Spectral Line Elimination and Spontaneous Emission Cancellation via Quantum Interference

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Quantum interference in spontaneous emission from a four-level atom is investigated. The atom has two upper levels coupled by the same vacuum modes to a common lower level and is driven by a coherent field to an auxiliary level. Interference can lead to the elimination of a spectral line in the spontaneous emission spectrum and spontaneous emission cancellation in steady state.

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Quantum interference in multilevel atomic systems can lead to many novel and unexpected effects, for example, absorption reduction and cancellation (or lasing without inversion) [1–3] and spontaneous emission reduction and cancellation (or population inversion without emission) [2]. In recent years, substantial attention has been paid to the effects associated with the absorption cancellation. In this work we will focus on another effect associated with the quantum interference effect, namely, spontaneous emission cancellation. A related problem, the suppression of autoionization in a three-level system when the two upper levels are degenerate, was pointed out by Harris [1]. In Ref. [2], we considered the interaction between a single mode (the signal field) and three-level atoms, and predicted the cancellation of spontaneous emission from the two nondegenerate upper levels into this mode. However, in that paper, the interaction between the vacuum modes and the atomic transition from the upper levels to the lower level was neglected, although the decay from the three levels to other levels was included. Thus the spontaneous emission into all the vacuum modes near the transition frequencies could not be considered. A question of interest is the following: Can the spontaneous emission into certain (not only one) or even all vacuum modes be canceled? We find that the answer is yes. In order to get the answer, the interaction between the vacuum modes and the transition between the three levels must be taken into account. On the other hand, the decay from the two upper levels to other levels can be neglected because the spectrum due to this decay is far away from the spectrum of the transition between the three levels. That is to say, we need to consider a closed system instead of an open system.

In this paper, we investigate the effect of quantum interference between the decay processes from the nondegenerate two upper levels of a four-level atom to the same lower level. This interference can lead to the elimination of a spectral line in the spontaneous emission spectrum and the cancellation of spontaneous emission in the steady state.

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 ν to another upper lying level $|b\rangle$; see Fig. 1. The interaction Hamiltonian of the system composed of the atom and the vacuum modes in the interaction picture can be written as

$$\begin{aligned} V = {} & i\sum_k [g_k^{(1)} e^{i(\omega_{a_1c}-\omega_k)t} b_k |a_1\rangle\langle c| + g_k^{(2)} e^{i(\omega_{a_2c}-\omega_k)t} b_k |a_2\rangle\langle c|] \\ & - i\sum_k [g_k^{(1)} e^{-i(\omega_{a_1c}-\omega_k)t} b_k^\dagger |c\rangle\langle a_1| + g_k^{(2)} b_k^\dagger e^{-i(\omega_{a_2c}-\omega_k)t} |c\rangle\langle a_2|] \\ & + i\Omega_1 e^{-i\Delta_1 t}|a_1\rangle\langle b| + i\Omega_2 e^{-i\Delta_2 t}|a_2\rangle\langle b| - i\Omega_1^* e^{i\Delta_1 t}|b\rangle\langle a_1| - i\Omega_2^* e^{i\Delta_2 t}|b\rangle\langle a_2|, \end{aligned} \tag{1}$$

where ω_{a_1c} and ω_{a_2c} are the frequency differences between levels $|a_1\rangle$ and $|a_2\rangle$ and $|c\rangle$, respectively, $\Delta_1 = \omega_{ba_1} - \nu$, $\Delta_2 = \omega_{ba_2} - \nu$, b_k ($b_k^\dagger$) is the annihilation (creation) operator for the kth vacuum mode with frequency ω_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 $|c\rangle$. In Eq. (1), Ω_1 and Ω_2 are the Rabi frequencies of the driving field corresponding to the two transitions from $|a_1\rangle$ and $|a_2\rangle$ 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

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

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

$$|\psi(t)\rangle = \{A^{(1)}(t)|a_1\rangle + A^{(2)}(t)|a_2\rangle + B(t)|b\rangle\}|0\rangle + \sum_k C_k(t)b_k^\dagger|0\rangle|c\rangle. \quad (3)$$

