Quantum theory of multiwave mixing. V. Two-photon two-level model

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We extend our theoretical formalism describing how one strong classical wave and one or two weak quantum-mechanical waves interact in a nonlinear medium to the "two-photon two-level" model. In this model, field transitions between two levels not connected by an electric dipole occur by means of a two-photon transition using off-resonant intermediate levels. This model has many similarities with, as well as significant differences from, the corresponding one-photon two-level model that has been treated in the previous papers in this series. The theory yields the first detailed calculation of the two-photon resonance fluorescence spectrum, as well as the complete set of four coefficients that describe the quantum theory of two-, three-, and four-wave mixing.

I. INTRODUCTION

Because of either selection rules or the unavailability of an appropriate laser frequency, many energy levels cannot be excited by means of a one-photon transition. Such excitations can only be performed by means of a multiphoton transition, and such transitions are now quite common in laser spectroscopy. Multiphoton transitions are also of interest because of their potential applications to isotope separation, laser chemistry, information transmission, and high-power lasers.

In previous papers in this series[1-5] we have derived and applied a theory describing quantum multiwave interactions in a nonlinear two-level medium, in which the levels are connected by an electric dipole. This theory unifies numerous areas of quantum optics—resonance fluorescence, saturation spectroscopy, modulation spectroscopy and phase conjugation. In this paper we extend our theory to the two-photon two-level model developed by a number of authors.[6-9] In this model, two levels not connected by an electric dipole interact with a field at approximately half the transition frequency by means of a two-photon transition. As we demonstrate, the form of the reduced field density operator equation of motion is the same as in the one-photon case, and so all of the results of the previous papers can now be obtained for this type of medium. In particular, we provide the first detailed calculation of the spectrum of two-photon resonance fluorescence[10] and find the set of four coefficients that describe the quantum theory of two, three, and four waves interacting in two-photon two-level media.

The two-photon two-level model is shown in Fig. 1. The dipole matrix element between levels a and b is zero, and the strong pump field frequency ν_2 is approximately one-half the frequency difference $\omega=\omega_a-\omega_b$. Dipole transitions from states a and b to the intermediate levels j are possible, but we assume that these levels are sufficiently far from resonance that they can always be treated to first order and hence they acquire no appreciable population.

Two major differences occur between the one- and two-photon models. First, dynamic Stark shifts of the level frequencies can play an important role in the two-photon case. The physical origin of the Stark shift comes from the frequency shifts of levels a and b induced by virtual transitions to the off-resonant j levels. In the one-photon model this shift can be neglected since such nonresonant interactions are small compared to resonant ones, but for the two-photon model, the shift is of the same magnitude as the other parameters and must be included. Second, the coherence R_{ab} induced between the two levels in the two-photon model does not contribute directly to the polarization of the medium; an additional atom-field interaction is required. These differences cause the algebraic expressions to be significantly more complicated than for the one-photon problem, and the resulting physics has considerably more variety.

The semiclassical multiwave mixing theory for the two-photon two-level model has recently been published by Sargent, Ovadia, and Lu.[11] In that paper the effects of the nonlinear coupling between the two-photon medium and a strong two-photon resonant field on one or two weak probe fields is considered. They thus obtain the two-photon theory of saturation spectroscopy and phase conjugation. The quantum theory presented here allows us to also consider the fluorescence from this system, instead of just the absorption. In addition, it is also possible to study the effects of quantum noise on such processes as phase conjugation and laser instabilities.[12-14] As in the

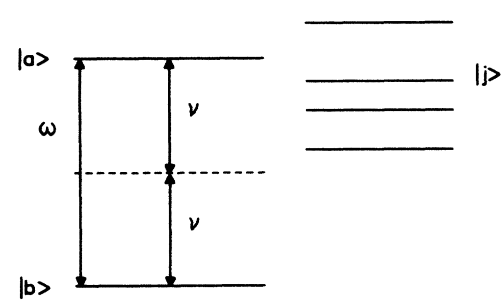

FIG. 1. Two-photon two-level model.

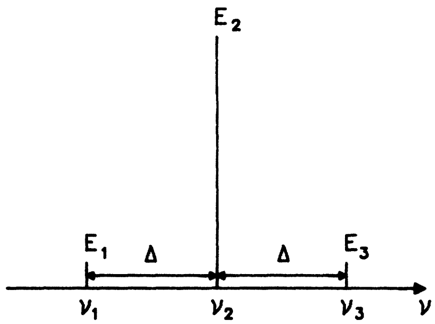

FIG. 2. Three-mode spectrum used for multiwave mixing such as in laser and/or optical bistabilities, phase conjugation, and modulation spectroscopy.

one-photon case, the semiclassical two-photon answers can be computed from our quantum theory, and we show that we obtain the same results.

Following the derivation presented in Refs. 1 and 2, before deriving the quantum-mechanical equations of motion for this model we need to first solve the semiclassical single-mode problem. This is done in Sec. II where we follow the procedure of Ref. 11. In Sec. III the quantum-mechanical model is presented. We follow the procedure of Ref. 2 in which the relevant quantities are expressed in terms of the operators of the quantized fields. Section IV derives the quantum-mechanical equations of motion for the quantized fields. The resulting expressions are considerably more complicated than either those of the semiclassical or the one-photon theory due to the many more interactions. Because of this complexity, in Sec. V we project these equations of motion for the quantum operators onto the atom-field basis states employed in Ref. 1. The equations for the components allow an immediate physical interpretation along the lines of their one-photon counterparts. For simplicity, the equations are then solved for the case of a single sidemode (one quantized field). Section VI discusses the consequences of this solution, one of which is the two-photon resonance fluorescence spectrum. The expression for the fluorescence is rather complicated, and we illustrate our results graphically. In Sec. VII we include a second sidemode placed symmetrically in frequency on the other side of the strong mode, as shown in Fig. 2. This case is applicable to three- and four-wave mixing and to laser instabilities. As we show, many more interactions arise in the double sidemode case, again resulting in effects absent in the one-photon case.

The emphasis in this paper is on the basic theoretical formalism. In Sec. VI we do present illustrations of two-wave mixing phenomena such as the resonance fluorescence spectrum and the associated Rayleigh scattering from this type of media, but we defer most of the discussion and applications of the three- and four-wave mixing interactions to subsequent papers in this series.

II. SEMICLASSICAL SINGLE-MODE SOLUTION

We consider the classical pump field to be of the form

$$E_2(\mathbf{r},t)=\tfrac{1}{2}\mathscr{E}_2(\mathbf{r},t)e^{-i\nu_2 t}+\text{c.c.}\ , \tag{1}$$

where ν_2 is the field frequency (in radians/sec) and where the complex amplitude $\mathscr{E}_2(\mathbf{r},t)$ varies little in a time $1/\nu_2$, but may have rapid spatial variations like $\exp(i\mathbf{K}_2\cdot\mathbf{r})$. This pump field induces the polarization

$$p(\mathbf{r},t)=\tfrac{1}{2}\mathscr{P}(\mathbf{r},t)e^{-i\nu_2 t}+\text{c.c.}\ , \tag{2}$$

where the complex polarization amplitude $\mathscr{P}(\mathbf{r},t)$ also varies little in the time $1/\nu_2$. The polarization for the two-photon two-level medium of Fig. 1 may be from the atomic density operator R. This yields

$$\begin{aligned}p(\mathbf{r},t)&=N\,\mathrm{Tr}(erR)\\&=N\sum_j(\mu_{aj}R_{ja}+\mu_{bj}R_{jb})+\text{c.c.}\ ,\end{aligned} \tag{3}$$

where μ_{aj} is the electric-dipole matrix element between the a and j levels, and R_{ja} is the density-matrix element between j and a. Combining Eqs. (2) and (3) we find

$$\mathscr{P}(\mathbf{r},t)=2N\sum_j(\mu_{aj}R_{ja}+\mu_{bj}R_{jb}+\text{c.c.})e^{i\nu_2 t}\ , \tag{4}$$

where only terms varying little in an optical frequency period $1/\nu_2$ are retained. The equations of motion for the matrix elements of the atomic density operator R are obtained from the usual equation of motion

$$i\dot{R}=[\mathscr{H}_{\text{sc}},R]+\Gamma(R)\ , \tag{5}$$

where the semiclassical Hamiltonian in this case is given by

$$\mathscr{H}_{\text{sc}}=\Omega+\mathscr{V}_{aj}\sigma_a^\dagger+\mathscr{V}_{jb}\sigma_b^\dagger+\mathscr{V}_{ja}\sigma_a+\mathscr{V}_{bj}\sigma_b\ , \tag{6}$$

where Ω, $\sigma_a^\dagger$, and $\sigma_b^\dagger$ are the matrices

$$\Omega=\begin{bmatrix}\omega_a&0&0\\0&\omega_b&0\\0&0&\omega_j\end{bmatrix},\quad \sigma_a^\dagger=\begin{bmatrix}0&0&1\\0&0&0\\0&0&0\end{bmatrix},\quad \sigma_b^\dagger=\begin{bmatrix}0&0&0\\0&0&0\\0&1&0\end{bmatrix}, \tag{7}$$

and where the interaction energies $\mathscr{V}_{aj}$ and $\mathscr{V}_{jb}$ are

$$\mathscr{V}_{aj}=-\frac{\mu_{aj}}{2\hbar}(\mathscr{E}_2e^{-i\nu_2 t}+\mathscr{E}_2^*e^{i\nu_2 t})\ , \tag{8}$$

$$\mathscr{V}_{jb}=-\frac{\mu_{jb}}{2\hbar}(\mathscr{E}_2e^{-i\nu_2 t}+\mathscr{E}_2^*e^{i\nu_2 t})\ , \tag{9}$$

with μ representing the appropriate dipole matrix element. Note that we have not made the rotating-wave approximation since the intermediate j levels are far from resonance. $\Gamma(R)$ is a matrix for relaxation processes. In this work we assume that the upper level a decays at the rate $\Gamma(=1/T_1)$ to the lower level b. In real systems, this decay generally occurs by dipole-allowed, one-photon transitions. We let $\gamma(=1/T_2)$ denote the decay rate of the coherence between levels a and b, and γ_{aj} and γ_{jb} the coherence decay rates between levels a and j and j and b, respectively. Accordingly, the matrix $\Gamma(R)$ becomes

$$\Gamma(R) = -\begin{bmatrix} \Gamma R_{aa} & \gamma R_{ab} & \gamma_{aj} R_{aj} \\ \gamma R_{ba} & -\Gamma R_{aa} & \gamma_{jb} R_{bj} \\ \gamma_{aj} R_{ja} & \gamma_{jb} R_{jb} & 0 \end{bmatrix} . \quad (10)$$

Projecting Eq. (5) onto the atomic basis states we obtain

$$\dot{R}_{ja} = -(\gamma_{ja} + i\omega_{ja}) R_{ja} + \frac{i}{2\hbar}(\mathscr{E}_2 e^{-i\nu_2 t} + \mathscr{E}_2^* e^{i\nu_2 t})(\mu_{ja} R_{aa} + \mu_{jb} R_{ba}) , \quad (11)$$

$$\dot{R}_{jb} = -(\gamma_{jb} + i\omega_{jb}) R_{jb} + \frac{i}{2\hbar}(\mathscr{E}_2 e^{i\nu_2 t} + \mathscr{E}_2^* e^{i\nu_2 t})(\mu_{jb} R_{bb} + \mu_{ja} R_{ab}) , \quad (12)$$

where $\hbar\omega_{ij} = \hbar(\omega_i - \omega_j)$ is the energy difference between levels i and j, and γ_{ij} is the corresponding decay constant.

We integrate Eqs. (11) and (12) to first order in $\mathscr{V}$ without making a rotating-wave approximation, since ν_2 differs substantially from all $\pm\omega_{ja}$ and $\pm\omega_{jb}$. Setting $R_{ba} = \tilde{R}_{ba} e^{2i\nu_2 t}$, where $\tilde{R}_{ba}$ varies little in an optical frequency period, we have

$$R_{ja} = \frac{i}{2\hbar}\int_{-\infty}^{t} dt' (\mathscr{E}_2 e^{-i\nu_2 t} + \mathscr{E}_2^* e^{i\nu_2 t}) \exp[-(\gamma_{ja} + i\omega_{ja})(t - t')](\mu_{ja} R_{aa} + \mu_{jb} R_{ba} e^{2i\nu_2 t'})$$

$$= \frac{1}{2\hbar}\left[\frac{\mathscr{E}_2 e^{-i\nu_2 t}}{\omega_{ja} - \nu_2} + \frac{\mathscr{E}_2^* e^{i\nu_2 t}}{\omega_{ja} + \nu_2}\right]\mu_{ja} R_{aa} + \frac{1}{2\hbar}\left[\frac{\mathscr{E}_2 e^{i\nu_2 t}}{\omega_{ja} + \nu_2} + \frac{\mathscr{E}_2^* e^{3i\nu_2 t}}{\omega_{ja} + 3\nu_2}\right]\mu_{jb}\tilde{R}_{ba} . \quad (13)$$

Since we assume $\omega_{ab} \equiv \omega \simeq 2\nu_2$, we have

$$\omega_{ja} + \nu_2 \simeq \omega_{jb} - \nu_2 , \quad (14)$$

which allows us to replace $\omega_{ja} + 3\nu_2$ in Eq. (13) by $\omega_{jb} + \nu_2$. Similarly integrating Eq. (12), we find

$$R_{jb} = \frac{1}{2\hbar}\left[\frac{\mathscr{E}_2 e^{-i\nu_2 t}}{\omega_{jb} - \nu_2} + \frac{\mathscr{E}_2^* e^{i\nu_2 t}}{\omega_{jb} + \nu_2}\right]\mu_{jb} R_{bb} + \frac{1}{2\hbar}\left[\frac{\mathscr{E}_2 e^{-3i\nu_2 t}}{\omega_{ja} - \nu_2} + \frac{\mathscr{E}_2^* e^{-i\nu_2 t}}{\omega_{jb} - \nu_2}\right]\mu_{ja}\tilde{R}_{ab} . \quad (15)$$

