# Multiphoton processes in two-level atoms in two intense pump beams

G. S. Agarwal and N. Nayak

*School of Physics, University of Hyderabad, Hyderabad—500134, India*

Received November 2, 1983; accepted November 29, 1983

The dynamical behavior of a two-level atom under the influence of two intense pump beams is examined. Exact equations for the nonlinear susceptibilities to all orders in the field strengths are derived. Numerical results for the nonlinear susceptibilities giving the absorption of the radiation from either of the pump beams and the generation of the four-wave mixing signal are given. The susceptibility for four-wave mixing, in addition to showing various saturation peaks, shows a peak at $\omega_1 = \omega_2$ even in the case of radiative relaxation. Higher-order multiphoton peaks in the absorption spectra are found to be extremely sensitive to the temporal fluctuations in the pumps.

## 1. INTRODUCTION

The dynamical behavior of a two-level atom in the presence of a single-mode field is well understood.[1] Such a dynamical behavior can be probed[2–8] by using another external beam of a different frequency, assuming that the probe field is weak. Such probe-field absorption spectra are known[2] to show regions of the parametric gain provided that the pumping field is strong enough to saturate the two-level transition. The predictions on the weak-probe-field absorption spectra have been experimentally verified[3,4] using both rf and optical fields. Some years ago Bonch-Bruevich *et al.* also investigated the effect of the temporal fluctuations[4] of the pump field on the weak-field absorption spectra. Their results are in good agreement with the theoretical models of the pump-field fluctuations.[9,10] Bonch-Bruevich *et al.* later carried out a series of experiments in which even the probe field was taken to be an intense one.[8] The new spectra, when both pump and probe were intense, exhibited a number of remarkable features, for example, the appearance of the subradiative structures. Toptygina and Fradkin[11] have recently presented a theoretical explanation for the appearance of subradiative structure and various multiphoton absorption peaks. Since fluctuations of the pump beams were an important factor in the weak-field spectra, the question arises: What happens to the multiphoton absorption peaks if the temporal fluctuations of the two intense fields pump and probe[12] are taken into account? This is the object of our present theoretical study. Another problem that has attracted considerable attention is four-wave mixing[13] in two-level atoms. If the probe and the pump fields are weak, then the four-wave mixing, the generation of a coherent signal at $2\omega_2 - \omega_1$, is described by the nonlinear susceptibility $\chi^{(3)}(-2\omega_2 + \omega_1, \omega_2, \omega_2, -\omega_1)$; such a nonlinear susceptibility in the usual perturbation treatment is independent of the field strengths. However, if the fields are strong enough to saturate the transition, then $\chi^{(3)}$ becomes intensity dependent. It is then desirable to know the variation of $\chi^{(3)}$ as a function of the intensities, relaxation parameters, etc. Our analysis also yields this information along with a host of other higher-order[14] $\chi$'s.

In Section 2 we formulate the basic problem. In order to keep the system of equations tractable, we adopt the phase diffusion model to describe the temporal fluctuations in each pump. Previous experience has shown that this model is quite adequate. Exact equations for the ensemble averages of various atomic operators are given. In Section 3 the results of the numerical calculations are given. The rate of the energy absorption from each field is calculated for arbitrary values of the intensities of the two fields. The effect of the field temporal fluctuations on the multiphoton absorption peaks is discussed in detail. In Section 4 we investigate numerically the behavior of $\chi^{(3)}$ and discuss the various saturation peaks that $\chi^{(3)}$ can show. We also discuss the effect of relaxation parameters on $\chi^{(3)}$ and show that, because of saturation, the type of extra resonance discussed by Bloembergen and coworkers[15] can also arise even in the case of radiative relaxation.

## 2. BASIC DYNAMICAL EQUATIONS IN THE FIELD OF TWO FLUCTUATING BEAMS

In this section we formulate dynamical equations giving the behavior of a two-level atom with states $|1\rangle$ and $|2\rangle$ separated by $\hbar\omega_0$, in the field of two fluctuating fields. The fluctuations in each field are described by the phase diffusion model, i.e., we write the fields in the form

$$\begin{aligned}\mathbf{E}(t) &= \epsilon_1 \exp[-i\omega_1 t - i\Phi_1(t)] \\ &\quad + \epsilon_2 \exp[-i\omega_2 t - i\Phi_2(t)] + \text{c.c.}, \\ &= \epsilon(t) + \text{c.c.},\end{aligned} \tag{2.1}$$

with

$$\dot{\Phi}_i(t) = \mu_i(t), \qquad i = 1,2. \tag{2.2}$$

The $\mu_i$'s are delta-correlated Gaussian random processes with zero mean:

$$\langle \mu_i(t) \rangle = 0, \qquad \langle \mu_i(t)\mu_j(t') \rangle = 2\Gamma_{ij}\delta(t - t'). \tag{2.3}$$

