Effects of cross relaxation and line mixing on third-order nonlinearities of resonant systems

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The effect of cross relaxation, i.e., the transfer of coherence between transitions coupled by inelastic collisions, on third-order nonlinear processes, namely, saturated absorption and four-wave mixing, is studied within the context of a model four-level system. Numerical results displaying changes in the resonant structures in four-wave-mixing signals for a range of collisional parameters are presented.

I. INTRODUCTION

There has been a lot of interest in the study of the nonlinear response of an atomic system to radiation in the presence of collisions. Collisions have been known to influence the atomic response in various ways. For instance, both elastic as well as inelastic collisions have been found to give rise to redistribution of radiation[1] emitted by an atomic system. These also give rise to extra resonances in the nonlinear susceptibilities[2,3] and fluorescence[4] produced by multilevel systems. In such studies, it is usually assumed that the function of inelastic collisions is merely to bring about a transfer of populations among the various energy levels of the system, and a decay of coherence induced between the levels. A point that is generally not taken into account is that there can be an interference[5] between two collision-induced transitions, which becomes significant when the rate of such transitions is appreciable compared to the frequency separation between those transitions. Such interferences give rise to couplings between different coherences or between coherences and populations in the equations of motion for the density operator of the system. A simple case of such couplings, namely, the coupling that gives rise to a transfer of coherence (or cross relaxation) between two pairs of levels, has been discussed in some detail in the literature.[6–8] Cross relaxation has been found to give rise to interesting effects in the linear response of a system to an external field, e.g., the collapse of the inversion spectrum of NH_3 at high pressures,[6] and the mixing of the two lines in the absorption spectrum, their merger into a single line and further, the narrowing of this line, with increasing pressure.[7] However, the question of how cross relaxation influences the nonlinear response of a system to external fields has not received enough consideration[7,9] so far. In this paper, we examine the effect of cross relaxation on third-order nonlinear processes.

In Sec. II we trace the origin of cross relaxation to certain nonsecular (or counter-rotating) terms in the master equation[10] describing the interaction of an atomic system with the bath of perturbers. In Sec. III we specialize the general formulation of Sec. II to a model four-level system with two optical transitions. We study the effect of cross relaxation on saturated absorption. In Sec. IV we calculate the nonlinear susceptibility for four-wave mixing in the model system of Sec. III. We present detailed numerical results for a range of collisional parameters. Our discussion includes the various resonances and, in special cases, our results reduce to those obtained by other methods.

II. ATOMIC DYNAMICS WITH CROSS RELAXATION

The relaxation behavior of an atomic system in the presence of collisions can be handled by using various methods. For example, one can consider the interaction of an atom with a heat bath and write the interaction in the form

$$H=\sum_m E_m \,|\, m \rangle\langle m \,| + \sum_{k,l} v_{kl} \,|\, k \rangle\langle l \,| + H_R \;, \tag{2.1}$$

where v describes the interaction of the atom with the heat bath or with the perturbers and H_R is the unperturbed Hamiltonian of the heat bath. Note that v_{kl} is an operator in the Hilbert space of the perturbers. Let ρ be the reduced density matrix for the multilevel atom of interest. In the weak-coupling limit and in the Markov (impact) approximation, the density matrix ρ^I in the interaction picture is found to obey the equation[10]

$$\dot{\rho}^I = \sum_{k,l,m,n} [(A_{mn}\rho^I A_{kl} - A_{kn}\rho^I \delta_{lm})\gamma^+_{klmn} + (A_{kl}\rho^I A_{mn} - \rho^I A_{ml}\delta_{nk})\gamma^-_{mnkl}] \times e^{i(\omega_{kl}+\omega_{mn})t} \;, \tag{2.2}$$

where

$$A_{kl} = |\, k \rangle\langle l \,| \;, \quad \omega_{kl} = E_k - E_l \tag{2.3}$$

and the coefficients $\gamma^\pm$ are related to the correlation functions of the perturber bath

$$\gamma^+_{klmn} = \int_0^\infty d\tau \langle v_{kl}(\tau) v_{mn}(0) \rangle e^{-i\omega_{mn}\tau} \;, \tag{2.4}$$

$$\gamma^-_{klmn} = \int_0^\infty d\tau \langle v_{mn}(0) v_{kl}(\tau) \rangle e^{-i\omega_{mn}\tau} = (\gamma^+_{lknm})^* \;. \tag{2.5}$$

At this point one usually makes the rotating-wave approximation and keeps only those terms in Eq. (2.2) for which $\omega_{kl}+\omega_{mn}$ is exactly equal to zero. Then, in the case when the atom has a nonequidistant and nondegenerate spectrum, one gets the well-known equation which has been extensively used in finding the nonlinear response of a driven system,

$$\dot{\rho}^I_{ij} = -\Gamma_{ij}(1-\delta_{ij})\rho^I_{ij} + \delta_{ij}\sum_{\substack{i,k\\(k\neq i)}} (\gamma_{ik}\rho^I_{kk} - \gamma_{ki}\rho^I_{ii}) , \tag{2.6}$$

where

$$\Gamma_{ij} = \tfrac{1}{2}\sum_{\substack{i,k\\(k\neq i)}} (\gamma_{ki}+\gamma_{kj}) + \Gamma^{\text{ph}}_{ij} . \tag{2.7}$$