Using Eqs. (13) and (15), we now derive the "two-level" equations of motion for R_{aa}, R_{bb}, and R_{ab} using the two-photon rotating-wave approximation, i.e., we neglect terms like $1/[\gamma + i(\omega + 2\nu_2)]$ compared to $1/[\gamma + i(\omega - 2\nu_2)]$. According to Eq. (5), we have

$$\dot{R}_{ab} = -(\gamma + i\omega) R_{ab} - i\sum_j (\mathscr{V}_{aj} R_{jb} - R_{aj}\mathscr{V}_{jb}) , \quad (16)$$

$$\dot{R}_{aa} = -\Gamma R_{aa} - \sum_j (i\mathscr{V}_{aj} R_{ja} + \text{c.c.}) . \quad (17)$$

For simplicity we take $\dot{R}_{bb} = -\dot{R}_{aa}$, since we assume $R_{jj} = 0$. We find the population difference equation of motion

$$\dot{D} = -2\Gamma R_{aa} - 2\sum_j (i\mathscr{V}_{aj} R_{ja} + \text{c.c.}) = -(D+1)/T_1 - 2\sum_j (i\mathscr{V}_{aj} R_{ja} + \text{c.c.}) , \quad (18)$$

where we write the population difference decay time $1/\Gamma$ as T_1. Substituting the dipole Eqs. (13) and (15) into (16), we have

$$\dot{R}_{ab} = -(\gamma + i\omega + i\omega_s I_2) R_{ab} - i k_{ab}\mathscr{E}_2^2/2 e^{-2i\nu_2 t} D , \quad (19)$$

where the two-photon dimensionless intensity

$$I_2 = |k_{ab}\mathscr{E}_2^2|\sqrt{T_1 T_2} \equiv |\mathscr{E}_2/\mathscr{E}_s|^2 , \quad (20)$$

the two-photon coherent decay time $T_2 \equiv 1/\gamma$, and the Stark shift parameter

$$\omega_s = (k_{bb} - k_{aa})/|k_{ab}|\sqrt{T_1 T_2} . \quad (21)$$

The two-photon coefficients k_{ab}, k_{aa}, and k_{bb} are given by

$$k_{ab} = (1/2\hbar^2)\sum_j \mu_{aj}\mu_{jb}/(\omega_{jb} - \nu_2) , \quad (22)$$

$$k_{aa} = (1/2\hbar^2)\sum_j |\mu_{ja}|^2\omega_{ja}/(\omega_{ja}^2 - \nu_2^2) , \quad (23)$$

$$k_{bb} = (1/2\hbar^2)\sum_j |\mu_{jb}|^2\omega_{jb}/(\omega_{jb}^2 - \nu_2^2) . \quad (24)$$

Similarly substituting Eqs. (13) and (15) into Eqs. (3) and (18), we find, respectively,

$$\mathscr{P} = 4\hbar N[\mathscr{E}_2(k_{aa} R_{aa} + k_{bb} R_{bb}) + \mathscr{E}_2^* k_{ab}^* R_{ab} e^{2i\nu_2 t}] , \quad (25)$$

$$\dot{D} = -(D+1)/T_1 + (i k_{ab}\mathscr{E}_2^2 e^{-2i\nu_2 t} R_{ba} + \text{c.c.}) . \quad (26)$$

As noted by a number of people,[6–9] Eqs. (19) and (26) are the same as those for a one-photon two-level system with the substitutions

$$\omega \to \omega + \omega_s I_2 ; \quad \mu_{21}\mathscr{E}_2/\hbar \to k_{ab}\mathscr{E}_2^2 ; \quad \nu_2 \to 2\nu_2 . \quad (27)$$

For single-frequency operation, we can solve these equations in the rate-equation approximation as done in Ref. 12. Specifically, we assume $\mathscr{E}_2$ and D vary little in the two-photon coherence decay time T_2, allowing Eq. (19) to be formally integrated with the value

$$R_{ab} = -i(k_{ab}\mathscr{E}_2^2/2)\mathscr{D}_2(\omega + \omega_s I - 2\nu_2) D e^{-2i\nu_2 t} , \quad (28)$$

where the complex denominator $\mathscr{D}_2$ is

$$\mathscr{D}_2 = \frac{1}{\gamma + i(\omega + \omega_s I_2 - \nu_2)} . \quad (29)$$

Substituting this into Eq. (26), we have

$$\dot{D} = -(D+1)/T_1 - 2\mathscr{R}D \,, \tag{30}$$

where the rate constant

$$\mathscr{R} = |k_{ab}\mathscr{E}_2^2|^2 \mathscr{L}_2(\omega+\omega_s I_2 - 2\nu_2)/4\gamma$$
$$= \tfrac{1}{2} I_2^2 \mathscr{L}_2(\omega+\omega_s I_2 - 2\nu_2)/T_1 \,, \tag{31}$$

and the Lorentzian

$$\mathscr{L}_2(\Delta_2) = 1/[1+(\Delta_2/\gamma)^2] \,. \tag{32}$$

Solving for D in steady state ($\dot{D}=0$), we have

$$D = -1/(1+I^2\mathscr{L}_2) \,. \tag{33}$$

Substituting this into Eq. (28), we have

$$R_{ab} = i\frac{k_{ab}\mathscr{D}_2\mathscr{E}_2^2}{2}\frac{e^{-2i\nu_2 t}}{1+I_2^2\mathscr{L}_2} \,, \tag{34}$$

where we have left off the frequency dependence on $\mathscr{D}_2$ and $\mathscr{L}_2$ for typographical simplicity. Finally, using Eq. (33) and trace relationship $R_{aa}+R_{bb}=1$, we have

$$R_{aa} = \frac{1}{2}\frac{I_2^2\mathscr{L}_2}{1+I_2^2\mathscr{L}_2} = \frac{f_a}{1+I_2^2\mathscr{L}_2} \,, \tag{35}$$

$$R_{bb} = \frac{1+\frac{1}{2}I_2^2\mathscr{L}_2}{1+I_2^2\mathscr{L}_2} = \frac{f_b}{1+I_2^2\mathscr{L}_2} \,. \tag{36}$$

The assumptions and method used to obtain Eqs. (34)–(36) are again employed in the next sections when the quantum-mechanical model is introduced, and we frequently refer to these results.

III. QUANTUM-MECHANICAL FORMALISM

The derivation of the quantum theory of multiwave mixing in Refs. 1 and 2 resulted in an equation of motion for the reduced field density operator P in terms of the creation and annihilation operators of the quantized fields and four complex coefficients, which we labeled A_1, B_1, C_1, and D_1. In those references we showed the physical meaning of those four coefficients. For example, $A_1+A_1^*$ is the resonance fluorescence spectrum, A_1-B_1 is the complex gain/absorption coefficient of a weak probe in the presence of a strong pump field, and C_1-D_1 is the semiclassical mode coupling coefficient, frequently denoted as $-i\kappa_1^*$. In this section we develop the quantum-mechanical model to derive the coefficients A_1, B_1, C_1, and D_1 for this medium. We solve for these coefficients in later sections.

Specifically, for the problem of resonance fluorescence, we calculate the spontaneous emission spectrum for frequencies ν_1 around the pump frequency ν_2 (see Fig. 2). Referring to Fig. 1, the spontaneous emission arises from the upper level a to the intermediate level j and from the intermediate level j to the lower level b. For each two-photon transition there is at most one spontaneous emission, either from level a to level j or from level j to level b, but not both. We thus neglect the two-photon spontaneous emission, which is negligible for our assumptions. Two-photon transitions also occur solely due to the strong pump field.

We formulate the problem by adopting the approach presented in Ref. 2 basing the calculation in terms of field operators. However, to gain additional insight into the physical problem and to express our results in familiar and meaningful notation, we project our operator equations onto an appropriate set of atom-field states as was done in Ref. 1. This also serves to demonstrate the connection between the approaches of the two references.

We separate our Hamiltonian into three parts, the semiclassical, the field, and the interaction. The semiclassical Hamiltonian is given by Eq. (6). The field Hamiltonian is

$$\mathscr{H}_f = \sum_k \nu_k a_k^\dagger a_k \,, \tag{37}$$

and the interaction Hamiltonian is

$$\mathscr{H}_{\text{int}} = -\left[\frac{\mu_{aj}}{2\hbar}(\sigma_a^\dagger+\sigma_a) + \frac{\mu_{jb}}{2\hbar}(\sigma_b^\dagger+\sigma_b)\right] \times \sum_k (\mathscr{E}_k a_k + \mathscr{E}_k^* a_k^\dagger) \,, \tag{38}$$

where ν_k is the frequency of mode k and $\mathscr{E}_k$ is the field amplitude of mode k, $\sqrt{\hbar\nu_k/\varepsilon_0 V}\,U_k(\mathbf{r})$, where $U_k(\mathbf{r})$ is the complex spatial component of the wave and V is the quantization volume. The σ matrices are given by Eq. (7) and $a_k^\dagger$ and a_k are the creation and annihilation operators for field mode k. The total Hamiltonian $\mathscr{H}$ is

$$\mathscr{H} = \mathscr{H}_{\text{sc}} + \mathscr{H}_f + \mathscr{H}_{\text{int}} \,. \tag{39}$$

We once again do not make the rotating-wave approximation since the intermediate j levels are far from resonance. We define ρ to be the atom-field density operator and this obeys the usual equation of motion

$$\dot{\rho} = -i[\mathscr{H},\rho] + \Gamma(\rho) \,, \tag{40}$$

where $\Gamma(\rho)$ is given by Eq. (10) with ρ replacing R. The aforementioned reduced field operator P for the quantized modes is defined by tracing ρ over the atomic states, $P=\text{Tr}(\rho)_{\text{atom}}$. From Eq. (40) this has the equation of motion

$$\dot{P} = -i[\mathscr{H}_f,P] - i\,\text{Tr}\sum_k \frac{\mu_{aj}}{2\hbar}[\mathscr{E}_k\phi_k(\sigma_a^\dagger+\sigma_a) + \mathscr{E}_k^*\phi^+k(\sigma_a^\dagger+\sigma_a)] + \frac{\mu_{bj}}{2\hbar}[\mathscr{E}_k\phi_k(\sigma_b^\dagger+\sigma_b) + \mathscr{E}_k^*\phi^+k(\sigma_b^\dagger+\sigma_b)] + \text{H.c.} \,, \tag{41}$$

where $\phi_k \equiv a_k\rho$ and $\phi_k^+ \equiv a_k^\dagger\rho$. Note that ϕ_k and ϕ_k^+ are operators of both the field and the atoms and are not Hermitian adjoints of one another.

Our method combines the techniques of Ref. 2 and the semiclassical theory of the previous section. From Eq. (40) we find the equations of motion for ϕ_k and ϕ_k^+ and then solve for their slowly varying components using the two-photon rotating-wave approximation, just as we did in the semiclassical problem of Sec. II for the atomic density-matrix elements R_{aa}, R_{bb}, and R_{ab}. Since we assume these elements vary little in atomic lifetimes, we may set their time derivatives to zero in steady state and obtain four coupled algebraic equations that are solved simultaneously.

IV. QUANTUM EQUATIONS OF MOTION

We begin by considering a single quantized sidemode. This corresponds in Fig. 2 to the modes at frequencies ν_1 and ν_2. We may then drop the k subscript from the ϕ^+ and ϕ operators, and we initially deal with the ϕ^+ operator. The solution for the ϕ operator is similar. From the equation of motion (40) and the definition of ϕ^+ we find

$$i\dot{\phi}^+ = -\nu_1 a^\dagger \rho + \frac{\mu_{ja}\mathscr{E}_1}{2\hbar}(\sigma_a^\dagger+\sigma_a)+\frac{\mu_{jb}\mathscr{E}_1}{2\hbar}(\sigma_b^\dagger+\sigma_b) + [\mathscr{H},\phi^+]+i\Gamma(\phi^+) \ . \quad (42)$$

This operator equation can be projected onto the atomic basis states to obtain the equations of motion for $\dot{\phi}^+_{aa}$, etc. These elements contain rapidly varying components and so we do not set $\dot{\phi}^+=0$. From Eq. (41) we find we need the following components of ϕ^+:

$$\mathrm{Tr}(\sigma_a\phi^+)=\phi^+_{aj} \quad (43a)$$

$$\mathrm{Tr}(\sigma_a^\dagger\phi^+)=\phi^+_{ja} \quad (43b)$$

$$\mathrm{Tr}(\sigma_b\phi^+)=\phi^+_{jb} \quad (43c)$$

$$\mathrm{Tr}(\sigma_b^\dagger\phi^+)=\phi^+_{bj} \ . \quad (43d)$$

For example, from Eq. (42) we find the equation of motion for ϕ^+_{aj} to be

$$i\dot{\phi}^+_{aj} = -(i\gamma_{ja}+\omega_{ja}+\nu_1)\phi^+_{aj} - \mathscr{V}_{aj}\phi^+_{aa} - \mathscr{V}_{jb}\phi^+_{ab} + \frac{\mu_{ja}\mathscr{E}_1}{2\hbar}a^\dagger\rho_{aa}a + \frac{\mu_{ja}\mathscr{E}_1^*}{2\hbar}a^\dagger\rho_{aa}a^\dagger + \frac{\mu_{jb}\mathscr{E}_1}{2\hbar}a^\dagger\rho_{ab}a + \frac{\mu_{jb}\mathscr{E}_1^*}{2\hbar}a^\dagger\rho_{ab}a^\dagger \quad (44)$$