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

$$\frac{d}{dt}A^{(1)}(t) = -\frac{\gamma_1}{2}A^{(1)}(t) - p\frac{\sqrt{\gamma_1\gamma_2}}{2}A^{(2)}(t)e^{i\omega_{12}t} + \Omega_1 e^{i\Delta_1 t}B(t), \quad (4a)$$

$$\frac{d}{dt}A^{(2)}(t) = -\frac{\gamma_2}{2}A^{(2)}(t) - p\frac{\sqrt{\gamma_1\gamma_2}}{2}A^{(1)}(t)e^{-i\omega_{12}t} + \Omega_2 e^{i\Delta_2 t}B(t), \quad (4b)$$

$$\frac{d}{dt}B(t) = -\Omega_1^* e^{-i\Delta_1 t}A^{(1)}(t) - \Omega_2^* e^{-i\Delta_2 t}A^{(2)}(t), \quad (4c)$$

$$\frac{d}{dt}C_k(t) = -g_k^{(1)}A^{(1)}(t)e^{-i(\omega_{a_1c}-\omega_k)t} - g_k^{(2)}A^{(2)}(t)e^{-i(\omega_{a_2c}-\omega_k)t}, \quad (5)$$

where ω_{12} is the frequency difference between the two upper levels, which is much smaller than the transition frequencies, and $p = \vec{\mu}_1 \cdot \vec{\mu}_2/|\vec{\mu}_1| \cdot |\vec{\mu}_2|$ with $\vec{\mu}_1$ and $\vec{\mu}_2$ being the dipole moments of the two transitions. Here γ_1 and γ_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. (4), 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. (4) into (5), and then integrating Eq. (5) we obtain,

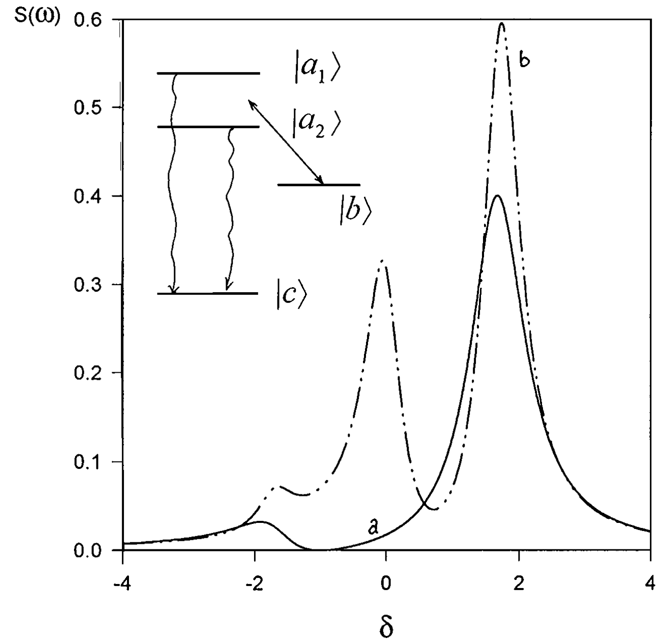

FIG. 1. The spontaneous emission spectra for $\omega_{12} = 2\gamma_1$, $\Omega_1 = \gamma_1$, $\gamma_2 = \gamma_1$, and $\Delta_1 = \gamma_1$, (a) $p = 1$, and (b) $p = 0$. The atom is initially in level $|a_1\rangle$. Inset shows upper levels $|a_1\rangle$ and $|a_2\rangle$ coupled to level $|b\rangle$ with Rabi frequency Ω while decaying to level $|c\rangle$.

$$C_k(\infty) = \sum_{i=1}^{3}\frac{g^{(1)}\alpha_i}{-\lambda_i + i(\Delta_1 - 0.5\omega_{12} + \delta_k)} + \frac{g^{(2)}\beta_i}{-\lambda_i + i(\Delta_2 - 0.5\omega_{12} + \delta_k)}, \quad (6)$$

where $\delta_k = \omega_k - 0.5(\omega_{a_1} + \omega_{a_2}) + \omega_c$ 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 λ_i ($i = 1, 2, 3$) are the three roots of a cubic equation.

$$\lambda^3 + (\Gamma_1 + \Gamma_2)\lambda^2 + (\Gamma_1\Gamma_2 - 0.25\gamma_1\gamma_2 + |\Omega_1|^2 + |\Omega_2|^2)\lambda - 0.5[\gamma_1|\Omega_2|^2 + \gamma_2|\Omega_1|^2 - p\sqrt{\gamma_1\gamma_2}(\Omega_1^*\Omega_2 + \Omega_1\Omega_2^*)] + i(\Delta_2|\Omega_1|^2 + \Delta_1|\Omega_2|^2) = 0, \quad (7)$$

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. (6). 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.

