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Four-wave mixing in strongly driven four-level systems

Fernando A. M. de Oliveira
Departamento de Física, Universidade Federal do Rio Grande do Norte, 59000 Natal, Rio Grande do Norte, Brazil and Optics Section, Blackett Laboratory, Imperial College, London SW7 2BZ, England

Cid B. de Araújo and José R. Rios Leite
Departamento de Física, Universidade Federal de Pernambuco, 50000 Recife, Pernambuco, Brazil
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A nonperturbative theoretical study of four-wave-mixing processes in four-level systems is presented. Using the density-matrix formalism, numerical results are obtained for two arbitrarily intense pump fields with the third field a weak probe. The fourth wave, radiated by the system, is obtained analytically for some special cases. We find, for different frequency detunings, Rabi frequencies, and relaxation rates that the line shapes exhibit: (i) dynamic Stark splitting and shifting, and merging of resonances; (ii) power broadening with two-photon antiresonance behavior resulting from a destructive interference between different contributions to the optical polarization; and (iii) extra resonances induced by field intensities and saturation processes.

I. INTRODUCTION

Four-wave mixing (FWM) is a nonlinear optical phenomenon that describes the combination of three propagating light waves in a medium, generating a fourth wave.[1] Among the several Fourier components of the nonlinear optical polarization created, one particular component has the frequency $\omega_4=\omega_1-\omega_2+\omega_3$, which generates a new field at the same frequency.

After the first observation of the FWM phenomenon by Maker and Terhune,[2] several nonlinear techniques such as coherent anti-Stokes Raman spectroscopy[3] (CARS), Raman-induced Kerr effect[3] (RIKE) and others[4–8] have been developed. Investigations in this area have provided a variety of new information on the nonlinear properties of matter. Using tunable laser sources from the ultraviolet to the infrared, line spectra of electronic, vibrational, and rotational transitions have been studied.[9] Since many of these interactions are enhanced on—or very near—resonance, saturation effects must be considered in order to understand the FWM spectra, whenever large Rabi frequencies[10] are used, to account for power broadening, line splitting and shift, and even new resonances due to higher-order effects.

As a consequence, wave-mixing studies in strongly driven systems have been of great interest in recent years.[3–9] Degenerate FWM in homogeneously and Doppler broadened media under a high-intensity pump has been investigated in atomic vapors.[11,12] Phase conjugation,[13] oscillation, and amplification of Stokes and anti-Stokes emission at the Rabi regime,[14(a)] extra resonances induced by saturation effects,[14(b)] and bistability phenomena[15] have also been reported. In a recent paper,[16] FWM by four-level systems was treated under conditions where relaxation processes could be ignored. Saturation effects in CARS (Ref. 17) and degenerate FWM (Ref. 18) by three-level systems have also been studied. Double-sided diagrams have been used to calculate perturbatively the FWM susceptibility.[19,20] However, although a considerable amount of work has been done, most of the studies of saturation effects in degenerate or near-degenerate four-wave mixing do not consider in detail the two-photon coherence, with the exception of Ref. 17 where some cases have been studied.

In this paper, we analyze intensity effects influencing a four-wave mixing process in which the generated field is proportional to the two-photon or Raman coherence between two levels of a four-level system. The calculation of the resonant susceptibility is made by using the optical Bloch equations.[10,21] Analytical and numerical results are presented. A solution is given, which is valid for two fields of arbitrary intensity and the third field considered as a weak probe and treated only to first order in the amplitude. An analysis is made for several values of the intensities of the pump fields, detunings, and features like line broadening, resonance shift and splitting, and other saturation effects are investigated. The role of the dynamic Stark effect is analyzed in detail. Destructive interferences between saturated optical coherences, leading to the total cancellation of the FWM polarization are studied in the presence of one- and two-photon (Raman or Zeeman) resonances. This effect is very interesting by itself and is complementary to the population trapping phenomenon.[22] The present steady-state theory is appropriate for homogeneously broadened systems, such as atomic beams. The analysis of the transient regime, its connection to transient saturated spectroscopy,[23] and the study of Doppler broadened systems, appropriate for gases in a cell, will be published elsewhere.

The paper is divided as follows. In Sec. II we present the four-level system and the interaction with the electromagnetic (e.m.) field. We show that with a suitable choice of the weak-field frequency, the four-wave-mixing polarization becomes proportional to a two-photon coherence between two atomic levels of the same parity, and the problem is reduced to the interaction of a three-level system with two e.m. fields. The Bloch equations are presented and solved. Two limits are discussed in Sec. III.

(i) *Only one intense field.* In this case, the results are shown to be asymmetrical: they depend on which one of the pump fields is the more intense one. A destructive interference is predicted at the onset of the Rabi regime.

(ii) *Two strong fields.* Here, we show that the line shapes obtained in Sec. III A change drastically, when both pump fields have comparable saturating intensities. The destructive interference predicted in Sec. III A is shown to extend also to the fully saturated regime for large values of the two strong field amplitudes.

Finally we make a summary of the principal results in Sec. IV.

II. DENSITY-MATRIX EQUATIONS

We consider a material system consisting of four-level atoms (see Fig. 1). Three laser fields $E_1(t)$, $E_2(t)$, and $E_3(t)$ couple to dipole transitions $|a\rangle-|c\rangle$, $|b\rangle-|c\rangle$, and $|b\rangle-|d\rangle$, respectively. The electric fields are given by

$$E_i(t)=E_i e^{-i\omega_i t}+\text{c.c.} \ , \tag{1}$$

where ω_i, E_i are the frequency and amplitude of field i $(i=1,2,3)$. States $|a\rangle$ and $|b\rangle$ ($|c\rangle$ and $|d\rangle$) are assumed to have the same parity. The electric-dipole matrix elements are denoted by μ_{ij}; the resonance frequencies are $\omega_{ij}=\hbar^{-1}(E_i-E_j)$, where E_i is the energy of level i. The atomic density operator ρ satisfies the equation[1]

$$\frac{d\rho}{dt}=-\frac{i}{\hbar}[H,\rho]+\left[\frac{d\rho}{dt}\right]_{\text{rel}}+\Lambda \ , \tag{2}$$

where H includes the free-atom Hamiltonian and electric-dipole interaction, which is given by $V=-\mu E$; the relaxation is described by

