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Four-wave mixing in a Λ system

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The four-wave mixing (4WM) signal is calculated for a three-level quantum system in a Λ configuration (upper level 2, lower levels 1 and 3) that is driven by three fields. Pump fields E and E' drive the 1-2 and 3-2 transitions, respectively. The probe field is detuned from field E by an amount Δ. If the detuning of field E from the 1-2 transition frequency is equal to that of field E' from the 3-2 transition frequency (dark state pumping), there is a dark state in the absence of the probe field. Rather surprisingly, even though the probe field destroys the dark state for $\Delta \neq 0$, the 4WM mixing vanishes for *all* Δ in the limit of dark-state pumping. The 4WM spectra are explained within the context of a semiclassical, dressed-state picture that involves the sequential emission of a probe and signal photon.

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I. INTRODUCTION

Pump-probe spectroscopy has been a mainstay of laser spectroscopy. In the cw domain one can drive an atomic transition with a strong pump field and probe the same or a coupled transition with a weaker probe field to obtain values for transition frequencies and relaxation rates that characterize the transitions. When the *same* transition is probed, interesting features arise resulting from interference effects between the pump and probe fields [1]. An extension of pump-probe techniques to a Λ system was carried out by Harshawardhan and Agarwal [2] and by Berman and Dubetsky [3]. In these schemes, *two* pump fields drive Raman transitions between the two lower states of a three-level Λ scheme. In the Berman-Dubetsky scheme, each of the fields is assumed to be far off resonance from the optical transitions, enabling one to use an effective two-level approximation. A probe field is applied having frequency close to that of one of the pump fields, and the probe absorption spectrum is monitored. Harshawardhan and Agarwal allowed for four fields (an additional probe field on the 2-3 transition) and obtained an expression for the off-diagonal density matrix element associated with probe field absorption or gain on one of the optical transitions. The resulting spectrum in both cases differs qualitatively from that of the pump-probe spectrum of a two-level atom [1], owing to the juxtaposition of both one-photon and two-photon transitions involving the probe field. Λ systems have received a great deal of attention owing to their applications for quantum information storage and slow light generation [4].

The atom-field system of interest is represented schematically in Fig. 1. A pump field E drives transitions between levels 1 and 2, while a second pump field E' drives transitions between levels 3 and 2. A probe field E_p is also applied, whose frequency is close to that of pump field E, also drives the 1-2 transition. This pump-probe geometry can be viewed as a type of double-Λ system, with fields E and E' providing one of the Λ's and fields E_p and E' providing the other. Double-Λ schemes [5] have been studied extensively since they can lead to index enhancement [6], slow light [7], information storage [8], and improved spin squeezing [9]. In this paper, we calculate the four-wave mixing (4WM) signal having frequency $2\omega-\omega_p$ as a function of the frequency difference Δ between the probe and pump field E frequencies. As will be seen, this signal reflects interesting and somewhat surprising dynamics of this Λ system. Motivation for this work arose in conjunction with experiments being carried out by Steel and co-workers at the University of Michigan [10].

If the detuning of the pump field E from the 1-2 transition frequency is equal to that of pump field E' from the 3-2 transition frequency, then the system evolves into a dark state in the absence of the probe field. For $\Delta=0$, the probe field does not modify this dark state and there is no probe absorption; however, for $\Delta \neq 0$, the dark state *is* modified and there can be an absorption doublet, but no gain, for the probe. If the conditions for a dark state (in the absence of the probe field) are not met, the probe absorption spectrum consists of three absorption and three gain components. These results can be understood easily within the context of a dressed-atom approach. In contrast, the 4WM signal vanishes for *all* probe frequencies when the dark-state conditions are met by the pump fields, *even though the probe field destroys the dark state*. This somewhat nonintuitive result can be understood in terms of a dressed-state interpretation of the signal that involves the sequential emission of a probe and signal

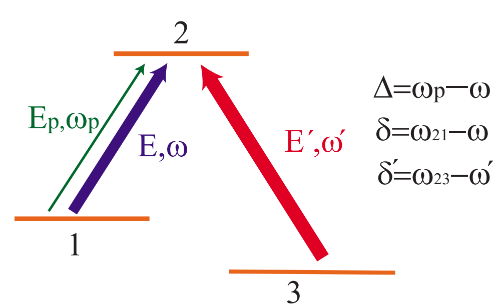

FIG. 1. (Color online) Pump field E and probe field E_p drive transitions between levels 1 and 2, while pump field E' drives transitions between levels 3 and 2. Levels 1 and 3 have infinite lifetimes, while level 2 decays to level 1 with rate $\gamma_{2,1}$ and to level 3 with rate $\gamma_{2,3}$.

photon. If the conditions for a dark state (in the absence of the probe field) are not met, we show that the 4WM signal consists of six components.

The paper is organized as follows: in Sec. II, the exact density matrix solution is obtained and a solution using semiclassical dressed states [11] is given in Sec. III. A discussion of the line shapes is given in Sec. IV in both the weak and strong pump field limits.

