

**Electromagnetically induced absorption due to transfer of coherence and to transfer of population**C. Goren,<sup>1</sup> A. D. Wilson-Gordon,<sup>2</sup> M. Rosenbluh,<sup>1</sup> and H. Friedmann<sup>2</sup><sup>1</sup>*Department of Physics, Bar-Ilan University, Ramat Gan 52900, Israel*<sup>2</sup>*Department of Chemistry, Bar-Ilan University, Ramat Gan 52900, Israel*

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The absorption spectrum of a weak probe, interacting with a driven degenerate two-level atomic system, whose ground and excited hyperfine states are  $F_{g,e}$ , can exhibit narrow peaks at line center. When the pump and probe polarizations are different,  $F_e = F_g + 1$  and  $F_g > 0$ , the electromagnetically induced absorption (EIA) peak has been shown to be due to the transfer of coherence (TOC) between the excited and ground states via spontaneous decay. We give a detailed explanation of why the TOC that leads to EIA (EIA-TOC) can only take place when ground-state population trapping does not occur, that is, when  $F_e = F_g + 1$ . We also explain why EIA-TOC is observed in open systems. We show that EIA can also occur when the pump and probe polarizations are identical and  $F_e = F_g + 1$ . This EIA is analogous to an effect that occurs in simple two-level systems when the collisional transfer of population (TOP) from the ground state to a reservoir is greater than that from the excited state. For a degenerate two-level system, the reservoir consists of the Zeeman sublevels of the ground hyperfine state, and of other nearby hyperfine states that do not interact with the pump. We will also discuss the four-wave mixing spectrum under the conditions where EIA-TOC and EIA-TOP occur.

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**I. INTRODUCTION**

The probe absorption spectrum of a degenerate two-level atomic system, interacting with a strong pump and weak probe, can exhibit narrow features at line center where the pump and probe have equal frequencies [1–5]. When the probe absorption spectrum is characterized by a sharp dip, the phenomenon is called electromagnetically induced transparency (EIT) [4–6]. However, when the absorption spectrum is characterized by a sharp peak, the effect is called electromagnetically induced absorption (EIA) [5]. EIA has been observed when copropagating, orthogonally polarized pump and probe beams interact with an  $F_g \rightarrow F_e = F_g + 1$  hyperfine transition [3–5,7] and also in the Hanle configuration [8–11]. It has been demonstrated that EIA is a consequence of the transfer of coherence (TOC) due to spontaneous emission between the excited and ground degenerate states [12].

In this paper, we explain why the TOC that leads to EIA (here called EIA-TOC) can only occur when the pump and probe lasers have different polarizations, and requires the presence of significant pump-induced population in the excited state. Through detailed arguments we will demonstrate that these criteria account for the experimental observation that EIA only occurs when  $F_g \rightarrow F_e = F_g + 1$  with  $F_g > 0$ . In addition, we will explain the observation of EIA in open transitions [10] where spontaneous decay can occur to other hyperfine levels close in energy to the ground state.

We also show that a mechanism that is completely different from TOC, namely, transfer of population (TOP), can also lead to EIA peaks in the absorption spectrum of a degenerate two-level system. It occurs when the pump and probe that interact with a closed  $F_g \rightarrow F_e = F_g + 1$  with  $F_g \geq 0$  hyperfine transition have the *same* polarization rather than *different* polarizations as in the case of EIA-TOC. The system can thus be considered as a series of two-level systems interlinked by spontaneous emission and collisions. In order to obtain EIA for this system (here called EIA-TOP),

we must include collisional transfer of population from each of the ground Zeeman sublevels to all the other sublevels of the same hyperfine state. We must also include collisional transfer of population from the ground Zeeman sublevels to other nearby hyperfine states that cannot be populated by spontaneous emission from the excited state. For a particular ground sublevel, all the states to which collisional relaxation takes place form a “reservoir,” allowing population to be transferred to and from the ground sublevel. We also include decay to the reservoir due to time of flight that occurs at the same rate for all the ground and excited sublevels. By including collisional effects, we can achieve the situation where the effective decay rate from the ground state to the reservoir is greater than that from the excited state. This leads to EIA-TOP in degenerate two-level systems which is analogous to an effect predicted by us for simple two-level systems [13].

Narrow features can also occur in the four-wave mixing (FWM) spectrum of simple [14] and degenerate two-level atoms [15,16]. We will discuss the FWM spectra that correspond to the two versions of EIA that can occur in degenerate two-level systems.

**II. EQUATIONS OF MOTION**

The system consists of one or more ground hyperfine states  $F_g$  and a single excited hyperfine state  $F_e$ , interacting with a pump of frequency  $\omega_1$  and a probe of frequency  $\omega_2$ . We use the equations for the time evolution of the Zeeman sublevels as formulated by Renzoni *et al.* [17], with the addition of decay from the ground and excited states to a reservoir [18,14], and collisions between the Zeeman sublevels of the ground states [19]:

$$\begin{aligned} \dot{\rho}_{e_i e_j} = & -(i\omega_{e_i e_j} + \Gamma)\rho_{e_i e_j} + (i/\hbar) \sum_{g_k} (\rho_{e_i g_k} V_{g_k e_j} - V_{e_i g_k} \rho_{g_k e_j}) \\ & - \gamma(\rho_{e_i e_j} - \rho_{e_i e_i}^{eq}) \delta_{e_i e_j}, \end{aligned} \quad (1)$$

$$\dot{\rho}_{e_i g_j} = -(i\omega_{e_i g_j} + \Gamma'_{e_i g_j})\rho_{e_i g_j} + (i/\hbar) \left( \sum_{e_k} \rho_{e_i e_k} V_{e_k g_j} - \sum_{g_k} V_{e_i g_k} \rho_{g_k g_j} \right), \quad (2)$$

$$\dot{\rho}_{g_i g_i} = (i/\hbar) \sum_{e_k} (\rho_{g_i e_k} V_{e_k g_i} - V_{g_i e_k} \rho_{e_k g_i}) + (\dot{\rho}_{g_i g_i})_{SE} - \gamma(\rho_{g_i g_i} - \rho_{g_i g_i}^{eq}) + \sum_{g_k, k \neq i} \Gamma_{g_k g_i} \rho_{g_k g_k} - \Gamma_{g_i} \rho_{g_i g_i}, \quad (3)$$

$$\dot{\rho}_{g_i g_j} = -(i\omega_{g_i g_j} + \Gamma'_{g_i g_j})\rho_{g_i g_j} + (i/\hbar) \sum_{e_k} (\rho_{g_i e_k} V_{e_k g_j} - V_{g_i e_k} \rho_{e_k g_j}) + (\dot{\rho}_{g_i g_j})_{SE}, \quad (4)$$

