Four-wave mixing under conditions when optical Bloch equations fail

S. Singh and G. S. Agarwal

School of Physics, University of Hyderabad, Hyderabad-500 134, India

Received August 18, 1987; accepted September 28, 1987

The nonlinear-optical properties of a system whose collisional dynamics are strongly affected by intense pump fields are calculated. The four-wave mixing signals can be affected in a dramatic way if the period of the Rabi oscillation is comparable with the correlation time of the collisional mechanism.

Relaxation processes[1] play an important role in nonlinear-optical problems. The structure of the nonlinear susceptibilities depends on the form of the relaxation matrix.[2,3] Collisional relaxation has been shown to lead to a variety of extra resonances in various nonlinear mixing problems. The form of the relaxation matrix that has generally been used in nonlinear-optical phenomena is taken to be independent of the applied fields. However, when the applied fields become strong, then the relaxation matrix should depend on the intensities of the applied fields. Such intensity-dependent relaxation would in turn change the form of the nonlinear-optical susceptibilities. It should be borne in mind that when the fields become strong, then one needs nonperturbative expressions for appropriately defined susceptibilities. Thus at high intensities one has to account for saturation effects and field-dependent changes in the relaxation matrix. These two problems were considered in detail[4–8] in the context of the experiments of Devoe and Brewer[9] on free induction decay. These authors discovered situations in which the usual optical Bloch equations with phenomenological relaxation constants cannot be used. Using microscopic considerations involving the competition between saturation and relaxation effects, one can derive a modified version[4–8] of optical Bloch equations. This modification can be used to understand phenomena that depend strongly on the intensity-dependent relaxation matrix. Clearly one should examine the changes in the nonlinear-optical effects for field intensities in a regime where the optical Bloch equations fail. In this paper we report our preliminary results on four-wave mixing under such conditions. Instead of using T_1 and T_2, we model the relaxation effects in a stochastic way. We assume that the relaxation arises from a stochastic modulation[4–8] of the frequency of the two-level atom, i.e., we take the instantaneous frequency of the atom as $\omega_0 + x(t)$, where $x(t)$ is a stochastic Markov process, with

$$\langle x(t)\rangle = 0, \qquad \langle x(t)x(t')\rangle = x_0{}^2 \exp(-\Gamma\,|t-t'|). \tag{1}$$

Let the total field acting on the atom be

$$\mathbf{E}(t) = \epsilon \exp(i\mathbf{k}_0\cdot\mathbf{r})\exp(-i\omega t) + \epsilon_3 \exp(i\mathbf{k}_3\cdot\mathbf{r} - i\omega_3 t) + \text{c.c.} \tag{2}$$

Then, in a frame rotating with the frequency ω of the pump, the equation of motion can be written in matrix form as

$$\dot{\psi} = C_0\psi + ix(t)C_1\psi + g + C_p(t)\psi, \tag{3}$$

where

$$\psi = \begin{bmatrix} \rho_{21}\exp(-i\omega t) \\ \rho_{12}\exp(i\omega t) \\ \tfrac{1}{2}(\rho_{11}-\rho_{22}) \end{bmatrix}, \qquad g = \begin{bmatrix} 0 \\ 0 \\ -\gamma_0 \end{bmatrix},$$

$$C_1 = \begin{bmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \qquad C_0 = \begin{bmatrix} i\Delta-\gamma_0 & 0 & 2i\alpha \\ 0 & -i\Delta-\gamma_0 & -2i\alpha \\ i\alpha & -i\alpha & -2\gamma_0 \end{bmatrix},$$

$$\Delta = \omega_0 - \omega, \qquad \alpha = -\mathbf{d}\cdot\epsilon \exp(i\mathbf{k}_0\cdot\mathbf{r}),$$

$$C_p(t) = \begin{bmatrix} 0 & 0 & 2i\alpha_p{}^*\exp(i\Omega t) \\ 0 & 0 & -2i\alpha_p\exp(-i\Omega t) \\ i\alpha_p\exp(-i\Omega t) & -i\alpha_p{}^*\exp(i\Omega t) & 0 \end{bmatrix},$$

$$\alpha_p = -\mathbf{d}\cdot\epsilon_3\exp(i\mathbf{k}_3\cdot\mathbf{r}), \qquad \Omega = \omega_3 - \omega. \tag{4}$$

