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Yakir Reuven, photograph and biography not available at the time of publication.

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Nearly Degenerate Four-Wave Mixing Enhanced by the ac Stark Effect

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Abstract—A model is developed to describe nondegenerate four-wave mixing in a medium characterized by a single strong atomic resonance. The nonlinear response is calculated using the density-matrix formalism, and propagation effects are included by solving coupled-wave equations. This model predicts large resonant responses for signals detuned from the pump frequency by approximately the generalized Rabi frequency. Potential applications of the effects predicted by this model include construction of tunable bandpass filters and four-wave parametric oscillators.

I. Introduction

Degenerate four-wave mixing in a nonlinear medium has recently become an important technique in phase conjugation and in aberration correction [1], [2]. Experimental studies have shown that very large nonlinear responses can be obtained by selecting an optical frequency close to a resonant frequency of an atomic system [3], [4]. A theoretical model for degenerate four-wave mixing in a gas of stationary two-level atoms has been presented by Abrams and Lind [5], who find a steady-state solution to the density-matrix equations of motion in the rotating-wave approximation for a single applied optical field. The influence of Doppler effects present in a real gas have also been studied [6].

In nondegenerate four-wave mixing, illustrated in Fig. 1, a signal field at frequency ω_4 interacts with two nearly counterpropagating pump waves at frequency ω_1 to produce a field of frequency $\omega_3 = 2\omega_1 - \omega_4$. For $|\omega_1 - \omega_4| \ll \omega_1$, nondegenerate four-wave mixing preserves in an approximate sense the aberration-correcting properties of degenerate four-wave mixing, and in addition is useful in producing a narrow bandpass filter and in obtaining parametric amplification and parametric oscillation. In the case where all optical frequencies are near one of the resonant frequencies of an atomic system, the two-level-atom approximation can be applied.

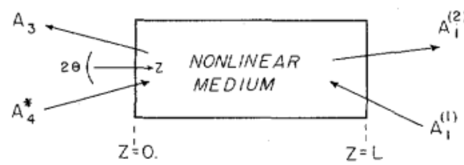

Fig. 1. Block diagram for four-wave mixing in a nonlinear medium.

A solution based on third-order, time-dependent perturbation theory has been presented by Nilsen and Yariv [7] for this case. However, a purely perturbative solution leaves out many of the more interesting features of nondegenerate mixing in a collection of two-level atoms. Recently Fu and Sargent [8] have performed a nonperturbative calculation and have applied it to the case of a pump laser tuned to the atomic resonance.

The present work generalizes that of Fu and Sargent by explicitly treating the case of a pump laser detuned from the atomic resonance. The atomic response to applied fields at frequencies ω_1 and ω_3 is calculated through the use of the density-matrix equations of motion with phenomenological decay terms in the rotating-wave approximation. A steady-state solution is found which is correct to all orders of the amplitude of the strong pump field at ω_1 and is correct to

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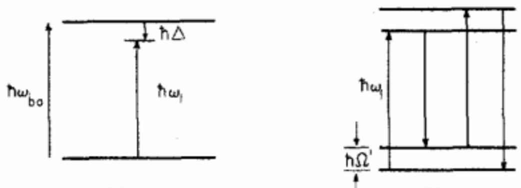

Fig. 2. (a) Two-level atom with energy levels a and b, separated by energy $\hbar\omega_{ba}$, interacts with an optical field of frequency ω_1, detuned from resonance by an amount $\Delta \equiv \omega_1 - \omega_{ba}$. (b) Each atomic energy level splits by an amount $\hbar\Omega'$ by the ac Stark effect. There is a strong nonlinear coupling between the strong field at ω_1 and weak fields at frequencies $\omega_1 \pm \Omega'$.

first order in the amplitudes of the weak signal fields at ω_3 and ω_4. Such an approximation should be adequate for most experimental situations. All effects of atomic motion are ignored.