The optical Bloch equations in the rotating wave approximation are given by

0740-3224/84/020164-05$02.00 

$$\frac{d}{dt}\begin{bmatrix}\rho_{21}\\ \rho_{12}\\ \rho_{11}-\rho_{12}\end{bmatrix}=\begin{bmatrix}i\omega_0-\frac{1}{T_2} & 0 & +i\mathbf{d}\cdot\epsilon^*(t) & \rho_{21}\\ 0 & -i\omega_0-\frac{1}{T_2} & -i\mathbf{d}\cdot\epsilon(t) & \rho_{12}\\ 2i\mathbf{d}\cdot\epsilon(t) & -2i\mathbf{d}\cdot\epsilon^*(t) & -\frac{1}{T_1} & \rho_{11}-\rho_{22}\end{bmatrix}+\begin{bmatrix}0\\ 0\\ \frac{\rho_{11}^{(0)}-\rho_{22}^{(0)}}{T_1}\end{bmatrix}, \quad (2.4)$$

where $\mathbf{d}$ is the dipole matrix element. On making the transformation to the rotating frame

$$\rho_{21}=\exp[i\omega_2 t+i\Phi_2(t)]\psi_1,$$
$$\rho_{12}=\exp[-i\omega_2 t-i\Phi_2(t)]\psi_2,$$
$$\rho_{11}-\rho_{22}=2\psi_3, \quad (2.5)$$

we can write Eq. (2.4) in the following matrix form:

$$\dot{\psi}=A\psi+i\mu_2(t)f\psi+I+B_+\exp\{i[\Phi(t)+\Omega t]\}\psi + B_-\exp\{-i[\Phi(t)+\Omega t]\}\psi,$$
$$\Omega=\omega_1-\omega_2, \qquad \Phi=\Phi_1-\Phi_2, \quad (2.6)$$

where the nonvanishing elements of $f$, $I$, and $B_\pm$ are

$$f_{11}=-1, \quad f_{22}=+1, \quad I_3=\eta\equiv(\rho_{11}^{(0)}-\rho_{22}^{(0)})/T_1,$$
$$(B_+)_{13}=2i\mathbf{d}\cdot\epsilon_1^*, \quad (B_+)_{32}=-i\mathbf{d}\cdot\epsilon_1^*,$$
$$(B_-)_{23}=-2i\mathbf{d}\cdot\epsilon_1, \quad (B_-)_{31}=i\mathbf{d}\cdot\epsilon_1, \quad (2.7)$$

and

$$A=\begin{bmatrix}i\Delta-\frac{1}{T_2} & 0 & 2i\mathbf{d}\cdot\epsilon_2^*\\ 0 & -i\Delta-\frac{1}{T_2} & -2i\mathbf{d}\cdot\epsilon_2\\ i\mathbf{d}\cdot\epsilon_2 & -i\mathbf{d}\cdot\epsilon_2^* & -\frac{1}{T_1}\end{bmatrix},$$
$$\Delta=\omega_0-\omega_2. \quad (2.8)$$

We next perform the ensemble average over the fluctuations of the fields, i.e., over the phases $\Phi_1$ and $\Phi_2$. For this purpose we define

$$\psi^{[n]}=\exp\{i[\Omega t+\Phi(t)]n\}\psi, \quad (2.9)$$

the equation of motion for which is given by

$$\dot{\psi}^{[n]}=A\psi^{[n]}+i\mu_2(t)f\psi^{[n]}+(i\Omega n+i\mu_1 n-i\mu_2 n)\psi^{[n]} + B_+\psi^{[n+1]}+B_-\psi^{[n-1]}+I\exp\{in[\Omega t+\Phi(t)]\}. \quad (2.10)$$

