Effects of ionization and cascade decay on two-photon two-level interactions

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We extend the semiclassical two-photon two-level model to include upper-level ionization and upper-level decay via an intermediate level. We examine the effects of these relaxation mechanisms on multiwave interactions, finding the generalized one- and two-wave two-photon absorption coefficients and the three-wave coupling coefficient. These mechanisms cause asymmetries to appear in the absorption spectra in addition to those caused by Stark shifts. The Stark-shift term becomes more complicated and fails to vanish even when it does in the simpler model. For recombination rates and ionization decay rates comparable to the coherence decay rate, coherent dips fill in.

I. INTRODUCTION

Two-photon two-level interactions receive much attention from both theorists and experimentalists. A frequently used model[1–3] assumes two-level atoms where transitions between the two levels are not dipole allowed but can occur via nonresonant intermediate states. The recent work of Sargent *et al.*[4] presents a semiclassical theory of two-photon multiwave mixing for this model for an arbitrarily strong pump field and weak signal and conjugate fields. A corresponding theory with quantized probe and conjugate fields has been given by Holm and Sargent.[5]

Most experiments with two-level media have made use of a near-resonant intermediate level (see, for example, Allen *et al.*[6]) to aid transitions between the two main levels. For example, the $5p$ level in the $5s$ to $5d$ transition of Rb and the $3p$ level of the $3s$ to $3d$ transition in Na both enhance the corresponding two-photon transition probabilities. In the present paper, we extend the theory of Ref. 4 to allow for nonzero population in such an intermediate level. Specifically the upper level a decays into the intermediate level l, which, in turn, decays to the lower level b. Level l is sufficiently nonresonant that its population is not directly affected by the two-photon field connecting levels a and b. We show that the existence of such a level can alter the absorption spectrum due to population of this level. A more general cascade decay scheme for the one-photon case has been examined by Sargent[7] and found to cause changes in the population-difference decay time as well as in the population pulsation factor. We find that for two-photon transitions, similar changes occur and in addition the intermediate level causes the Stark shift to become more complicated and inherently nonzero. Sargent *et al.*[4] showed how Stark shifts cause the absorption spectra to become asymmetric. Thus it is not surprising that we find addition of the third populated level causes asymmetries in the spectra.

Multiphoton ionization is currently receiving a lot of attention in laser spectroscopy.[8–10] In this paper we also allow for upper-level ionization as an additional decay path in order to study the effect of this decay process on the complex absorption and coupling coefficients. Other authors have treated the energy spectrum of the continuum electrons[10] and coherent effects in multiple-level systems.[8] In this work we use a very simple model ignoring the structure of the continuum and assuming recombination to the lower level. This approximates a cascade of decays through the intermediate levels by one straight to the lower level. Since we expect the number of recombinations to be very small, we let the recombination rate in general be much lower than any of the other rates. Simple first-order perturbation theory describing the photoionization as is commonly used in astrophysics[11] assumes the ionization rate is intensity dependent. We show that this effect can lead to significant changes in the complex absorption spectrum.

In Sec. II of this paper we derive the polarization of the two-level medium including the two new upper-level decay paths. We assume a simple ionization decay rate proportional to the intensity. In Sec. III we derive the corresponding single-mode steady-state complex absorption. In Sec. IV we allow for two sidemodes and a pump wave with different frequencies and solve for the sidemode polarization. In Sec. V we calculate the single sidemode complex probe-absorption spectrum and in Sec. VI we discuss three-wave mixing. Section VII illustrates the effects of the new decay processes on the complex absorption and coupling factor spectra in different limits. Due to the new degrees of freedom allowed by these new processes the effects can be quite complicated. For this reason we examine them independently by allowing either ionization decay or the third level to become populated. We examine both the short-coherence-lifetime limit and that for comparable coherence and population difference lifetimes. We also look in both the low- and high-intensity limits.

II. POLARIZATION OF THE MEDIUM

In general the polarization of the medium with the level scheme in Fig. 1 is given by

$$P(\mathbf{r},t)=\mathrm{tr}(\mathscr{O}\rho)=\sum_j(\wp_{aj}\rho_{ja}+\wp_{bj}\rho_{jb})+\text{c.c.}\,, \qquad (1)$$

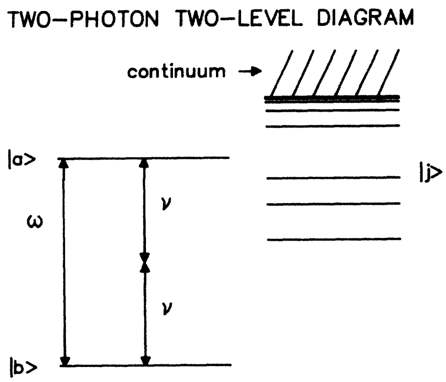

FIG. 1. Two-photon two-level model including continuum.

where $\wp_{aj}$ is the electric-dipole matrix element between the a and j states, $\mathcal{O}$ is the atomic electric dipole operator, and ρ_{aj} is the population matrix element between a and j. Since $a \leftrightarrow b$ is a two-photon transition, $\wp_{ab}$ vanishes. We consider cases in which the polarization (1) is induced by the electric field

$$E(\mathbf{r},t)=\frac{1}{2}\mathcal{E}(\mathbf{r},t)e^{-i\nu t}+\text{c.c.}\ , \tag{2}$$

where $\mathcal{E}(\mathbf{r},t)$ varies little in a time $1/\nu$, but has rapid spatial variations like $\exp(i\mathbf{K}\cdot\mathbf{r})$. This field induces the polarization

$$P(\mathbf{r},t)=\tfrac{1}{2}\mathcal{P}(\mathbf{r},t)e^{-i\nu t}+\text{c.c.}\ , \tag{3}$$

where the complex polarization $\mathcal{P}(\mathbf{r},t)$ also varies little in the time $1/\nu$. Combining Eqs. (1) and (3), we find

$$\mathcal{P}(\mathbf{r},t)=2\sum_j(\wp_{aj}\rho_{ja}+\wp_{bj}\rho_{jb}+\text{c.c.})e^{i\nu t}\ , \tag{4}$$

where we keep only terms varying little in an optical frequency period $(1/\nu)$.

The electric-dipole coherences ρ_{ja} are induced by the interaction energies

$$\mathcal{V}_{ja}=-\frac{1}{2\hbar}\wp_{ja}[\mathcal{E}(\mathbf{r},t)e^{-i\nu t}+\text{c.c.}]\ , \tag{5}$$

with a similar formula for ρ_{jb}. Using the general Schrödinger equation of motion

$$\dot{\rho}_{ij}=-(\gamma_{ij}+i\omega_{ij})\rho_{ij}-i[\mathcal{V},\rho]_{ij}\ , \tag{6}$$

we have

$$\dot{\rho}_{ja}=-(\gamma_{ja}+i\omega_{ja})\rho_{ja}+\frac{i}{2\hbar}[\wp_{ja}(\rho_{aa}-\rho_{jj}\delta_{lj})+\wp_{jb}\rho_{ba}]\times(\mathcal{E}e^{-i\nu t}+\mathcal{E}^*e^{i\nu t})\ , \tag{7}$$

and

$$\dot{\rho}_{jb}=-(\gamma_{jb}+i\omega_{jb})\rho_{jb}+\frac{i}{2\hbar}[\wp_{jb}(\rho_{bb}-\rho_{jj}\delta_{lj})+\wp_{ja}\rho_{ab}]\times(\mathcal{E}e^{-i\nu t}+\mathcal{E}^*e^{i\nu t})\ , \tag{8}$$

where $\hbar\omega_{ij}=\hbar(\omega_i-\omega_j)$ is the energy difference between levels i and j, γ_{ij} is the corresponding decay constant, and l identifies the intermediate state that may acquire population. We see that wherever we had ρ_{aa} in Ref. 4 we now have $\rho_{aa}-\rho_{jj}\delta_{lj}$ and similarly $\rho_{bb}\rightarrow\rho_{bb}-\rho_{jj}\delta_{lj}$.

We integrate Eqs. (7) and (8) to first order in $\mathcal{V}$ without making a rotating-wave approximation (RWA), since ν differs substantially from all $\pm\omega_{ja}$ and $\pm\omega_{jb}$. Using the ω_{ij} to drop the lower limit of integration, and setting $\rho_{ba}=R_{ba}e^{2i\nu t}$, where R_{ba} varies little in an optical frequency period, we have

$$\begin{aligned}\rho_{ja}&=\frac{i}{2\hbar}\int_{-\infty}^{t}dt'(\mathcal{E}e^{-i\nu t'}+\mathcal{E}^*e^{i\nu t'})\exp[-(\gamma_{ja}+i\omega_{ja})(t-t')][\wp_{ja}(\rho_{aa}-\rho_{jj}\delta_{lj})+\wp_{jb}R_{ba}e^{2i\nu t'}]\\&=\frac{1}{2\hbar}\left[\frac{\mathcal{E}e^{-i\nu t}}{\omega_{ja}-\nu}+\frac{\mathcal{E}^*e^{i\nu t}}{\omega_{ja}+\nu}\right]\wp_{ja}(\rho_{aa}-\rho_{jj}\delta_{lj})+\frac{1}{2\hbar}\left[\frac{\mathcal{E}e^{i\nu t}}{\omega_{ja}+\nu}+\frac{\mathcal{E}^*e^{3i\nu t}}{\omega_{ja}+3\nu}\right]\wp_{jb}R_{ba}\ .\end{aligned} \tag{9}$$

