CHAPTER II

CROSSRELAXATION UNDER STRONG PUMPING CONDITIONS

In Chapter VI, we had examined the nonlinear response of a model four-level system which had two transitions coupled by inelastic collisions. Considering the pump to be weak (treated to second-order in perturbation theory) we studied the effect of crossrelaxation on the saturated absorption and four-wave mixing spectra. For instance, we had shown that crossrelaxation tends to suppress the collision-induced resonant structures, both in the saturated absorption as well as the four-wave mixing spectra. It is of interest to examine how crossrelaxation would affect the response of a system to a probe field in presence of an intense pump field. In the present chapter, we consider the pump to be strong and treat it as a dressing field. Using the methods developed in Chapter VII, we obtain analytical expressions for the intensity-dependent susceptibilities that describe probe absorption and four-wave mixing in the model system of Fig.4. Using these, we examine the effect of crossrelaxation on the probe absorption and four-wave mixing spectra under conditions when the pump is strong. The analytical results clearly show the existence of the line mixing and line narrowing phenomena in the strong-field spectra similar to those in the weak-field spectra discussed in Chapter VI.
Consider the four-level system described in Chapter VI (see Fig.4) to be interacting with a strong field $\omega_s$ and a weak probe field $\omega_b$. The total field is given by (6.24). The unperturbed Hamiltonian, the strong-field and the weak-field interactions for this model are respectively given by

$$H_0 = \frac{\omega_{12}}{2} (A_{11} - A_{22}) + \frac{\omega_{24}}{2} (A_{33} - A_{44})$$ \hspace{1cm} (9.1)

$$V(t) = G(A_{12} + A_{34}) e^{-i\omega_b t} + \text{H.c.}, \quad G = -\alpha \omega$$ \hspace{1cm} (9.2)

$$F(t) = -\alpha \omega (A_{12} + A_{34}) e^{-i\omega_b t} + \text{H.c.},$$ \hspace{1cm} (9.3)

where $A_{ij}$ stands for the operator $|i\rangle \langle j|$. Note that in (9.2)-(9.3), we have taken $\hat{d}_{12} \hat{e} = \hat{d}_{34} \hat{e} = \text{d (real)}$. On making the canonical transformations on the basic dynamical equation (7.1) [with $H_0$, $V$ and $F$ given by Eqs. (9.1)-(9.3) and $L_0 \rho = -i[H_0, \rho] + L_D \rho$ given by Eqs. (6.14)] successively with

$$U(t) = e^{-i\omega_s (A_{11} - A_{22} + A_{33} - A_{44}) t/2}$$ \hspace{1cm} (9.4)

and

$$s = \begin{pmatrix} M_1 & 0 \\ 0 & M_2 \end{pmatrix}, \quad M_i = \begin{pmatrix} \cos \theta_i & -\sin \theta_i \\ \sin \theta_i & \cos \theta_i \end{pmatrix},$$

$$\tan \theta_i = \frac{2|G|}{\Delta_i + \mathcal{O}_{R_i}}.$$
\[ \Omega_{R1} = j(\Delta_i^2 + 4|G|^2), \quad i = 1, 2, \]

\[ \Delta_1 = \omega_{12} - \omega_c, \quad \Delta_2 = \omega_{34} - \omega_c \]  \hspace{1cm} (9.5)

we obtain the equations of motion in the dressed-atom picture

\[ \frac{\partial}{\partial t} \tilde{\beta} = L_o \tilde{\beta} - i[\tilde{\beta}, \tilde{\beta}] \]  \hspace{1cm} (9.6)

\[ L_o = -i[\beta, \beta] + L_D \]  \hspace{1cm} (9.7)

where \( L_D \tilde{\beta} \) gives the relaxation terms when the dressed-atom approximation is made. Note that in (9.7), \( \beta \) is the unperturbed Hamiltonian of the dressed system

\[ \beta = \sum_i \beta_i |i\rangle \langle i|, \quad i = 1, 2, 3, 4, \]

\[ \beta_{1,2} = \pm \frac{1}{2} \Omega_{R1}, \quad \beta_{3,4} = \pm \frac{1}{2} \Omega_{R2} \]  \hspace{1cm} (9.8)

