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 \( \omega_3 \) interacts with two nearly counterpropagating pump waves at frequency \( \omega_1 \) to produce a field of frequency \( \omega_2 = 2\omega_1 - \omega_3 \). For \( |\omega_1 - \omega_3| \ll |\omega_3| \), 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.

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 detuned 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 \( \omega_3 \) and \( \omega_1 \) 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 \( \omega_1 \) and is correct to

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first order in the amplitudes of the weak signal fields at \( \omega_3 \) and \( \omega_4 \). 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. This phenomenon is known as the ac Stark effect. This splitting suggests that large nonlinear responses occur for weak signal fields at frequencies \( \omega_3 \) and \( \omega_4 \). Such an approximation should be adequate for most experimental situations. All effects of atomic motion are ignored.

In this section, the response of an atom to a strong pump field at frequency \( \omega_1 \) and a weak probe field at frequency \( \omega_3 \) is calculated by use of the density-matrix formalism. The equations of motion for the elements \( \rho_{ij} \) of the density matrix have the form [9]

\[
\begin{align*}
\hbar \dot{\rho}_{ab} &= -\hbar (\omega_{ba} + i/T_2) \rho_{ab} + V_{ab}(\rho_{bb} - \rho_{aa}) \quad (1a) \\
\hbar \dot{\rho}_{ba} &= \hbar (\omega_{ab} - i/T_2) \rho_{ba} - V_{ba}(\rho_{bb} - \rho_{aa}) \quad (1b) \\
\hbar (\dot{\rho}_{bb} - \dot{\rho}_{aa}) &= 2(V_{ba} \rho_{ab} - \rho_{bb} V_{ab}) \\
&- (\hbar/T_1)(\rho_{bb} - \rho_{aa} - (\rho_{bb} - \rho_{aa})^0) \quad (1c)
\end{align*}
\]

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

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

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

\[
E_i(r, t) = e^{i(k_i r)} e^{-i\omega_i t} + e^{i(k_i r)} e^{i\omega_i t}
\]

where

\[
E_1 = A_1^{(1)} e^{i(k_1 r)} + A_2^{(1)} e^{i(k_1 r)}
\]

and where \( \hat{e} \) is the complex polarization unit vector. For convenience, the inner product of \( \hat{e} \) with the matrix element of the dipole operator is denoted as \( \mu_{ba} \). 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 \( \omega_3 \), \( \omega_4 \), and \( \omega_1 \), 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

\[
\begin{align*}
(\omega_3 - \omega_{ba} + i/T_2) \rho_{ba}(\omega_3) &= -\hbar^{-1} \mu_{ba} E_3 (\rho_{bb} - \rho_{aa}) e^{i\omega_3 t} + \hbar^{-1} \mu_{ba} E_1 (\rho_{bb} - \rho_{aa}) e^{i\omega_1 t} \\
(\omega_3 - 2\omega_1 + \omega_{ba} + i/T_2) \rho_{ba}(\omega_3 - 2\omega_1) &= -\hbar^{-1} \mu_{ab} E_3^* (\rho_{bb} - \rho_{aa}) e^{i\omega_3 t} - \hbar^{-1} \mu_{ba} E_1^* (\rho_{bb} - \rho_{aa}) e^{i\omega_1 t} \\
(\omega_3 - \omega_1 + i/T_2) (\rho_{bb} - \rho_{aa}) e^{i\omega_3 t} &= 2\hbar^{-1} \mu_{ab} E_3 (\rho_{bb} - \rho_{aa}) e^{i\omega_1 t} - 2\hbar^{-1} \mu_{ba} E_1 (\rho_{bb} - \rho_{aa}) e^{i\omega_3 t}
\end{align*}
\]

where \( (\rho_{bb} - \rho_{aa}) e^{i\omega_3 t} \) is the familiar solution to the Bloch equations for the steady-state saturated population difference given by

\[
(\rho_{bb} - \rho_{aa}) e^{i\omega_1 t} = \frac{1 + (\omega_1 - \omega_{ba})^2 T_1^2}{1 + (\omega_1 - \omega_{ba})^2 T_1^2 + 4 \hbar^2 |\mu_{ba}|^2 |E_1|^2 T_1 T_2}
\]