Ensemble averaging can now be done by using the theory of multiplicative stochastic processes as explained in Ref. 9, with the result that

$$\langle\dot{\psi}^{[n]}\rangle=A\langle\psi^{[n]}\rangle+B_+\langle\psi^{[n+1]}\rangle+B_-\langle\psi^{[n-1]}\rangle+\delta_{n0}I + [in\Omega-\Gamma_1 n^2-\Gamma_2(f-n)^2 - 2\Gamma_{12}n(f-n)]\langle\psi^n\rangle. \quad (2.11)$$

where $\Gamma_1=\Gamma_{11}$ and $\Gamma_2=\Gamma_{22}$ represent the bandwidths of the beams at frequencies $\omega_1$ and $\omega_2$, respectively, and $\Gamma_{12}$ represents the correlation between the two beams [Eqs. (2.3)]. The steady-state situation is simple as $\langle\dot{\psi}^{[n]}\rangle=0$. These equations are strictly valid for $\Phi(t)\neq 0$, so that the well-defined steady states exist. Equation (2.11) is a recursion relation involving $\langle\psi^{[n]}\rangle$ and $\langle\psi^{[n\pm1]}\rangle$. We now reduce Eq. (2.11) to be a single equation for $\langle\psi_3{}^{[n]}\rangle$, whose solution is written in the form of a continued fraction.

A calculation using the components of Eq. (2.11) shows that, in the steady state,

$$\langle\psi_1{}^{[n]}\rangle=(2ig_2\langle\psi_3{}^{[n]}\rangle+2ig_1\langle\psi_3{}^{[n+1]}\rangle)/D_n, \quad (2.12)$$
$$\langle\psi_2{}^{[n]}\rangle=(-2ig_2\langle\dot{\psi}_3{}^{[n]}\rangle-2ig_1\langle\psi_3{}^{[n-1]}\rangle)/D_{-n}{}^*, \quad (2.13)$$

with $g_i$ ($\equiv\mathbf{d}\cdot\epsilon$) representing coupling of the field at $\omega_i$ with the atom end

$$D_n=\frac{1}{T_2}-i\Delta_2-in\Omega+\Gamma_1 n^2 + \Gamma_2(n+1)^2+2\Gamma_{12}n(n+1). \quad (2.14)$$

The equation for $\langle\psi_3{}^{[n]}\rangle\equiv X_n$ has the form

$$a_n X_n+b_n X_{n+1}+c_n X_{n-1}=\eta\delta_{n0}, \quad (2.15)$$

where

$$a_n=\frac{1}{T_1}+n^2(\Gamma_1+\Gamma_2-2\Gamma_{12})-in\Omega + \frac{2g_2{}^2}{D_{-n}{}^*}+\frac{2g_1{}^2}{D_{n-1}}+\frac{2g_1{}^2}{D_{-n-1}{}^*}+\frac{2g_2{}^2}{D_n},$$
$$b_n=2g_1g_2\left[\frac{1}{D_n}+\frac{1}{D_{-n-1}{}^*}\right],$$
$$c_n=2g_1g_2\left[\frac{1}{D_{n-1}}+\frac{1}{D_{-n}{}^*}\right]. \quad (2.16)$$

The recursion relation [Eq. (2.15)] for $X_n$ can be solved in terms of the continued fraction, with the result that

$$X_0=\eta[a_0+2\,\mathrm{Re}\,(b_0Y_1)]^{-1}, \quad (2.17)$$
$$Y_n=-c_n\,[a_n+b_nY_{n+1}]^{-1}, \quad Y_n\equiv X_n/X_{n-1}, \quad n\neq 0. \quad (2.18)$$

The system of equations is thus completely solved once the solution of Eqs. (2.17) and (2.18) is known. The off-diagonal elements of the density matrix are then obtained from Eqs. (2.12) and (2.13).

The rate of absorption of the energy $W_i$ from the $i$th field is obtained by using

$$W_1\cong 2\omega_1 g_1\,\mathrm{Im}\,\langle\psi_1{}^{[-1]}\rangle, \quad (2.19)$$
$$W_2\cong 2\omega_2 g_2\,\mathrm{Im}\,\langle\psi_1{}^{[0]}\rangle. \quad (2.20)$$

We close this section by commenting on the case $\Phi=0$, i.e., the case when both the lasers are completely correlated. In such a case Eq. (2.6) can be directly averaged as

$$\langle\dot{\psi}\rangle=A\langle\psi\rangle-\Gamma_2 f^2\langle\psi\rangle+I + B_+\exp(i\Omega t)\,\langle\psi\rangle+B_-\exp(-i\Omega t)\,\langle\psi\rangle, \quad (2.21)$$

whose steady-state solution is

$$\langle\psi\rangle = \Sigma\langle\psi^{[n]}\rangle \exp(-in\Omega t), \tag{2.22}$$

with

$$A\langle\psi^{[n]}\rangle + B_+\langle\psi^{[n+1]}\rangle + B_-\langle\psi^{[n-1]}\rangle + \delta_{n0} I + (in\Omega - \Gamma_2 f^2)\langle\psi^{[n]}\rangle = 0. \tag{2.23}$$

The set [Eq. (2.23)] is the same as Eq. (2.11) with $\Gamma_1 = \Gamma_2 = \Gamma_{12}$.

