Quenching of spontaneous emission via quantum interference

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A four-level atom, driven by a coherent field, is considered. We show that under certain conditions complete quenching of spontaneous emission is possible. Hence the population inversion on some specific atomic transitions can be created using a very weak incoherent pumping. We investigate the physics of the effect using bare and dressed states. The proposed scheme may be useful, in principle, for generation of high-frequency and/or high power laser light. [S1050-2947(97)01306-1]

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I. INTRODUCTION

Modification of spontaneous emission is an active research topic in quantum optics. The three-peak spectrum of resonance fluorescence was one of the first developments in this area [1]. Cavity electrodynamics has made possible the enhancement [2] and suppression [3] of spontaneous emission from an atom via a ‘‘tailoring’’ of the mode density. Elimination of resonance fluorescence from a driven three-level atom was proposed in [4]. Dynamical suppression has also been achieved from a driven, cavity-confined, two-level atom [5]. Furthermore, it has been predicted [6] and experimentally demonstrated [7] that three-level atoms can exhibit a narrowing of spectral linewidth on one transition controlled by coherent driving another transition.

Cancellation of emission into a single mode was demonstrated in [8], when an atom is excited to a certain coherent superposition of two upper levels, as in Fig. 1. In such a case both spontaneous and stimulated emission on one specific frequency are suppressed. Elimination of steady-state resonance fluorescence was proposed in [4]. Furthermore, it has been shown [9] that the emission spectrum can be substantially modified via atomic coherence and interference even for an atom in an ordinary vacuum (see Fig. 2). The coherent preparation of atomic states essential for this effect can be realized via Autler-Townes splitting, as in Fig. 3 [10]. In a recent paper [11] it was shown that spectral line elimination and cancellation of spontaneous emission is possible under certain conditions, and experiment has also been performed to observe this phenomenon [12]. It was also shown [13] that the spectrum of resonance fluorescence, under certain conditions, can have spectral lines which are very narrow compared to the natural width of individual levels.

All spontaneous emission suppression effects mentioned above have one common origin: the quantum interference of spontaneous transitions from two closely lying atomic levels to a third level. But the emission cancellation in the model of Ref. [11] (see Fig. 4), which is the subject of the present paper, is different in that the emission can be suppressed for all modes near the transition frequencies and this cancellation is possible even if the upper levels are well separated. That is, the atoms can be ‘‘trapped’’ in the upper states without decaying. It is our goal to give a simple dressed-state description of this phenomenon. These studies suggest that one can, in principle, ‘‘control’’ the amount of fluorescence as well as the population of the upper levels.

We note that if spontaneous emission on some atomic transition is quenched, it might become possible to create a population inversion on this transition using a very weak incoherent pumping. Thus the control of spontaneous emission can be potentially useful in order to achieve high frequency lasing, since the spontaneous emission rate is typically proportional to the frequency cubed (ω^3), and creating the inversion on the high frequency transition with allowed spontaneous emission is therefore problematic.

The paper is organized as follows. In Sec. II we study the quenching of spontaneous emission in the driven four-level atom using bare states. In Sec. III we introduce the dressed states and explain how a spectral line can be eliminated from the emission spectrum. In order to describe the other additional decay processes from the upper levels and the incoherent pumping processes, we approach the problem via density matrix formalism in Sec. IV. In Sec. V we illustrate the mechanism of the upper level decay along the driven transitions and discuss how these decays influence the effect of spontaneous emission quenching. Section VI contains a summary of the results.

II. QUENCHING OF SPONTANEOUS EMISSION AND SPECTRAL LINE ELIMINATION

We consider a four-level model atom as shown in Fig. 4. It has two upper levels $|a_1\rangle$ and $|a_2\rangle$, which are coupled by the same vacuum modes to the lower level $|c\rangle$. The two upper levels are coupled by strong coherent field with frequency ν_0 to another upper lying level $|b\rangle$. The interaction picture Hamiltonian can be written as

$$\begin{aligned}\mathcal{V}=&\hbar\Omega_1 e^{i\Delta_1 t}|a_1\rangle\langle b|+\hbar\Omega_2 e^{i\Delta_2 t}|a_2\rangle\langle b|+\text{H.c.}\\&+\hbar\sum_k g_k^{(1)} e^{i(\omega_{1c}-\nu_k)t}|a_1\rangle\langle c|\hat{b}_k\\&+g_k^{(2)} e^{i(\omega_{2c}-\nu_k)t}|a_2\rangle\langle c|\hat{b}_k+\text{H.c.}\end{aligned} \tag{1}$$

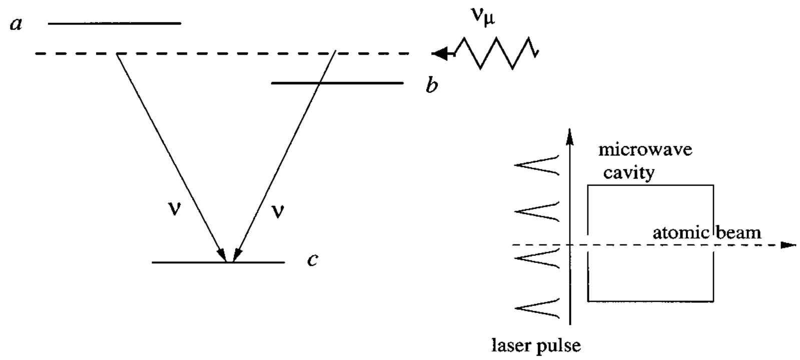

FIG. 1. Scheme of emission cancellation into a single mode. The V-type three-level atom is excited by the pulsed laser light and then brought through the high-Q-factor micromaser cavity. If microwave radiation appropriately mixes two upper states, then emission (both spontaneous and stimulated) of the photon with frequency ν is canceled [8].

The interaction of driven transitions with the vacuum modes is neglected in this section. $\Omega_{1,2}$ are the Rabi frequencies of the field coupling $|a_{1,2}\rangle$ and $|b\rangle$; in general, Ω_1 and Ω_2 can be different since they are proportional to the matrix elements of the corresponding dipole moments. The frequency differences between levels $|a_1\rangle$, $|a_2\rangle$, $|b\rangle$, and $|c\rangle$ are denoted by ω_{1c}, ω_{2c}, ω_{bc}, respectively. Detunings of the driving field are $\Delta_1=(\omega_{1c}-\omega_{bc})-\nu_0$, $\Delta_2=(\omega_{2c}-\omega_{bc})-\nu_0$. $g_k^{(1,2)}$ are the coupling constants between the kth vacuum mode and the atomic transitions from $|a_1\rangle$ and $|a_2\rangle$ to $|c\rangle$; they are assumed to be real. $\hat{b}_k$ ($\hat{b}_k^\dagger$) is the annihilation (creation) operator for the kth vacuum mode with frequency ν_k; k here represents both the momentum and polarization of the vacuum mode. If the atom is initially excited, we can write the initial state vector as

$$\Psi(0)=[A_1(0)|a_1\rangle+A_2(0)|a_2\rangle+B(0)|b\rangle]|\{0\}\rangle, \quad (2)$$

where $|\{0\}\rangle$ represents the absence of photons in all vacuum modes. The state vector at time t can be written as

$$\Psi(t)=[A_1(t)|a_1\rangle+A_2(t)|a_2\rangle+B(t)|b\rangle]|\{0\}\rangle + \sum_k C_k(t)|c\rangle|1_k\rangle. \quad (3)$$

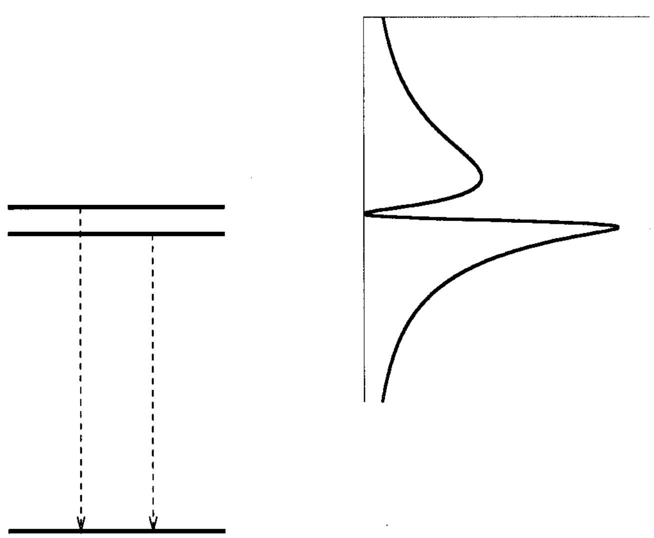

FIG. 2. Emission spectrum from an atom that has two closely spaced upper levels, coupled by the same vacuum modes to the common lower level. The spectrum shows a Fano zero at a certain frequency [9].