Here γ_{ij} is the rate of transition from $|j\rangle$ to $|i\rangle$ due to spontaneous emission as well as due to inelastic collisions, and Γ^{ph}_{ij} is the rate of dephasing due to elastic collisions. However, the counter-rotating terms in Eq. (2.2) can be important in certain cases as discussed below. When counter-rotating terms are retained, then Eq. (2.2) can be written in the Schrödinger picture as

$$\dot{\rho}_{ij} = -i\Lambda_{ij}\rho_{ij} + \sum_{n,k}\xi_{kjin}\rho_{nk} - \sum_k (\eta^+_{ik}\rho_{kj} + \eta^-_{kj}\rho_{ik}), (i\neq j) , \tag{2.8}$$

$$\dot{\rho}_{ii} = \sum_{\substack{k,i\\(k\neq i)}} (\gamma_{ik}\rho_{kk} - \gamma_{ki}\rho_{ii}) + \sum_{n,k}(1-\delta_{nk})\xi_{kiin}\rho_{nk} - \sum_k (\eta^+_{ik}\rho_{ki} + \eta^-_{ki}\rho_{ik}) , \tag{2.9}$$

where

$$\xi_{kjin} = \gamma^+_{kjin} + \gamma^-_{kjin} ,$$
$$\eta^{+(-)}_{ij} = (1-\delta_{ij})\sum_k \gamma^{+(-)}_{ikkj} , \tag{2.10}$$
$$\Lambda_{ij} = \omega_{ij} - i\Gamma_{ij} .$$

In Eqs. (2.8) and (2.9) the ξ and η terms are the counter-rotating terms that bring about coherence-coherence and coherence-population coupling. Let us now look at the origin and meaning of such additional terms. Expressing Eq. (2.4) in a representation in which the bath Hamiltonian is diagonal, i.e., $H_R\,|R\rangle = R\,|R\rangle$, we get[11]

$$\gamma^+_{klmn} = \pi\sum_R f_0(R)\langle R,k\,|V|\,R-\omega_{mn},l\rangle \times \langle R,n\,|V|\,R-\omega_{mn},m\rangle^* , \tag{2.11}$$

and we also have

$$\gamma_{lk} = \gamma^+_{kllk} + \gamma^-_{kllk} = 2\pi\sum_R f_0(R)\,|\langle R,k\,|V|\,R-\omega_{lk},l\rangle|^2 , \tag{2.12}$$

where $f_0(R)$ is the probability distribution of the bath states. One can see from Eqs. (2.11) and (2.12), that while γ^+_{klmn} is related to a product of two transition amplitudes of the form $c_{lk}c^*_{mn}$ (where c_{ij} denotes the probability amplitude that a transition takes place from $|j\rangle$ to $|i\rangle$), γ_{lk} is related to the modulus squared of the transition amplitude of the form $|c_{lk}|^2$. Thus the terms γ^+_{klmn} can be seen to arise from a quantum-mechanical interference between the two transitions $|k\rangle\to|l\rangle$ and $|n\rangle\to|m\rangle$. One can take the magnitude of $\gamma^{+(-)}_{klmn}$ to be roughly equal to $\frac{1}{2}\sqrt{\gamma_{lk}\gamma_{mn}}$. By doing a simple first-order perturbation theory with any particular counter-rotating term as the perturbation parameter, one can estimate the order of its contribution to $\rho(t)$. For instance, the contribution from the term ξ_{kjin} that couples the coherences ρ_{ij} and ρ_{nk} can be seen to be important when $\xi_{kjin}(\sim\sqrt{\gamma_{jk}\gamma_{in}})$ is appreciable compared to $|\Lambda_{ij}-\Lambda_{nk}|$. There are many systems for which some of the nonsecular couplings become important (at high pressures) when the inelastic collision rates become sufficiently large. In Sec. II we study a model system where a simple coherence-coherence coupling between two pairs of levels is important. Such a model system has been of considerable interest. Other systems can also be studied similarly.

III. EFFECT OF CROSS RELAXATION ON THIRD-ORDER NONLINEAR SUSCEPTIBILITIES

Consider a frequently studied[5,7,8] four-level system consisting of two transitions $|2\rangle\leftrightarrow|3\rangle$ and $|1\rangle\leftrightarrow|4\rangle$ coupled by inelastic collisions (see Fig. 1) where a central frequency ω_0 is defined so that $\omega_1=\omega_0-\delta$ and $\omega_2=\omega_0+\delta$. The time evolution of an atomic system in the presence of external fields is described by the equation

$$\dot{\rho} = -i[H',\rho] + L_R\rho , \tag{3.1}$$

where $H'(t)$ is the atom-field interaction in the dipole approximation, and $L_R\rho$ is given by Eqs. (2.8) and (2.9). For the above system, Eq. (3.1) leads explicitly to the system of equations