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. 1, 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. 2, we show the elimination of the 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

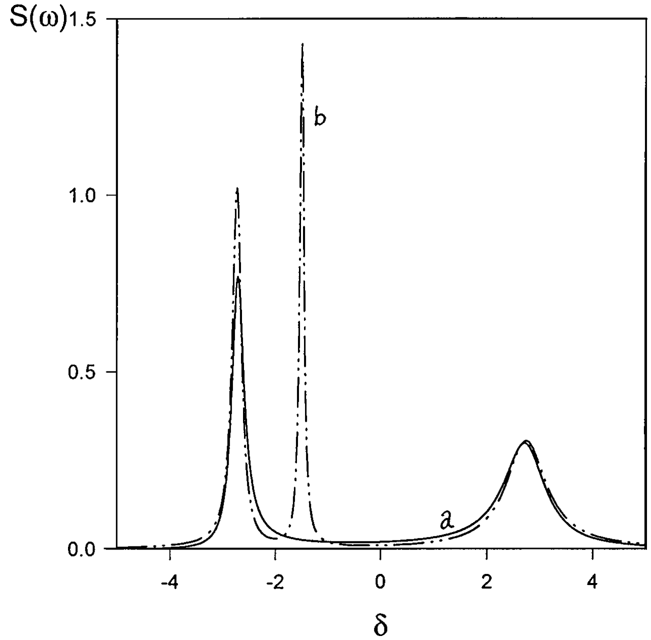

FIG. 2. The spontaneous spectra for $\Delta_1 = 4\gamma_1$, $\omega_{12} = 5\gamma_1$, $\Omega_1 = \gamma_1$, and $\gamma_2 = 0.25\gamma_1$, (a) $p = 1$, and (b) $p = 0$. The atom is initially in $(|a_1\rangle - |a_2\rangle)/\sqrt{2}$.

eliminated if

$$\Delta_2 = -\chi^2 \Delta_1 \,, \tag{8}$$

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 result of a destructive interference. As opposed to destructive interference, a constructive interference can also be found as shown in Fig. 3, where the central peak increases while the other two peaks decrease.

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 Figs. 1 and 2) 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. 4, 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 $|a_1\rangle$ and $|a_2\rangle$, $|\alpha_3|^2$, and $|\beta_3|^2$, are determined by

$$\begin{aligned} \alpha_1 + \alpha_2 + \alpha_3 &= A_1(0)\,, \\ \beta_1 + \beta_2 + \beta_3 &= A_2(0)\,, \\ b_1 + b_2 + b_3 &= B(0)\,, \qquad (i = 1,2,3)\,, \\ \lambda_i - \Omega_1^* \alpha_i - \Omega_2^* \beta_i &= 0\,, \\ \Omega_1 b_i - (\Gamma_1 - \lambda_i)\alpha_i - {} & \\ 0.5\sqrt{\gamma_1 \gamma_2}\,\beta_i &= 0\,, \end{aligned} \tag{9}$$

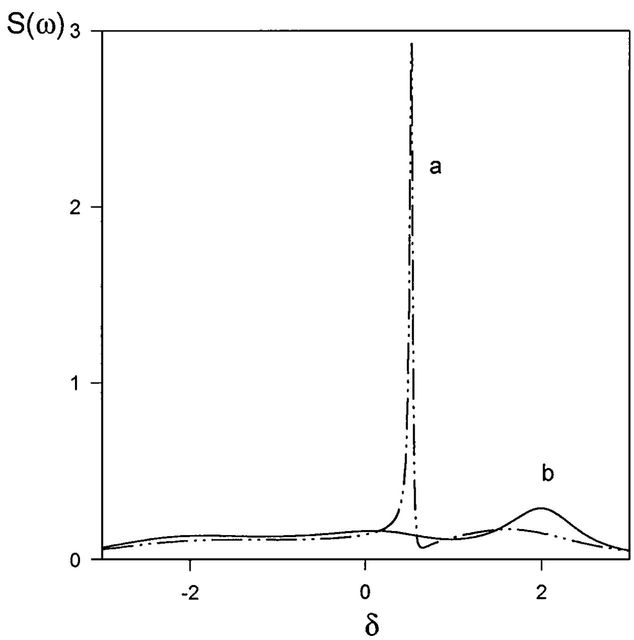

FIG. 3. The spectra with a constructive interference for $\omega_{12} = 2\gamma_1$, $\Delta_1 = \gamma_1$, $\Omega_1 = \gamma_1$, and $\gamma_2 = 4\gamma_1$, (a) $p = 1$, and (b) $p = 0$. The atom is initially in $(|a_1\rangle - 3|b\rangle)/\sqrt{10}$.

