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Spontaneous emission quenching

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# Effects of Quantum Interference in Spontaneous Emission: Spectral line elimination and spontaneous emission cancellation

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# I. Introduction

Recently quantum interference and coherence in a multilevel atomic system have attracted a lot of attention, because they can lead to absorption cancellation, electromagnetically induced transparency (EIT), and population inversion without emission [3]. These quantum interference effects may result in a new type of laser system operating without population inversion (LWI) and transparent high-index materials. The spectrum of spontaneous emission from a two-level atom is Lorentzian with a peak at the atomic transition frequency, and the width of the spectrum depends on the decay rate of the upper level. It was pointed out that the noise of the radiation field from the new-type laser systems (LWI) might be less compared to the noise of a laser light from two-level laser systems. We disease the spontaneous emission from a three-level atom with two upper levels coupled by the same vacuum modes and how the quantum interference affects the spontaneous emission process and its spectrum.

# II. Pure Three Level Systems

# (A) Basic Equations

Consider a-three level atom with two upper levels  $|a_1\rangle$  and  $|a_2\rangle$  as shown in Fig. 1a. The two upper levels are coupled by the same vacuum modes to the lower level  $|b\rangle$ .

The interaction Hamiltonian of the system composed of the atom and the vacuum modes in the interaction picture can be written as

$$V = i \sum_{k} \left[ g_{k}^{(1)} e^{i(\omega_{\alpha_{l}b} - \omega_{k})t} b_{k} |\alpha_{1}\rangle\langle b| + g_{k}^{(2)} e^{i(\omega_{\alpha_{2}b} - \omega_{k})t} b_{k} |\alpha_{2}\rangle\langle b| \right]$$

$$-i \sum_{k} \left[ g_{k}^{(1)} e^{-i(\omega_{\alpha_{l}b} - \omega_{k})t} b_{k}^{+} |b\rangle\langle a_{1}| + g_{k}^{(2)} b_{k}^{+} e^{-i(\omega_{\alpha_{2}b} - \omega_{k})t} |b\rangle\langle a_{2}| \right]$$

$$(1)$$

where  $\omega_{a,b}$ ,  $\omega_{a,b}$  are the frequency difference between level  $|a_1\rangle$ ,  $|a_2\rangle$  and  $|b\rangle$ ,  $b_k(b_k^+)$  is the annihilation(creation) operator for the kth vacuum mode with frequency  $\omega_k$ , and  $g_k^{(1,2)}$  are the coupling constants between the kth vacuum mode and the atomic transitions from  $|a_1\rangle$  and  $|a_2\rangle$  to  $|b\rangle$ . Here k stands for both momentum and polarization of the vacuum modes and  $\hbar=1$  and real  $g_k^{(1,2)}$  have been assumed. This Hamiltonian controls the spontaneous emission of the atom initially in the upper levels.

The initial state vector can be written as

$$\left|\psi(0)\right\rangle = A^{(1)}(0)\left|a_{1}\right\rangle \left|0\right\rangle + A^{(2)}(0)\left|a_{2}\right\rangle \left|0\right\rangle. \tag{2}$$

The evolution of the state vector obeys the Schrodinger equation,

$$\frac{d}{dt}|\psi(t)\rangle = -iV|\psi(t)\rangle. \tag{3}$$

The state vector at time t can be written as

$$|\psi(t)\rangle = A^{(1)}(t)|a_1\rangle|0\rangle + A^{(2)}(t)|a_2\rangle|0\rangle + \sum_{k} B_k(t)b_k^+|0\rangle|b\rangle. \tag{4}$$

Substituting Eq. (4) into (3), we can obtain

$$\frac{d}{dt}A^{(1)}(t) = \sum_{k} g_{k}^{(1)} e^{i(\omega_{\alpha_{1}b} - \omega_{k})t} B_{k}(t), \tag{5a}$$

$$\frac{d}{dt}A^{(2)}(t) = \sum_{k} g_{k}^{(2)} e^{i(\omega_{\alpha_{2}b} - \omega_{k})t} B_{k}(t), \tag{5b}$$

$$\frac{d}{dt}B_k(t) = -g_k^{(1)}A^{(1)}(t)e^{-i(\omega_{a_1b}-\omega_k)t} - g_k^{(2)}A^{(2)}(t)e^{i(\omega_{a_2b}-\omega_k)t}.$$
 (5c)

Formally integrating Eq. (5c), and then substituting into Eqs. (5a) and (5b), we find

$$\frac{d}{dt}A^{(1)}(t) = -\frac{\gamma_1}{2}A^{(1)}(t) - \frac{\sqrt{\gamma_1\gamma_2}}{2}A^{(2)}(t)e^{t\omega_{12}t},$$

\_\_\_\_ (6a)

$$\frac{d}{dt}A^{(2)}(t) = -\frac{\gamma_2}{2}A^{(2)}(t) - \frac{\sqrt{\gamma_1\gamma_2}}{2}A^{(1)}(t)e^{-t\omega_{12}t},$$
 (6b)

where  $\omega_{12}$  is the frequency difference between the two upper levels and is assumed to be much less than  $\omega_{a_1b}$ ,  $\gamma_1 = 2\pi g^{(1)^2} D(\omega_1)$ , and  $\gamma_2 = 2\pi g^{(2)^2} D(\omega_2)$ . Here  $g^{(1)}$ ,  $D(\omega_1)$ and  $g^{(2)}$ ,  $D(\omega_2)$  are calculated at frequencies  $\omega_{a_1b}$ , and  $\omega_{a_2b}$ , respectively, and  $D(\omega)$ is the mode density. In obtaining Eqs. (6)  $\omega_{12} << \omega_{a_1b}$ ,  $\omega_{a_2b}$  have been assumed (but not  $\omega_{12} << \gamma_1, \gamma_2$ ) and we have assumed that the two dipole moments of the two transitions are parallel to each other (anti parallel will be the same). Here we did not neglect the time dependent exponential factors  $(e^{\pm \omega_{11}t})$  as was done in a previous similar work [6]. From Eqs. (6) we can obtain the equation of motion for the reduced density matrix of the atom [15]. In order to obtain the spontaneous spectrum, however, Eq. (5c) is necessary, which includes the information of the field radiated by the atom. Similar equations can be found in the problem of photoionization [16].

Solving Eq.(6), we obtain the solution for  $A^{(1)}(t)$  and  $A^{(2)}(t)$ ,

$$A^{(1)}(t) = (C_1 e^{s_1 t} + C_2 e^{S_2 t}) e^{-\frac{y_1}{2}t}, \tag{7a}$$

$$A^{(2)}(t) = -\frac{2}{\sqrt{\gamma_1 \gamma_2}} (S_1 C_1 e^{S_1 t} + S_2 C_2 e^{S_2 t}) e^{-(\frac{\gamma_1}{2} + i\omega_{12})t},$$
 (7b)

where  $S_{1,2}$  are two roots of the equation  $S^2 - \lambda S - 0.25\gamma_1\gamma_2 = 0$ ,

$$S_{1,2} = \frac{1}{2} \left( \lambda \pm \sqrt{\lambda^2 + \gamma_1 \gamma_2} \right), \tag{8a}$$

$$\lambda = \frac{1}{2} (\gamma_1 - \gamma_2) + i\omega_{12},$$

(8b)

$$C_1 = \frac{S_2 A^{(1)}(0) + 0.5 \sqrt{\gamma_1 \gamma_2} A^{(2)}(0)}{S_2 - S_1},$$
(8c)

$$C_2 = \frac{S_1 A^{(1)}(0) + 0.5 \sqrt{\gamma_1 \gamma_2} A^{(2)}(0)}{S_1 - S_2}.$$
 (8d)

From Eqs. (7) we can calculate, the evolution of the populations. The population in the two upper levels are equal to  $\left|A^{(1)}(t)\right|^2$  and  $\left|A^{(2)}(t)\right|^2$ , respectively, which can be obtained from Eqs. (7).