Thus the \( \beta_i, i=1,2,3,4 \) in (9.8) are the energies of the dressed system while the eigenstates \( |\beta_i\rangle \) of \( \beta \) are given by the \( i \)th column of the matrix \( S \) defined in (9.5). The operator \( L_o \tilde{\beta} \) (9.7) is explicitly given by

\[
\begin{pmatrix}
(L_o \tilde{\beta})_{12} \\
(L_o \tilde{\beta})_{34}
\end{pmatrix} = M \begin{pmatrix}
\tilde{\beta}_{12} \\
\tilde{\beta}_{34}
\end{pmatrix}, \quad \begin{pmatrix}
(L_o \tilde{\beta})_{21} \\
(L_o \tilde{\beta})_{43}
\end{pmatrix} = M^\times \begin{pmatrix}
\tilde{\beta}_{21} \\
\tilde{\beta}_{43}
\end{pmatrix},
\]

\[
\begin{pmatrix}
(L_o \tilde{\beta})_{12} \\
(L_o \tilde{\beta})_{34}
\end{pmatrix} = M \begin{pmatrix}
\tilde{\beta}_{12} \\
\tilde{\beta}_{34}
\end{pmatrix}, \quad \begin{pmatrix}
(L_o \tilde{\beta})_{21} \\
(L_o \tilde{\beta})_{43}
\end{pmatrix} = M^\times \begin{pmatrix}
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\tilde{\beta}_{43}
\end{pmatrix},
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(L_o \tilde{\beta})_{43}
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\tilde{\beta}_{21} \\
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\end{pmatrix},
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\tilde{\beta}_{34}
\end{pmatrix}, \quad \begin{pmatrix}
(L_o \tilde{\beta})_{21} \\
(L_o \tilde{\beta})_{43}
\end{pmatrix} = M^\times \begin{pmatrix}
\tilde{\beta}_{21} \\
\tilde{\beta}_{43}
\end{pmatrix},
\]

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(L_o \tilde{\beta})_{12} \\
(L_o \tilde{\beta})_{34}
\end{pmatrix} = M \begin{pmatrix}
\tilde{\beta}_{12} \\
\tilde{\beta}_{34}
\end{pmatrix}, \quad \begin{pmatrix}
(L_o \tilde{\beta})_{21} \\
(L_o \tilde{\beta})_{43}
\end{pmatrix} = M^\times \begin{pmatrix}
\tilde{\beta}_{21} \\
\tilde{\beta}_{43}
\end{pmatrix},
\]

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\begin{pmatrix}
(L_o \tilde{\beta})_{12} \\
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\tilde{\beta}_{12} \\
\tilde{\beta}_{34}
\end{pmatrix}, \quad \begin{pmatrix}
(L_o \tilde{\beta})_{21} \\
(L_o \tilde{\beta})_{43}
\end{pmatrix} = M^\times \begin{pmatrix}
\tilde{\beta}_{21} \\
\tilde{\beta}_{43}
\end{pmatrix},
\]

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\begin{pmatrix}
(L_o \tilde{\beta})_{12} \\
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\tilde{\beta}_{12} \\
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\end{pmatrix}, \quad \begin{pmatrix}
(L_o \tilde{\beta})_{21} \\
(L_o \tilde{\beta})_{43}
\end{pmatrix} = M^\times \begin{pmatrix}
\tilde{\beta}_{21} \\
\tilde{\beta}_{43}
\end{pmatrix},
\]

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\end{pmatrix}, \quad \begin{pmatrix}
(L_o \tilde{\beta})_{21} \\
(L_o \tilde{\beta})_{43}
\end{pmatrix} = M^\times \begin{pmatrix}
\tilde{\beta}_{21} \\
\tilde{\beta}_{43}
\end{pmatrix},
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\begin{pmatrix}
(L_o \tilde{\beta})_{12} \\
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\end{pmatrix} = M \begin{pmatrix}
\tilde{\beta}_{12} \\
\tilde{\beta}_{34}
\end{pmatrix}, \quad \begin{pmatrix}
(L_o \tilde{\beta})_{21} \\
(L_o \tilde{\beta})_{43}
\end{pmatrix} = M^\times \begin{pmatrix}
\tilde{\beta}_{21} \\
\tilde{\beta}_{43}
\end{pmatrix},
\]
\[
\mathbf{\tilde{M}} = \begin{pmatrix}
-\mathbf{A}_{12} & \mathbf{\xi} \\
\mathbf{\mathbf{\xi}} & -\mathbf{A}_{34}
\end{pmatrix}
\]