Although the nonlinear response is calculated by means of the density-matrix equations of motion, many of the results can be understood qualitatively as resulting from the ac Stark effect. If the atom of Fig. 2(a) with ground state a, upper level b, and energy separation $\hbar\omega_{ba}$ is subjected to a strong optical field of amplitude E_1 and frequency ω_1, the population will oscillate between the two levels at an angular frequency given by the generalized Rabi frequency

$$\Omega' = \sqrt{(\omega_{ba} - \omega_1)^2 + \Omega^2}$$

(where $\Omega = 2|\mu_{ba}E_1|/\hbar$ is the Rabi frequency and where the dipole moment μ_{ba} and pump-field amplitude E_1 will be defined more precisely in the following section). This oscillation can, in a sense, be viewed as causing a splitting of each of the energy levels by an amount $\hbar\Omega'$, as shown in Fig. 2(b). This phenomenon is known as the ac Stark effect. This splitting suggests that large nonlinear responses occur for weak signal fields at frequencies $\omega_1 \pm \Omega'$. A more precise mathematical description of this effect and its experimental implications are the primary purposes of this paper.

II. Nonlinear Response of a Two-Level Atom in an Intense Optical Field

In this section, the response of an atom to a strong pump field at frequency ω_1 and a weak probe field at frequency ω_3 is calculated by use of the density-matrix formalism. The equations of motion for the elements ρ_{ij} of the density matrix have the form [9]

$$i\hbar\dot{\rho}_{ab} = -\hbar(\omega_{ba} + i/T_2)\rho_{ab} + V_{ab}(\rho_{bb} - \rho_{aa}) \quad (1a)$$

$$i\hbar\dot{\rho}_{ba} = \hbar(\omega_{ba} - i/T_2)\rho_{ba} - V_{ba}(\rho_{bb} - \rho_{aa}) \quad (1b)$$

$$i\hbar(\dot{\rho}_{bb} - \dot{\rho}_{aa}) = 2(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}) - (i\hbar/T_1)[\rho_{bb} - \rho_{aa} - (\rho_{bb} - \rho_{aa})^0] \quad (1c)$$

where a and b denote, respectively, the ground and upper atomic levels separated by energy $\hbar\omega_{ba}$; T_1 and T_2 are the longitudinal and transverse relaxation times, respectively; and $(\rho_{bb} - \rho_{aa})^0$ is the equilibrium population difference between levels b and a in the absence of the optical fields.

The matrix elements of the interaction energy are given in the rotating-wave approximation by

$$V_{ba} = -\mu_{ba}(E_1 e^{-i\omega_1 t} + E_3 e^{-i\omega_3 t}) = V_{ab^*}. \quad (2)$$

Here the frequency components of the electric field at the position r of the atomic nucleus are given by

$$E_i(r, t) = \hat{\epsilon} E_i e^{-i\omega_i t} + \hat{\epsilon}^* E_i^* e^{i\omega_i t} \quad (3a)$$

where

$$E_1 = A_1^{(1)} e^{ik_1^{(1)}\cdot r} + A_1^{(2)} e^{ik_1^{(2)}\cdot r}$$

$$E_j = A_j e^{ik_j \cdot r}, \quad j = 3, 4 \quad (3b)$$

and where $\hat{\epsilon}$ is the complex polarization unit vector. For convenience, the inner product of $\hat{\epsilon}$ with the matrix element of the dipole operator is denoted as μ_{ba}. The propagation vectors of the two pump beams are designated here as $k_1^{(1)}$ and $k_1^{(2)}$.

Steady-state solutions to these equations of motion can be found using the Fourier transforms of (1a)-(1c). Equations for the Fourier components of the density matrix oscillating at frequencies ω_3, $\omega_3 - \omega_1$, and $\omega_3 - 2\omega_1$, which are correct to all orders of the strong-field amplitude E_1 and correct to first order in the weak-field amplitude E_3, are given by

$$(\omega_3 - \omega_{ba} + i/T_2)\rho_{ba}(\omega_3) = \hbar^{-1}\mu_{ba}E_3(\rho_{bb} - \rho_{aa})^{dc} + \hbar^{-1}\mu_{ba}E_1(\rho_{bb} - \rho_{aa})^{(\omega_3 - \omega_1)} \quad (4a)$$