The spontaneous spectrum  $S(\omega)$  is the Fourier transform of

$$\langle E^{-}(t+\tau)E^{+}(t)\rangle_{t=\infty} = \langle \psi(t)|\sum_{k,k} b_{k}^{+} b_{k}^{-} e^{i\omega_{k}(t+\tau)} e^{-i\omega_{k}^{-}t} |\psi(t)\rangle_{t=\infty}. \tag{10}$$

Substituting Eq. (4) into (10) and we have

$$\langle E^{-}(t+\tau)E^{+}(t)\rangle_{t=\infty} = \sum_{k} B_{k}^{*}(\infty)B_{k}(\infty)e^{i\omega_{k}\tau} = \int_{-\infty}^{\infty} d\omega D(\omega)B_{k}^{*}(\infty)B_{k}(\infty)e^{i\omega\tau}. \tag{11}$$

From Eq(11) we find

$$S(\omega_k) = \gamma \left| B_k(\infty) \right|^2 / 2\pi g^2. \tag{12}$$

The spontaneous spectrum is proportional to  $|B_k(\infty)|^2$ . Substituting Eqs(7) into (5c), and then integrating Eq. (5c) we can obtain,

$$B_{k}(\infty) = \frac{g_{k}^{(1)}C_{1}(1-2S_{1}/\gamma_{1})}{S_{1}-\gamma_{1}/2-i(.5\omega_{12}-\delta_{k})} + \frac{g_{k}^{(1)}C_{2}(1-2S_{2}/\gamma_{1})}{S_{2}-\gamma_{1}/2-i(.5\omega_{12}-\delta_{k})},$$
(13)

where  $\delta_k = \omega_k - .5(\omega_{a_1} + \omega_{a_2}) + \omega_b$  is the detuning of the *k*th vacuum mode with respect to the central frequency (from the middle point of the two upper levels to the lower level). The spontaneous spectrum can be obtained by taking absolutely square of Eq. (10), which not only depends on the square of each terms in above equation, but also on their interference terms. The interference results in some very interesting features.

#### (B). Dark lines

For a two-level atom, its spontaneous spectrum is Lorentzian and peaked at its transition frequency due to the population transfer from its upper level to the lower level. In the three-level atom case, the population initially in one upper level (say  $|a_1\rangle$ ) is partly transferred to another upper level (say  $|a_2\rangle$ ) during the time evolution. It is expected that the spontaneous spectrum of the three-level atom will differ from its counterpart of a two-level atom due to the transferred population in  $|a_2\rangle$ . This population in  $|a_2\rangle$  will eventually decay to the lower level. Therefore, a major difference will be the weight of the frequency components around the transition frequency from  $|a_2\rangle$  to lower  $(\omega_{a_2}-\omega_b)$  in the spontaneous emission spectrum. The weight of these components might be larger for the three-level atom than that for a two-level atom. In Fig. 5, we plot two spontaneous emission spectra as functions of

the detuning  $\delta_k$ , one for the three-level atom, and one for a two-level atom. The three-level atom is initially in upper level  $|a_1\rangle$ . Comparing the two curves, we find that some components of the three-level spectrum at the neighborhood of  $\omega_{a_1} - \omega_b$  are much larger than their counterpart of a two-level spectrum. If we simply added the two spontaneous decay processes together, we might conclude that the spectrum would have two peaks at the two transition frequencies. However, the spontaneous emission spectrum of the three-level atom is not a simple two-peak distribution peaked at the two transitions from levels  $|a_1\rangle$  and  $|a_2\rangle$  to the lower level. There is strong interference between the two processes. Therefore, the spectrum of the three-level atom is not a two peak one with the two peak located on the two sides of the central frequency. The interference leads to a dark line in the spontaneous spectrum. In Fig. 2, we plot the spontaneous spectra for a three-level atom and a two level atom, where it can be seen very clear that there is a dark line in the spectrum of the three-level atom at the frequency of the transition from  $|a_2\rangle$  to the lower level. The dark line results from the interference. In fact, it can been proven that,  $B_k(\infty) = 0$  at  $\omega_k$  being equal to the frequency of one transition, when the atom is initially in the upper level of the other transition.

Assume the atom initially in level  $|a_1\rangle$ , and consider  $B_k(\infty)$  at  $\omega_k = \omega_{a_2b}$   $(\delta_k = -0.5\omega_{12})$ . Substituting  $\delta_k = -0.5\omega_{12}$  in Eq. (13), we can obtain,

$$B_{k}(\infty) = \frac{C_{1}(1-2S_{1}/\gamma_{1})}{S_{1}-0.5\gamma_{1}-i\omega_{12}} + \frac{C_{2}(1-2S_{2}/\gamma_{1})}{S_{2}-0.5\gamma_{1}-i\omega_{12}}$$

$$= \frac{4C_{1}S_{1}(-S_{2}-0.5\gamma_{2})/\gamma_{1}\gamma_{2}}{S_{1}-0.5\gamma_{1}-i\omega_{12}} + \frac{4C_{2}S_{2}(-S_{1}-0.5\gamma_{2})/\gamma_{1}\gamma_{2}}{S_{2}-0.5\gamma_{1}-i\omega_{12}}$$

$$= \frac{4(S_{1}C_{1}+S_{2}C_{2})}{\gamma_{1}\gamma_{2}}.$$
(14)

In obtaining the above equation

$$S_1 S_2 = -\frac{\gamma_1 \gamma_2}{4},\tag{15a}$$

$$S_1 + S_2 = 0.5(\gamma_1 - \gamma_2) + i\omega_{12},$$
 (15b)

have been used. Because the atom is initially in  $|a_1\rangle$ , we have  $A^{(1)}(0) = -$  and  $A^{(2)}(0) = 0$ . From Eqs. (8c) and (8d), we can find  $S_1C_1 + S_2C_2 = 0$ , which yields  $B_k(\infty)=0$ . From Eq. (12) we get  $S(\omega_k=\omega_{a_1b})=0$ . This tells us that not only is there a dark lines in the spontaneous emission spectrum, but also that the center of the dark line is absolutely black, independent of  $\gamma_1, \gamma_2$  and  $\omega_{12}$ , as long as the two coupling constants are not equal to zero (if one of them is zero, the three-level atom reduces to a two-level one). In addition, we can see in Fig. 2 that there was an attempt to build a peak at the dark line position due to the transferred population.

The width of the dark line depends on the decay rate of the upper level of corresponding transition. In Fig. 3, we show two spontaneous emission spectra of the three-level atom initially in  $|a_1\rangle$ , with the same  $\gamma_1$  and  $\omega_{12}$ , but different  $\gamma_2$  (=0.5 $\gamma_1$  and 0.05 $\gamma_1$ ). It is clear that the larger the decay rate of the corresponding upper level ( $|a_2\rangle$  in this example), the wider the width of the dark line will be. As  $\gamma_2$  (or  $\gamma_1$ ) tends to zero, the dark line becomes narrower and narrower (and will finally disappear), and the spectrum becomes closer and closer to a Lorentzian distribution.

# (C). Spectral narrowing

As mentioned above, the dark line is absolutely black at its center, and its width depends on the decay rate of the corresponding upper level. A larger decay rate results in wider width. In the above example we use a small value for  $\gamma_2$  (more precisely  $\gamma_2/\gamma_1$ ) in order to show clear dark lines. On the other hand, if  $\gamma_2$  is of the same order or is ever larger than  $\gamma_1$ , the width of the dark line will be big enough to depress one of the two wings of the spectrum. Consequently, the spontaneous emission spectrum can be greatly narrowed. In Fig. 4, we plot the spectra of the three-level atom for different values of the ratios  $\gamma_2/\gamma_1 = 0.01$ , 0.5, and 2, where we can see that the width of the dark line increases and the spectrum becomes narrower as the ratio increase. In Fig. 5, we compare the spectrum of the three-level atom with that of a two-level atom. The parameters used for the two-level atom are the same as

those used for the three-level atom, except  $\gamma_2=0$  (because there is no level  $|a_2\rangle$ ). The width of the spontaneous spectrum for the three-level atom is much narrower than that for a two-level atom.

The spectrum may have three peaks as shown in Fig. 2, and may have two peaks, as shown in Fig. 5, depending on the parameters  $\gamma_1, \gamma_2$  and  $\omega_{12}$ . From  $dS(\omega_k)/d\omega_k = 0$  we can obtain a fifth-order polynomial, which may have five real roots corresponding to three peaks, and may have three real roots corresponding to two peaks. One of the roots corresponds to the dark line.

