the higher-order Stokes or anti-Stokes emission may appear strongly in such coherent process.

We assume that two monochromatic plane-wave classical fields are given as

$$E_i(z,t)=\mathcal{E}_i(z,t)\cos[\omega_i t-k_i z+\phi_i(z,t)] \quad (i=L,S)\,, \tag{1}$$

where the electric fields E_L and E_S induce the transitions between the energy levels 1-3 and 2-3 in the three-level system consisting the ground state 1, a phonon or electronic excited state 2, and the far-off-resonance virtual state 3. The frequencies ω_L and ω_S are assumed to be far-off-resonance from the eigenfrequencies Ω_L and Ω_S for each transition.

The interacting Hamiltonian may be written

$$\mathcal{H}'=\begin{bmatrix}0 & 0 & V_L\\ 0 & 0 & V_S\\ V_L^* & V_S^* & 0\end{bmatrix}, \tag{2}$$

where $V_i=-\mu_i\mathcal{E}_i\cos\Psi_i$, μ_i is the matrix element of the electric dipole moment in the relevant transition, and $\Psi_i=\omega_i t-k_i z+\phi_i$. The density matrix obeys the equation of motion

$$\frac{\partial\rho}{\partial t}=-\frac{i}{\hbar}[\mathcal{H},\rho]\,, \tag{3}$$

where $\mathcal{H}=\mathcal{H}^0+\mathcal{H}'$; $\mathcal{H}^0$ is the unperturbed Hamiltonian. Here, the rotating-wave approximation is assumed. It is also postulated that the slowly-varying amplitude and phase approximation is valid for ρ_{mn}, $\mathcal{E}_i$, and ϕ_i in the usual sense and also for a variation of off-resonance frequency $\Delta\omega_0=|\Delta\omega_L|=|\Delta\omega_S|$ with respect to the virtual state 3, where $\Delta\omega_i=\omega_i-\Omega_i$. The elements of ρ_{13} and ρ_{23} are implicitly transfered to ρ_{12}, yielding an induced two-photon polarization at the phonon frequency by means of the integral of the components of $\dot\rho_{13}$ and $\dot\rho_{23}$ in Eq. (3) under the above assumptions. This integral leads to a resulting explicit form for the two-photon Raman coupling.

Thus, one can obtain the working equations for the medium as follows:

$$\frac{d}{dt}(\rho_{22}-\rho_{11})=\frac{\mu_S\mu_L}{2\hbar^2\Delta\omega_0}\mathcal{E}_S\mathcal{E}_L[i(\sigma_{12}-\sigma_{12}^*)\cos\overline{\Psi}_\lambda-(\sigma_{12}+\sigma_{12}^*)\sin\overline{\Psi}_\lambda]\,, \tag{4a}$$

$$\frac{d}{dt}(\sigma_{12}+\sigma_{12}^*)=-\frac{\mu_S\mu_L}{2\hbar^2\Delta\omega_0}(\rho_{22}-\rho_{11})\mathcal{E}_S\mathcal{E}_L\sin\overline{\Psi}_\lambda-\frac{i}{4\hbar^2\Delta\omega_0}(\mu_L^2\mathcal{E}_L^2-\mu_S^2\mathcal{E}_S^2)(\sigma_{12}-\sigma_{12}^*)\,, \tag{4b}$$

$$\frac{d}{dt}(\sigma_{12}-\sigma_{12}^*)=\frac{i\mu_S\mu_L}{2\hbar^2\Delta\omega_0}(\rho_{22}-\rho_{11})\mathcal{E}_S\mathcal{E}_L\cos\overline{\Psi}_\lambda+\frac{i}{4\hbar^2\Delta\omega_0}(\mu_L^2\mathcal{E}_L^2-\mu_S\mathcal{E}_S^2)(\sigma_{12}+\sigma_{12}^*)\,, \tag{4c}$$

$$\sigma_{13}+\sigma_{13}^*=\frac{\mu_L}{\hbar\Delta\omega_0}(\rho_{33}-\rho_{11})\mathcal{E}_L\cos\overline{\Psi}_L+\frac{\mu_S}{2\hbar\Delta\omega_0}\mathcal{E}_S[(\sigma_{12}+\sigma_{12}^*)\cos\overline{\Psi}_S+i(\sigma_{12}-\sigma_{12}^*)\sin\overline{\Psi}_S]\,, \tag{4d}$$

$$\sigma_{23}+\sigma_{23}^*=-\frac{\mu_S}{\hbar\Delta\omega_0}(\rho_{33}-\rho_{22})\mathcal{E}_S\cos\overline{\Psi}_S-\frac{\mu_L}{2\hbar\Delta\omega_0}\mathcal{E}_L[(\sigma_{12}+\sigma_{12}^*)\cos\overline{\Psi}_L+i(\sigma_{12}-\sigma_{12}^*)\sin\overline{\Psi}_L]\,, \tag{4e}$$

where

$$\sigma_{mn} = \rho_{mn} e^{i\Omega_j} (j = L, S, \lambda; \Omega_\lambda = \Omega_L - \Omega_S),$$

$$\overline{\Psi}_i = \Delta\omega_0 t - k_i z + \phi_i,$$

$$\overline{\Psi}_\lambda = \Delta\omega t - (k_L - k_S)z + (\phi_L - \phi_S),$$

and

$$\Delta\omega = \Delta\omega_L - \Delta\omega_S.$$

Some variables are defined like the Bloch vector as follows:

$$\rho_L = \rho_{33} - \rho_{11}, \quad \rho_S = \rho_{33} - \rho_{22}, \quad \rho_\lambda = \rho_{22} - \rho_{11}, \tag{5}$$

$$(\sigma_{12} + \sigma_{12}^*) = u_\lambda \cos\Psi_\lambda + v_\lambda \sin\Psi_\lambda, \tag{6a}$$