$$\begin{aligned}
\dot{\rho}_{14} &= -[i(\omega_0+\delta)+\gamma+\sigma]\rho_{14} + \xi\rho_{23} - iH'_{14}(\rho_{44}-\rho_{11}) ,\\
\dot{\rho}_{23} &= -[i(\omega_0-\delta)+\gamma+\sigma]\rho_{23} + \xi\rho_{14} - iH'_{23}(\rho_{33}-\rho_{22}) ,\\
\dot{\rho}_{11} &= -(\sigma+2\gamma)\rho_{11} + \sigma\rho_{22} - i(H'_{14}\rho_{41}-\rho_{14}H'_{41}) ,\\
\dot{\rho}_{22} &= \sigma\rho_{11} - (\sigma+2\gamma)\rho_{22} - i(H'_{23}\rho_{32}-\rho_{23}H'_{32}) ,\\
\dot{\rho}_{33} &= 2\gamma\rho_{22} - \sigma\rho_{33} + \sigma\rho_{44} + i(H'_{23}\rho_{32}-\rho_{23}H'_{32}) ,\\
\dot{\rho}_{44} &= 2\gamma\rho_{11} + \sigma\rho_{33} - \sigma\rho_{44} + i(H'_{14}\rho_{41}-\rho_{14}H'_{41}) ,
\end{aligned} \tag{3.2}$$

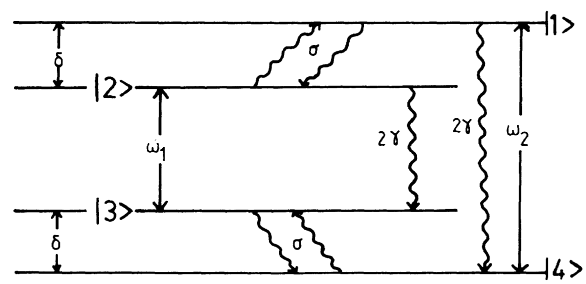

FIG. 1. Energy diagram of the model system with various relaxation rates. 2γ is the radiative relaxation rate of each transition and σ is the strength of collisional coupling between the two components.

where we have taken $\xi_{3412}=\xi_{4321}=\xi_{1234}=\xi_{2143}=\xi$ (all other ξ and η terms are negligible), $\Gamma_{14}=\Gamma_{23}=\gamma+\sigma$, $\Gamma^{\mathrm{ph}}=0$. Here σ is thus the rate of the inelastic collisions between two nearby levels as shown in Fig. 1. The parameter ξ couples coherences to coherences. These parameters ξ and σ can be obtained in terms of the matrix elements of V [see Eq. (2.11)]. These can also be related[5] to the collision matrix S, for example, $\xi=\int (S^+)_{12}(S)_{34}F(g)dg$, where $F(g)$ can, for example, be the distribution of impact parameters g. Let the system be interacting with two monochromatic fields with frequencies ω_s and ω_p. We write the total field as

$$\mathbf{E}(t)=\hat{e}(e_s e^{-i\omega_s t}+e_p e^{-i\omega_p t}+\text{c.c.}) . \tag{3.3}$$

The macroscopic polarization induced in the system is given by

$$\langle \mathbf{P}(t)\rangle = N\,\mathrm{Tr}[\rho(t)\mathbf{p}] , \tag{3.4}$$

where N is the atomic number density, $\mathbf{p}$ is the dipole moment operator, and $\rho(t)$ is determined from Eqs. (3.2) with

$$\begin{aligned} H'(t)&=-\mathbf{p}\cdot\mathbf{E}(t)\\ &=-d[(\,|1\rangle\langle 4|+|2\rangle\langle 3|\,)\\ &\quad\times(e_s e^{-i\omega_s t}+e_p e^{-i\omega_p t})+\text{H.c.}] \end{aligned} \tag{3.5}$$

in the rotating-wave approximation. It should be borne in mind that the cross relaxation in Eqs. (3.2) is represented by the terms involving ξ. We will now obtain the results for the nonlinear response of a system when cross relaxation is important.

A. Linear response

To first order in the probe field e_s (in the absence of any pump) one has

$$P^{(1)}(t)=\chi^{(1)}(\omega_s)e_s e^{-i\omega_s t}+\text{c.c.} \tag{3.6}$$

The linear absorption is related to the imaginary part of $\chi^{(1)}$ by the relation

$$I_1(\omega_s)=2\omega_s\,|e_s|^2\mathrm{Im}\chi^{(1)}(\omega_s) . \tag{3.7}$$

Using the system of equations (3.2) we have shown that

$$\chi^{(1)}(\omega_s)=-\frac{Nd^2}{2}\left[\frac{A_+}{\Delta_s-i\lambda_+}+\frac{A_-}{\Delta_s-i\lambda_-}\right] , \tag{3.8}$$

where

$$\begin{aligned} &\Delta_s=\omega_s-\omega_0 ,\\ &\lambda_\pm=-(\gamma+\sigma)\pm i(\delta^2-\xi^2)^{1/2} ,\\ &A_\pm=1\pm\xi/(\xi^2-\delta^2)^{1/2} . \end{aligned} \tag{3.9}$$