II. DENSITY MATRIX SOLUTION

Three fields are incident on an ensemble of stationary, three-level atoms, having the level scheme shown in Fig. 1. The electric field vectors of the two pump fields are given by

$$\mathbf{E}(\mathbf{R},t) = E\boldsymbol{\epsilon}\cos(\mathbf{k}\cdot\mathbf{R}-\omega t), \tag{1}$$

$$\mathbf{E}'(\mathbf{R},t) = E'\boldsymbol{\epsilon}'\cos(\mathbf{k}'\cdot\mathbf{R}-\omega' t), \tag{2}$$

while that of the probe field is given by

$$\mathbf{E}_p(\mathbf{R},t) = E_p\boldsymbol{\epsilon}_p\cos(\mathbf{k}_p\cdot\mathbf{R}-\omega_p t), \tag{3}$$

where (E,E',E_p) are the amplitudes, $(\mathbf{k},\mathbf{k}',\mathbf{k}_p)$ are the propagation vectors, $(\boldsymbol{\epsilon},\boldsymbol{\epsilon}',\boldsymbol{\epsilon}_p)$ are the polarizations, and $(\omega,\omega',\omega_p)$ are the frequencies of the applied fields. Owing to the difference in level spacings or polarization selection rules, one can neglect any effects arising from fields E and E_p driving the 3-2 transition and field E' driving the 1-2 transition.

The Hamiltonian for the atom-field system is

$$H = H_0 - \boldsymbol{\mu}\cdot[\mathbf{E}(\mathbf{R},t)+\mathbf{E}'(\mathbf{R},t)+\mathbf{E}_p(\mathbf{R},t)], \tag{4}$$

where H_0 is the free-atom Hamiltonian and $\boldsymbol{\mu}$ is the atomic dipole moment operator. The density matrix in the Schrödinger picture, $\rho^S(t)$, characterizing this atom-field system evolves as

$$i\hbar\dot{\rho}^S = [H,\rho^S] + (\text{relaxation terms}), \tag{5}$$

where the relaxation terms result from spontaneous emission.

It is convenient to introduce a field interaction representation in which

$$\rho_{12}^S = \rho_{12}e^{-i(\mathbf{k}\cdot\mathbf{R}-\omega t)},$$

$$\rho_{32}^S = \rho_{32}e^{-i(\mathbf{k}'\cdot\mathbf{R}-\omega' t)},$$

$$\rho_{13}^S = \rho_{13}e^{-i[(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}-(\omega-\omega')t]},$$

$$\rho_{ij}^S = \rho_{ji}^{S*}. \tag{6}$$

In this representation and in the rotating-wave approximation, one can write the time evolution equations for the density matrix elements in matrix form as

$$\dot{\boldsymbol{\rho}} = -\mathbf{A}\boldsymbol{\rho} + \mathbf{B}_+\boldsymbol{\rho}e^{-i(\mathbf{k}_p-\mathbf{k})\cdot\mathbf{R}}e^{i\Delta t} + \mathbf{B}_-\boldsymbol{\rho}e^{i(\mathbf{k}_p-\mathbf{k})\cdot\mathbf{R}}e^{-i\Delta t} + \boldsymbol{\lambda}, \tag{7}$$

with

$$\boldsymbol{\rho} = \begin{pmatrix} \rho_{11} \\ \rho_{13} \\ \rho_{31} \\ \rho_{12} \\ \rho_{21} \\ \rho_{32} \\ \rho_{23} \\ \rho_{22} \end{pmatrix}, \quad \boldsymbol{\lambda} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ i\chi' \\ -i\chi' \\ 0 \end{pmatrix}, \tag{8}$$

$$\mathbf{A} = \begin{pmatrix} 0 & 0 & 0 & -i\chi & i\chi & 0 & 0 & -\gamma_{2,1} \\ 0 & -i(\delta-\delta') & 0 & -i\chi' & 0 & 0 & i\chi & 0 \\ 0 & 0 & i(\delta-\delta') & 0 & i\chi' & -i\chi & 0 & 0 \\ -i\chi & -i\chi' & 0 & \gamma-i\delta & 0 & 0 & 0 & i\chi \\ i\chi & 0 & i\chi' & 0 & \gamma+i\delta & 0 & 0 & -i\chi \\ i\chi' & 0 & -i\chi & 0 & 0 & \gamma-i\delta' & 0 & 2i\chi' \\ -i\chi' & i\chi & 0 & 0 & 0 & 0 & \gamma+i\delta' & -2i\chi' \\ 0 & 0 & 0 & i\chi & -i\chi & i\chi' & -i\chi' & \gamma_2 \end{pmatrix}, \tag{9}$$

$$\mathbf{B}_+ = i\chi_p \begin{pmatrix} 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 & 0 & 0 & -1 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \end{pmatrix}, \qquad (10)$$