where

$$\begin{aligned} (\dot{\rho}_{g_i g_j})_{SE} &= (2F_e + 1) \Gamma_{F_e \rightarrow F_g} \\ &\times \sum_{q=-1,0,1} \sum_{m_e, m'_e = -F_e}^{F_e} (-1)^{-m_e - m'_e} \\ &\times \begin{pmatrix} F_g & 1 & F_e \\ -m_{g_i} & q & m_e \end{pmatrix} \\ &\times \rho_{m_e m'_e} \begin{pmatrix} F_e & 1 & F_g \\ -m'_e & q & m_{g_j} \end{pmatrix}, \end{aligned} \quad (5)$$

with

$$\Gamma_{F_e \rightarrow F_g} = (2F_g + 1)(2J_e + 1) \begin{Bmatrix} F_e & 1 & F_g \\ J_g & I & J_e \end{Bmatrix}^2 \Gamma \equiv b\Gamma. \quad (6)$$

In Eqs. (1)–(6),  $\Gamma$  is the *total* spontaneous emission rate from each  $F_e m_e$  sublevel whereas  $\Gamma_{F_e \rightarrow F_g}$  is the decay rate from  $F_e$  to one of the  $F_g$  states,  $\Gamma_{g_i}$  is the total collisional decay rate from sublevel  $g_i$ , and  $\Gamma_{g_i g_j}$  is the rate of transfer from sublevel  $g_i \rightarrow g_j$ . The dephasing rates of the excited- to ground-state coherences are given by  $\Gamma'_{e_i g_j} = \frac{1}{2}(\Gamma + \Gamma_{g_j}) + \Gamma^*$ , where  $\Gamma^*$  is the rate of phase-changing collisions. The dephasing rates of the ground-state coherences are given by  $\Gamma'_{g_i g_j} = \frac{1}{2}(\Gamma_{g_i} + \Gamma_{g_j}) + \Gamma^*_{g_i g_j}$ , where  $\Gamma^*_{g_i g_j}$  is the rate of phase-changing collisions. The frequency separation between levels  $a_i$  and  $b_j$ , including Zeeman splitting of the ground and excited levels due to an applied magnetic field, is given by  $\omega_{a_i b_j} = (E_{a_i} - E_{b_j})/\hbar$ , with  $a, b = (g, e)$ , and  $\rho_{a_i a_i}^{eq}$  with  $a = (g, e)$  is the equilibrium population of state  $a_i$ , in the absence of any electrical fields. The interaction energy in the rotating wave approximation for the transition from level  $g_j$  to  $e_i$  is written as

$$\begin{aligned} V_{e_i g_j} &= -\mu_{e_i g_j} (E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t}) \\ &\equiv -\hbar [V_{e_i g_j}(\omega_1) e^{-i\omega_1 t} + V_{e_i g_j}(\omega_2) e^{-i\omega_2 t}], \end{aligned} \quad (7)$$

where  $2V_{e_i g_j}(\omega_{1,2})$  are the pump and probe Rabi frequencies for the  $F_e m_e \rightarrow F_g m_g$  transition, given by

$$\begin{aligned} 2V_{e_i g_j}(\omega_{1,2}) &= \frac{2\mu_{e_i g_j} E_{1,2}}{\hbar} \\ &= (-1)^{F_e - m_e} \begin{pmatrix} F_e & 1 & F_g \\ -m_e & q & m_g \end{pmatrix} \Omega_{1,2}, \end{aligned} \quad (8)$$

where  $\Omega_{1,2} = 2\langle F_e || \mu || F_g \rangle E_{1,2}/\hbar$  [20] are the general pump and probe Rabi frequencies for the  $F_e \rightarrow F_g$  transition. In Eqs. (1)–(4), we include two mechanisms by which atoms may fail to interact with the laser beams. First, they may leave the region of interaction at a rate  $\gamma$  (decay due to time of flight). Second, they can be transferred collisionally from a specific ground sublevel to all the other sublevels of the same  $F_g$  state, and to those of any other nearby hyperfine states that cannot be populated by spontaneous emission from the excited state. We can envisage a reservoir consisting of all the atoms which do not interact with the pump or probe. When collisions are significant,  $\gamma_g^{eff}$ , the effective decay rate to this reservoir from the ground state, will be greater than  $\gamma_g^{eff}$ , the effective decay rate of the excited state, thus enabling EIA-TOP to occur.

Equations (1)–(4) are solved in two stages. In the first, the pump interacts with the system, to all orders in its Rabi frequency. Since  $\rho_{e_i g_j}$  oscillates at the pump frequency  $\omega_1$ , we can write  $\rho_{e_i g_j} = \rho_{e_i g_j}(\omega_1) \exp(-i\omega_1 t)$  and  $\rho_{a_i a_j} = \rho_{a_i a_j}^0$  with  $a = (g, e)$ , where  $\rho_{a_i a_i}^0$  are the populations of the various sublevels and  $\rho_{a_i a_j}^0$  with  $i \neq j$  are the coherences between ground- or excited-state Zeeman sublevels, determined by the pump [14,21,22]. We thus obtain

$$\begin{aligned} \dot{\rho}_{e_i e_j}^0 &= -(i\omega_{e_i e_j} + \Gamma)\rho_{e_i e_j}^0 + i \sum_{g_k} [\rho_{e_i g_k}(\omega_1) V_{g_k e_j}(-\omega_1) \\ &\quad - V_{e_i g_k}(\omega_1) \rho_{g_k e_j}(-\omega_1)] - \gamma(\rho_{e_i e_j}^0 - \rho_{e_i e_i}^{eq}) \delta_{e_i e_j}, \end{aligned} \quad (9)$$

$$\begin{aligned} \dot{\rho}_{e_i g_j}(\omega_1) &= -[i(\omega_{e_i g_j} - \omega_1) + \Gamma'_{e_i g_j}] \rho_{e_i g_j} \\ &\quad + i \left[ \sum_{e_k} \rho_{e_i e_k}^0 V_{e_k g_j}(\omega_1) - \sum_{g_k} V_{e_i g_k}(\omega_1) \rho_{g_k g_j}^0 \right], \end{aligned} \quad (10)$$

$$\begin{aligned} \dot{\rho}_{g_i g_i}^0 &= \sum_{e_k} [\rho_{g_i e_k}(-\omega_1) V_{e_k g_i}(\omega_1) - V_{g_i e_k}(-\omega_1) \rho_{e_k g_i}(\omega_1)] \\ &\quad + (\dot{\rho}_{g_i g_i})_{SE} - \gamma(\rho_{g_i g_i}^0 - \rho_{g_i g_i}^{eq}) + \sum_{g_k, k \neq i} \Gamma_{g_k g_i} \rho_{g_k g_k}^0 \\ &\quad - \Gamma_{g_i} \rho_{g_i g_i}^0, \end{aligned} \quad (11)$$

$$\begin{aligned} \dot{\rho}_{g_i g_j}^0 = & -(i\omega_{g_i g_j} + \Gamma'_{g_i g_j})\rho_{g_i g_j}^0 + i \sum_{e_k} [\rho_{g_i e_k}(-\omega_1) V_{e_k g_j}(\omega_1) \\ & - V_{g_i e_k}(-\omega_1)\rho_{e_k g_j}(\omega_1)] + (\dot{\rho}_{g_i g_j}^0)_{SE}. \end{aligned} \quad (12)$$