# (D) Evoluations of Populations

The population in the two upper levels tends to zero as time goes to infinity. If  $|\omega_{12}|$  is larger than  $\gamma_1$  and  $\gamma_2$ , the last term in Eq. (6a) or (6b) can be neglected, and consequently the population of the upper levels decays to the lower level. For  $|\omega_{12}|$  much less than  $\gamma_1$  and  $\gamma_2$ , assuming  $|\omega_{12}| << 0.5$  ( $\gamma_1 + \gamma_2$ ), it can be proven that the real parts of  $S_i$ -0.5 $\gamma_j$  (i, j, = 1, 2) are negative. That is to say, from Eqs. (6), no population is in the upper levels at time equal to infinity.

However, during the time evoluation one of the upper-level populations may increase first, even if initially there is no population in it. In Fig. 6, we plot the evolution of an initially empty upper level with  $\omega_{12}=0.2\gamma_1$ ,  $\gamma_2=\gamma_1$ . The maximum population in this level is 0.237. In some situations, the population of an initially empty level (also the other upper level) oscillates for several cycles, and then tends to zero [see Fig. 7].

# III. Driven Three-level Systems

# (A) Basic Equations

Consider a four-level atom that consists of two upper levels  $|a_1\rangle$  and  $|a_2\rangle$ , and one lower level  $|c\rangle$ . The two upper levels are coupled by the same vacuum modes to the lower level  $|c\rangle$  and are driven by a strong field with frequency  $\nu$  to another lower lying level  $|b\rangle$ ; see Fig. 8. The interaction Hamiltonian of the system composed of the atom and the vacuum modes in the interaction picture can be written as

$$V = i \sum_{k} \left[ g_{k}^{(1)} e^{i(\omega_{\alpha_{1}c} - \omega_{k})t} b_{k} \middle| a_{1} \middle\rangle \langle c \middle| + g_{k}^{(2)} e^{i(\omega_{\alpha_{2}c} - \omega_{k})t} b_{k} \middle| a_{2} \middle\rangle \langle c \middle| \right]$$

$$-i \sum_{k} \left[ g_{k}^{(1)} e^{-i(\omega_{\alpha_{1}c} - \omega_{k})t} b_{k}^{+} \middle| c \middle\rangle \langle a_{1} \middle| + g_{k}^{(2)} b_{k}^{+} e^{-i(\omega_{\alpha_{2}c} - \omega_{k})t} \middle| c \middle\rangle \langle a_{2} \middle| \right]$$

$$+i \Omega_{1} e^{-i\Delta_{1}t} \middle| a_{1} \middle\rangle \langle b \middle| + i \Omega_{2} e^{-i\Delta_{2}t} \middle| a_{2} \middle\rangle \langle b \middle| -i \Omega_{1}^{*} e^{i\Delta_{1}t} \middle| b \middle\rangle \langle a_{1} \middle| -i \Omega_{2}^{*} e^{i\Delta_{2}t} \middle| b \middle\rangle \langle a_{2} \middle|,$$

$$(16)$$

where  $\omega_{a_1c}$ ,  $\omega_{a_2c}$  are the frequency differences between level  $|a_1\rangle$ ,  $|a_2\rangle$  and  $|c\rangle$ , respectively,  $\Delta_1 = \omega_{ba_1} - v$ ,  $\Delta_2 = \omega_{ba_2} - v$ ,  $b_k(b_k^+)$  is the annihilation(creation) operator for the k-th vacuum mode with frequency  $\omega_k$ , and  $g_k^{(1,2)}$  are the coupling constants between the k-th vacuum mode and the atomic transitions from  $|a_1\rangle$ ,  $|a_2\rangle$  to  $|c\rangle$ . In Eq. (16),  $\Omega_1$  and  $\Omega_2$  are the Rabi frequencies of the driving field corresponding to the two transitions from  $|a_1\rangle$ ,  $|a_2\rangle$  to  $|b\rangle$ , respectively. Here k stands for both the momentum and polarization of the vacuum modes and  $\hbar=1$  and real  $g_k^{(1,2)}$  are assumed. This Hamiltonian describes the spontaneous emission of the atom initially in the two upper and  $|b\rangle$  levels.

The initial state vector can be written as

$$|\psi(0)\rangle = \left\{ A^{(1)}(0)|a_1\rangle + A^{(2)}(0)|a_2\rangle + B(0)|b\rangle \right\} |0\rangle. \tag{17}$$

The evolution of the state vector obeys the Schrodinger equation, and the state vector at time t can be written as

$$|\psi(t)\rangle = \left\{ A^{(1)}(t)|a_1\rangle + A^{(2)}(t)|a_2\rangle + B(t)|b\rangle \right\} |0\rangle + \sum_{k} C_k(t)b_k^+|0\rangle |c\rangle. \tag{18}$$

By using the Weisskopf-Wigner approximation, we obtain[3, 4]

$$\frac{d}{dt}A^{(1)}(t) = -\frac{\gamma_1}{2}A^{(1)}(t) - p\frac{\sqrt{\gamma_1\gamma_2}}{2}A^{(2)}(t)e^{i\omega_{12}t} + \Omega_1e^{i\Delta_1t}B(t), \tag{19a}$$

$$\frac{d}{dt}A^{(2)}(t) = -\frac{\gamma_2}{2}A^{(2)}(t) - p\frac{\sqrt{\gamma_1\gamma_2}}{2}A^{(1)}(t)e^{-i\omega_{12}t} + \Omega_2e^{i\Delta_2t}B(t), \tag{19b}$$

$$\frac{d}{dt}B(t) = -\Omega_1^* e^{-i\Delta_1 t} A^{(1)}(t) - \Omega_1^* e^{-i\Delta_2 t} A^{(2)}(t), \qquad (19c)$$

$$\frac{d}{dt}C_k(t) = -g_k^{(1)}A^{(1)}(t)e^{-i(\omega_{\alpha_1e^{-\omega_k}})t} - g_k^{(2)}A^{(2)}(t)e^{-i(\omega_{\alpha_2e^{-\omega_k}})t},$$
(20)

where  $\omega_{12}$  is the frequency difference between the two upper levels, which is much smaller than the transition frequencies, and  $p = \mu_1 \cdot \mu_2 / |\mu_1| \cdot |\mu_2|$  with  $\mu_1$  and  $\mu_2$  being the dipole moments of the two transitions. Here  $\gamma_1$  and  $\gamma_2$  are the decay rates from the two upper levels to the lower level. If the dipole moments of the two transitions are parallel, we have p = 1, while for orthogonal dipole moments we have p = 0. On solving Eq. (19), we obtain a solution for  $A^{(1)}(t)$ ,  $A^{(2)}(t)$ , or B(t), which can be written as a sum of three terms.

The spontaneous emission spectrum,  $S(\omega)$ , is the Fourier transform of  $\langle E^-(t+\tau)E^+(t)\rangle_{t=\infty}$ , and is equal to  $S(\omega_k) = \gamma_i |C_k(\infty)|^2 / 2\pi g^{(i)^2}$  ( i=1 or 2). On substituting the solution of Eqs. (19) into (20), and then integrating Eq. (20) we obtain,

$$C_{k}(\infty) = \sum_{i=1}^{3} \frac{g^{(1)}\alpha_{i}}{-\lambda_{i} + i(\Delta_{1} - 0.5\omega_{12} + \delta_{k})} + \frac{g^{(2)}\beta_{i}}{-\lambda_{i} + i(\Delta_{2} + 0.5\omega_{12} + \delta_{k})},$$
(21)

where  $\delta_k = \omega_k - .5(\omega_{a_1} + \omega_{a_2}) + \omega_c$  is the detuning of the kth vacuum mode with respect to the central frequency (from the middle point of the two upper levels to level  $|c\rangle$ ). Here  $\lambda_i$  (i = 1,2,3) are the three roots of a cubic equation.

$$\lambda^{3} - (\Gamma_{1} + \Gamma_{2})\lambda^{2} + (\Gamma_{1}\Gamma_{2} - 0.25\gamma_{1}\gamma_{2} + |\Omega_{1}|^{2} + |\Omega_{2}|^{2})\lambda$$

$$- 0.5(\gamma_{1}|\Omega_{2}|^{2} + \gamma_{2}|\Omega_{1}|^{2} - p\sqrt{\gamma_{1}\gamma_{2}}(\Omega_{1}^{\bullet}\Omega_{2} + \Omega_{1}\Omega_{2}^{\bullet})) + i(\Delta_{2}|\Omega_{1}|^{2} + \Delta_{1}|\Omega_{2}|^{2}) = 0,$$
(22)

where  $\Gamma_{1,2} = 0.5\gamma_{1,2} + i\Delta_{1,2}$ . The spontaneous emission spectrum can be obtained by taking the absolute square of Eq. (21). For p = 1, quantum interference can cancel spontaneous emission, but for p = 0, there is no cancellation. This interference results in some very interesting features, e.g., spectral peak elimination and the cancellation of spontaneous emission.