$$(\omega_3 - 2\omega_1 + \omega_{ba} + i/T_2)\rho_{ab}(\omega_3 - 2\omega_1) = -\hbar^{-1}\mu_{ab}E_1^*(\rho_{bb} - \rho_{aa})^{(\omega_3 - \omega_1)} \quad (4b)$$

$$(\omega_3 - \omega_1 + i/T_1)(\rho_{bb} - \rho_{aa})^{(\omega_3 - \omega_1)} = 2\hbar^{-1}\mu_{ab}E_1^*\rho_{ba}(\omega_3) - 2\hbar^{-1}\mu_{ba}E_1\rho_{ab}(\omega_3 - 2\omega_1) - 2\hbar^{-1}\mu_{ba}E_3\rho_{ab}(-\omega_1) \quad (4c)$$

where $(\rho_{bb} - \rho_{aa})^{dc}$ is the familiar solution to the Bloch equations for the steady-state saturated population difference given by

$$(\rho_{bb} - \rho_{aa})^{dc} = \frac{(1 + (\omega_1 - \omega_{ba})^2 T_2^2)(\rho_{bb} - \rho_{aa})^0}{1 + (\omega_1 - \omega_{ba})^2 T_2^2 + 4\hbar^{-2}|\mu_{ab}|^2|E_1|^2 T_1 T_2}. \quad (5)$$

The off-diagonal matrix element at frequency ω_1

$$\rho_{ba}(\omega_1) = \frac{\hbar^{-1}\mu_{ba}E_1(\rho_{bb} - \rho_{aa})^{dc}}{\omega_1 - \omega_{ba} + i/T_2} \quad (6)$$

was used above as was the general result

$$\rho_{ba}^*(\omega_i) = \rho_{ab}(-\omega_i). \quad (7)$$

Equations (4a)-(4c) are similar to [9, eqs. (3-7) through (3-9)] except for a term which is missing from the referenced equations. Furthermore, (4a)-(4c) are identical to equations which Mollow [10] introduced in his discussion of the ac Stark effect. The intimate connection between near-resonant four-wave mixing and the ac Stark effect will become more apparent in the following development.

Equations (4a)-(4c) can be solved algebraically to give the response at frequencies ω_3 and $\omega_3 - 2\omega_1$ as

$$\rho_{ba}(\omega_3) = \frac{\hbar^{-1}\mu_{ba}E_3(\rho_{bb} - \rho_{aa})^{dc}}{D(\omega_3)} \cdot \left[(\omega_3 - \omega_1 + i/T_1)(\omega_3 - 2\omega_1 + \omega_{ba} + i/T_2) - \frac{2\hbar^{-2}|\mu_{ab}|^2|E_1|^2(\omega_3 - \omega_1)}{(\omega_1 - \omega_{ba} - i/T_2)}\right] \tag{8a}$$

$$\rho_{ab}(\omega_3 - 2\omega_1) = 2\hbar^{-3}\mu_{ab}|\mu_{ab}|^2E_1^{*2}E_3 \cdot \frac{(\rho_{bb} - \rho_{aa})^{dc}(\omega_3 - \omega_1 + 2i/T_2)}{D(\omega_3)(\omega_1 - \omega_{ba} - i/T_2)} \tag{8b}$$

in terms of the function $D(\omega_3)$ defined by

$$D(\omega_3) \equiv (\omega_3 - \omega_1 + i/T_1)(\omega_3 - \omega_{ba} + i/T_2) \cdot (\omega_3 - 2\omega_1 + \omega_{ba} + i/T_2) - 4\hbar^{-2}|\mu_{ab}|^2|E_1|^2(\omega_3 - \omega_1 + i/T_2). \tag{9}$$