In the second stage, interaction with the probe field  $E_2$  is included to first order so that  $\rho_{e_i g_j}$  now oscillates at three frequencies [14,21,22]: the pump frequency  $\omega_1$ , the probe frequency  $\omega_2$ , and the four-wave mixing frequency  $2\omega_1 - \omega_2$ . We therefore express  $\rho_{e_i g_j}$  in terms of its Fourier amplitudes as

$$\begin{aligned} \rho_{e_i g_j} = & \rho_{e_i g_j}(\omega_1)\exp(-i\omega_1 t) + \rho_{e_i g_j}(\omega_2)\exp(-i\omega_2 t) \\ & + \rho_{e_i g_j}(2\omega_1 - \omega_2)\exp[-i(2\omega_1 - \omega_2)t]. \end{aligned} \quad (13)$$

Similarly, the populations and coherences within the same hyperfine level can be written as

$$\begin{aligned} \rho_{a_i a_j} = & \rho_{a_i a_j}^0 + \rho_{a_i a_j}(\omega_2 - \omega_1)\exp[-i(\omega_2 - \omega_1)t] \\ & + \rho_{a_i a_j}(\omega_1 - \omega_2)\exp[-i(\omega_1 - \omega_2)t], \end{aligned} \quad (14)$$

where  $\rho_{a_i a_j}(\omega_2 - \omega_1)$  and  $\rho_{a_i a_j}(\omega_1 - \omega_2)$  are population and coherence oscillations at frequencies  $\omega_2 - \omega_1$  and  $\omega_1 - \omega_2$ . Substituting Eqs. (13) and (14) into Eqs. (1)–(4), we obtain the following set of linear equations for the Fourier amplitudes:

$$\begin{aligned} \dot{\rho}_{e_i e_j}(\omega_2 - \omega_1) = & -[i(\omega_{e_i e_j} - \omega_2 + \omega_1) + \Gamma + \gamma\delta_{e_i e_j}] \\ & \times \rho_{e_i e_j}(\omega_2 - \omega_1) + i \sum_{g_k} [\rho_{e_i g_k}(\omega_2) V_{g_k e_j} \\ & (-\omega_1) - V_{e_i g_k}(\omega_1)\rho_{g_k e_j}(\omega_2 - 2\omega_1) \\ & - V_{e_i g_k}(\omega_2)\rho_{g_k e_j}(-\omega_1)], \end{aligned} \quad (15)$$

$$\begin{aligned} \dot{\rho}_{e_i g_j}(\omega_2) = & -[i(\omega_{e_i g_j} - \omega_2) + \Gamma'_{e_i g_j}]\rho_{e_i g_j}(\omega_2) \\ & + i \left[ \sum_{e_k} \rho_{e_i e_k}^0 V_{e_k g_j}(\omega_2) - \sum_{g_k} V_{e_i g_k}(\omega_2)\rho_{g_k g_j}^0 \right. \\ & + \sum_{e_k} \rho_{e_i e_k}(\omega_2 - \omega_1) V_{e_k g_j}(\omega_1) \\ & \left. - \sum_{g_k} V_{e_i g_k}(\omega_1)\rho_{g_k g_j}(\omega_2 - \omega_1) \right], \end{aligned} \quad (16)$$

$$\begin{aligned} \dot{\rho}_{e_i g_j}(2\omega_1 - \omega_2) = & -[i(\omega_{e_i g_j} - 2\omega_1 + \omega_2) + \Gamma'_{e_i g_j}]\rho_{e_i g_j}(2\omega_1 \\ & - \omega_2) + i \left[ \sum_{e_k} \rho_{e_i e_k}(\omega_1 - \omega_2) V_{e_k g_j}(\omega_1) \right. \\ & \left. - \sum_{g_k} V_{e_i g_k}(\omega_1)\rho_{g_k g_j}(\omega_1 - \omega_2) \right], \end{aligned} \quad (17)$$

$$\begin{aligned} \dot{\rho}_{g_i g_j}(\omega_2 - \omega_1) = & [i(\omega_2 - \omega_1) + \gamma - \Gamma_{g_i}] \rho_{g_i g_j}(\omega_2 - \omega_1) \\ & + i \sum_{e_k} [\rho_{g_i e_k}(-\omega_1) V_{e_k g_j}(\omega_2) + V_{g_i e_k}(\omega_2 \\ & - 2\omega_1)\rho_{e_k g_j}(\omega_1) - V_{g_i e_k}(-\omega_1)\rho_{e_k g_j}(\omega_2)] \\ & + (\dot{\rho}_{g_i g_j})_{SE} + \sum_{g_k, k \neq i} \Gamma_{g_k g_i} \rho_{g_k g_j}(\omega_2 - \omega_1), \end{aligned} \quad (18)$$

$$\begin{aligned} \dot{\rho}_{g_i g_j}(\omega_2 - \omega_1) = & -[i(\omega_{g_i g_j} - \omega_2 + \omega_1) + \Gamma'_{g_i g_j}] \\ & \times \rho_{g_i g_j}(\omega_2 - \omega_1) + i \sum_{e_k} [\rho_{g_i e_k}(-\omega_1) \\ & \times V_{e_k g_j}(\omega_2) + V_{g_i e_k}(\omega_2 - 2\omega_1)\rho_{e_k g_j}(\omega_1) \\ & - V_{g_i e_k}(-\omega_1)\rho_{e_k g_j}(\omega_2)] + (\dot{\rho}_{g_i g_j})_{SE}. \end{aligned} \quad (19)$$

The equations for  $\rho_{a_i a_j}(\omega_1 - \omega_2)$  for  $a = (g, e)$  can easily be written by analogy with Eqs. (15) and (18). As we are only interested in the steady-state results in this paper, we set the time derivatives of the Fourier amplitudes in Eqs. (9)–(12) and (15)–(19) equal to zero. The time evolution of EIT and EIA has recently been discussed by Valente *et al.* [23].