#### (B) Dark Lines and Spectral Narrowing

The spontaneous emission spectra for the two cases (p=1 and 0) are quite different due to interference. It is well known that the spontaneous emission spectrum for p=0 is a three peak distribution. For p=1, we have interference, which can lead to the elimination of one of the three peaks. In Fig. 1b, we plot spectra for p=1 and p=0 with the atom initially being in  $|a_1\rangle$ . We can see the disappearance of the central peak as a result of interference. The elimination of the central peak can also be observed for the atom being initially in a superposition state. In Fig. 9, we show the elimination of central peak for the atom initially in the state  $(|a_1\rangle - |a_2\rangle)/\sqrt{2}$ . It can be proven analytically that the central peak is eliminated if

$$\Delta_2 = -\chi^2 \Delta_1,\tag{23}$$

where  $\chi = |\Omega_2/\Omega_1| = g^{(2)}/g^{(1)}$  is the ratio of dipole moments between the two upper levels and level  $|b\rangle$ , which is also equal to the ratio of the dipole moments between the two upper levels and level  $|c\rangle$ . The elimination of the central peak indicates the cancellation of the spontaneous emission into those modes with their frequencies near the central peak (in the neighborhood of the driving field frequency) and is the resulted of a destructive interference. As opposed to destructive interference, a constructive interference can also be found as shown in Fig. 10, where the central peak increases while the other two peaks decrease.

## (C) Spontaneous Emission Cancellation and Populations Trapping in the Upper Levels

The area under the spectral curve is proportional to the energy emitted by the atom to the vacuum modes. For p=0, the area is always equal to unity (energy conservation), that is to say, the atom finally will be in the lower level  $|c\rangle$  and no population in the upper levels. For p=1, we find the area may be less than unity (see Fig. 8 and 9) if condition (8) is satisfied, which means that some population is still in the upper levels at  $t=\infty$ . That is to say, in the steady state the spontaneous emission is canceled due to interference. In Fig. 11, we plot the evolution of the population in level  $|a_1\rangle$  for p=1 and p=0. It is clear that the population goes to zero for p=0, while it tends to a steady value 0.51 for p=1. The cancellation of the spontaneous emission in the steady state is another phenomenon of the interference.

The populations trapped in levels 
$$|\alpha_1\rangle$$
 and  $|\alpha_2\rangle$ ,  $|\alpha_3|^2$  and  $|\beta_3|^2$ , are determined by  $\alpha_1 + \alpha_2 + \alpha_3 = A_1(0)$ ,  $\beta_1 + \beta_2 + \beta_3 = A_2(0)$ ,  $b_1 + b_2 + b_3 = B(0)$ ,  $(i = 1,2,3)$ , (24)  $\lambda_i - \Omega_1^* \alpha_i - \Omega_2^* \beta_i = 0$ ,  $\Omega_1 b_i - (\Gamma_1 - \lambda_i) \alpha_i - 0.5 \sqrt{\gamma_1 \gamma_2} \beta_i = 0$ ,

where the condition Eq. (23) and p=1 have been used. For arbitrary  $\chi$  the expressions for  $|\alpha_3|^2$  and  $|\beta_3|^2$  are complicated. For  $\chi=1$  and the atom initially being in  $|a_1\rangle$ , we can find the populations trapped in  $|a_1\rangle$  (or  $|a_2\rangle$ ) and  $|b\rangle$  are  $\Omega^4/(\Delta^2+2\Omega^2)^2$ , and  $\Omega^2\Delta^2/(\Delta^2+2\Omega^2)^2$  ( $\Omega=\Omega_1$  and  $\Delta=\Delta_1$ ), respectively. In Fig. 11 we see more than 50% of population is trapped in  $|a_1\rangle$  (also 13% in  $|a_2\rangle$ ). The population trapping in upper levels depends on the driving field and the detunings. Without the driving field, there will be no trapping [5] if the two upper levels are not degenerate. How much population can be trapped in the upper levels depends on the separation between the two upper levels, the ratio of the two decay rates, and the Rabi frequency. In Fig. 12 we plot the population in the level  $|a_1\rangle$  versus the Rabi frequency

for three cases with  $\chi = 2$  and Eq. (23) being satisfied. It can been seen that 30% of the population will be trapped in level  $|a_1\rangle$  if the Rabi frequency is  $10\gamma_1$  for a large separation ( $\omega_{12} = 40\gamma_1$ ). In order to trap more population we need high Rabi frequency.

The spectral peak elimination and cancellation of spontaneous emission in the steady state can be understood in the dressed state picture. On diagonalizing the Hamiltonian for  $|a_1\rangle$  and  $|a_2\rangle$ ,  $|b\rangle$  and the driving field, we get three dressed states. The decay from  $|a_1\rangle$  and  $|a_2\rangle$  to  $|c\rangle$  becomes the decay from three dressed states to  $|c\rangle$ . The decay rates for the three dressed states depend on the interference (terms of the type  $p\sqrt{\gamma_1\gamma_2}/2$ ). Under the condition  $\Delta_2=-\chi^2\Delta_1$ , the decay rate of one dressed state (with intermediate energy) is proportional to  $\frac{\gamma_1^2}{\Delta_1}+\frac{\gamma_2^2}{\Delta_2}+\frac{2p\gamma_1\gamma_2}{\Delta_1\Delta_2}$ . For p=1, this decay rate is proportional to  $(\Delta_2+\chi^2\Delta_1)^2=0$ . The interference results in a zero decay rate. The population in this dressed state will not decay to lower level  $|c\rangle$ , and consequently we have the central peak elimination and spontaneous emission cancellation in the steady state.

In order to experimentally observe the elimination of the spectral line, we need two closely separated levels with parallel dipole moments. Mixing two different parity levels by a static electric field or other means can produce such two closely separated upper levels with parallel dipole moments.