## 3. EFFECT OF PUMP TEMPORAL FLUCTUATIONS ON MULTIPHOTON PEAKS IN THE ABSORPTION SPECTRA

We now use the dynamical equations of Section 2 to study how the temporal fluctuations of the two pump fields affect the multiphoton absorption peaks. It is clear that $\langle\psi_1^{[-1]}\rangle$ depends on the intensities of the two fields to all orders. $\langle\psi_1^{[-1]}\rangle$ is expected to have the structure

$$\langle\psi_1^{[-1]}\rangle = \sum_{n=0}^{\infty} g_1^{2n+1}\,\alpha_{2n+1}, \tag{3.1}$$

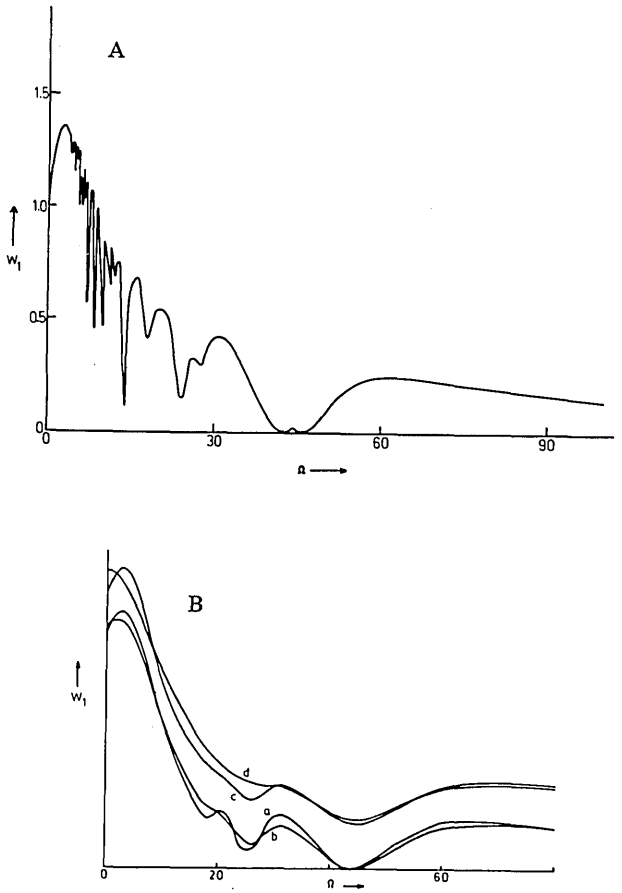


Fig. 1. A, rate of absorption $W_1$ [$\propto \langle\psi_1^{(-1)}\rangle$] as a function of $\Omega$ for $g_1 = 30$, $g_2 = 20$, $\Delta = 0$, $\Gamma_1 = \Gamma_2 = 0$, and $T_1 = T_2 = 1$. B, $W_1$ for various values of $\Gamma_1$ and $\Gamma_2$ as a function of $\Omega$ for $g_1 = 30$, $g_2 = 20$, $\Delta = 0$, and $T_1 = T_2 = 1$. Curve a, $\Gamma_1 = 0.4$, $\Gamma_2 = 0.0$. Curve b, $\Gamma_1 = \Gamma_2 = 0.4$. Curve c, $\Gamma_1 = 0.0$, $\Gamma_2 = 0.8$. Curve d, $\Gamma_1 = \Gamma_2 = 0.8$. Note that the spectrum for $\Gamma_1 = 0.0$ and $\Gamma_2 = 0.4$ is similar to that for curve a. For convenience of illustration, curves c and d have been shifted.