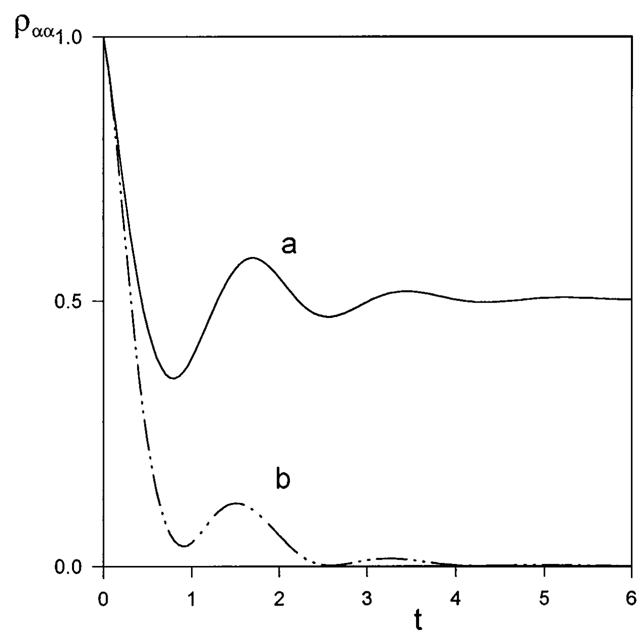

FIG. 4. Time evolution of populations in $|a_1\rangle$, for $\omega_{12} = 4\gamma_1$, $\gamma_2 = 4\gamma_1$, $\Delta_1 = 0.8\gamma_1$, and $\Omega_1 = 2\gamma_1$, (a) $p = 1$, and (b) $p = 0$. The atom is initially in level $|a_1\rangle$.

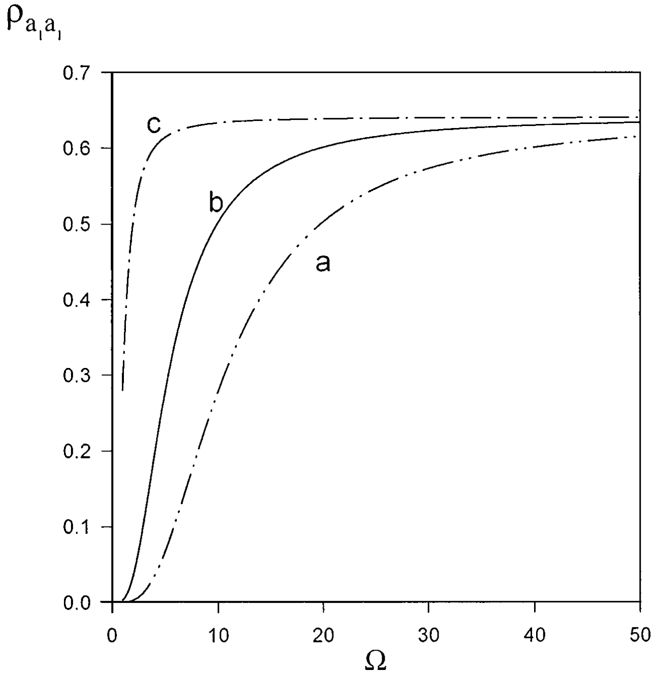

FIG. 5. The trapped population in level $|a_1\rangle$ versus the Rabi frequency (in unit of γ_1) $\omega_{12} =$ (a) $40\gamma_1$, (b) $20\gamma_1$, and (c) $4\gamma_1$.

where the conditions Eq. (8) and $p = 1$ have been used. For arbitrary χ 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. 4 we see more than 50% of the 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. 5 we plot the population in level $|a_1\rangle$ versus the Rabi frequency for three cases with $\chi = 2$ and Eq. (8) 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 $\gamma_1^2/\Delta_1 + \gamma_2^2/\Delta_2 + 2p\gamma_1\gamma_2/\Delta_1\Delta_2$. 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 can produce such two closely separated upper levels with parallel dipole moments, for example the $|2s\rangle$ and $|2p\rangle$ states of a hydrogen atom. This mixing results in two upper levels with a separation of $40\gamma_1$ [6]. Furthermore, two closely separated upper levels can be created from a single upper level by applying another field to couple it with an additional level. The detailed calculation and results to such considerations will be published elsewhere.

The cancellation of spontaneous emission has potential application in the generation of high power laser pulses and x-ray lasers. When the driving field is on, there is no spontaneous emission and the population in the upper levels could be extremely large. If we switch off the driving field, the upper-level population could transit into the lower level in a short time to generate a high power pulse. One of the difficulties in the generation of x-ray lasers is the lifetime of the upper levels. This spontaneous emission cancellation might provide an effective method to overcome this difficulty.

In conclusion, we studied the interference between spontaneous decay processes from two upper levels which are driven to another level by a strong field. Destructive interference results in the elimination of a spectral line and cancellation of spontaneous emission in the steady state. Constructive interference can enhance one of the spectral peaks with the others being depressed.

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