After a substitution $A_j(t)=a_j(t)e^{i\Delta_j t}$ we have (see Appendix A) the equations of motion for the probability amplitudes:

$$\frac{d}{dt}\hat{R}(t)=-M\hat{R}(t), \quad (4)$$

where

$$\hat{R}=\begin{pmatrix} a_1(t) \\ a_2(t) \\ B(t) \end{pmatrix}, \quad M=\begin{pmatrix} \Gamma_1 & p\sqrt{\gamma_1\gamma_2}/2 & i\Omega_1 \\ p\sqrt{\gamma_1\gamma_2}/2 & \Gamma_2 & i\Omega_2 \\ i\Omega_1^* & i\Omega_2^* & 0 \end{pmatrix}, \quad (5)$$

and $\Gamma_{1,2}=\gamma_{1,2}/2+i\Delta_{1,2}$. γ_1 and γ_2 are the radiative decay rates from the two upper levels to the lower level given by $\gamma_j=|\vec{\mu}_{jc}|^2\omega_{jc}^3/3\pi\epsilon_0\hbar c^3$ ($j=1,2$), and $\vec{\mu}_{jc}$'s are the matrix elements of the dipole moments of the two transitions. p denotes the alignment of the matrix elements of the two dipole moments and is given by

$$p=\frac{\langle a_1|\mathbf{r}|c\rangle\cdot\langle a_2|\mathbf{r}|c\rangle}{|\langle a_1|\mathbf{r}|c\rangle||\langle a_2|\mathbf{r}|c\rangle|}. \quad (6)$$

We say that the matrix elements of the two dipole moments are parallel (or antiparallel), if $p=1$ (or $p=-1$) and when they are orthogonal, $p=0$.

The solutions for $a_1(t)$, $a_2(t)$, and $B(t)$ are now given by

$$a_1(t)=\sum_{j=1}^{3}\alpha_j e^{-\lambda_j t}, \quad a_2(t)=\sum_{j=1}^{3}\beta_j e^{-\lambda_j t},$$

$$B(t)=\sum_{j=1}^{3} b_j e^{-\lambda_j t}, \quad (7)$$

where λ_j's are the three roots of the secular equation corresponding to matrix M. The coefficients α_j, β_j, b_j in Eq. (7) are determined by the initial state of the atom.

If we have nonzero $a_1(t=\infty)$, $a_2(t=\infty)$, and $B(t=\infty)$, this means that some population is trapped in the upper levels, which indicates that the spontaneous emission from the

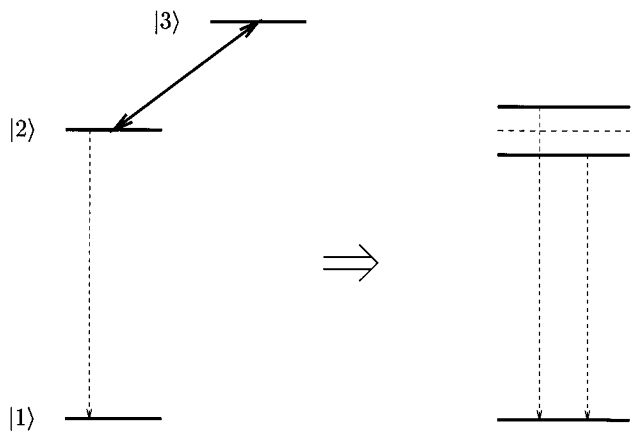

FIG. 3. Three-level driven atom and its ''dressed'' analog. If the initial state is state $|2\rangle$ then the emission spectrum has a dip at the central frequency [10].

two upper levels to level $|c\rangle$ is canceled. To have nonzero steady-state solution of Eq. (4) one needs

$$\det M \equiv \frac{1}{2}(\gamma_1|\Omega_2|^2+\gamma_2|\Omega_1|^2)-\frac{1}{2}p\sqrt{\gamma_1\gamma_2}(\Omega_1^*\Omega_2+\Omega_1\Omega_2^*)+i(\Delta_1|\Omega_2|^2+\Delta_2|\Omega_1|^2)=0. \tag{8}$$

If $|p|\neq 1$, Eq. (8) cannot be satisfied. For $p=\pm 1$, we find two conditions for having the nonzero upper level populations in the steady state:

$$\Delta_1|\Omega_2|^2+\Delta_2|\Omega_1|^2=0, \tag{9a}$$

$$p\frac{\Omega_1}{\Omega_2}=\sqrt{\frac{\gamma_1}{\gamma_2}} \quad (p=\pm 1). \tag{9b}$$

For Eq. (9b) to be true, one needs $p=1$ if Ω_1 and Ω_2 have the same sign, or $p=-1$ when Ω_1 and Ω_2 have the opposite signs.

Now, in steady state, the probability amplitude $C_k(\infty)$ of the atom being in the lower level with one photon emitted is given by [see Eq. (A9)]

$$C_k(\infty)=\sum_{j=1}^{3}\frac{i(g_k^{(1)}\alpha_j+g_k^{(2)}\beta_j)}{-\lambda_j+i[\nu_k-(\omega_{bc}+\nu_0)]}. \tag{10}$$

Since the spontaneous emission spectrum is proportional to $|C_k(t=\infty)|^2$, one might expect that there would be three peaks in the spectrum corresponding to the three resonant denominators in Eq. (10). This is the case if $p=0$. However, if $|p|=1$ and conditions (9a) and (9b) are fulfilled, the spectrum can have only two peaks. Mathematically this simply follows from the fact that under conditions (9a) and (9b) one of the three numerators in Eq. (10) is identically zero *at arbitrary initial state of the atom*. The physical reason for the spectral line elimination is the quantum interference of the two spontaneous transitions $|a_1\rangle\to|c\rangle$ and $|a_2\rangle\to|c\rangle$.

III. PHYSICS OF THE EFFECT VIA DRESSED STATES

In Sec. II and Ref. [11] it was shown that the general three-peak spontaneous emission spectrum, typical for such a

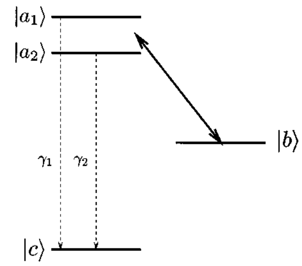

FIG. 4. Level scheme of a model atom. Upper levels $|a_1\rangle$ and $|a_2\rangle$ are coupled to $|b\rangle$ by a coherent field of frequency ν_0.

scheme, can be modified under certain conditions involving the intensity and detuning of the driving field. In particular, we saw that the central peak can be eliminated. A simple explanation of the peak elimination and cancellation of spontaneous emission can be given within the dressed-state picture. We now rewrite the interaction picture Hamiltonian in a rotating frame such as

$$\mathcal{V}=\mathcal{V}_0+\mathcal{V}_1,$$

$$\mathcal{V}_0=\hbar\Delta_1|a_1\rangle\langle a_1|+\hbar\Delta_2|a_2\rangle\langle a_2|+(\hbar g_1|a_1\rangle\langle b|\hat{a}+\hbar g_2|a_2\rangle\langle b|\hat{a}+\text{H.c.}),$$

$$\mathcal{V}_1=\hbar\sum_k g_k^{(1)}e^{-i\nu_k t}e^{i(\nu_0+\omega_{bc})t}|a_1\rangle\langle c|\hat{b}_k+g_k^{(2)}e^{-i\nu_k t}e^{i(\nu_0+\omega_{bc})t}|a_2\rangle\langle c|\hat{b}_k+\text{H.c.}, \tag{11}$$

where $\mathcal{V}_1$ describes the interaction with vacuum modes. $g_{1,2}$ are the coupling constants between $|a_{1,2}\rangle$ and $|b\rangle$. In spite of the fact that basic physics of spontaneous emission cancellation can be understood even if we consider the driving field classically, here we quantize it since we are going to use the formulas of this section later, when this quantization will be necessary. So, $\hat{a}$ ($\hat{a}^\dagger$) is the annihilation (creation) operator for the one-mode driving field. Diagonalizing $\mathcal{V}_0$ which corresponds to the interaction of the atom with the driving field, we arrive at the characteristic equation

$$x_n^3-x_n^2(\Delta_1+\Delta_2)-x_n[g_1^2(n+1)+g_2^2(n+1)-\Delta_1\Delta_2]+\Delta_1 g_2^2(n+1)+\Delta_2 g_1^2(n+1)=0. \tag{12}$$