Here $2\gamma_0$ is the Einstein A coefficient of the excited state, $|1\rangle$. We will now calculate the four-wave mixing signal valid to all orders in the pump intensity $|\epsilon|^2$ but valid only to second order in the probe intensity $|\epsilon_3|^2$. The modulation $x(t)$ is also to be accounted for exactly. The intense field changes the basic time scales of the system. For example, several Rabi oscillations are possible within the correlation time of the modulation, and this is responsible for the breakdown of the optical Bloch equations. The next step is to eliminate the stochastic character of Eq. (3), i.e., to obtain the equation for the ensemble average of ψ. To do so one needs the statistical properties of $x(t)$. Techniques exist for treating Gaussian processes[10]; however, this requires extensive numerical work. A reasonable understanding of the optical phenomena when time-dependent changes in the relaxation matrix are important can be obtained by assuming $x(t)$ to be a dichotomic Markov process.[7,8] In such a case one can get an exact equation[11] for $\langle\psi(t)\rangle$.

The four-wave mixing signal can be obtained if we evaluate ψ to first order in α_p. The first-order contribution $\psi^{(1)}$ to ψ is given by

$$\dot{\psi}^{(1)} = C_0\psi^{(1)} + ix(t)C_1\psi^{(1)} + C_p(t)\psi^{(0)}. \tag{5}$$

0740-3224/88/020254-05$02.00

For the dichotomic Markov process $x(t)$, Eq. (5) leads to the exact result (cf. Ref. 11)

$$\langle \dot{\psi}^{(1)} \rangle = C_0 \langle \psi^{(1)} \rangle + C_p(t) \langle \psi^{(0)} \rangle - x_0^2 \int_0^t d\tau C_1 \exp[C_0(t-\tau) - \Gamma(t-\tau)] C_1 \langle \psi^{(1)}(\tau) \rangle - x_0^2 \int_0^t d\alpha C_1 \exp[C_0(t-\alpha)] C_p(\alpha) \int_0^\alpha d\beta \times \exp[C_0(\alpha-\beta)] C_1 \langle \psi^{(0)}(\beta) \rangle \exp[-\Gamma(t-\beta)]. \quad (6)$$

The zeroth-order response is obtained from the solution of

$$\dot{\psi}^{(0)} = C_0 \psi^{(0)} + ix(t) C_1 \psi^{(0)} + g, \quad (7)$$

which leads to the exact equation for $\langle \psi^{(0)} \rangle$:

$$\langle \dot{\psi}^{(0)} \rangle = C_0 \langle \psi^{(0)} \rangle + g - x_0^2 \int_0^t d\tau C_1 \times \exp[(C_0 - \Gamma)(t-\tau)] C_1 \langle \psi^{(0)}(\tau) \rangle. \quad (8)$$

Note that $C_p(t)$ has the form

$$C_p(t) = \exp(-i\Omega t) C_+ + \exp(i\Omega t) C_-. \quad (9)$$

By using Eqs. (9) and (6), the steady-state response can be written as

$$\psi^{(1)}(t) = \exp(-i\Omega t) \psi_+ + \exp(i\Omega t) \psi_-, \quad (10)$$

where

$$\psi_- = [i\Omega - C_0 + x_0^2 C_1 (i\Omega + \Gamma - C_0)^{-1} C_1]^{-1} \times [C_- \langle \psi^{(0)}(\infty) \rangle - x_0^2 C_1 (i\Omega + \Gamma - C_0)^{-1} \times C_-(\Gamma - C_0)^{-1} C_1 \langle \psi^{(0)}(\infty) \rangle], \quad (11)$$

with

$$\langle \psi^{(0)}(\infty) \rangle = [x_0^2 C_1 (\Gamma - C_0)^{-1} C_1 - C_0]^{-1} g. \quad (12)$$