\[
\mathbf{A}_{12} = \mathbf{D}_{R1} - \mathbf{iq}_{12}, \quad \mathbf{A}_{34} = \mathbf{D}_{R2} - \mathbf{iq}_{34}
\]

and

\[
(\mathbf{L}_0 \mathbf{\tilde{g}})_{ll} = \sum_{k} (\mathbf{p}_{lk} \mathbf{\tilde{p}}_{kk} - \mathbf{p}_{kl} \mathbf{\tilde{p}}_{ll}) \quad l=1,2,3,4
\]

The various relaxation parameters can be calculated by using (9.5) in (7.8) and these are given by

\[
\mathbf{p}_{12} = 2\gamma \sin^4 \theta_1, \quad \mathbf{p}_{21} = 2\gamma \cos^4 \theta_1
\]

\[
\mathbf{p}_{34} = 2\gamma \sin^4 \theta_2, \quad \mathbf{p}_{43} = 2\gamma \cos^4 \theta_2
\]

\[
\mathbf{p}_{13} = \mathbf{p}_{31} = \mathbf{p}_{24} = \mathbf{p}_{42} = \sigma \alpha + \xi c = \gamma_1
\]

\[
\mathbf{p}_{14} = \mathbf{p}_{41} = \mathbf{p}_{23} = \mathbf{p}_{32} = \sigma \beta - \xi c = \gamma_2
\]

\[
\mathbf{\xi} = \sigma c + \xi a
\]

\[
\mathbf{q}_{12} = \sigma + \gamma(1 + 2\sin^2 \theta_1 \cos^2 \theta_1)
\]

\[
\mathbf{q}_{34} = \sigma + \gamma(1 + 2\sin^2 \theta_2 \cos^2 \theta_2)
\]

where we have defined \(a, b\) and \(c\) as
\[ a = \sin^2 \theta_1 \sin^2 \theta_2 + \cos^2 \theta_1 \cos^2 \theta_2 \]

\[ b = \sin^2 \theta_1 \cos^2 \theta_2 + \cos^2 \theta_1 \sin^2 \theta_2 \]

\[ c = \frac{1}{2} \sin 2\theta_1 \sin 2\theta_2 \]  \hspace{1cm} (9.12)

Note that \( \sigma \) is the rate of inelastic collisions and \( \xi \) is the cross-relaxation parameter. The parameter \( \xi \) couples the coherences \( \tilde{\rho}_{12} \) and \( \tilde{\rho}_{34} \) in the dressed-atom picture. Note that this coupling is important since the factor \( \xi / |Q_{R_1} - Q_{R_2}| \) is quite significant and it cannot be thrown away in the dressed-atom approximation. The steady-state level populations of the dressed system are given by

\[ \tilde{\rho}^{(o)}_{11} = \kappa [2\gamma_1 \gamma_2 + \gamma_1 P_{34} + \gamma_2 P_{43} + P_{12}(\sigma + P_{34} + P_{43})] \]

\[ \tilde{\rho}^{(o)}_{22} = \kappa [2\gamma_1 \gamma_2 + \gamma_1 P_{34} + \gamma_2 P_{43} + P_{21}(\sigma + P_{34} + P_{43})] \]  \hspace{1cm} (9.13)

and \( \tilde{\rho}^{(o)}_{33} \) and \( \tilde{\rho}^{(o)}_{44} \) are determined from (9.13) by interchanging \( P_{12} \leftrightarrow P_{34}, \ P_{21} \leftrightarrow P_{43} \) in the expressions for \( \tilde{\rho}^{(o)}_{11} \) and \( \tilde{\rho}^{(o)}_{22} \) respectively. The normalization constant \( \kappa \) is easily determined by requiring that \( \text{Tr} \ \tilde{\rho}^{(o)} = 1 \). The operator \( \tilde{F}(t) \) in (9.6) which represents the interaction of the dressed system with the weak field is given by