$$(\sigma_{23} + \sigma_{23}^*) = u_S \cos\Psi_S + v_S \sin\Psi_S, \tag{6b}$$

$$(\sigma_{13} + \sigma_{13}^*) = u_L \cos\Psi_L + v_L \sin\Psi_L, \tag{6c}$$

where $\Psi_\lambda = (\omega_L - \omega_S)t - (k_L - k_S)z + (\phi_L - \phi_S)$. In Eqs. (6), u_n and v_n are related to the respective in-phase and out-of-phase components of the induced polarizations. Substituting Eqs. (5) and (6) into Eqs. (4a)–(4c) and eliminating u_L, u_S, v_L, and v_S, the response of the atomic system to the propagating electric fields is described by

$$\dot{\rho}_\lambda = \kappa \mathcal{E}_L \mathcal{E}_S v_\lambda - (1/T_1)\rho_\lambda, \tag{7a}$$

$$\dot{u}_\lambda = \left(\Delta\omega + \dot{\phi}_\lambda + \frac{\mu_S^2 \mathcal{E}_S^2 - \mu_L^2 \mathcal{E}_L^2}{4\hbar^2 \Delta\omega_0}\right) v_\lambda - \frac{1}{T_2'} u_\lambda, \tag{7b}$$

$$\dot{v}_\lambda = -\left(\Delta\omega + \dot{\phi}_\lambda + \frac{\mu_S^2 \mathcal{E}_S^2 - \mu_L^2 \mathcal{E}_L^2}{4\hbar^2 \Delta\omega_0}\right) u_\lambda - \kappa \mathcal{E}_L \mathcal{E}_S \rho_\lambda - \frac{1}{T_2'} v_\lambda, \tag{7c}$$

where $\kappa = \mu_L \mu_S / 2\hbar^2 \Delta\omega_0$, $\phi_\lambda = \phi_L - \phi_S$.

Damping effects have been introduced phenomenologically by means of a longitudinal relaxation time T_1 of the Raman excited state 2, and by means of a homogeneous, transverse relaxation time T_2'. The behavior of the atomic system represented by Eqs. (7) is found to be analogous to that for a one-photon transition in a two-level system except for the fields involving Raman (two-photon) coupling and for the frequency shift $(\mu_S^2 \mathcal{E}_S^2 - \mu_L^2 \mathcal{E}_L^2)/4\hbar^2 \Delta\omega_0$. Therefore, the above equations may be refered to as "Raman Bloch equations" analogous to "Two-photon Bloch equations" in Refs. 10 and 11.

The induced macroscopic polarizations at the input frequencies ω_L and ω_S may be described as

$$P_L = N_0 \mu_L \langle u_L \cos\Psi_L + v_L \sin\Psi_L \rangle, \tag{8a}$$

$$P_S = N_0 \mu_S \langle u_S \cos\Psi_S + v_S \sin\Psi_S \rangle, \tag{8b}$$

where N_0 is the particle density, and $\langle \cdots \rangle$ stands for the averaging over the inhomogeneous broadening $g(\Delta\omega)$. The expressions of u_i and v_i are derived by substituting Eqs. (5) and (6) into Eqs. (4d) and (4e). The above macroscopic polarizations act as sources in the self-consistent form for the electric field in accordance with Maxwell's equations. In general, one could represent the evolution of the plane-wave light pulses of Eq. (1) by the second-order wave equations

$$\frac{\partial^2 E_i}{\partial z^2} - \frac{\eta_i}{c^2} \frac{\partial^2 E_i}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 P_i}{\partial t^2}, \tag{9}$$

where η_i is the host refractive index at ω_i and c is the velocity of light in vacuum. Under the slowly varying amplitude and phase approximation as mentioned above, one can replace Eq. (9) with the reduced wave equations after substituting Eqs. (6b), (6c), (4d), and (4e) into (8), and Eqs. (1) and (8) into (9), and equating the coefficients of $\cos\Psi_i$ and $\sin\Psi_i$. Thus, the reduced wave equations are

$$\frac{\partial \mathcal{E}_L}{\partial z} + \frac{\eta_L}{c} \frac{\partial \mathcal{E}_L}{\partial t} = -\tfrac{1}{2}\beta_L \langle \mathcal{E}_S v_\lambda \rangle, \tag{10a}$$

$$\frac{\partial \mathcal{E}_S}{\partial z} + \frac{\eta_S}{c} \frac{\partial \mathcal{E}_S}{\partial t} = \tfrac{1}{2}\beta_S \langle \mathcal{E}_L v_\lambda \rangle, \tag{10b}$$

$$\left(\frac{\partial \phi_L}{\partial z} + \frac{\eta_L}{c} \frac{\partial \phi_L}{\partial z}\right) \mu_S \mathcal{E}_L = -\tfrac{1}{2}\beta_L \langle 2\mu_L \mathcal{E}_L \rho_\lambda + \mu_S \mathcal{E}_S u_\lambda \rangle, \tag{11a}$$

$$\left(\frac{\partial \phi_S}{\partial z} + \frac{\eta_S}{c} \frac{\partial \phi_S}{\partial z}\right) \mu_L \mathcal{E}_S = \tfrac{1}{2}\beta_S \langle 2\mu_S \mathcal{E}_S \rho_\lambda + \mu_L \mathcal{E}_L u_\lambda \rangle, \tag{11b}$$

where

$$\beta_i = 2\pi N_0 \Omega_i \mu_S \mu_L / c\hbar\eta_i \Delta\omega_0.$$

Equations (7), (10), and (11) characterize the dynamics of the Raman process in the fully self-consistent form without approximation in which a variation of the field is treated to be constant with respect to space and time "in the interaction with matter." Although the atomic system seems to behave in appearance in the same way as for a two-level system represented by the Bloch-Maxwell equations, the polarization at each frequency appearing on the right-hand side of Eqs. (10) results from the product of another field and the term v_λ arising from the second-order induced polarization of the effective Raman coupling due to the off-diagonal elements. This feature is a novel effect belonging to Raman transitions in the dynamic self-consistent form: We proceed with a discussion of the coherent propagation of two photons by analyzing only the dynamics of this nonlinear process. Therefore, for the present analysis the "zero-phase" solution ($\phi_i = 0$) is assumed in a self-consistent fashion, neglecting the frequency shift due to the fields, which was shown to be satisfactory for an amplifier when $g(\Delta\omega)$ is a symmetrical function.[25] For the Raman transition, let $\Delta\omega = 0$ in the usual sense and $T_1 = T_2' = \infty$ in Eqs. (7), and let $g(\Delta\omega) \sim \delta(\Delta\omega)$ as a sharp line for the averaging over $g(\Delta\omega)$ in Eqs. (10). This restriction of $g(\Delta\omega) \simeq \delta(\Delta\omega)$ does not change the essential response of the inhomogeneously broadened medium under the atomic coherence condition.[1] Under the above assumption, the solutions for the medium are