$$\mathbf{B}_- = i\chi_p \begin{pmatrix} 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ -1 & 0 & 0 & 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \end{pmatrix}, \qquad (11)$$

where

$$\chi = -\boldsymbol{\mu}_{21} \cdot \boldsymbol{\epsilon} E/2\hbar, \quad \chi' = -\boldsymbol{\mu}_{31} \cdot \boldsymbol{\epsilon}' E'/2\hbar,$$

$$\chi_p = -\boldsymbol{\mu}_{21} \cdot \boldsymbol{\epsilon}_p E_p/2\hbar \qquad (12)$$

are (one-half of the) Rabi frequencies (all assumed real) associated with the various transitions,

$$\delta = \omega_{21} - \omega, \quad \delta' = \omega_{23} - \omega' \qquad (13)$$

are atom-field detunings,

$$\Delta = \omega_p - \omega \qquad (14)$$

is the detuning of the probe field from the first pump field, γ_2 is the excited-state decay rate resulting from spontaneous emission, $\gamma = \gamma_2/2$ is the decay rate of the optical coherences ρ_{12} and ρ_{32}, and $\gamma_{2,1}/\gamma_2$ is the branching ratio for emission into state 1. The fact that $\rho_{11}+\rho_{22}+\rho_{33}=1$ has been used to eliminate ρ_{33} from the equations.

We wish to obtain a solution of Eq. (7) to first order in χ_p. After all transients have died away, such a solution to these equations can be written in the form

$$\boldsymbol{\rho} = \boldsymbol{\rho}^{(0)} + \boldsymbol{\rho}^{(+)} e^{-i(\mathbf{k}_p - \mathbf{k})\cdot \mathbf{R}} e^{i\Delta t} + \boldsymbol{\rho}^{(-)} e^{i(\mathbf{k}_p - \mathbf{k})\cdot \mathbf{R}} e^{-i\Delta t}. \qquad (15)$$

Substituting Eq. (15) into Eq. (7) and equating coefficients of e^0, $e^{\mp i(\mathbf{k}_p-\mathbf{k})\cdot\mathbf{R}} e^{\pm i\Delta t}$, we obtain

$$\boldsymbol{\rho}^{(0)} = \mathbf{A}^{-1}\boldsymbol{\lambda}, \qquad (16)$$

which is the solution in the absence of the probe field, and

$$\boldsymbol{\rho}^{(+)} = (\mathbf{A} + i\Delta \mathbf{1})^{-1}\mathbf{B}_+\boldsymbol{\rho}^{(0)}, \qquad (17)$$

$$\boldsymbol{\rho}^{(-)} = (\mathbf{A} - i\Delta \mathbf{1})^{-1}\mathbf{B}_-\boldsymbol{\rho}^{(0)}, \qquad (18)$$

where $\mathbf{1}$ is an 8×8 unit matrix, which are contributions to $\boldsymbol{\rho}$ of order χ_p.

It is simple enough to obtain expressions for $\boldsymbol{\rho}^{(0)}$, $\boldsymbol{\rho}^{(+)}$, and $\boldsymbol{\rho}^{(-)}$ using programs such as MATHEMATICA, but they are unwieldy and are not reproduced here. We are interested in the 4WM signal propagating in the direction $\mathbf{k}_s = 2\mathbf{k} - \mathbf{k}_p$ with frequency $\omega_s = 2\omega - \omega_p$ (it is assumed that $\mathbf{k}$ and $\mathbf{k}_p$ are nearly parallel such that phase matching can be approximately satisfied). Using the Maxwell-Bloch equations, one can show that the 4WM signal is proportional to

$$S_f = |\gamma \rho_{12}^{(-)}/\chi_p|^2. \qquad (19)$$

(The probe absorption coefficient, not calculated in this work, is proportional to $\alpha_p = -\mathrm{Im}[\gamma \rho_{12}^{(+)}/\chi_p]$.)

In general, the 4WM signal consists of six components, consisting of three equal-intensity doublets centered about $\Delta = 0$, as shown in Fig. 2. If $\delta = \delta'$, which is the condition for a dark state to exist in the absence of the probe field, then the 4WM signal *vanishes identically* for all probe detunings. That is, the 4WM signal vanishes for $\Delta \neq 0$, *even though the dark state no longer exists*. In contrast, the corresponding equation for probe absorption is nonvanishing for $\Delta \neq 0$ under conditions of dark-state pumping. The physical origin of the 4WM signals is discussed in Sec. IV. Note that, for $\delta \neq \delta'$, there is also a small, "nonsecular" contribution to the line shapes centered at $\Delta = 0$.