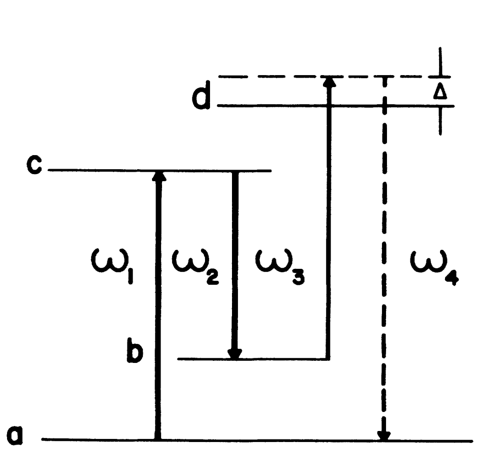

FIG. 1. Four-level system used to calculate $\chi(\omega_4)$. E_4 is the field generated by four-wave mixing.

$$\left[\frac{d\rho_{ij}}{dt}\right]_{\text{re}}=-\Gamma_{ij}\rho_{ij} \ ,$$

where the relaxation rates are denoted by Γ_{ij} $(i,j=a,b,c,d)$; the incoherent pumping of population to the levels is represented by the diagonal matrix $\Lambda_{ij}=\Gamma_i\rho_{ii}^{(0)}\delta_{ij}$. In order to describe optical transitions we assume that the dipole transition frequencies are of the order of 10^{14} Hz, the Raman transition frequency is $\leq 10^{12}$ Hz, and all the relaxations rates characterizing the system are small ($\Gamma_{ij}\leq 10^8$ Hz) compared to the Raman frequency. The nonlinear optical mixing generates a polarization which has, among others, one component given by[1]

$$P(\omega_4)=\mu_{ad}\rho_{da}=\chi(\omega_4)E_1E_2^*E_3 \ , \tag{3a}$$

with

$$\omega_4=\omega_1-\omega_2+\omega_3 \ , \tag{3b}$$

where $\chi(\omega_4)$ is the nonlinear susceptibility at frequency ω_4. The direction of propagation for the FWM signal can be determined by the phase-matching condition $\mathbf{k}_4\approx\mathbf{k}_1-\mathbf{k}_2+\mathbf{k}_3$, where $\mathbf{k}_i$ is the wave vector of field i $(i=1,2,3,4)$. Under the experimental conditions of an optically thin sample of length L, $\Delta kL\ll 1$, where $\Delta k=|\mathbf{k}_4-\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3|$, when pump depletion is negligible, and in the slowly varying envelope approximation, the intensity of the generated wave is proportional to $|\chi(\omega_4)|^2$ (see, e.g., Bloch and Ducloy in Ref. 12, and references therein).

In order to have each laser interacting with only one transition, as assumed above, one can adjust the beam polarizations[11,12] or assume that each laser is much closer to one particular resonance than to the other ones. Hence, $|\omega_1-\omega_{ca}|\ll|\omega_2-\omega_{ca}|$, $|\omega_3-\omega_{ca}|$, $|\omega_4-\omega_{ca}|$, with similar relations for $|\omega_4-\omega_{cb}|$ and $|\omega_3-\omega_{db}|$ and $|\omega_4-\omega_{da}|$ (see Fig. 1). Nevertheless, the detuning Δ of field E_3 with respect to ω_{db} is assumed to be much larger than the relaxation rates of the atomic system, the detunings of the other fields to their respective resonances, and the Rabi frequency of the field E_3, defined by $\Omega_3=\mu_{db}E_3/\hbar$, so that the field E_3 can be treated perturbatively. Therefore the four-wave-mixing polarization at frequency ω_4 can be calculated up to first order of the field E_3. From Eq. (3a), $P(\omega_4)$ is proportional to the density-matrix element ρ_{da}. This can be calculated using Eq. (2), and in steady state is given by

$$\rho_{da}=\frac{i}{\hbar}\frac{\mu_{db}E_3\rho_{ba}}{\omega_{da}-\omega_4-i\Gamma_{da}} \ . \tag{4}$$

By Eq. (3b) (see Fig. 1), and because ω_3 is far from a resonance frequency of the atom, we can neglect Γ_{da} in the denominator of this equation, and consider $\omega_{da}-\omega_4$ as approximately constant, so that the dominant frequency behavior of $\chi(\omega_4)$ as a function of ω_1 and ω_2 is described by ρ_{ba}. Therefore, with the help of Eqs. (3) and (4), the four-wave-mixing susceptibility can be written as

$$\chi(\omega_4) \approx \frac{i\mu_{ad}\mu_{db}}{\hbar(\omega_{da}-\omega_4)E_1E_2^*}\rho_{ba} \ . \tag{5}$$

This susceptibility $\chi(\omega_4)$ is independent of the field amplitude E_3, and so hereafter we can concentrate in the reduced three-level system interacting with two fields of arbitrary intensity. The equations of motion for the density-matrix elements related to levels $|a\rangle$, $|b\rangle$, and $|c\rangle$ are obtained from Eq. (2). The rotating-wave approximation gives the optical Bloch equations for the slowly varying quantities ρ_{ij} (e.g., Refs. 10 and 21):

$$\dot\rho_{ba} = -i\Delta_{ba}\rho_{ba} - i\Omega_1\rho_{bc} + i\Omega_2^*\rho_{ca} \ , \tag{6a}$$

$$\dot\rho_{ca} = -i\Delta_{ca}\rho_{ca} + i\Omega_1(\rho_{aa}-\rho_{cc}) + i\Omega_2\rho_{ba} \ , \tag{6b}$$

$$\dot\rho_{bc} = +i\Delta_{cb}^*\rho_{bc} - i\Omega_2^*(\rho_{bb}-\rho_{cc}) - i\Omega_1^*\rho_{ba} \ , \tag{6c}$$

$$\dot\rho_{aa} = -\Gamma_a(\rho_{aa}-\rho_{aa}^{(0)}) + i(\Omega_1^*\rho_{ca} - \text{c.c.}) \ , \tag{6d}$$

$$\dot\rho_{bb} = -\Gamma_b(\rho_{bb}-\rho_{bb}^{(0)}) - i(\Omega_2\rho_{bc} - \text{c.c.}) \ , \tag{6e}$$

$$\dot\rho_{cc} = -\Gamma_c(\rho_{cc}-\rho_{cc}^{(0)}) - i(\Omega_1^*\rho_{ca} - \text{c.c.}) + i(\Omega_2\rho_{bc} - \text{c.c.}) \ , \tag{6f}$$