If we assume, for simplicity, that

$$\Delta_1 g_2^2+\Delta_2 g_1^2=0, \tag{13}$$

then there is one trivial eigenvalue $x_n^0=0$ and we find the eigenstates such as

$$|0,n\rangle=N_{0,n}\bigg[g_2\sqrt{n+1}|a_1,n\rangle-g_1\sqrt{n+1}|a_2,n\rangle-\frac{g_2}{g_1}\Delta_1|b,n+1\rangle\bigg],$$

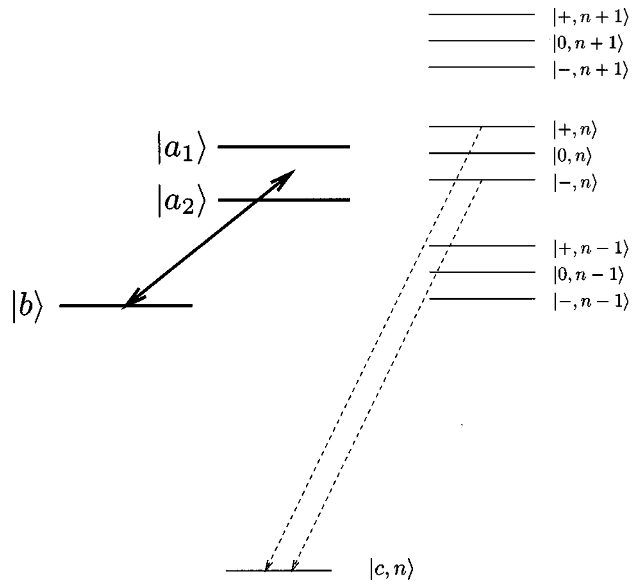

FIG. 5. Atomic transitions in view of dressed states.

$$|\pm,n\rangle=N_{\pm,n}\bigg[g_1\sqrt{n+1}\bigg(\mu\pm\frac{\omega_{12}}{2}\bigg)|a_1,n\rangle + g_2\sqrt{n+1}\bigg(\mu\mp\frac{\omega_{12}}{2}\bigg)|a_2,n\rangle \pm(g_1^2+g_2^2)(n+1)|b,n+1\rangle\bigg], \tag{14}$$

where

$$\mu=\sqrt{g_1^2(n+1)+g_2^2(n+1)+\frac{\omega_{12}^2}{4}}, \tag{15}$$

$\omega_{12}=\omega_{1c}-\omega_{2c}$ is the spacing between two upper levels, and $N_{0,n}$ and $N_{\pm,n}$ are the normalization constants. Corresponding eigenvalues are

$$x_n^0=0, \quad x_n^{\pm}=\frac{\Delta_1+\Delta_2}{2}\pm\mu. \tag{16}$$

Once we have a ''dressed''-state description of the two upper levels $|a_1\rangle$, $|a_2\rangle$ connected to $|b\rangle$ by the driving field, we can describe spontaneous emission from the upper levels in terms of decay from the atom–driving-field combined system (see Fig. 5). For the matrix element of the atom–vacuum-field interaction between the state $|0,n\rangle$ and the ground state $|c,n\rangle$, we have

$$\begin{aligned}V_{c,0;n}(t)&=\langle c,n|\langle 1_k|\mathcal{V}_1|0,n\rangle|\{0\}\rangle\\&=N_{0,n}[\hbar g_2\sqrt{n+1}g_k^{(1)}\\&\quad-\hbar g_1\sqrt{n+1}g_k^{(2)}]e^{i\nu_k t}e^{-i(\nu_0+\omega_{bc})t}.\end{aligned} \tag{17}$$

This matrix element vanishes if

$$\frac{g_k^{(1)}}{g_k^{(2)}}=\frac{g_1}{g_2} \tag{18}$$

for arbitrary mode of the vacuum field. Since, by definition,

$$\frac{g_k^{(1)}}{g_k^{(2)}}=\frac{\vec{\mu}_{1c}\cdot\hat{\mathbf{e}}_k}{\vec{\mu}_{2c}\cdot\hat{\mathbf{e}}_k} \tag{19}$$

(where $\hat{\mathbf{e}}_k$ is the unit polarization vector of the kth radiation mode and $\vec{\mu}_{jc}$'s are the matrix elements of the dipole moments of the two transitions), the parallel matrix elements of the two dipole moments are needed for vanishing of $V_{c,0;n}$ for arbitrary polarization of the vacuum field, assuming that g_1 and g_2 have the same sign. In the case when g_1 and g_2 have opposite signs, matrix element (17) can be zero for each vacuum mode, if the dipole moments are antiparallel.

An explicit expression for the decay rate from the dressed state $|0,n\rangle$ to the state $|c,n\rangle$ can be obtained as

$$\begin{aligned}\gamma_{0;n}&=\frac{d}{dt}\sum_k\left|-\frac{i}{\hbar}\int_0^t dt' V_{c,0;n}\right|^2\\&=\frac{d}{dt}N_{0,n}^2\sum_k\int_0^t dt'\int_0^t dt''[g_2^2(n+1)\\&\quad\times g_k^{(1)}g_k^{(1)}e^{i(\nu_k-\nu)(t'-t'')}+g_1^2(n+1)\\&\quad\times g_k^{(2)}g_k^{(2)}e^{i(\nu_k-\nu)(t'-t'')}-\{g_2g_1(n+1)\\&\quad\times g_k^{(1)}g_k^{(2)}e^{i(\nu_k-\nu)(t'-t'')}+\text{c.c.}\}],\end{aligned} \tag{20}$$

where $\nu=\nu_0+\omega_{bc}$. Replacing the summation over k by an integration and using the Weisskopf-Wigner approximation [14], we have

$$\begin{aligned}\gamma_{0;n}=N_{0,n}^2[&\gamma_1g_2^2(n+1)+\gamma_2g_1^2(n+1)\\&-2p\sqrt{\gamma_1\gamma_2}g_2g_1(n+1)].\end{aligned} \tag{21}$$

Similarly, we can find the decay rates from the other dressed states:

$$\begin{aligned}\gamma_{\pm,n}=N_{\pm,n}^2\bigg[&\gamma_1g_1^2(n+1)\bigg(\mu\pm\frac{\omega_{12}}{2}\bigg)^2+\gamma_2g_2^2(n+1)\\&\times\bigg(\mu\mp\frac{\omega_{12}}{2}\bigg)^2+2p\sqrt{\gamma_1\gamma_2}g_2g_1(n+1)\bigg(\mu^2-\frac{\omega_{12}^2}{4}\bigg)\bigg].\end{aligned} \tag{22}$$

One can see that γ_0 can be zero only if $|p|=1$ (parallel or antiparallel case). Then the condition for zero transition rate is

$$\frac{g_1}{g_2}=p\sqrt{\frac{\gamma_1}{\gamma_2}}, \tag{23}$$

together with Eq. (13). We will call $|p|=1$ and Eqs. (13) and (23) the *trapping conditions*. ''Trapping conditions'' simply mean that atom–vacuum-field interaction does not couple the transition between the dressed state $|0,n\rangle$ and the state $|c,n\rangle$. Note here that if we introduce the Rabi frequencies of the driving field such as $\Omega_{1,2}=g_{1,2}\sqrt{n+1}$ (n is the number of photons in the one-mode driving field), then the trapping conditions will exactly coincide with the conditions (9a) and (9b) obtained in the preceding section with classical driving field.

Assume that g_1 and g_2 have the same sign. Then $p=1$ is required for destructive interference in spontaneous emission from the dressed state $|0\rangle$ and for suppression of the corresponding peak in spectrum. However, a constructive interference is also possible. From Eq. (21) one can see that for $p=-1$, the decay rate γ_0 increases. In particular, if $g_1=g_2$ and $p=-1$, γ_0 becomes twice greater than in the case when interference of spontaneous transitions is absent $(p=0)$. Thus, for the central peak we have destructive interference if $p=1$ and constructive interference if $p=-1$.