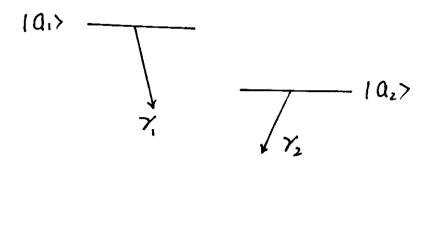
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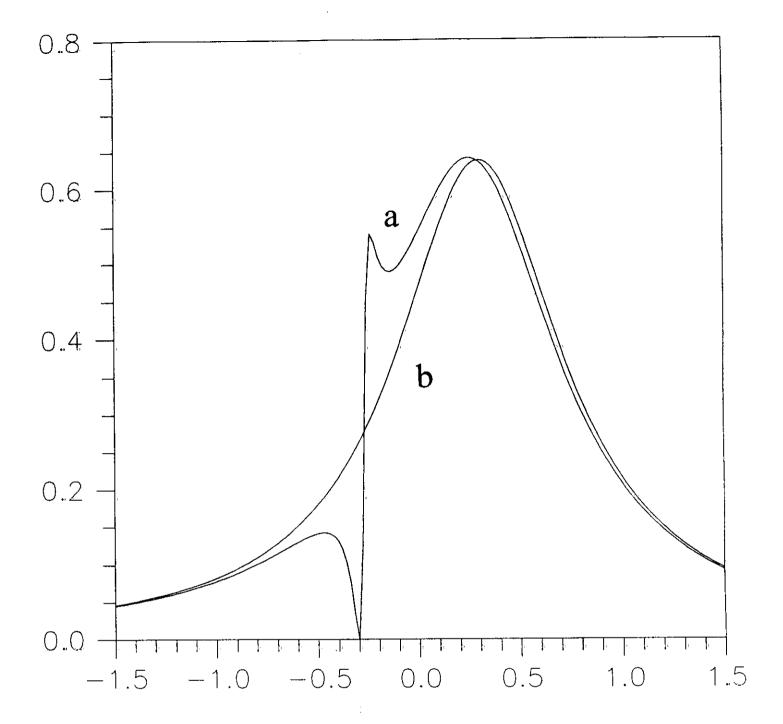
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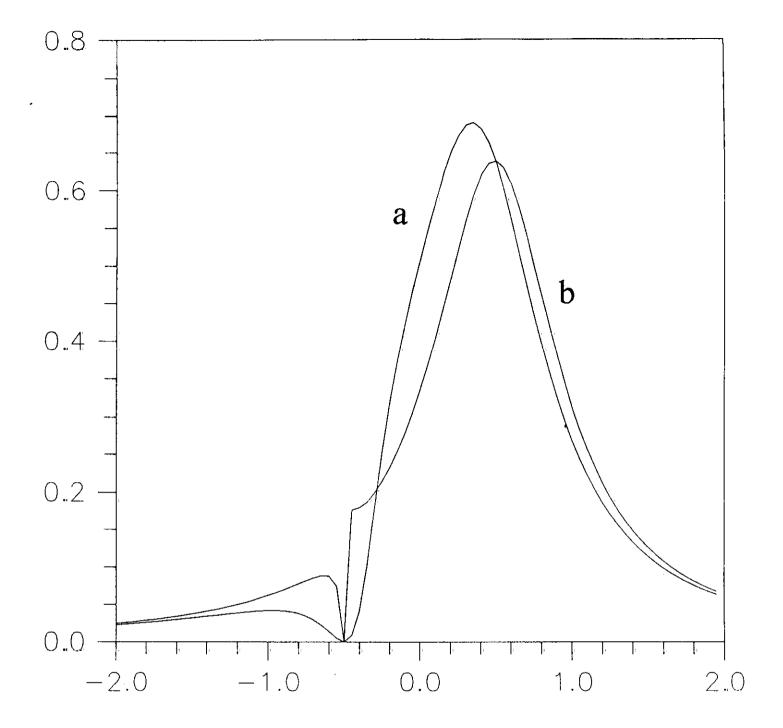
- 1. The three-level atom.
- 2. Spontaneous emission spectrum for (a) the three-level atom with  $\omega_{12} = 0.6\gamma_1$  and  $\gamma_2 = 0.1\gamma_1$ , and (b) a two-level atom.
- 3. The dark lines in the spontaneous emission spectrum of the three-level atom with  $\omega_{12} = \gamma_1$ , and (a)  $\gamma_2 = 0.5\gamma_1$ , and (b)  $\gamma_2 = 0.05\gamma_1$ .
- 4. Spectral narrowing by increasing the ratio  $\gamma_2/\gamma_1 = (a)$ . 0.05, (b). 1, and (c). 5, with  $\Delta = \gamma_1$ .
- 5. Spectral narrowing, (a) the spectrum of the three-level atom with  $\omega_{12} = \gamma_1$  and  $\gamma_2 = 2\gamma_1$ , and (b) the spectrum of a two level atom with a decay rate  $\gamma_1$ .
- 6. The temporary population in  $|a_2\rangle$  reaches a maximum of .237 ( $\omega_{12} = 0.2\gamma_1$ ,  $\gamma_2 = \gamma_1$ ).
- 7. The oscillation of the population in  $|a_2\rangle$  ( $\omega_{12} = 5\gamma_1$ ,  $\gamma_2 = \gamma_1$ ).
- 8. The spontaneous emission spectra for  $\omega_{12} = 2\gamma_1$ ,  $\Omega_1 = \gamma_1$ ,  $\gamma_2 = \gamma_1$ , and  $\Delta_1 = \gamma_1$ , (a) p = 1, and (b) p = 0. The atom is initially in level  $|a_1\rangle$ . Inset shows uppers levels  $|a_1\rangle$  and  $|a_2\rangle$  coupled to level  $|b\rangle$  with Rabi frequency  $\Omega$  while decaying to level  $|c\rangle$ .
- 9. The spontaneous spectra for  $\Delta_1 = 4\gamma_1$ ,  $\omega_{12} = 5\gamma_1$ ,  $\Omega_1 = \gamma_1$ , and  $\gamma_2 = 0.25\gamma_1$ , (a) p = 1, and (b) p = 0. The atom is initially in  $(|a_1\rangle |a_2\rangle)/\sqrt{2}$ .
- 10. The spectra with a constructive interference for  $\omega_{12}=2\gamma_1$ ,  $\Delta_1=\gamma_1$ ,  $\Omega_1=\gamma_1$ , and  $\gamma_2=4\gamma_1$ , (a) p=1, and (b) p=0. The atom is initially in  $(|a_1\rangle-3|b\rangle)/\sqrt{10}$ .
- 11. Time evolution of populations in  $|a_1\rangle$ , for  $\omega_{12}=4\gamma_1$ ,  $\gamma_2=4\gamma_1$ ,  $\Delta_1=0.8\gamma_1$ , and  $\Omega_1=2\gamma_1$ , (a) p=1, and (b) p=0. The atom is initially in  $|a_1\rangle$ .
- 12. The trapped population in level  $|a_1\rangle$  versus the Rabi frequency (in unit of  $\gamma_1$ )  $\omega_{12}$  = (a)  $40\gamma_1$ , (b)  $20\gamma_1$ , and (c)  $4\gamma_1$ .