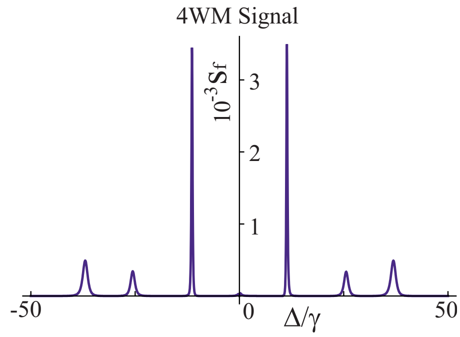

FIG. 2. (Color online) Four-wave mixing signal as a function of Δ/γ for $\chi/\gamma = \chi'/\gamma = 10$, $\delta/\gamma = -25$, $\delta'/\gamma = -15$, and $\gamma_{2,1}/\gamma_2 = 0.5$. For these parameters the corresponding dressed-state frequencies and populations are $\omega_A/\gamma = 29.4$, $\omega_B/\gamma = 18.1$, $\omega_C/\gamma = -7.5$ and $\rho_{AA} = 0.300$, $\rho_{BB} = 0.684$, $\rho_{CC} = 0.016$.

III. DRESSED-STATE SOLUTION

The dressed-state approach is formulated in terms of semiclassical dressed states [11]. To introduce this dressed-state representation, we arbitrarily set the energy of state 2 equal to zero and write the state vector as

$$|\psi(t)\rangle = \tilde{c}_1 e^{-i(\mathbf{k}\cdot\mathbf{R} - \omega t)}|1\rangle + |2\rangle + \tilde{c}_3 e^{-i(\mathbf{k}'\cdot\mathbf{R} - \omega' t)}|3\rangle \qquad (20)$$

such that, in the absence of relaxation, the probability amplitudes $\tilde{\mathbf{c}} = (\tilde{c}_1, \tilde{c}_2, \tilde{c}_3)$, written as a column vector, evolve according to

$$i\hbar \frac{d\tilde{\mathbf{c}}}{dt} = (\mathbf{H}_1 + \mathbf{V})\tilde{\mathbf{c}}, \qquad (21)$$

where

$$\mathbf{H}_1 = \hbar \begin{pmatrix} -\delta & \chi & 0 \\ \chi & 0 & \chi' \\ 0 & \chi' & -\delta' \end{pmatrix}, \tag{22a}$$

$$\mathbf{V} = \hbar \chi_p \begin{pmatrix} 0 & a(\mathbf{R},t) & 0 \\ b(\mathbf{R},t) & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{22b}$$

and

$$a(\mathbf{R},t) = e^{-i(\mathbf{k}_p-\mathbf{k})\cdot\mathbf{R}} e^{i\Delta t} = b^*(\mathbf{R},t). \tag{23}$$

The semiclassical dressed states are obtained by diagonalizing $\mathbf{H}_1$. In other words, we set

$$\mathbf{c}_D = \begin{pmatrix} c_A \\ c_B \\ c_C \end{pmatrix} = \mathbf{T}\tilde{\mathbf{c}}, \tag{24}$$

where

$$\mathbf{T} = \begin{pmatrix} T_{A1} & T_{A2} & T_{A3} \\ T_{B1} & T_{B2} & T_{B3} \\ T_{C1} & T_{C2} & T_{C3} \end{pmatrix} \tag{25}$$

is an orthogonal matrix defined such that

$$\mathbf{T}\mathbf{H}_1\mathbf{T}^\dagger = \mathbf{\Lambda} = \hbar \begin{pmatrix} \omega_A & 0 & 0 \\ 0 & \omega_B & 0 \\ 0 & 0 & \omega_C \end{pmatrix} \tag{26}$$

and $\hbar\omega_\alpha$ ($\alpha=A,B,C$) are the energies of the semiclassical dressed states obtained by diagonalizing $\mathbf{H}_1$ (the eigenvalues are chosen such that $\omega_A > \omega_B > \omega_C$).

It follows from Eqs. (21)–(25) that, in the absence of relaxation, the dressed-state amplitudes evolve according to

$$i\hbar\dot{\mathbf{c}}_D = \mathbf{\Lambda}\mathbf{c}_D + \mathbf{V}_D\mathbf{c}_D, \tag{27}$$

where $\mathbf{V}_D=\mathbf{T}\mathbf{V}\mathbf{T}^\dagger$ is given by

$$\mathbf{V}_D = \hbar\chi_p a(\mathbf{R},t) \begin{pmatrix} T_{A1}T_{A2} & T_{A1}T_{B2} & T_{A1}T_{C2} \\ T_{B1}T_{A2} & T_{B1}T_{B2} & T_{B1}T_{C2} \\ T_{C1}T_{A2} & T_{C1}T_{B2} & T_{C1}T_{C2} \end{pmatrix} + \hbar\chi_p b(\mathbf{R},t) \begin{pmatrix} T_{A1}T_{A2} & T_{B1}T_{A2} & T_{C1}T_{A2} \\ T_{A1}T_{B2} & T_{B1}T_{B2} & T_{C1}T_{B2} \\ T_{A1}T_{C2} & T_{B1}T_{C2} & T_{C1}T_{C2} \end{pmatrix}. \tag{28}$$

It is possible to get analytic expressions for the ω_α and the $T_{\alpha j}$, but they are very complicated unless $\delta=\delta'$.