where $\Delta_{ca}=\omega_{ca}-\omega_1-i\Gamma_{ca}$, $\Delta_{cb}=\omega_{cb}-\omega_2-i\Gamma_{cb}$, and $\Delta_{ba}=\omega_{ba}-(\omega_1-\omega_2)-i\Gamma_{ba}$ are the complex detunings and $\Omega_1=\mu_{ca}E_1/\hbar$, $\Omega_2=\mu_{cb}E_2/\hbar$ are the Rabi frequencies. Equations similar to Eqs. (6a)–(6f) have been studied before, with different relaxation models and incoherent pumping, in connection with various phenomena, such as free-induction decay,[23] three-level laser theory,[24] laser-induced line narrowing,[25] double optical resonance,[26] phase conjugation,[11,12] population trapping,[22] and squeezing in resonance fluorescence,[27] among others.[10,21,28] In the present paper, the relaxation rates are assumed to be independent of each other: Γ_i ($i=a,b,c$) are the population relaxations, Γ_{ca} and Γ_{cb} are the optical coherence dephasing rates, and Γ_{ba} is the two-photon (Zeeman or Raman) relaxation rate. They must satisfy $\Gamma_{ij}\geq(\Gamma_i+\Gamma_j)/2$ (for a discussion of relaxations, see, e.g., Cohen-Tannoudji in Ref. 29).

We are interested in the steady-state regime, obtained by dropping all time derivatives in Eq. (6). From Eqs. (6a)–(6c), one obtains the Raman coherence

$$\rho_{ba} = \frac{\Omega_1\Omega_2^*[\Delta_{cb}^*(\rho_{aa}-\rho_{cc}) - \Delta_{ca}(\rho_{bb}-\rho_{cc})]}{\Delta_{ba}\Delta_{ca}\Delta_{cb}^* + |\Omega_1|^2\Delta_{ca} - |\Omega_2|^2\Delta_{cb}^*} \ . \tag{7}$$

In Eq. (7) the lowest-order terms contributing in the perturbation limit are the second-order terms (proportional to $\Omega_1\Omega_2^*$). The denominator of Eq. (7) is associated with the dynamic Stark effect and has been studied in the context of two-photon absorption, saturated absorption spectroscopy,[30–33] and phase conjugation by FWM.[11,12] The closed solution for ρ_{ba} is obtained if the values for ρ_{aa}, ρ_{bb}, and ρ_{cc} are determined. The calculation is lengthy, but straightforward, and we just quote the results in the Appendix. The equations developed in this section, particularly Eq. (7), contain the essential results of this paper. They can be applied for arbitrary intensities of the fields E_1 and E_2. In Sec. III we discuss two particularly relevant limits which enable us to obtain a clearer physical understanding of Eq. (7).

III. DISCUSSION

A. Limit of one strong pump field

In this section the behavior of the four-wave mixing is examined under the conditions of only one arbitrary intense field. (Some results of this section have been discussed before by the authors.[20,34]) We anticipate that by choosing either E_1 or E_2 as the dominant field, very different behaviors are predicted for the four-wave-mixing signal. In the rest of this paper, we will assume that $\rho_{bb}^{(0)}=\rho_{cc}^{(0)}=0$.

1. $\Omega_2\to 0$

First we consider the high-intensity effects produced by field E_1. In the limit $\Omega_2\to 0$, Eqs. (A11)–(A13) lead to

$$W_{ca} = 2|\Omega_1/\Delta_{ca}|^2\Gamma_{ca} \ , \tag{8}$$

$$W_{cb}, W_{ba} \to 0 \ . \tag{9}$$

From Eqs. (8) and (9), one sees that E_1 induces transitions between $|a\rangle$ and $|c\rangle$, but the rate of transitions to state $|b\rangle$ is negligible. We define the one-photon detunings $\delta_1=\omega_{ca}-\omega_1$ and $\delta_2=\omega_{cb}-\omega_2$. The rate of transitions induced by E_1 increases near resonance ($\delta_1\sim 0$). The populations can be calculated substituting Eqs. (8) and (9) into the expressions given in the Appendix. The result is

$$\frac{\rho_{aa}}{\rho_{aa}^{(0)}} = 1 - \frac{2|\Omega_1|^2\Gamma_{ca}\Gamma_a^{-1}}{|\Delta_{ca}|^2 + 2|\Omega_1|^2\Gamma_{ca}(\Gamma_c^{-1}+\Gamma_a^{-1})} \ , \tag{10}$$

$$\frac{\rho_{bb}}{\rho_{aa}^{(0)}} = 0 \ , \tag{11}$$

$$\frac{\rho_{cc}}{\rho_{aa}^{(0)}} = \frac{2|\Omega_1|^2\Gamma_{ca}\Gamma_c^{-1}}{|\Delta_{ca}|^2 + 2|\Omega_1|^2\Gamma_{ca}(\Gamma_c^{-1}+\Gamma_a^{-1})} \ . \tag{12}$$

Using Eqs. (10) and (12) in Eq. (7), we obtain

$$\rho_{ba} = \rho_{aa}^{(0)}\Omega_1\Omega_2^* \frac{(\Delta_{cb}^*\Delta_{ca}^* + 2|\Omega_1|^2\Gamma_{ca}\Gamma_{ca}\Gamma_c^{-1})}{(\Delta_{bc}\Delta_{cb}^* + |\Omega_1|^2)[\,|\Delta_{ca}|^2 + 2|\Omega_1|^2\Gamma_{ca}(\Gamma_a^{-1}+\Gamma_c^{-1})]} \ . \tag{13}$$

For the saturation parameter $2|\Omega_1|^2(\Gamma_a^{-1}+\Gamma_c^{-1})\Gamma_{ca}^{-1} \geq 1$, the terms in the denominator of Eq. (13) containing $(\Gamma_a^{-1}+\Gamma_c^{-1})$ are responsible for power broadening. On the other hand, the intensity effects due to the first term in parentheses in the denominator of Eq. (13) are responsible for the ac Stark effect on the coupled transition.

The first term inside the parentheses in the numerator of Eq. (13) results from the coupling of the field E_2 to the saturated optical polarization (ρ_{ca} coherence) induced by E_1 on the two-level $|a\rangle$-$|c\rangle$ subsystem. This term is independent of the population relaxation rates only at the lowest order in the power expansion described above. Saturation effects introduce nonparametric, yet coherent contributions.

The second term inside the parentheses in the numerator of Eq. (13) is due to the coupling of the field E_1 to the optical polarization (ρ_{bc} coherence) induced by E_2 on the $|b\rangle$-$|c\rangle$ transition. The population involved here is the change in the common level (ρ_{cc}) population due to the E_1 field saturation of the two-level $|a\rangle$-$|c\rangle$ subsystem. Thus this term is not of parametric character (Γ_c is explicit there). It is, though, coherent and leads to an interesting interference feature capable of total signal cancellation at detuning $\delta_1=0$.