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

$$\omega_{ja}+\nu\simeq\omega_{jb}-\nu\ , \tag{10}$$

which allows us to replace $\omega_{ja}+3\nu$ in Eq. (9) by $\omega_{jb}+\nu$. Similarly integrating Eq. (8), we find

$$\rho_{jb}=\frac{1}{2\hbar}\left[\frac{\mathcal{E}e^{-i\nu t}}{\omega_{jb}-\nu}+\frac{\mathcal{E}^*e^{i\nu t}}{\omega_{jb}+\nu}\right]\wp_{jb}(\rho_{bb}-\rho_{jj}\delta_{lj})+\frac{1}{2\hbar}\left[\frac{\mathcal{E}e^{-3i\nu t}}{\omega_{ja}-\nu}+\frac{\mathcal{E}^*e^{-i\nu t}}{\omega_{jb}-\nu}\right]\wp_{ja}R_{ab}\ . \tag{11}$$

Substituting Eqs. (9) and (11) into the polarization (4) we have

$$\begin{aligned}\mathcal{P}=\frac{1}{\hbar}\Bigg[\sum_j&|\wp_{aj}|^2\left[\frac{\mathcal{E}e^{-i\nu t}}{\omega_{ja}-\nu}+\frac{\mathcal{E}^*e^{i\nu t}}{\omega_{jb}-\nu}\right](\rho_{aa}-\rho_{jj}\delta_{lj})+\wp_{aj}\wp_{jb}\left[\frac{\mathcal{E}e^{i\nu t}}{\omega_{ja}+\nu}+\frac{\mathcal{E}^*e^{3i\nu t}}{\omega_{ja}+3\nu}\right]R_{ba}\\&+|\wp_{aj}|^2\left[\frac{\mathcal{E}^*e^{i\nu t}}{\omega_{ja}-\nu}+\frac{\mathcal{E}e^{-i\nu t}}{\omega_{jb}-\nu}\right](\rho_{aa}-\rho_{jj}\delta_{lj})+\wp_{aj}^*\wp_{jb}^*\left[\frac{\mathcal{E}^*e^{-i\nu t}}{\omega_{ja}+\nu}+\frac{\mathcal{E}e^{-3i\nu t}}{\omega_{ja}+3\nu}\right]R_{ba}\Bigg]e^{i\nu t}\\&+\frac{1}{\hbar}\Bigg[\sum_j|\wp_{bj}|^2\left[\frac{\mathcal{E}e^{-i\nu t}}{\omega_{jb}-\nu}+\frac{\mathcal{E}^*e^{i\nu t}}{\omega_{jb}+\nu}\right](\rho_{bb}-\rho_{jj}\delta_{lj})+\wp_{bj}\wp_{ja}\left[\frac{\mathcal{E}e^{-3i\nu t}}{\omega_{ja}-\nu}+\frac{\mathcal{E}^*e^{-i\nu t}}{\omega_{ja}+\nu}\right]R_{ab}\\&+|\wp_{jb}|^2\left[\frac{\mathcal{E}^*e^{i\nu t}}{\omega_{jb}-\nu}+\frac{\mathcal{E}e^{-i\nu t}}{\omega_{jb}+\nu}\right](\rho_{bb}-\rho_{jj}\delta_{lj})+\wp_{bj}^*\wp_{ja}^*\left[\frac{\mathcal{E}^*e^{3i\nu t}}{\omega_{ja}-\nu}+\frac{\mathcal{E}e^{i\nu t}}{\omega_{jb}-\nu}\right]R_{ba}\Bigg]e^{i\nu t}\ .\end{aligned} \tag{12}$$

Now keeping only terms that vary little in the time $1/\nu$, we have

$$\mathscr{P}=\frac{1}{\hbar}\left[\sum_j |\wp_{aj}|^2\left[\frac{\mathscr{E}}{\omega_{ja}-\nu}+\frac{\mathscr{E}}{\omega_{jb}-\nu}\right](\rho_{aa}-\rho_{jj}\delta_{lj})+\frac{\wp^*_{aj}\wp^*_{jb}\mathscr{E}^*R_{ab}}{\omega_{ja}+\nu}\right.$$

$$\left.+|\wp bj|^2\left[\frac{\mathscr{E}}{\omega_{jb}-\nu}+\frac{\mathscr{E}}{\omega_{jb}+\nu}\right](\rho_{bb}-\rho_{jj}\delta_{lj})+\frac{\wp_{bj}\wp_{ja}\mathscr{E}^*R_{ab}}{\omega_{jb}-\nu}\right]$$

$$=\frac{\mathscr{E}}{\hbar}\sum_j\left[|\wp_{aj}|^2\frac{2\omega_{ja}}{\omega_{ja}^2-\nu^2}(\rho_{aa}-\rho_{jj}\delta_{lj})+|\wp_{bj}|^2\frac{2\omega_{jb}}{\omega_{jb}^2-\nu^2}(\rho_{bb}-\rho_{jj}\delta_{lj})\right]+\frac{2}{\hbar}\sum_j\frac{\wp^*_{aj}\wp^*_{jb}\mathscr{E}^*R_{ab}}{\omega_{jb}-\nu}\ . \quad (13)$$

Performing the summation in Eq. (13) we obtain the slowly varying polarization of the medium

$$\mathscr{P}=\mathscr{E}(k_{aa}\rho_{aa}+k_{bb}\rho_{bb}-k'_{ll}\rho_{ll})+2\mathscr{E}^*k^*_{ab}\rho_{ab}e^{2i\nu t}\ , \quad (14)$$

where the two-photon coefficients k_{ab}, k_{aa}, and k_{bb} are given by the usual values

$$k_{ab}=\hbar^{-1}\sum_j\wp_{aj}\wp_{jb}/(\omega_{jb}-\nu)$$

$$\simeq\hbar^{-1}\sum_j\wp_{aj}\wp_{jb}/(\omega_{ja}+\nu)\ , \quad (15)$$

$$k_{aa}=2\hbar^{-1}\sum_j|\wp_{ja}|^2\omega_{ja}/(\omega_{ja}^2-\nu^2)\ , \quad (16)$$

$$k_{bb}=2\hbar^{-1}\sum_j|\wp_{jb}|^2\omega_{jb}/(\omega_{jb}^2-\nu^2)\ , \quad (17)$$

and k'_{ll} is given by

$$k'_{ll}=2\hbar^{-1}\left[|\wp_{al}|^2\frac{\omega_{la}}{\omega_{la}^2-\nu^2}+|\wp_{bl}|^2\frac{\omega_{lb}}{\omega_{lb}^2-\nu^2}\right]. \quad (18)$$

Note that the sum over j implicitly includes an integral over all j levels in the continuum. We see the ρ_{ll} term adds an extra nonlinearity to the index similar to the $k_{aa}\rho_{aa}$ term. Using Eqs. (9) and (11), we now derive the equations of motion for the level populations ρ_{aa}, ρ_{bb}, ρ_{ll}, the continuum population N^+, and the coherence ρ_{ab} using the two-photon rotating-wave approximation, i.e., we neglect terms like $1/[\gamma+i(\omega+2\nu)]$ compared to $1/[\gamma+i(\omega-2\nu)]$. According to Eq. (6), we have

$$\dot\rho_{ab}=-(\gamma+i\omega)\rho_{ab}-i\sum_j(\mathscr{V}_{aj}\rho_{jb}-\rho_{aj}\mathscr{V}_{jb})\ , \quad (19)$$

$$\dot\rho_{aa}=-(\gamma_a+\gamma_I I)\rho_{aa}-\sum_j(i\mathscr{V}_{aj}\rho_{ja}+\text{c.c.})\ , \quad (20)$$

$$\dot\rho_{ll}=\gamma_a\rho_{aa}-\gamma_l\rho_{ll}\ , \quad (21)$$

$$\dot\rho_{bb}=\gamma_l\rho_{ll}+\lambda N^+-i\sum_j(\mathscr{V}_{bj}\rho_{jb}-\rho_{bj}\mathscr{V}_{jb})\ , \quad (22)$$

and

$$\dot N^+=\gamma_I I\rho_{aa}-\lambda N^+\ . \quad (23)$$

where N^+ is the population of the continuum and $\gamma_I I$ represents the decay rate to the continuum which is proportional to the intensity I (in units of the saturation intensity) and has the decay constant γ_I. The recombination rate is λ which is generally small. For the steady-state solution (i.e., single mode) we can set

$$\dot\rho_{ll}=\dot N^+=0\ , \quad (24)$$

which then gives us $\rho_{ll}=\gamma_a\rho_{aa}/\gamma_l$ and $N^+=\gamma_I I\rho_{aa}/\lambda$. These together with

$$\rho_{aa}+\rho_{bb}+\rho_{ll}+N^+=N \quad (25)$$

give us ρ_{aa} and ρ_{bb} in terms of the population difference $D=\rho_{aa}-\rho_{bb}$,

$$\rho_{aa}=\frac{D+N}{2+\gamma_a/\gamma_l+\gamma_I I/\lambda}\ , \quad (26)$$

and

$$\rho_{bb}=\frac{N-D(1+\gamma_a/\gamma_l+\gamma_I I/\lambda)}{2+\gamma_a/\gamma_l+\gamma_I I/\lambda}\ . \quad (27)$$

It is not true in general that the populations in level l and in the continuum will come to equilibrium before the other populations. However we make that assumption here merely for the purpose of deriving an approximate population difference decay time T_1. Thus, we find the population difference equation of motion

$$\dot D=-(\gamma_a+\gamma_I I)\rho_{aa}-\gamma_l\rho_{ll}-\lambda N^+$$

$$-\sum_j[i(\mathscr{V}_{aj}\rho_{ja}+\mathscr{V}_{bj}\rho_{jb})+\text{c.c.}]$$

$$=-2(\gamma_a+\gamma_I I)\rho_{aa}-\sum_j[i(\mathscr{V}_{aj}\rho_{ja}+\mathscr{V}_{bj}\rho_{jb})+\text{c.c.}]\ . \quad (28)$$