The behavior of $\chi^{(1)}$ depends on the magnitude of cross relaxation. For $\xi/\delta<1$, one has

$$\begin{aligned} \mathrm{Im}\chi^{(1)}(\omega_s)\propto &\frac{\gamma+\sigma+\alpha[\Delta_s+(\delta^2-\xi^2)^{1/2}]}{[\Delta_s+(\delta^2-\xi^2)^{1/2}]^2+(\gamma+\sigma)^2}\\ &+\frac{\gamma+\sigma-\alpha[\Delta_s-(\delta^2-\xi^2)^{1/2}]}{[\Delta_s-(\delta^2-\xi^2)^{1/2}]^2+(\gamma+\sigma)^2} ,\\ \alpha=\xi/(\delta^2-\xi^2)^{1/2} . \end{aligned} \tag{3.10}$$

One can see from Eq. (3.10) that for $\xi/\delta<1$, the linear-absorption spectrum consists of two lines exhibiting both absorptive and dispersive character. These lines get mixed more and more as σ increases (static mixing) and as ξ increases (dynamic mixing). In the limit $\xi/\delta\to 0$, Eq. (3.10) reduces to a sum of two Lorentzians,

$$\begin{aligned} \mathrm{Im}\chi^{(1)}(\omega_s)\propto &\frac{\gamma+\sigma}{(\Delta_s+\delta)^2+(\gamma+\sigma)^2}\\ &+\frac{\gamma+\sigma}{(\Delta_s-\delta)^2+(\gamma+\sigma)^2} . \end{aligned} \tag{3.11}$$

As $\xi/\delta=1$ one has a single line peaked at $\Delta_s=0$ and with width $\gamma+\sigma$. In general, the first-order spectrum is symmetrical around $\Delta_s=0$. For $\xi/\sigma>1$, one has

$$\mathrm{Im}\chi^{(1)}(\omega_s)\propto\left[\frac{A_+\lambda_+}{\Delta_s^2+\lambda_-^2}+\frac{A_+\lambda_-}{\Delta_s^2+\lambda_-^2}\right] , \tag{3.12}$$

where

$$\begin{aligned} &\lambda_\pm=-(\gamma+\sigma)\pm(\xi^2-\delta^2)^{1/2} ,\\ &A_\pm=1\pm\xi/(\xi^2-\delta^2)^{1/2} . \end{aligned} \tag{3.13}$$

Thus, as ξ/δ increases beyond 1, one of the two lines becomes narrower and stronger while the other line becomes broader and weaker and what survives is the narrower line. For $\xi/\delta\sim\sigma/\delta\gg 1$, one has

$$\begin{aligned} &\lambda_+\sim-\gamma-\delta^2/2\sigma,\quad A_+\sim 2+\delta^2/2\sigma ,\\ &\lambda_-\sim-(\gamma+2\sigma)+\delta^2/2\sigma,\quad A_-\sim-\delta^2/2\sigma . \end{aligned} \tag{3.14}$$

Hence, at very high pressures, one observes a single narrow line with width approaching the natural linewidth. This is the well-known line-narrowing phenomena[5-8] resulting from the mixing of the lines.

B. Susceptibility for saturated absorption with cross relaxation

We next consider absorption from a probe in the presence of a pump field. The absorption spectrum in such a case is given by

$$I_3(\omega_s)=2\omega_s\,|e_s|^2\mathrm{Im}[\chi^{(1)}(\omega_s)+|e_p|^2\chi^{(3)}(\omega_p,-\omega_p,\omega_s)] , \tag{3.15}$$

where $\chi^{(1)}(\omega_s)$ is as given by Eq. (3.8). We have proved using Eqs. (3.2) that the third-order contribution to Eq. (3.15) is given by

$$\chi^{(3)}(\omega_p,-\omega_p,\omega_s)$$
$$=-\frac{Nd^4}{2}\left\{\frac{1}{2i\gamma+\Delta_s-\Delta_p}\left[\frac{A_+}{\Delta_s-i\lambda_+}+\frac{A_-}{\Delta_s-i\lambda_-}\right]\left[\left[\frac{A_+}{\Delta_s-i\lambda_+}+\frac{A_-}{\Delta_s-i\lambda_-}\right]-\left[\frac{A_+}{\Delta_p+i\lambda_+}+\frac{A_-}{\Delta_p+i\lambda_-}\right]\right]\right.$$
$$+\frac{1}{2i\gamma}\left[\frac{A_+}{\Delta_s-i\lambda_+}+\frac{A_-}{\Delta_s-i\lambda_-}\right]\left[\left[\frac{A_+}{\Delta_p-i\lambda_+}+\frac{A_-}{\Delta_p-i\lambda_-}\right]-\left[\frac{A_+}{\Delta_p+i\lambda_+}+\frac{A_-}{\Delta_p+i\lambda_-}\right]\right]$$
$$+\frac{A_+A_-}{2i(\gamma+\sigma)+\Delta_s-\Delta_p}\left[\frac{1}{\Delta_s-i\lambda_+}-\frac{1}{\Delta_s-i\lambda_-}\right]\left[\frac{1}{\Delta_s-i\lambda_+}-\frac{1}{\Delta_s-i\lambda_-}+\frac{1}{\Delta_p+i\lambda_+}-\frac{1}{\Delta_p+i\lambda_-}\right]$$
$$\left.+\frac{A_+A_-}{2i(\gamma+\sigma)}\left[\frac{1}{\Delta_s-i\lambda_+}-\frac{1}{\Delta_s-i\lambda_-}\right]\left[\frac{1}{\Delta_p-i\lambda_+}-\frac{1}{\Delta_p-i\lambda_-}+\frac{1}{\Delta_p+i\lambda_+}-\frac{1}{\Delta_p+i\lambda_-}\right]\right\}, \quad (3.16)$$