$$v_\lambda(z,t) = -\sin\varphi(z,t)\,, \tag{12}$$

$$\rho_\lambda(z,t) = -\cos\varphi(z,t)\,. \tag{13}$$

The wave equations are then

$$\frac{\partial \mathcal{E}_L(z,t)}{\partial z} + \frac{\eta_L}{c}\frac{\partial \mathcal{E}_L(z,t)}{\partial t} = \tfrac{1}{2}\beta_L \mathcal{E}_S \sin\varphi(z,t) - \gamma_L \mathcal{E}_L\,, \tag{14a}$$

$$\frac{\partial \mathcal{E}_S(z,t)}{\partial z} + \frac{\eta_S}{c}\frac{\partial \mathcal{E}_S(z,t)}{\partial t} = -\tfrac{1}{2}\beta_S \mathcal{E}_L \sin\varphi(z,t) - \gamma_S \mathcal{E}_S\,, \tag{14b}$$

where

$$\varphi(z,t) = -\kappa\int_{-\infty}^{t} \mathcal{E}_L(z,t')\mathcal{E}_S(z,t')\,dt'\,, \tag{15}$$

γ_i is the loss factor due to impurities and scattering. The minus sign in Eq. (15) implies an inverse phase relation between $\mathcal{E}_L$ and $\mathcal{E}_S$. Equations (14) easily lead to $\mathcal{E}_L^2/\beta_L + \mathcal{E}_S^2/\beta_S = \text{const}$, namely the Manley-Rowe relation if we ignore the loss terms. It is worthwhile to note that simple equations describing the usual stimulated Raman scattering is easily derived from Eqs. (14) for a small area $\sin\varphi \simeq \varphi$ after carrying out an integral involving the two-photon coupling.

Equations (14) are the basic equations necessary in order to discuss some properties of coherent Raman propagation or coherent stimulated Raman scattering. If we define coherent gain factors for the two-photon Raman coupling, they are written as follows:

$$g_{S,L} = \frac{2\pi N_0 \Omega_{S,L}\mu_S\mu_L}{c\hbar\eta_{S,L}\Delta\omega_0}\sin\kappa\int \mathcal{E}_L\mathcal{E}_S\,dt\,. \tag{16}$$

It may be useful to define the out-of-phase components of the induced second-order macroscopic polarization P_{ip} contributing to the respective fields at ω_L and ω_S:

$$P_{Lp} = -\frac{N_0\mu_L\mu_S}{2\hbar\Delta\omega_0}\mathcal{E}_S \sin\left(-\kappa\int \mathcal{E}_L\mathcal{E}_S\,dt\right), \tag{17a}$$

$$P_{Sp} = +\frac{N_0\mu_L\mu_S}{2\hbar\Delta\omega_0}\mathcal{E}_L \sin\left(-\kappa\int \mathcal{E}_L\mathcal{E}_S\,dt\right). \tag{17b}$$

Of particular interest is that for Gaussian input pulses yielding a large value of $\varphi(0,t)$, the above induced polarizations cause a repetition of two modes of absorption and emission with respect to time like a variation of a wave packet in accordance with a repetition of the sign of the sinusoidal function.

Here we study a coherent amplification of the Raman pulse energy along the propagating distance for the small pulse area. Let $\mathcal{E}_i(z,t)$ be a rectangular pulse with pulse width τ' along the time axis. Equations (14), after integrating out the time dependence, lead to

$$\frac{d}{dz}\mathcal{E}_L^2 = -\tfrac{1}{2}\kappa\beta_L\tau'\mathcal{E}_S^2\mathcal{E}_L^2\,, \tag{18a}$$

$$\frac{d}{dz}\mathcal{E}_S^2 = \tfrac{1}{2}\kappa\beta_S\tau'\mathcal{E}_S^2\mathcal{E}_L^2\,, \tag{18b}$$

neglecting the additional loss terms. These equations differ from the ordinary steady-state rate equation because of no influence of T_1 and T_2, and the present equations involve only the pulse width τ', while the rate equation is valid for the pulse envelope over the relaxation time. Accordingly, the coherent amplification of the Raman pulse could be solved in consequence. The solutions for the energy transmissions $T_L(z)$ and $T_S(z)$ are given as follows:

$$T_L(z) = \frac{\mathcal{E}_L^2(z)\tau'}{\mathcal{E}_L^2(0)\tau'} = \frac{1+\delta}{\delta + \exp(\frac{1}{2}\kappa\tau'\beta_0 I_0 z)}\,, \tag{19}$$

$$T_S(z) = \frac{\mathcal{E}_S^2(z)\tau'}{\mathcal{E}_L^2(z)\tau'} = \frac{1+\delta}{1+\delta\exp(-\frac{1}{2}\kappa\tau'\beta_0 I_0 z)}\,, \tag{20}$$

where

$$\delta = \beta_S\mathcal{E}_L^2(0)/\beta_L\mathcal{E}_S^2(0)\,,$$

$$\beta_0 I_0 = \beta_S\mathcal{E}_L^2(0) + \beta_L\mathcal{E}_S^2(0) = \beta_S\mathcal{E}_L^2(z) + \beta_L\mathcal{E}_S^2(z)\,.$$

The parameter δ is the input intensity ratio. The above expression for I_0 shows the sum of the powers of the respective fields. The pulse energy transmissions $T_i(z)$ are plotted against distance in Fig. 1.