The four-wave mixing susceptibility $\chi^{(3)}(\omega, \omega, -\omega_3)$ is obtained from the second component of the column matrix, ψ_-. Note that ψ_- depends on all powers of the pump intensity (this dependence is through C_0). In the limit $\Gamma \to \infty$, Eqs. (11) and (12) reduce to the results Φ_- and $\langle \Phi^{(0)}(\infty) \rangle$, obtained from optical Bloch equations:

$$\langle \Phi^{(0)}(\infty) \rangle = -\left(C_0 - \frac{x_0^2}{\Gamma} C_1 C_1 \right)^{-1} g, \quad (13)$$

$$\Phi_- = \left(i\Omega - C_0 + \frac{x_0^2}{\Gamma} C_1 C_1 \right)^{-1} C_- \langle \Phi^{(0)}(\infty) \rangle. \quad (14)$$

The differences between the predictions based on Eqs. (11) and (14) start becoming more and more pronounced as the eigenvalues of C_0 start becoming comparable in magnitude with Γ. Note that x_0^2/Γ defines the usual transverse relaxation width. Thus we define

$$\frac{1}{T_2} = \gamma_0 + \frac{x_0^2}{\Gamma}, \qquad \frac{1}{T_1} = 2\gamma_0 \quad (15)$$

and express all the other parameters in units of $1/T_2$. In Figs. 1 and 2 we show the real and the imaginary parts of the four-wave mixing susceptibility $(\psi_-)_2$. We also show the differences in $\chi^{(3)}(\omega, \omega, -\omega_3)$ as predicted by the exact result [Eq. (11)] and the one following from optical Bloch equations. We see that the nonlinear susceptibility $\chi^{(3)}$ changes dramatically as the random modulation starts acquiring a larger correlation time. In Fig. 1 we show the comparison between the results of the two theories. Even for $\Gamma = 10$ the differences between the two theories are quite noticeable. In Figs. 2(a) and 2(b) we display the real and imaginary parts