To obtain the 4WM intensity using the dressed-state representation, one must (a) express S_f in terms of the dressed-state density matrix elements $\rho_{\alpha\beta}$, (b) obtain the evolution equations for the dressed-state density matrix elements including decay, and (c) solve these equations in "steady state" to first order in χ_p. This method is particularly useful only in the so-called secular approximation, where all the frequency spacings $|\omega_{\alpha\beta}|=|\omega_\alpha-\omega_\beta|$ are much greater than the decay rate γ_2 [12]. For dark-state pumping ($\delta=\delta'$), it is not necessary to make the secular approximation [2].

In analogy with Eq. (15), we write

$$\boldsymbol{\rho}_D = \boldsymbol{\rho}_D^{(0)} + a(\mathbf{R},t)\boldsymbol{\rho}_D^{(+)} + b(\mathbf{R},t)\boldsymbol{\rho}_D^{(-)}. \tag{29}$$

Using the adjoint of Eq. (24), Eq. (23), and the fact that $\rho_{\alpha\beta}=c_\alpha c_\beta^*$, one finds that

$$\begin{aligned}\rho_{12}^{(\pm)} &= T_{A2}(T_{B1}\rho_{BA}^{(\pm)} + T_{C1}\rho_{CA}^{(\pm)}) + T_{B2}(T_{A1}\rho_{AB}^{(\pm)} + T_{C1}\rho_{CB}^{(\pm)}) \\ &\quad + T_{C2}(T_{A1}\rho_{AC}^{(\pm)} + T_{B1}\rho_{BC}^{(\pm)}) + T_{A1}T_{A2}\rho_{AA}^{(\pm)} \\ &\quad + T_{B1}T_{B2}\rho_{BB}^{(\pm)} + T_{C1}T_{C2}\rho_{CC}^{(\pm)},\end{aligned} \tag{30}$$

giving the density matrix elements needed to evaluate S_f [see Eq. (19)]. The dressed-state density matrix in the absence of the probe field is given by

$$\boldsymbol{\rho}_D^{(0)} = \mathbf{T}\boldsymbol{\rho}^{(0)}\mathbf{T}^\dagger, \tag{31}$$

where elements of $\rho^{(0)}$ are given by Eq. (16).

The next step is to obtain the equations for $\rho_{\alpha\beta}^{(\pm)}$. The evolution equation for the dressed state density matrix is

$$i\hbar\dot{\boldsymbol{\rho}}_D = [(\mathbf{\Lambda} + \mathbf{V}_D), \boldsymbol{\rho}_D] + \mathbf{\Gamma}\boldsymbol{\rho}_D, \tag{32}$$

where the relaxation terms $\mathbf{\Gamma}\rho$ are expressed conveniently as

$$\begin{aligned}\mathbf{\Gamma}\boldsymbol{\rho}_D &= -\gamma[(\boldsymbol{\sigma}_0)_D\boldsymbol{\rho}_D + \boldsymbol{\rho}_D(\boldsymbol{\sigma}_0)_D] \\ &\quad + \gamma_{2,1}(\boldsymbol{\sigma}_-^{12})_D\boldsymbol{\rho}_D(\boldsymbol{\sigma}_+^{12})_D + \gamma_{2,3}(\boldsymbol{\sigma}_-^{32})_D\boldsymbol{\rho}_D(\boldsymbol{\sigma}_+^{32})_D,\end{aligned} \tag{33}$$

with

$$(\boldsymbol{\sigma})_D = \mathbf{T}\boldsymbol{\sigma}\mathbf{T}^\dagger \tag{34}$$

and

$$\boldsymbol{\sigma}_0 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{35}$$

$$\boldsymbol{\sigma}_-^{12} = \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \boldsymbol{\sigma}_+^{12} = \begin{pmatrix} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{36}$$

$$\boldsymbol{\sigma}_-^{32} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}, \quad \boldsymbol{\sigma}_+^{32} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 0 & 0 \end{pmatrix}. \tag{37}$$

Equation (29) substituted into Eqs. (32) and (23) is used, and the coefficients of e^0 and $e^{\pm i(\mathbf{k}_p-\mathbf{k})\cdot\mathbf{R}\mp i\Delta t}$ are equated. In this manner, one obtains equations for $\boldsymbol{\rho}_D^{(0)}$, $\boldsymbol{\rho}_D^{(+)}$, and $\boldsymbol{\rho}_D^{(-)}$. Admittedly, these equations are horrendously complicated; however, in the secular approximation where terms of order $\gamma/\sqrt{\chi^2+\chi'^2+\delta^2}$ are neglected, one has