The real and imaginary parts of $\rho_{ba}(\omega_3)$ and the modulus of $\rho_{ab}(\omega_3 - 2\omega_1)$ are shown in Fig. 3(a)-(c), for $T_2/T_1 = 2$, indicating pure radiative damping. Since ρ_{ab} is proportional to the expectation value of the dipole moment induced by the applied fields, the imaginary part of $\rho_{ba}(\omega_3)$ corresponds to the gain or loss experienced by a weak beam at ω_3 in the presence of a strong beam at ω_1, whereas the real part of $\rho_{ba}(\omega_3)$ contributes to the refractive index for a field at frequency ω_3 in the presence of a strong field at ω_1. The response represented by the real and imaginary parts of $\rho_{ab}(\omega_3 - 2\omega_1)$ corresponds to the generation of a signal at $\omega_3 - 2\omega_1$ due to fields at ω_1 and ω_3, and thus corresponds to four-wave parametric amplification. The experimental implications of these results are discussed in the conclusion.

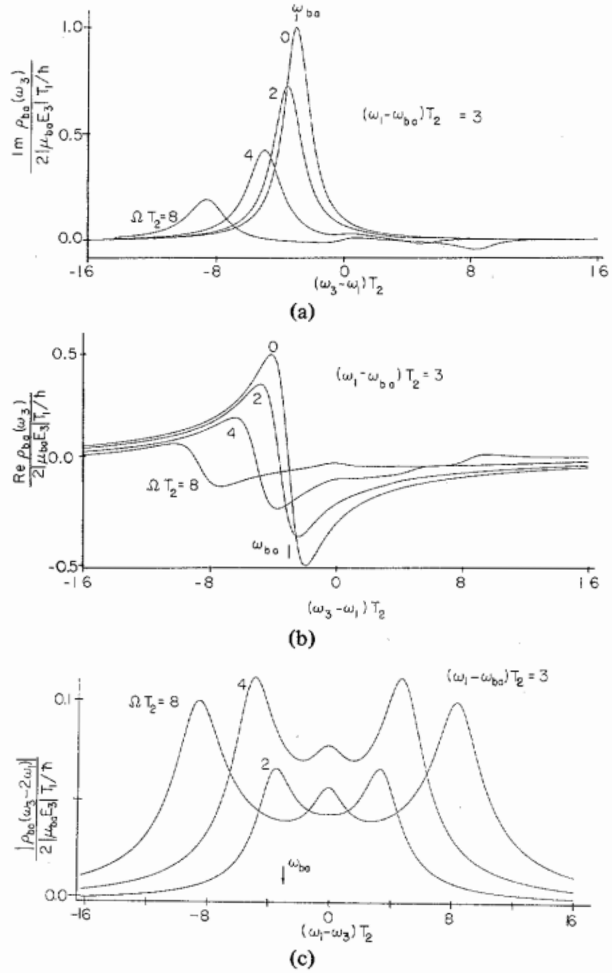

Fig. 3. (a) The imaginary part of $\rho_{ba}(\omega_3)$. As the pump-field amplitude, and thus Ω increases, positive absorption developes at negative values of $\omega_3 - \omega_1$; negative absorption develops at positive values of $\omega_3 - \omega_1$; and the positive absorption at $\omega_3 = \omega_{ba}$ decreases due to saturation. (b) The real part of $\rho_{ba}(\omega_3)$. (c) The modulus of $\rho_{ab}(\omega_3 - 2\omega_1)$, which gives the coupling between the fields at ω_1 and ω_3 and the field at $\omega_3 - 2\omega_1$. $T_2/T_1 = 2$ in all cases.

III. Solution to the Wave Equation with the Nonlinear Polarization

The treatment thus far has dealt with the response of a single atom to the applied optical fields. In this section, propagation effects are considered by treating the nonlinear polarization proportional to ρ_{ba} as a source term in the Helmholtz wave equation and finding approximate solutions under the assumption of slowly varying probe-field amplitudes.