For a given input ratio δ the Stokes pulse energy is amplified along the distance, while the laser pulse is attenuated. For an input laser pulse large compared with the input Stokes pulse, i.e., a large value of δ, the laser pulse decays remarkably, yielding the complementary amplification for the Stokes pulse congruent with the Manley-Rowe relation. Equations (19) and (20) indicate the fact that initially the transmitting pulse energy for the laser decays exponentially with distance and the pulse energy for the Stokes is amplified in the associated way. However, finally the energy of the laser pulse goes to zero and the energy of the Stokes pulse to the constant value of $\beta_0 I_0$. Therefore, all of a given input energy of the laser is ultimately transfered to the Stokes pulse at an infinitely large distance. This coherent amplification and attenuation in the small area is significantly analogous to a result using the rate equation.[16]

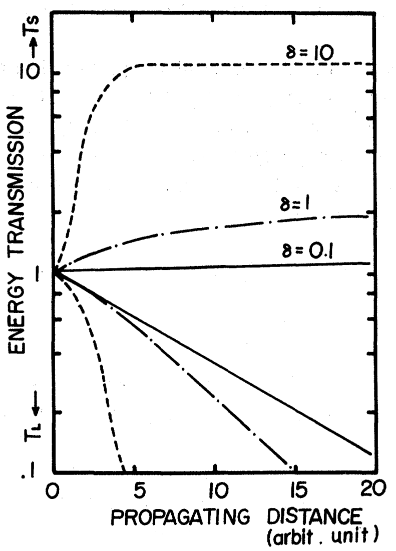

FIG. 1. Amplification rate T_S for the Stokes pulse and absorption rate T_L for the laser for a small input area, calculated from Eqs. (19) and (20), plotted against the propagating distance keeping the input intensity ratio δ constant.

III. COHERENT PROPAGATION IN A STEADY STATE

Stimulated Raman scattering in a stationary state has been studied by many researchers. In this section, we wish to investigate the property of steady-state propagation in the coherent Raman process. Analytical solutions of Eqs. (14) in the steady state predict the evolution of specially shaped pulses with pulse breakup even for step-function input pulses.

In the moving frame $\zeta = t - z/V$, where the pulse velocity V is conveniently assumed to be equal for both the Stokes and the laser pulses in the steady state, Eqs. (14) lead to

$$\frac{\partial \mathcal{E}_L}{\partial \zeta} = -\tfrac{1}{2}\bar{\beta}_L \mathcal{E}_S \sin\left(-\kappa \int_{-\infty}^{\zeta} \mathcal{E}_S \mathcal{E}_L \, d\zeta\right), \tag{21a}$$

$$\frac{\partial \mathcal{E}_S}{\partial \zeta} = \tfrac{1}{2}\bar{\beta}_S \mathcal{E}_L \sin\left(-\kappa \int_{-\infty}^{\zeta} \mathcal{E}_S \mathcal{E}_L \, d\zeta\right), \tag{21b}$$

where $\bar{\beta}_i = cV\beta_i/(c - \tau V)$, and $\eta \equiv \eta_i$ was assumed. We take into account simultaneous coherent propagation of both of the externally generated coherent fields. To describe the evolution of the two fields in the steady state, the respective amplitudes of the initial electric fields would be set equal to constants $\mathcal{E}_{S0}$, $\mathcal{E}_{L0}$ for $\zeta = -\infty$. Thus, the first integrals of Eqs. (21) are calculated from the initial condition

$$\mathcal{E}_L^2(\zeta) = \mathcal{E}_{L0}^2 - (\bar{\beta}_L/\kappa)[1 - \cos\varphi(\zeta)], \tag{22a}$$

$$\mathcal{E}_S^2(\zeta) = \mathcal{E}_{S0}^2 - (\bar{\beta}_S/\kappa)[1 - \cos\varphi(\zeta)]. \tag{22b}$$

Carrying out integrals after coupling $\varphi(\zeta)$ of Eqs. (15) and (22), the $\mathcal{E}_L^2(\zeta)$ and $\mathcal{E}_S^2(\zeta)$ are given as

$$\mathcal{E}_L^2(\zeta) = \frac{2\bar{\beta}_0 I_0 + \mathcal{E}_{S0}^2(\kappa\mathcal{E}_{L0}^2 - 2\bar{\beta}_L)\{1 + \mathrm{tn}^2[\tau^{-1}(\zeta - \zeta_0); m]\}}{\kappa\mathcal{E}_{S0}^2\{1 + \mathrm{tn}^2[\tau^{-1}(\zeta - \zeta_0); m]\} + 2\bar{\beta}_S}, \tag{23a}$$

$$\mathcal{E}_S^2(\zeta) = \frac{\mathcal{E}_{S0}^2(\kappa\mathcal{E}_{S0}^2 + 2\bar{\beta}_S)\{1 + \mathrm{tn}^2[\tau^{-1}(\zeta - \zeta_0); m]\}}{\kappa\mathcal{E}_{S0}^2\{1 + \mathrm{tn}^2[\tau^{-1}(\zeta - \zeta_0); m]\} + 2\bar{\beta}_S}, \tag{23b}$$

and the population difference ρ_λ is obtained from Eq. (13) under the initial condition of $\rho_{11} = 1$ in the ground state,

$$\rho_\lambda(\zeta) = -\frac{\kappa\mathcal{E}_{S0}^2\{1 - \mathrm{tn}^2[\tau^{-1}(\zeta - \zeta_0); m]\} + 2\bar{\beta}_S}{\kappa\mathcal{E}_{S0}^2\{1 + \mathrm{tn}^2[\tau^{-1}(\zeta - \zeta_0); m]\} + 2\bar{\beta}_S}, \tag{24}$$

where tn is the Jacobian elliptic function: m is the modulus

$$m = \frac{(2\bar{\beta}_S\bar{\beta}_L)^{1/2} I_0}{\mathcal{E}_{L0}(\kappa\mathcal{E}_{S0}^2 + 2\bar{\beta}_S)^{1/2}}; \tag{25}$$

and the definition

$$1/\tau = (\tfrac{1}{2}\kappa)^{1/2}\mathcal{E}_{L0}(\kappa\mathcal{E}_{S0}^2 + 2\bar{\beta}_S)^{1/2} \tag{26}$$

has been made. Physically, the above solutions correspond to Stokes and laser fields whose amplitudes are modulated periodically with a period $T = 2\tau K(m)$, where $K(m)$ is the complete elliptic integral of the first kind. It becomes evident from Eqs. (23) and Fig. 2 that the modulation is caused by an alternation of the amplification for the Stokes wave and the absorption for the laser on the basis