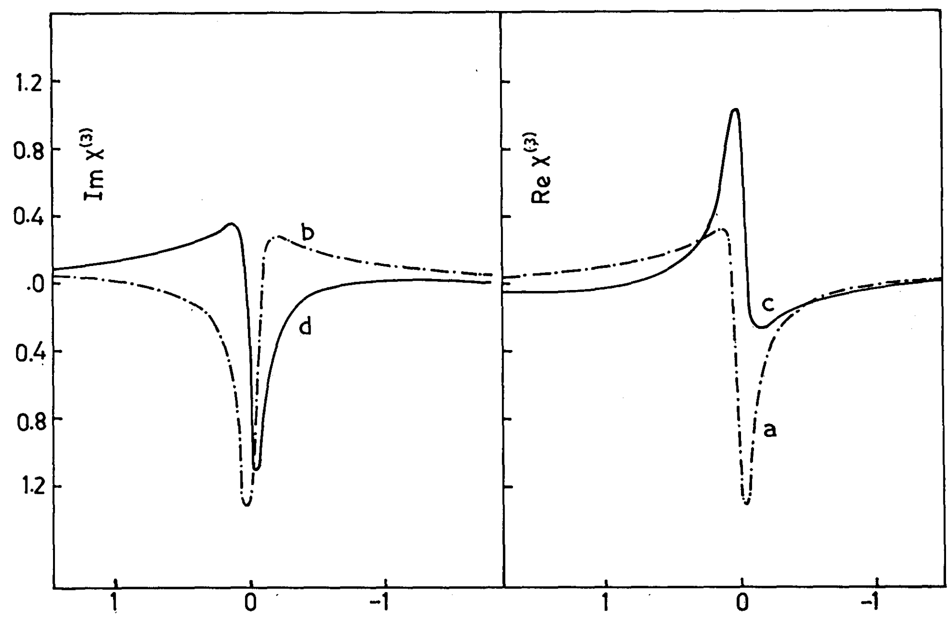
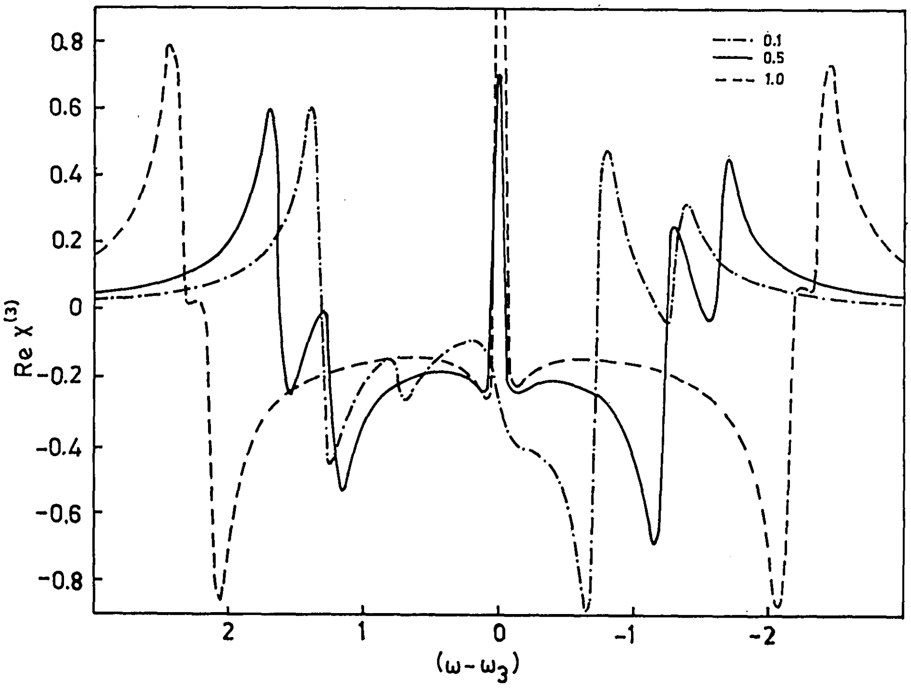
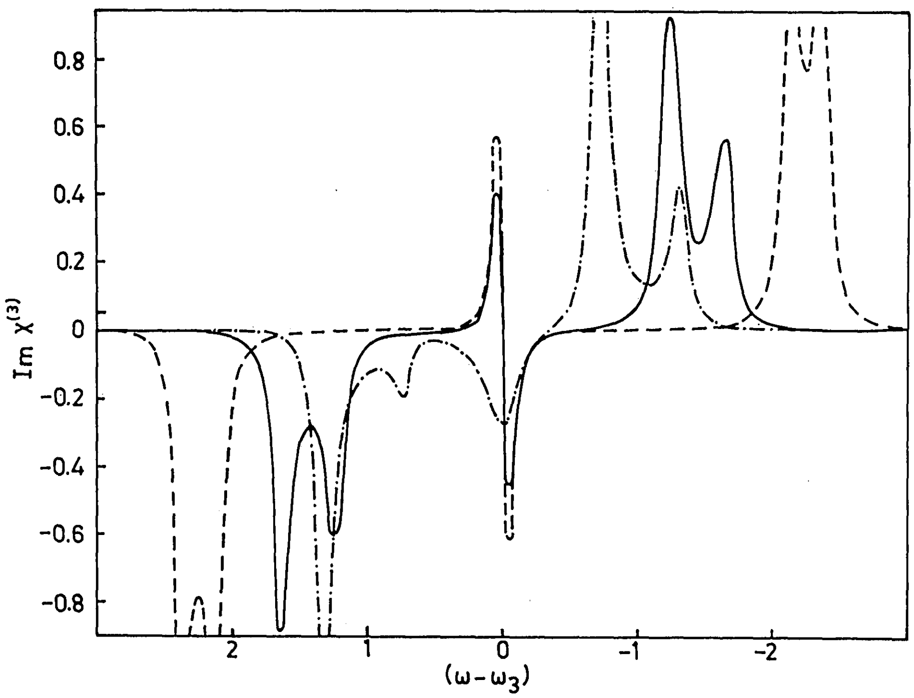


Fig. 1. The real (curves a and c) and the imaginary (curves b and d) parts of the four-wave mixing susceptibility as a function of $\omega - \omega_3$, where ω_3 is the frequency of the probe. Curves a and b were obtained from the present theory [Eq. (11)] for $\Gamma = 10$, whereas curves c and d are based on Eq. (14), obtained from the conventional Bloch equations. All the parameters are scaled in terms of $1/T_2 = \gamma_0 + (x_0^2/\Gamma)$, $\alpha = 0.1$, and $\omega_0 - \omega = 1$.