Substituting in the coherence equations (9) and (11) we then obtain

$$\dot D=-2(\gamma_a+\gamma_I I)\rho_{aa}+\frac{1}{2\hbar}(i\mathscr{E}^2k_{ab}\rho_{ba}e^{-2i\nu t}+\text{c.c.})\ , \quad (29)$$

which in terms of D becomes

$$\dot D=\frac{-2(\gamma_a+\gamma_I I)(D+N)}{2+\gamma_a/\gamma_l+\gamma_I I/\lambda}$$

$$+\frac{1}{2\hbar}(i\mathscr{E}^2k_{ab}e^{-2i\nu t}\rho_{ba}+\text{c.c.})\ . \quad (30)$$

We can write this in terms of a "population difference decay time" T_I analogous to the traditional NMR name T_1,

$$T_I=\frac{2+\gamma_a/\gamma_l+\gamma_I I/\lambda}{2(\gamma_a+\gamma_I I)}\ , \tag{31}$$

$$\dot{D}=-\frac{D+N}{T_I}+\frac{1}{2\hbar}(i\mathscr{E}^2 k_{ab}e^{-2i\nu t}\rho_{ba}+\text{c.c.})\ . \tag{32}$$

We note that T_I differs from the normal value for T_1 in that it is intensity dependent. For zero intensity we recover the cascade population difference decay time

$$T_1=\tfrac{1}{2}(2\gamma_a^{-1}+\gamma_l^{-1})\ , \tag{33}$$

found by Sargent.[7] Note that this expression for $\dot{D}$ agrees with that of Sargent *et al.*[4] provided T_1 is replaced by T_I. Substituting the coherence equations (9) and (11) into (19), we have

$$\dot{\rho}_{ab}=-(\gamma+i\omega+i\omega_s I)\rho_{ab}-i(k_{ab}\mathscr{E}^2/4\hbar)e^{-2i\nu t}D\ , \tag{34}$$

where the two-photon dimensionless intensity

$$I=|k_{ab}\mathscr{E}^2|(T_1T_2)^{1/2}/2\hbar\equiv|\mathscr{E}/\mathscr{E}_s|^2\ , \tag{35}$$

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

$$\omega_s=(k_{bb}-k_{aa})/2|k_{ab}|(T_1T_2)^{1/2}\ . \tag{36}$$

This expression for $\dot{\rho}_{ab}$ also agrees with Eq. (20) of Ref. 4. Here I is written in units of decay processes independent of intensity.

For single-frequency operation, we can solve these equations in the rate-equation approximation. Specifically, we assume $\mathscr{E}$ and D vary little in the two-photon coherence decay time T_1, allowing Eq. (34) to be formally integrated with the value

$$\rho_{ab}=-i(k_{ab}\mathscr{E}^2/4\hbar)\mathscr{D}(\omega+\omega_s I-2\nu)De^{-2i\nu t}\ , \tag{37}$$

where the complex denominator

$$\mathscr{D}(\Delta)=(\gamma+i\Delta)^{-1}\ . \tag{38}$$

Substituting this into Eq. (32), we have

$$\dot{D}=-(D+N)/T_I-2RD\ , \tag{39}$$

where the rate constant

$$\begin{aligned}R&=|k_{ab}\mathscr{E}^2|^2\mathscr{L}(\omega+\omega_s I-2\nu)/8\gamma\hbar^2\\&=\tfrac{1}{2}I^2\mathscr{L}(\omega+\omega_s I-2\nu)/T_1\ ,\end{aligned} \tag{40}$$

and the Lorentzian

$$\mathscr{L}(\Delta)=1/[1+(\Delta/\gamma)^2]\ . \tag{41}$$

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

$$D=-N/[1+I^2\mathscr{L}(\omega+\omega_s I-2\nu)T_I/T_1]\ . \tag{42}$$

Substituting this into Eq. (37) we have

$$\rho_{ab}=i\frac{N(k_{ab}\mathscr{D}\mathscr{E}^2)}{4\hbar}\frac{e^{-2i\nu t}}{1+I^2\mathscr{L}T_I/T_1}\ . \tag{43}$$

Equations (42) and (43) are identical with those in Ref. 4 except for the different saturation factor $S=(1+I^2\mathscr{L}T_I/T_1)^{-1}$. This term is identical with the normal saturation factor $(1+I^2\mathscr{L})^{-1}$ in both the low- and high-intensity limits. However, in between these limits it is enhanced (i.e., less saturation) or decreased (more saturation) depending on the relative values of T_I and T_1. Thus when

$$\gamma_a>\frac{2\lambda}{1-\lambda/\gamma_l}\ , \tag{44}$$

the medium is easier to saturate and when the inequality in (44) does not hold it is harder to saturate. In particular, we see that in the limit $\gamma_l\to\lambda$, it is always harder to saturate, which is expected because of the extra decay path. In Fig. (2) we have shown this new saturation factor ($\gamma_I/\gamma_a=0.01$) along with the standard one ($\gamma_I/\gamma_a=0$) as a function of intensity for $\gamma_a=\gamma_l=1$ and $\lambda=\gamma_a/100$ where all decay constants are in units of $2/T_2$. This new saturation factor follows through to all our results.

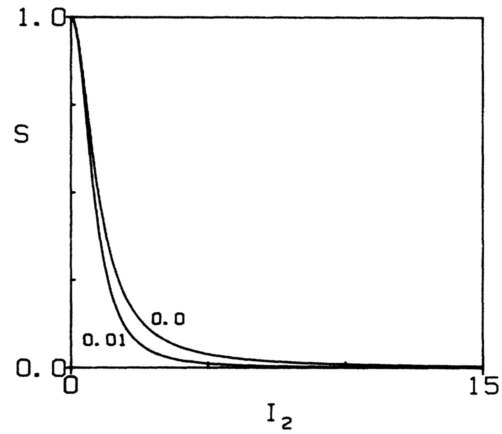

FIG. 2. Saturation factor $S=(1+I_2^2\mathscr{L}_2T_I/T_1)^{-1}$ vs I_2 for $\gamma_a=\gamma_l=2\gamma$, $\lambda=\gamma_a/100$, and γ_I/γ_a increases from 0 to 0.01.

Substituting Eq. (43) along with Eqs. (26) and (27) into the polarization (14), we have

$$\mathscr{P}(\mathbf{r})=\mathscr{E}\left[\frac{(k_{aa}+k_{bb}-k_{ll})(D+N)}{2T_I(\gamma_a+\gamma_I I)}-k_{bb}D\right]-\frac{i\mathscr{E}|k_{ab}|I\mathscr{D}D}{(T_1T_2)^{1/2}}\ , \tag{45}$$

where $k_{ll}=\gamma_a k_{ll}'/\gamma_l$. Using Eq. (42) for D we can write this as

$$\mathscr{P}(\mathbf{r})=N\mathscr{E}\left[\frac{k_{aa}+k_{bb}-k_{ll}}{2T_I(\gamma_a+\gamma_I I)}+\frac{|k_{ab}|(T_2/T_1)^{1/2}}{1+I^2\mathscr{L}T_I/T_1}[\Omega(0)T_1+iI\gamma\mathscr{D}]\right]\ , \tag{46}$$

where we now introduce the generalized Stark-shift parameter

$$\Omega(0)=\frac{k_{aa}-k_{ll}+k_{bb}[1-2T_I(\gamma_a+\gamma_I I)]}{-2T_I(\gamma_a+\gamma_I I)|k_{ab}|(T_1T_2)^{1/2}}\ . \tag{47}$$

Although considerably more complicated this expression reduces to ω_s of the two-level, nonionization model of Ref. 4 when $T_I \equiv \gamma_I^{-1}$ and γ_I both go to zero. In this limit k_{ll} also goes to zero and we recover Eq. (22) of Ref. 4. Stark shifts are known to cause asymmetries in spectra. The fact that this new term is a function of intensity is significant because, while it may equal zero for a given combination of the k coefficients and for a specific intensity, it does not equal zero in general. This parameter also appears in the multimode case where it is complicated by being a function of Δ, the detuning between the modes. For our single-mode case $\Delta=0$.

III. SINGLE RUNNING-WAVE SATURATION

A simple special case of the electric field (2) is that for the single-frequency running wave [Fig. 3(a)]

$$E(z,t)=\tfrac{1}{2}A_2(z)e^{i(K_2 z-\nu_2 t)}+\text{c.c.}\ , \tag{48}$$

where $A_2(z)$ varies little in an optical wavelength and $K_2=\nu_2/c$. We use mode 2, since in multiwave mixing mode 2 is the large-intensity mode. This field induces the polarization

$$P(z,t)=\tfrac{1}{2}\mathscr{P}_2(z)e^{i(K_2 z-\nu_2 t)}+\text{c.c.}\ , \tag{49}$$

where $\mathscr{P}_2(z)$ too varies little in a wavelength. We substitute Eqs. (48) and (49) without complex conjugates into the wave equation

$$\frac{\partial^2 E}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2 E}{\partial t^2}=\frac{1}{\epsilon_0 c^2}\frac{\partial^2 P}{\partial t^2}\ , \tag{50}$$

neglect small terms like $\partial^2 A_2/\partial z^2$, etc., equate coefficients of $e^{i(K_2 z-\nu_2 t)}$, and find the propagation equation