The off-diagonal matrix element at frequency \( \omega_1 \)

\[
\rho_{ba}(\omega_1) = \frac{\hbar^{-1} \mu_{ba} E_1 (\rho_{bb} - \rho_{aa}) e^{i\omega_1 t}}{\omega_1 - \omega_{ba} + i/T_2}
\]

was used above as was the general result

\[
\rho_{ba}(\omega) = \rho_{ab}(\omega).
\]

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-reso-
nant 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 \( \omega_3 \) and \( \omega_3 - 2\omega_1 \) as

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

(8a)

\[
\rho_{ab}(\omega_3 - 2\omega_1) = 2 \hbar^{-3} \mu_{ab} |\mu_{ab}|^2 E_1^* E_3 \cdot \left[ (\omega_{ba} - \rho_{ba}) de (\omega_3 - \omega_1 + 2i/T_2) \right] \]

(8b)

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

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

(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 \( \rho_{ab} \) 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 \( \omega_3 \) in the presence of a strong beam at \( \omega_1 \), whereas the real part of \( \rho_{ba}(\omega_3) \) contributes to the refractive index for a field at frequency \( \omega_3 \) in the presence of a strong field at \( \omega_1 \). The response represented by the real and imaginary parts of \( \rho_{ba}(\omega_3 - 2\omega_1) \) corresponds to the generation of a signal at \( \omega_3 - 2\omega_1 \) due to fields at \( \omega_1 \) and \( \omega_3 \), and thus corresponds to four-wave parametric amplification. The experimental implications of these results are discussed in the conclusion.

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 \( \rho_{ba} \) 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 \( \omega_3 \) and at \( \omega_3 = 2\omega_1 - \omega_3 \) interact with each other and with strong nearly counterpropagating pump waves of equal frequency \( \omega_1 \). 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

\[
\Delta k = k^{(1)}_1 + k^{(2)}_{-1} - k_3 - k_4
\]

(10)
\[ \frac{\partial^2 E_3}{\partial z^2} + k_3^2 E_3 = -4\pi Nk_3^2 \mu_{ab} \rho_{ba}(\omega_3) + \rho_{ab}(2\omega_1 - \omega_4) \]  

(12a)

\[ \frac{\partial^2 E_3^*}{\partial z^2} + k_3^2 E_3^* = -4\pi Nk_3^2 \mu_{ba} \rho_{ab}(-\omega_4) + \rho_{ab}(\omega_5 - 2\omega_1) \]  

(12b)

where

\[ k_i = \omega_i/c, \quad i = 3, 4. \]  

(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 \( \omega_1 \). To display this variation it is necessary to introduce (8a) and (8b) for \( \rho_{ba}(\omega_3) \). The Helmholtz equation for the wave at \( \omega_3 \) then becomes

\[ \delta \frac{\partial^2 E_3}{\partial z^2} = \frac{\delta^2 E_3}{\partial z^2} + k_3^2 E_3 = \left[ \frac{\delta}{\gamma_3} \Omega_3^2 |A_1|^2 \left( 1 + \frac{4\delta |A_1|^2}{\gamma_3} \right)^{1/2} - \gamma_3 \Omega_3^2 |A_1|^2 \left( 1 + \frac{4\delta |A_1|^2}{\gamma_3} \right)^{1/2} \right] \]  

(18b)

and

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 \( A_2 \) are small on the scale of a wavelength so that

\[ \left| \frac{d^2 A_i}{dz^2} \right| \ll \left| \frac{d^2 A_i}{dz^2} \right| \quad i = 3, 4 \]  

then the wave equation (17) can be approximated by

\[ \frac{d^2 A_3}{dz^2} = \frac{\left( 4\alpha_3 N(\omega_3) + 4\alpha_3 |A_1|^2 A_3 - 4\kappa_3 A_3^* A_3 e^{i\Delta k z} \right)}{D'(\omega_3)} \]  

(19)

where \( \Delta k \) is the z-component of \( \Delta k \).