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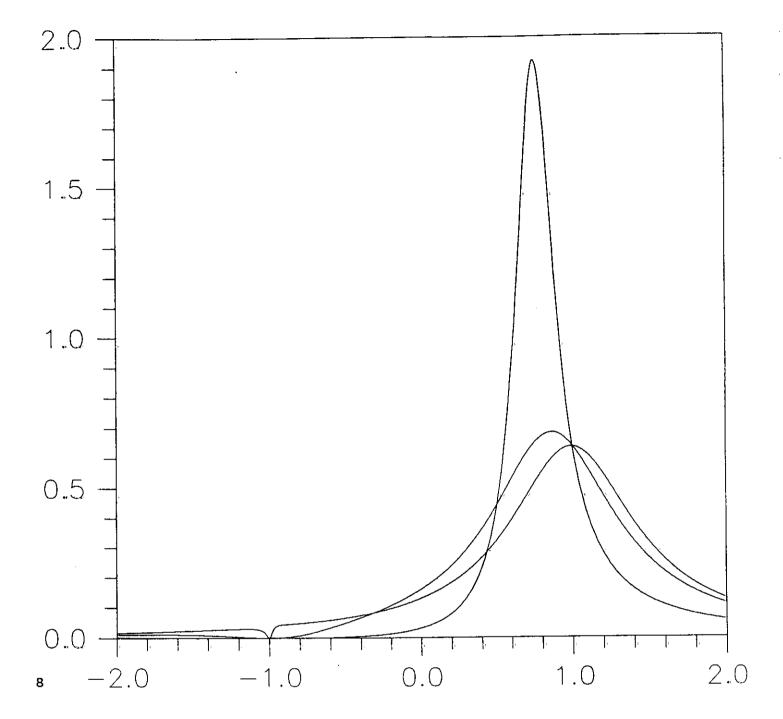
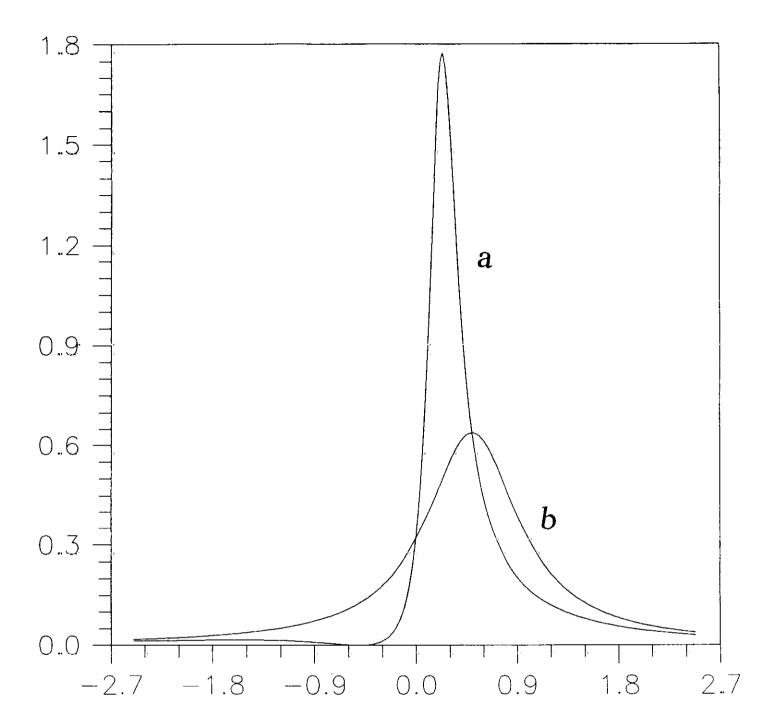
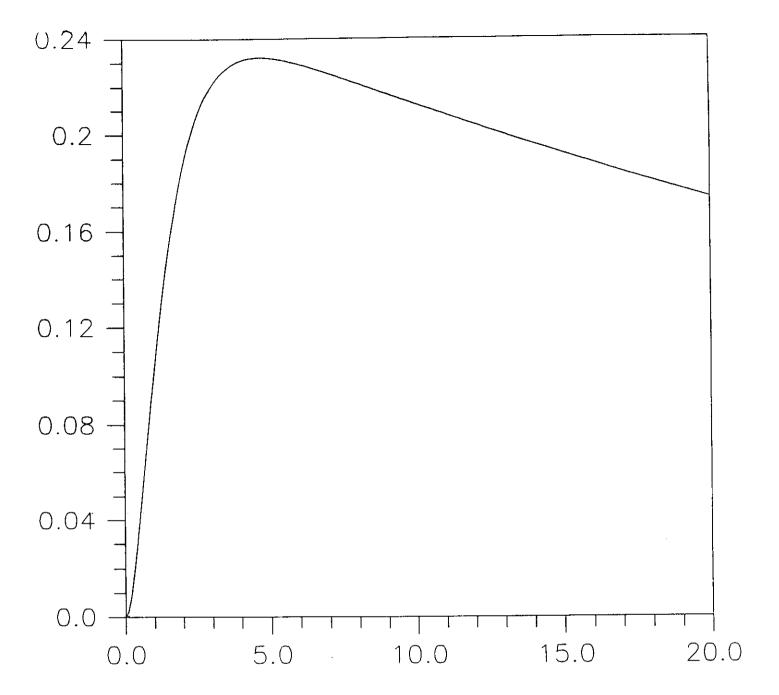
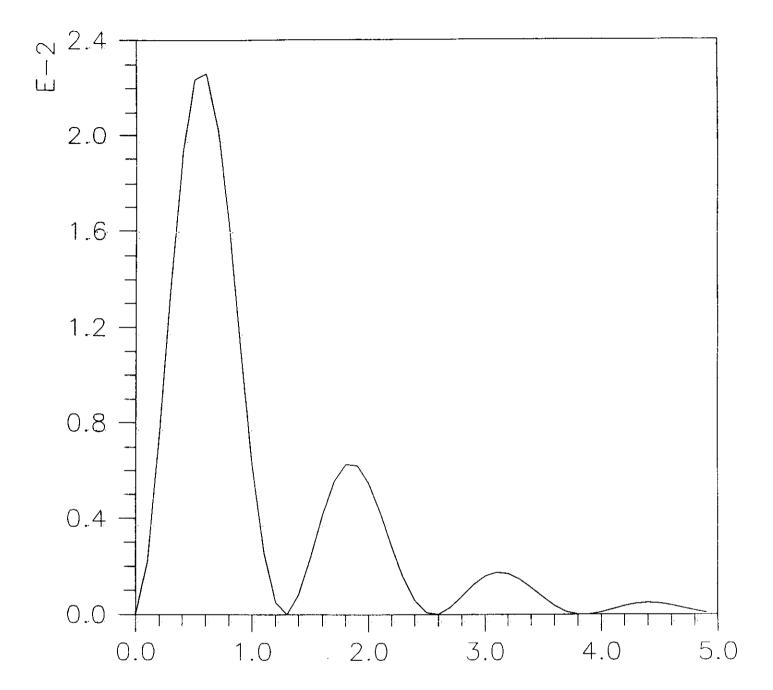


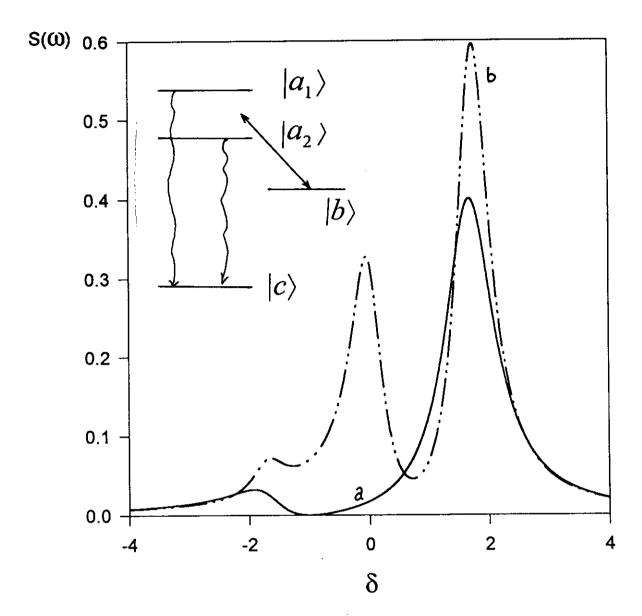
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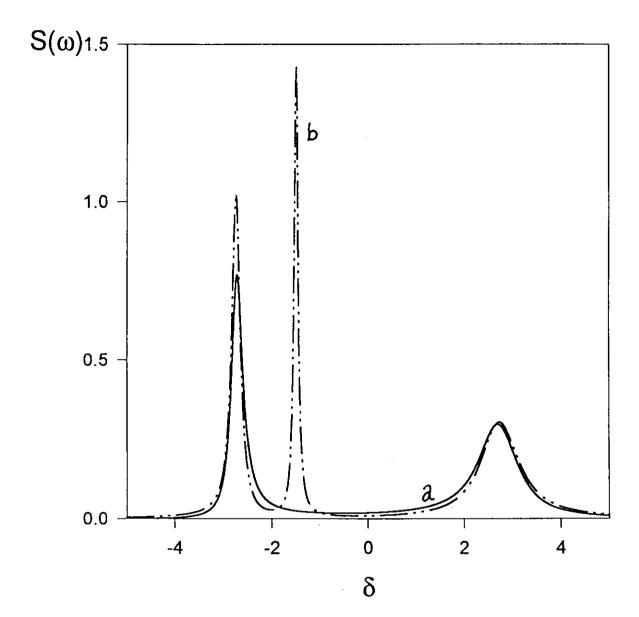




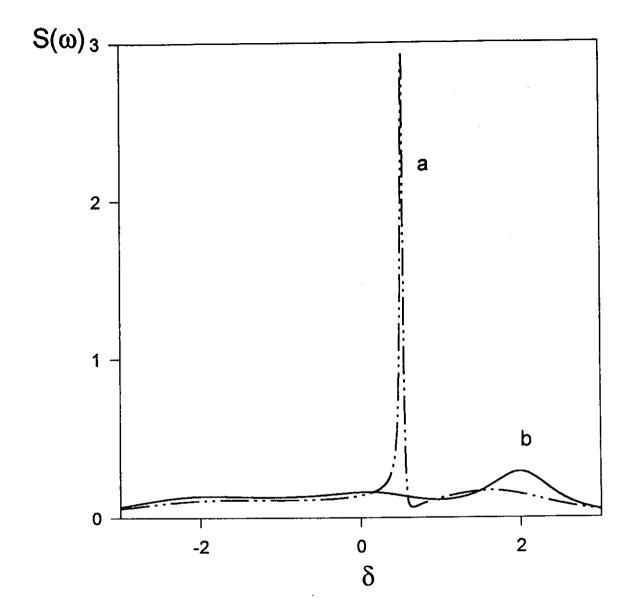
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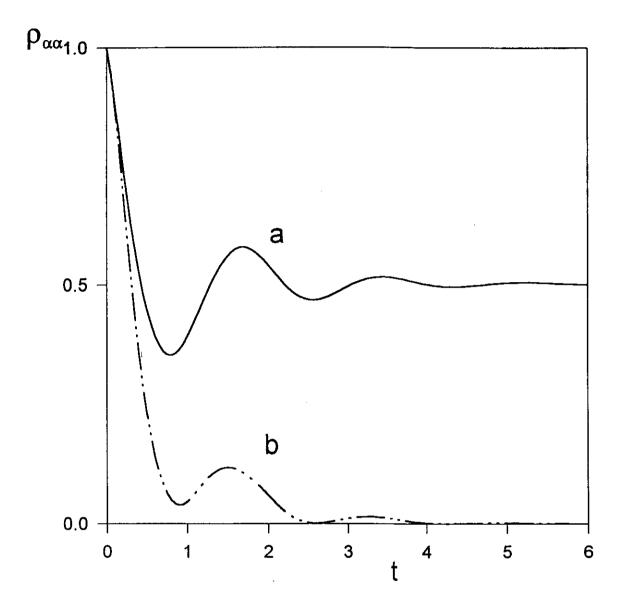