The assumed geometry is shown in Fig. 4 where weak, nearly counterpropagating signal waves at frequencies ω_3 and at $\omega_4 = 2\omega_1 - \omega_3$ interact with each other and with strong nearly counterpropagating pump waves of equal frequency ω_1. It is assumed that the pump waves are unaffected by the probe waves and by absorption, and thus the two pump waves can be assumed to have the equal and constant amplitudes A_1. The phase mismatch is proportional to the propagation-vector mismatch which is given by

$$\Delta k = k_1^{(1)} + k_1^{(2)} - k_3 - k_4 \tag{10}$$

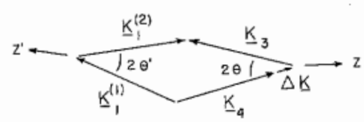

Fig. 4. The geometry of the four-wave interaction in terms of the wave vectors.

and can be varied by changing the angle $2\theta'$ between the pump waves or by changing the angle between $k_1^{(2)}$ and k_4.

The polarizations at ω_3 and ω_4 are given by

$$P(\omega_3) = -N\mu_{ab}(\rho_{ba}(\omega_3) + \rho_{ba}(2\omega_1 - \omega_4)) \tag{11a}$$

and

$$P^*(\omega_4) = -N\mu_{ba}(\rho_{ab}(-\omega_4) + \rho_{ab}(\omega_3 - 2\omega_1)) \tag{11b}$$

where N is the number density of atoms, and where $\rho_{ab}(-\omega_4)$ and $\rho_{ba}(2\omega_1 - \omega_4)$ are found by replacing the appropriate frequencies and field in (8). These polarizations can be used as source terms in the one-dimensional Helmholtz equations for frequencies ω_3 and ω_4 which, for $\theta \ll 1$, are given by

$$\frac{\partial^2 E_3}{\partial z^2} + k_3^2 E_3 = -4\pi N k_3^2 \mu_{ab} [\rho_{ba}(\omega_3) + \rho_{ba}(2\omega_1 - \omega_4)] \tag{12a}$$

$$\frac{\partial^2 E_4^*}{\partial z^2} + k_4^2 E_4^* = -4\pi N k_4^2 \mu_{ba} [\rho_{ab}(-\omega_4) + \rho_{ab}(\omega_3 - 2\omega_1)] \tag{12b}$$

where

$$k_i = \omega_i/c, \quad i = 3, 4. \tag{13}$$

The integration of these equations is complicated by their coupling through their source terms and also by the rapid spatial variation of the source terms caused by the interference between the two waves at frequency ω_1. To display this variation it is necessary to introduce (8a) and (8b) for $\rho_{ba}(\omega_3)$ and $\rho_{ba}(2\omega_1 - \omega_3)$ into (12). The resulting expressions are greatly simplified through the use of the notation

$$\delta \equiv T_2^2(\omega_1 - \omega_{ba})^2 + 1 \tag{14a}$$

$$\gamma_3 \equiv [(\omega_3 - \omega_1)T_1 + i][(\omega_3 - \omega_{ba})T_2 + i] \cdot [T_2(\omega_3 - 2\omega_1 + \omega_{ba}) + i] \tag{14b}$$

$$\xi_3 \equiv -[(\omega_3 - \omega_1)T_2 + i] \tag{14c}$$

$$\Omega_0^2 \equiv 4\hbar^{-2}|\mu_{ab}|^2 T_1 T_2 \tag{14d}$$

$$\alpha_0 \equiv 2\pi\hbar^{-1} k_3 N |\mu_{ab}|^2 T_2 (\rho_{aa} - \rho_{bb})^0 \tag{14e}$$

$$\alpha_3' \equiv -4\alpha_0 k_3 \hbar^{-2} |\mu_{ab}|^2 T_2 T_1 (\omega_3 - \omega_1) \times (1 + (\omega_1 - \omega_{ba})^2 T_2^2)(\omega_1 - \omega_{ba} + i/T_2) \tag{14f}$$

$$\alpha_3 \equiv 2\alpha_0 k_3 T_2 T_1 (\omega_3 - \omega_1 + i/T_1) \cdot (\omega_3 - 2\omega_1 + \omega_{ba} + i/T_2) \times (1 + (\omega_1 - \omega_{ba})^2 T_2^2) \tag{14g}$$

$$\kappa_3 \equiv -4\alpha_0 \hbar^{-2} |\mu_{ab}|^2 T_2 (T_2^2(\omega_1 - \omega_{ba})^2 + 1) \times (\omega_3 - \omega_1 + 2i/T_2)/(\omega_1 - \omega_{ba} + i/T_2). \tag{14h}$$