$$\rho_{\alpha\beta}^{(0)} \sim 0, \quad \alpha \neq \beta, \tag{38a}$$

$$\rho_{\alpha\alpha}^{(\pm)} \sim 0, \quad \alpha = A,B,C, \tag{38b}$$

and

$$\rho_{AB}^{(+)} = \frac{-i\chi_p T_{A1}T_{B2}(\rho_{BB}^{(0)} - \rho_{AA}^{(0)})}{\gamma_{AB} + i(\Delta + \omega_{AB})},$$

$$\rho_{BA}^{(+)}=\frac{-i\chi_p T_{B1}T_{A2}(\rho_{AA}^{(0)}-\rho_{BB}^{(0)})}{\gamma_{AB}+i(\Delta-\omega_{AB})},$$

$$\rho_{AC}^{(+)}=\frac{-i\chi_p T_{A1}T_{C2}(\rho_{CC}^{(0)}-\rho_{AA}^{(0)})}{\gamma_{AC}+i(\Delta+\omega_{AC})},$$

$$\rho_{CA}^{(+)}=\frac{-i\chi_p T_{C1}T_{A2}(\rho_{AA}^{(0)}-\rho_{CC}^{(0)})}{\gamma_{AC}+i(\Delta-\omega_{AC})},$$

$$\rho_{BC}^{(+)}=\frac{-i\chi_p T_{B1}T_{C2}(\rho_{CC}^{(0)}-\rho_{BB}^{(0)})}{\gamma_{BC}+i(\Delta+\omega_{BC})},$$

$$\rho_{CB}^{(+)}=\frac{-i\chi_p T_{C1}T_{B2}(\rho_{BB}^{(0)}-\rho_{CC}^{(0)})}{\gamma_{BC}+i(\Delta-\omega_{BC})},$$

$$\rho_{\alpha\beta}^{(-)}=-\rho_{\beta\alpha}^{(+)}, \tag{39}$$

where

$$\gamma_{\alpha\beta}=\gamma(T_{\alpha2}^2+T_{\beta2}^2)+\gamma_{2,1}T_{\alpha2}T_{\beta2}T_{\alpha1}T_{\beta1}+\gamma_{2,3}T_{\alpha2}T_{\beta2}T_{\alpha3}T_{\beta3} \tag{40}$$

and $\gamma_{2,3}/\gamma_2$ is the branching ratio for emission into state 3. It then follows that

$$\begin{aligned}S_f=&|(T_{A2}T_{B1}T_{B2}T_{A1})(\rho_{AA}^{(0)}-\rho_{BB}^{(0)})|^2\\&\times\left[\frac{\gamma^2}{(\Delta-\omega_{AB})^2+\gamma_{AB}^2}+\frac{\gamma^2}{(\Delta+\omega_{AB})^2+\gamma_{AB}^2}\right]\\&+|(T_{C2}T_{A1}T_{A2}T_{C1})(\rho_{CC}^{(0)}-\rho_{AA}^{(0)})|^2\\&\times\left[\frac{\gamma^2}{(\Delta-\omega_{AC})^2+\gamma_{AC}^2}+\frac{\gamma^2}{(\Delta+\omega_{AC})^2+\gamma_{AC}^2}\right]\\&+|(T_{B2}T_{C1}T_{C2}T_{B1})(\rho_{BB}^{(0)}-\rho_{CC}^{(0)})|^2\\&\times\left[\frac{\gamma^2}{(\Delta-\omega_{BC})^2+\gamma_{BC}^2}+\frac{\gamma^2}{(\Delta+\omega_{BC})^2+\gamma_{BC}^2}\right].\end{aligned} \tag{41}$$

The quantities $T_{\alpha\beta}$ and $\omega_{\alpha\beta}$ are obtained by diagonalizing the matrix H_1, while $\rho_{\alpha\beta}^{(0)}$ are obtained from Eqs. (16) and (31).

As in the case of pump-probe absorption on a two-level atom [11], the widths $\gamma_{\alpha\beta}$ are equal to one-half the sum of the decay rates of the α and β levels [first term in Eq. (40)] *plus* a contribution (which can be positive or negative) from the repopulation of the ground states resulting from spontaneous emission [second two terms in Eq. (40)].

IV. DISCUSSION

A. Weak fields

If the Rabi frequencies of the pump fields are much less than the relaxation rates, then the probe absorption and 4WM signals can be obtained by calculating $\rho_{12}^{(\pm)}$ to lowest order in the probe Rabi frequency, assuming that the density matrix in the absence of the probe field is equal to $\boldsymbol{\rho}^{(0)}$. However, since we are looking for a new signal generated in the $\mathbf{k}_s=2\mathbf{k}-\mathbf{k}_p$ direction, we must include pump field contributions *in addition* to those provided by $\boldsymbol{\rho}^{(0)}$. There are *three* terms that contribute to $\rho_{12}^{(-)}$, which can be obtained as a perturbative solution of Eqs. (7)–(18). The first results from a chain in which we start from $(\rho_{11}^{(0)}-\rho_{22}^{(0)})$, the probe field acts to produce $\rho_{21}^{(-)}$, pump field E acts to produce $\rho_{11}^{(-)}$ and $\rho_{22}^{(-)}$ (repopulation of state 1 by spontaneous emission also enters), and pump field E acts once again to produce $\rho_{12}^{(-)}$. The second results from a perturbative chain in which we start from $\rho_{13}^{(0)}$, the probe field acts to produce $\rho_{23}^{(-)}$, pump field E acts to produce $\rho_{13}^{(-)}$, and pump field E' acts to produce $\rho_{12}^{(-)}$. The third results from a perturbative chain in which we start from $\rho_{13}^{(0)}$, the probe field acts to produce $\rho_{23}^{(-)}$, pump field E' acts to produce $\rho_{22}^{(-)}$ and $\rho_{11}^{(-)}$ (via spontaneous emission), and pump field E acts to produce $\rho_{12}^{(-)}$. Explicitly, one finds