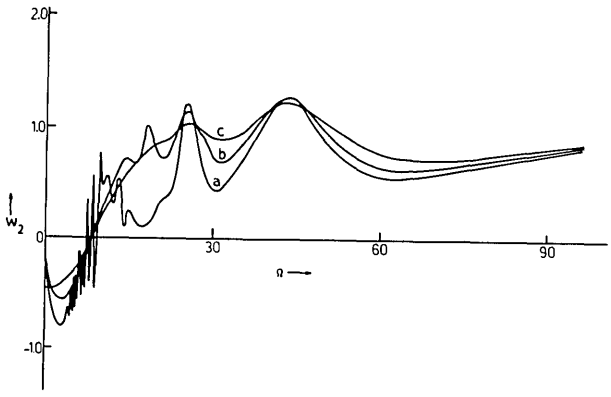


Fig. 2. Absorption spectrum $W_2$ [$\propto \langle\psi_1^{(0)}\rangle$] as a function of $\Omega$ for $g_1 = 30$, $g_2 = 20$, $\Delta = 0$, and $T_1 = T_2 = 1$. Curve a, $\Gamma_1 = \Gamma_2 = 0.0$. Curve b, $\Gamma_1 = 0.2$, $\Gamma_2 = 0.0$. Curve c, $\Gamma_1 = 0.8$, $\Gamma_2 = 0.0$.

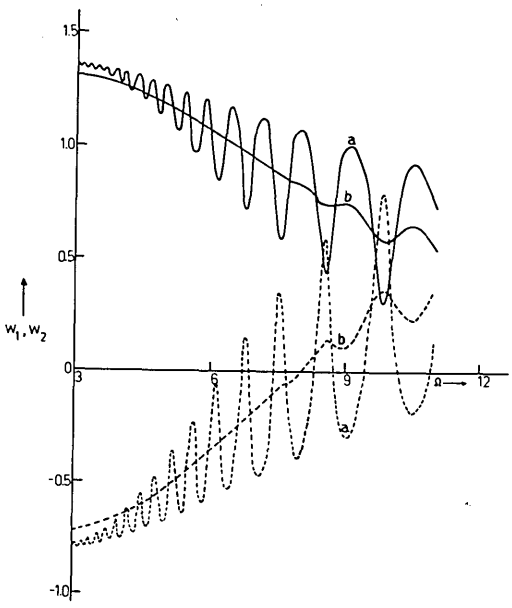


Fig. 3. Multiphoton absorption region of the spectrum $W_1$ (solid line) and $W_2$ (dashed line) for $g_1 = 30$, $g_2 = 20$, $\Delta = 0$, and $T_1 = T_2 = 1$. Curve a, $\Gamma_1 = \Gamma_2 = 0.0$. Curve b, $\Gamma_1 = 0.05$, $\Gamma_2 = 0.0$.

where $\alpha$'s depend on $g_2^2$ to all orders. $\alpha_1$ represents the absorption of a single photon from the beam at $\omega_1$ in the presence of another field of arbitrary strength and this is the quantity measured in the experiments of Wu *et al.*[3] The field $\epsilon_2$ can be imagined to dress the eigenstates of the atom. Thus $\alpha_{2n+1}$ will represent the absorption of $(n+1)$ photons of frequency $\omega_1$ by the atom dressed by the field $\epsilon_2$. The effect of the temporary fluctuations on $\alpha_1$ is well known both experimentally and theoretically.

In the case when both the pumps are completely correlated, $\Gamma_1 = \Gamma_2 = \Gamma_{12} = \gamma$, then the multiphoton absorption spectra in the presence of fluctuations can be obtained from those in the absence of fluctuations by using the simple substitution rule[16]

$$\frac{1}{T_2} \rightarrow \frac{1}{T_2} + \Gamma. \tag{3.2}$$

The general case is extremely involved, and it does not appear to be possible to obtain a simple result even for the line center $\omega_1 = \omega_2 = \omega_0$. Hence we have numerically investigated the

solution of Eq. (2.18), and the results are shown in a number of figures. The parameters (all in frequency units) used in our numerical computations are based on the ones used in the experimental investigations of the Russian workers.[8,11] We also assume that the two fields are uncorrelated, $\Gamma_{12} = 0$. Figure 1 gives the behavior of the absorption spectra for the absorption of the energy from field 1 for $g_1 > g_2$ and when both the fields are strong. The figure shows that the multiphoton absorption peaks are wiped out with increase in $\Gamma_1$. The effect of $\Gamma_2$ is similar. Only the peak corresponding to the frequency $2(g_1^2 + g_2^2)^{1/2}$ and the first few multiphoton peaks survive. The behavior of the energy absorption from field 2 is similar, as is shown in Fig. 2. Figure 3 shows in detail the effect of the laser fluctuations on the multiphoton absorption peaks. In order to see these absorption peaks, the laser bandwidths must not be more than few percent of the natural line width. Figure 4 is for much larger values of the bandwidths, whence only the structure at the line center survives. The case when $g_1 < g_2$ is illustrated in Fig. 5.