The Helmholtz equation for the wave at ω_3 then becomes

$$\frac{\partial^2 E_3}{\partial z^2} + k_3^2 E_3 = \frac{(\alpha_3 + \alpha_3'|E_1|^2)E_3 - \kappa_3 E_1^2 E_4^*}{(\delta + \Omega_0^2|E_1|^2)(\gamma_3 + \xi_3 \Omega_0^2 |E_1|^2)} \tag{15}$$

where

$$E_1 = 2A_1 \cos(k_1 \cos\theta' z). \tag{16}$$

The right-hand side of (15) contains several spatial-frequency components in addition to those phase-matched terms which vary at nearly $e^{ik_3 z}$ and which can thus act as efficient source terms for the field E_3. If the right-hand side of (15) is Fourier analyzed and the nonphase-matched components are neglected, the wave equation (15) becomes

$$\frac{\partial^2 E_3}{\partial z^2} + k_3^2 E_3 = \left(4\alpha_3 \frac{N(\omega_3)}{D'(\omega_3)} + \frac{4\alpha_3'|A_1|^2}{D'(\omega_3)}\right) E_3 - 4\kappa_3 A_1^2 \frac{E_4^*}{D'(\omega_3)} \tag{17}$$

where

$$D'(\omega_3) = \delta\gamma_3 \left\{ \left(1 + \frac{4\Omega_0^2|A_1|^2}{\delta}\right)^{1/2} \left(1 + \frac{4\xi_3\Omega_0^2|A_1|^2}{\gamma_3}\right)^{1/2} \cdot \left[\left(1 + \frac{4\Omega_0^2|A_1|^2}{\delta}\right)^{1/2} + \left(1 + \frac{4\xi_3\Omega_0^2|A_1|^2}{\gamma_3}\right)^{1/2} \right] \right\} \tag{18a}$$

and where

$$N(\omega_3) = \frac{\delta\xi_3\Omega_0^2|A_1|^2 \left(1 + \frac{4\Omega_0^2|A_1|^2}{\delta}\right)^{1/2} - \gamma_3\Omega^2|A_1|^2 \left(1 + \frac{4\xi_3\Omega_0^2|A_1|^2}{\gamma_3}\right)^{1/2}}{\gamma_3\delta \left[\left(1 + \frac{4\xi_3\Omega_0^2|A_1|^2}{\gamma_3}\right)^{1/2} - \left(1 + \frac{4\Omega_0^2|A_1|^2}{\delta}\right)^{1/2} \right]}. \tag{18b}$$

This procedure of dropping the nonphase-matched terms is equivalent to averaging the right-hand side of (15) over a region of several wavelengths extent [5], but gives physical justification to this procedure.

If it is further assumed that, in contrast to the rapid spatial variation displayed by the standing pump waves, the spatial variations of the signal amplitudes A_3 and A_4^* are small on the scale of a wavelength so that

$$\left| \frac{d^2 A_i}{dz^2} \right| \ll \left| k_i \frac{dA_i}{dz} \right| \quad i = 3, 4$$

then the wave equation (17) can be approximated by

$$ik_3 \frac{\partial A_3}{\partial z} = \frac{(4\alpha_3 N(\omega_3) + 4\alpha_3'|A_1|^2)A_3 - 4\kappa_3 A_1^2 A_4^* e^{i\Delta kz}}{D'(\omega_3)} \tag{19}$$

where Δk is the z-component of $\Delta \boldsymbol{k}$.