(a)



(b)

Fig. 2. (a) The real and (b) the imaginary parts of the four-wave mixing susceptibility [Eq. (11)] as a function of $\omega - \omega_3$ for $\Gamma = 0.1$, pump atom detuning, $\omega_0 - \omega = 1$, and various values of the pump-field strength α: 0.1 for the dashed–dotted curve, 0.5 for the solid curve, and 1.0 for the dashed curve. All parameters are in units of $1/T_2$.

of $\chi^{(3)}$ for the case when the deviations from the predictions of the optical Bloch equations are most remarkable. This can be seen from an examination of Figs. 1 and 2 for $\alpha = 0.1$. The nonlinear susceptibility exhibits many new resonances. These new resonances can be understood in terms of the roots of the determinant of the matrix $[z - C_0 + x_0{}^2C_1(z + \Gamma - C_0)^{-1}C_1]$. For large α, small Γ, and $\Delta = 0$, such roots, to order Γ^2/α^2, are given by

$$\pm 2i\alpha - \Gamma \pm \frac{i\Gamma}{4\alpha}, \pm 2i\alpha \pm \frac{i\Gamma}{4\alpha}, 0, -\Gamma. \tag{16}$$

Thus the signal would be dominated by very narrow lines at

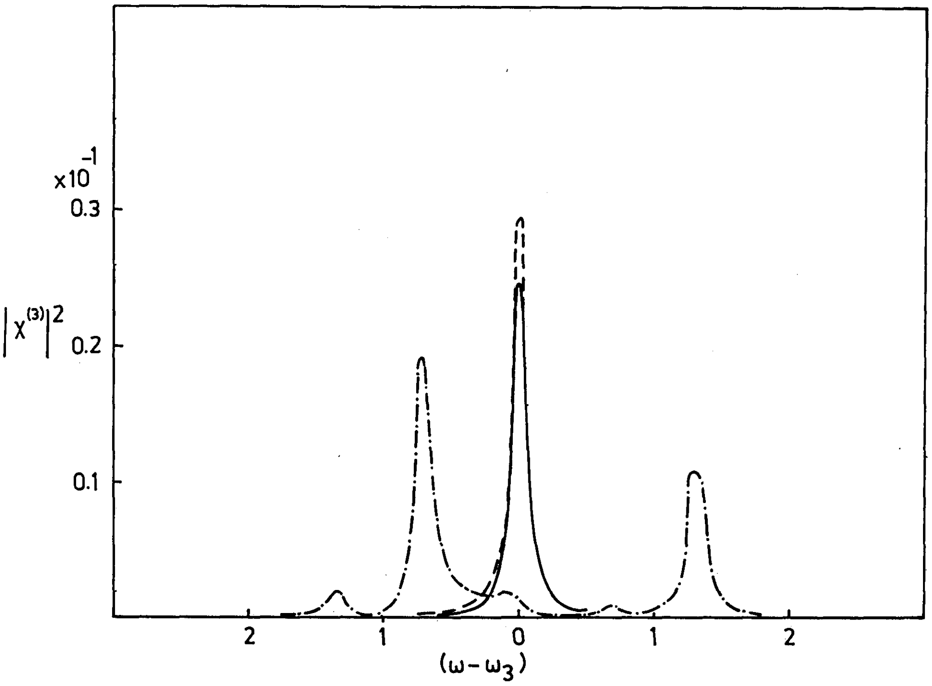


Fig. 3. The four-wave mixing signal as a function of $\omega - \omega_3$ for $\alpha = 0.1$ and $\omega_0 - \omega = 1$. The solid line is the signal obtained from the conventional Bloch equations. The dashed line and the dashed-dotted lines are those obtained by the present theory for $\Gamma = 10$ and $\Gamma = 0.1$, respectively.