To integrate Eq. (44), we must first determine the time dependences of each term, just as was done in the semiclassical theory for R_{ja}, Eq. (13). Since we are making the two-photon rotating-wave approximation with respect to levels a and b, the matrix elements ϕ^+_{aa}, ϕ^+_{bb}, ϕ^+_{ab}, and ϕ^+_{ba} may be written in an interaction picture rotating at the strong field frequency ν_2. We find from Eq. (42) that this can be done with the transformations

$$\phi^+_{ab}=\widetilde{\phi}^+_{ab}e^{-i\nu_2 t} \quad (45a)$$

$$\phi^+_{aa}=\widetilde{\phi}^+_{aa}e^{i\nu_2 t} \quad (45b)$$

$$\phi^+_{ab}=\widetilde{\phi}^+_{bb}e^{i\nu_2 t} \quad (45c)$$

$$\phi^+_{ba}=\widetilde{\phi}^+_{ba}e^{3i\nu_2 t} \ , \quad (45d)$$

where the tilde represents the slowly varying quantity in our interaction picture. Note that Eq. (44) contains terms like $a^\dagger\rho_{aa}a^\dagger$. As shown in Ref. 2, terms such as this give rise to the C_1 and D_1 coefficients and hence it is worthwhile to retain these. We note that these coupled mode terms $a^\dagger\rho_{aa}a^\dagger$ really arise from expressions such as $\mathscr{E}_1^*\mathscr{E}_3^* e^{i(\nu_1+\nu_3)t}$, and thus the time dependence of these is proportional to $e^{2i\nu_2 t}$. The matrix elements ρ_{aa}, ρ_{ab}, etc., can be expressed in terms of the semiclassical results and the field operator P. For example, $\rho_{ab}=PR_{ab}=P\widetilde{R}_{ab}e^{-2i\nu_2 t}$. This gives the time dependences of these terms. Including all of these in Eq. (44), it is then possible to integrate it just as for R_{ja} and R_{jb} in Eqs. (13) and (15) to yield

$$\phi^+_{aj} = \frac{\mu_{ja}\mathscr{E}_2\widetilde{\phi}^+_{aa}+\mu_{jb}\mathscr{E}_2\widetilde{\phi}^+_{ab}+\mu_{ja}\mathscr{E}_1 a^\dagger\rho_{aa}a+\mu_{jb}\mathscr{E}_1^* a^\dagger P a^\dagger\widetilde{R}_{ab}}{2\hbar(\omega_{ja}+\nu_2)} + \frac{\mu_{ja}\mathscr{E}_2^*\widetilde{\phi}^+_{aa}+\mu_{ja}\mathscr{E}_1^* a^\dagger\rho_{aa}a^\dagger}{2\hbar(\omega_{ja}-\nu_2)}e^{2i\nu_2 t} + \frac{\mu_{jb}\mathscr{E}_2\widetilde{\phi}^+_{ab}+\mu_{jb}\mathscr{E}_1 a^\dagger P a\widetilde{R}_{ab}}{2\hbar(\omega_{ja}+3\nu_2)}e^{-2i\nu_2 t} \ , \quad (46)$$

where the γ_{ja} terms have been dropped since they are small compared to $\omega_{ja}-\nu_2$ and where we approximate $\nu_1\simeq\nu_2$ for the same reason. Similarly, we may integrate the equations of motion for the other components of ϕ^+ in the $\dot{P}$ equation, ϕ^+_{ja}, ϕ^+_{bj}, and ϕ^+_{jb}. We find

$$\phi^+_{ja} = \frac{\mu_{ja}\mathscr{E}_2^*\widetilde{\phi}^+_{aa}+\mu_{jb}\mathscr{E}_2\widetilde{\phi}^+_{ba}+\mu_{ja}\mathscr{E}_1^* a^\dagger a^\dagger\rho_{aa}+\mu_{jb}\mathscr{E}_1 a^\dagger a P\widetilde{R}_{ba}}{2\hbar(\omega_{ja}+\nu_2)}e^{2i\nu_2 t} + \frac{\mu_{ja}\mathscr{E}_2\widetilde{\phi}^+_{aa}+\mu_{ja}\mathscr{E}_1 a^\dagger a\rho_{aa}}{2\hbar(\omega_{ja}-\nu_2)} + \frac{\mu_{jb}\mathscr{E}_2^*\widetilde{\phi}^+_{ba}+\mu_{jb}\mathscr{E}_1^* a^\dagger a^\dagger P\widetilde{R}_{ba}}{2\hbar(\omega_{ja}+3\nu_2)}e^{4i\nu_2 t} \ , \quad (47)$$

$$\phi^+_{bj} = \frac{\mu_{jb}\mathscr{E}_2^*\widetilde{\phi}^+_{bb}+\mu_{jb}\mathscr{E}_2\widetilde{\phi}^+_{ba}+\mu_{ja}\mathscr{E}_1 a^\dagger P a\widetilde{R}_{ba}+\mu_{jb}\mathscr{E}_1^* a^\dagger\rho_{bb}a^\dagger}{2\hbar(\omega_{jb}-\nu_2)}e^{2i\nu_2 t} + \frac{\mu_{jb}\mathscr{E}_2\widetilde{\phi}^+_{bb}+\mu_{jb}\mathscr{E}_1 a^\dagger\rho_{bb}a}{2\hbar(\omega_{jb}+\nu_2)} + \frac{\mu_{ja}\mathscr{E}_2^*\widetilde{\phi}^+_{ba}+\mu_{ja}\mathscr{E}_1^* a^\dagger P a^\dagger\widetilde{R}_{ba}}{2\hbar(\omega_{ja}-3\nu_2)}e^{4i\nu_2 t} \ , \quad (48)$$

$$\phi_{jb}^{+}=\frac{\mu_{jb}\mathscr{E}_2\tilde{\phi}_{bb}^{+}+\mu_{ja}\mathscr{E}_2^{*}\tilde{\phi}_{ab}^{+}+\mu_{jb}\mathscr{E}_1a^{\dagger}a\rho_{bb}+\mu_{ja}\mathscr{E}_1^{*}a^{\dagger}a^{\dagger}P\tilde{R}_{ab}}{2\hbar(\omega_{ja}-\nu_2)}+\frac{\mu_{jb}\mathscr{E}_2^{*}\tilde{\phi}_{bb}^{+}+\mu_{jb}\mathscr{E}_1^{*}a^{\dagger}a^{\dagger}\rho_{bb}}{2\hbar(\omega_{ja}+\nu_2)}e^{2i\nu_2 t}+\frac{\mu_{ja}\mathscr{E}_2\tilde{\phi}_{ab}^{+}+\mu_{ja}\mathscr{E}_1a^{\dagger}aP\tilde{R}_{ab}}{2\hbar(\omega_{ja}-3\nu_2)}e^{-2i\nu_2 t}\ . \tag{49}$$

We wish to derive the equations of motion for the slowly varying components of ϕ as defined by Eqs. (45). We proceed by again using Eq. (42) to obtain the equations of motion for ϕ_{aa}^{+}, ϕ_{ab}^{+}, ϕ_{ba}^{+}, and ϕ_{bb}^{+}. For example, the equation of motion for ϕ_{aa}^{+} is

$$i\dot{\phi}_{aa}^{+}=-(\nu_1+i\Gamma)\phi_{aa}^{+}+\mathscr{V}_{aj}(\phi_{jb}^{+}-\phi_{aj}^{+})-1/(2\hbar)[\mu_{ja}\mathscr{E}_1a^{\dagger}a\rho_{ja}-\mu_{ja}\mathscr{E}_1a^{\dagger}\rho_{aj}a+\mu_{ja}\mathscr{E}_1^{*}(a^{\dagger}a^{\dagger}\rho_{ja}+a^{\dagger}\rho_{aj}a^{\dagger})e^{2i\nu_2 t}]\ . \tag{50}$$

We then substitute the expressions for $\mathscr{V}_{aj},\phi_{jb}^{+},\phi_{aj}^{+},\rho_{ja}=PR_{ja}$ from Eqs. (8), (49), (47), and (13) into Eq. (50) and retain only those terms varying like $e^{i\nu_2 t}$. We then use Eq. (45a) to determine the equation of motion for $\tilde{\phi}_{aa}^{+}$. We find, after some algebra,

$$\begin{aligned}i\dot{\tilde{\phi}}_{aa}^{+}={}&-(i\Gamma-\Delta)\tilde{\phi}_{aa}^{+}+(k_{ab}/2)(\mathscr{E}_2^{*2}\tilde{\phi}_{ab}^{+}-\mathscr{E}_2^{2}\tilde{\phi}_{ba}^{+})\\&+k_{aa}\mathscr{E}_1\mathscr{E}_2^{*}R_{aa}(a^{\dagger}Pa-a^{\dagger}aP)\\&-k_{ab}\mathscr{E}_1\mathscr{E}_2\tilde{R}_{ba}a^{\dagger}aP+k_{ab}\mathscr{E}_1^{*}\mathscr{E}_2^{*}a^{\dagger}Pa^{\dagger}\tilde{R}_{ab}\\&+k_{aa}\mathscr{E}_1^{*}\mathscr{E}_2R_{aa}(a^{\dagger}Pa^{\dagger}-a^{\dagger}a^{\dagger}P)\ ,\end{aligned} \tag{51}$$

where $\Delta=\nu_2-\nu_1$ as in the previous papers. This procedure is then repeated for the other three components $\tilde{\phi}_{ab}^{+}$, $\tilde{\phi}_{ba}^{+}$, and $\tilde{\phi}_{bb}^{+}$. This yields

$$\begin{aligned}i\dot{\tilde{\phi}}_{ab}^{+}={}&(\Delta_2+\Delta+\omega_sI_2-i\gamma)\tilde{\phi}_{ab}^{+}+(k_{ab}/2)\mathscr{E}_2^{2}(\tilde{\phi}_{aa}^{+}-\tilde{\phi}_{bb}^{+})\\&+k_{ab}\mathscr{E}_1\mathscr{E}_2(a^{\dagger}PaR_{aa}-a^{\dagger}aPR_{bb})\\&-\mathscr{E}_1\mathscr{E}_2^{*}\tilde{R}_{ab}(k_{aa}a^{\dagger}aP-k_{bb}a^{\dagger}Pa)\\&-\mathscr{E}_1^{*}\mathscr{E}_2\tilde{R}_{ab}(k_{aa}a^{\dagger}a^{\dagger}P-k_{bb}a^{\dagger}Pa^{\dagger})\ ,\end{aligned} \tag{52}$$

$$\begin{aligned}i\dot{\tilde{\phi}}_{ba}^{+}={}&-(\Delta_2-\Delta+\omega_sI_2+i\gamma)\tilde{\phi}_{ba}^{+}+(k_{ab}/2)\mathscr{E}_2^{*2}(\tilde{\phi}_{bb}^{+}-\tilde{\phi}_{aa}^{+})\\&-\mathscr{E}_1\mathscr{E}_2^{*}\tilde{R}_{ba}(k_{bb}a^{\dagger}aP-k_{aa}aPa^{\dagger})\\&+\mathscr{E}_1^{*}\mathscr{E}_2\tilde{R}_{ba}(k_{aa}a^{\dagger}Pa^{\dagger}-k_{bb}a^{\dagger}a^{\dagger}P)\\&+k_{ab}\mathscr{E}_1^{*}\mathscr{E}_2^{*}(a^{\dagger}Pa^{\dagger}R_{bb}-a^{\dagger}a^{\dagger}PR_{aa})\ ,\end{aligned} \tag{53}$$

$$\begin{aligned}i\dot{\tilde{\phi}}_{bb}^{+}={}&i\Gamma\tilde{\phi}_{aa}^{+}+\Delta\tilde{\phi}_{bb}^{+}-(k_{ab}/2)(\mathscr{E}_2^{*2}\tilde{\phi}_{ab}^{+}-\mathscr{E}_2^{2}\tilde{\phi}_{ba}^{+})\\&+k_{bb}\mathscr{E}_1\mathscr{E}_2^{*}R_{bb}(a^{\dagger}Pa-a^{\dagger}aP)\\&+k_{ab}\mathscr{E}_1\mathscr{E}_2\tilde{R}_{ba}a^{\dagger}Pa-k_{ab}\mathscr{E}_1^{*}\mathscr{E}_2^{*}a^{\dagger}a^{\dagger}P\tilde{R}_{ab}\\&+k_{bb}\mathscr{E}_1^{*}\mathscr{E}_2R_{bb}(a^{\dagger}Pa^{\dagger}-a^{\dagger}a^{\dagger}P)\ .\end{aligned} \tag{54}$$

Because $\tilde{\phi}^{+}$ decays at the rate Γ in our interaction picture and hence rapidly reaches a steady state compared with the reduced field operator P, we solve Eqs. (51)–(54) in steady state. We then have four equations for the four unknowns ϕ_{aa}^{+}, $\tilde{\phi}_{ab}^{+}$, $\tilde{\phi}_{ba}^{+}$, and $\tilde{\phi}_{bb}^{+}$.

We now repeat this procedure for the operator ϕ. In this case the integrated equations of motion for the components ϕ_{ja}, ϕ_{aj}, ϕ_{jb}, and ϕ_{bj} are

$$\phi_{ja}=\frac{\mu_{ja}\mathscr{E}_2^{*}\tilde{\phi}_{aa}+\mu_{jb}\mathscr{E}_2\tilde{\phi}_{ba}+\mu_{ja}\mathscr{E}_1^{*}aa^{\dagger}\rho_{aa}+\mu_{jb}\mathscr{E}_1aaP\tilde{R}_{ba}}{2\hbar(\omega_{ja}+\nu_2)}+\frac{\mu_{ja}\mathscr{E}_2\tilde{\phi}_{aa}+\mu_{ja}\mathscr{E}_1aa\rho_{aa}}{2\hbar(\omega_{ja}-\nu_2)}e^{-2i\nu_2 t}+\frac{\mu_{jb}\mathscr{E}_2^{*}\tilde{\phi}_{ba}+\mu_{jb}\mathscr{E}_1^{*}aa^{\dagger}P\tilde{R}_{ba}}{2\hbar(\omega_{ja}+3\nu_2)}e^{2i\nu_2 t}\ , \tag{55}$$

$$\phi_{aj}=\frac{\mu_{ja}\mathscr{E}_2\tilde{\phi}_{aa}+\mu_{jb}\mathscr{E}_2^{*}\tilde{\phi}_{ab}+\mu_{ja}\mathscr{E}_1a\rho_{aa}a+\mu_{jb}\mathscr{E}_1^{*}aPa^{\dagger}\tilde{R}_{ab}}{2\hbar(\omega_{ja}+\nu_2)}e^{-2i\nu_2 t}+\frac{\mu_{ja}\mathscr{E}_2^{*}\tilde{\phi}_{aa}+\mu_{ja}\mathscr{E}_1^{*}a\rho_{aa}a^{\dagger}}{2\hbar(\omega_{ja}-\nu_2)}+\frac{\mu_{jb}\mathscr{E}_2\tilde{\phi}_{ab}+\mu_{jb}\mathscr{E}_1aPa\tilde{R}_{ab}}{2\hbar(\omega_{ja}+3\nu_2)}e^{-4i\nu_2 t}\ , \tag{56}$$