### III. EIA: TRANSFER OF COHERENCE

#### A. Pump and probe polarizations

It was shown by Taichenachev *et al.* [12] that transfer of coherence from the excited state to the ground state due to spontaneous emission is responsible for EIA. However, this explanation alone does not account for the fact that there are systems where the TOC occurs but EIA is not observed. For example, the case where the pump and probe have different polarizations but  $F_e = F_g$ . In this section, we rationalize the experimental conditions needed to achieve EIA as a result of the TOC. These include pump and probe lasers of different polarizations,  $F_e > F_g$  and  $F_g > 0$  [5].

The probe absorption is proportional to the imaginary part of [22,24]

$$\sum_{e_i, g_j} \mu_{e_i g_j} \rho_{e_i g_j}(\omega_2). \quad (20)$$

An explicit expression for  $\rho_{e_i g_j}(\omega_2)$  in the steady state is found from Eq. (16) to be

$$\begin{aligned} \rho_{e_i g_j}(\omega_2) = & \frac{1}{(\omega_{e_i g_j} - \omega_1) - i\Gamma'_{e_i g_j}} \left[ \sum_{e_k} \rho_{e_i e_k}^0 V_{e_k g_j}(\omega_2) \right. \\ & - \sum_{g_k} V_{e_i g_k}(\omega_2)\rho_{g_k g_j}^0 + \sum_{e_k} \rho_{e_i e_k}(\omega_2 - \omega_1) \\ & \left. \times V_{e_k g_j}(\omega_1) - \sum_{g_k} V_{e_i g_k}(\omega_1)\rho_{g_k g_j}(\omega_2 - \omega_1) \right]. \end{aligned} \quad (21)$$

Let us examine the various contributions to the probe absorption [19]. The first and second terms contain the ground- and excited-state populations ( $i=j$ ) and coherences ( $i \neq j$ ) induced by the pump laser. These terms only contribute a constant to the probe absorption. The third and fourth terms involve the ground- and excited-state populations and coherences that oscillate at the pump-probe detuning. It should be noted that the excited-state coherences  $\rho_{e_i e_j}(\omega_2 - \omega_1)$  appear in the fourth term as well as in the third term. This can be seen by writing an explicit expression for  $\rho_{g_i g_j}(\omega_2 - \omega_1)$  in the steady state using Eqs. (19) and writing out the spontaneous emission term of Eq. (5) in detail. The TOC from the excited to ground state via spontaneous emission [12] derives from this spontaneous emission term, which only appears in the Bloch equations for degenerate systems, the simplest of which is the  $N$  system [25].

When we take the selection rules for the pump  $\Delta m_1$  and probe  $\Delta m_2$  into account, we find that the only nonzero terms oscillating at the pump-probe detuning frequency are  $\rho_{a_m a_{m'}}(\omega_2 - \omega_1)$ , where  $a=(g,e)$  and

$$m' = m + \Delta m_1 - \Delta m_2. \quad (22)$$

When the pump and probe have the *same polarization* so that  $\Delta m_2 = \Delta m_1$ , we see from Eq. (22) that  $m' = m$ . Consequently, all the oscillating coherences are zero and only the population oscillations survive. Thus EIA-TOC cannot occur when the fields have the same polarization. By contrast, as we will show later, EIA-TOP occurs precisely when both fields have the same polarization.

However, when the pump and probe have *different polarizations* so that  $\Delta m_2 \neq \Delta m_1$ , we see from Eq. (22) that  $m' \neq m$ . Thus, all the oscillating populations are zero and only the oscillating ground- and excited-state coherences can be nonzero. This explains why EIA-TOC only occurs when the fields have different polarizations.

### B. Oscillating coherences

Here we will show that the value of the excited-state oscillating coherence that leads to TOC depends on the excited-state population produced by the pump. As a consequence, only systems where the population is not trapped in the ground state, either coherently or incoherently, can exhibit EIA. In Fig. 1, we show that trapping occurs in all degenerate two-level systems, pumped by a circularly polarized ( $\Delta m = +1$  or  $-1$ ) or  $\pi$ -polarized ( $\Delta m = 0$ ) laser, for which  $F_e \leq F_g$ . We note that Renzoni *et al.* [17] have shown that coherent population trapping occurs for all cases where an  $F_g \rightarrow F_e \leq F_g$  transition is pumped by a  $\sigma$ -polarized laser. In Fig. 2, we show that there is significant population in the excited state when an  $F_g \rightarrow F_e = F_g + 1$  transition is pumped by a  $\sigma_{\pm}$  or  $\pi$ -polarized laser [26]. The correlation between the magnitude of the coherence oscillations and that of the pump-induced populations is established in Fig. 3 for an artificially closed  $F_g = 1 \rightarrow F_e = 2$  transition interacting with a  $\sigma_+$  pump and a  $\sigma_-$  probe, where decay due to time of flight is included but collisions are excluded. The ground- and excited-state oscillating coherences are shown in Figs. 3(a)

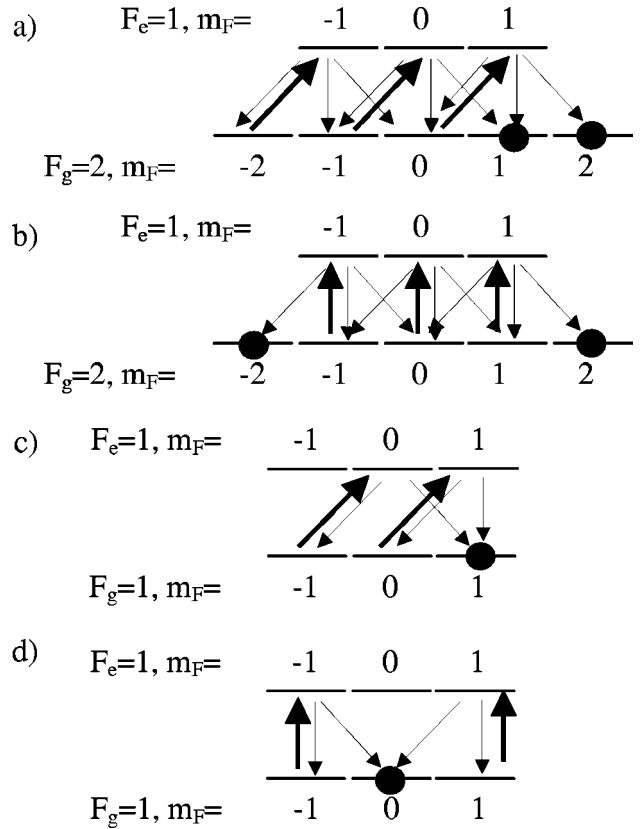


FIG. 1. Population trapping due to interaction of (a),(b)  $F_g = 2 \rightarrow F_e = 1$  and (c),(d)  $F_g = 1 \rightarrow F_e = 1$  transitions with (a),(c)  $\sigma_+$ - and (b),(d)  $\pi$ -polarized pump, in the absence of collisions and decay to the reservoir. The thick arrows indicate interaction with the pump and the thin arrows indicate spontaneous emission. Note that in (d), the transition between  $m = 0$  sublevels is forbidden so that the population is trapped in  $m_g = 0$  sublevel.