where Δ_s, $A_\pm$, and $\lambda_\pm$ are defined as before [Eq. (3.9)] and $\Delta_p=\omega_p-\omega_0$. It is found that $\text{Im}\chi^{(3)}(\omega_p,-\omega_p,\omega_s)$ can be negative. This can be seen by considering a special case of Eq. (3.16) with $\xi/\delta=\sigma/\delta=0$. In this limit, we have

$$\chi^{(3)}(\omega_p,-\omega_p,\omega_s)=-\frac{Nd^4}{2}\left[\frac{-2}{(\Delta_s+\delta+i\gamma)(\Delta_p+\delta-i\gamma)}\left[\frac{1}{\Delta_s+\delta+i\gamma}+\frac{1}{\Delta_p+\delta+i\gamma}\right]+(\delta\to-\delta)\right], \quad (3.17)$$

(where $\delta\to-\delta$ represents the same terms with δ interchanged with $-\delta$) which is just the sum of $\chi^{(3)^s}$ for two independent two-level systems with resonance frequencies $\omega_0-\delta$ and $\omega_0+\delta$. Equation (3.17) leads to

$$\text{Im}\chi^{(3)}(\omega_p,-\omega_p,\omega_s)=-\frac{Nd^4}{2}\left[\frac{4\gamma(\gamma^2+\Delta_{1p}\Delta_{1s})}{(\Delta_{1s}^2+\gamma^2)(\Delta_{1p}^2+\gamma^2)}\right]+(1\to2), \quad (3.18)$$
$$\Delta_{ip,is}=\omega_i-\omega_{p,s},\quad i=1,2 .$$

From Eq. (3.18) one can see that for $\Delta_{1p}=0$, for instance, $\text{Im}\chi^{(3)}$ remains negative for all values of Δ_{1s} for which it is significant. Hence it turns out that the third-order process gives rise to the possibility of amplification[12] of the probe wave. However the total absorption will still be positive since the contribution of $\chi^{(3)}$ terms is small. This is seen from Fig. 2. The amplification can become apparent by, for example, examining the absorption at two different values of the pump intensity. Further, one can see from Eq. (3.16) that $\chi^{(3)}$ has resonances at

$$\Delta_s=\Delta_p,\ \pm(\delta^2-\xi^2)^{1/2}, \quad (3.19)$$

which should be well resolved if Δ_p is large and if $\delta\gg\xi$. Note that the resonance $\Delta_s=\Delta_p$ is the pressure-induced extra resonance[2] (PIER), which has been discussed extensively in the context of four-wave mixing. Figure 2 shows the resonances at $\Delta_s=\pm(\delta^2-\xi^2)^{1/2}$. The resonance at $\Delta_s=\Delta_p$ is rather weak and gets washed away by the line-mixing effects. This can be seen from the analysis of Eq. (3.16). In the limit of large Δ_s and Δ_p the coefficient of the resonant term $(2i\gamma+\Delta_s-\Delta_p)^{-1}$ has an overall coefficient $[\Delta_s-\Delta_p+2i(\gamma+\sigma-\xi)]$. Thus the resonant structure corresponding to PIER will disappear if $\sigma=\xi$ and

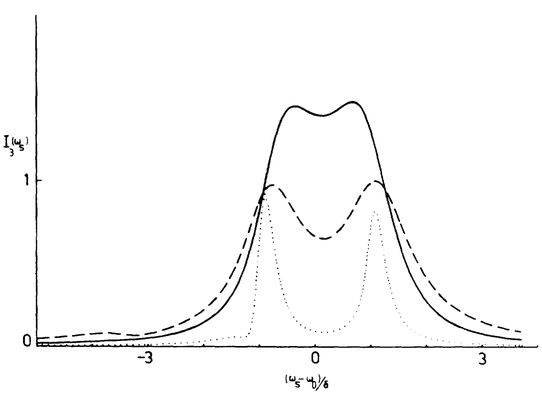

FIG. 2. Saturated absorption spectrum $I_3(\omega_s)$ as a function of $(\omega_s-\omega_0)/\delta$ for various parameters given by $\sigma/\delta=0.5$, $\gamma/\delta=0.25$, $\Delta_p=-7/2\delta$, $g/2\gamma=1.0$ ($g=de/\hbar$ is the pump Rabi frequency). The solid (dashed) line corresponds to the spectrum with cross relaxation, $\xi=\sigma$ (without cross relaxation, $\xi=0$); the dotted line denotes the case of no collisions ($\sigma=\xi=0$). The normalization in the last case is four times larger than what is shown in the figure.