A first-order differential equation for A_4^* can similarly be derived by equations analogous to (14)–(19) giving the result

$$-ik_4 \frac{\partial A_4^*}{\partial z} = \frac{(4\alpha_4 N(\omega_4) + 4\alpha_4'|A_1|^2)A_4^* + 4\kappa_4 A_1^2 A_3 e^{-i\Delta kz}}{D'(\omega_4)}. \tag{20}$$

These coupled equations can be solved if specific boundary conditions are assumed. For comparison with the work of other authors, boundary conditions are taken of the form $A_4^*(0) = A_4^*(\text{input}) \neq 0$ and $A_3(L) = A_3(\text{input}) = 0$, where L is the length of the medium. For this case we can obtain a nonlinear reflectance R and nonlinear transmittance T given by

$$R \equiv \left|\frac{A_3(\text{output})}{A_4^*(\text{input})}\right|^2 = \left|\frac{A_3(0)}{A_4^*(0)}\right|^2 = \left|\frac{2\kappa_3^0 \sin \beta L}{2\beta \cos \beta L + (\alpha_4^0 + \alpha_3^0 - i\Delta k)\sin \beta L}\right|^2 \quad (21)$$

and

$$T \equiv \left|\frac{A_4^*(\text{output})}{A_4^*(\text{input})}\right|^2 = \left|\frac{A_4^*(L)}{A_4^*(0)}\right|^2 = \left|\frac{2\beta \exp \frac{1}{2}(\alpha_3^0 - \alpha_4^0 - i\Delta k)L}{2\beta \cos \beta L + (\alpha_3^0 + \alpha_4^0 - i\Delta k)\sin \beta L}\right|^2 \quad (22)$$

where

$$\beta = \left(\kappa_4^0 \kappa_3^0 - \left(\frac{\alpha_4^0 + \alpha_3^0}{2} - \frac{i\Delta k}{2}\right)^2\right)^{1/2} \quad (23a)$$

$$\alpha_3^0 = \frac{4\alpha_3 N(\omega_3) + 4\alpha_3' |A_1|^2}{ik_3 D'(\omega_3)} \quad (23b)$$

$$\alpha_4^0 = \frac{4\alpha_4 N(\omega_4) + 4\alpha_4' |A_1|^2}{-ik_4 D'(\omega_4)} \quad (23c)$$

$$\kappa_3^0 = \frac{-4\kappa_3}{ik_3 D'(\omega_3)} \quad (23d)$$

$$\kappa_4^0 = \frac{-4\kappa_4}{ik_4 D'(\omega_4)}. \quad (23e)$$

These rather complicated expressions for R and T are displayed for typical cases in Figs. 5 and 6, respectively, where the curves are labeled according to their spatially averaged Rabi frequency $\overline{\Omega} \equiv |\mu_{ba} A_1|/\hbar$ and their pump-wave detuning $(\omega_1 - \omega_{ba})T_2$.

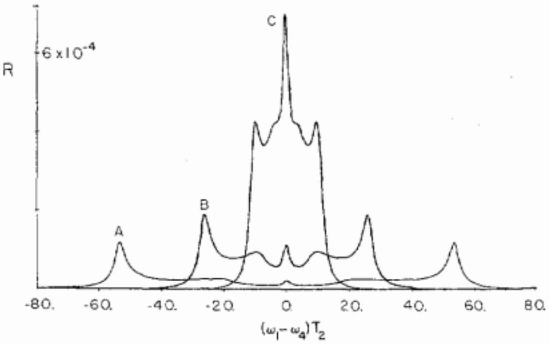

Fig. 5. Nonlinear reflectance $R \equiv |A_3(\text{output})/A_4^*(\text{input})|^2$ as a function of the weak-field detuning. The peaks at the origin correspond to degenerate four-wave mixing; the peaks at larger values of $(\omega_1 - \omega_4)T_2$ are resonantly enhanced by the ac Stark effect. Curve A: $(\omega_1 - \omega_{ba})T_2 = 19$, $\overline{\Omega}T_2 = 25$. Curve B: $(\omega_1 - \omega_{ba})T_2 = 8$, $\overline{\Omega}T_2 = 12.5$. Curve C: $(\omega_1 - \omega_{ba})T_2 = 3$, $\overline{\Omega}T_2 = 5$. $\alpha_0 L = 1$, $\Delta k = 0$, and $T_2/T_1 = 2$ in all cases.