and 3(b), as a function of the pump-probe detuning, and the populations of the various sublevels are shown in Fig. 3(c). We see that the highest excited-state coherence is about a third of the ground-state coherence, and that a sixth of the total population is in the excited state. The other two excited-

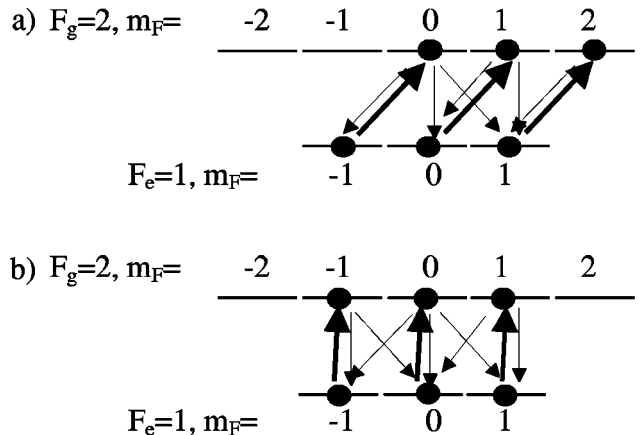


FIG. 2. Ground- and excited-state pump-induced populations for  $F_g = 1 \rightarrow F_e = 2$  transition, in the absence of collisions and decay to the reservoir.

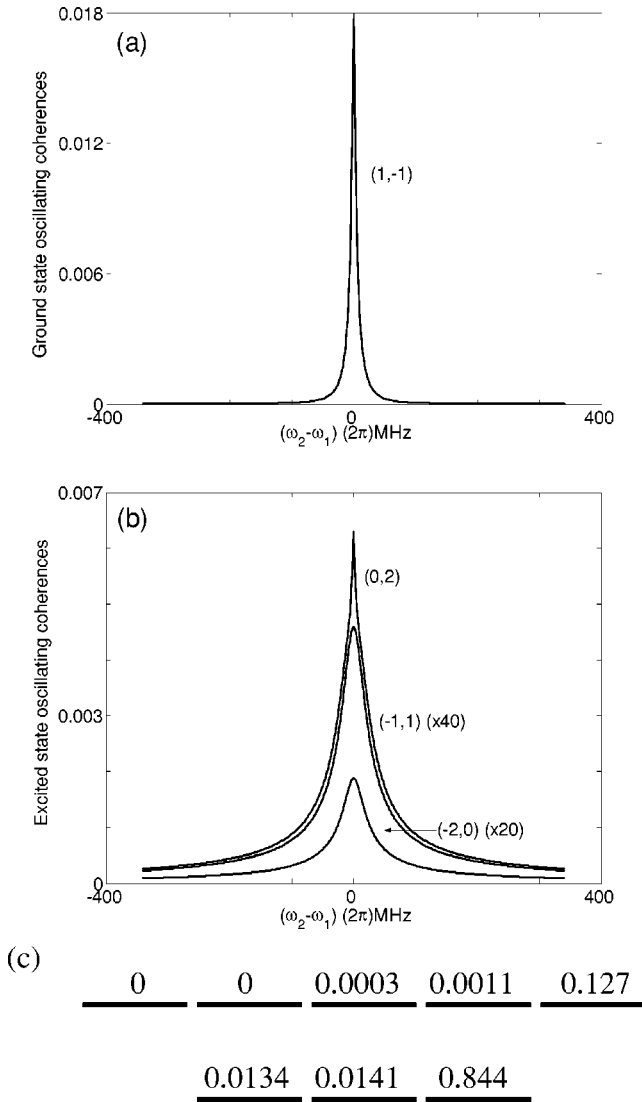


FIG. 3. (a) Ground- and (b) excited-state coherences  $\rho_{a_m a_{m'}}(\omega_2 - \omega_1)$  with  $a = (g, e)$  oscillating at pump-probe detuning as a function of pump-probe detuning for an artificially closed  $F_g = 1 \rightarrow F_e = 2$  transition (for  $^{87}\text{Rb}$ ), interacting with a  $\sigma_+$ -polarized pump with intensity  $1 \text{ mW cm}^{-2}$  and a  $\sigma_-$ -polarized probe, with  $\Gamma = 34 \text{ rad sec}^{-1}$ . Transitions are labeled according to  $(m, m')$ . (c) Fraction of population in each Zeeman sublevel. Note correlation between the coherences and the populations.

state coherences between excited states with almost no population are about 40 times smaller. Similarly, it can be shown that the excited-state coherences are very small for population-trapped systems for which the pump-induced steady-state population in the excited states is very small. We thus conclude that TOC, and hence EIA, can only take place for systems where the population is not trapped in the ground state, that is, in  $F_g \rightarrow F_e = F_g + 1$  transitions.

### C. Open systems

In a *closed* system, atoms excited to the state  $F_e$  can only decay spontaneously to the pumped ground state  $F_g$ , whereas in an *open* system, they can decay to other ground

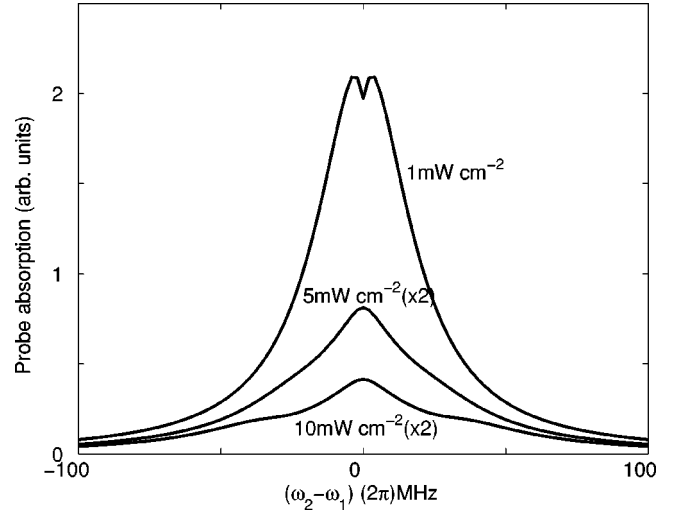


FIG. 4. Calculated probe absorption for the open transition,  $F_g = 1 \rightarrow F_e = 2$  of  $^{87}\text{Rb}$  ( $b = 0.5$ ) for increasing pump intensity in the absence of collisions. Pump is  $\pi$  polarized ( $\Delta m = 0$ ) and probe is  $\sigma$  polarized ( $\Delta m = \pm 1$ ) and  $\gamma_e = \gamma_g = 0.001\Gamma$ .