\[ \tilde{F}(t) = -\tilde{d}^+ e^e e^{-i\Omega t} + H. c. \]

\[ (\tilde{d}^+)^+ = \tilde{d}^- \ , \quad \Omega = \omega_e - \omega_r \]  \hspace{1cm} (9.14)
where

\[ \tilde{d}^+ = dS^{-1} (A_{12} + A_{34}) S \]

\[ = d \left\{ -\sin \theta_1 \cos \theta_1 \langle 11 \rangle < 11 \rangle - \langle 12 \rangle < 21 \rangle + \cos^2 \theta_1 \langle 11 \rangle < 21 \rangle - \sin^2 \theta_1 \langle 12 \rangle < 11 \rangle + \langle 11 \rangle < 13 \rangle , \langle 12 \rangle < 14 \rangle , \theta_1 \leftrightarrow \theta_2 \right\} \]

(9.15)

The induced polarization is given by

\[ P(t) = \left\{ \tilde{d}(t) \right\} = \left\{ \tilde{\tilde{d}}(t) \tilde{\tilde{\rho}}(t) \right\} \]

(9.16)

where

\[ \tilde{\tilde{d}}(t) = \tilde{\tilde{d}}^- e^{-i\omega_1 t} + \tilde{\tilde{d}}^+ e^{i\omega_1 t} = \sum_a Q_a e^{-i\omega_a t} \]

(9.17)

The induced polarization to first order in the weak field and to all orders in the strong (dressing) field can be expressed as

\[ P^{(1)}(t) = e^{-i(\omega_1 + \Omega) t} I^{(1)}_{\text{abs}}(\Omega) + e^{-i(\omega_1 - \Omega) t} I^{(1)}_{\text{FWM}}(-\Omega) + \text{c.c.} \]

(9.18)

where \( I^{(1)}_{\text{abs}}(\Omega) \) and \( I^{(1)}_{\text{FWM}}(-\Omega) \) are respectively the intensity-dependent susceptibilities that describe probe absorption and four-wave mixing. These intensity-dependent susceptibilities can be obtained from the bare-atom result (6.28) by the usual renormalization procedure which in the present case consists of letting \( d_{\tilde{u}a} \rightarrow Q_a, d_{\tilde{a}a} \rightarrow q_a, \xi \rightarrow \xi' \), and using the translation table (7.16).

In what follows, we use the analytical expressions for these inten-
sity-dependent susceptibilities to examine the effect of crossrelaxation on the probe absorption and four-wave mixing spectra of the model system of Fig. 4.

A. Effect of crossrelaxation on probe absorption in presence of a strong pump

We have shown that the intensity-dependent susceptibility \( I_{\text{abs}}^{(1)}(\Omega) \) that describes energy absorption from probe \( \omega_2 \) in presence of the strong pump \( \omega_1 \) is given by expression

\[
I_{\text{abs}}^{(1)}(\Omega) = d^2 \left[ \frac{A_+}{(\Omega-n_2+\eta)+i(q_{34}-\kappa)} + \frac{A_-}{(\Omega-n_1-\eta)+i(q_{12}+\kappa)} \right.
- \frac{B_+}{(\Omega+n_2-\eta)+i(q_{34}-\kappa)} - \frac{B_-}{(\Omega+n_1+\eta)+i(q_{12}+\kappa)} \right], \quad (9.20)
\]

where

\[
A_{\pm} = \left( \tilde{p}_{11}^{(0)} - \tilde{p}_{22}^{(0)} \right) \cos^2 \theta_1 \left[ \cos^2 \theta_2 \tilde{a}_x \pm \cos^2 \theta_2 \tilde{b} \right]
+ \left( \tilde{p}_{33}^{(0)} - \tilde{p}_{44}^{(0)} \right) \cos^2 \theta_2 \left[ \cos^2 \theta_2 \tilde{a}_x \pm \cos^2 \theta_2 \tilde{b} \right],
\]