On the contrary, for the two side peaks, we have constructive interference if $p=1$ and destructive interference if $p=-1$. This is indicated by the increase of the widths of the side peaks for the case $p=1$ (which means the increase of the decay rates from the dressed states $|\pm,n\rangle$) and by the decrease of these widths for the case $p=-1$. It can be shown that the decay rates $\gamma_\pm$ are not equal to zero for any p, therefore two side peaks cannot be eliminated for arbitrary initial state of the atom.

Consider a simple particular case: $g_1\sqrt{n+1}=g_2\sqrt{n+1}=\Delta_1=-\Delta_2\equiv\gamma$. [If we additionally require Eq. (23), the trapping conditions will be fulfilled]. Then three dressed states read

$$|0,n\rangle=\frac{1}{\sqrt{3}}[|a_1,n\rangle-|a_2,n\rangle-|b,n+1\rangle],$$

$$|\pm,n\rangle=\frac{1}{\sqrt{3}}\left[\frac{\sqrt{3}\pm1}{2}|a_1,n\rangle+\frac{\sqrt{3}\mp1}{2}|a_2,n\rangle\pm|b,n+1\rangle\right]. \tag{24}$$

Corresponding energies are $x_n=0,\pm\sqrt{3}\gamma$. Now if the initial atomic state is $|a_1\rangle$, it can be rewritten as

$$|a_1,n\rangle=\frac{1}{2\sqrt{3}}[(\sqrt{3}+1)|+,n\rangle-(\sqrt{3}-1)|-,n\rangle+2|0,n\rangle]. \tag{25}$$

Then we expect that the spontaneous emission spectrum will have three peaks centered at $-\sqrt{3}\gamma$, 0, and $\sqrt{3}\gamma$ which correspond to the three dressed states $|-\rangle$, $|0\rangle$, and $|+\rangle$. For $p=1$, however, there is no coupling between $|0,n\rangle$ and $|c,n\rangle$ [see Eq. (17)]. The entire initial population in $|0,n\rangle$ will stay there and give no contribution to the spontaneous emission. That is why we have the central peak eliminated from the spectrum (Fig. 6). (Note that under the trapping conditions there will be no central peak in the spectrum for any initial state of the atom.) If the atomic state is $|0,n\rangle$ initially, one can clearly see the difference between $p=0$ and $p=1$ cases. If $p=0$, we simply expect one peak at the center since it is the dressed state $|0,n\rangle$ which is responsible for the central peak. On the other hand, there is no spontaneous emission at all for $p=1$ and the entire population is trapped in the state $|0,n\rangle$. In this last case spontaneous emission is completely canceled.

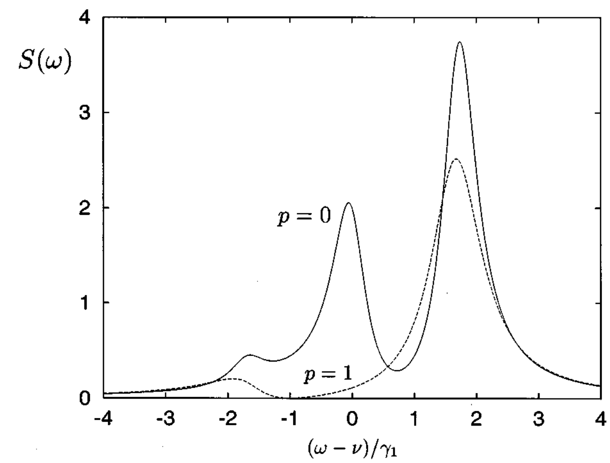

FIG. 6. Spontaneous emission spectra for $\omega_{12}=2\gamma_1$, $\Omega_1=\Omega_2=\gamma_2=\Delta_1=\gamma_1$, and (a) $p=0$ and (b) $p=1$. The atom is initially in level $|a_1\rangle$ [11].

If the trapping conditions are fulfilled, then the population trapped in the upper levels can be calculated as a portion of the state $|0,n\rangle$ in the initial state. For example, at $g_1\sqrt{n+1}=g_2\sqrt{n+1}\equiv\Omega_n$ and $\Delta_1=-\Delta_2=\omega_{12}/2$, state $|a_1\rangle$ can be rewritten in terms of the dressed states as

$$|a_1,n\rangle=\frac{1}{2\mu}\left[\left(\mu+\frac{\omega_{12}}{2}\right)|+,n\rangle+\left(\mu-\frac{\omega_{12}}{2}\right)|-,n\rangle+2\Omega_n|0,n\rangle\right], \tag{26}$$

with $\mu=\sqrt{2\Omega_n^2+\omega_{12}^2/4}$. Hence for the atom initially prepared in the state $|a_1\rangle$, we can find that trapped populations are

$$|A_1(\infty)|^2=|A_2(\infty)|^2=\frac{16\Omega_n^4}{(8\Omega_n^2+\omega_{12}^2)^2},$$

$$|B(\infty)|^2=\frac{\Omega_n^2\omega_{12}^2}{(8\Omega_n^2+\omega_{12}^2)^2}. \tag{27}$$

Here we can see that even if two upper levels are well separated, a significant amount of population can be trapped in the upper levels provided that the driving field is strong enough.

IV. MASTER EQUATION APPROACH

In the preceding section the decays along the driven transitions $|a_1\rangle\to|b\rangle$ and $|a_2\rangle\to|b\rangle$ were not taken into account and, to have population trapped in the upper levels, the atom was assumed to be initially prepared in a specific (dark) state. To describe more realistic situations one needs to include the decays from upper levels $|a_1\rangle$ and $|a_2\rangle$ to $|b\rangle$, see Fig. 7. In other words, levels $|a_1\rangle$ and $|a_2\rangle$ are coupled by the same vacuum modes also to the level $|b\rangle$ (these modes are supposed to be different from those relevant to the transitions from $|a_1\rangle$ and $|a_2\rangle$ to $|c\rangle$). For this purpose, it is more convenient to describe the system by standard density matrix formalism, and the spontaneous emission spectrum can be

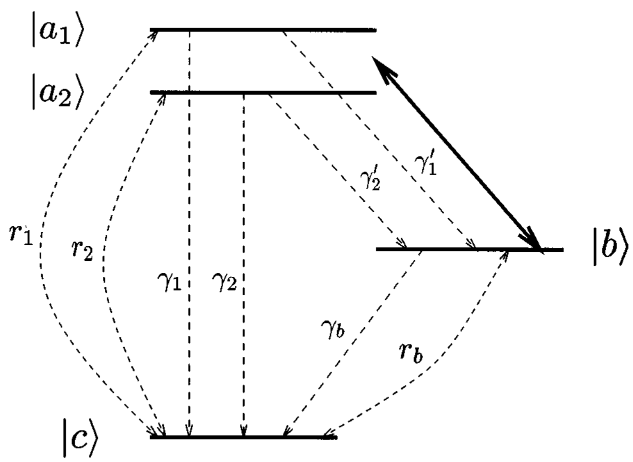

FIG. 7. Atomic level scheme including all the decay and incoherent pumping processes. The incoherent pumping r_j's are supposed to be in both directions.

calculated with use of the quantum regression theorem. In the following we will treat the driving field classically. It is difficult to obtain simple analytical expressions in the master equation approach, therefore we find the populations and spectrum numerically. Since we are now interested in a steady-state solution, we also include the incoherent pumping from level $|c\rangle$ to levels $|a_1\rangle$ and $|a_2\rangle$.

Including the interaction of the atom and the vacuum modes which couple the transitions between $|a_{1,2}\rangle$ and $|b\rangle$, the total interaction Hamiltonian in this case can be written as

$$\mathcal{V}=\hbar\Delta_1|a_1\rangle\langle a_1|+\hbar\Delta_2|a_2\rangle\langle a_2| +(\hbar\Omega_1|a_1\rangle\langle b|+\hbar\Omega_2|a_2\rangle\langle b|+\text{H.c.})+\mathcal{V}_1+\mathcal{V}_2, \tag{28}$$

$$\mathcal{V}_1=\hbar\sum_k g_k^{(1)}e^{-i\nu_k t}e^{i(\nu_0+\omega_{bc})t}|a_1\rangle\langle c|\hat{b}_k +g_k^{(2)}e^{-i\nu_k t}e^{i(\nu_0+\omega_{bc})t}|a_2\rangle\langle c|\hat{b}_k+\text{H.c.},$$

$$\mathcal{V}_2=\hbar\sum_q \tilde{g}_q^{(1)}e^{i(\nu_0-\nu_q)t}|a_1\rangle\langle b|\hat{a}_q +\tilde{g}_q^{(2)}e^{i(\nu_0-\nu_q)t}|a_2\rangle\langle b|\hat{a}_q+\text{H.c.},$$

where Ω_1 and Ω_2 are Rabi frequencies of the driving field corresponding to the two transitions from $|a_1\rangle$ and $|a_2\rangle$ to $|b\rangle$, respectively, $\tilde{g}_q^{(1,2)}$ are coupling constants between the qth vacuum mode and the atomic transitions from $|a_{1,2}\rangle$ to $|b\rangle$ (for simplicity they are assumed to be real), $\hat{a}_q$ ($\hat{a}_q^\dagger$) is the annihilation (creation) operator for the qth vacuum mode with frequency ν_q. In the following the Rabi frequencies are assumed to be real and positive (which corresponds to the case of positive coupling constants $g_{1,2}$ in Sec. II).