hyperfine states. In Eq. (6), the decay rate for a specific transition was written as  $\Gamma_{F_e \rightarrow F_g} \equiv b\Gamma$  with  $b = 1$  for a closed system and  $b < 1$  for an open system. Let us examine the effect of introducing additional decay channels, thereby reducing the value of  $b$  from unity. First, the total steady-state pump-induced population in the original  $F_g$  and  $F_e$  states is now less than unity and the excited-state oscillating coherences which correlate with the populations are also reduced. Second, the TOC from the excited state to the ground state decreases, leading to reduced EIA. However, the effect of reducing  $b$  can be mitigated by increasing the pump intensity. In Fig. 4, we plot the absorption spectrum for the  $D_1$  transition of  $^{87}\text{Rb}$  ( $F_g = 1 \rightarrow F_e = 2$ ) which forms an open system ( $b = 0.5$ ), since the excited state can also decay to  $F_g = 2$ . We see that as the pump intensity increases from  $1 \text{ mW cm}^{-2}$  (corresponding to a Rabi frequency of  $0.94\Gamma$ ) to  $10 \text{ mW cm}^{-2}$  (corresponding to a Rabi frequency of  $2.98\Gamma$ ), the overall probe absorption decreases as a result of increased excited-state population, and hence increased decay from the excited state to the noninteracting ground state. However, we also see that EIT is replaced at higher intensity by EIA. This is due to the relatively larger population in the excited state, which results in higher excited-state coherence and thus increased TOC. This may explain some of the seemingly contradictory results obtained for open systems. According to the calculations of Akulshin and co-workers [5,27], EIA does not take place in open systems. However, Alzetta *et al.* [10] have observed EIA peaks in Hanle experiments on open systems. Our calculations suggest that Akulshin and co-workers would have obtained EIA for open systems had they used higher Rabi frequencies.

### D. The role of decay to the reservoir

We pointed out in Sec. III A that TOC only occurs when the polarizations of the pump and probe are different, since only in this case are the coherence oscillations nonzero. It

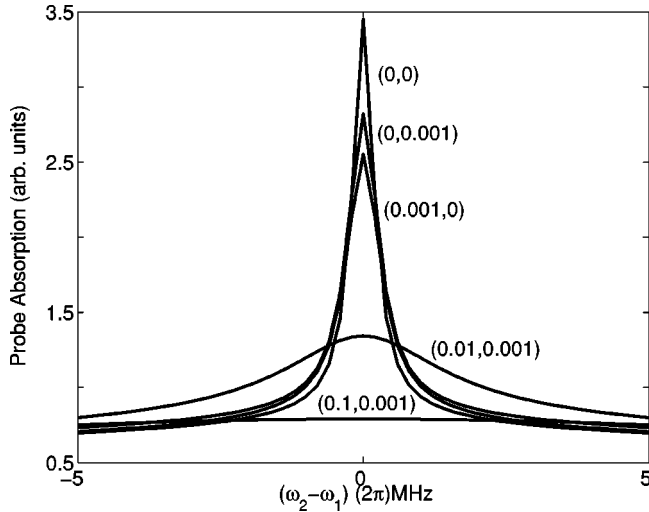


FIG. 5. Calculated probe absorption (EIA-TOC) for the closed transition  $F_g=2 \rightarrow F_e=3$  of  $^{87}\text{Rb}$  ( $b=1$ ), with the addition of collisions. Curves are labeled according to  $(\Gamma_{\text{same}F}^{\text{coll}}/\Gamma, \Gamma_{\text{diff}F}^{\text{coll}}/\Gamma)$ . Pump intensity is  $10 \text{ mW/cm}^2$ , the pump is  $\pi$  polarized ( $\Delta m=0$ ) and the probe is  $\sigma$  polarized ( $\Delta m=\pm 1$ ), and  $\gamma_e = \gamma_g = 0.001\Gamma$ .

can be seen from Eqs. (15) and (19) that decay due to time of flight does not directly affect the coherence oscillations, whereas collisions do affect them through the transverse decay rate. The time-of-flight decay affects the coherence oscillations indirectly through the pump-induced coherences  $\rho_{e_i g_j}(\omega_1)$  that depend on the pump-induced populations of the ground and excited states [see Eq. (10)] which are influenced by decay to the reservoir, both due to time of flight and collisions. Thus the main effect of the reservoir is to redistribute the population among the sublevels which interact with the pump field and those which do not. In the presence of decay to the reservoir, some of the sublevels that are unoccupied in Figs. 1 and 2 will be occupied to some extent and the associated coherences will be different from zero (see Fig. 3). In our numerical calculations, we take the decay rates between the sublevels of the  $F_g$  state that interacts with the pump to be different from those to the sublevels of any nearby hyperfine states. In Fig. 5, we show the absorption spectrum for the  $F_g=2 \rightarrow F_e=3$  closed transition of  $^{87}\text{Rb}$  interacting with a  $\pi$ -polarized ( $\Delta m=0$ ) pump and a  $\sigma$ -polarized ( $\Delta m=\pm 1$ ) probe, for various values of the collision rate  $\Gamma_{g_i g_j}$  between the  $F_g=2$  sublevels ( $\Gamma_{\text{same}F}^{\text{coll}}$ ) and between  $F_g=2$  and  $F_g=1$  ( $\Gamma_{\text{diff}F}^{\text{coll}}$ ). We see that collisions to  $F_g=1$  sublevels decrease the EIA-TOC peak. Collisions between the  $F_g=2$  sublevels decrease the EIA-TOC peak substantially and may even wash it out completely due to collisional broadening.