$$\phi_{jb}=\frac{\mu_{jb}\mathscr{E}_2\tilde{\phi}_{bb}+\mu_{ja}\mathscr{E}_2^{*}\tilde{\phi}_{ab}+\mu_{ja}\mathscr{E}_1^{*}aa^{\dagger}P\tilde{R}_{ab}+\mu_{jb}\mathscr{E}_1aa\rho_{bb}}{2\hbar(\omega_{ja}-\nu_2)}e^{-2i\nu_2 t}+\frac{\mu_{ja}\mathscr{E}_2\tilde{\phi}_{ab}+\mu_{ja}\mathscr{E}_1aaP\tilde{R}_{ab}}{2\hbar(\omega_{ja}-3\nu_2)}e^{-4i\nu_2 t}+\frac{\mu_{jb}\mathscr{E}_2^{*}\tilde{\phi}_{bb}+\mu_{jb}\mathscr{E}_1^{*}aa^{\dagger}\rho_{bb}}{2\hbar(\omega_{jb}+\nu_2)}\ , \tag{57}$$

$$\phi_{bj}=\frac{\mu_{jb}\mathscr{E}_2^*\tilde{\phi}_{bb}+\mu_{ja}\mathscr{E}_2\tilde{\phi}_{ba}+\mu_{jb}\mathscr{E}_1^*a\rho_{bb}a^\dagger+\mu_{ja}\mathscr{E}_1a^\dagger Pa^\dagger\tilde{R}_{ba}}{2\hbar(\omega_{ja}-\nu_2)}$$
$$+\frac{\mu_{jb}\mathscr{E}_2\tilde{\phi}_{bb}+\mu_{jb}\mathscr{E}_1a\rho_{bb}a}{2\hbar(\omega_{ja}+\nu_2)}e^{-2i\nu_2t}+\frac{\mu_{ja}\mathscr{E}_2^*\tilde{\phi}_{ba}+\mu_{ja}\mathscr{E}_1^*aPa^\dagger\tilde{R}_{ba}}{2\hbar(\omega_{ja}-3\nu_2)}e^{2i\nu_2t}, \tag{58}$$

where again the γ_{ja} and γ_{jb} terms have been dropped since they are small compared to ω_{ja} and where we approximate $\nu_1\simeq\nu_2$. Equations (55)—(58) are analogous to Eqs. (46)—(49). We again choose the appropriate time dependences from these equations and obtain the slowly varying components of ϕ in our interaction picture. The equations of motion for these components are

$$i\dot{\tilde{\phi}}_{aa}=-(i\Gamma+\Delta)\tilde{\phi}_{aa}+(k_{ab}/2)(\mathscr{E}_2^{*2}\tilde{\phi}_{ab}-\mathscr{E}_2^2\tilde{\phi}_{ba})+k_{aa}\mathscr{E}_1^*\mathscr{E}_2R_{aa}(aPa^\dagger-aa^{\dagger P})+k_{ab}\mathscr{E}_1^*\mathscr{E}_2^*\tilde{R}_{ab}aPa^\dagger$$
$$-k_{ab}\mathscr{E}_1\mathscr{E}_2aaP\tilde{R}_{ba}+k_{aa}\mathscr{E}_1\mathscr{E}_2^*R_{aa}(aPa-aaP), \tag{59}$$

$$i\dot{\tilde{\phi}}_{ba}=(\Delta-\Delta_2-\omega_sI_2-i\gamma)\tilde{\phi}_{ba}-(k_{ab}/2)\mathscr{E}_2^{*2}(\tilde{\phi}_{aa}-\tilde{\phi}_{bb})+k_{ab}\mathscr{E}_1^*\mathscr{E}_2^*(aPa^\dagger R_{bb}-aa^\dagger PR_{aa})$$
$$-\mathscr{E}_1^*\mathscr{E}_2\tilde{R}_{ba}(k_{bb}aa^\dagger P-k_{aa}aPa^\dagger)+\mathscr{E}_1\mathscr{E}_2^*\tilde{R}_{ba}(k_{aa}aPa-k_{bb}aaP), \tag{60}$$

$$i\dot{\tilde{\phi}}_{ab}=(\Delta_2-\Delta+\omega_sI_2-i\gamma)\tilde{\phi}_{ab}+(k_{ab}/2)\mathscr{E}_2^2(\tilde{\phi}_{aa}-\tilde{\phi}_{bb})+\mathscr{E}_1^*\mathscr{E}_2\tilde{R}_{ab}(k_{bb}aPa^\dagger-k_{aa}aa^\dagger P)$$
$$-\mathscr{E}_1\mathscr{E}_2^*\tilde{R}_{ab}(k_{aa}aaP-k_{bb}aPa)-k_{ab}\mathscr{E}_1\mathscr{E}_2(aaPR_{bb}-aPaRP_{aa}), \tag{61}$$

$$i\dot{\tilde{\phi}}_{bb}=i\Gamma\tilde{\phi}_{22}-\Delta\tilde{\phi}_{11}-(k_{ab}/2)(\mathscr{E}_2^{*2}\tilde{\phi}_{ab}-\mathscr{E}_2^2\tilde{\phi}_{ba})+k_{bb}\mathscr{E}_1^*\mathscr{E}_2R_{bb}(aPa^\dagger-aa^\dagger P)$$
$$-k_{ab}\mathscr{E}_1^*\mathscr{E}_2^*\tilde{R}_{ab}aa^\dagger P+k_{ab}\mathscr{E}_1\mathscr{E}_2aPa\tilde{R}_{ba}+k_{bb}\mathscr{E}_1\mathscr{E}_2^*R_{bb}(aPa-aaP). \tag{62}$$

Equations (59)—(62) may be solved in steady state just as Eqs. (51)—(54). Before attempting this, we first rewrite Eq. (41) in terms of the interaction picture components of ϕ^+ and ϕ. Substituting these components into the slowly varying terms of Eqs. (47)—(50) and Eqs. (55)—(58) yields the $\dot{P}$ equation

$$i\dot{P}=\Bigg\{\Bigg[-k_{aa}\mathscr{E}_1^*\mathscr{E}_2(\tilde{\phi}_{aa}^+-\tilde{\phi}_{aa}^\dagger)-k_{ab}\mathscr{E}_1^*\mathscr{E}_2^*(\tilde{\phi}_{ab}^+-\tilde{\phi}_{ba}^\dagger)-k_{bb}\mathscr{E}_1^*\mathscr{E}_2(\tilde{\phi}_{bb}^+-\tilde{\phi}_{bb}^\dagger)$$
$$-\sum_j\left|\frac{\mu_{ja}\mathscr{E}_1}{2\hbar}\right|^2R_{aa}\left[\frac{a_1^\dagger a_1P-a_1Pa_1^\dagger}{\omega_{ja}-\nu_2}+\frac{a_1^\dagger Pa_1-Pa_1a_1^\dagger}{\omega_{ja}+\nu_2}\right]$$
$$-\sum_j\left|\frac{\mu_{jb}\mathscr{E}_1}{2\hbar}\right|^2R_{bb}\left[\frac{a_1^\dagger a_1P-a_1Pa_1^\dagger}{\omega_{jb}-\nu_2}+\frac{a_1^\dagger Pa_1-Pa_1a_1^\dagger}{\omega_{jb}+\nu_2}\right]$$
$$-(k_{ab}\mathscr{E}_1^*\mathscr{E}_3^*/2)\tilde{R}_{ab}(a_1^\dagger Pa_3^\dagger+a_1^\dagger Pa_3^\dagger-a_3^\dagger Pa_1^\dagger-Pa_1^\dagger a_3^\dagger)\Bigg]+(1\leftrightarrow3)\Bigg\}-\text{H.c.} \tag{63}$$

V. SINGLE SIDEMODE SOLUTION OF THE EQUATIONS OF MOTION

In this section we project the operator equations Eqs. (51)—(54) and Eqs. (59)—(62) onto the basis states $|n_1n_3\rangle$. This gives us equations of motion for the components of ρ in terms of the atom-field states of Ref. 1. Due to the complexity of our equations, in this section we limit our analysis to the single sidemode solution. The coupled mode terms such as $a^\dagger\rho a^\dagger$ are considered in Sec. VII. Consider the four atom-field states depicted in Fig. 3. The states are labeled for numerical simplicity. For example, the state $|5\rangle=|an_1n_2\rangle$ means that the medium is in the upper level a, the quantized field 1 has n_1 photons, and the classical field 2 has n_2 photons. Because n_2 is treated classically, n_2 is large so that $n_2\simeq n_2+1$. The levels are connected by means of two-photon transitions

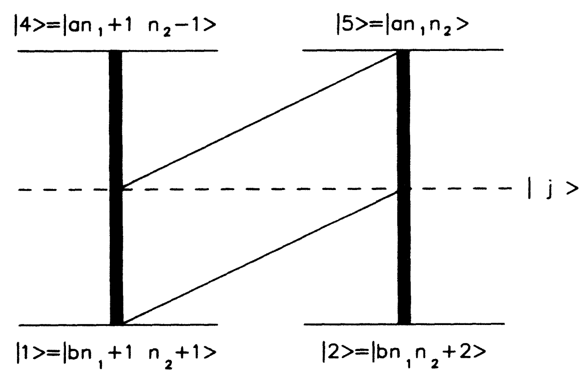

FIG. 3. Two-photon four-level atom-field energy level diagram valid for single-sidemode interactions. Extra levels enter with two sidemodes as shown in Fig. 10.

through the intermediate atomic level j. In particular, the strong pump field 2 connects levels $|1\rangle$ with $|4\rangle$, and $|2\rangle$ with $|5\rangle$.

Spontaneous emission can occur in two ways: from level $|5\rangle$ to the intermediate level j and from the intermediate level j to the level $|1\rangle$. These transitions are shown in Fig. 3 by the lines slanting down to the left. Figure 3 is similar to the one-photon diagram (see Fig. 3 in Ref. 1). For the one-photon problem, the transition between the upper and lower levels is accomplished in one step, whereas in the two-photon problem two steps are required. This leads to the appearance of new interaction potentials as compared to the one-photon problem. From Fig. 3, spontaneous emission occurs when n_1 becomes n_1+1. In the one-photon case, this always occurs by means of a transition from level $|5\rangle$ to level $|1\rangle$ since only one photon number could change in the transition, and there is only the one quantized field interaction potential, $\mathscr{V}_{51}$. In the two-photon problem, however, the photon number for the fields must change twice for each transition between any pair of the four levels. The quantum number n_1 can go to n_1+1 in any of three ways: From $|5\rangle$ to j to $|1\rangle$, from $|5\rangle$ to j to $|4\rangle$, and from $|2\rangle$ to j to $|1\rangle$. Figure 3 readily shows each of these "two-step" processes. Hence we expect three quantized field interaction potentials for the two-photon problem, $\mathscr{V}_{51}$, $\mathscr{V}_{54}$, and $\mathscr{V}_{21}$.

These additional interaction potentials account for the increased complexity of our equations. The potentials $\mathscr{V}_{54}$ and $\mathscr{V}_{21}$ represent processes that have no single photon analog. A $\mathscr{V}_{54}$ transition implies an atom in the upper level spontaneously emits but still remains in the upper level, and a $\mathscr{V}_{21}$ transition represents spontaneous emission originating from the ground state. This is possible for a two-photon transition, of course, because in each case a photon is also absorbed from the classical pump field, preserving energy conservation. These transitions thus represent an exchange of photons between the quantized and pump fields.

To justify these assertions, we now show that the projections of the operator $\tilde{\phi}^+$ are related to the components of the atom-field density operator ρ. For example, by taking the field component of $\tilde{\phi}^+_{ab}$ we have

$$\begin{aligned}\langle n_1 n_2|\tilde{\phi}^+_{ab}|n_1 n_2\rangle &= \langle a\, n_1 n_2|a^\dagger\rho|b\, n_1 n_2\rangle \\ &= \sqrt{n_1}\langle a\, n_1-1\, n_2|\rho|b\, n_1 n_2\rangle \\ &= \sqrt{n_1}\rho'_{51}\,, \qquad (64a)\end{aligned}$$

where the prime means let $n_1 \to n_1-1$ and where we approximate $n_2 \simeq n_2+1$. Similarly, for the other components

$$\langle n_1 n_2|\tilde{\phi}^+_{aa}|n_1 n_2\rangle = \langle a\, n_1 n_2|a^\dagger\rho|a\, n_1 n_2\rangle = \sqrt{n_1}\langle a\, n_1-1\, n_2|\rho|a\, n_1 n_2\rangle = \sqrt{n_1}\rho'_{54} \qquad (64b)$$

$$\langle n_1 n_2|\tilde{\phi}^+_{bb}|n_1 n_2\rangle = \langle b\, n_1 n_2|a^\dagger\rho|b\, n_1 n_2\rangle = \sqrt{n_1}\langle b\, n_1-1\, n_2|\rho|b\, n_1 n_2\rangle = \sqrt{n_1}\rho'_{21} \qquad (64c)$$

$$\langle n_1 n_2|\tilde{\phi}^+_{ba}|n_1 n_2\rangle = \langle b\, n_1 n_2|a^\dagger\rho|a\, n_1 n_2\rangle = \sqrt{n_1}\langle b\, n_1-1\, n_2|\rho|a\, n_1 n_2\rangle = \sqrt{n_1}\rho'_{24}\,. \qquad (64d)$$