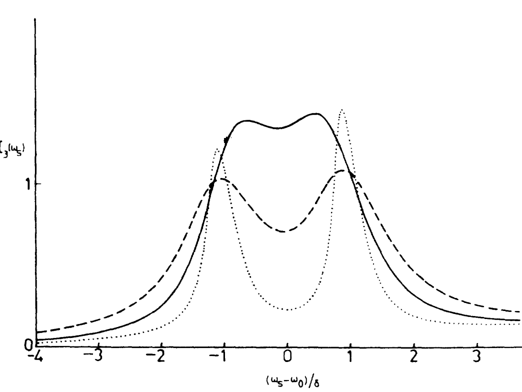

FIG. 3. Same as Fig. 2, except that now $\Delta_p=-\delta$ and $g/2\gamma=0.1$. The normalization in the case of no collisions ($\xi=\sigma=0$) is two times larger than what is shown in the figure.

this is the case in Fig. 2. We have checked numerically the existence of PIER for $\xi=\sigma/2$, etc. We also observe a small asymmetry between the two resonances at $\pm(\delta^2-\xi^2)^{1/2}$. Note also that the character of the resonance changes if Δ_p, say, is chosen close to $(\delta^2-\xi^2)^{1/2}$. Some of the resonances take the form of the square of a Lorentzian. The behavior of the absorption spectrum when the pump is tuned to one of the atomic transitions is shown in Fig. 3. This tuning results in a smaller absorption. For $\xi/\delta=1$, Eq. (3.16) reduces to

$$\chi^{(3)}(\omega_p,-\omega_p,\omega_s)=-\frac{Nd^4}{2}\left[\frac{2}{(2i\gamma+\Delta_s-\Delta_p)[\Delta_s+i(\gamma+\sigma)]}\left[\frac{1}{\Delta_s+i(\gamma+\sigma)}-\frac{1}{\Delta_p-i(\gamma+\sigma)}\right]+\frac{2}{2i\gamma[\Delta_s+i(\gamma+\sigma)]}\left[\frac{1}{\Delta_p+i(\gamma+\sigma)}-\frac{1}{\Delta_p-i(\gamma+\sigma)}\right]\right], \quad (3.20)$$

which has the same structure as that of $\chi^{(3)}$ for a two-level system with excited state lifetime $\gamma+\sigma$. For $\xi/\delta\sim\sigma/\delta\gg1$,

$$\chi^{(3)}(\omega_p,-\omega_p,\omega_s)=-\frac{Nd^4}{2}\left[\frac{-2}{(\Delta_s+i\gamma)(\Delta_p-i\gamma)}\left[\frac{1}{\Delta_s+i\gamma}+\frac{1}{\Delta_p+i\gamma}\right]\right] \quad (3.21)$$

resembles the $\chi^{(3)}$ for a two-level system when only radiative relaxation is present. Hence, for $\xi/\delta\geq1$, the third-order spectrum will consist of a single narrow symmetrical line at $\Delta_s=0$, in much the same way as in the first-order spectrum, except for a slight reduction in the peak height depending on pump detuning and pump strength.

IV. NONLINEAR SUSCEPTIBILITY FOR FOUR-WAVE MIXING WITH CROSS RELAXATION

We next examine in detail the nonlinear susceptibility $\chi^{(3)}(\omega_p,\omega_p,-\omega_s)$ describing four-wave mixing (FWM). For the model of Fig. 1 we have shown that

$$\begin{aligned}\chi^{(3)}(\omega_p,\omega_p,-\omega_s)&=-\frac{Nd^4}{2}\left\{\frac{1}{\Omega-2i\gamma}\left[\frac{A_+}{\Omega-\Delta_p+i\lambda_+}+\frac{A_-}{\Omega-\Delta_p+i\lambda_-}\right]\left[\left[\frac{A_+}{\Delta_p-i\lambda_+}+\frac{A_-}{\Delta_p-i\lambda_-}\right]-\left[\frac{A_+}{\Omega+\Delta_p+i\lambda_+}+\frac{A_-}{\Omega+\Delta_p+i\lambda_-}\right]\right]\right.\\&\quad+\frac{A_+A_-}{\Omega-2i(\gamma+\sigma)}\left[\frac{1}{\Omega-\Delta_p+i\lambda_+}-\frac{1}{\Omega-\Delta_p+i\lambda_-}\right]\left[\left[\frac{1}{\Delta_p-i\lambda_+}-\frac{1}{\Delta_p-i\lambda_-}\right]\right.\\&\quad\left.\left.+\left[\frac{1}{\Omega+\Delta_p+i\lambda_+}-\frac{1}{\Omega+\Delta_p+i\lambda_-}\right]\right]\right\}, \quad (4.1)\end{aligned}$$

where $\Omega=\omega_s-\omega_p$, $\Delta_p=\omega_p-\omega_0$, and $A_\pm,\lambda_\pm$ are defined by Eq. (3.9). One can see from Eq. (4.1) that for $\xi/\delta<1$, the resonances in $\chi^{(3)}(\omega_p,\omega_p,-\omega_s)$ are at

$$\Omega=\Delta_p-(\delta^2-\xi^2)^{1/2},\ \Delta_p+(\delta^2-\xi^2)^{1/2},\ 0,\ -[\Delta_p+(\delta^2-\xi^2)^{1/2}],\ -[\Delta_p-(\delta^2-\xi^2)^{1/2}]\,. \quad (4.2)$$