$$\begin{aligned}\rho_{12}^{(-)}=&\frac{i\chi}{\gamma-i(\Delta+\delta)}\frac{\chi}{\Delta}\left[1-\frac{\gamma_{2,1}+i\Delta}{\gamma_2-i\Delta}\right]\frac{-i\chi_p}{\gamma-i(\Delta-\delta)}(\rho_{11}^{(0)}-\rho_{22}^{(0)})\\&+\frac{i\chi'}{\gamma-i(\Delta+\delta')}\left[\frac{\chi}{(\delta-\delta'+\Delta)}-\left(\frac{\chi}{\Delta}\right)\frac{\gamma_{2,1}+i\Delta}{\gamma_2-i\Delta}\right]\\&\times\frac{-i\chi_p}{\gamma-i(\Delta-\delta')}\rho_{13}^{(0)}.\end{aligned} \tag{42}$$

If $\delta=\delta'$ (dark-state pumping), Eq. (42) reduces to

$$\begin{aligned}\rho_{12}^{(-)}=&\frac{i\chi}{\gamma-i(\Delta+\delta)}\frac{\chi}{\Delta}\left[1-\frac{\gamma_{2,1}+i\Delta}{\gamma_2-i\Delta}\right]\frac{-i\chi_p}{\gamma-i(\Delta-\delta)}\frac{\chi'^2}{\chi^2+\chi'^2}\\&+\frac{i\chi'}{\gamma-i(\Delta+\delta)}\frac{\chi}{\Delta}\left[1-\frac{\gamma_{2,1}+i\Delta}{\gamma_2-i\Delta}\right]\frac{-i\chi_p}{\gamma-i(\Delta-\delta)}\frac{-\chi\chi'}{\chi^2+\chi'^2}\\&=0,\end{aligned} \tag{43}$$

in agreement with the exact solution. Thus the vanishing of the 4WM signal in the weak field limit can be viewed as an cancellation between three terms, one originating from the ground-state population $\rho_{11}^{(0)}$ ($\rho_{22}^{(0)}=0$ for dark-state pumping) and the other two from the ground-state coherence $\rho_{13}^{(0)}$.

B. Strong fields

In the limit that $\sqrt{(\chi^2+\chi'^2)}\gg\gamma$, it is possible to use the dressed-state picture to obtain approximate solutions for the 4WM signals. Even for weak fields, such an approach can be used provided $|\delta|\gg\gamma$ [12]. We have seen already that this signal has six components, in general. In contrast to the quantized dressed-state picture, there is no infinite "ladder" of dressed states in the semiclassical dressed-state picture. Nevertheless, one can use a three-step ladder as a device for obtaining the 4WM signal [each "step" itself consists of a triplet of steps (see below)]. The corresponding ladder "rungs" are separated by the pump field E frequency ω. There is a corresponding ladder of states separated by the pump field E' frequency ω'; however, this ladder of states is not needed when one considers 4WM associated with the 1-2 transition.

The "three-step" ladder is shown in Fig. 3. The 4WM signal can be viewed as arising from a *sequential* emission process in which the first step involves a coupling from the state $|2\rangle$ component of a given dressed state in the upper