$$\frac{dA_2(z)}{dz}=i\frac{K_2}{2\epsilon_0}\mathscr{P}_2(z)\ . \tag{51}$$

Writing this in the complex Beer's-law form, we have

$$\frac{dA_2(z)}{dz}=-\alpha_2 A_2(z)\ , \tag{52}$$

where

$$\alpha_2=-i\frac{K_2}{2\epsilon_0}\mathscr{P}_2(z)/A_2(z)\ . \tag{53}$$

For the two-photon polarization (46), this gives

$$\alpha_2=-i\frac{K_2 N}{4\epsilon_0}\frac{k_{aa}+k_{bb}-k_{ll}}{T_I(\gamma_a+\gamma_I I_2)}+\alpha_0\frac{I_2\gamma\mathscr{D}_2-i\Omega(0)T_1}{1+I_2^2\mathscr{L}_2 T_I/T_1}\ , \tag{54}$$

where for typographical simplicity it is assumed that $I\to I_2$ in the expression for T_I. The real part of α_2 determines the absorption in the medium. This is given by

$$\text{Re}(\alpha_2)=\alpha_0\frac{\gamma^2 I_2}{\gamma^2(1+I_2^2T_I/T_1)+(\omega+\omega_s I_2-2\nu_2)^2}\ , \tag{55}$$

where the two-photon absorption parameter

$$\alpha_0=K_2 N\,|\,k_{ab}\,|\,(T_2/T_1)^{1/2}/2\epsilon_0\ . \tag{56}$$

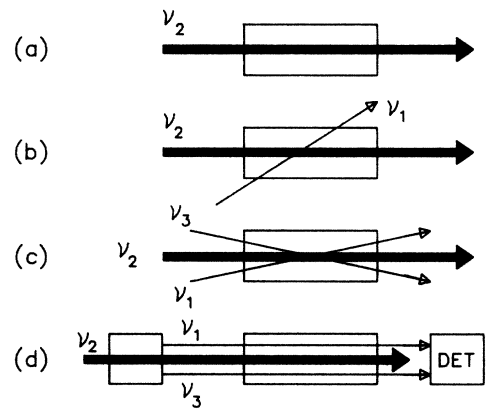

FIG. 3. One-, two-, and three-wave two-photon interactions treated in this paper.

The imaginary part of α_2 adds to the wave vector K_2 and hence changes the index of refraction. It has the value

$$\text{Im}(\alpha_2)=-\frac{K_2 N}{4\epsilon_0}\frac{k_{aa}+k_{bb}-k_{ll}}{T_I(\gamma_a+\gamma_I I_2)}-\alpha_0\frac{(\omega+\omega_s I_2-2\nu)I_2\mathscr{L}_2/\gamma+\Omega(0)T_1}{1+I_2^2\mathscr{L}_2T_I/T_1}\ . \tag{57}$$

IV. TWO- AND THREE-WAVE POLARIZATIONS

We now suppose the electric field has the form

$$E(\mathbf{r},t)=\tfrac{1}{2}[\mathscr{E}_1(\mathbf{r})e^{i\Delta t}+\mathscr{E}_2(\mathbf{r})+\mathscr{E}_3(\mathbf{r})e^{-i\Delta t}]e^{-i\nu_2 t}+\text{c.c.}\ , \tag{58}$$

i.e., the slowly varying complex field amplitude $\mathscr{E}(\mathbf{r},t)$ in Sec. II is given by

$$\mathscr{E}(\mathbf{r},t)=\mathscr{E}_1(\mathbf{r})e^{i\Delta t}+\mathscr{E}_2(\mathbf{r})+\mathscr{E}_3(\mathbf{r})^{-i\Delta t}\ . \tag{59}$$

$\mathscr{E}_2$ is the pump-wave amplitude, while $\mathscr{E}_1$ and $\mathscr{E}_3$ are typically the signal and conjugate waves, respectively, and $\Delta=\nu_2-\nu_1$ is the pump-signal detuning. Similarly, we write the corresponding induced polarization in Eq. (3) as

$$\mathscr{P}(\mathbf{r},t)=\mathscr{P}_1(\mathbf{r})e^{i\Delta t}+\mathscr{P}_2(\mathbf{r})+\mathscr{P}_3(\mathbf{r})e^{-i\Delta t}\ . \tag{60}$$

Here we assume both $\mathscr{E}_1$ and $\mathscr{E}_3$ are sufficiently small that other Fourier components of the polarization are negligible. As discussed in the single-photon theory,[7] this occurs if $\mathscr{P}(\mathbf{r})$ is a linear function of $\mathscr{E}_1$ and $\mathscr{E}_3$. To determine $\mathscr{P}$, we need to solve the polarization (14) and equations of motion (30) and (34) to first order in $\mathscr{E}_1$ and $\mathscr{E}_3$, while keeping all orders in the pump amplitude $\mathscr{E}_2$. As in the one-photon, two-level problem (see Ref. 7), it is clear that the field (59) induces a set of Fourier components in the two-photon coherence ρ_{ab} and in the populations ρ_{aa}, ρ_{bb}, ρ_{ll}, and N^+, as well as in the probability difference D. Here we expand these as

$$\rho_{ab}=e^{-2i\nu_2 t}\sum_m p_m e^{im\Delta t}\ , \tag{61}$$

$$\rho_{\alpha\alpha}=\sum_k n_{\alpha k}e^{ik\Delta t},\quad \alpha=a,b,l \tag{62}$$

$$N^+=\sum_k n_{pk}e^{ik\Delta t}, \tag{63}$$

and

$$D=\sum_k d_k e^{ik\Delta t}, \tag{64}$$

where obviously $d_k=n_{ak}-n_{bk}$.

We note however that the population difference equation of motion (30) no longer holds because the populations are no longer in steady state but rather are responding to the beat frequency between the fields with population pulsations. In fact the populations act like forced damped harmonic oscillators of resonant frequency zero.[7] For example, in Eq. (21) the term $-\gamma_l\rho_{ll}$ acts as the damping term while $\gamma_a\rho_{aa}$ acts as the forcing term. Driving an oscillator off resonance gives rise to phase shifts and we show later how these phase shifts between the population Fourier components affect the probe absorption. We need equations relating the n_{ak}. Substituting Eq. (62) into Eq. (21) we obtain

$$n_{lk}=\gamma_a\mathscr{D}_l(k\Delta)n_{ak}\,. \tag{65}$$

Thus we see the population in level l responds to being driven by level a with a complex phase shift contained in the $\mathscr{D}_l$ term.

We next obtain an equation for the n_{pk} which is more complicated due to the fact that Eq. (23) contains a term with I. The assumption that $\mathscr{E}_1$ and $\mathscr{E}_3$ are weak allows us to let

$$\mathscr{E}^2\simeq\mathscr{E}_2(\mathscr{E}_2+2\mathscr{E}_1e^{i\Delta t}+2\mathscr{E}_3e^{-i\Delta t}), \tag{66}$$

and

$$|\mathscr{E}|^2=|\mathscr{E}_2|^2+[(\mathscr{E}_1\mathscr{E}_2^*+\mathscr{E}_2\mathscr{E}_3^*)e^{i\Delta t}+\text{c.c.}] \tag{67}$$

so that

$$I=I_2+\mathscr{I}e^{i\Delta t}+\mathscr{I}^*e^{-i\Delta t}, \tag{68}$$

where

$$\mathscr{I}=\mathscr{E}_s^{-2}(\mathscr{E}_1\mathscr{E}_2^*+\mathscr{E}_2\mathscr{E}_3^*)\,. \tag{69}$$

Substituting expansion (63) along with Eqs. (66)–(68) into Eq. (23) and equating exponents of $e^{i\Delta t}$ gives

$$n_{pk}=\gamma_I\mathscr{D}_\lambda(k\Delta)(I_2n_{ak}+\mathscr{I}n_{ak-1}+\mathscr{I}^*n_{ak+1}), \tag{70}$$

where $\mathscr{D}_\lambda(k\Delta)=(\lambda+ik\Delta)^{-1}$. The Fourier components of the continuum population not only exhibit a phase shift relative to the corresponding Fourier component of the upper-level population, but also a dependence (with a phase shift) on the other Fourier components of level a. This dependence arises due to the beat terms in the intensity [Eq. (68)]. By substituting expansions (61)–(64) along with Eqs. (65) and (70) into Eq. (25) we obtain the following equation for the d_k:

$$d_k=-N\delta_{0k}+B_2(k\Delta)n_{ak}+\gamma_I\mathscr{D}_\lambda(k\Delta)(\mathscr{I}n_{ak-1}+\mathscr{I}^*n_{ak+1}), \tag{71}$$

where the complex dimensionless factor

$$B_2(k\Delta)=2+\gamma_a\mathscr{D}_l(k\Delta)+\gamma_I\mathscr{D}_\lambda(k\Delta)I_2 \tag{72}$$

can be interpreted as the phase lag of the kth Fourier component of level a relative to the kth component of the population difference.