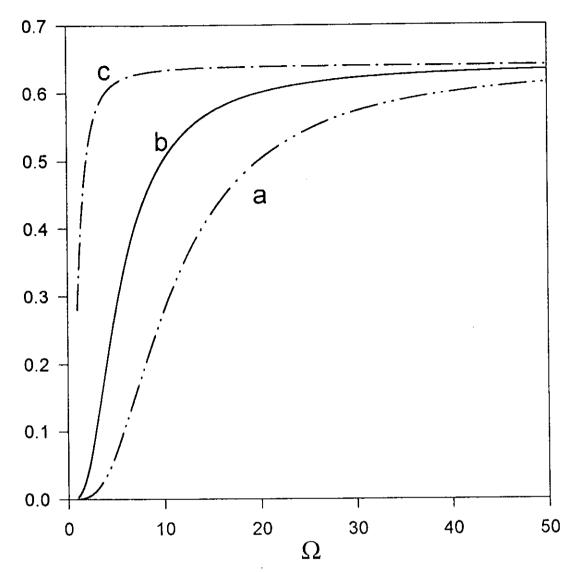
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Quantum Interference Effects in Spontaneous Emission from an Atom Embedded in a Photonic band gap structure

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Abstract

The spontaneous emission from a three-level atom embedded in a photonic band gap structure is studied. Interference between two transitions leads to qausi-periodical oscillations of population between the two upper levels with large amplitudes. The spontaneous emission of the atom is characterized by three components in the radiated field; a localized part, a travelling pulse, and a  $1/\sqrt{t}$  decaying part. An analytical expression for the localization distance for the localized field is obtained. The phase velocity for the travelling pulse is larger than vacuum speed of light, and its energy velocity could be close to zero. By selecting a certain initial superposition state, a large amount of population trapping can be achieved.

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Quantum interference between different atomic transitions and atomic coherence can lead to various effects such as change of spectra, population trapping, phase-sensitive amplification, and laser without inversion[1-3]. In the past, these phenomena were studied mostly in systems with coherent driving field. In a four-level system the spontaneous emission from two neighbouring levels (which emit photons of the same polarization) to the lower level can be totally supressed when they are coupled to the fourth level by a driving field [2]. In the case, population trapping and oscillation of population in upper levels can be observed. In the absence of external coherent field, oscillation of population in the upper levels also exists due to the emission and re-absorption of a single photon from the two upper levels as the result of the interference [3]. However, such external-field-free oscillation is very weak as the spontaneously emitted photon travels away from the atom in free space with the vacuum speed of light c.

It was shown that, in a photonic band gap structure (PBGS), the prohibition of light wave transmission can be achieved for some frequency range in all directions [4]. As a consequence the energy of field can be localized in a space domain without propagating away. Recently a lot of effort has been concentrated in the study of photonic crystals in which a three-dimensional periodic dielectric structure is used to create one or several forbidden frequency bands [5].

An atom (impurity) embedded in such a structure will interact with field modes in the propagating frequency band as well as those in the forbidden band, localized field modes created by the atom [6,7]. Since emitted photon can be trapped in the vicinity of the atom, the exchange of energy between the atom and field can be significant. It has been shown that a two-level atom embedded in a PBGS could retain some population in the upper level, even when the transition frequency was in the transmitting band [6.8]. The final state is a dressed state of the atom with a localized field mode, which lies in the forbidden band. A natural question arises: how to use the localized field to enhance

quantum interference effect. Furthermore, one might ask what the properties of the field emitted by the atom are.

In this letter we report the effect of quantum interference without coherent driving field in the spontaneous emission of a three-level atom embedded in a photonic band structure. The two upper levels are coupled to the lower one via the same field continuum. The intereference between the two transitions leads to an exchange of population between the two upper levels, extended oscillations of energy distribution between the field and the atom, and a large population trapping in the upper levels. The field emitted by the atom is composed of three parts; a localized field, a travelling wave with very slow energy propagation velocity, and a decaying field.

Our model consists of a three-level atom with two upper levels  $|a_1>$ ,  $|a_2>$  and a lower level |b>. The dipole vectors between  $|a_1>$  and |b> and between  $|a_2>$  and |b> are parallel. The dispersion relationship of the band gap material near the band gap edge  $\omega_c$  can be approximated by [6,8]

$$\omega_k = \omega_c + A(k - k_0)^2$$
.  $A = \omega_c/k_0^2$ . (1)

The Hamiltonian for the system, after carrying out the rotating wave approximation at  $\omega_c$ , is

$$H = \sum_{k} \hbar(\omega_{k} - \omega_{c}) a_{k}^{\dagger} a_{k} + i \hbar \left[ \sum_{k} (g_{k}^{(1)} a_{k}^{\dagger} | b > < a_{1}| + g_{k}^{(2)} a_{k}^{\dagger} | b > < a_{2}|) - H.c. \right]. \tag{2}$$

The coefficients  $g_k^{(1)}, g_k^{(2)}$  are related to the decay coefficients of each upper level. We assume here as in practical situations  $|\omega_{a_1} - \omega_{a_2}| << \omega_c$ . For a special case  $\omega_{a_1b} - \omega_c = -(\omega_{a_2b} - \omega_c) = \Delta$  with  $g_k^{(1)} = g_k^{(2)}$ , analytic results can be obtained for the evolution of an atom from an arbitrary initial excited state. The state vector at a time t is given by

$$|\psi(t)>=[A^{(1)}(t)|a_1>+A^{(2)}(t)|a_2>]_a|0>_f+\sum_k B_k(t)|b>_a|1_k>_f. \tag{3}$$

with  $A^{(1)}(0)$ ,  $A^{(2)}(0) \neq 0$ , and  $B_k(0) = 0$ .

Following procedures similar to Ref. [8] we obtain the Laplace transform for the amplitudes  $A^{(1)}(t)$ ,  $A^{(2)}(t)$ :

$$\widetilde{A}^{(1)}(s) = \frac{A^{(1)}(0)(s - i\Delta) - [A^{(1)}(0) - A^{(2)}(0)](i\gamma)^{3/2}\sqrt{s}}{s^2 + \Delta^2 - 2(i\gamma)^{3/2}\sqrt{s}},$$

$$\widetilde{A}^{(2)}(s) = \frac{A^{(2)}(0)(s + i\Delta) - [A^{(2)}(0) - A^{(1)}(0)](i\gamma)^{3/2}\sqrt{s}}{s^2 + \Delta^2 - 2(i\gamma)^{3/2}\sqrt{s}}.$$
(4)

Here  $\gamma = \omega_{ab}^{7/2} d_{ab}^2/(6\pi\epsilon_0 \hbar c^3)$ . The RHS contains four poles,  $s_i = x_i^2, i = 1, 2, 3, 4$ , where  $x_i$  are the roots of the equation  $x^4 + \Delta^2 - 2(i\gamma)^{3/2}x = 0$  and are located in the quadratures II, IV, III, and I respectively. The inverse of Eq. (4) can be expressed as

$$A^{(1,2)}(t) = \sum_{i=1}^{4} a_i^{(1,2)} \left[ (x_i + y_i) e^{x_i^2 t} + y_i \left[ 1 - erf(\sqrt{x_i^2 t}) \right] e^{x_i^2 t} \right]. \tag{5}$$

where  $a_i^{(1,2)}$  is the expansion coefficient corresponding to the pole  $x_i$ , which depends on both  $\Delta$  and  $A^{(1,2)}(0)$ . Here  $y_i = \sqrt{x_i^2}$  is on the right half plane, thus only two purely exponential terms due to  $x_4$  (oscillatory without decay at a fixed space point) and  $x_2$  (oscillatory with decay at a fixed space point) survive. The second half containing  $erf(\sqrt{x_i^2 t})$  decays usually as  $1/\sqrt{t}$ , while similar terms in Ref. [8] decays at a faster pace as  $(1/\sqrt{t})^3$ .