#### IV. EIA: TRANSFER OF POPULATION

As we explained in Sec. III A, EIA-TOC cannot occur when the polarizations of the pump and probe are the same. It is precisely in this case that EIA-TOP, which is analogous to EIA in two-level systems [13], occurs. The reason for this is that the degenerate two-level system then reduces to a series of two-level systems, which are interlinked due to

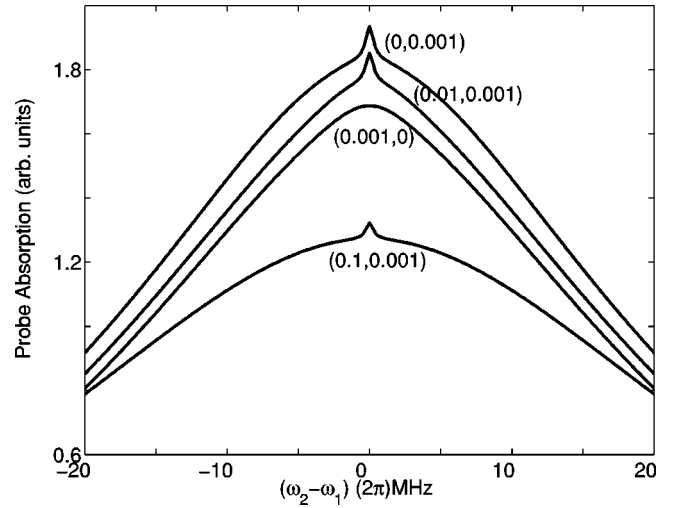


FIG. 6. Calculated probe absorption (EIA-TOP) for the closed transition  $F_g=2 \rightarrow F_e=3$  of  $^{87}\text{Rb}$  ( $b=1$ ), with the addition of collisions. Curves are labeled according to  $(\Gamma_{\text{same}F}^{\text{coll}}/\Gamma, \Gamma_{\text{diff}F}^{\text{coll}}/\Gamma)$ . Pump intensity is  $1 \text{ mW/cm}^2$ , pump and probe are  $\pi$  polarized ( $\Delta m=0$ ), and  $\gamma_e = \gamma_g = 0.001\Gamma$ .

spontaneous emission and population transfer to and from the reservoir. It can be seen from Figs. 1 and 2 that describing the system in terms of two-level systems is only possible when  $F_e = F_g + 1$ .

It will be recalled that EIA-TOP requires  $\gamma_g^{\text{eff}} > \gamma_e^{\text{eff}}$ . This can be achieved by including collisional transfer of population between the ground-state sublevels and also to the sublevels of nearby noninteracting hyperfine states. Obviously, this effect can only occur in closed systems, since in an open system the spontaneous decay from the excited state to the reservoir will ensure that  $\gamma_e^{\text{eff}} > \gamma_g^{\text{eff}}$ . In Fig. 6, we show the absorption spectrum for the same closed transition as in Fig. 5, interacting with a  $\pi$ -polarized pump and probe, for various values of  $\Gamma_{\text{same}F}^{\text{coll}}$  and  $\Gamma_{\text{diff}F}^{\text{coll}}$ . The collisions between  $F_g=2$  and  $F_g=1$  are crucial for producing the EIA-TOP peak. However, the collisions within the  $F_g=2$  state alter the contributions of the individual two-level systems due to changes in the pump-induced populations and population oscillations. As a result, the overall absorption becomes smaller and broader with increasing  $\Gamma_{\text{same}F}^{\text{coll}}$ .

#### V. FWM

It is interesting to compare the FWM signals that arise in cases where EIA-TOC and EIA-TOP are obtained. The four-wave mixing signal is given by the absolute value squared of

$$\sum_{e_i, g_j} \mu_{e_i g_j} \rho_{e_i g_j} (2\omega_1 - \omega_2). \quad (23)$$

An explicit expression for  $\rho_{e_i g_j}(2\omega_1 - \omega_2)$  in the steady state can be obtained from Eq. (17):

$$\rho_{e_i g_j}(2\omega_1 - \omega_2) = \frac{1}{(\omega_{e_i g_j} - 2\omega_1 + \omega_2) - i\Gamma'_{e_i g_j}} \times \left[ \sum_{e_k} \rho_{e_i e_k}(\omega_1 - \omega_2) V_{e_k g_j}(\omega_1) - \sum_{g_k} V_{e_i g_k}(\omega_1) \rho_{g_k g_j}(\omega_1 - \omega_2) \right]. \quad (24)$$

By the same analysis as in Sec. III A, we find that the FWM signal obtained with fields of different polarizations [16] depends on the ground- and excited-state oscillating coherences and will be called FWM-TOC for convenience. On the other hand, the FWM signal [14] obtained with the same polarizations depends on the population oscillations and will be called FWM-TOP.

#### A. FWM: Transfer of coherence

It can be shown by comparing Eqs. (16) and (17) that the contribution of the coherence oscillations to the imaginary part of  $\rho_{e_i g_j}(\omega_2)$  has the opposite sign to the imaginary part of  $\rho_{e_i g_j}(2\omega_1 - \omega_2)$ . Thus the sharp EIA-TOC peak obtained for closed systems [see Fig. 7(a)] leads to a deep dip of negative sign in the imaginary part of  $\rho_{e_i g_j}(2\omega_1 - \omega_2)$  and hence to a sharp peak in its contribution to the FWM signal. The real part of  $\rho_{e_i g_j}(2\omega_1 - \omega_2)$  is dispersive passing through zero when  $\omega_2 = \omega_1$ , so that its square contributes a peak with a sharp dip in the center. If the dip in the imaginary part is sufficiently negative, it will show up as a sharp peak in the FWM signal (see Fig. 7(b) and also Fig. 9 of Ref. [27]). We showed in Sec. III C that the EIA-TOC signal decreases as  $b$  decreases, that is, as the system becomes more open. Thus, the dip in the imaginary part of  $\rho_{e_i g_j}(2\omega_1 - \omega_2)$  also decreases and is overtaken by the background so that the FWM is now characterized by a dip. The FWM-TOC spectrum for the same open system as in Fig. 4 is shown in Fig. 8. There we see that a dip in the absorption for pump intensity  $1 \text{ mW cm}^{-2}$  corresponds to a peak in the FWM spectrum, whereas the small peaks in the absorption at higher intensities correspond to small dips in the FWM spectrum.

#### B. FWM: Transfer of population

The FWM spectrum of a two-level system whose absorption spectrum has a small sharp peak is characterized by a deep dip [14]. The same is true for the FWM-TOP spectrum as shown in Fig. 9, which was calculated for the same parameters as in Fig. 6. We see a correlation between the heights of the sharp EIA-TOP peaks in Fig. 6 and the depths of the dips in the FWM-TOP spectra of Fig. 9.