Substituting Eq. (71) and the expansions of Eqs. (62) and (64) into the polarization equation (14) we then obtain

$$\mathscr{P}=\mathscr{E}\sum_k e^{ik\Delta t}\{[k_{aa}+k_{bb}-k'_{ll}\gamma_a\mathscr{D}_l(k\Delta)][d_k+N\delta_{0k}-\gamma_I\mathscr{D}_\lambda(k\Delta)(\mathscr{I}n_{ak-1}+\mathscr{I}^*n_{ak+1})]/B_2(k\Delta)-k_{bb}d_k\}+2\mathscr{E}^*k_{ab}^*\sum_m p_me^{im\Delta t}\,. \tag{73}$$

The assumption that $\mathscr{E}_1$ and $\mathscr{E}_3$ are weak limits these expansions to the nine Fourier coefficients $d_{\pm1},d_0$, $n_{a\pm1},n_{a0}$, and $p_{\pm1},p_0$. By equating the exponents of $e^{i\Delta t}$ in Eq. (73) we find

$$\begin{aligned}\mathscr{P}_1(\mathbf{r})=&\frac{\mathscr{E}_1}{B_2(0)}(\{k_{aa}+k_{bb}[1-B_2(0)]-k_{ll}\}d_0+(k_{aa}+k_{bb}-k_{ll})N)\\&+\frac{\mathscr{E}_2}{B_2(\Delta)}(-[k_{aa}+k_{bb}-k'_{ll}\gamma_a\mathscr{D}_l(\Delta)]\gamma_I\mathscr{D}_\lambda(\Delta)\mathscr{I}n_{a0}\\&\qquad+\{k_{aa}+k_{bb}[1-B_2(\Delta)]-k'_{ll}\gamma_a\mathscr{D}_l(\Delta)\}d_1)+2k_{ab}^*(\mathscr{E}_2^*p_1+\mathscr{E}_3^*p_0)\,.\end{aligned} \tag{74}$$

Using Eq. (71) without reference to $\mathscr{E}_1$ or $\mathscr{E}_3$ since they cannot saturate the response we obtain the following expression for n_{a0},

$$n_{a0}=\frac{d_0+N}{B_2(0)}, \tag{75}$$

which then gives us the following expression for $\mathscr{P}_1$ in terms of d_0, d_1, p_0, and p_1:

$$\mathscr{P}_1=\frac{N\mathscr{E}_1}{B_2(0)}(k_{aa}+k_{bb}-k_{ll})+|k_{ab}|\left[\frac{T_2}{T_1}\right]^{1/2}$$
$$\times\left\{-\Omega(0)\mathscr{E}_1T_1d_0+\frac{\gamma_I\mathscr{D}_\lambda(\Delta)\mathscr{I}\mathscr{E}_2}{B_2(0)}\left[\Omega(\Delta)T_1-\frac{k_{bb}}{|k_{ab}|}\left[\frac{T_1}{T_2}\right]^{1/2}\right](N+d_0)-\Omega(\Delta)\mathscr{E}_2T_1d_1\right\}$$
$$+2k_{ab}^*(\mathscr{E}_2^*p_1+\mathscr{E}_3^*p_0)\;, \tag{76}$$

where the generalized Stark-shift parameter, which as promised is now a function of Δ, is

$$\Omega(\Delta)=-\frac{k_{aa}+k_{bb}[1-B_2(\Delta)]-k_{ll}'\gamma_a\mathscr{D}_l(\Delta)}{B_2(\Delta)\,|k_{ab}|\,(T_1T_2)^{1/2}}\;. \tag{77}$$

Again we see that when $\gamma_I\to\infty$ and $\gamma_I\to 0$, $B_2(\Delta)\to 2$ and $\Omega(\Delta)\to\omega_s$. It is significant to note that the Stark shift becoming Δ dependent is not solely due to the ionization as can be seen by letting $\gamma_I\to 0$. We then still have the same expression only now $B_2(\Delta)$ simplifies to $2+\gamma_a\mathscr{D}_l(\Delta)$. $\Omega(\Delta)$ may equal zero for a specific value of Δ but not in general. Thus, either of these decay mechanisms causes in effect a built-in Stark shift.

Similar to above we can equate the exponents of $e^{-i\Delta t}$ in Eq. (73) and obtain the expression for $\mathscr{P}_3$

$$\mathscr{P}_3=\frac{N\mathscr{E}_3}{B_2(0)}(k_{aa}+k_{bb}-k_{ll})+|k_{ab}|\left[\frac{T_2}{T_1}\right]^{1/2}$$
$$\times\left\{-\Omega(0)\mathscr{E}_3T_1d_0+\frac{\gamma_I\mathscr{D}_\lambda(-\Delta)\mathscr{I}\mathscr{E}_2}{B_2(0)}\left[\Omega(-\Delta)T_1-\frac{k_{bb}}{|k_{ab}|}\left[\frac{T_1}{T_2}\right]^{1/2}\right](N+d_0)-\Omega(-\Delta)\mathscr{E}_2T_1d_{-1}\right\}$$
$$+2k_{ab}^*(\mathscr{E}_2^*p_{-1}+\mathscr{E}_1^*p_0)\;. \tag{78}$$

To find the required p_m we substitute expansions (61) and (64) along with Eqs. (66) and (67) into Eq. (34). Equating coefficients of $\exp(-2i\nu_2t+mi\Delta t)$ we find

$$p_m=-i\frac{k_{ab}}{4\hbar}\mathscr{D}_{2-m}(\mathscr{E}_2^2d_m+2\mathscr{E}_1\mathscr{E}_2d_{m-1}+2\mathscr{E}_2\mathscr{E}_3d_{m+1})$$
$$-i\omega_s\mathscr{D}_{2-m}(\mathscr{I}p_{m-1}+\mathscr{I}^*p_{m+1})\;, \tag{79}$$

where the two-photon complex denominator (differs from single-photon definition)

$$\mathscr{D}_{2-m}=1/[\gamma+i(\omega+\omega_sI_2-2\nu_2+m\Delta)]\;. \tag{80}$$

We solve for p_0 without reference to $\mathscr{E}_1$ or $\mathscr{E}_3$, since they cannot saturate the response. Hence p_0 is given by the coefficient of $e^{-2i\nu t}$ in (37) with $\mathscr{E}$ replaced by $\mathscr{E}_2$,

$$p_0=-i(k_{ab}\mathscr{E}_2^2/4\hbar)\mathscr{D}_2d_0\;. \tag{81}$$

Keeping only terms linear in $\mathscr{E}_1$ and $\mathscr{E}_3$ and their complex conjugates and noting that $d_{\pm1}$ must already have such a field amplitude, we find from (79)

$$p_1=-i\frac{k_{ab}}{4\hbar}\mathscr{D}_1[\mathscr{E}_2^2d_1+(2\mathscr{E}_1\mathscr{E}_2-i\omega_s\mathscr{D}_2\mathscr{I}\mathscr{E}_2^2)d_0] \tag{82}$$

and

$$p_{-1}=-i\frac{k_{ab}}{4\hbar}\mathscr{D}_3[\mathscr{E}_2^2d_{-1}+(2\mathscr{E}_2\mathscr{E}_3-i\omega_s\mathscr{D}_2\mathscr{I}^*\mathscr{E}_2^2)d_0]\;. \tag{83}$$

To obtain d_k we first solve for the n_{ak} by substituting expressions (61) and (62) into Eq. (20) and then relate the d_k to the n_{ak} using Eq. (71). Thus

$$\sum_k(ik\Delta+\gamma_a+\gamma_II_2)n_{ak}e^{ik\Delta t}$$
$$=-\gamma_1(\mathscr{I}e^{i\Delta t}+\mathscr{I}^*e^{-i\Delta t})\sum_m n_{am}e^{im\Delta t}$$
$$+\left[\frac{ik_{ab}/4\hbar}{ik\Delta+\gamma_a+\gamma_II_2}(\mathscr{E}_2^2+2\mathscr{E}_1\mathscr{E}_2e^{i\Delta t}+2\mathscr{E}_3\mathscr{E}_2e^{-i\Delta t})\right.$$
$$\left.\times\sum_m p_m^*e^{-im\Delta t}+\text{c.c.}\right]\;. \tag{84}$$

Similar to the case for p_0, we obtain n_{a0} from Eq. (84) ignoring terms containing $\mathscr{E}_1$ or $\mathscr{E}_3$,

$$n_{a0}=\frac{1}{\gamma_a+\gamma_II_2}\left[\frac{ik_{ab}}{4\hbar}\mathscr{E}_2^2p_0^*+\text{c.c.}\right]\;. \tag{85}$$

Together with Eq. (81) this gives

$$n_{a0}=-\frac{I_2^2\mathscr{L}_2d_0}{2T_1(\gamma_a+\gamma_II_2)}\;, \tag{86}$$

where $\mathscr{L}_2$ is the $m=0$ case of the Lorentzian

$$\mathscr{L}_{2-m}=\frac{1}{1+(\omega+\omega_sI_2-2\nu_2+m\Delta)^2/\gamma^2}\;. \tag{87}$$

Now, using Eqs. (86) and (71) we can solve for d_0 to obtain

$$d_0=-\frac{N}{1+I_2^2\mathscr{L}_2T_I/T_1}\;, \tag{88}$$

where again it is assumed that $I \rightarrow I_2$ in the expression for T_I. Now returning to Eqs. (84) and (71) and retaining the terms linear in $\mathscr{E}_1$ and $\mathscr{E}_3$ we obtain a general expression for d_k,

$$d_k = -N\delta_{0k} + (\mathscr{I} n_{ak-1} + I^* n_{ak+1})\gamma_I[\mathscr{D}_\lambda(k\Delta) - B_2(k\Delta)\mathscr{D}_I(k\Delta)] + B_2(k\Delta)\mathscr{D}_I(k\Delta)\left[\frac{ik_{ab}}{4\hbar}(\mathscr{E}_2^2 p^*_{-k} + 2\mathscr{E}_1\mathscr{E}_2 p^*_{-k+1} + 2\mathscr{E}_3\mathscr{E}_2 p^*_{-k-1}) - \frac{ik^*_{ab}}{4\hbar}(\mathscr{E}_2^{*2} p_k + 2\mathscr{E}_1^*\mathscr{E}_2^* p_{k+1} + 2\mathscr{E}_3^*\mathscr{E}_2^* p_{k-1})\right], \tag{89}$$

where we have defined $\mathscr{D}_I(k\Delta) = (ik\Delta + \gamma_a + \gamma_I I_2)^{-1}$. Comparing Eq. (89) with Eq. (57) in Ref. 4 allows us to identify $B_2(k\Delta)\mathscr{D}_I(k\Delta)/2$ with $T_1\mathscr{F}(k\Delta)$. This then gives us the following generalized expression for the complex population pulsation factor $\mathscr{F}(k\Delta)$,

$$\mathscr{F}(k\Delta) = \frac{B_2(k\Delta)\mathscr{D}_I(k\Delta)}{2T_1} = \frac{2 + \gamma_a \mathscr{D}_I(k\Delta) + \gamma_I I_2 \mathscr{D}_\lambda(k\Delta)}{(2\gamma_a^{-1} + \gamma_I^{-1})(ik\Delta + \gamma_a + \gamma_I I_2)}. \tag{90}$$