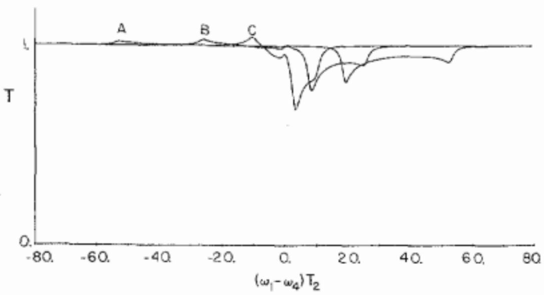

Fig. 6. Nonlinear transmittance $T \equiv |A_4^*(\text{output})/A_4^*(\text{input})|^2$ as a function of the weak-field detuning. If $\omega_1 - \omega_4 = -\overline{\Omega}$, the field ω_4 is amplified; while if $\omega_1 - \omega_4 = \overline{\Omega}$, it is attenuated. Curve A: $(\omega_1 - \omega_{ba})T_2 = 19$, $\overline{\Omega}T_2 = 25$. Curve B: $(\omega_1 - \omega_{ba})T_2 = 8$, $\overline{\Omega}T_2 = 12.5$. Curve C: $(\omega_1 - \omega_{ba})T_2 = 3$, $\overline{\Omega}T_2 = 5$. $\alpha_0 L = 1$, $\Delta k = 0$, and $T_2/T_1 = 2$ in all cases.

IV. Conclusions

Some of the predictions of the previous sections have already been verified by experimental studies. The imaginary part of $\rho_{ba}(\omega_3)$ shown in Fig. 3(a) corresponds to absorption of a field at frequency ω_3. The negative absorption corresponding to negative values of $\rho_{ba}(\omega_3)$ has been observed [11] and has been called the three-photon effect. This process corresponds to a transition from the upper level of the ground-state ac Stark-split doublet to the lower level of the excited-state ac Stark-split doublet by the absorption of two photons of frequency ω_1 and the emission of a photon of energy $\omega_1 + \Omega'$. In other studies, a field at frequency ω_3 has been observed to grow from noise both by the three-photon effect [12] and by the phase-matched parametric interaction [13], [14].

The calculations presented here also predict some new experimental effects. The nonlinear reflectance has a single symmetrical peak for low values of the pump field strength as is predicted by a perturbative treatment [7]. Fig. 5 shows that as the pump field strength increases, the reflectance curve broadens and develops additional peaks in the wings. These new peaks result from the ac Stark effect as illustrated in Fig. 2(b). Due to the interference of the pump beams, the local Rabi frequency shows rapid spatial variations. Thus, in Figs. 5 and 6, the enhancement due to the ac Stark effect extends over the range $0 \leqslant (\omega_3 - \omega_1)T_2 \leqslant 2\overline{\Omega}T_2$. The enhancement is greatest at the maximum and near the minimum values of the Rabi frequency since the most likely values of a harmonically varying Rabi frequency are its maximum and minimum.

For a pump laser tuned to the atomic resonance, our numerical results agree with those of Fu and Sargent [8]. If the pump laser is tuned off the atomic resonance frequency, large values of $\alpha_0 L$ can be used without pump-wave depletion. [α_0 is the line-center, weak-field, amplitude absorption coefficient of (14e)]. With sufficiently large values of $\alpha_0 L$, we find that the nonlinear reflectance and transmittance can be larger than unity for certain values of ω_1, ω_2, and E_1, thus leading to gain. Since the peak of the nonlinear reflectance curve changes with pump power, we predict that a tunable narrow-bandpass filter can be constructed using nondegenerate four-wave mixing resonantly enhanced by the ac Stark effect. In addition, it should be possible to utilize the high gain in the transmitted signal wave as shown in Fig. 6 to construct a tunable, four-wave parametric oscillator.

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