If one takes the strong laser on resonance ($\delta_1=0$), the susceptibility will be a function of two variables—the weak laser detuning δ_2 and the strong laser Rabi frequency Ω_1. Figure 2 shows the behavior of $|\chi(\omega_4)|^2$ as a function of these variables. At low intensity, one has a single peak centered at $\delta_2=0$. This signal is due to the first contribution in Eq. (13) and is purely parametric. It has a Lorentzian line shape of width Γ_{ba}. As saturation effects become important, the first factor of the denomi-

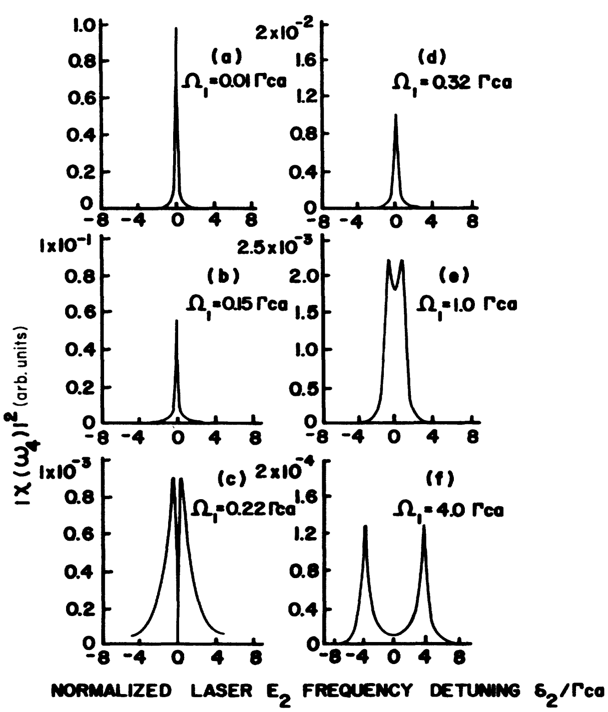

FIG. 2. $|\chi|^2$ as a function of the weak pump laser normalized detuning δ_2/Γ_{ca} and strong pump laser Rabi frequency $|\Omega_1|$. Parameters are $\Gamma_{cb}=\Gamma_{ca}$, $\Gamma_{ba}=\Gamma_a=\Gamma_b=\Gamma_c=0.1\Gamma_{ca}$, $|\Omega_2|=0.001\Gamma_{ca}$, $\omega_{ca}=\omega_1$, $\rho_{aa}^{(0)}=1$, $\rho_{bb}^{(0)}=\rho_{cc}^{(0)}=0$.

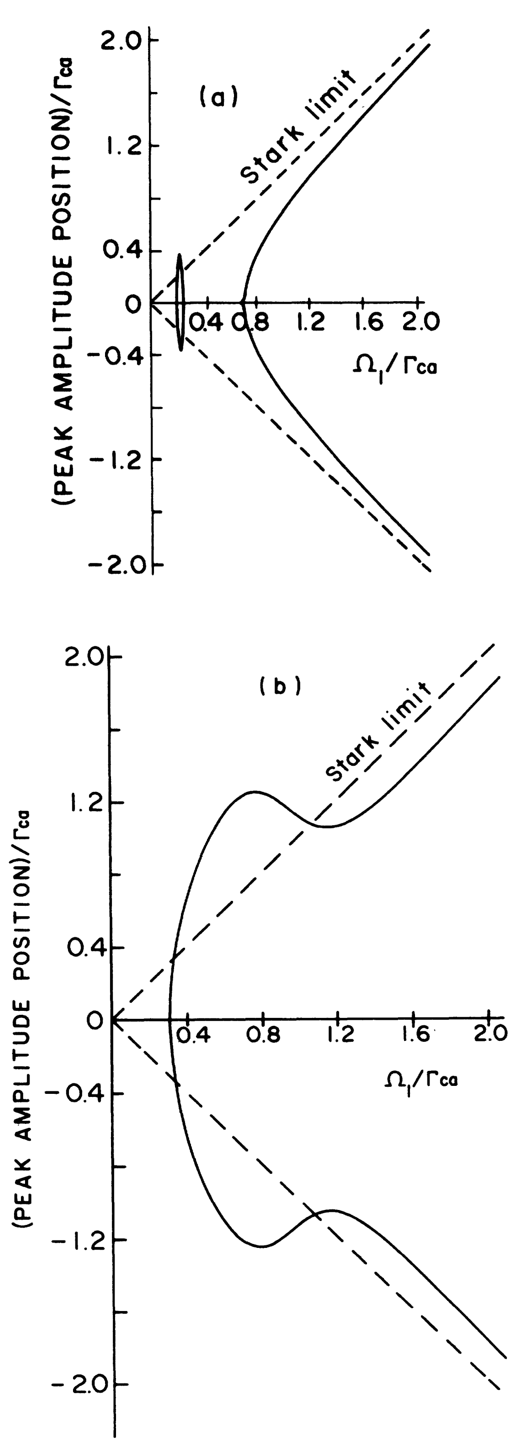

FIG. 3. Peak amplitude position of $|\rho_{ba}/\Omega_1\Omega_2^*|^2$ as a function of the pump laser amplitude. Line splitting is indicated by the curve bifurcation. Parameters are (a) as in Fig. 2; (b) all relaxation rates are set equal to Γ_{ca}.

nator of Eq. (13) begins to split the single line obtained at low intensity. For this condition, the term resulting from population saturation becomes significant and coherently adds to the parametric one. The sign of the second term is negative while the saturated population term is positive. Thus for $\delta_2=0$ there will be a value of the strong laser intensity for which the numerator of Eq. (13) completely cancels. With the value of the Rabi frequency $|\Omega_1^c|=(\Gamma_{bc}\Gamma_c/2)^{1/2}$, the signal appears as a power broadened line with a hole of width

$$[(\Gamma_{ba}+\Gamma_{bc})^2+4\Gamma_{ba}\Gamma_{bc}+2\Gamma_c\Gamma_{bc}]^{1/2}-(\Gamma_{ba}+\Gamma_{bc})$$

at its center [Fig. 2(c)]. This antiresonance behavior arises from a destructive interference between the contributions of ρ_{ca} and ρ_{bc} to the element ρ_{ba}, and has been reported before.[20,34] The interference occurs because $\mathrm{Im}(\rho_{ca}/\Omega_1)$ and $\mathrm{Im}(\rho_{bc}/\Omega_2^*)$ as functions of $|\Omega_1|$ for $\delta_1=\delta_2=0$ have the same value at the critical value $|\Omega_1^c|=(\Gamma_{bc}\Gamma_c/2)^{1/2}$ and contribute with opposite sign to ρ_{ba}. At the value Ω_1^c, the relative importance of these contributions changes and, in consequence, $\mathrm{Re}(\rho_{ba})$ changes sign. The values of $\mathrm{Re}(\rho_{ca})$ and $\mathrm{Re}(\rho_{bc})$ are null in this doubly resonant case. The cancellation of ρ_{ba} corresponding to the interference of two dipole coherences is, in a sense, the complementary phenomena to population trapping,[22,27] because in the latter the population of level $|c\rangle$ and the dipole coherences vanish, ρ_{ba} is maximized; while in the former ρ_{ba} is zero but the other terms are nonvanishing.