We note that in the limit $\gamma_I \rightarrow 0$ we recover the $\mathscr{F}(k\Delta)$ found by Sargent.[7] Using $\mathscr{F}(k\Delta)$ in Eq. (89) gives for d_1 and d_{-1},

$$d_1 = -\gamma_I \mathscr{I}[\mathscr{D}_\lambda(\Delta) - 2T_1\mathscr{F}(\Delta)]\frac{I_2^2 \mathscr{L}_2 d_0}{2T_1(\gamma_a + \gamma_I I_2)} + 2T_1\mathscr{F}(\Delta)\left[\frac{ik_{ab}}{4\hbar}\mathscr{E}_2(\mathscr{E}_2 p^*_{-1} + 2\mathscr{E}_1 p_0^*) - \frac{ik^*_{ab}}{4\hbar}\mathscr{E}_2^*(\mathscr{E}_2^* p_1 + 2\mathscr{E}_3^* p_0)\right], \tag{91}$$

and $d_{-1} = d_1^*$. Substituting Eqs. (82) and (83) into (91) and solving for d_1, we find

$$d_1 = -d_0 I_2 \mathscr{E}_s^{-2}\frac{\mathscr{E}_1\mathscr{E}_2^* f_1 + \mathscr{E}_2\mathscr{E}_3^* f_3^*}{1 + I_2^2\mathscr{F}(\Delta)\frac{\gamma}{2}(\mathscr{D}_1 + \mathscr{D}_3^*)}, \tag{92}$$

where $\mathscr{E}_s$ is given by Eq. (35) and

$$f_1 = \gamma\mathscr{F}(\Delta)\left[\mathscr{D}_1 + \mathscr{D}_2^* + \tfrac{1}{2}i\omega_s I_2(\mathscr{D}_2^*\mathscr{D}_3^* - \mathscr{D}_1\mathscr{D}_2) - \frac{\gamma_I I_2 \mathscr{L}_2}{\gamma(\gamma_a + \gamma_I I_2)}\left[1 - \frac{\mathscr{D}_\lambda(\Delta)}{2T_1\mathscr{F}(\Delta)}\right]\right] \tag{93}$$

and $f_3^* = f_1^*(-\Delta)$.

Substituting p_0 and p_1 of Eqs. (81) and (82) into the polarization (76) we find

$$\mathscr{P}_1(\mathbf{r}) = \frac{N\mathscr{E}_1}{B_2(0)}(k_{aa} + k_{bb} - k_{ll}) + |k_{ab}|(T_2/T_1)^{1/2}\left[\frac{\gamma_I \mathscr{D}_\lambda(\Delta)\mathscr{I}\mathscr{E}_2}{B_2(0)}\left[\Omega(\Delta)T_1 - \frac{k_{bb}}{|k_{ab}|}(T_1/T_2)^{1/2}\right](N + d_0) - id_1\mathscr{E}_2[I_2\gamma\mathscr{D}_1 - i\Omega(\Delta)T_1] + d_0\{-\Omega(0)T_1\mathscr{E}_1 - i\mathscr{E}_2\mathscr{E}_s^{-2}\gamma[\mathscr{E}_1\mathscr{E}_2^*\mathscr{D}_1(2 - i\omega_s I_2\mathscr{D}_2) + \mathscr{E}_2\mathscr{E}_3^*\mathscr{D}_2(1 - i\omega_s I_2\mathscr{D}_1)]\}\right]. \tag{94}$$

Substituting Eq. (91) along with Eqs. (81)–(83) into Eq. (94), we have

$$\mathscr{P}_1(\mathbf{r}) = \frac{N\mathscr{E}_1}{B_2(0)}(k_{aa} + k_{bb} - k_{ll}) + \frac{N|k_{ab}|}{1 + I_2^2\mathscr{L}_2 T_I/T_1}\left[\frac{T_2}{T_1}\right]^{1/2}\left\{\frac{I_2^2\mathscr{L}_2 T_I/T_1}{B_2(0)}\gamma_I\mathscr{D}_\lambda(\Delta)\mathscr{I}\mathscr{E}_2\left[\Omega(\Delta)T_1 - \frac{k_{bb}}{|k_{ab}|}\left[\frac{T_1}{T_2}\right]^{1/2}\right] + \Omega(0)T_1\mathscr{E}_1 + i\mathscr{E}_2\mathscr{E}_s^{-2}[\mathscr{E}_1\mathscr{E}_2^*\gamma\mathscr{D}_1(2 - i\omega_s I_2\mathscr{D}_2) + \mathscr{E}_2\mathscr{E}_3^*\gamma\mathscr{D}_2(1 - i\omega_s I_2\mathscr{D}_1)] - i\mathscr{E}_2\mathscr{E}_s^{-2}I_2[I_2\gamma\mathscr{D}_1 - i\Omega(\Delta)T_1]\frac{\mathscr{E}_1\mathscr{E}_2^* f_1 + \mathscr{E}_2\mathscr{E}_3^* f_3^*}{1 + I_2^2\mathscr{F}(\Delta)\frac{\gamma}{2}(\mathscr{D}_1 + \mathscr{D}_3^*)}\right\}, \tag{95}$$

$\mathscr{P}_3(\mathbf{r})$ is given by interchanging the subscripts 1 and 3 on the $\mathscr{E}$'s, $\mathscr{D}$'s, and f's and replacing Δ by $-\Delta$.

V. TWO-WAVE, TWO-PHOTON ABSORPTION

Consider first the simple two-wave case of the field (59) [Fig. 3(b)]

$$E(\mathbf{r},t)=\tfrac{1}{2}(A_1e^{i\mathbf{K}_1\cdot\mathbf{r}+i\Delta t}+A_2e^{i\mathbf{K}_2\cdot\mathbf{r}})e^{-i\nu_2 t}+\text{c.c.} \tag{96}$$

Although this does induce a conjugate polarization at the frequency $\nu_3=\nu_2+\Delta$, we assume that either the length of the interaction is small enough, or the angle between $\mathbf{K}_1$ and $\mathbf{K}_2$ is large enough so that the ν_3 component is not phase matched and therefore can be neglected (this becomes clearer in the three-wave discussion). Substituting Eq. (96) into the wave equation (50) and projecting on $\exp(-i\mathbf{K}_1\cdot\mathbf{r})$, we find the propagation Eq. (51), where the temporally and spacially slowly varying

$$\mathscr{P}_1(z)=\frac{K_1}{2\pi}\int_0^{2\pi/K_1}d\zeta\, e^{-iK_1(z+\zeta)}\mathscr{P}_1(\mathbf{r}) , \tag{97}$$

where $\mathbf{K}_1\cdot\mathbf{r}=K_1z$.

Substituting Eq. (95) into Eq. (97), neglecting the $\mathscr{E}_3^*$ terms and using Eq. (53), we find the complex absorption coefficient

$$\begin{aligned}\alpha_1=&\frac{-i\alpha_0(k_{aa}+k_{bb}-k_{ll})}{|k_{ab}|B_2(0)(T_2/T_1)^{1/2}}-\frac{i\alpha_0\gamma_I\mathscr{D}_\lambda(\Delta)I_2^3\mathscr{L}_2T_I/T_1}{B_2(0)(1+I_2^2\mathscr{L}_2T_I/T_1)}\left[\Omega(\Delta)T_1-\frac{k_{bb}}{|k_{ab}|}\left[\frac{T_1}{T_2}\right]^{1/2}\right]\\&+\frac{\alpha_0}{1+I_2^2\mathscr{L}_2T_I/T_1}\left\{-i\Omega(0)T_1+I_2\gamma\mathscr{D}_1(2-i\omega_sI_2\mathscr{D}_2)-I_2^2\frac{[I_2\gamma\mathscr{D}_1-i\Omega(\Delta)T_1]\mathscr{F}(\Delta)\gamma}{1+I_2^2\mathscr{F}(\Delta)\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right.\\&\left.\times\left[\mathscr{D}_1+\mathscr{D}_2^*+\frac{1}{2}i\omega_sI_2(\mathscr{D}_2^*\mathscr{D}_3^*-\mathscr{D}_1\mathscr{D}_2)-\frac{\gamma_II_2\mathscr{L}_2}{\gamma(\gamma_a+\gamma_II_2)}\left[1-\frac{\mathscr{D}_\lambda(\Delta)}{2T_1\mathscr{F}(\Delta)}\right]\right]\right\} ,\end{aligned} \tag{98}$$

where α_0 is given by Eq. (56) as $KN\,|k_{ab}|(T_2/T_1)^{1/2}/2\epsilon_0$. It is useful to compare this result to the absorption coefficient of Sargent *et al.*[4] In addition to the previously discussed changes in $\mathscr{F}(\Delta)$, T_1, $\Omega(\Delta)$, and the saturation factor, we have two entirely new terms. Of the previously mentioned effects, all but the new saturation factor arise from the addition of the population in level l and are further complicated by the ionization route. The new saturation factor, like these other two new factors, arises solely due to the ionization. The first of the new terms [the second term in Eq. (98)] affects the incoherent part of the expression while the second [last term in Eq. (98)] affects the coherent part.