\[
B_{\pm} = \left( \tilde{p}_{11}^{(0)} - \tilde{p}_{22}^{(0)} \right) \sin^2 \theta_1 \left[ \sin^2 \theta_1 \tilde{a}_x \pm \sin^2 \theta_1 \tilde{b} \right]
+ \left( \tilde{p}_{33}^{(0)} - \tilde{p}_{44}^{(0)} \right) \sin^2 \theta_2 \left[ \sin^2 \theta_2 \tilde{a}_x \pm \sin^2 \theta_2 \tilde{b} \right], \quad (9.21)
\]

\[
\tilde{a}_x = \frac{1}{2} \left( 1 \pm \frac{i\tilde{b}}{\sqrt{(\xi^2-\tilde{p}^2)}} \right), \quad \tilde{b} = \frac{\xi}{2\sqrt{(\xi^2-\tilde{p}^2)}},
\]
\[ \bar{\beta} = \frac{1}{2} (\tilde{\Lambda}_{12} - \tilde{\Lambda}_{34}) . \]

In the expression (9.20), \( \kappa \) and \( \eta \) are respectively the real and imaginary parts of the quantity

\[ z = 1 \exp \left( \bar{\beta}^2 - \xi^2 \right) - \bar{\beta} \equiv \kappa + i\eta \quad (9.23) \]

Note that \( z \) is zero in the absence of collisions. The expression (9.20) has a simple structure in the absence of collisions (\( \sigma = \xi = 0 \)).

\[ I^{(1)}_{\text{abs}}(\Omega) = d^2 \left\{ \begin{array}{ccc}
\bar{\beta}^{(o)}_{11} & - & \bar{\beta}^{(o)}_{22} \\
\bar{\beta}^{(o)}_{33} & - & \bar{\beta}^{(o)}_{44}
\end{array} \right\} \left[ \begin{array}{cc}
\frac{\cos^2 \theta_1}{\Omega - \Omega_{R1} + i\xi_{12}} & - \frac{\sin^2 \theta_1}{\Omega + \Omega_{R1} + i\xi_{12}} \\
\frac{\cos^2 \theta_2}{\Omega - \Omega_{R2} + i\xi_{34}} & - \frac{\sin^2 \theta_2}{\Omega + \Omega_{R2} + i\xi_{34}}
\end{array} \right] , \quad (9.24) \]

Hence in absence of collisions, the probe absorption spectrum contains resonances at

\[ \Omega = -\Omega_{R2} , \quad -\Omega_{R1} , \quad \Omega_{R1} , \quad \Omega_{R2} \quad (9.25) \]

characterized by widths \( q_{34} , q_{12} , q_{12} \) and \( q_{34} \) respectively. The weights as well as the widths of these resonances depend on the detunings \( \Delta_1 , \Delta_2 \) and the strength of the pump \( G \). However when collisions are present, it can be seen from (9.20) that the probe absorption spectrum will in general exhibit resonances at
\[ \Omega = -(\Omega_{R2} - \eta), -(\Omega_{R1} + \eta), \Omega_{R1} + \eta, \Omega_{R2} - \eta, \quad (9.26) \]

which are respectively characterized by widths \( |q_{34} - \kappa| \), \( |q_{12} + \kappa| \), \( |q_{12} + \kappa| \) and \( |q_{34} - \kappa| \). The quantities \( \eta \) and \( \kappa \) are thus the collision-induced contributions respectively to the shifts and widths of the resonances in (9.25). The weights of the resonances in (9.26) are determined by the quantities \( A_{\pm} \) and \( B_{\pm} \) (9.21) and these depend on the collisional and the pump parameters in a complicated way. The general expression (9.20) already shows the mixing and narrowing of the various lines which arises due to collisions. For instance if \( \kappa \) and \( \eta \) are both positive, then the lines at \( \Omega_{R1} \) and \( \Omega_{R2} \) [or at \( -\Omega_{R1} \) and \( -\Omega_{R2} \)] are shifted closer to each other by an amount \( 2\eta \) and further, one of the lines has a narrower width than the other if \( |q_{34} - \eta| < |q_{12} + \eta| \). This is the usual mixing and narrowing phenomena which we had discussed in detail in Chapter VI in the case when the pump was absent, or when it was weak. Hence it is evident from the expression (9.20) that such mixing and narrowing phenomena occur in the probe absorption spectrum even when the pump is intense. In the following we examine a few special situations where the result (9.20) has a simple structure. Consider the case when the pump is tuned to the center of the two transitions, i.e., \( \Delta_1 = -\delta, \Delta_2 = +\delta \). In this case, Eq. (9.20) reduces to