Thus, depending on the magnitude of $\Delta_p\pm(\delta^2-\xi^2)^{1/2}$ some of the lines may overlap. The widths of the two resonances at $\Omega=0$ are 2γ and $2(\gamma+\sigma)$, while the width of each of the other resonances is $\gamma+\sigma$. Consider the case when Δ_p is negative, and $\Delta_p+(\delta^2-\xi^2)^{1/2}$ is far from zero. In such a case, all the five lines corresponding to the resonances in (4.2) are distinctly present. It can be seen from (4.2) that the lines corresponding to the first (third) and the second (fourth) resonances dynamically mix with each other as ξ/δ increases. As in the linear-absorption spectrum, here too the dynamic mixing is due to the fact that the frequency separation between the lines in each of the above pairs [equal to $2(\delta^2-\xi^2)^{1/2}$] becomes smaller and smaller and approaches zero as ξ/δ approaches 1. On the contrary, the separation between the lines corresponding to the second (fourth) and the third resonances [in (4.2)], which is equal to $|\Delta_p+(\delta^2-\xi^2)|$, increases as ξ/δ approaches 1, for any given $\Delta_p<0$. Hence, one can say that

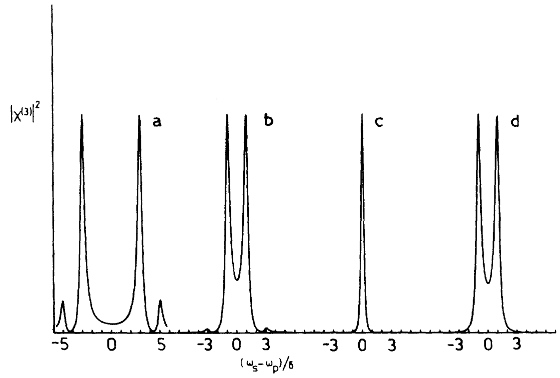

FIG. 4. Four-wave-mixing signal for the case of no collisions ($\sigma/\delta=\xi/\delta=0$) and $\gamma/\delta=0.25$ as a function of $(\omega_s-\omega_0)/\delta$. Different curves in this figure correspond to different values of pump detuning. $\Delta_p=$ (a) -4δ, (b) -2δ, (c) $-\delta$, (d) 0.

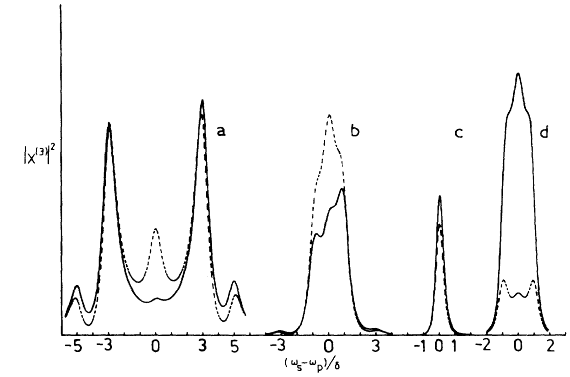

FIG. 5. Effect of change in the rate of inelastic collisions on the four-wave-mixing signal as a function of $(\omega_s-\omega_p)/\delta$. $\sigma/\delta=0.25$, $\gamma/\delta=0.25$. Solid (dashed) line corresponds to the case with cross relaxation, $\xi=\sigma$ (without cross relaxation, $\xi=0$). Different curves in this figure correspond to different values of pump detuning. $\Delta_p=$ (a) -4δ, (b) -2δ, (c) $-\delta$, (d) 0. The normalization for curves (c) and (d) is, respectively, two and four times larger than what is shown in the figure.

these lines dynamically recede from each other, or unmix, as ξ/δ approaches unity. At $\xi/\delta=1$, the mixing (or unmixing) of lines has reached a maximum, and we have just three lines at

$$\Omega=\Delta_p,\ \ 0,\ -\Delta_p \tag{4.3}$$

with widths $\gamma+\sigma$, 2γ, and $\gamma+\sigma$, respectively, since, in the limit $\xi/\delta=1$, Eq. (4.1) reduces to

$$\chi^{(3)}(\omega_p,\omega_p,-\omega_s) \propto \frac{1}{(\Omega-2i\gamma)[\Omega-\Delta_p-i(\gamma+\sigma)]} \times \left[\frac{1}{\Delta_p+i(\gamma+\sigma)}-\frac{1}{\Omega+\Delta_p-i(\gamma+\sigma)}\right]. \tag{4.4}$$