We may also project the adjoint of $\phi,\phi^\dagger$, onto these states and

$$\begin{aligned}\langle n_1 n_2|(\tilde{\phi}_{ab})^\dagger|n_1 n_2\rangle &= \langle b\, n_1 n_2|\rho a^\dagger|a\, n_1 n_2\rangle = \sqrt{n_1+1}\langle b\, n_1 n_2|\rho|a\, n_1+1\, n_2\rangle \\ &= \sqrt{n_1+1}\rho_{24} \qquad (65a)\end{aligned}$$

$$\begin{aligned}\langle n_1 n_2|\tilde{\phi}^\dagger_{aa}|n_1 n_2\rangle &= \langle a\, n_1 n_2|\rho a^\dagger|a\, n_1 n_2\rangle = \sqrt{n_1+1}\langle a\, n_1 n_2|\rho|a\, n_1+1\, n_2\rangle \\ &= \sqrt{n_1+1}\rho_{54} \qquad (65b)\end{aligned}$$

$$\begin{aligned}\langle n_1 n_2|\tilde{\phi}^\dagger_{bb}|n_1 n_2\rangle &= \langle b\, n_1 n_2|\rho a^\dagger|b\, n_1 n_2\rangle = \sqrt{n_1+1}\langle b\, n_1 n_2|\rho|b\, n_1+1\, n_2\rangle \\ &= \sqrt{n_1+1}\rho_{21} \qquad (65c)\end{aligned}$$

$$\begin{aligned}\langle n_1 n_2|(\tilde{\phi}_{ba})^\dagger|n_1 n_2\rangle &= \langle a\, n_1 n_2|\rho a^\dagger|b\, n_1 n_2\rangle = \sqrt{n_1+1}\langle a\, n_1 n_2|\rho|b\, n_1+1\, n_2\rangle \\ &= \sqrt{n_1+1}\rho_{51}\,. \qquad (65d)\end{aligned}$$

Projecting Eq. (63) onto the basis states and substituting Eqs. (64) and (65), we find the equation of motion for the probability of the photon number $\langle n_1\rangle$ to be

$$\dot{\rho}_{n_1} = [\mathscr{V}_{15}\rho_{51} + i\mathscr{V}_{45}\rho_{54} + i\mathscr{V}_{12}\rho_{21} - i\,|\mathscr{E}_1|^2(n_1+1)(k^-_{aa}\rho_{44} + k^+_{aa}\rho_{55} + k^-_{bb}\rho_{11} + k^+_{aa}\rho_{22}) - (n_1 \to n_1-1)] + \text{c.c.}\,, \qquad (66)$$

where $\mathscr{V}_{51} = -k_{ab}\mathscr{E}_1\mathscr{E}_2\sqrt{n_1+1}$, $\mathscr{V}_{54} = -k_{aa}\mathscr{E}_1\mathscr{E}^*_2\sqrt{n_1+1}$, $\mathscr{V}_{21} = -k_{bb}\mathscr{E}_1\mathscr{E}^*_2\sqrt{n_1+1}$, and the k^+_{aa}, k^+_{bb}, k^-_{aa}, and k^-_{bb} terms are the k_{aa} and k_{bb} sums in Eqs. (23) and (24) with only the $\omega_{aj}+\nu_2$, $\omega_{jb}+\nu_2$, $\omega_{aj}-\nu_2$, or $\omega_{jb}-\nu_2$ terms, respectively.

We have thus proven the existence of these new interaction potentials $\mathscr{V}_{54}$ and $\mathscr{V}_{21}$ and have demonstrated how these are related to the two-photon coefficients k_{aa} and k_{bb}. In the semiclassical solution of Sec. II and Ref. 11, it is shown how these lead to the occurrence of the dynamic Stark shift. Here in the quantum calculation we see they also play a major role in the process of spontaneous emission. Note that the presence of the $\mathscr{E}^*_2$ in the $\mathscr{V}_{54}$ and $\mathscr{V}_{21}$ potentials represents an absorption of a classical field photon, and is depicted in Fig. 3 as an upward transition.

We project Eqs. (51)–(54) onto the basis states $|n_1 n_2\rangle$ and make use of Eqs. (64). We find

$$\dot\rho_{51}=-(\gamma+i\Delta_1)\rho_{51}-i[\mathscr{V}_{51}(\rho_{11}-\rho_{55})+i\mathscr{V}_2(\rho_{21}-\rho_{54})-\rho_{52}\mathscr{V}_{21}+\mathscr{V}_{54}\rho_{41}]\ , \tag{67}$$

$$\dot\rho_{54}=-(\Gamma+i\Delta)\rho_{54}-i[\mathscr{V}_{51}\rho_{14}-\rho_{51}\mathscr{V}_{54}+\mathscr{V}_{52}\rho_{24}-i\mathscr{V}_{54}(\rho_{55}-\rho_{44})]\ , \tag{68}$$

$$\dot\rho_{21}=\Gamma\rho_{54}-i\Delta\rho_{21}+i[\rho_{25}\mathscr{V}_{51}-\mathscr{V}_{25}\rho_{51}+\rho_{24}\mathscr{V}_{41}+i\mathscr{V}_{21}(\rho_{22}-\rho_{11})]\ , \tag{69}$$

$$\dot\rho_{24}=-(\gamma-i\Delta_3)\rho_{24}-i(\mathscr{V}_{25}\rho_{54}-\rho_{21}\mathscr{V}_{14}+\mathscr{V}_{21}\rho_{14}-\rho_{25}\mathscr{V}_{54})\ , \tag{70}$$

where $\mathscr{V}_2=\mathscr{V}_{52}=\mathscr{V}_{41}=-k_{ab}\mathscr{E}_2^2$ is the classical interaction potential. Substituting for ρ_{54} given by the trace condition

$$P_{n_1;n_1+1}\equiv P_{01}=\rho_{54}+\rho_{21} \tag{71}$$

into Eq. (69), we have

$$\dot\rho_{21}=-(\Gamma+i\Delta)\rho_{21}+\Gamma P_{01}+i[\rho_{25}\mathscr{V}_{51}-\mathscr{V}_{25}\rho_{51}+\rho_{24}\mathscr{V}_{41}+i\mathscr{V}_{21}(\rho_{22}-\rho_{11})]\ . \tag{72}$$

As we asserted earlier, we assume the atoms relax quickly compared to the field variations and so we solve Eqs. (67), (68), (70), and (72) in steady state. We then substitute these solutions along with the semiclassical equations for ρ_{25}, ρ_{41}, etc., into the photon number equation of motion (66). Solving Eq. (67) in steady state, we have

$$\rho_{51}=i\mathscr{D}_1(\mathscr{V}_{51}d_{051}+\mathscr{V}_{21}\rho_{52}-\mathscr{V}_{54}\rho_{41})+i\mathscr{D}_1\mathscr{V}_2d_{154}=\rho_{51}^0+i\mathscr{D}_1\mathscr{V}_2d_{154}\ , \tag{73}$$

where in general $d_{0ij}=\rho_{ii}-\rho_{jj}$ and $d_{154}=\rho_{54}-\rho_{21}$, and $\mathscr{D}_1$ is the $n=1$ case of the complex denominator

$$\mathscr{D}_n=\frac{1}{\gamma+i\Delta_n}\ . \tag{74}$$

Similarly Eq. (70) yields

$$\rho_{24}=-i\mathscr{D}_3^*(\mathscr{V}_{21}\rho_{14}-\mathscr{V}_{54}\rho_{25})-i\mathscr{D}_3^*\mathscr{V}_2d_{154}=\rho_{24}^0-i\mathscr{D}_3^*\mathscr{V}_2d_{154}\ . \tag{75}$$

Subtracting the steady-state solution of Eq. (72) from that of (68), we have

$$d_{154}=\mathscr{F}P_{01}-i\mathscr{D}_a[\mathscr{V}_{51}(\rho_{14}+\rho_{25})-2\mathscr{V}_2^*\rho_{51}+2\mathscr{V}_2\rho_{24}-\mathscr{V}_{54}d_{054}+\mathscr{V}_{21}d_{021}]\ , \tag{76}$$

where the complex denominators

$$\mathscr{D}_a=\frac{1}{\Gamma+i\Delta}\ , \tag{77}$$

$$\mathscr{F}=\Gamma\mathscr{D}_a\ . \tag{78}$$

Substituting Eqs. (73) and (75) into (76) and solving for d_{154}, we have

$$d_{154}=\frac{-i\mathscr{D}_a[\mathscr{V}_{51}(\rho_{14}+\rho_{25})-2\mathscr{V}_2^*\rho_{51}^0+2\mathscr{V}_2\rho_{24}^0-\mathscr{V}_{54}d_{054}+\mathscr{V}_{21}d_{021}]-\mathscr{F}P_{01}}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}=d_{154}^0-\frac{\mathscr{F}P_{01}}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\ . \tag{79}$$

Using the explicit values for the various density-matrix elements inside the [ ], we have (for typographical simplicity, we set $n=n_1$)

$$[\ ]=\mathscr{V}_{51}(p_{n+1}+p_n)\tilde R_{ba}-2i\mathscr{V}_{51}\mathscr{V}_2^*\mathscr{D}_1(R_{aa}p_n-R_{bb}p_{n+1})-2i\mathscr{D}_1\mathscr{V}_2^*\tilde R_{ab}(\mathscr{V}_{21}p_n-\mathscr{V}_{54}p_{n+1})+2i\mathscr{D}_3^*\mathscr{V}_2\tilde R_{ba}(\mathscr{V}_{54}p_n-\mathscr{V}_{21}p_{n+1})-(p_n-p_{n+1})(\mathscr{V}_{54}R_{aa}-\mathscr{V}_{21}R_{bb})\ . \tag{80}$$

From the trace condition (71) we also have

$$\rho_{54}=\tfrac{1}{2}(P_{01}+d_{154})\ , \tag{81}$$

$$\rho_{21}=\tfrac{1}{2}(P_{01}-d_{154})\ . \tag{82}$$

Substituting these equations and Eqs. (73), (75), and (79) into (66), we find

$$\dot{p}_{n_1}=\frac{i}{2}(\mathscr{V}_{54}^*+\mathscr{V}_{21}^*)P_{01}+i\mathscr{V}_{15}\rho_{51}^0+\left[\frac{i}{2}(\mathscr{V}_{45}-\mathscr{V}_{12})-\mathscr{D}_1\mathscr{V}_2\mathscr{V}_1^*\right]d_{154}$$

$$-i\frac{|\mathscr{E}_1|^2}{4h}-(k_{aa}^+\rho_{55}+k_{aa}^-\rho_{44}+k_{bb}^+\rho_{11}+k_{bb}^-\rho_{22})-(\text{same with } n_1\to n_1-1)+\text{c.c.}$$

$$=P_{01}\left[\frac{i}{2}(\mathscr{V}_{45}+\mathscr{V}_{12})-\frac{\left[\frac{i}{2}(\mathscr{V}_{45}-\mathscr{V}_{12})-\mathscr{D}_1\mathscr{V}_2\mathscr{V}_{15}\right]\mathscr{F}}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right]+i\mathscr{V}_1^*\rho_{51}^0+\left[\frac{i}{2}(\mathscr{V}_{45}-\mathscr{V}_{12})-\mathscr{D}_1\mathscr{V}_2\mathscr{V}_{15}\right]d_{154}^0$$

$$-i\frac{|\mathscr{E}_1|^2}{4\hbar}(k_{aa}^+\rho_{55}+k_{aa}^-\rho_{44}+k_{bb}^+\rho_{11}+k_{bb}^-\rho_{22})-(\text{same with } n_1\to n_1-1)+\text{c.c.} \tag{83}$$

Further taking the derivative of the trace condition (71) and using (68) and (69), we have

$$\dot{P}_{01}=\dot{\rho}_{54}+\rho_{21}$$
$$=-i\Delta P_{01}+i\mathscr{V}_{51}(\rho_{25}-\rho_{14})+i\mathscr{V}_{54}d_{054}+i\mathscr{V}_{21}d_{021}\ . \tag{84}$$

In steady state (assuming the field varies little in a time $1/\Delta$), we have the off-diagonal matrix element

$$P_{01}=\frac{p_n-p_{n+1}}{\Delta}(\mathscr{V}_{51}\tilde{R}_{ba}+\mathscr{V}_{54}R_{aa}+\mathscr{V}_{21}R_{bb})\ . \tag{85}$$

Substituting this along with Eq. (73) for ρ_{51}^0, (75) for ρ_{24}^0, (79) and (80) for d_{154}^0, we have the sidemode photon-number equation of motion

$$\dot{p}_{n_1}=\{-(n+1)[A_1p_n-B_1p_{n+1}]+n[A_1p_{n-1}-B_1p_n]\}+\text{c.c.}\ , \tag{86}$$

where

$$A_1=-\frac{i(v_{51}\tilde{R}_{ba}+v_{54}R_{aa}+v_{21}R_{bb})}{2\Delta}\left[v_{45}+v_{12}-\frac{(v_{45}-v_{12}+2i\mathscr{D}_1\mathscr{V}_2v_{15})\mathscr{F}}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right]$$

$$+i|\mathscr{E}_1|^2(k_{aa}^+R_{aa}+k_{bb}^+R_{bb})+v_{15}\mathscr{D}_1(v_{51}R_{aa}+v_{21}\tilde{R}_{ab})$$

$$-\frac{\mathscr{D}_a}{2}\left[\frac{v_{45}-v_{12}+2i\mathscr{D}_1\mathscr{V}_2v_{15}}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right](v_1+2i\mathscr{D}_3^*\mathscr{V}_2v_{54})\tilde{R}_{ba}$$

$$-2i\mathscr{D}_1\mathscr{V}_2^*v_{21}\tilde{R}_{ab}-(2i\mathscr{V}_2^*v_{51}\mathscr{D}_1+v_{54})R_{aa}+v_{21}R_{bb}\ , \tag{87}$$

$$B_1=-\frac{i(v_1\tilde{R}_{ba}+v_{54}R_{aa}+v_{21}R_{bb})}{2\Delta}\left[v_{45}+v_{12}-\frac{(v_{45}-v_{12}+2i\mathscr{D}_1\mathscr{V}_2v_{15})\mathscr{F}}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right]$$

$$-i|\mathscr{E}_1|^2(k_{bb}^-R_{bb}+k_{aa}^-R_{aa})+v_{15}\mathscr{D}_1(v_1R_{bb}+v_{54}\tilde{R}_{ab})$$

$$-\frac{\mathscr{D}_a}{2}\left[\frac{v_{45}-v_{12}+2i\mathscr{D}_1\mathscr{V}_2v_{15}}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right](-v_{51}+2i\mathscr{D}_3^*\mathscr{V}_2v_{21})\tilde{R}_{ba}$$

$$-2i\mathscr{D}_1\mathscr{V}_2^*v_{54}\tilde{R}_{ab}-(2i\mathscr{V}_2^*v_{51}\mathscr{D}_1-v_{21})R_{bb}-v_{54}R_{aa}\ , \tag{88}$$

and where we set $v_{51}=\mathcal{V}_{51}/\sqrt{n_1+1}$, $v_{54}=\mathcal{V}_{54}/\sqrt{n_1+1}$, etc. We note that Eq. (86) is identical to the result derived in Ref. 1, Eq. (64), except for the cavity loss constant which we have neglected here for simplicity. The corresponding expressions for the A_1 and B_1 terms are considerably more complicated, however.