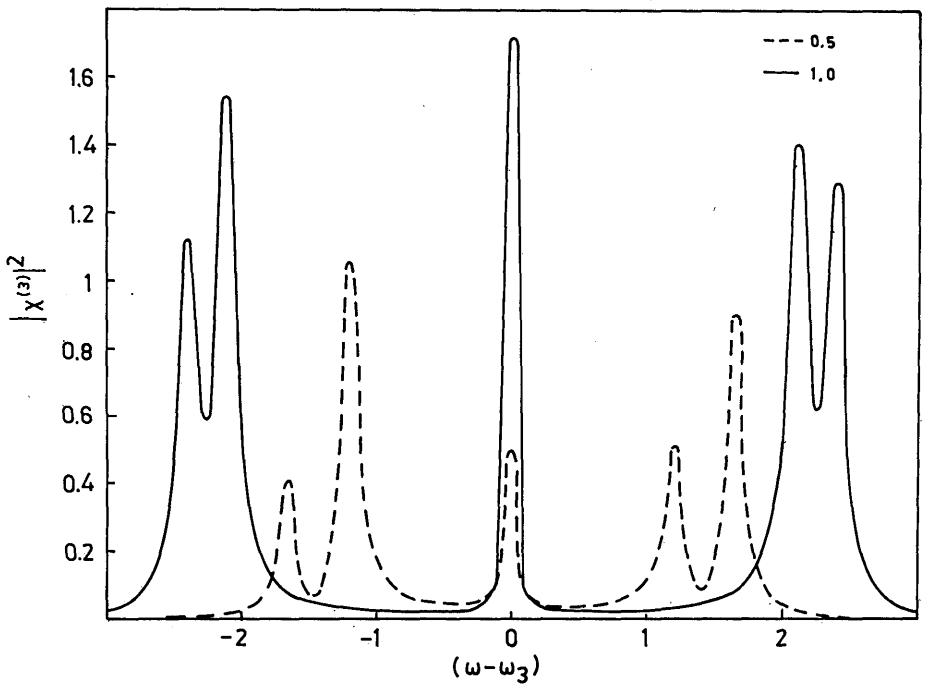


Fig. 4. The four-wave mixing signal as a function of $\omega - \omega_3$ for $\Gamma = 0.1$ and $\omega_0 - \omega = 1$, as obtained from the present theory. Dashed and solid lines are for $\alpha = 0.5$ and $\alpha = 1.0$, respectively.

$\pm 2i\alpha$ and 0 with widths $\sim(\Gamma^2/\alpha^2)$. There is a splitting of the side peaks if $\Delta \neq 0$. The full four-wave mixing signal $S \propto |\chi^{(3)}|^2$ is shown in Figs. 3 and 4. Figure 3 shows that, even for $(\Gamma/\alpha) = 1$, the deviations from the predictions of optical Bloch equations are quite significant. The new resonances become better resolved with an increase in α, as shown in Fig. 4.

Thus, in conclusion, we have shown in the framework of a model calculation the changes in the coherent signals generated by four-wave mixing in pump fields that are strong enough to invalidate the optical Bloch equations. Similar changes are expected for the other nonlinear processes and for more-complex media.

REFERENCES

1. C. Flytzanis, in *Quantum Electronics*, H. Rabin and C. L. Tang, eds. (Academic, New York, 1975), p. 9.
2. N. Bloembergen, H. Lotem, and R. T. Lynch, Ind. J. Pure Appl. Phys. **16**, 151 (1978).
3. M. S. Kumar and G. S. Agarwal, Phys. Rev A **33**, 1817 (1986).
4. E. Hanamura, J. Phys. Soc. Jpn. **52**, 2267, 3265, 3678 (1983); M. Yamanoi and J. H. Eberly, Phys. Rev. Lett. **52**, 1353 (1984), J. Opt. Soc. Am. B **1**, 751 (1984); H. Tsunetsugu, T. Taniguchi, and E. Hanamura, Solid State Commun. **52**, 663 (1984).
5. P. R. Berman, J. Opt. Soc. Am. B **3**, 564 (1986); S. Yakilin and A. P. Nizovtsev, J. Phys. B **19**, 3457 (1986).
6. A. Schenzle, M. Mitsunaga, R. G. Devoe, and R. G. Brewer, Phys. Rev. A **30**, 325 (1984).
7. K. Wodkiewicz and J. H. Eberly, Phys. Rev. A **32**, 992 (1985).
8. G. S. Agarwal, Opt. Acta **32**, 981 (1985).
9. R. G. Devoe and R. G. Brewer, Phys. Rev. Lett. **50**, 1269 (1983).
10. A. T. Georges, Phys. Rev. A **21**, 2034 (1980); application of these techniques to the present problem will be reported elsewhere.
11. G. S. Agarwal, Z. Phys. B **33**, 111 (1979).