For larger values of $|\Omega_1|$, an important contribution to the generated signal comes from ρ_{bc} which is strongly affected by the population of level $|c\rangle$. The behavior of the FWM signal for $|\Omega_1|\geq|\Omega_1^c|$ is illustrated in Fig. 2(d), and the line shape becomes single peaked again.

In the high-power limit (i.e., the Rabi regime) with $\delta_1=0$, the signal has two peaks at $\delta_2=\pm|\Omega_1|$ with linewidth equal to $(\Gamma_{ba})/2$. This is due to the dynamic Stark effect induced by E_1,[10] and was previously studied in connection with other nonlinear phenomena in three-level systems.[14(a),23–26,30,32,35,36] The dominant term in the numerator of Eq. (13) is still the saturated population one, and dominates the observed line shape, showing that in the Rabi regime the wave mixing is not a pure parametric phenomenon.

In Figs. 3(a) and 3(b), the peak position of $|\rho_{ba}|^2$ is shown as a function of $|\Omega_1|$, for $\delta_1=0$. In Fig. 3(a) we make the same choice for the relaxation rates as in Fig. 2. In Fig. 3(b) all the relaxations are equal to Γ_{ca}. In the low-power regime, the splitting has a nonlinear dependence on $|\Omega_1|$, contrary to the result obtained at high intensity. It should be emphasized that this effect is physically different from other line splittings observed in degenerate four-wave mixing, because here there is a true destructive interference, while in Refs. 11 and 12 the line-center signal only vanishes asymptotically, for infinite values of the Rabi frequencies. The position of the antiresonance is dependent on the values of the relaxation rates, so it could occur in the Rabi regime, as shown in Fig. 3(b), in which case the single-peak regime is obtained only for small values of $|\Omega_1|$. This behavior implies that in general a model neglecting the relaxation of the system works only in the limit of very high intensities.

From Eq. (13), it can also be shown that for Rabi frequencies greater than $|\Omega_1^c|$, the value of $|\chi(\omega_4)|^2$ at line center increases until a maximum occurs at the value

$$|\Omega_1|=\{\tfrac{1}{2}\Gamma_c\Gamma_{bc}+\tfrac{1}{8}[16\Gamma_c^2\Gamma_{bc}^2+2(\Gamma_a^{-1}+\Gamma_c^{-1})^{-1}\Gamma_{ca}\Gamma_{cb}(\Gamma_c+2\Gamma_{ba})+4\Gamma_{cb}^2\Gamma_c\Gamma_{ba}]^{1/2}\}^{1/2}\ .$$

For larger amplitudes, the central line signal at $\delta_2=0$ decreases monotonically, as expected.

Figure 4 displays the behavior of $|\chi(\omega_4)|^2$ for the nonresonant case $|\delta_1|\gg\Gamma$. The same parameters as in Fig. 2 were used except for $\delta_1=-40\Gamma_{ca}$ and $|\Omega_2|=0.001\Gamma_{ca}$. The higher peak at $\delta_2=\delta_1+|\Omega_1|^2/\delta_1$ has width Γ_{ba} and is due to the Raman resonance. The one-photon peak appears at $\delta_2=-|\Omega_1|^2/\delta_1$ and has linewidth Γ_{cb}. The Raman resonance appears even for small values of $|\Omega_1|$, while the one-photon peak is induced by saturation effects and appears only if an appreciable amount of population is transferred to level $|c\rangle$.

2. $\Omega_l\to 0$

In the preceding discussion only saturation effects induced by E_1 were considered. Another case of interest is the one with saturation induced by the field E_2 with a weak field E_1. Taking this limit in the equations of the Appendix, the expression for ρ_{ba} reduces to the following simple equations:

$$\rho_{ba}=\frac{\Omega_1\Omega_2^*\rho_{aa}^{(0)}}{\Delta_{ba}\Delta_{ca}-|\Omega_2|^2} \tag{14}$$

$$=\frac{\rho_{aa}^{(0)}\Omega_1\Omega_2^*}{\Delta_{ca}}\,\frac{-1}{\delta_2-\delta_1+\delta_1\left|\dfrac{\Omega_2}{\Delta_{ca}}\right|^2+i\Gamma_{ba}\left\{1+\dfrac{\Gamma_{ca}}{\Gamma_{ba}}\left|\dfrac{\Omega_2}{\Delta_{ca}}\right|^2\right\}}\ . \tag{15}$$

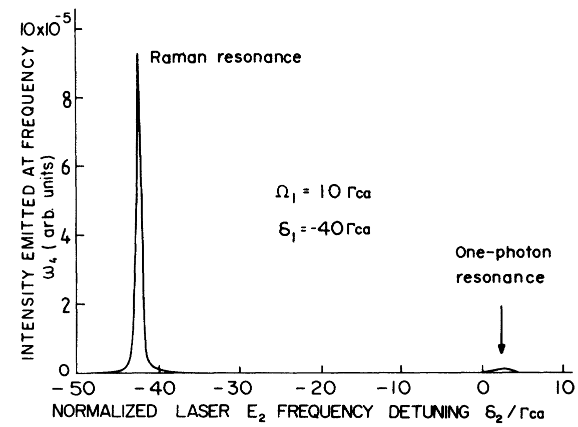

FIG. 4. $|\chi|^2$ as a function of δ_2/Γ_{ca}. Laser E_2 is supposed to have small amplitude ($|\Omega_2|=0.001\Gamma_{ca}$). The detuning of the strong laser E_1 is fixed at $\delta_1=-40\Gamma_{ca}$. Notice the one-photon peak at $\delta_2=-|\Omega_1|^2/\delta_1=2.5\Gamma_{ca}$ and the Raman peak at $\delta_2=\delta_1+|\Omega_1|^2/\delta_1=-42.5\Gamma_{ca}$.