Equations (87) and (88) may be written in a form more closely resembling the one-photon coefficients derived in Ref. 1, Eqs. (70) and (71) of that paper, by substituting the equations for the atomic density elements from Eqs. (34), (35), and (36) and for the interaction potentials v_{54}, v_{21}, and v_{51}. We find

$$\begin{aligned} A_1=\frac{|k_{ab}|\,|\mathcal{E}_1|^2}{2(1+I_2^2\mathcal{L}_2)}\Bigg[& \sqrt{\gamma\Gamma}I_2\mathcal{D}_1(2f_a+i\sqrt{\gamma\Gamma}I_2\mathcal{D}_2k_{bb}/|k_{ab}|) \\ & +\frac{2i(k_{aa}^{+}f_a+k_{bb}^{+}f_b)}{|k_{ab}|}-I_2^2\mathcal{F}\sqrt{\gamma\Gamma}\frac{\gamma I_2\mathcal{D}_1-i\omega_s/\Gamma}{1+I_2^2\mathcal{F}\frac{\gamma}{2}(\mathcal{D}_1+\mathcal{D}_3^*)} \\ & \times\left[\mathcal{D}_1f_a-\frac{\mathcal{D}_2^*}{2}+i\sqrt{\gamma\Gamma}I_2\frac{k_{aa}\mathcal{D}_2^*\mathcal{D}_3^*+k_{bb}\mathcal{D}_1\mathcal{D}_2}{2|k_{ab}|}-\frac{i(k_{bb}f_b-k_{aa}f_a)}{\sqrt{\gamma\Gamma}I_2|k_{ab}|}\right] \\ & +\frac{\sqrt{\gamma\Gamma}I_2\mathcal{D}_2^*/2+i(k_{aa}f_a+k_{bb}f_b)/|k_{ab}|}{i\Delta}\left[\frac{I_2\mathcal{F}(\gamma\Gamma I_2\mathcal{D}_1-i\omega_s)}{1+I_2^2\mathcal{F}\frac{\gamma}{2}(\mathcal{D}_1+\mathcal{D}_3^*)}-i\sqrt{\gamma\Gamma}\frac{I_2(k_{aa}+k_{bb})}{|k_{ab}|}\right]\Bigg] \end{aligned} \tag{89}$$

$$\begin{aligned} B_1=\frac{|k_{ab}|\,|\mathcal{E}_1|^2}{2(1+I_2^2\mathcal{L}_2)}\Bigg[& \sqrt{\gamma\Gamma}I_2\mathcal{D}_1(2f_b+i\sqrt{\gamma\Gamma}I_2\mathcal{D}_2k_{aa}/|k_{ab}|)-\frac{2i(k_{aa}^{-}f_a+k_{bb}^{-}f_b)}{|k_{ab}|} \\ & -I_2^2\mathcal{F}\sqrt{\gamma\Gamma}\frac{\gamma I_2\mathcal{D}_1-i\omega_s/\Gamma}{1+I_2^2\mathcal{F}\frac{\gamma}{2}(\mathcal{D}_1+\mathcal{D}_3^*)} \\ & \times\left[\mathcal{D}_1f_b+\frac{\mathcal{D}_2^*}{2}+i\sqrt{\gamma\Gamma}I_2\frac{k_{bb}\mathcal{D}_2^*\mathcal{D}_3^*+k_{aa}\mathcal{D}_1\mathcal{D}_2}{2|k_{ab}|}-\frac{i(k_{bb}f_b-k_{aa}f_a)}{\sqrt{\gamma\Gamma}I_2|k_{ab}|}\right] \\ & +\frac{\sqrt{\gamma\Gamma}I_2\mathcal{D}_2^*/2+i(k_{aa}f_a+k_{bb}f_b)/|k_{ab}|}{i\Delta} \\ & \times\left[\frac{I_2\mathcal{F}(\gamma\Gamma I_2\mathcal{D}_1-i\omega_s)}{1+I_2^2\mathcal{F}\frac{\gamma}{2}(\mathcal{D}_1+\mathcal{D}_3^*)}-i\sqrt{\gamma\Gamma}\frac{I_2(k_{aa}+k_{bb})}{|k_{ab}|}\right]\Bigg]\,. \end{aligned} \tag{90}$$

VI. THE RESONANCE FLUORESCENCE SPECTRUM

Following the same procedure as in Ref. 1, we calculate the build up of mode 1, which can be described by the average photon number $\langle n_1\rangle=\sum_{n_1}n_1p_{n_1}$. Using Eq. (86), we find the equation of motion

$$\begin{aligned} \frac{d}{dt}\langle n_1\rangle &=[-A_1(\langle n_1^2\rangle+\langle n_1\rangle)-B_1\langle n_1^2\rangle+B_1(\langle n_1^2\rangle-\langle n_1\rangle)+A_1(\langle n_1^2\rangle+2\langle n_1\rangle+1)]+\text{c.c.} \\ &=[(A_1-B_1)\langle n_1\rangle+A_1]+\text{c.c.} \end{aligned} \tag{91}$$

As for the one-photon case, this consists of the gain coefficient A_1-B_1 and the resonance fluorescence coefficient A_1. The gain coefficient given by Eqs. (89) and (90) is

$$A_1 - B_1 = -\frac{|k_{ab}|\,|\mathscr{E}_1|^2}{2(1+I_2^2\mathscr{L}_2)}\sqrt{\Gamma/\gamma}\left[\gamma I_2\mathscr{D}_1(2-i\omega_s I_2\mathscr{D}_2)+2i(k_{aa}f_a+k_{bb}f_b)/|k_{ab}| - \frac{\gamma I_2^2\mathscr{F}(\gamma I_2\mathscr{D}_1 - i\omega_s/\Gamma)}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\left[\mathscr{D}_1+\mathscr{D}_2^*+\frac{i\omega_s I_2}{2}(\mathscr{D}_2^*\mathscr{D}_3^*-\mathscr{D}_1\mathscr{D}_2)\right]\right]. \tag{92}$$

This agrees with the two-photon semiclassical sidemode absorption coefficient calculated in Ref. 11 that adopts a completely semiclassical approach.

Some discussion of the two-photon resonance fluorescence spectrum as given by Eq. (91) has already been given in Ref. 10. Due to the complexity of the expression, in this section we present additional examples. We first consider the case of zero Stark shift. As discussed in the semiclassical case of Ref. 11, for no Stark shift the spectra closely resemble the equivalent one-photon problem, and we find this to be the case in the quantum calculation as well. Figure 4 plots the inelastic spectrum of $A_1+A_1^*=A$ for the strong pump field of $I_2=10$, $T_2=2T_1$, and $\omega_s=0$ for the detunings $\omega-2\nu_2=0$, $2T_1^{-1}$, and $5T_1^{-1}$. We note this has the same appearance and behavior of the one-photon result of Mollow.[15] In particular, for central tuning, the heights and widths of the peaks have the same proportion as the one-photon spectrum and it remains symmetric when detuned. Note also that the side peaks are at the detuned Rabi flopping frequency and that the central peak rapidly drops with detuning.

We now consider the fluorescence spectrum from Eq. (89) for nonzero ω_s. This is by far more realistic since practically all two-photon systems have a nonzero Stark shift. Initially considering central tuning, Fig. 5 depicts the spectrum vs ΔT_1 for $I_2=10$, $T_2=2T_1$, and for $\omega_s=0$, $0.2T_1^{-1}$, and $0.5T_1^{-1}$. This means the actual Stark shifts $\omega_s I_2$ are 0, $2T_1^{-1}$, and $5T_1^{-1}$. In a manner similar to varying the detuning in Fig. 4, we see once again the side peaks move outward and the central peak drops. More obviously, one sideband is increased in intensity and the other is attenuated—the Stark shift leads to a noticeably asymmetric spectrum. Thus we see that in some respects $\omega_s I_2$ behaves like a detuning, yet is is also quite different in other respects. That it is similar to a detuning is understandable because of the appearance of $\omega_s I_2$ in $\mathscr{L}_2$ and the complex Lorentzians $\mathscr{D}_n$. However, the asymmetry is not like a detuning, and so this is evidently due to the ω_s and k_{aa}, k_{ab}, and k_{bb} terms appearing elsewhere in Eq. (89). Figure 6, which is also presented in Ref. 10, shows the effect of "balancing" the Stark shift versus detuning, i.e., $\omega_s I_2=5T_1^{-1}$, but $\omega-2\nu_2=-5T_1^{-1}$. We see that the main effect is to raise the central peak back to its centrally tuned, zero Stark shifted value. In addition, the sidebands are brought back in, but the asymmetry remains.

We may gain some insight into the cause of this asymmetry from the two-photon probe gain/absorption coefficient $A-B$ of Eq. (92). This quantity has already been graphed and discussed in Ref. 11. In Fig. 7 we plot $A-B$ from Eq. (92) for the following parameters: $I_2=10$, $\omega_s I_2=5T_1^{-1}$, $\omega-2\nu_2=0$, and $T_2=2T_1$. The spectral gain (gain is positive) is almost antisymmetric; there is strong gain at one sideband and an almost equal absorption at the other. Note that the probe gain appears at almost the same place in frequency as the sharply increased sideband of the fluorescent spectrum, and vice versa for

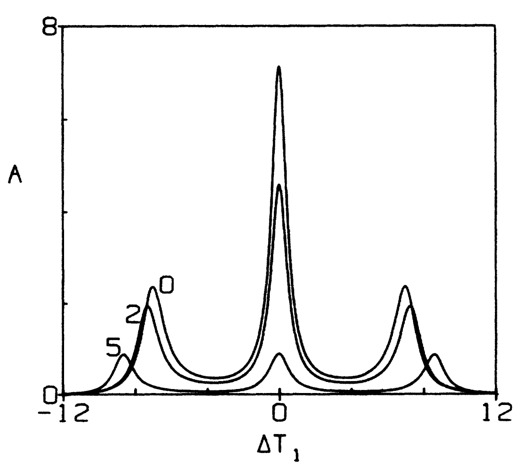

FIG. 4. $A_1+\text{c.c.}=A$ of Eq. (89) vs ΔT_1 for $I_2=10$, $\omega_s=0$, $T_2=2T_1$ for detunings of 0, $2T_1^{-1}$, and $5T_1^{-1}$.

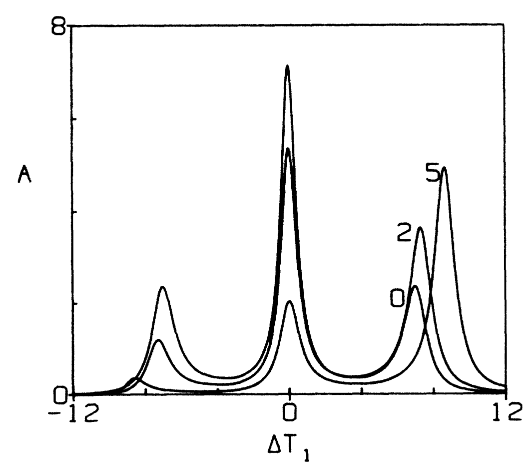

FIG. 5. A vs ΔT_1 for $I_2=10$, $\omega-2\nu_2=0$, $T_2=2T_1$ for $\omega_s I_2=0$, $2T_1^{-1}$, and $5T_1^{-1}$.

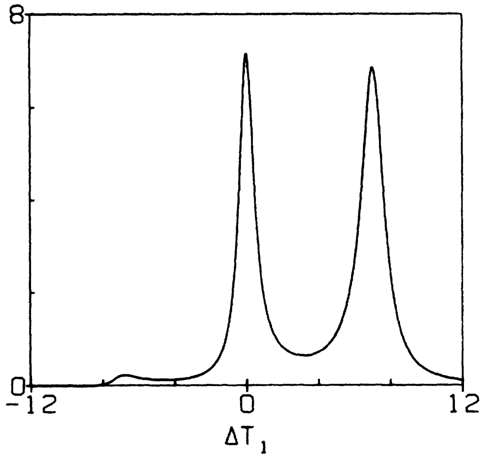

FIG. 6. A vs ΔT_1 for $I_2=10$, $T_2=2T_1$, $\omega-2\nu_2=-5T_1^{-1}$, and $\omega_s I_2=5T_1^{-1}$.

the other sideband. Thus the increase of one sideband and the decrease of the other is related to the asymmetric shape of the gain/absorption spectrum.

One other property of the inelastic scattering spectrum of Eq. (89), unlike the one-photon result of Mollow, is that the total energy scattered does not saturate as $I_2\to\infty$, but instead increases linearly with I_2. This can be seen from the expression for A_1. This is characteristic of two-photon processes, and is due to our first-order perturbation treatment of the off-resonance j levels.

The two-photon two-level resonance fluorescence spectrum also has a significantly more complex expression for the elastic portion of the spectrum, also called the Rayleigh peak. The Rayleigh peak is given by the term in the expression for the A_1 coefficient, Eq. (89), that is divided by $i\Delta$. To evaluate the Rayleigh scattering from this model, we allow $\Delta\to 0$ in this term, which gives a real numerator. This gives a delta function spectrum because $i/\Delta+\text{c.c.}=2\pi\delta(\Delta)$. The elastic contribution is then

$$A_{\text{el}}=2\pi\sqrt{\gamma\Gamma}\frac{I_2|\mathscr{E}_1|^2}{(1+I_2^2\mathscr{L}_2)^2}\times\left[\frac{(k_{aa}f_a+k_{bb}f_b)^2}{|k_{ab}|}+\frac{|k_{ab}|\Gamma f_a}{2\gamma}+\frac{(k_{aa}f_a+k_{bb}f_b)I_2\mathscr{L}_2(\gamma\Gamma)^{1/2}\Delta_2}{\gamma^2}\right]\delta(\Delta)\,, \quad (93)$$

where Δ_2 is the detuning (including the Stark shift) of the strong mode, $\omega+\omega_s I_2-2\nu_2$. The Rayleigh scattering consists of three contributions, (1) the off-resonant dipoles, (2) the two-photon two-level coherence R_{ab}, and (3) the interference between the dipoles and R_{ab}. Because of this third term, the elastic scattering is asymmetric with respect to detuning, totally unlike the one-photon resonance fluorescence case. Depending upon the relative values of k_{aa}, k_{bb}, and k_{ab}, this part of the spectrum may either dominate the total emission, or, for an appropriate detuning and intensity I_2, be very small. We recall that the elastic scattering of the one-photon case bleaches to zero for strong, resonant fields, and is symmetric with detuning. The two-photon result of Eq. (93) is considerably different.