The extreme sensitivity of the multiphoton absorption peaks on the temporal fluctuations of pump beams can be understood from the structure of the coefficients $D_n$ [defined by Eq. (21.4)]. The position and the width of the multiphoton absorption peaks are approximately determined from the zeros of $a_n$. For large $n$ and in the absence of any temporal fluctuations, these are extremely narrow—for $T_1 = T_2$ these are located at $\pm(g_1^2 + g_2^2)^{1/2}/n$ with widths $\sim(1/nT_2)$. Note that the laser temporal fluctuations (bandwidths) enter as $n^2\Gamma$ in the coefficients $D_n$ and $a_n$. Hence these multiphoton absorption peaks acquire an additional width of the order of $n\Gamma$. Since the relative separation of such peaks is of the order of $gT_2/n$, it is clear that a small fluctuation will wash away such multiphoton structures.

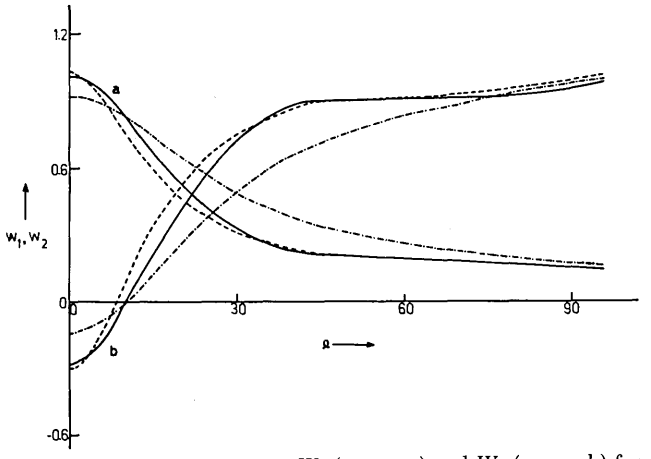


Fig. 4. Absorption spectrum $W_1$ (curves a) and $W_2$ (curves b) for high values of $\Gamma_1$ and $\Gamma_2$. $g_1 = 30$, $g_2 = 20$, $\Delta = 0$, and $T_1 = T_2 = 1$. (Solid line, $\Gamma_1 = 10.0$, $\Gamma_2 = 0.0$; dashed line, $\Gamma_1 = 0.0$, $\Gamma_2 = 10.0$; and dashed-dotted line, $\Gamma_1 = \Gamma_2 = 10.0$.)

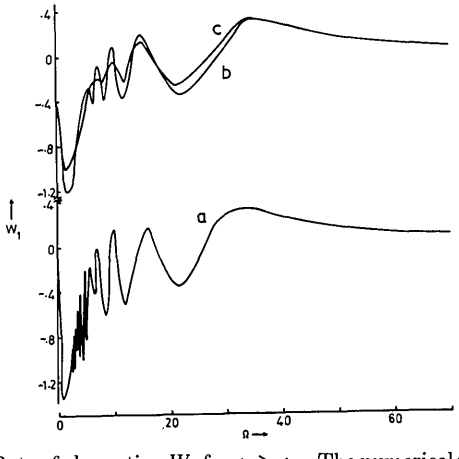


Fig. 5. Rate of absorption $W_1$ for $g_2 > g_1$. The numerical values are $g_1 = 9.52$, $g_2 = 11.9$, $\Delta = 0$, and $T_1 = T_2 = 1$. The various values for $\Gamma_1$ and $\Gamma_2$ are curve a, $\Gamma_1 = \Gamma_2 = 0.0$; curve b, $\Gamma_1 = 0.0$, $\Gamma_2 = 0.05$; curve c, $\Gamma_1 = 0.0$, $\Gamma_2 = 0.2$.