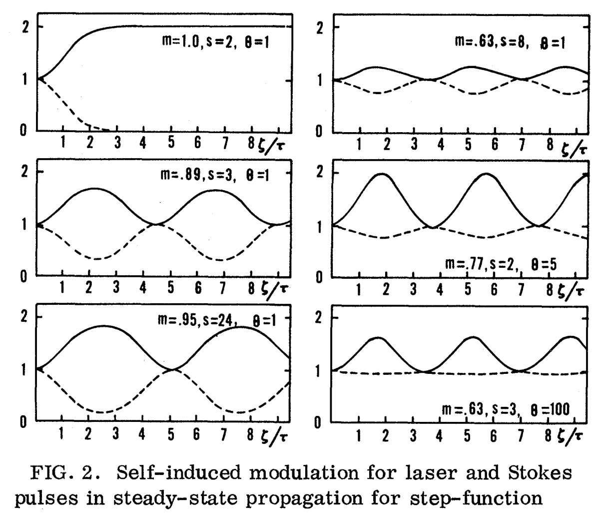

FIG. 2. Self-induced modulation for laser and Stokes pulses in steady-state propagation for step-function input pulses, estimated from Eqs. (23). Solid lines, periodic coherent amplification for the Stokes input pulse. Dashed line, complementary absorption for the laser. Both ordinates are normalized by the respective input intensity. The parameter m is the modulus of the Jacobian elliptic function tn and s is the normalized input intensity of the Stokes pulse $\kappa\mathcal{E}_{S0}^2/\bar{\beta}_S$; θ indicates the input-intensity ratio $\bar{\beta}_S\mathcal{E}_{L0}^2/\bar{\beta}_L\mathcal{E}_{S0}^2$.

of the periodic population inversion as apparent in Eq. (24). In this alternation, the energy conservation law for the incident pulses is found to be satisfied. The amplitude modulation for the both fields is of interest when the modulus m approaches unity. The period of both propagating pulses then approaches infinity and the elliptic solutions are given approximately by the well-known hyperbolic-secant solutions,[12] as illustrated in Fig. 2. From the figures and Eq. (26) one notices that the pulse narrowing and the high repetition of the pulse train appear with increasing the incident input-intensity ratio θ, where $\theta = \overline{\beta}_S \mathcal{E}_{L0}^2 / \overline{\beta}_L \mathcal{E}_{S0}^2$. Therefore, such an intense Raman field coherently interacts with a collection of atoms or molecules, resulting in the self-induced periodic modulation whose period depends upon the incident input powers.

IV. COHERENT EFFECT ON TRANSIENT PULSE BEHAVIOR

An analysis of the pulse evolution exhibiting coherent transient effects is performed by numerically integrating Eqs. (14). The numerical calculations were achieved by an application of a finite-difference method to the basic equations. As a result, computer-generated output pulses of $\mathcal{E}_L(z,t)$ and $\mathcal{E}_S(z,t)$ for the coherent Raman propagation are illustrated in Fig. 3. The initial pulse shapes, which have a given input pulse area $A_0 \equiv \varphi(0,\infty)$ estimated from Eq. (15), were taken as Gaussian functions with the full pulse width of 1.2 τ_0, where τ_0 is the time unit, taken as 10 psec. Equal peak intensities for the simultaneous incident beams at the Stokes and laser frequencies $[\xi = \mathcal{E}_L(0,t)/\mathcal{E}_S(0,t) = 1.0]$ were assumed. The intensities of $\mathcal{E}_i^2(z,t)$ are normalized to the height of the input pulse. The time is measured in units of τ_0 and the propagating distance z in β^{-1} $[\beta = 10^{-2} \times (\beta_S \beta_L)^{1/2}]$ for convenience. The parameters used here were conveniently estimated from a Raman transition in CS_2 at an incident wavelength 6943 Å characterized by the typical values of $\mu_{S,L} \simeq 1.6 \times 10^{-18}$ cm esu, $N_0 = 10^{22}/\text{cm}^3$, and $\Delta\omega_0 = 1.6 \times 10^4$ cm^{-1}. Thus, the evaluation of these values yields an order of 10^4 cm^{-1} for $\beta_{S,L}$ so that we can expect a very large amount of energy exchange between the Stokes and the laser fields, which causes a deep modulation within a very short propagation distance.

Figures 3(a)–3(c) exhibit a monotonic coherent amplification for the Stokes wave and accompanying attenuation for the laser wave in any input area less than $A_0 = \pi$. The amplification apparently increases with respect to the propagating distance, in contrast to the attenuation. The pulse energy of the Stokes wave at infinite distance increases with higher input area until the area π is reached (see Fig. 5). This process naturally implies that the pulse energy of the laser wave is ultimately changed into the Stokes pulse as long as the input pulse area is less than π. This property agrees with the result for the energy transmission for a small pulse area consisting of the step-function input pulses discussed in Sec. III. In this sense,