The radiated field amplitude at a particular space point is [9]

$$E(r,t) = \sum_{k} \sqrt{\frac{\hbar \omega_{k}}{2\epsilon_{0} V}} e^{-i(\omega_{k} t - k \cdot r)} B_{k}(t), \tag{6}$$

where the one-photon state amplitude

$$B_k(t) = -\int_0^t dt' \left[ g_k^{(1)} A^{(1)}(t') + g_k^{(2)} A^{(2)}(t') \right]. \tag{7}$$

We find the right hand side of Eq. (6) can be expressed as the sum of three contributions  $I_1, I_2$ , and  $I_3$  for large times  $(t \to \infty)$ . The first part,  $I_1$ , comes from the  $x_4$  term in Eq. (5) and does not decay in time. Its amplitude drops exponentially as  $e^{-r/l_c}$ , and its frequency  $(\omega_c - |x_4|^2)$  is within the forbidden band. It represents a localized field. The size of the localized photon mode is  $l_c = \sqrt{A/|x_4|^2}$ . The maximum amplitude of the localized photon

mode is proportional to  $(1-i\sqrt{\omega_c/|x_4|^2})$ . The  $I_2$  part comes from the  $x_2$  term. Spatially it is an exponential pulse with the phase velocity  $v_p$  and energy velocity  $v_e$ .

$$v_{p} = \left[1 - \text{Re}(\sqrt{ix_{2}^{2}/\omega_{c}})\right] k_{0}/\alpha,$$

$$v_{e} = \text{Im}(\sqrt{ix_{2}^{2}/\omega_{c}}) k_{0}/\alpha.$$
(8)

The phase and amplitude propagation is proportional to  $e^{-i\alpha(t-r/v_p)}e^{-\beta(t-r/v_e)}$ . Its frequency  $\alpha = \omega_c - \text{Im}(x_2^2)$  is in the transmitting band. The phase velocity  $v_p$  is greater than the vacuum speed of light c, while  $v_e$  is considerably smaller than c (could be close to zero).

For the third part,  $I_3$ , the exact result can not be found. An approximate expression was obtained for large  $\gamma t$ . The  $I_3$  term decays to zero, but unlike  $I_2$ , it does not have the form of an exponential of  $e^{\eta(t-r/v)}$  representing a wave-packet travelling away from the atom. It only contains a phase propagating factor  $e^{-i(\omega_c t - k_0 r)}$ . At any fixed time, the amplitude of the third part decreases to zero exponentially as distance from the atom increases. At any space point, the amplitude decays to zero as time goes to infinity. Therefore, it represents a decaying field. The amplitude decay is usually very slow, proportional to  $\frac{1}{\sqrt{t}}$  (see Fig. 2) due to the interference between the two transitions from the two upper levels to the lower level.

We examined the evolution behaviors and the final state with various initial superposition states to analyze the roles played by the coupling of the decaying field to the travelling wave and the localized wave, and by the interference due to the two interaction channels. A picture of how the population transfers between levels, and how the energy is transferred from the atom to the travelling wave and localized field is thus obtained.

Interference leads to the transfer of population from  $|a_1\rangle$  to  $|a_2\rangle$  or vice-versa, as witnessed by the oscillations in Fig. 2, (initially the atom in  $|a_2\rangle$ ). The population trapping in the two upper levels is due to the photonic band gap with a non-decaying component to form the final dressed state. In Fig. 2, the dominating part  $(I_3)$  decays

at a rate  $(\sqrt{\gamma t})^{-1}$  due to the interference. The populations oscillate many cycles ( $\sim 10^2$ ) before eventually decaying to their final values (for other initial states, the oscillations are similar.)

This quasi-oscillation has a quite large amplitude of the order of 0.5, a feature significantly different from the two-level case. In the current situation, the interference between the two transitions is enhanced by the localized field. Consequently, we have larger oscillation amplitude compared to either a two-level atom in a PBGS or a three-level atom in vacuum. From the pattern of decay we determined that the strong quasi-oscillation is mainly due to the  $I_3$  part. The energy in the decaying localized field (given by the atom initially) will transfer back to the atom, and then become the energy of the travelling wave and the localized field. This explains the small value of  $v_e$ .

It can be proven analytically that the amplitude of the oscillations can be minimized by choosing a special initial state  $A^{(1)}(0) = A^{(2)}(0)$  (minimizing the interference), and with this initial state the amplitude of the third part  $(I_3)$  decays as  $(1/\sqrt{t})^3$ , as shown in Fig. 3. (Note decay as  $(1/\sqrt{t})^3$  is the situation for a two-level atom.)

The amount of population trapped in the upper levels depends on the initial condition. The population in the upper level within the band gap ( $|a_2\rangle$ ) could be transferred to the upper level in the transmitting band ( $|a_1\rangle$ ) from which it could emit the travelling wave. The final state contains an upper level part (trapped excited state population) and a lower level part with one photon in the localized mode. The phase difference between the two upper levels at a final state is always zero. The dependence of the ratio of the populations in the two upper levels,  $A^{(1)}(\infty)/A^{(2)}(\infty)$ , on the initial state is weak. This ratio, as well as the portion in the lower level, depends on  $\Delta/\gamma$ .

Since the true trapped final state is a dressed state with a lower level component, no superposition of the two upper levels can evade decay completely. This is in contrast with the dark state of a driven four-level system. However, if the atom is prepared in a state, which is a renormalized state of the final state projected onto the manifold of the two

upper levels starting from any initial state (i.e. by making  $B_k(t = \infty) = 0$  in Eq. (3) and renormalizing the resulting state), we can minimize the energy emitted into the travelling wave, and could have more population trapped in the upper levels (even more than the case that the atom is initially in the level in the gap, i.e.,  $|a_2\rangle$ ). This final state population trapped in the two upper level is plotted in Fig. 4 (where for comparison we also plotted the final population trapped in the two upper levels starting from the atom initially in the upper level in the band gap). We have established that the trapped population for each value of  $\Delta$  is higher than that from any other initial states (see Fig. 4). This is similar to the situation of a driven four-level system. If the four-level system is prepared in a non-decaying dressed state, the four-level atom will never decay (no travelling wave). In the current case, the atom gives energy mainly to the localized field to form the required component of lower level with one photon.

In conclusion, we found in a photonic band gap structure, the interference of spontaneous emission from a three-level atom with the two upper levels coupled to the same continium can be significantly enhanced without the help of a driving field, which is essential in free space. The re-absorption and re-emission of photons in the three-level system embedded in PBGS are more pronounced. The atom releases its energy during the spontaneous emission process in three forms, a localized field with a localization distance  $l_c$ , a travelling wave (an exponential pulse with a phase velocity larger than c and very lower energy velocity), and a slowly decaying field. Since there is one level in the forbidden band and one level in the transmitting band, it could also serve as a medium to study the coupling of external field and the local trapped field.

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# Figure Captions

- A three-level atom in a photonic band gap structure. The two upper levels (|a<sub>1</sub>| · · and |a<sub>2</sub> >) are symmetrically placed from the band gap edge by Δ.
- 2. Upper state population evolution for the initial state  $|\psi(0)\rangle = \frac{1}{\sqrt{2}}(|a_1\rangle + |a_2\rangle)$ .  $\Delta = \gamma = 1$ .  $p_1, p_2$  are the populations in levels  $|a_1\rangle$  and  $|a_2\rangle$  respectively.
- 3. Upper state population evolution for the initial state  $|\psi(0)\rangle = \frac{1}{\sqrt{2}}(|a_1\rangle |a_2\rangle)$ .  $\Delta = \gamma = 1$ . Notice the significantly different decay time scale compared with Fig. 2.
- 4. Trapped population in the two upper levels as a function of  $\Delta$  for the initial state which is a renormalized state of the final state projected onto the manifold of the two upper levels and started from any initial state (solid line). For comparison the trapped population for the initial state  $|\psi(0)| = |a_2|$  is also plotted (dashed line).

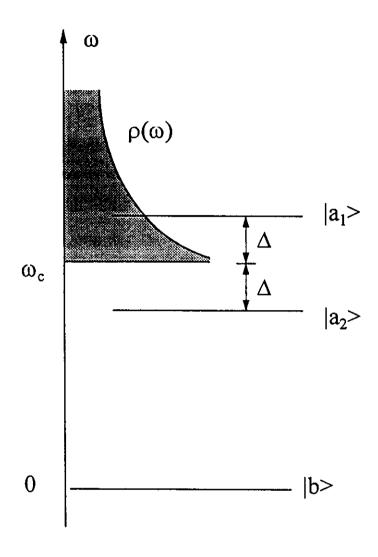


Figure 1