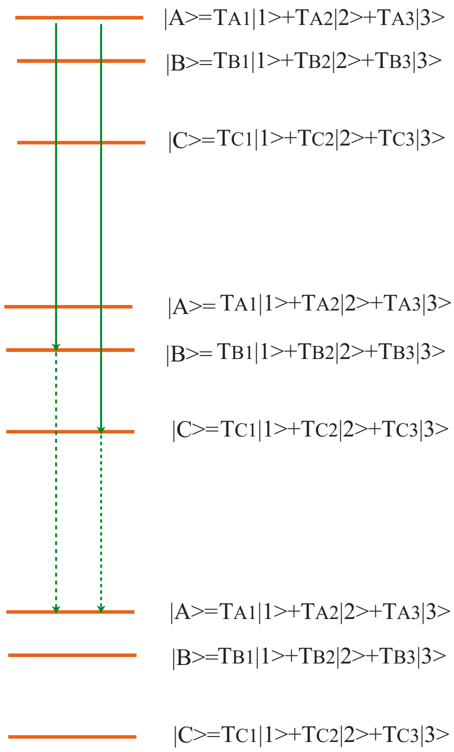

FIG. 3. (Color online) Semiclassical dressed states: four-wave mixing occurs by the sequential emission of a probe and signal photon in this dressed-state picture. The probe field connects a component $T_{\alpha 2}$ in the upper manifold of levels to a component $T_{\beta 1}$ while the signal photon connects the $T_{\beta 2}$ component of the *same* level in the middle manifold of levels to the $T_{\alpha 1}$ component in the lower manifold of levels. The final level of this sequential emission must be the *same* as the initial level from which we started to satisfy the energy conservation condition $2\hbar\omega=\hbar(\omega_p+\omega_s)$. Two of the six possible chains are shown in the figure.

manifold of levels to the state $|1\rangle$ component of a given level in the middle manifold, followed by an emission of the signal photon from the state $|2\rangle$ component of the *same* level in the middle manifold of levels to the state $|1\rangle$ component in the lower manifold of levels. The final level of this sequential emission must be the *same* as the initial level from which we started. In other words, transitions of the type $A\to B\to A$, or $A\to C\to A$ constitute two of the six possible chains. The restriction to begin and end on the same dressed state guarantees that the sum of the probe field plus the signal field frequencies is centered at twice the frequency of pump field E, as is required in the 4WM process. The emission of the probe photon must be weighted with the population difference of the upper-level and middle-level populations. In this manner, one obtains contributions to the 4WM signal:

$$(S_f)_{ABA}=\frac{\gamma^2|(T_{A2}T_{B1}T_{B2}T_{A1})(\rho_{AA}^{(0)}-\rho_{BB}^{(0)})|^2}{(\Delta-\omega_{AB})^2+\gamma_{AB}^2}, \quad (44a)$$

$$(S_f)_{ACA}=\frac{\gamma^2|(T_{A2}T_{C1}T_{C2}T_{A1})(\rho_{AA}^{(0)}-\rho_{CC}^{(0)})|^2}{(\Delta-\omega_{AC})^2+\gamma_{AC}^2}, \quad (44b)$$

$$(S_f)_{BAB}=\frac{\gamma^2|(T_{B2}T_{A1}T_{A2}T_{B1})(\rho_{BB}^{(0)}-\rho_{AA}^{(0)})|^2}{(\Delta+\omega_{AB})^2+\gamma_{AB}^2}, \quad (44c)$$

$$(S_f)_{BCB}=\frac{\gamma^2|(T_{B2}T_{C1}T_{C2}T_{B1})(\rho_{BB}^{(0)}-\rho_{CC}^{(0)})|^2}{(\Delta-\omega_{BC})^2+\gamma_{BC}^2}, \quad (44d)$$

$$(S_f)_{CAC}=\frac{\gamma^2|(T_{C2}T_{A1}T_{A2}T_{C1})(\rho_{CC}^{(0)}-\rho_{AA}^{(0)})|^2}{(\Delta+\omega_{AC})^2+\gamma_{AC}^2}, \quad (44e)$$

$$(S_f)_{CBC}=\frac{\gamma^2|(T_{C2}T_{B1}T_{B2}T_{C1})(\rho_{CC}^{(0)}-\rho_{BB}^{(0)})|^2}{(\Delta+\omega_{BC})^2+\gamma_{BC}^2}, \quad (44f)$$

One sees immediately that $(S_f)_{\alpha\beta\alpha}=(S_f)_{\beta\alpha\beta}$ such that the signal consists of three pairs of signals; the amplitudes of each component of the pair are equal and are symmetrically centered about $\Delta=0$. If $\delta=\delta'$ (dark-state pumping), then $T_{B2}=0$, $\rho_{CC}^{(0)}=\rho_{AA}^{(0)}=0$, $\rho_{BB}^{(0)}=1$, and $(S_f)_{\alpha\beta\alpha}=0$—consistent with the exact result given above—there is no 4WM for dark-state pumping.

We could equally well have examined the 4WM signal on the 2-3 transition. In that case one finds a 4WM signal in the $\mathbf{k}'+\mathbf{k}_p-\mathbf{k}$ direction that is the analog of the probe absorption; that is, it vanishes under dark-state pumping for $\Delta=0$, but not for $\Delta\neq 0$. There is also a 4WM signal in the $\mathbf{k}'+\mathbf{k}-\mathbf{k}_p$ that is the analog of the 4WM signal on the 1-2 transition; that is, it vanishes for all Δ under dark-state pumping.

In summary, we have presented what we believe to be an interesting pump-probe scheme involving three incident fields interacting with atoms whose level structure can be approximated as a three-level Λ configuration. Interference phenomena have been described for the 4WM signal. A semiclassical dressed-state picture has been introduced that allows one to obtain a semiquantitative description of the atom-field dynamics.

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