VI. THREE-WAVE MIXING

We now consider three-wave phase conjugation diagrammed in Fig. 3(c). This also pertains to modulation spectroscopy of two-photon media [Fig. 3(d)] and to two-photon laser and optical bistability instabilities.[12] The signal (at frequency $\nu_1=\nu_2-\Delta$) and conjugate (at frequency $\nu_3=\nu_2+\Delta$) in combination with the pump (at frequency ν_2) induce polarizations affecting one another. If the angles and frequencies between the waves are small enough, these induced polarizations are phase matched to the corresponding waves, leading to a coupled-mode problem. For this case, we write the field (59) as

$$E(\mathbf{r},t)=\tfrac{1}{2}(A_1e^{i\mathbf{K}_1\cdot\mathbf{r}+i\Delta t}+A_2e^{i\mathbf{K}_2\cdot\mathbf{r}}+A_3e^{i\mathbf{K}_3\cdot\mathbf{r}-i\Delta t})e^{-i\nu_2 t}+\text{c.c.} , \tag{99}$$

in which $\mathbf{K}_1$, $\mathbf{K}_2$, and $\mathbf{K}_3$ are nearly parallel to one another as depicted in Fig. 3(c).

Substituting this field with the corresponding polarization into the wave equation (50) and projecting onto $\exp(-i\mathbf{K}_1\cdot\mathbf{r})$, we find

$$\frac{dA_1}{dz}=-\alpha_1A_1+\chi_1A_3^*\exp[i(2\mathbf{K}_2-\mathbf{K}_1-\mathbf{K}_3)\cdot\mathbf{r}] , \tag{100}$$

where α_1 is given by (98), z is taken along $\mathbf{K}_2$ for convenience (technically this z value should be divided by $\mathbf{K}_2\cdot\mathbf{K}_1/K_2K_1$) and the coupling coefficient (which is often called $-i\kappa_1^*$)

$$\chi_1 = \frac{iA_2^2}{\mathscr{E}_s^2} \frac{\alpha_0 \gamma_I \mathscr{D}_\lambda(\Delta) I_2^2 \mathscr{L}_2 T_I / T_1}{B_2(0)(1 + I_2^2 \mathscr{L}_2 T_I / T_1)} \left[\Omega(\Delta) T_1 - \frac{k_{bb}}{|k_{ab}|} \left[\frac{T_1}{T_2} \right]^{1/2} \right]$$

$$- \frac{\alpha_0}{1 + I_2^2 \mathscr{L}_2 T_I / T_1} \frac{A_2^2}{\mathscr{E}_s^2} \left\{ \gamma \mathscr{D}_2 (1 - i\omega_s I_2 \mathscr{D}_1) - \frac{I_2 [I_2 \gamma \mathscr{D}_1 - i\Omega(\Delta) T_1] \mathscr{F}(\Delta) \gamma}{1 + I_2^2 \mathscr{F}(\Delta) \frac{\gamma}{2} (\mathscr{D}_1 + \mathscr{D}_3^*)} \right.$$

$$\left. \times \left[\mathscr{D}_3^* + \mathscr{D}_2 + \tfrac{1}{2} i\omega_s I_2 (\mathscr{D}_2^* \mathscr{D}_3^* - \mathscr{D}_1 \mathscr{D}_2) - \frac{\gamma_I I_2 \mathscr{L}_2}{\gamma(\gamma_a + \gamma_I I_2)} \left[1 - \frac{\mathscr{D}_\lambda(\Delta)}{2 T_1 \mathscr{F}(\Delta)} \right] \right] \right\} .$$

In the limit that $\gamma_l^{-1} = \gamma_I = \omega_s = 0$, this answer is analogous to the one-photon χ_1 except for some extra factors of I_2 and the first term proportional to $\mathscr{D}_2$. This term results from the term in the polarization $\mathscr{P}_1$ [Eq. (74)] proportional to $\mathscr{E}_3^* p_0$ and arises in the two-photon case due to conjugate scattering off the pump-induced two-photon coherence ρ_{ab}. In the one-photon case this extra degree of freedom does not arise and all components of $\mathscr{P}_1$ are proportional to p_1. However, here the field $\mathscr{E}_3^*$ can interact with p_0 and give the correct frequency dependence for $\mathscr{P}_1$.

VII. EFFECTS OF NEW DECAY PATHS

In this section we examine the effects of the new decay paths on the probe absorption and coupling coefficients. We look in several different coherence-lifetime and intensity limits. The ionization decay and the cascade decay cause different types of effects so we examine the two independently.

The coherent-dip limit is when the coherence lifetime is short relative to the population difference lifetime. In this limit and for a low-intensity field, the probe-absorption spectrum develops a dip of approximate width $2(1 + I_2^2)/T_1$ caused by the limited bandwidth of the population pulsations and thus provides a means of measuring T_1. Decreasing γ_l causes the dip to turn into a dispersive-type feature. This is shown in Fig. 4(a) where we have plotted the real part of the complex probe-absorption spectrum versus pump-probe detuning in units of $1/T_2$ for the case where $T_1 = 10T_2$ and $I_2 = 1$. We let $\gamma_I T_2 = 0$ and decrease $\gamma_l T_2$ from 10 000 to 0.1. We have previously shown how atomic detuning and Stark shifts cause the coherent dip to become asymmetric.[13] Here the asymmetry is due to a generalized Stark shift $\Omega(\Delta)$ caused by population in the intermediate level. As γ_l decreases, the population of level l increases and the phase lag it experiences has more influence on the spectrum. Hence the dispersive characteristic increases. In the presence of ionization decay the coherent dip also becomes asymmetric. This is shown in Fig. 4(b) where we let $\gamma_l T_2 \rightarrow 10\,000$ so we can just examine the effect of increasing ionization decay. We increase $\gamma_I T_2$ from 0 to 0.1 letting $\lambda T_2 = 0.01$ since in general we do not expect many recombinations. The coherent dip remains but becomes more dispersive due to the influence of the phase-shifted continuum, and the magnitude of the absorption peak decreases due to fewer atoms in the unionized systems.

It is interesting to compare this result to another regime handled by the theory, i.e., that for larger λ. Although in general for a beam we do not expect many ionized electrons to recombine, this case may describe a gas cell where collisions can enhance the recombination rate. This limit is illustrated in Fig. 4(c) where the parameters are the same as in Fig. 4(b) except that now $\lambda T_2 = \gamma_a$ and $\gamma_I T_2$ increases from 0 to 0.5. The significant change is the nearly immediate filling in of the coherent dip. By the time γ_I reaches $\gamma/2$ there is no longer any evidence of the dip. The dip is dependent on a coherent interaction. Increasing λ increases the population of level b but does it in an incoherent fashion (i.e., not by stimulated transitions). Thus the overall absorption increases and the coherent dip disappears. Thus we see that these new decay paths can make it difficult to use coherent-dip spectroscopy as a means of measuring T_1.

It is also important to notice that due to the dependence of T_1 on γ_l, by decreasing γ_l we are effectively increasing T_1. The increase of T_1 with decreasing γ_l is more apparent when we go to the high-intensity limit. Here we expect the Mollow-type[14] spectrum with gain inside the Rabi sidebands. As shown in Fig. 5(a) as γ_l is decreased, the Rabi sidebands move in due to the decrease in the Rabi frequency [$I_2/(T_1T_2)^{1/2}$ for the two-photon case]. In addition the spectrum develops a slight asymmetry. In order for this asymmetry to become significant γ_l must be substantially smaller than γ_a as shown in the curve of Fig. 5(a) where $\gamma_l = \gamma_a/10$. The effect of ionization is shown in the high-intensity limit in Fig. 5(b) where we have let $\gamma_I T_1$ vary from 0 to 1. Since Eq. (33) shows no dependence of T_1 on the ionization decay rate we do not expect the location of the Rabi sidebands to change as we vary γ_I. The increase of ionization causes asymmetries to develop and rapidly causes the absorption and gain to decrease. This is the expected result since we are decreasing the number of atoms in levels a, b, and l by allowing ionization and only a very small recombination ($\lambda = \gamma_a/100$). The changes in the spectra caused by these two new decay terms are not as drastic as those caused by introducing a simple ω_s-type Stark shift.[4] However, due to the dependence of the Stark shift on T_1, by decreasing γ_l for a given difference in k_{aa} and k_{bb} we can decrease the effective Stark shift. This is illustrated in Fig. 6.

Finally, we examine the effect of the new decay paths on the coupling coefficient $\mathcal{X}_1$. In Ref. 4 it was shown that Stark shifts cause substantial asymmetries but maintain narrow resonances at the Rabi sidebands. The effect of the third level is similar as shown in Figs. 7(a) and 7(b) which give the real and imaginary parts of $\mathcal{X}_1$ for increasing lifetime in level l with no ionization. The effects of ionization are quite similar, although once again this mechanism tends to decrease $\mathcal{X}_1$ due to the loss of atoms. Increasing γ_I when λ is large causes the resonances at the Rabi sidebands to broaden substantially. The result of these effects on phase conjugation reflection coefficients will be examined in a forthcoming paper.