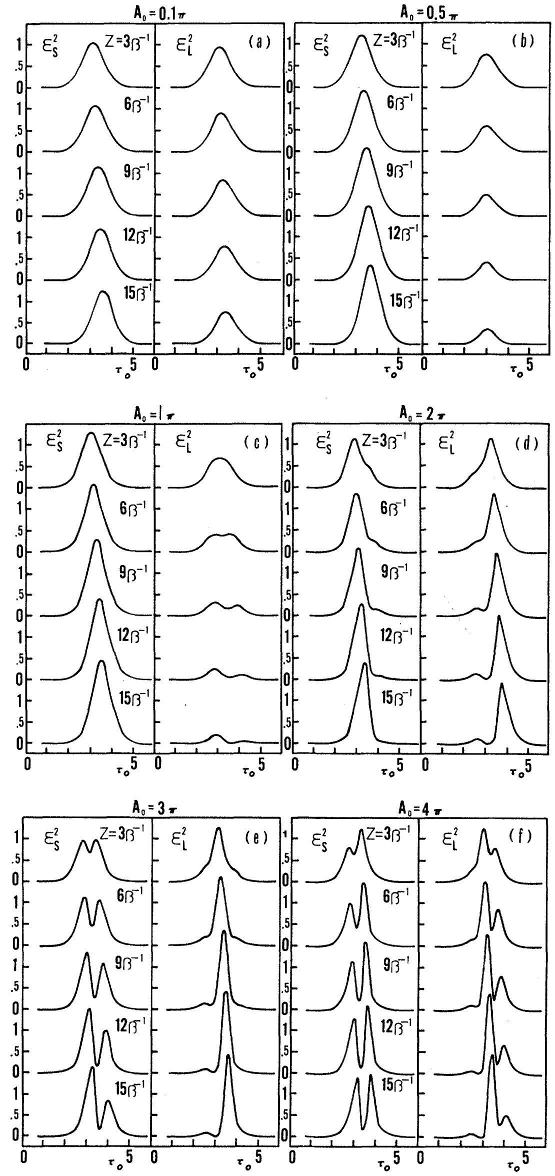

FIG. 3. Computer-generated output pulses showing transient pulse evolution for Gaussian input pulses at the respective frequency. Equal input intensity $\mathcal{E}_S^2(0,t) = \mathcal{E}_L^2(0,t)$ was assumed (i.e., the pulse-height ratio $\xi = 1.0$). The pulse height is normalized to one of the input pulses at $z=0$. A_0 $[=\varphi(0,\infty)]$ in (a)–(f) shows the various input areas in units of π radians. Time is measured in units of $\tau_0 = 10$ psec and the propagating distance z in β^{-1}.

the amplification of the incident Stokes beams corresponds to the usual stimulated Raman scattering in the nonstationary process.[14,16] However, for an area greater than π the laser pulse reasonably affects its own pulse shape with the peak amplification and the pulse narrowing due to the coherently regenerative response of a collection of atomic systems. Attention should be drawn to the interesting fact that the forming of laser pulses takes place with a time lag, in comparison with the Stokes pulse evolution, and also at a higher input area the total number of breakup pulses for both fields is proportional to a number m for an input area $m\pi$. For instance, in Figs. 3(d)–3(f) the peak amplification of Stokes and laser waves alternately appears as a function of time and consequentially yields the pulse breakup. Physically, the anomalous behavior indicates that firstly, for an initial area π, one transition producing the population inversion is achieved accompanying the stimulated absorption of the laser pulse and the relevant stimulated emission to the Stokes pulse. Secondly, in the next π area the population from the excited state causes absorption of the subsequent section of the amplified Stokes wave and amplification for the laser pulse. As a result, the population goes back to the initial ground state at the final time of the 2π cycle. Accordingly, it is not surprising that a given input pulse area more than 2π yields the interesting transient phenomenon of pulse breakup with peak amplification.

We next take into account how the previous results are modified when we vary the input-pulse intensity ratio ξ keeping the input area A_0 constant. Typical pulse formation for $\xi = 10.0$ is plotted in Fig. 4. In this case, sharp peak amplification for the Stokes pulses conspicuously appears in contrast to the case for the laser pulses which maintain a smooth variation in distance and time. It should be noticed that the pulse forming takes place in the leading portion of the laser pulse and does not show marked pulse breakup at any higher input pulse area, analogous to the usual stimulated Raman scattering. The amplification of the pulse energy is a maximum at an input pulse area of π. The reason for this unilateral deep modulation is that a Raman transition within a 2π cycle is ascribed to simultaneous annihilation of a laser pulse and creation of a Stokes pulse with equivalent photons, and the reciprocal process. Accordingly, the modulation of the laser becomes less marked in appearance.

The energy transmission of the incident Stokes and laser pulses are described by the quantities Γ_S and Γ_L, where

$$\Gamma_{S,L} = \int_{-\infty}^{\infty} dt\, \mathcal{E}_{S,L}(z,t) \Big/ \int_{-\infty}^{\infty} dt\, \mathcal{E}_{S,L}(0,t)\,. \qquad (27)$$

The values of Γ_i are plotted against the input area A_0 in Fig. 5 for the equal-intensity input of both beams. The pulse velocity is measured as the shift to the peak of the velocity of light in an inert background as illustrated in Figs. 6 and 7 using the delay and advance times t_{pi}.

The behavior of transmission summarizes the consequence of the above pulse evolutions. Maximum amplification and absorption clearly take place for an input pulse area of π. As concerns the pulse speed, a small delay of the Stokes pulse is seen in a region less than the area π. For an area more than π a pulse advance takes place. Since the pulse evolution originates in the formation of the second-order nonlinear polarization characterized by a sinusoidal function involving the time integral of the product of the two fields, peak evolution due to the stimulated emission in an area less than π is delayed compared with the peak velocity of the incident wave, and also the first peak within the multiple pulses for more than an input pulse area of π may appear near or in front of the