Using a Weisskopf-Wigner approximation in the generalized reservoir theory [15], we derive the equations of motion for the atomic density matrix elements which are [16]

$$\begin{aligned}
\dot{\rho}_{a_1c}&=-\left[\frac{1}{2}(\gamma_1+\gamma_1'+r_1+r)+i\Delta_1\right]\rho_{a_1c}-\frac{1}{2}(p\sqrt{\gamma_1\gamma_2}+p'\sqrt{\gamma_1'\gamma_2'})\rho_{a_2c}-i\Omega_1\rho_{bc},\\
\dot{\rho}_{a_1a_2}&=-\left[\frac{1}{2}(\gamma_1+\gamma_1'+\gamma_2+\gamma_2'+r_1+r_2)+i(\Delta_1-\Delta_2)\right]\rho_{a_1a_2}-\frac{1}{2}(p\sqrt{\gamma_1\gamma_2}+p'\sqrt{\gamma_1'\gamma_2'})(\rho_{a_1a_1}+\rho_{a_2a_2})+i(\Omega_2\rho_{a_1b}-\Omega_1\rho_{ba_2}),\\
\dot{\rho}_{a_1a_1}&=-(\gamma_1+\gamma_1'+r_1)\rho_{a_1a_1}-\frac{1}{2}(p\sqrt{\gamma_1\gamma_2}+p'\sqrt{\gamma_1'\gamma_2'})(\rho_{a_1a_2}+\rho_{a_2a_1})+r_1\rho_{cc}-i\Omega_1(\rho_{ba_1}-\rho_{a_1b}),\\
\dot{\rho}_{a_1b}&=-\left[\frac{1}{2}(\gamma_1+\gamma_1'+\gamma_b+r_1+r_b)+i\Delta_1\right]\rho_{a_1b}-\frac{1}{2}(p\sqrt{\gamma_1\gamma_2}+p'\sqrt{\gamma_1'\gamma_2'})\rho_{a_2b}-i\Omega_1(\rho_{bb}-\rho_{a_1a_1})+i\Omega_2\rho_{a_1a_2},\\
\dot{\rho}_{a_2c}&=-\left[\frac{1}{2}(\gamma_2+\gamma_2'+r_2+r)+i\Delta_2\right]\rho_{a_2c}-\frac{1}{2}(p\sqrt{\gamma_1\gamma_2}+p'\sqrt{\gamma_1'\gamma_2'})\rho_{a_1c}-i\Omega_2\rho_{bc},\\
\dot{\rho}_{a_2a_2}&=-(\gamma_2+\gamma_2'+r_2)\rho_{a_2a_2}-\frac{1}{2}(p\sqrt{\gamma_1\gamma_2}+p'\sqrt{\gamma_1'\gamma_2'})(\rho_{a_2a_1}+\rho_{a_1a_2})+r_2\rho_{cc}-i\Omega_2(\rho_{ba_2}-\rho_{a_2b}),\\
\dot{\rho}_{a_2b}&=-\left[\frac{1}{2}(\gamma_2+\gamma_2'+\gamma_b+r_2+r_b)+i\Delta_2\right]\rho_{a_2b}-\frac{1}{2}(p\sqrt{\gamma_1\gamma_2}+p'\sqrt{\gamma_1'\gamma_2'})\rho_{a_1b}-i\Omega_2(\rho_{bb}-\rho_{a_2a_2})+i\Omega_1\rho_{a_2a_1},\\
\dot{\rho}_{bc}&=-\frac{1}{2}(\gamma_b+r_b+r)\rho_{bc}-i\Omega_2\rho_{a_2c}-i\Omega_1\rho_{a_1c},
\end{aligned} \tag{29}$$

$$\dot{\rho}_{bb}=-(\gamma_b+r_b)\rho_{bb}+\gamma_1'\rho_{a_1a_1}+\gamma_2'\rho_{a_2a_2}+r_b\rho_{cc}+p'\sqrt{\gamma_1'\gamma_2'}(\rho_{a_1a_2}+\rho_{a_2a_1})-i\Omega_2(\rho_{a_2b}-\rho_{ba_2})-i\Omega_1(\rho_{a_1b}-\rho_{ba_1}),$$

$$1=\rho_{a_1a_1}+\rho_{a_2a_2}+\rho_{bb}+\rho_{cc},$$

where γ_j' is the decay rate from level $|a_j\rangle$ to $|b\rangle$ and the level $|b\rangle$ decays to $|c\rangle$ (due to collisions, for example) with a decay rate γ_b. p stands for the alignment of the dipole moments along the transitions from levels $|a_1\rangle$ and $|a_2\rangle$ to $|c\rangle$ as in Eq. (6) and, similarly, p' denotes the alignment of the dipole moments along the transitions from $|a_1\rangle$ and $|a_2\rangle$ to $|b\rangle$. We assume the incoherent pumping to be in both directions; the pumping rate from the level $|c\rangle$ to level $|a_1\rangle$ ($|a_2\rangle$) and back is denoted by r_1 (r_2). For generality we also include the incoherent pumping from level $|c\rangle$ to level $|b\rangle$ and back (r_b), but in our numerical calculations we set it equal to zero. r denotes the sum of all three pumping rates: $r\equiv r_1+r_2+r_b$. The atom is assumed to be a closed system, therefore the total population is conserved.

Solving Eq. (29), we can obtain the time evolution of the populations and the steady state population in each level. Thus we can define how much population inversion we will have in the steady state with a certain incoherent pumping. In Fig. 8 we plot the time evolution of the population in each level under the trapping conditions. At $t=0$, a weak incoherent pumping is switched on ($r_1=r_2=0.01\gamma_1$); at $t=500/\gamma_1$, the system almost reaches steady state. In the steady state we have 80% of population trapped in the two upper levels, with the lower level having only 0.2% of the total population. In Fig. 9 we plot the population inversion ($\rho_{a_1a_1}+\rho_{a_2a_2}-\rho_{cc}$) as a function of the Rabi frequency for different upper level separations. Larger upper level separation needs a larger Rabi frequency for the driving field in order to get the same amount of inversion in the steady state. For a quite large separation, $\omega_{12}=20\gamma_1$, we need Rabi frequency of $\approx 2\gamma_1$ to have an inversion. Note that in the limit of very large intensity of the driving field, when $\Omega_1\gg\omega_{12}$, both levels $|b\rangle$ and $|c\rangle$ are not populated and the whole population is distributed between only two upper levels $|a_1\rangle$ and $|a_2\rangle$. Actually, in this limit the steady state of the atomic system is the antisymmetric combination of these two upper levels.

Figures 8 and 9 are the main result of the present paper. They show that for the proposed scheme (Fig. 4) under the certain trapping conditions specified in Sec. III, it is possible to establish the population inversion and hold it up in the steady state even with a very weak incoherent pumping.