## 4. EFFECT OF SATURATION ON THE NONLINEAR SUSCEPTIBILITY $\chi^{(3)}(-2\omega_2 + \omega_1, \omega_2, \omega_2, -\omega_1)$

As was mentioned in the introduction, $\chi^{(3)}$ yields the four-wave mixing signal in the direction $2\mathbf{k}_2 - \mathbf{k}_1$. The theory described in Section 2 is fairly general and should also enable us to calculate $\chi^{(3)}$. We show that $\chi^{(3)}$ has an interesting saturation behavior. In order to keep the analysis simple, we ignore the complications that arise because of finite temporal fluctuations[17] of the fields. From Eqs. (2.22) and (2.5) it is clear that

$$\rho_{12}(t) = \sum_n \psi_2^{[n]} \exp\{-it[\omega_2(1-n) + n\omega_1]\} \tag{4.1}$$

$$= \psi_2^{[-1]} \exp[-it(2\omega_2 - \omega_1)] + \text{other terms}. \tag{4.2}$$

The four-wave mixing signal is related to the induced polarization at $2\omega_2 - \omega_1$. The induced polarization is proportional to $\rho_{12}$, and thus the component responsible for four-wave mixing is $\psi_2^{[-1]} = \psi_1^{[1]*}$. Note that, in the limit of weak fields (Rabi frequencies $\ll$ detunings), the induced polarization at $2\omega_2 - \omega_1$ is known[13] to be proportional to $\epsilon_2^2\epsilon_1^*$, and hence $|\psi_1^{[1]}|^2 \propto |\epsilon_2|^4|\epsilon_1|^2$. In general, $\psi_1^{[1]}$ will depend on all powers of $\epsilon$. The component $\langle\psi_1^{[1]}\rangle$ is given by Eq. (2.12) with $n = 1$, which when Eq. (2.18) is used, becomes

$$\langle\psi_1^{[1]}\rangle = 2i(g_2 + g_1Y_2)Y_1X_0D_1^{-1}. \tag{4.3}$$

Thus the continued fraction of Section 2 will give the four-wave mixing signal if Eq. (4.3) is used. The continued fraction can be summed up exactly for $\omega_1 = \omega_2$ leading to

$$Y_1 = -\frac{a}{2b}\left[1 - \left(1 - \frac{4cb}{a^2}\right)^{1/2}\right], \tag{4.4}$$

where

$$a = \frac{1}{T_1} + \frac{4(g_1^2 + g_2^2)}{T_2\left[\Delta_2^2 + \left(\frac{1}{T_2}\right)^2\right]}, \quad b = c = \frac{4g_1g_2}{T_2\left[\left(\frac{1}{T_2}\right)^2 + \Delta_2^2\right]}. \tag{4.5}$$

Bloembergen *et al.*[15] have shown that $S$, in the limit of weak fields, has an interesting resonance at $\omega_1 = \omega_2$ with a width $T_1^{-1}$ provided that the system has collisions; otherwise, for the case of radiative relaxation, $S$ has no structure. This is illustrated in Fig. 6, curve a. We also show in Fig. 6 (curves b and c) the effect of the saturation, which leads to the peak at $\omega_1 = \omega_2$, even in the case of the radiative relaxation,[18] in

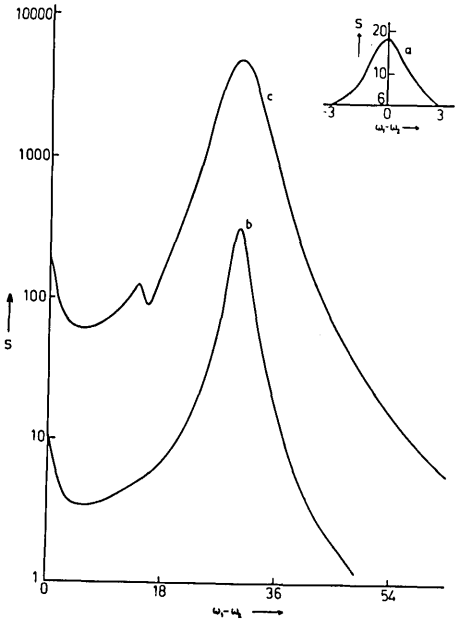


Fig. 6. Four-wave mixing signal $S(\alpha|\psi_1^{[1]}|^2)$ as a function of $(\omega_1 - \omega_2)$ for curve a, inset: $\Delta = 200$, $T_1 = T_2 = 1$, $g_1 = 4$, and $g_2 = 15$ and for $T_1 = 0.5$, $T_2 = 1.0$, $\Delta = 0$; curve b, $g_1 = 1$, $g_2 = 15$; curve c, $g_1 = 4$, $g_2 = 15$.