Equations (14) and (15) show that the saturation effect due to E_2 appears only in the denominator $(\Delta_{ba}\Delta_{ca}-|\Omega_2|^2)$. Saturation of population does not occur in this case and the spectrum of ρ_{ba} as a function of δ_2 shows a Lorentzian peak centered at $\delta_2=0$ with linewidth given by $\Gamma_{ba}(1+\Gamma_{ca}^{-1}\Gamma_{ba}^{-1}|\Omega_2|^2)$, for $\delta_1=0$. On the other hand, if E_1 is not on-resonance with the transition $|a\rangle$-$|c\rangle$, the broadening is small and can be neglected although the resonance appears shifted to $\delta_2=\delta_1-|\Omega_2|^2/\delta_1$.

The line-shape behavior of ρ_{ba} as a function of δ_1 can also be obtained from Eq. (14) (see Fig. 5). When E_2 is on-resonance ($\delta_2=0$) and $|\Omega_2|\gg\Gamma$, the Stark effect splits the line into two Lorentzian peaks at $\delta_1=\pm|\Omega_2|$ with linewidth $(\Gamma_{ca}+\Gamma_{ba})/2$ [see Fig. 5(a)]. When E_2 is off-resonance ($|\delta_2|\gg|\Omega_2|\gg\Gamma$), the spectrum has two peaks at $\delta_1^A=\delta_2+|\Omega_2|^2/\delta_2$ and $\delta_1^B=-|\Omega_2|^2/\delta_2$ [see Fig. 5(b), where $\Gamma_{ba}=\Gamma_{ca}$]. The first peak at δ_1^A is due to the Raman resonance that is Stark shifted and has width $\sim\Gamma_{ba}$. The second resonance at δ_1^B corresponds to the one-photon resonance between $|a\rangle$ and $|c\rangle$. In the present case the one-photon peak appears in the FWM spectrum, even for small E_2, because $\rho_{aa}\neq0$.

B. Limit of two strong fields

When both laser fields are strong, the expressions become more complex and higher-order saturation effects appear. Therefore a numerical analysis of Eq. (7) is appropriate in order to study the various effects. The steady-state saturated populations are given in the Appendix.

1. $\delta_1=0$

First, the resonant E_1 field case is considered ($\delta_1=0$) and the study of $|\chi|^2$ as a function of δ_2 is presented. For increasing values of $|\Omega_2|$, line broadening and new structures appear as illustrated in Fig. 6 for $|\Omega_1|=4.0\Gamma_{ca}$. In Fig. 6(a), the two Stark peaks which were discussed in the Sec. III A are shown, the individual peaks saturate, broaden, and shift toward line center, as $|\Omega_2|$ increases. At $|\Omega_2|=0.25\Gamma_{ca}$, a small structure appears in each individual peak, and for increasing values of $|\Omega_2|$, the line shape has four peaks. In Fig. 6(b) we see that the inside peaks merge into only one structure, and at $|\Omega_2|=0.7\Gamma_{ca}$ the line shape has three peaks. For $|\Omega_2|>2.5\Gamma_{ca}$, the central peak decreases [see Fig. 6(c)] and there is a particular value of $|\Omega_2|\sim2.8\Gamma_{ca}$ in which $|\chi|^2$ *is zero* at line center [see Fig. 6(c)]. This occurs due to a competition between the contributions of the two coherences ρ_{bc} and ρ_{ca}, [see Eq. (7)]. At higher values of $|\Omega_2|$, all the structures are merged together and only one peak appears [Fig. 6(d)], because the broadening due to the field E_2 is of the same order as the Stark splitting due to the field E_1. We will now derive an analytical expression giving the relationship between the intensities of E_1 and E_2 which induce, in steady state, the cancellation in Fig. 6(c). We assume as before that

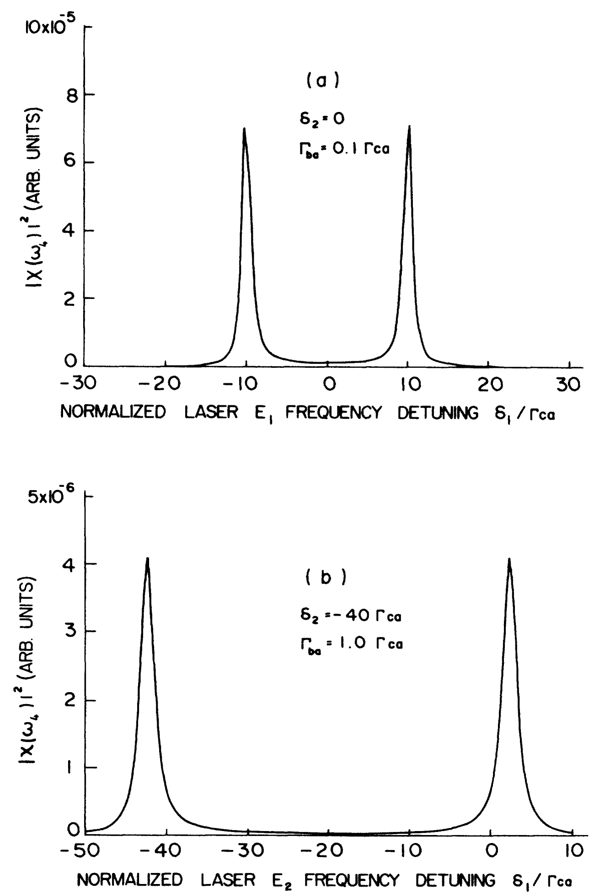

FIG. 5. $|\chi|^2$ as a function of δ_1/Γ_{ca} for strong E_2 field. Parameters as in Fig. 2, except for $|\Omega_1|=0.01\Gamma_{ca}$, $|\Omega_2|=10\Gamma_{ca}$. (a) Resonant case ($\delta_2=0$). (b) Nonresonant case $\delta_2=-40\Gamma_{ca}$. [Note that $\Gamma_{ba}=\Gamma_{ca}$ in (b).]