### VI. CONCLUSION

We have shown that there are two kinds of EIA in degenerate two-level systems: EIA-TOC and EIA-TOP. EIA-TOC

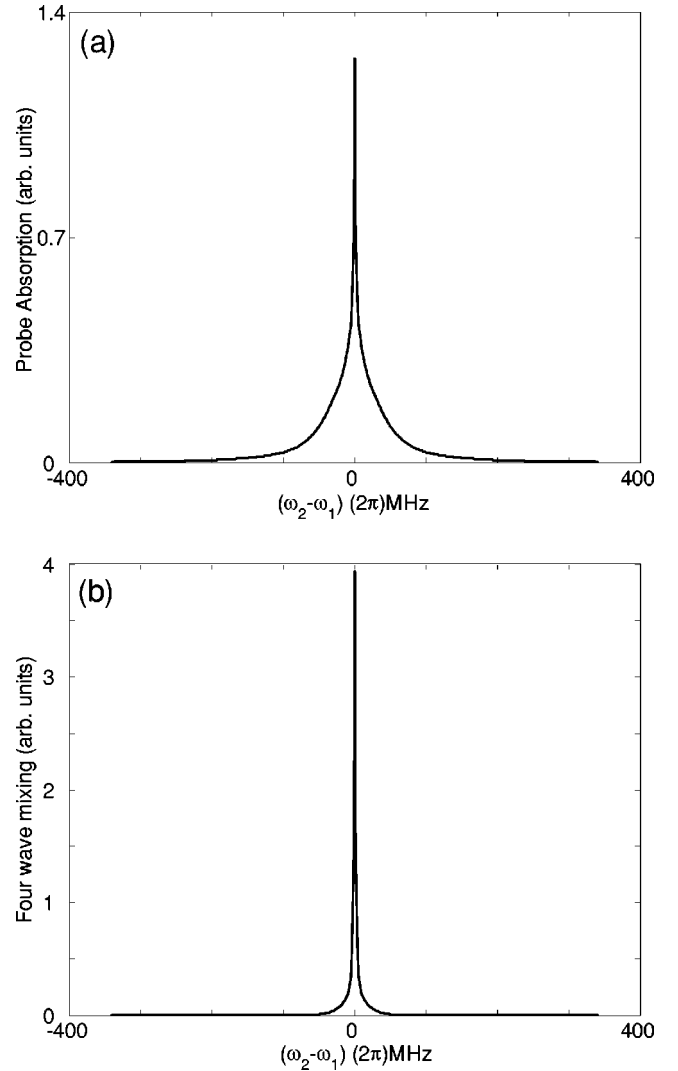


FIG. 7. (a) Calculated probe absorption and (b) four-wave mixing for the closed transition  $F_g=1 \rightarrow F_e=2$ . Pump intensity is  $5 \text{ mW/cm}^2$ , the pump is  $\pi$ -polarized ( $\Delta m=0$ ), the probe is  $\sigma$  polarized ( $\Delta m=\pm 1$ ), and  $\gamma_e = \gamma_g = 0.001\Gamma$ .

is due to TOC from the excited state to the ground state via spontaneous emission whereas EIA-TOP is due to the collisional TOP to Zeeman sublevels that do not interact with the pump. From an analysis of the equations of motion, we have shown that there is a correlation between the magnitude of the excited-state oscillating coherences and the pump-induced population in the excited state so that EIA-TOC can only occur for  $F_g \rightarrow F_e = F_g + 1$  transitions. We demonstrated that the excited-state oscillating coherences only exist when the pump and probe have different (not necessarily perpendicular) polarizations. EIA-TOC was shown to occur on open systems as well as closed systems provided the pump Rabi frequency is sufficiently high.

EIA-TOP which is characterized by smaller sharp peaks than EIA-TOC is analogous to an effect that was predicted in simple two-level systems. It occurs when the degenerate two-level system reduces to a series of two-level systems interlinked by spontaneous emission and collisional TOP

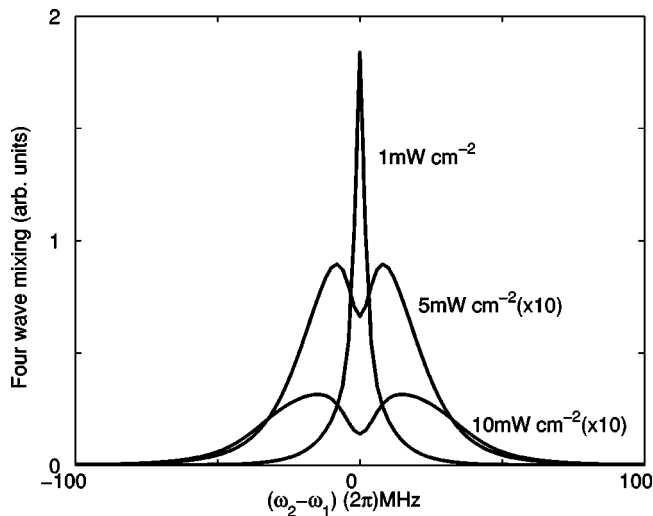


FIG. 8. Four-wave mixing (FWM-TOC) spectrum for the same system as in Fig. 4.

between the ground Zeeman sublevels of the pumped hyperfine state and, more importantly, to other unpumped hyperfine states. The analogy with the simple two-level system explains why EIA-TOP only occurs for closed systems interacting with a pump and probe that have the same polariza-

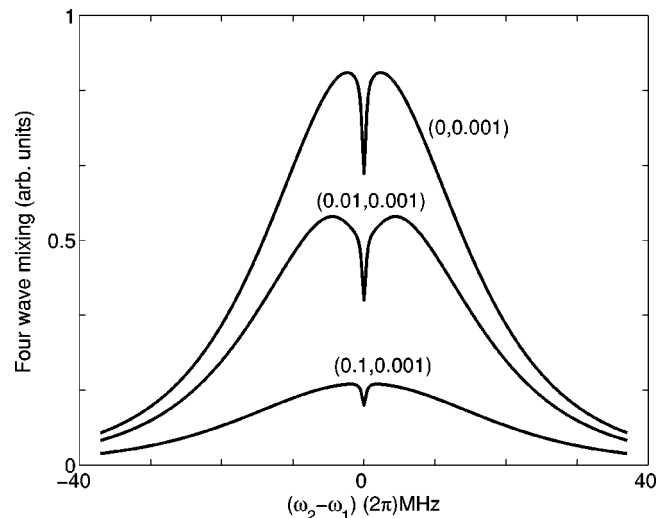


FIG. 9. Four wave mixing (FWM-TOP) spectrum for the same system as in Fig. 6.

tion. Furthermore, it explains why EIA-TOP, like EIA-TOC, takes place for  $F_g \rightarrow F_e = F_g + 1$  transitions where there is significant population in the excited states. Finally, we discussed the FWM spectra that are obtained in the presence of TOC and TOP.

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