The expression for the elastic scattering intensity, Eq. (93), may also be obtained from the semiclassical expression for the complex amplitude of the polarization of the medium, Eq. (25). This provides an additional check on our calculations. From the slowly varying amplitude and phase relationship, the scattered radiation field amplitude is proportional to the complex polarization $\mathscr{P}$ of Eq. (25). Hence, the intensity of the elastic scattering is proportional to the squared modulus of $\mathscr{P}$. Evaluating the square of Eq. (25) we find that it agrees with the expression of Eq. (93).

Figure 8 plots Eq. (93) vs the detuning $\omega-2\nu_2$ for $I_2=10$, $k_{ab}=k_{aa}$, $T_2=2T_1$ for $\omega_s I_2=-5T_1^{-1}$, $-2T_1^{-1}$, 0, $2T_1^{-1}$, and $5T_1^{-1}$. The asymmetry of each curve is evident. Figure 9 is similar except only the $\omega_s I_2=-2T_1^{-1}$,

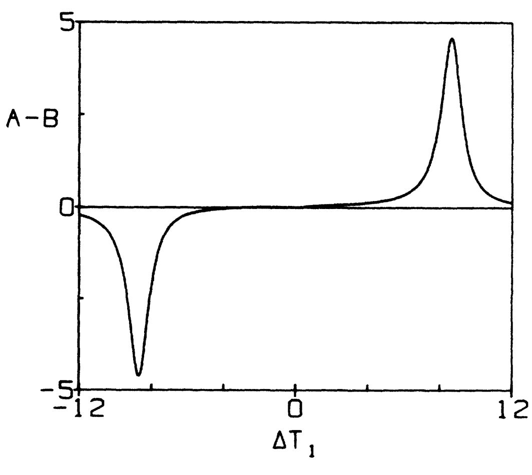

FIG. 7. $A_1-B_1+\text{c.c.}=A-B$ vs ΔT_1 for $I_2=10$, $\omega-2\nu_2=0$, and $\omega_s I_2=5T_1^{-1}$.

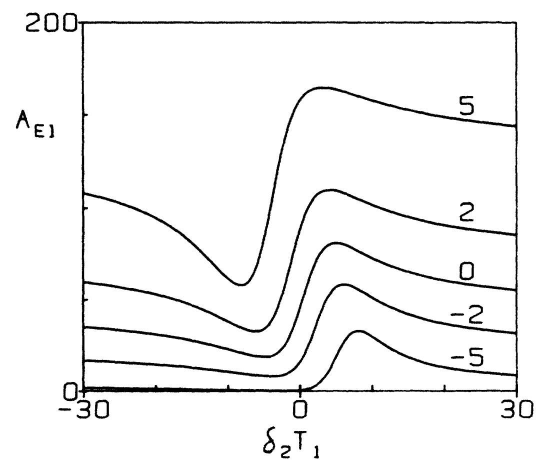

FIG. 8. The elastic scattering A_{el} vs $(\omega-2\nu_2)T_1=\delta_2 T_1$ for $I_2=10$, $k_{ab}=k_{aa}$, and $\omega_s I_2=-5T_1^{-1}$, $-2T_1^{-1}$, 0, $2T_1^{-1}$, $5T_1^{-1}$.

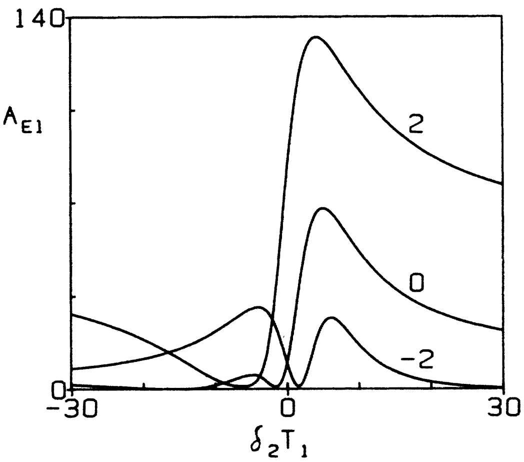

FIG. 9. The elastic scattering A_{el} vs $(\omega-2\nu_2)T_1=\delta_2 T_1$ for $I_2=10$, $k_{ab}=5k_{aa}$, and $\omega_s I_2=-2T_1^{-1}$, 0, $2T_1^{-1}$.

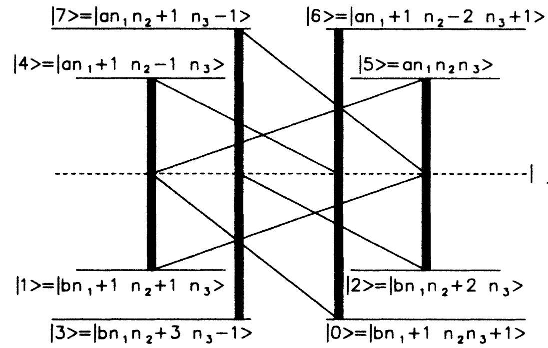

FIG. 10. Two-photon eight-level atom-field energy level diagram valid for one strong central mode and two weak sidemodes. This case treats coupled-mode phenomena such as phase conjugation and modulation spectroscopy.

0, and $2T_1^{-1}$ curves are plotted and now $k_{ab}=5k_{bb}$. In addition, the intensity of Rayleigh scattering does not bleach to zero as in the one-photon case, but increases linearly with I_2, just as for the inelastic spectrum.

VII. DOUBLE SIDEMODE SOLUTION

We now include the second sidemode of frequency $\nu_3=\nu_2+\Delta$. This means that we consider mode-coupling contributions arising from terms like a_1a_3 in Eqs. (51)–(55) that were dropped in Sec. V. We again project these terms onto a set of basis states like those of Fig. 3, only now we must include four additional atom-field states to account for the coupling to the third mode. Following Ref. 1, we denote the new states as

$$|0\rangle=|b\,n_1+1\,n_2\,n_3+1\rangle$$

and

$$|7\rangle=|a\,n_1\,n_2+1\,n_3-1\rangle$$

that are coupled by the strong mode potential $\mathscr{V}_2$ to the states

$$|6\rangle=|a\,n_1+1\,n_2-2\,n_3+1\rangle$$

and

$$|3\rangle=|b\,n_1\,n_2-1\,n_3-1\rangle ,$$

respectively. These are shown in Fig. 10. Note that the states of Fig. 10 are almost identical to those of Fig. 4 of Ref. 1, except that the lower-level states have an additional n_2 photon because of the two-photon transition. In Fig. 10, a mode 3 transition is shown by a line slanting downward to the right, in comparison to mode 1 transitions that are down to the left, and mode 2 transitions that are straight down and in boldface.

In the one-photon problem the second sidemode introduces the new potentials $\mathscr{V}_{04}$ and $\mathscr{V}_{27}$. In the two-photon theory we are presenting here, we also have these potentials, but analogous to the new appearance of the $\mathscr{V}_{54}$ and $\mathscr{V}_{21}$ potentials, we again have additional interaction potentials due to the two-photon process. Referring to Fig. 10, we see that by emitting an n_2 photon and absorbing an n_3 photon, $|5\rangle$ is connected to $|7\rangle$, $|6\rangle$ is connected to $|4\rangle$, $|0\rangle$ is connected to $|1\rangle$, and $|2\rangle$ is connected to $|3\rangle$. Thus we expect to acquire the new potentials $\mathscr{V}_{57}$, $\mathscr{V}_{64}$, $\mathscr{V}_{01}$, and $\mathscr{V}_{23}$. We also see from Fig. 10 that the pairs of levels $|7\rangle$ and $|1\rangle$ and $|5\rangle$ and $|0\rangle$ are connected by a transition of the form $\mathscr{E}_1\mathscr{E}_3$, independent of $\mathscr{E}_2$. These potentials connect the upper and lower levels (and thus involve k_{ab}) even in the absence of the pump field, and they are also unique to the two-photon model. As we demonstrate below, these do not enter into the equations of motion for the atom-field density-matrix elements, but do appear in the equation of motion for the reduced field density operator P.

To justify these assertions, we now find the components of our operator equations (51)–(54) using the $|n_1n_2n_3\rangle$ states of Fig. 10 as a basis. Because the single sidemode case has been presented, in all of the equations that follow only the coupled mode terms are shown. We designate these terms by the subscript a_3. Using Eqs. (65), we find

$$\dot\rho_{51}|_{a_3}=-(\gamma+i\Delta_1)\rho_{51}-i(\mathscr{V}_{57}\rho_{71}-\mathscr{V}_{01}\rho_{50}) , \tag{94}$$

$$\dot\rho_{54}|_{a_3}=-(\Gamma+i\Delta)\rho_{54} -i(\mathscr{V}_{57}\rho_{74}-\mathscr{V}_{64}\rho_{56}-\mathscr{V}_{04}\rho_{50}) , \tag{95}$$

$$\dot\rho_{21}|_{a_3}=-i\Delta\rho_{21}+\Gamma\rho_{54} -i(\mathscr{V}_{23}\rho_{31}+\mathscr{V}_{27}\rho_{71}-\mathscr{V}_{01}\rho_{20}) , \tag{96}$$

$$\dot\rho_{24}|_{a_3}=-(\gamma-i\Delta_3)\rho_{24} -i(\mathscr{V}_{23}\rho_{34}+\mathscr{V}_{27}\rho_{74}-\mathscr{V}_{64}\rho_{26}-\mathscr{V}_{04}\rho_{20}) , \tag{97}$$

where the interactions potentials are

$$\mathscr{V}_{57}=-k_{aa}\mathscr{E}_2\mathscr{E}_3^*\sqrt{n_3} \tag{98a}$$

$$\mathscr{V}_{01}=-k_{bb}\mathscr{E}_2\mathscr{E}_3^*\sqrt{n_3+1} \tag{98b}$$

$$\mathscr{V}_{64}=-k_{aa}\mathscr{E}_2\mathscr{E}_3^*\sqrt{n_3+1} \tag{98c}$$

$$\mathscr{V}_{23}=-k_{bb}\mathscr{E}_2\mathscr{E}_3^*\sqrt{n_3} \tag{98d}$$

$$\mathscr{V}_{04}=-k_{ab}\mathscr{E}_2^*\mathscr{E}_3^*\sqrt{n_3+1} \tag{98e}$$

$$\mathscr{V}_{27}=-k_{ab}\mathscr{E}_2^*\mathscr{E}_3^*\sqrt{n_3}\ . \tag{98f}$$

In a similar manner, we project the $\dot{P}$ equation, Eq. (63), to obtain

$$\dot{p}_{n_1}=\Big\{i\mathscr{V}_1^*\rho_{51}+i\mathscr{V}_{54}^*\rho_{54}+i\mathscr{V}_{21}^*\rho_{21} -i\,|\mathscr{E}_1|^2(n+1)(k_{aa}^-\rho_{44}+k_{aa}^+\rho_{55}+k_{bb}^-\rho_{11}+k_{aa}^+\rho_{22}) +\frac{i}{2}(\mathscr{V}_{17}\rho_{71}+\mathscr{V}_{05}\rho_{50})-(n_1\to n_1-1)\Big\}+\text{c.c.}\ , \tag{99}$$

where

$$\mathscr{V}_{17}=-k_{ab}\mathscr{E}_1^*\mathscr{E}_3^*\sqrt{n_1+1}\sqrt{n_3}$$

and

$$\mathscr{V}_{05}=-k_{ab}\mathscr{E}_1^*\mathscr{E}_3^*\sqrt{n_1+1}\sqrt{n_3+1}\ .$$

We proceed as before, solving Eqs. (98)–(101) in steady state. In particular, for ρ_{51} we obtain

$$\rho_{51}\,|_{a_3}=i\mathscr{D}_1(\mathscr{V}_2d_{154}+\mathscr{V}_{01}\rho_{50}-\mathscr{V}_{57}\rho_{71})\ . \tag{100}$$

We eliminate the ρ_{54} term in Eq. (100) by using

$$P_{0010}=\langle n_1n_3\,|P\,|\,n_1+1\,n_3\rangle=\rho_{54}+\rho_{21}\ .$$

The equation for d_{154} in this case becomes

$$\begin{aligned}d_{154}\,|_{a_3}=&-\mathscr{F}P_{0010}-i\mathscr{D}_a-2i\mathscr{V}_2^*\mathscr{D}_1(\mathscr{V}_2d_{154}+\mathscr{V}_{01}\rho_{50}-\mathscr{V}_{57}\rho_{71})\\&-2i\mathscr{V}_2\mathscr{D}_3^*(\mathscr{V}_2^*d_{154}+\mathscr{V}_{23}\rho_{34}+\mathscr{V}_{27}\rho_{74}-\mathscr{V}_{64}\rho_{26}-\mathscr{V}_{04}\rho_{20})\\&+(\mathscr{V}_{57}\rho_{74}-\mathscr{V}_{64}\rho_{56}-\mathscr{V}_{04}\rho_{50}-\mathscr{V}_{23}\rho_{31}-\mathscr{V}_{27}\rho_{71}+\mathscr{V}_{01}\rho_{20})\ ,\end{aligned} \tag{101}$$

which can be solved to yield

$$\begin{aligned}d_{154}\,|_{a_3}=&\frac{-\mathscr{F}P_{0010}-i\mathscr{D}_a[-2i\mathscr{V}_2^*\mathscr{D}_1(\mathscr{V}_{01}\rho_{50}-\mathscr{V}_{57}\rho_{71})]}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\\&-\frac{i\mathscr{D}_a[-2i\mathscr{V}_2\mathscr{D}_3^*(\mathscr{V}_{23}\rho_{34}+\mathscr{V}_{27}\rho_{74}-\mathscr{V}_{64}\rho_{26}-\mathscr{V}_{04}\rho_{20})]}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\\&+\frac{\mathscr{V}_{57}\rho_{74}-\mathscr{V}_{64}\rho_{56}-\mathscr{V}_{04}\rho_{50}-\mathscr{V}_{23}\rho_{31}-\mathscr{V}_{27}\rho_{71}+\mathscr{V}_{01}\rho_{20}}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\ .\end{aligned} \tag{102}$$