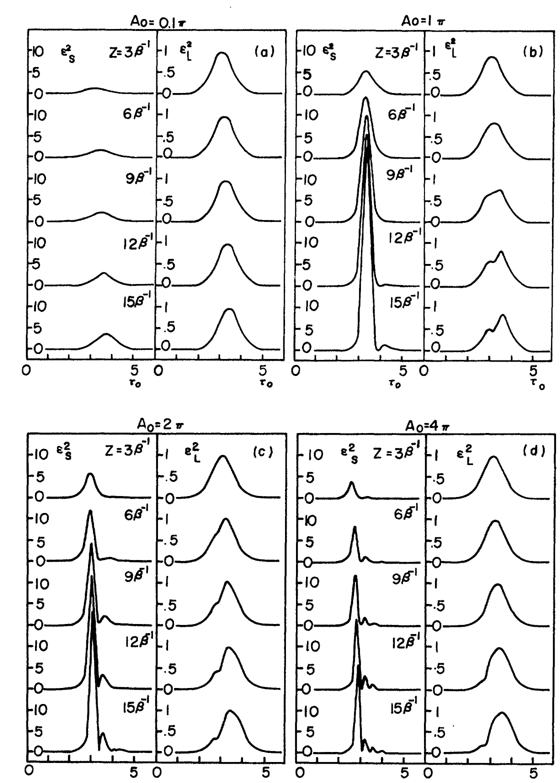

FIG. 4. Transient pulse evolution at the respective frequency plotted for a different input intensity with the ratio $\xi = 10.0$, keeping the input pulse area A_0 constant. (a)–(d) correspond to input areas $A_0 = (0.1 - 4.0)\pi$. Note the factor of 10 increase in the ordinate for the Stokes pulse height. The time and the distance are the same as in Fig. 3.

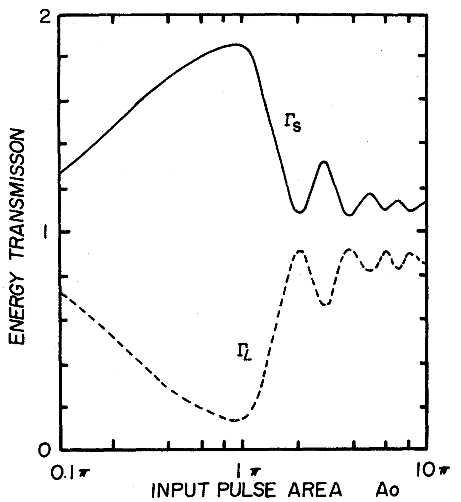

FIG. 5. Summary of the result of the computer calculation for the energy transmission Γ_i from Eq. (27) vs A_0, corresponding to the various pulse evolutions in Fig. 3. Solid line, for the Stokes pulse; dashed curve, for the laser pulse, for an equal input intensity $\xi = 1.0$. No occurrence of full transformation of the input energy from the laser to the Stokes at $A_0 = \pi$ is due to the propagation limit.

pulse center. The pulse advance of the laser wave is ascribed to the residue in the leading portion of the incident pulse after the stimulated absorption expressed by the nonlinear functional polarization of Eq. (16a). The occurrence of the maximum delay at an input pulse area 2π is due to field regeneration on the basis of coherent transition by virtue of the inverted population. These characteristics

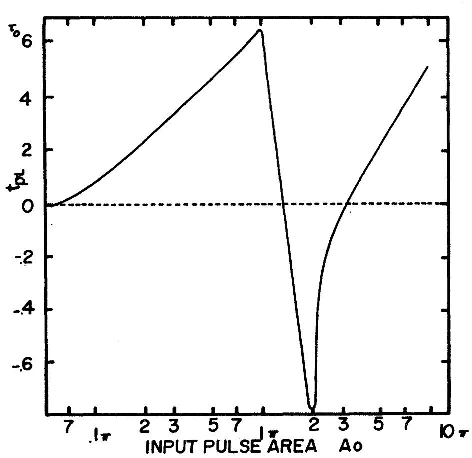

FIG. 6. Here and in Fig. 7 we show plots of pulse advance and delay time of both frequency pulses vs the input area A_0. t_{pL} shows the relative time for the laser. The upper half (plus sign) corresponds to pulse advance, and the lower half (minus sign) to pulse delay with respect to the velocity of light in vacuum.

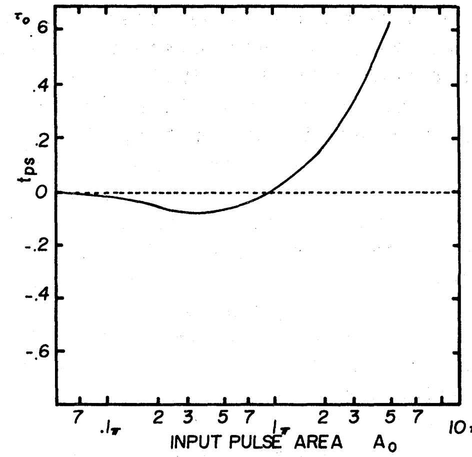

FIG.7. Pulse advance and delay time for the Stokes pulse t_{pS} vs the input pulse area A_0. The sign is the same as in Fig. 6.

are found to be analogous to that of two-photon coherent propagation.[10,11]

The pulse-narrowing property, which depends on the input intensity, is shown in Fig. 8. It appears from the figure that in the region above an area of π, remarkable narrowing of the pulse width for the laser and the Stokes pulses occurs where the coherent effect predominates for the both fields. The coherent Raman process hardly yields the pulse narrowing for the equal-intensity input pulses in a lower-intensity input pulse area. However, for the relatively low-intensity input of the Stokes

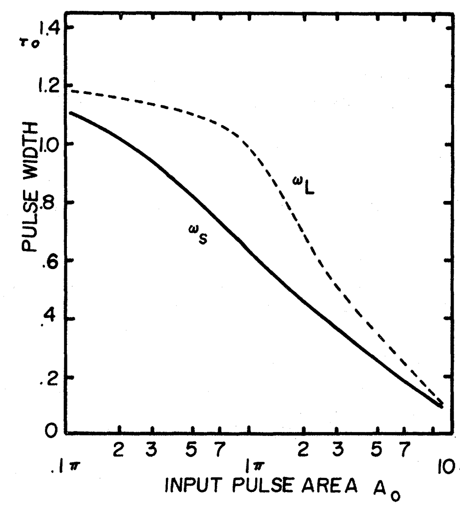

FIG. 8. Summary of the variation of the pulse width at the final propagation stage $z = 15\beta^{-1}$ vs the input area A_0, corresponding to the various pulse evolutions in Fig. 3 for $\xi = 1.0$. The average pulse width for the break-up pulses is plotted. Pulse narrowing of the laser is not remarkable for less than input π-area because of no coherent regeneration for the laser field.

compared with that of the laser, the remarked pulse narrowing is expected as shown in Fig. 4.