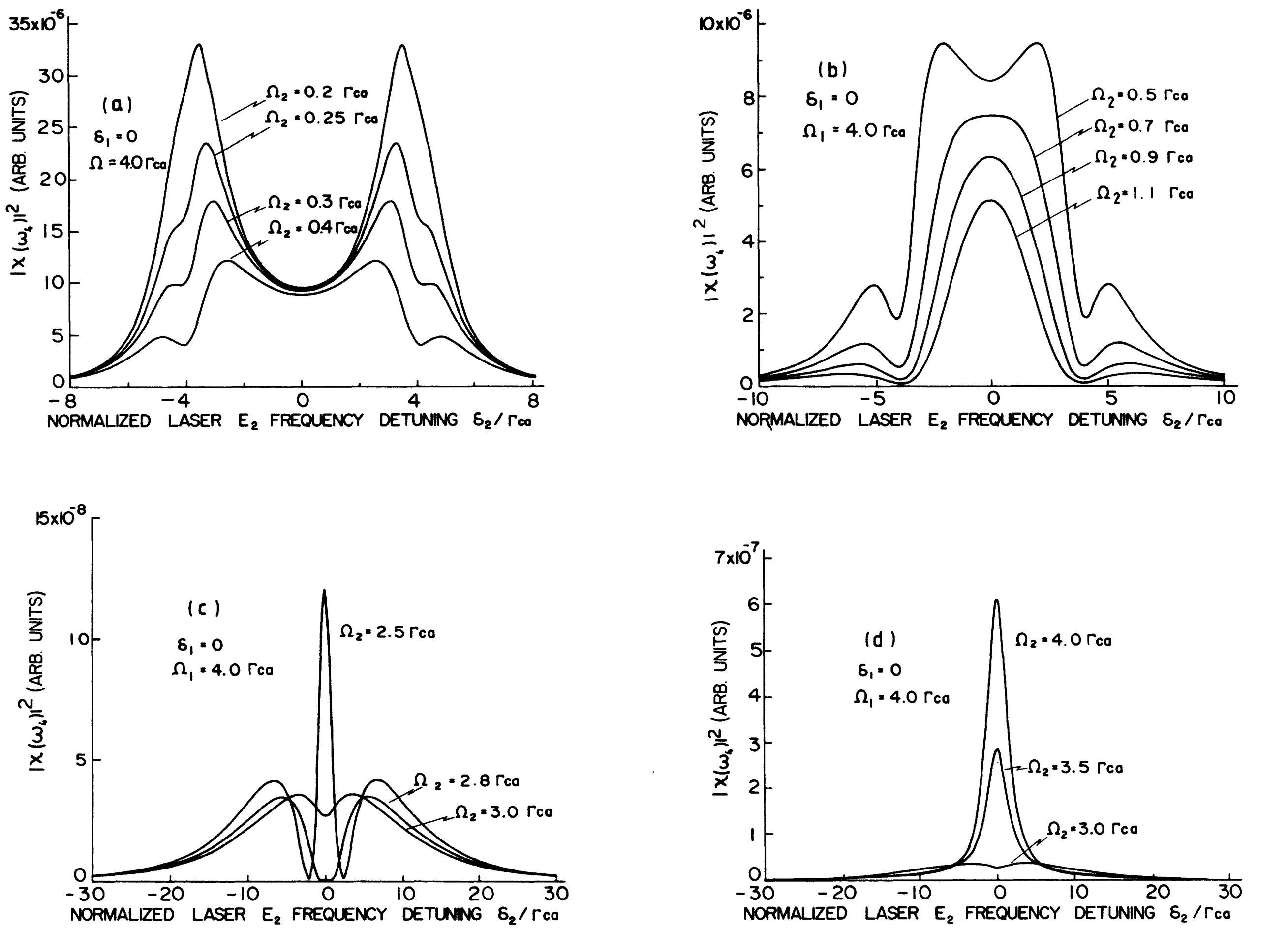

FIG. 6. Evolution of $|\chi|^2$ with $|\Omega_2|$ as a function of δ_2/Γ_{ca}, for $\delta_1=0$, and $|\Omega_1|=4.0\Gamma_{ca}$. Other parameters the same as in Fig. 2.

$\rho_{ii}^{(0)}=0$ ($i=b,c$), and the two fields E_1 and E_2 are on-resonance. The condition for cancellation is given formally by

$$\rho_{ba}=0\ . \tag{16}$$

With the extra condition Eq. (16), Eqs. (6) can be divided into two sets of equations: (6a)–(6e) and (6b)–(6f). For each set, one solution is obtained for each element of the density matrix, e.g., for ρ_{cc}. In order to have both solutions corresponding to the same physical situation, we compare them, obtaining, therefore, the following condition:

$$(\Gamma_b+\Gamma_c)|\Omega_2|^2=\Gamma_b|\Omega_1|^2-\Gamma_c\Gamma_b\Gamma_{cb}\ . \tag{17}$$

Thus, the intensities of the two fields needed to obtain the destructive interference are linearly related. Equation (17) is a generalization of the result given in Sec. III A 1, showing that the destructive interference is not restricted to a point, but occurs along a semi-infinite interval, beginning from the threshold value $|\Omega_1^c|^2$. In a plot of $|\Omega_2|^2$ versus $|\Omega_1|^2$, the intersection with the horizontal axis gives the threshold value of $|\Omega_1^c|^2$ obtained in Sec. III A 1. This gives the value of the saturating field inducing a destructive interference for negligible E_2. Therefore, for $|\Omega_1|\geq(\Gamma_c\Gamma_{cb}/2)^{1/2}$, there is always one value of $|\Omega_2|$, given by Eq. (17), that destroys the Raman coherence. As an example, using the value $|\Omega_1|=4\Gamma_{ca}$ in Eq. (17), we obtain $|\Omega_2|\approx 2.824\Gamma_{ca}$, for the destructive interference to occur, with the values of the relaxation rates as in Fig. 2.

2. $\delta_l \neq 0$

Finally, the evolution with increasing $|\Omega_2|$ of the Raman (or Zeeman) peak as a function of δ_2 is illustrated in Fig. 7, for $\delta_1=-40\Gamma_{ca}$ and $|\Omega_1|=10\Gamma_{ca}$. In Fig. 7(a) we see that for $|\Omega_2|=4\Gamma_{ca}$ the two-photon resonance splits into two peaks. This is because the absorptive component saturates faster than the dispersive one. This faster saturation of the absorption has been reported in other studies of saturation phenomena of nonlinear spectroscopy (see, for example, Refs. 11 and 12). The dispersive behavior is clearly seen in Fig. 7(b).