Calculation of spontaneous emission spectrum using the quantum regression theorem is described in detail in Appendix B. The spectrum of radiation, spontaneously emitted by the transitions from $|a_1\rangle$ and $|a_2\rangle$ to $|c\rangle$, is given by Eq. (B14). As we expect from the discussion of the previous sections, we have a three-peak spectrum for $p=p'=0$ ($\Omega_1\neq 0,\Omega_2\neq 0$) as shown in Fig. 10(a). Now compare two cases: $p=-1,p'=1$ and $p=p'=1$. First, as shown in Fig. 10(b), for the case of $p=-1,p'=1$, the central peak is broadened while the two side peaks are narrowed. A broadening (or narrowing) of the peak means an increase (or decrease) of the corresponding dressed-state decay rate. Therefore we have decay rate enhancement for the dressed state corresponding to the central peak at the expense of decay rate decrease for the states corresponding to the side peaks. There is no population trapping in the upper levels for the case $p=-1$, $p'=1$, and the steady-state populations are only slightly different from those of the case $p=p'=0$ corresponding to the absence of interference in spontaneous emission. However, in the case of $p=p'=1$ we have almost

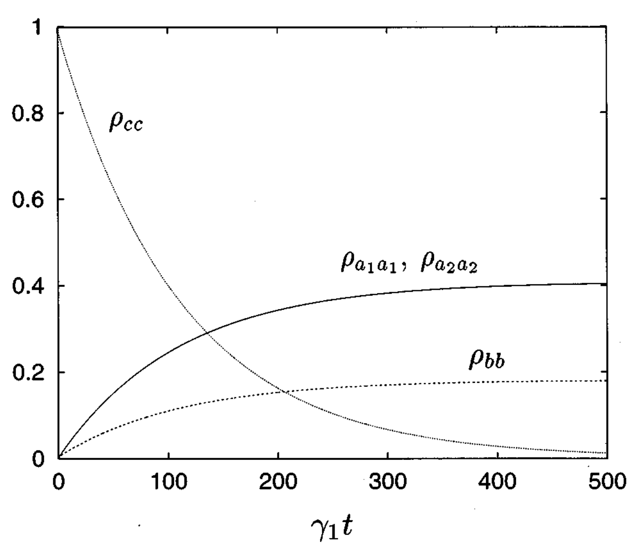

FIG. 8. Time evolution of the populations for $\omega_{12}=2\gamma_1$, $\Delta_1=-\Delta_2=\gamma_2=\gamma_1$, $\gamma_1'=\gamma_2'=0.5\gamma_1$, $\Omega_1=\Omega_2=1.5\gamma_1$, $\gamma_b=10^{-4}\gamma_1$, and $r_1=r_2=0.01\gamma_1$.

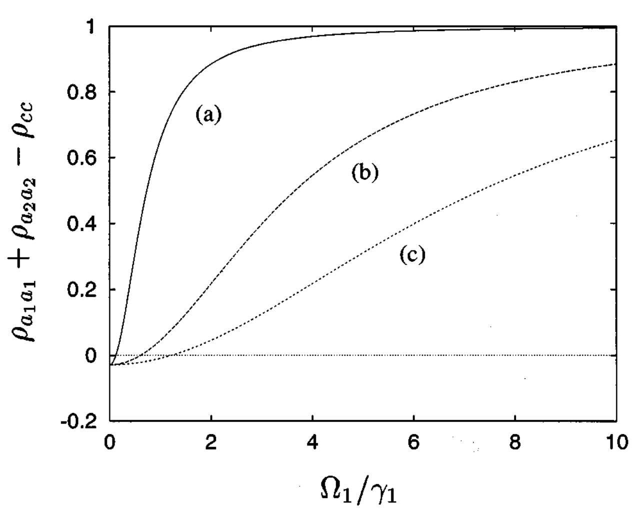

FIG. 9. Population inversion ($\rho_{a_1a_1}+\rho_{a_2a_2}-\rho_{cc}$) vs Rabi frequency Ω_1. $\Delta_1=-\Delta_2=\omega_{12}/2$, $\gamma_2=\gamma_1$, $\gamma_1'=\gamma_2'=0.5\gamma_1$, $\gamma_b=10^{-4}\gamma_1$, $r_1=r_2=0.01\gamma_1$, and $\Omega_2=\Omega_1$, (a) $\omega_{12}=2\gamma_1$, (b) $\omega_{12}=10\gamma_1$ and (c) $\omega_{12}=20\gamma_1$.

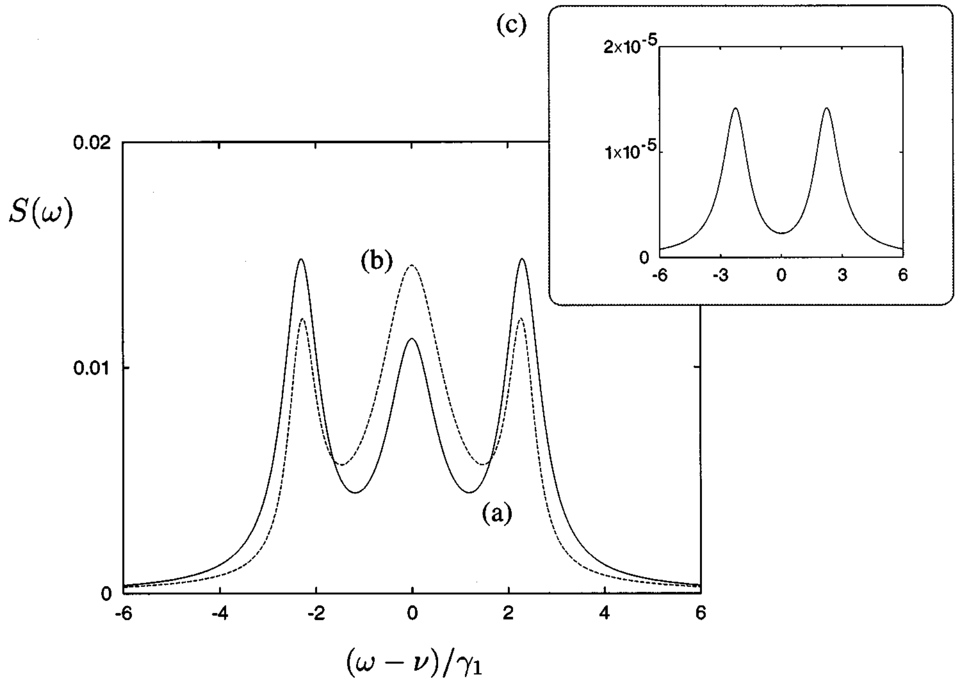

FIG. 10. Spontaneous emission spectra for $\omega_{12}=2\gamma_1$, $\Delta_1=-\Delta_2=\gamma_2=\gamma_1$, $\Omega_1=\Omega_2=1.5\gamma_1$, $\gamma_1'=\gamma_2'=0.5\gamma_1$, $\gamma_b=10^{-4}\gamma_1$, $r_1=0.01\gamma_1$, and (a) $p=p'=0$, with steady-state population $\rho_{a_1a_1}=0.010$, $\rho_{a_2a_2}=0.010$, $\rho_{bb}=0.012$, and $\rho_{cc}=0.969$, (b) $p=-1,p'=1$, with steady-state population $\rho_{a_1a_1}=0.017$, $\rho_{a_2a_2}=0.017$, $\rho_{bb}=0.024$, and $\rho_{cc}=0.941$; and (c) $p=p'=1$, with steady-state population $\rho_{a_1a_1}=0.408$, $\rho_{a_2a_2}=0.408$, $\rho_{bb}=0.181$, and $\rho_{cc}=0.002$.

zero emission from levels $|a_1\rangle$ and $|a_2\rangle$ to level $|b\rangle$ [the area under the spectrum curve is almost zero, as shown in Fig. 10(c)], which indicates almost complete cancellation of the spontaneous emission. (The fact that very weak emission is still present in this last case is only due to both two-way incoherent pumping and small decay rate from $|b\rangle$ to $|c\rangle$. If we used one-way upward pumping with no decay along $|b\rangle\to|c\rangle$ transition, there would be no emission at all in the steady state, and the population of the level $|c\rangle$ would be identically zero.)

V. THE EFFECT OF UPPER LEVEL DECAYS ALONG THE DRIVEN TRANSITIONS

In the preceding section we have shown that even if we include the decays of the upper levels to the level $|b\rangle$, the effect of spontaneous emission cancellation is not destroyed. Moreover, these decays make it possible to have population inversion in the steady state, independently of the initial state of the system at very low incoherent pumping rates. We now discuss, in more detail, how these decays actually work. To do this in the simplest way we again switch to the quantized description of the driving field. A very clear picture can again be provided in terms of the dressed states introduced in Sec. III.