For $\xi/\delta>1$, each of the resonances $\Omega=\Delta_p$ and $\Omega=-\Delta_p$ in (4.3) corresponds to a pair of lines with widths $\gamma+\sigma\pm(\xi^2-\delta^2)^{1/2}$. Hence, as in the linear-absorption spectrum, as ξ/δ increases beyond 1, the lines centered at $\Omega=\pm\Delta_p$ become narrower and narrower. In the limit $\xi/\delta\sim\sigma/\delta\gg1$ one has

$$\chi^{(3)}(\omega_p,\omega_p,-\omega_s) \propto \frac{1}{(\Omega-\Delta_p-i\gamma)(\Delta_p+i\gamma)(\Omega+\Delta_p-i\gamma)}. \tag{4.5}$$

Hence in this limit the extra resonance $\Omega=0$ disappears, and the two lines centered at $\Omega=\pm\Delta_p$ become narrowed to a width approaching natural linewidth.

It is also interesting to look at the case when there is no collisional coupling between the two components ($\sigma/\delta=\xi/\delta=0$). In this limit, Eq. (4.1) reduces to

$$\chi^{(3)}(\omega_p,\omega_p,-\omega_s) \propto \left[\frac{1}{(\Omega-\Delta_p-\delta-i\gamma)(\Delta_p+\delta+i\gamma)(\Omega+\Delta_p+\delta-i\gamma)}+(\delta\leftrightarrow-\delta)\right], \tag{4.6}$$

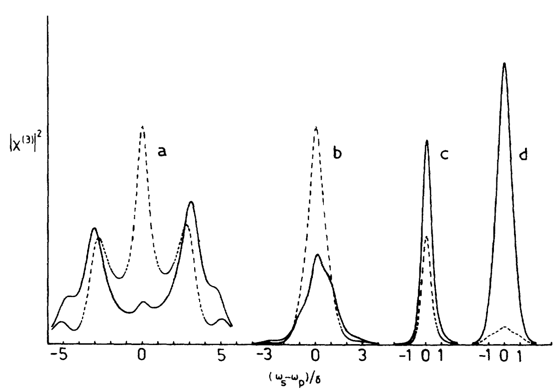

FIG. 6. Same as in Fig. 5 but for increased value of the rate of collisions $\sigma/\delta=0.5$. The normalization for curves (c) and (d) is, respectively, two and 12 times larger than what is shown in the figure.

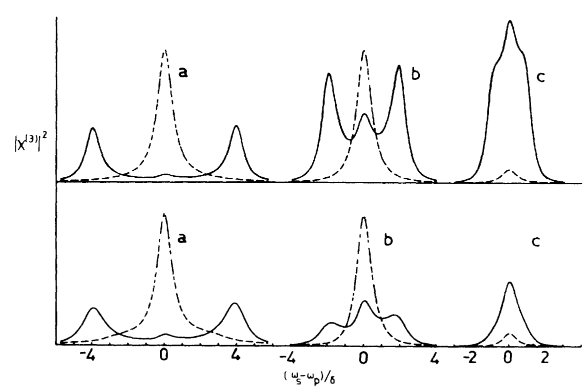

FIG. 7. Same as in Fig. 5 but now the case (d) dropped. The lower (upper) set of curves correspond to $\sigma/\delta=1.25$ ($\sigma/\delta=2.25$). The normalization for curves (c) is ten times larger than what is shown in the figure.

which is just the sum of $\chi^{(3)'s}$ for two independent two-level systems. Note that the extra resonance $\Omega=0$ now disappears, so that one has only four resonances in general, at

$$\Omega=\Delta_p-\delta,\ \ \Delta_p+\delta,\ \ -(\Delta_p+\delta),\ \ -(\Delta_p-\delta)\ . \qquad (4.7)$$

In Figs. 4–7 we plot the FWM signal $|\chi^{(3)}(\omega_p,\omega_p,-\omega_s)|^2$ versus Ω $(=\omega_s-\omega_p)$ for various values of pump detuning and cross-relaxation rates ξ $(=\sigma)$. Figure 4 shows the usual FWM signal in the absence of collisional coupling between the two components $(\xi/\delta=\sigma/\delta=0)$. In Figs. 5–7, we display the changes in the FWM signals as the strength of the collisions increases. We show results both with and without cross relaxation. When all the five lines are resolved, then cross relaxation reduces the strength of the collision-induced coherence at $\Omega=0$. The mixing and narrowing of lines as discussed above are seen. The effect of cross relaxation is seen to be more dramatic when the pump is detuned far away from the resonant frequencies of either of the two components.

In conclusion we have studied the effect of cross relaxation on saturated absorption and four-wave mixing. We have based our calculation on the general atomic relaxation equations which follow by considering the interaction of the atoms with the bath of perturbers. Cross relaxation makes a difference in the pump-induced asymmetry in the third-order absorption spectrum. The effect of cross relaxation on four-wave mixing is more dramatic than on saturated absorption. Line-mixing and line-narrowing phenomena occur in four-wave mixing as well, as in the linear-absorption spectrum.

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