A pulse width within the multiply divided pulses is found to be compressed to about 1 psec at higher input pulse areas. In this case the peak power is estimated to be 8×10^{11} W/cm^2 for a π square pulse for the both fields from the above typical values for a Raman-active medium. For an electronic Raman transition in atomic gases, the peak power for the pulse width 1 psec becomes a reasonable value of 2×10^9 W/cm^2 for the typical values of $\mu_{S,L} = 7\times 10^{-18}$ cm esu (oscillator strength $f_{S,L} \simeq 0.6$), $N_0 = 10^{14}/\mathrm{cm}^3$, and $\Delta\omega_0 = 4\times 10^2$ cm^{-1}. This peak amplification due to the pulse breakup and the compression may be easily attained within a self-focusing filament where the high-density field 10^{8-10} W/cm^2 is initially obtained, as pointed out by other researchers.[18-20]

V. SUMMARY AND CONCLUSIONS

In the present paper we have given a theoretical approach for the coherent transient effect in the propagation of the simultaneously entering Stokes and laser pulses in a Raman-active medium. As a result, novel and interesting features were obtained with respect to the pulse evolutions of the laser and the Stokes pulses by introducing the coherent Raman propagation equations, i.e., "Raman Bloch-Maxwell equations." These effects are understood from the interaction of the individual fields by virtue of the second-order induced polarizations on the basis of periodic population inversion. Such intense Raman and laser fields coherently interact with a collection of atoms or molecules. Therefore, the coherent propagation effect causes a periodic amplified self-induced modulation which does produce an ultrashort pulse train within the relaxation times.

Experimental preparation for the observation requires a sufficiently long transverse relaxation time and a high-density incident power. This latter condition may be obtained under the circumstances of a self-focusing filament in a liquid or rarefield gases. Atomic or molecular gases may be available because of the long relaxation times of 10^{-9}–10^{-11} sec. In this case the condition for a coherent two-photon interaction, namely $\kappa \mathcal{E}_S \mathcal{E}_L \gg 1/T_2$, $1/T_1$, which is derived from the Raman-Bloch equations, reasonably holds for the appropriate high-density field.

The present analysis lacks the phase evolution and the frequency chirping at different pulse velocities. However, it appears that the present Raman Bloch-Maxwell equations wholly involve the origin of the analysis for the coherent Raman interaction under the self-consistency requirement. A recent experimental study of coherent Raman scattering in CdS has yielded an interesting phenomenon.[26] Further analysis of coherent Raman scattering with the use of the present equations will be reported elsewhere.

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[1]S. L. McCall and E. L. Hahn, Phys. Rev. 183, 457 (1969).

[2]S. R. Hartmann, IEEE J. Quantum Electron. QE-4, 802 (1968).

[3]N. Tan-no, K. Yokoto, and H. Inaba, Transaction of the Technical Group on Quantum Electronics, Institute of Electronics and Communication Engineers of Japan, Report No. QW70-21, 1970 (in Japanese) (unpublished).

[4]M. Aihara and H. Inaba, J. Phys. A 6, 1709 (1973).

[5]M. Aihara and H. Inaba, J. Phys. A 6, 1725 (1973).

[6]E. M. Belenov and I. A. Poluektov, Zh. Eksp. Teor. Fiz. 56, 1407 (1969) [Sov. Phys.—JETP 29, 754 (1969)].

[7]M. Takatsuji, Phys. Rev. A 4, 808 (1971).

[8]N. Tan-no, K. Yokoto, and H. Inaba, IEEE J. Quantum Electron. QE-8, 591 (1972).

[9]N. Tan-no, K. Yokoto, and H. Inaba, Phys. Rev. Lett. 29, 1211 (1972).

[10]N. Tan-no, K. Yokoto, and H. Inaba, J. Phys. B (to be published).

[11]N. Tan-no, K. Yokoto, and H. Inaba, J. Phys. B (to be published).

[12]N. Tan-no, T. Shirahata, K. Yokoto, and H. Inaba, Phys. Lett. 47A, 241 (1974).

[13]S. L. Shapiro, J. A. Giordmaine, and K. W. Wecht, Phys. Rev. Lett. 19, 1093 (1967).

[14]R. L. Carman, F. Shimizu, C. S. Wang, and N. Bloembergen, Phys. Rev. A 2, 60 (1970).

[15]R. L. Carman, J. Reintjes, and N. Bloembergen, Appl. Phys. Lett. 16, 209 (1970).

[16]S. A. Akhmanov, K. N. Drabovich, A. P. Suchorukov, and A. C. Chirkin, Zh. Eksp. Teor. Fiz. 59, 485 (1970) [Sov. Phys.—JETP 32, 266 (1971)].

[17]M. Maier, W. Kaiser, and J. A. Giordmaine, Phys. Rev. 177, 580 (1969).

[18]S. A. Akhmanov, A. S. Chirkin, K. N. Drabovich, A. I. Kovrigin, R. V. Khokhlov, and A. P. Sukhorukov, IEEE J. Quantum Electron. QE-4, 598 (1968).

[19]K. Shimoda, Japan. J. Appl. Phys. 8, 1499 (1969); Z. Phys. 234, 293 (1970).

[20]E. Courtens, in *Laser Handbook*, edited by F. T. Arecchi *et al*. (North-Holland, Amsterdam, 1972), p. 1261.

[21]F. De Martini and G. Giuliani, Opt. Commun. 11, 42 (1974).

[22]R. L. Shoemaker and R. G. Brewer, Phys. Rev. Lett. 28, 1430 (1972).

[23]F. A. Hopf, R. F. Shea, and M. O. Scully, Phys. Rev.

A 7, 2105 (1973).
[24]R. G. Brewer and E. L. Hahn, Phys. Rev. A 8, 464 (1973).
[25]F. A. Hopf and M. O. Scully, Phys. Rev. 179, 399 (1969).
[26]R. Romestain, G. Geshwind, G. E. Devlin, and P. A. Wolff, Phys. Rev. Lett. 33, 10 (1974).