A first-order differential equation for \( A_2^* \) can similarly be derived by equations analogous to (14)-(19) giving the result

\[ -i k_3 \frac{\partial A_2^*}{\partial z} = \left( 4\alpha_3 N(\omega_3) + 4\alpha_3 |A_1|^2 A_2 - 4\kappa_3 A_2^* A_2 e^{i\Delta k z} \right) \]  

(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

\[ A_3^*(0) = A_3^*(\text{input}) \neq 0 \quad \text{and} \quad A_3(L) = A_3(\text{input}) = 0, \]  

where \( L \) is the length of the medium. For this case we can obtain a nonlinear reflectance \( R \) and nonlinear transmittance \( T \) given by
Due local Rabi frequency shows rapid spatial variations. Thus, in 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

\[ R \equiv \left| \frac{A_3(\text{output})}{A_4(\text{input})} \right|^2 = \left| \frac{A_3(0)}{A_4(0)} \right|^2 = \frac{2\kappa^2}{2\beta \cos \beta L + (\alpha^2 + \alpha^2 - i\Delta k) \sin \beta L} \]

and

\[ T \equiv \left| \frac{A_3(\text{output})}{A_4(\text{input})} \right|^2 = \left| \frac{A_3(0)}{A_4(0)} \right|^2 = \frac{2\beta \exp \frac{1}{2} (\alpha^2 - \alpha^2 - i\Delta k) L}{2\beta \cos \beta L + (\alpha^2 + \alpha^2 - i\Delta k) \sin \beta L} \]

where

\[ \beta = \left( \kappa^2 - \frac{(\alpha^2 + \alpha^2 - i\Delta k)^2}{2} \right)^{1/2} \]

\[ \alpha^2 = \frac{4\alpha_0 N(\omega_2) + 4\alpha'_2 |A_1|^2}{ik_2 D(\omega_2)} \]

\[ \alpha^2 = \frac{4\alpha_0 N(\omega_3) + 4\alpha'_3 |A_1|^2}{-ik_2 D(\omega_3)} \]

\[ \kappa^2 = \frac{-4k_2}{ik_2 D(\omega_2)} \]

\[ \kappa^2 = \frac{-4k_2}{ik_2 D(\omega_3)} \]

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 \( \Omega \equiv |\mu_{ba}A_1|/h \) and their pump-wave detuning \( (\omega_1 - \omega_{ba})T_2 \).

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 \( \omega_3 \). 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 \( \omega_1 \) and the emission of a photon of energy \( \omega_2 + \Omega' \). In other studies, a field at frequency \( \omega_3 \) 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

\[ R = |A_3(\text{output})/A_4(\text{input})|^2 \]

\[ T = |A_3(\text{output})/A_4(\text{input})|^2 \]

Fig. 5. Nonlinear reflectance \( R = |A_3(\text{output})/A_4(\text{input})|^2 \) 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 \( (\omega_1 - \omega_{ba})T_2 \) are resonantly enhanced by the ac Stark effect. Curve A: \( (\omega_1 - \omega_{ba})T_2 = 19, \Omega T_2 = 25 \). Curve B: \( (\omega_1 - \omega_{ba})T_2 = 8, \Omega T_2 = 12.5 \). Curve C: \( (\omega_1 - \omega_{ba})T_2 = 3, \Omega T_2 = 5. \) \( \alpha_0L = 1, \Delta k = 0 \), and \( T_3/T_1 = 2 \) in all cases.

Figs. 5 and 6, the enhancement due to the ac Stark effect extends over the range \( 0 \leq (\omega_3 - \omega_1)T_2 \leq 2\Omega T_2 \). 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 \( \omega_3 \) can be used without pump-wave depletion. \( \omega_0L = 1 \) is the line-center, weak-field, amplitude absorption coefficient of (14e)]. With sufficiently large values of \( \omega_0L \), we find that the nonlinear reflectance and transmittance can be larger than unity for certain values of \( \omega_1 \), \( \omega_2 \), and \( \Delta k \), thus leading to gain. Since the peak of the nonlinear reflectance curve changes with pump power, we predict that a tunable narrowbandpass 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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REFERENCES


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