\[
\chi^{(1)}_{abs} (\Omega) = \frac{g^2}{2} \left[ \tilde{p}_{11}^{(2)} - \tilde{p}_{22}^{(2)} \right] \frac{\delta}{\sqrt{\delta^2 + \gamma^2}} \left\{ \frac{1}{\Omega - \Omega_{R2} + i\gamma} + \frac{1}{\Omega - \Omega_{R1} + i\gamma} + \frac{1}{\Omega + \Omega_{R2} + i\gamma} + \frac{1}{\Omega + \Omega_{R1} + i\gamma} \right\}, \quad (9.27)
\]
where

\[ \Gamma_\pm = \sigma + \gamma \left( 1 + \frac{-2G}{\delta^2 + 4G^2} \right) \pm \left( \frac{\sigma + \xi}{2} \right) \frac{-4G^2}{\delta^2 + 4G^2}, \]  

(9.28)

Note that in the case when \( \Delta_1 = -\delta, \Delta_2 = \delta \), the mixing parameter \( \eta \) is zero. Hence the probe absorption spectrum in this case has resonances as in (9.25). However, some of these resonances can be narrower than the others. For instance the resonances at \( \Omega = \pm \Omega_{R2} \) have a narrower width \( \Gamma_\pm \) (9.28). Note that here both the inelastic collision rate \( \sigma \) as well as the crossrelaxation \( \xi \) contribute to the narrowing of the lines, unlike in the linear absorption spectrum in the absence of pump where only the crossrelaxation \( \xi \) contributed to the narrowing. In the case when \( \xi = \sigma \) and \( \frac{\delta}{G} \ll 1 \), the resonances at \( \Omega = \pm \Omega_{R2} \) have a width \( \Gamma_\pm \approx 3\gamma/2 \) which is independent of the collisional parameters \( \sigma \) and \( \xi \). Another situation of interest is when the pump is tuned to one of the transitions, for example, the \( |11\rangle \leftrightarrow |12\rangle \) transition so that \( \Delta_1 = 0 \). In the case when \( \Delta_1 = 0 \) and \( \frac{\delta}{G} \ll 1 \), the expression (9.20) reduces to

\[ I_{\text{abs}}^{(1)}(\Omega) = d^2 \left\{ \frac{A_+}{\Omega - 2G - \frac{\delta}{2G} + i\Gamma_-} + \frac{A_-}{\Omega - 2G - \frac{\delta}{2G} + i\Gamma_+} - \frac{B_+}{\Omega + 2G + \frac{\delta}{2G} + i\Gamma_-} \right\}, \]

(9.29)

where

\[ A_\pm = \frac{1}{2} \left( \bar{p}_{33}^{(0)} - \bar{p}_{44}^{(0)} \right) \left[ 1 \pm \frac{1}{4} + \frac{\delta^2}{2G^2} \left( 1 \pm \frac{2G + i\gamma - (3\sigma + \xi)}{\sigma + \xi} \right) \right]. \]
\[ B_\pm = \pm \frac{1}{4} \left[ \bar{\tilde{p}}^{(o)}_{33} - \bar{\tilde{p}}^{(o)}_{44} \right] \frac{\delta^2}{4G^2} \]  

(9.30)

and the widths \( \Gamma_+ \) and \( \Gamma_- \) are now given by

\[ \Gamma_+ = \frac{3\lambda}{2} - \frac{\lambda^2}{4G^2} + \frac{3\delta + \frac{3\delta^2}{2G}}{4G^2} \]

\[ \Gamma_- = \frac{3\lambda}{2} - \frac{\lambda^2}{4G^2} + \frac{\delta - \delta^2}{2G} \]

(9.31)

Thus one can see from Eqs. (9.29)-(9.30) that the probe absorption spectrum in the limit \( \Delta_1 = 0, \frac{\delta}{G} \ll 1 \), consists of a doublet at \( \Omega = 2G + \frac{\delta^2}{2G} \) and \( \Omega = -2G - \frac{\delta^2}{2G} \) with a width given by \( \Gamma_- \). However the strength of the peak at \( \Omega = -2G - \frac{\delta^2}{2G} \) is quite small compared to that of the peak at \( \Omega = 2G + \frac{\delta^2}{2G} \) since \( |B_\pm| \ll |A_\pm| \) (9.30). The effect of the other Lorentzian terms with \( \Gamma_+ \) in (9.29) is merely to reduce the strength of the two lines.