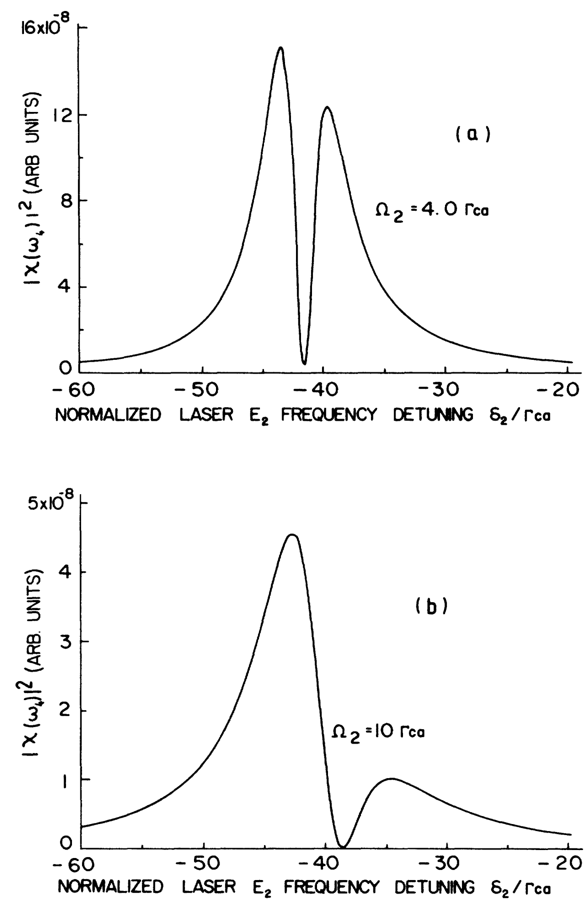

FIG. 7. Raman resonance of $|\chi|^2$ as a function of δ_2/Γ_{ca}. Nonresonant case, with $\delta_1=-40\Gamma_{ca}$, and $|\Omega_1|=10\Gamma_{ca}$. The other parameters are the same as in Fig. 2.

IV. SUMMARY

In this paper, steady-state four-wave mixing has been analyzed for homogeneously broadened four-level systems coupled to three laser fields (E_1 and E_2 of arbitrary intensity and E_3, weak and nonresonant). The relaxation of the system is included phenomenologically in the Bloch equations. The four-wave-mixing signal was shown to be proportional to ρ_{ba}—the coherence between two levels of the same parity. The behavior of $\chi(\omega_4)$—the saturated nonlinear susceptibility—has been studied in detail through an analysis of ρ_{ba}. The present study applies to the whole range of E_1 and E_2 laser intensities extending from the small-intensity regime to the high-intensity Rabi regime.

The dynamic Stark effect due to two of the pump fields and its consequences on the FWM spectrum have been studied in detail. For only one field with increasing intensity, antiresonance behavior resulting from a destructive interference between two one-photon coherences was obtained. This effect leads to a total cancellation of the FWM signal and to a power broadened line shape with a narrow dip in its center. For an even stronger resonant field, the line shape as a function of the detuning of the weak field is Stark split and Stark shifted for a nonresonant excitation. In the case of a nonresonant strong field interacting with an initially nonpopulated transition, the one-photon peak appears at the same order as the two-photon peak does, whereas for the initially populated transition case, the two-photon peak appears for lower intensities of the strong field than the one-photon peak does.

Cases when two fields have saturating intensities were also analyzed. For a resonant interaction, the line shape has a complicated behavior which may show from one up to four resonance peaks. The study of the cancellation of the susceptibility at line center was extended to the regime of two saturating fields. An analytical expression was given relating the intensities of the two fields, for which the destructive interference occurs, to the atomic relaxation rates. For a resonant excitation, we have shown that the Raman resonance is split due to the saturation of the absorptive component of the nonlinear susceptibility. All the predicted spectral lineshapes should be observable in atomic beam experiments or in narrow lines of rare-earth impurities in solids. In both cases, available cw or long-pulse dye lasers are capable of inducing steady-state saturation effects in four-wave-mixing experiments.[14] Further extensions of this work for the case of inhomogeneously broadened systems as well as the transient FWM behavior will be reported in the future.

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APPENDIX

We give here the expressions for the populations ρ_{ii} ($i=a,b,c$), used in Eq. (7), which are obtained by solving Eq. (6) in steady state:

$$\rho_{aa}=\frac{A_3B_1-A_2B_3}{A_1B_1-A_2B_2}\,, \tag{A1}$$

$$\rho_{bb}=\frac{A_1B_3-A_3B_2}{A_1B_2-A_2B_2}\,, \tag{A2}$$

$$\rho_{cc}=P-\Gamma_c^{-1}(\Gamma_a\rho_{aa}+\Gamma_b\rho_{bb})\,, \tag{A3}$$

where the parameter P is related to the incoherent pump of population, and given by the conservation law

$$\Gamma_cP\equiv\Gamma_a\rho_{aa}^{(0)}+\Gamma_b\rho_{bb}^{(0)}+\Gamma_c\rho_{cc}^{(0)}$$
$$=\Gamma_a\rho_{aa}+\Gamma_b\rho_{bb}+\Gamma_c\rho_{cc}\,, \tag{A4}$$

and the other coefficients are

$$A_1=\Gamma_a[1+W_{ca}(\Gamma_a^{-1}+\Gamma_c^{-1})]+W_{ba} \ , \tag{A5}$$

$$A_2=W_{ca}\Gamma_b\Gamma_c^{-1}-W_{ba} \ , \tag{A6}$$

$$A_3=\Gamma_a\rho_{aa}^{(0)}+W_{ca}P \ , \tag{A7}$$

$$B_1=\Gamma_b[1+W_{cb}(\Gamma_b^{-1}+\Gamma_c^{-1})] \ W_{ba} \ , \tag{A8}$$

$$B_2=W_{cb}\Gamma_a\Gamma_c^{-1}-W_{ba} \ , \tag{A9}$$

$$B_3=\Gamma_b\rho_{bb}^{(0)}+W_{cb}P \ . \tag{A10}$$

In Eqs. (A5)–(A10), W_{ij} are transitions rates between levels $|i\rangle$ and $|j\rangle$ $(ij=ca,cb,ba)$, given by

$$W_{ca}=|\Omega_1|^2\left|\frac{-i(\Delta_{ba}\Delta_{cb}^*+|\Omega_1|^2-|\Omega_2|^2)}{D}+\text{c.c.}\right| \ , \tag{A11}$$

$$W_{cb}=|\Omega_2|^2\left|\frac{i(\Delta_{ba}\Delta_{ca}+|\Omega_1|^2-|\Omega_2|^2)}{D}+\text{c.c.}\right| \ , \tag{A12}$$

$$W_{ba}=|\Omega_1\Omega_2|^2\left[-\frac{i}{D}+\text{c.c.}\right] \ , \tag{A13}$$

where the generalized Stark denominator is

$$D=\Delta_{ca}\Delta_{cb}^*\Delta_{ba}+\Delta_{ca}|\Omega_1|^2-\Delta_{cb}^*|\Omega_2|^2 \ .$$

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