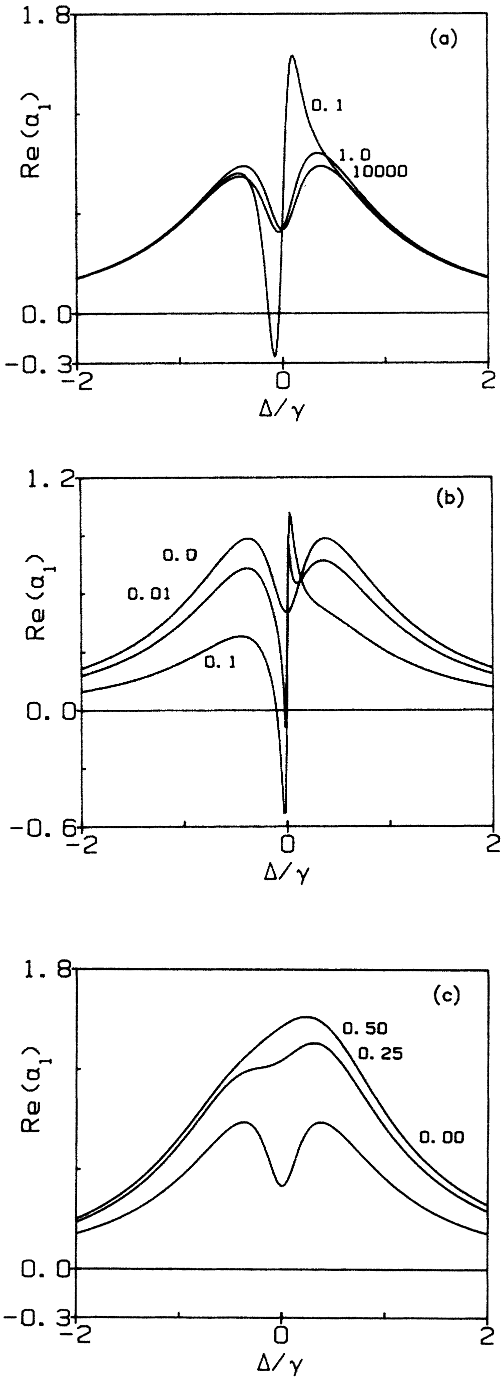

FIG. 4. $\mathrm{Re}(\alpha_1)$ vs Δ/γ for $I_2=1$ and $\gamma_a=\gamma/10$ and $\lambda=\gamma/100$. In (a) we increase the lifetime of level l ($\gamma_l/\gamma=10\,000$, 1, and 0.1) for $\gamma_I/\gamma=0$ and in (b) we increase the ionization ($\gamma_I/\gamma=0$, 0.01, and 0.1) for $\gamma_l/\gamma=10\,000$. In (c) all parameters are the same as in (b) except $\lambda=\gamma$ and γ_l/γ increases from 0 to 0.5.

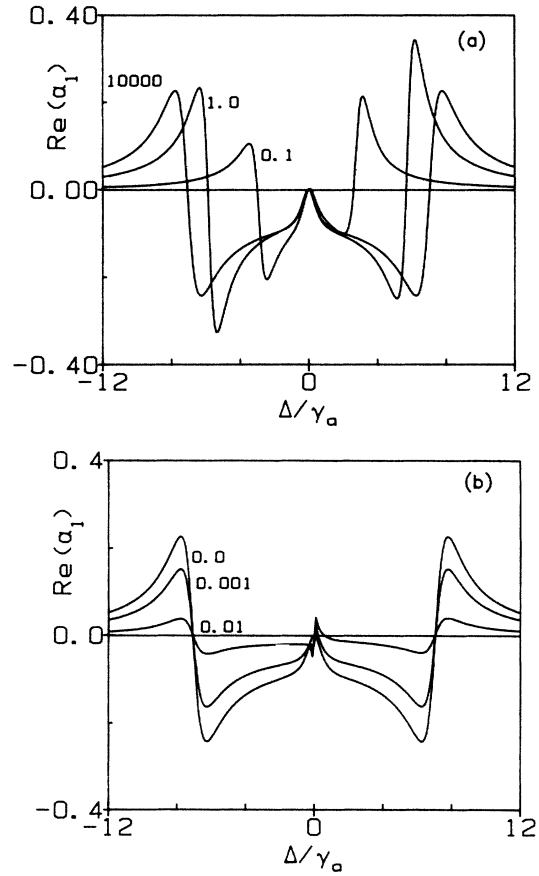

FIG. 5. $\mathrm{Re}(\alpha_1)$ vs Δ/γ_a for $I_2=10$, $\gamma_a=2\gamma$, and $\lambda=\gamma_a/100$. In (a) we decrease γ_l/γ_a from 10 000 to 0.1 for $\gamma_I/\gamma_a=0$ and in (b) we increase γ_I/γ_a from 0 to 0.01 while $\gamma_l/\gamma_a=10\,000$.

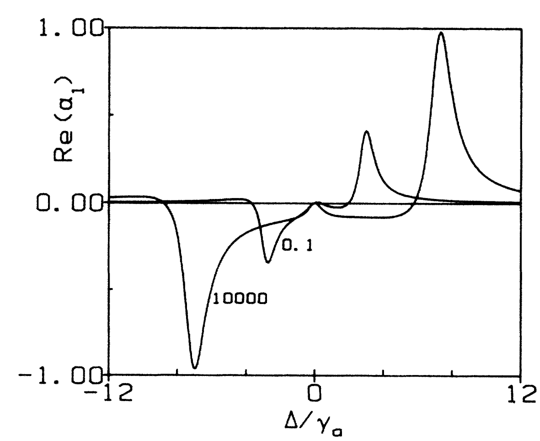

FIG. 6. $\mathrm{Re}(\alpha_1)$ vs Δ/γ_a showing the effects of decreasing γ_l when $k_{aa}-k_{bb}$ is nonzero. $k_{aa}-k_{bb}=0.283\,|k_{ab}|$, $\gamma_l/\gamma_a=10\,000$ and 0.1 and all other parameters are the same as in Fig. 5(a).

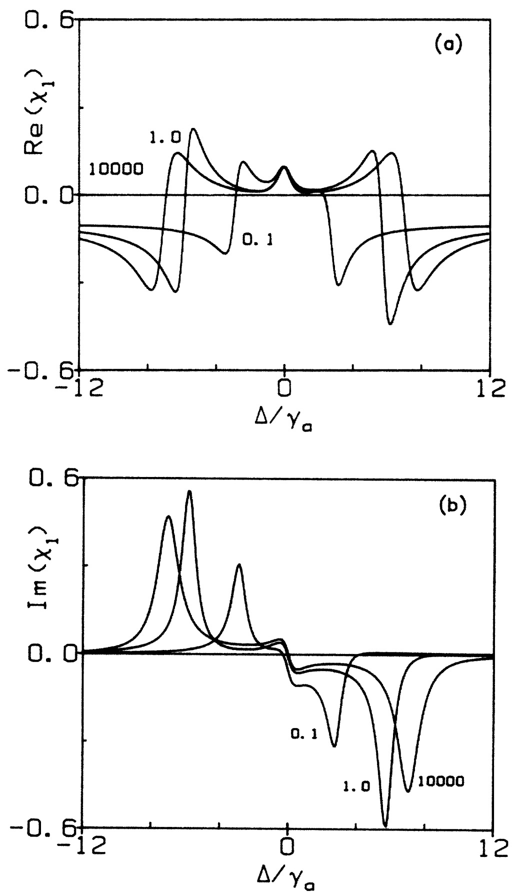

FIG. 7. The real (a) and imaginary (b) parts of χ_1 vs Δ/γ_a for $I_2=10$ for decreasing γ_I. All other parameters are the same as in Fig. 5(a).

VIII. CONCLUSIONS

We have examined the effects of two new decay paths on two-photon two-level interactions. The inclusion of a perturbation-type ionization decay and a decay to a third intermediate level cause some significant changes in the probe absorption and coupling coefficients. Population in the third level, in addition to changing T_1 and $\mathscr{F}(\Delta)$ causes phase lags between the population Fourier components giving rise to a new more generalized Stark-shift term which is Δ dependent and therefore not zero. This causes asymmetries in the spectra. The ionization decay adds new terms to both the coherent and incoherent parts of the spectrum and causes the amplitude of the spectral features to decrease. Another important ionization regime treated by the theory is that for which recombination is negligible but for which the ionization rate $\gamma_I I$ is substantially slower than the other rates. Provided one observes over times short compared to significant ionization, the problem then reduces to the case with no ionization, but with an overall linear absorption coefficient that diminishes slowly in time as the medium ionizes. Hence the present theory treats the effects of ionization on two-photon two-level media interactions except in transient regimes, notably when the medium is ionizing fast compared to the other relaxation processes.

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[1]M. Takatsuji, Phys. Rev. A **4**, 808 (1971).

[2]B. R. Mollow, Phys. Rev. A **4**, 1666 (1971).

[3]D. Grishkowsky, M. M. T. Loy, and P. F. Liao, Phys. Rev. A **12**, 2514 (1975).

[4]M. Sargent III, S. Ovadia, and M. S. Lu, Phys. Rev. A **32**, 1596 (1985).

[5]D. A. Holm and M. Sargent III, Phys. Rev. A **33**, 1073 (1986).

[6]L. Allen, R. W. Boyd, J. Krasinski, M. S. Malcuit, and C. R. Stroud, Jr., Phys. Rev. Lett. **54**, 309 (1985).

[7]M. Sargent III, Phys. Rep. **43C**, 223 (1978).

[8]G. L. Peterson and C. D. Cantrell, Phys. Rev. A **31**, 807 (1985).

[9]W. K. Bischel, L. J. Jusinski, M. N. Spencer, and D. J. Eckstrom, J. Opt. Soc. B **2**, 877 (1985).

[10]Z. Deng and J. H. Eberly, J. Opt. Soc. B **1**, 102 (1984).

[11]D. E. Osterbrock, *Astrophysics of Gaseous Nebulae* (Freeman, San Francisco, 1974), especially Ch. 2.

[12]S. Ovadia and M. Sargent III, Opt. Commun. **49**, 447 (1984).

[13]B. A. Capron and M. Sargent III, preceding paper, Phys. Rev. A **34**, 3034 (1986).

[14]B. R. Mollow, Phys. Rev. A **5**, 2217 (1972).