The equation for $\dot{P}_{0010}$ is also complicated by the additional potentials. We define $P_{0010}\,|_{a_3}$ to be the part of P_{0010} involving only the coupled-mode terms. Using Eqs. (99) and (100) we obtain

$$\dot{P}_{0010}\,|_{a_3}=-i\Delta P_{0010}\,|_{a_3}-i\mathscr{V}_{57}\rho_{74}+i\mathscr{V}_{64}\rho_{56}+i\mathscr{V}_{04}\rho_{50}-i\mathscr{V}_{23}\rho_{31}-i\mathscr{V}_{27}\rho_{71}+i\mathscr{V}_{01}\rho_{20}\ , \tag{103}$$

and hence

$$P_{0010}\,|_{a_3}=\frac{i(-\mathscr{V}_{57}\rho_{74}+\mathscr{V}_{64}\rho_{56}+\mathscr{V}_{04}\rho_{50}-\mathscr{V}_{23}\rho_{31}-\mathscr{V}_{27}\rho_{71}+\mathscr{V}_{01}\rho_{20})}{i\Delta}\ , \tag{104}$$

where we again assume the field varies little in a time $1/\Delta$. Substituting Eqs. (100) and (102) into (103) and making use of Eqs. (81) and (82) yields

$$\begin{aligned}\dot{p}_{n_1}\,|_{a_3}=\Bigg\{&i\mathscr{V}_1^*[i\mathscr{D}_1(\mathscr{V}_{01}\rho_{50}-\mathscr{V}_{57}\rho_{71})]+\frac{i}{2}(\mathscr{V}_{54}^*+\mathscr{V}_{21}^*)P_{0010}\,|_{a_3}\\&+[\mathscr{V}_1^*+\tfrac{1}{2}(\mathscr{V}_{54}^*-\mathscr{V}_{21}^*)]\frac{-\mathscr{F}P_{0010}\,|_{a_3}-i\mathscr{D}_a[-2i\mathscr{V}_2^*\mathscr{D}_1(\mathscr{V}_{01}\rho_{50}-\mathscr{V}_{57}\rho_{71})]}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\\&-\frac{i\mathscr{D}_a[-2i\mathscr{V}_2\mathscr{D}_3^*(\mathscr{V}_{23}\rho_{34}+\mathscr{V}_{27}\rho_{74}-\mathscr{V}_{64}\rho_{26}-\mathscr{V}_{04}\rho_{20})]}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\\&+\frac{\mathscr{V}_{57}\rho_{74}-\mathscr{V}_{64}\rho_{56}-\mathscr{V}_{04}\rho_{50}-\mathscr{V}_{23}\rho_{31}-\mathscr{V}_{27}\rho_{71}+\mathscr{V}_{01}\rho_{20}}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}+\frac{i}{2}(\mathscr{V}_{17}\rho_{71}+\mathscr{V}_{05}\rho_{50})-(n_1\to n_1-1)\Bigg\}+\text{c.c.}\end{aligned} \tag{105}$$

Analogous to Ref. 1, Eqs. (98) and (99), we introduce the C_1 and D_1 coefficients by letting

$$\dot{p}_{n_1}\,|_{a_3}=\left[-\sum_{n_3}\sqrt{n_1+1}(C_1\sqrt{n_3}P_{0-110}-D_1\sqrt{n_3+1}P_{0011})\right.$$
$$\left.+\sum_{n_3}\sqrt{n_1}(C_1\sqrt{n_3}P_{-1-100}-D_1\sqrt{n_3+1}P_{-1001})\right]+\text{c.c.} \tag{106}$$

The coupled mode matrix elements ρ_{71}, ρ_{74}, etc., are found in exactly the same manner as in Ref. 1. Because they are always multiplied by the weak double sidemode interaction potential, they are calculated in steady state from the strong mode alone. Thus the atomic and field dependencies factor and

$$\rho_{71}=\tilde{R}_{ab}P_{0-110} \tag{107a}$$
$$\rho_{34}=\tilde{R}_{ba}P_{0-110} \tag{107b}$$
$$\rho_{31}=R_{bb}P_{0-110} \tag{107c}$$
$$\rho_{74}=R_{aa}P_{0-110} \tag{107d}$$
$$\rho_{50}=\tilde{R}_{ab}P_{0011} \tag{107e}$$
$$\rho_{26}=\tilde{R}_{ba}P_{0011} \tag{107f}$$
$$\rho_{20}=R_{bb}P_{0011} \tag{107g}$$
$$\rho_{56}=R_{aa}P_{0011}\;. \tag{107h}$$

Selecting the proper terms in Eq. (105) by comparing to Eq. (106), with the help of Eqs. (107), we have, after substituting the expressions for the interaction potentials and the atomic density-matrix components,

$$C_1=-\frac{|k_{ab}|\,\mathscr{E}_1^*\mathscr{E}_3^*}{2(1+I_2^2\mathscr{L}_2)}\frac{\mathscr{E}_2^2}{\mathscr{E}_s^2}\left[\frac{\sqrt{\gamma\Gamma}\mathscr{D}_2}{2}(1+2i\sqrt{\gamma\Gamma}I_2\mathscr{D}_1k_{aa}/|k_{ab}|)-\sqrt{\gamma\Gamma}\frac{I_2\mathscr{F}(\gamma I_2\mathscr{D}_1-i\omega_s/\Gamma)}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right.$$
$$\times\left[\frac{\mathscr{D}_2}{2}-\mathscr{D}_3^*f_a+i\sqrt{\gamma\Gamma}\frac{I_2(k_{bb}\mathscr{D}_2^*\mathscr{D}_3^*+k_{aa}\mathscr{D}_1\mathscr{D}_2)}{2|k_{ab}|}-\frac{i}{\sqrt{\gamma\Gamma}}\frac{k_{bb}f_b-k_{aa}f_a}{I_2|k_{ab}|}\right]$$
$$+\frac{-\sqrt{\gamma\Gamma}I_2\mathscr{D}_2/2+i(k_{aa}f_a+k_{bb}f_b)/|k_{ab}|}{i\Delta}$$
$$\left.\times\left[\frac{\mathscr{F}(\gamma\Gamma I_2\mathscr{D}_1-i\omega_s)}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}-i\sqrt{\gamma\Gamma}\frac{k_{aa}+k_{bb}}{|k_{ab}|}\right]\right], \tag{108}$$

$$D_1=-\frac{|k_{ab}|\,\mathscr{E}_1^*\mathscr{E}_3^*}{2(1+I_2^2\mathscr{L}_2)}\frac{\mathscr{E}_2^2}{\mathscr{E}_s^2}\left[-\frac{\sqrt{\gamma\Gamma}\mathscr{D}_2}{2}(1-2i\sqrt{\gamma\Gamma}I_2\mathscr{D}_1k_{bb}/|k_{ab}|)-\sqrt{\gamma\Gamma}\frac{I_2\mathscr{F}(\gamma I_2\mathscr{D}_1-i\omega_s/\Gamma)}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right.$$
$$\times\left[-\frac{\mathscr{D}_2}{2}-\mathscr{D}_3^*f_b+i\sqrt{\gamma\Gamma}\frac{I_2(k_{aa}\mathscr{D}_2^*\mathscr{D}_3^*+k_{bb}\mathscr{D}_1\mathscr{D}_2)}{2|k_{ab}|}-\frac{i}{\sqrt{\gamma\Gamma}}\frac{k_{bb}f_b-k_{aa}f_a}{I_2|k_{ab}|}\right]$$
$$+\frac{-\sqrt{\gamma\Gamma}I_2\mathscr{D}_2/2+i(k_{aa}f_a+k_{bb}f_b)/|k_{ab}|}{i\Delta}$$
$$\left.\times\left[\frac{\mathscr{F}(\gamma\Gamma I_2\mathscr{D}_1-i\omega_s)}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}-i\sqrt{\gamma\Gamma}\frac{k_{aa}+k_{bb}}{|k_{ab}|}\right]\right]. \tag{109}$$

We express our results for both the single and double sidemode coefficients by combining Eq. (86) and its coupled-mode addition, Eq. (106), and converting to an operator equation. This also may be obtained from Eq. (66). We find that the equation of motion for the reduced field operator P is given by

$$\dot{P}=\{[-A_1(Pa_1a_1^\dagger-a_1^\dagger Pa_1)-B_1(a_1^\dagger a_1P-a_1Pa_1^\dagger)+C_1(a_1^\dagger a_3^\dagger P-a_3^\dagger Pa_1^\dagger)+D_1(Pa_3^\dagger a_1^\dagger-a_1^\dagger Pa_3^\dagger)]+(1\to 3)\}+\text{H.c.} \tag{110}$$

which has exactly the same form as Eq. (100) of Ref. 1.

We may recover the semiclassical three-mode coupling coefficient $-i\kappa_1^*$ by computing the difference C_1-D_1, just as in the single photon calculation. We find

$$C_1-D_1=-\frac{|k_{ab}|\,\mathscr{E}_1^*\mathscr{E}_3^*}{2(1+I_2^2\mathscr{L}_2)}\frac{\mathscr{E}_2^2}{\mathscr{E}_s^2}\sqrt{\Gamma/\gamma}\times\left[\gamma\mathscr{D}_2(1-i\omega_s\mathscr{D}_1)-\frac{\gamma I_2\mathscr{F}(\gamma I_2\mathscr{D}_1-i\omega_s/\Gamma)}{1+I_2^2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\left[\mathscr{D}_2+\mathscr{D}_3^*+\frac{i\omega_sI_2}{2}(\mathscr{D}_2^*\mathscr{D}_3^*-\mathscr{D}_1\mathscr{D}_2)\right]\right], \tag{111}$$

which agrees with the calculation in Ref. 11.

Figure 11 depicts $C_1+C_1^*=C$, Eq. (108) plus its complex conjugate, for $I_2=10$, $T_2=2T_1$, $\omega_s=0$, and $\omega=2\nu_2$. The shape of the curve closely resembles the one-photon result of Fig. 8 of Ref. 4. Note, however, the slight asymmetry of the spectral dependence, even for a zero Stark shift. From Eq. (108) we see this arises from the $\mathscr{D}_2$ term in the first line, which is absent in the one-photon case, Eq. (98) of Ref. 1. This term results from the presence of the $\mathscr{V}_{17}$ potential in the $\dot{P}$ equation, and is due to the presence of all of the off-resonant j levels. In the semiclassical two-photon laser instability work of Ref. 14, this is interpreted to result from a scattering off the field induced two-photon coherence R_{ab}, and it is shown in that work to have significant consequences. When a Stark shift is introduced, the spectral dependence is significantly altered. This is depicted in Fig. 12 for $\omega_sI_2=5/T_1$, $\omega-2\nu_2=-5/T_1$, and $I_2=10$. In comparison to Fig. 11, the central peak is practically unchanged, one sideband is amplified, and the other is reduced, reminiscent of Fig. 6 for the A coefficient. The "balancing" effect of the Stark shift versus the detuning is also evident for this case.

In conclusion, this paper generalizes our quantum theory of multiwave mixing to the two-photon two-level model. The resulting expressions for the four coefficients A_1, B_1, C_1, and D_1 are all considerably more complex than their one-photon counterparts due to the increased number of interaction potentials and the appearance of the dynamic Stark shifts. We have calculated and discussed the two-photon resonance fluorescence spectrum, and have shown that for vanishing Stark shifts, the spectrum is very similar to the one-photon result, but that with a Stark shift present, the spectrum becomes quite different.

Because the $\dot{P}$ equation, Eq. (110), has the same for this two-photon level scheme, all of the solutions derived in Refs. 4 and 5 also apply to the two-photon two-level model. Thus coefficients derived in this paper, Eqs. (89),

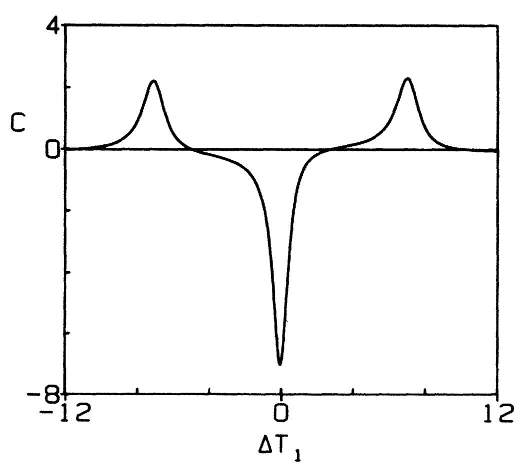

FIG. 11. C_1+c.c.$=C$ vs ΔT_1 for $I_2=10$, $\omega-2\nu_2=\omega_s=0$, and $T_2=2T_1$.

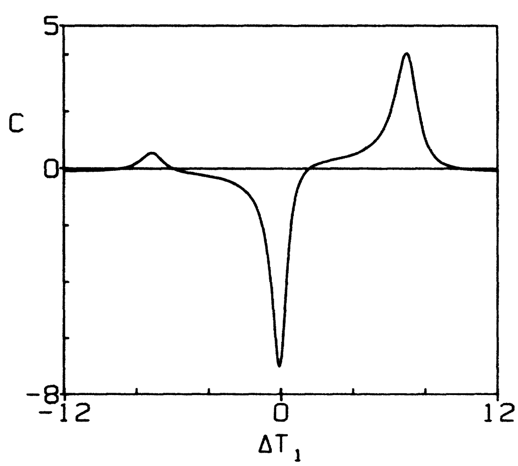

FIG. 12. C vs ΔT_1 for $I_2=10$, $T_2=2T_1$, $\omega-2\nu_2=5T_1^{-1}$, and $\omega_sI_2=5T_1^{-1}$.

(90), (108), and (109) can be used in the formulas of those references to predict cavity and propagation effects for two-photon two-level media. Due to the increased complexity of the two-photon model, more complicated calculations would be required to analyze this case, especially if a thorough understanding of the role of the Stark shifts is desired. The theory derived in this paper, however, makes this potential work possible.

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