B. **Effect of crossrelaxation on the four-wave mixing signal in presence of a strong pump**

We have shown that the intensity-dependent susceptibility, \( I_{FWM}^{(1)}(-\Omega) \) that describes four-wave mixing in the model system of Fig.4 is given by

\[ I_{FWM}^{(1)}(-\Omega) = d^2 \left[ \frac{C_+}{(-\Omega-\Omega_{R2}+\eta)+i(q_{34}-\kappa)} + \frac{C_-}{(-\Omega-\Omega_{R1}-\eta)+i(q_{12}+\kappa)} \right. \]

\[ - \frac{D_+}{(-\Omega+\Omega_{R2}-\eta)+i(q_{34}-\kappa)} - \frac{D_-}{(-\Omega+\Omega_{R1}+\eta)+i(q_{12}+\kappa)} \right] \]

(9.32)
where

\[ C_\pm = -\left( \tilde{p}_{11}^{(o)} - \tilde{p}_{22}^{(o)} \right) \sin^2 \theta_1 \left( \sin^2 \theta_1 \tilde{a}_\pm \pm \cos^2 \theta_2 \tilde{b} \right) \]

\[ - \left( \tilde{p}_{33}^{(o)} - \tilde{p}_{44}^{(o)} \right) \sin^2 \theta_2 \left( \cos^2 \theta_2 \tilde{a}_\pm \pm \cos^2 \theta_1 \tilde{b} \right) \]

\[ D_\pm = -\left( \tilde{p}_{11}^{(o)} - \tilde{p}_{22}^{(o)} \right) \cos^2 \theta_1 \left( \sin^2 \theta_2 \tilde{a}_\pm \pm \sin^2 \theta_1 \tilde{b} \right) \]

\[ - \left( \tilde{p}_{33}^{(o)} - \tilde{p}_{44}^{(o)} \right) \cos^2 \theta_2 \left( \sin^2 \theta_2 \tilde{a}_\pm \pm \sin^2 \theta_1 \tilde{b} \right) \]

(9.33)

Here the quantities \( \tilde{a}_\pm, \tilde{b}_\pm \) are as given by Eq. (9.22). Note, that the expression in (9.32) has the same structure as that in (9.20) except for the fact that \( \Omega \) is now replaced by \(-\Omega\), and \( A_\pm, B_\pm \) are respectively replaced by \( C_\pm \) and \( D_\pm \). Hence the resonances in the four-wave mixing spectrum occur at the same frequencies [as in (9.26)] as in the probe absorption spectrum. Thus, the resonances in the four-wave mixing spectrum are at

\[ -\Omega = -(\Omega_{R2} - \eta), -(\Omega_{R1} + \eta), \Omega_{R1} + \eta, \Omega_{R2} - \eta \]

(9.34)

which are respectively characterized by widths \(|q_{34} - \kappa|, |q_{12} + \kappa|, |q_{12} + \kappa| \) and \(|q_{12} - \kappa|\). In the absence of collisions, the expression in (9.32) reduces to
\[ I_{\text{FWM}}^{(1)}(-\Omega) = d^2 \left\{ \left[ \tilde{p}_{11}^{(0)} - \tilde{p}_{22}^{(0)} \right] \left[ \frac{\sin^2 \theta_1 \cos^2 \theta_1}{\Omega + \Omega_{R_1} + i \eta_{12}} - \frac{\sin^2 \theta_1 \cos^2 \theta_1}{\Omega - \Omega_{R_1} - i \eta_{12}} \right] \right. \\
+ \left[ \tilde{p}_{33}^{(0)} - \tilde{p}_{44}^{(0)} \right] \left[ \frac{\sin^2 \theta_2 \cos^2 \theta_2}{\Omega + \Omega_{R_2} + i \eta_{34}} - \frac{\sin^2 \theta_2 \cos^2 \theta_2}{\Omega - \Omega_{R_2} - i \eta_{34}} \right] \right\} \]  