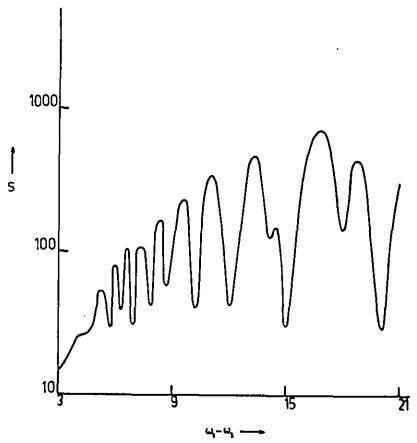


Fig. 7. Multiphoton absorption region of $S$ as a function of $(\omega_1 - \omega_2)$ for $g_1 = 30$, $g_2 = 20$, $T_1 = T_2 = 1$, and $\Delta = 0$.

addition to the multiphoton (saturation) peaks.[19] The effect of collisions is not dramatic if the saturation effects are important. Note that, as the field intensities are increased further, more and more multiphoton absorption peaks start appearing as shown in Fig. 7.

## ACKNOWLEDGMENT

This research has been supported in part by a grant from the Department of Science and Technology, Government of India.

## REFERENCES

1. B. R. Mollow, Phys. Rev. **188**, 1969–1975 (1969); R. P. Hackel and S. Ezekiel, in *Laser Spectroscopy IV*, H. Walther and K. W. Rothe, eds. (Springer-Verlag, Berlin, 1979), p. 88.
2. B. R. Mollow, Phys. Rev. A**5**, 2217–2222 (1972).
3. F. Y. Wu, S. Ezekiel, M. Ducloy, and B. R. Mollow, Phys. Rev. Lett. **38**, 1077–1080 (1977).
4. A. M. Bonch-Bruevich, S. G. Przhibelskii, V. A. Khodovoi, and N. A. Chigir, Sov. Phys. JETP **43**, 230–236 (1976).
5. R. G. Gush and H. P. Gush, Phys. Rev. A**10**, 1474–1487 (1974).
6. N. Tsukada and T. Nakayama, Phys. Rev. A**25**, 964–997 (1982).
7. A. M. Bonch-Bruevich, S. G. Przhibelskii, and N. A. Chigir, Sov. Phys. JETP **53**, 285–291 (1981).
8. A. M. Bonch-Bruevich, T. A. Vartanyan, and N. A. Chigir, Sov. Phys. JETP **50**, 901–906 (1979).
9. G. S. Agarwal, Phys. Rev. A**18**, 1490–1506 (1978).
10. G. S. Agarwal, Z. Physik B**33**, 111 (1979).
11. G. I. Toptygina and E. E. Fradkin, Sov. Phys. JETP **55**, 246–251 (1982).
12. Note that, since both the fields can be strong, there is really no distinction between probe and pump.
13. N. Bloembergen, in *Laser Spectroscopy*, H. Walther and K. W. Rothe, eds. (Springer-Verlag, Berlin, 1979), p. 340.
14. For example, the saturation effects in the susceptibility $\psi^{(5)}(-3\omega_2 + 2\omega_1, \omega_2, \omega_2, \omega_2, -\omega_1, -\omega_1)$, which describes higher-order coherent anti-Stokes Raman Spectroscopy [A. Compaan, E. Wiener-Avnear, and S. Chandra, Phys. Rev. A**17**, 1083 (1978)] can be studied.
15. N. Bloembergen, A. R. Bogdan, and M. W. Downer, in *Laser Spectroscopy V*, A. R. W. McKellar, T. Oka, and B. P. Stoicheff, eds. (Springer-Verlag, Berlin, 1981), p. 157.
16. Substitution rules in the context of optical resonance have been emphasized by J. H. Eberly, in *Laser Spectroscopy IV*, H. Walther and K. W. Rothe, eds. (Springer-Verlag, Berlin, 1979), p. 80.
17. Complications that arise because of the temporal fluctuations are discussed in G. S. Agarwal and C. V. Kunasz, Phys. Rev. A**27**, 996–1012 (1983).
18. In the framework of higher-order perturbation theory, H. Friedmann and A. D. Wilson-Gordon [Phys. Rev. A**26**, 2768–2777 (1982)] have also found the presence of the extra resonance, even in the case of radiative relaxation.
19. The values at the peak $\omega_1 = \omega_2$ calculated from the analytic formulas (4.4) and (4.5) are in agreement with the numerical computations.