Let us consider mutual decays between the dressed states due to the upper level decays to the level $|b\rangle$ (Fig. 5). The interaction Hamiltonian for these transitions is given by $\mathcal{V}_2$ in Eq. (28),

$$\mathcal{V}_2=\hbar\sum_q \tilde{g}_q^{(1)}e^{i(\nu_0-\nu_q)t}|a_1\rangle\langle b|\hat{a}_q+\tilde{g}_q^{(2)}e^{i(\nu_0-\nu_q)t}|a_2\rangle\langle b|\hat{a}_q+\text{adj.} \tag{30}$$

The matrix element of the transition from the dressed state $|\pm,n+1\rangle$ to the state $|0,n\rangle$ can be written as

$$\begin{aligned}V_{0,\pm;n}(t)&=\langle 0,n|\langle 1_q|\mathcal{V}_2|\pm,n+1\rangle|\{0\}\rangle\\&=N_{0,n}N_{\pm,n+1}\left\langle 0,n\left|\left[\hbar\tilde{g}_q^{(1)}g_1\sqrt{n+2}\left(\mu\pm\frac{\omega_{12}}{2}\right)\right.\right.\right.\\&\quad\left.\left.+\hbar\tilde{g}_q^{(2)}g_2\sqrt{n+2}\left(\mu\mp\frac{\omega_{12}}{2}\right)e^{i(\nu_q-\nu_0)t}\right|b,n+1\right\rangle\\&=N_{0,n}N_{\pm,n+1}\left(-\frac{g_2}{g_1}\Delta_1\right)\left[\hbar\tilde{g}_q^{(1)}g_1\sqrt{n+2}\right.\\&\quad\times\left(\mu\pm\frac{\omega_{12}}{2}\right)+\hbar\tilde{g}_q^{(2)}g_2\sqrt{n+2}\\&\quad\times\left(\mu\mp\frac{\omega_{12}}{2}\right)e^{i(\nu_q-\nu_0)t}.\end{aligned} \tag{31}$$

On the other hand, matrix elements of the transitions from $|0,n+1\rangle$ to $|\pm,n\rangle$ are given by

$$\begin{aligned}V_{\pm,0;n}(t)&=\langle\pm,n|\langle 1_q|\mathcal{V}_2|0,n+1\rangle|\{0\}\rangle\\&=N_{\pm,n}N_{0,n+1}\langle\pm,n|[\hbar\tilde{g}_q^{(1)}g_2\sqrt{n+2}\\&\quad-\hbar\tilde{g}_q^{(2)}g_1\sqrt{n+2}]e^{i(\nu_q-\nu_0)t}|b,n+1\rangle\\&=N_{\pm,n}N_{0,n+1}[\pm(g_1^2+g_2^2)(n+1)][\hbar\tilde{g}_q^{(1)}g_2\sqrt{n+2}\\&\quad-\hbar\tilde{g}_q^{(2)}g_1\sqrt{n+2}e^{i(\nu_q-\nu_0)t}.\end{aligned} \tag{32}$$

One can see that matrix element (32) can be zero and the condition for this is similar to Eq. (18):

$$\frac{\tilde{g}_q^{(1)}}{\tilde{g}_q^{(2)}}=\frac{g_1}{g_2}. \tag{33}$$

Hence we can have no decay from $|0,n+1\rangle$ to $|\pm,n\rangle$. On the other hand, under the condition (33) the matrix element (31) is maximal, and the decay rates from states $|\pm,n+1\rangle$ to the state $|0,n\rangle$ are not zero. Therefore if Eq. (33) is valid, dressed states $|\pm\rangle$ can decay into the dressed state $|0\rangle$, but not vice versa [4]. By analogy with Eq. (23), condition (33) can be rewritten as

$$\frac{g_1}{g_2}=p'\sqrt{\frac{\gamma_1'}{\gamma_2'}}, \tag{34}$$

where γ_1' and γ_2' are the radiative decay rates from the two upper levels to the level $|b\rangle$ and p' was defined in the preceding section as the alignment of the dipole moments corresponding to the driven transitions. Now one can see that if the trapping conditions (specified in Sec. III) are fulfilled together with Eq. (34), then for any initial state of the atom and arbitrarily small incoherent pumping rates from level $|c\rangle$ to levels $|a_{1,2}\rangle$, the decays from the two upper levels to the level $|b\rangle$ eventually put the atom into a coherent superposition state, which is nothing but the nondecaying state $|0,n\rangle$. The atom stays in this dark state as long as the driving field is turned on.

VI. CONCLUSION

In this paper we have presented the physical origin of the cancellation of spontaneous emission in terms of the atom-field dressed states. One of the dressed states can be a nondecaying one under certain conditions, and it is therefore possible to hold the population in the upper levels even for the case when the two bare states are well separated. The spontaneous emission processes along the driven transitions can make this effect even more pronounced. These additional decays add population to the nondecaying state and consequently we can have complete cancellation of spontaneous emission together with population inversion in steady state. This effect could be potentially useful for high frequency and high power laser systems.

ACKNOWLEDGMENTS

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APPENDIX A: DERIVATION OF EQ. (7)

The Schrödinger equation with the Hamiltonian given by Eq. (1) reads

$$i\dot{A}_1(t)=\sum_k C_k(t)g_k^{(1)}e^{i(\omega_1c-\nu_k)t}+B(t)\Omega_1e^{i\Delta_1t},$$

$$i\dot{A}_2(t)=\sum_k C_k(t)g_k^{(2)}e^{i(\omega_2c-\nu_k)t}+B(t)\Omega_2e^{i\Delta_2t},$$

$$i\dot{B}(t)=A_1(t)\Omega_1^*e^{-i\Delta_1t}+A_2(t)\Omega_2^*e^{-i\Delta_2t}, \tag{A1}$$

and

$$i\dot{C}_k(t)=A_1(t)g_k^{(1)}e^{-i(\omega_1c-\nu_k)t}+A_2(t)g_k^{(2)}e^{-i(\omega_2c-\nu_k)t}. \tag{A2}$$

By formal integration of Eq. (A2), the probability amplitude $C_k(t)$ can be written as

$$C_k(t)=-ig_k^{(1)}\int_0^t dt'A_1(t')e^{-i(\omega_1c-\nu_k)t'} -ig_k^{(2)}\int_0^t dt'A_2(t')e^{-i(\omega_2c-\nu_k)t'}. \tag{A3}$$

After inserting Eq. (A3) into the first equation of Eq. (A1) we find

$$\dot{A}_1(t)=-\sum_k g_k^{(1)}e^{i(\omega_1-\nu_k)t}\left(g_k^{(1)}\int_0^t dt'A_1(t')e^{-i(\omega_1-\nu_k)t'} +g_k^{(2)}\int_0^t dt'A_2(t')e^{-i(\omega_2-\nu_k)t'}\right)-iB(t)\Omega_1e^{i\Delta_1t}. \tag{A4}$$

We now in a usual manner replace the summation over k by an integration, which gives

$$\begin{aligned}\dot{A}_1(t)=&-\frac{V}{(2\pi)^3}\int k^2dk\int d\phi\,\sin\theta\,d\theta\sum_\sigma\\ &\times\left(g_k^{(1)}g_k^{(1)}\int_0^t dt'A_1(t')e^{-i(\omega_{1c}-\nu_k)(t-t')}\right.\\ &\left.+g_k^{(1)}g_k^{(2)}\int_0^t dt'A_2(t')e^{-i(\omega_{2c}-\nu_k)(t-t')}e^{i\omega_{12}t'}\right)\\ &-iB(t)\Omega_1e^{i\Delta_1t}\\ =&-\frac{V}{(2\pi)^3}\int\left(\frac{\nu_k^2}{c^3}\right)d\nu_k\int d\phi\,\sin\theta\,d\theta\left[\left(\frac{|\mu_1|^2\sin^2\theta}{\hbar^2}\right)\right.\\ &\times\left(\frac{\hbar\nu_k}{2\epsilon_0V}\right)\int_0^t dt'A_1(t')e^{-i(\omega_{1c}-\nu_k)(t-t')}\\ &+\left(\frac{|\mu_1||\mu_2|p\sin^2\theta}{\hbar^2}\right)\left(\frac{\hbar\nu_k}{2\epsilon_0V}\right)\\ &\left.\times\int_0^t dt'A_2(t')e^{-i(\omega_{1c}-\nu_k)(t-t')}e^{i\omega_{12}t'}\right]\\ &-iB(t)\Omega_1e^{i\Delta_1t},\end{aligned} \tag{A5}$$

where the factors of $\sin^2\theta$ come from the summation over the polarization vectors, and the cross term has the factor of p, given by Eq. (6). Integration over ϕ and θ gives