(9.35)

Thus the four-wave mixing spectrum in the absence of collisions consists of four resonances, namely at \( \Omega = \pm \Omega_{R_1} \), and \( \Omega = \pm \Omega_{R_2} \). However, when collisions are present, the mixing and narrowing phenomena occur due to the presence of the collision-induced intensity-dependent parameters \( \eta \) and \( \kappa \) in the expression (9.32). In the case when the pump is tuned to the center of the two transitions, i.e., \( \Delta_1 = -\delta \), \( \Delta_2 = \delta \), the quantities \( C_{\pm} \) and \( D_{\pm} \) in Eq. (9.32) become zero. Hence the four-wave mixing spectrum in this case vanishes. Note that this is so because in deriving (9.32) we have made the dressed-atom approximation. However, the non-secular terms which have been thrown away under the dressed-atom approximation give rise to nonvanishing contribution to the four-wave mixing spectrum. In the other situation where the pump is resonant with one of the transitions, namely the \( |11\rangle \leftrightarrow |12\rangle \) transition, and in the limit \( \frac{\delta}{G} \ll 1 \), the expression in (9.32) reduces to

\[ I_{\text{FWM}}^{(1)}(-\Omega) = d^2 \left[ \frac{C_+}{\Omega - 2G - \frac{\delta^2}{2G} + i \Gamma_-} - \frac{D_+}{\Omega + 2G + \frac{\delta^2}{2G} + i \Gamma_-} \right. \\
- \frac{D_-}{\Omega + 2G - \frac{\delta^2}{2G} + i \Gamma_+} \right] , \]  

(9.36)
where

\[ C_+ = - \left( \tilde{p}^{(o)}_{33} - \tilde{p}^{(o)}_{44} \right) \frac{\delta^2}{4G^2}, \]

\[ D_\pm = - \left( \tilde{p}^{(o)}_{33} - \tilde{p}^{(o)}_{44} \right) \left[ \pm \frac{1}{4} + \frac{\delta^2}{8G^2} \left( 1 \mp \frac{3\sigma + \xi}{2(\sigma + \xi)} \right) \right], \quad (9.37) \]

and the widths \( \Gamma_+ \) and \( \Gamma_- \) are given by

\[ \Gamma_+ = \frac{3\delta}{2} - \frac{\delta^2}{4G^2} + \frac{3\sigma + \xi}{2} - \frac{\delta^2}{4G^2}, \]

\[ \Gamma_- = \frac{3\delta}{2} - \frac{\delta^2}{4G^2} + \frac{\sigma - \xi}{2} + \frac{\delta^2}{4G^2} \quad (9.38) \]

Thus in this limit, the four-wave mixing spectrum consists of two lines at \( \Omega = \pm (2G + \frac{\delta^2}{2G}) \) characterized by a width \( \Gamma_- \). However since \( |C_+| < |D_+| \), the line at \( \Omega = -(2G + \frac{\delta^2}{2G}) \) is expected to be weak compared to the line at \( \Omega = +(2G + \frac{\delta^2}{2G}) \). The effect of the other Lorentzian at \( \Omega = +(2G + \frac{\delta^2}{2G}) \) with the much broader width \( \Gamma_+ \) is merely to reduce the strength of the line at \( \Omega = +(2G + \frac{\delta^2}{2G}) \).

Thus, in conclusion, we have studied the effect of cross-relaxation on probe absorption and four-wave mixing under conditions when the pump is strong. Analytical expressions for the intensity-dependent susceptibilities describing probe absorption and four-wave mixing are obtained. These analytical expressions enable us to obtain a qualitative understanding of the effect of cross-relaxation on the probe absorption and the four-wave mixing.
spectra. Various special cases are discussed. It is evident from the structure of these expressions that crossrelaxation enhances the line mixing and the line narrowing effects in the strong-field spectra.