$$\begin{aligned}\dot{A}_1(t) = & -\frac{1}{6\pi^2\epsilon_0\hbar c^3}\Bigg(|\mu_1|^2\int_0^\infty d\nu_k\nu_k^3 \\ & \times\int_0^t dt' A_1(t')e^{-i(\omega_{1c}-\nu_k)(t-t')} \\ & +|\mu_1||\mu_2|p\int_0^\infty d\nu_k\nu_k^3\int_0^t dt' A_2(t') \\ & \times e^{-i(\omega_{1c}-\nu_k)(t-t')}e^{i\omega_{12}t'}\Bigg)-iB(t)\Omega_1 e^{i\Delta_1 t} \\ \simeq & -\frac{1}{6\pi^2\epsilon_0\hbar c^3}\Bigg(|\mu_1|^2\omega_{1c}^3\int_{-\infty}^\infty d\nu_k \\ & \times\int_0^t dt' A_1(t')e^{-i(\omega_{1c}-\nu_k)(t-t')} \\ & +|\mu_1||\mu_2|p\,\omega_{1c}^3\int_{-\infty}^\infty d\nu_k\int_0^t dt' A_2(t') \\ & \times e^{-i(\omega_{1c}-\nu_k)(t-t')}e^{i\omega_{12}t'}\Bigg)-iB(t)\Omega_1 e^{i\Delta_1 t},\end{aligned} \tag{A6}$$

where we have used the Weisskopf-Wigner approximation in the last step. Hence we have

$$\begin{aligned}\dot{A}_1(t) = & -\frac{1}{6\pi^2\epsilon_0\hbar c^3}\Bigg(|\mu_1|^2\omega_{1c}^3\int_0^t dt' A_1(t')2\pi\delta(t-t') \\ & +|\mu_1||\mu_2|p\,\omega_{1c}^3\int_0^t dt' A_2(t')2\pi\delta(t-t')e^{i\omega_{12}t'}\Bigg) \\ & -iB(t)\Omega_1 e^{i\Delta_1 t} \\ \simeq & -\frac{\gamma_1}{2}A_1(t)-p\frac{\sqrt{\gamma_1\gamma_2}}{2}A_2(t)e^{i\omega_{12}t}-i\Omega_1 B(t)e^{i\Delta_1 t}.\end{aligned} \tag{A7}$$

[We replaced $(|\mu_1||\mu_2|\omega_{1c}^3)/(3\pi\epsilon_0\hbar c^3)$ by $\sqrt{\gamma_1\gamma_2}$, using the assumption that the upper level separation ω_{12} is much smaller than the optical frequencies ω_{1c} and ω_{2c}.] Similarly, we obtain

$$\dot{A}_2(t)=-p\frac{\sqrt{\gamma_1\gamma_2}}{2}A_1(t)e^{-i\omega_{12}t}-\frac{\gamma_2}{2}A_2(t)-i\Omega_2 B(t)e^{i\Delta_2 t}. \tag{A8}$$

After transforming $A_j(t)=a_j(t)e^{i\Delta_j t}$ we arrive at Eq. (4). Now, substituting Eqs. (7) into Eq. (A3), in the steady state we find

$$C_k(\infty)=\sum_{j=1}^{3}\frac{i(g_k^{(1)}\alpha_j+g_k^{(2)}\beta_j)}{-\lambda_j+i[\nu_k-(\omega_{bc}+\nu_0)]}. \tag{A9}$$

APPENDIX B: SPONTANEOUS EMISSION SPECTRUM

Spontaneous emission spectrum can be calculated as a Fourier transform of the two-time correlation function of electric field intensity:

$$S_{\mathbf{r}t}(\omega)=\frac{1}{2\pi}\int_0^\infty d\tau\, e^{-i\omega\tau}\langle\mathbf{E}^{(-)}(\mathbf{r},t+\tau)\cdot\mathbf{E}^{(+)}(\mathbf{r},t)\rangle+\text{c.c.}, \tag{B1}$$

where $\mathbf{E}^{(+)}(\mathbf{r},t)$ $[\mathbf{E}^{(-)}(\mathbf{r},t)]$ is the positive (negative) part of the electric field operator at time t and position $\mathbf{r}$. In the far-zone approximation this operator takes the form

$$\mathbf{E}^{+}(\mathbf{r},t)=\frac{\omega_0^2}{4\pi\epsilon_0 c^2 r}\hat{\mathbf{n}}\times[\hat{\mathbf{n}}\times\mathbf{P}^{(+)}(t-r/c)], \tag{B2}$$

where $\hat{\mathbf{n}}$ is a unit vector in the direction of observation, $\mathbf{P}^{(+)}$ is the positive part of the atomic polarization operator in the Heisenberg picture. We are interested in the spectrum of radiation emitted by the transitions $|a_1\rangle\rightarrow|c\rangle$ and $|a_2\rangle\rightarrow|c\rangle$ (Fig. 4). In this case $\omega_0=(\omega_{a_1c}+\omega_{a_2c})/2$ and

$$\begin{aligned}\mathbf{P}^{(-)}(t)&=\vec{\mu}_{1c}(|a_1\rangle\langle c|)^H(t)+\vec{\mu}_{2c}(|a_2\rangle\langle c|)^H(t),\\ \mathbf{P}^{(+)}(t)&=[\mathbf{P}^{(-)}]^\dagger,\end{aligned} \tag{B3}$$

where superscript H denotes that the operators are taken in the Heisenberg picture. Note that

$$\frac{\vec{\mu}_{1c}\cdot\vec{\mu}_{2c}}{\mu_{1c}\mu_{2c}}=p \tag{B4}$$

according to Eq. (6). From Eqs. (B2) and (B3) it follows that the spontaneous emission spectrum is proportional to the Fourier transform of the atomic two-time correlation function

$$\Gamma^{(1)}(t,\tau)=\langle\mathbf{P}^{(-)}(t+\tau)\cdot\mathbf{P}^{(+)}(t)\rangle. \tag{B5}$$

Calculation of Eq. (B5) involves a straightforward application of the quantum regression theorem [17]. This theorem states that if, for some operator $\hat{O}_i$,

$$\langle\hat{O}_i(t+\tau)\rangle=\sum_j c_j(t,\tau)\langle\hat{O}_j(t)\rangle, \tag{B6}$$

where $\{\hat{O}_j\}$ is a complete set of system operators and c_j's are c-number functions of time, then

$$\langle\hat{O}_i(t+\tau)\hat{O}_k(t)\rangle=\sum_j c_j(t,\tau)\langle\hat{O}_j(t)\hat{O}_k(t)\rangle. \tag{B7}$$

Rewrite the equations of motion (29) in the following vector form:

$$\frac{d}{dt}\hat{\psi}=L\hat{\psi}+\hat{\mathbf{C}}, \tag{B8}$$

where

$$\hat{\psi}=\begin{pmatrix}\rho_{ca_2}\\ \rho_{ca_1}\\ \rho_{cb}\\ \rho_{a_2c}\\ \rho_{a_2a_2}\\ \rho_{a_2a_1}\\ \rho_{a_2b}\\ \rho_{a_1c}\\ \rho_{a_1a_2}\\ \rho_{a_1a_1}\\ \rho_{a_1b}\\ \rho_{bc}\\ \rho_{ba_2}\\ \rho_{ba_1}\\ \rho_{bb}\end{pmatrix}, \tag{B9}$$

and $\hat{\mathbf{C}}$ is the inhomogeneous part arising from elimination of ρ_{cc} from Eqs. (29) by the normalization condition $\Sigma_i\rho_{ii}=1$. Explicit expressions for the matrix L and vector $\hat{\mathbf{C}}$ are too bulky to be presented here, but they can be easily derived from Eqs. (29). The solution of the system (B9) can be written as

$$\hat{\psi}(t)=\exp[L(t-t_0)]\hat{\psi}(t_0)+\int_{t_0}^{t}dt'\exp[L(t-t')]\hat{\mathbf{C}}, \tag{B10}$$

and the steady-state solution reads

$$\hat{\psi}(t=\infty)=-L^{-1}\hat{\mathbf{C}}. \tag{B11}$$

The first step in the application of the regression theorem is to find the one-time expectation value of the atomic polarization operator. The expectation values calculated in Schrödinger and Heisenberg pictures coincide, therefore

$$\begin{aligned}\langle\mathbf{P}^{(-)}(t+\tau)\rangle&=\vec{\mu}_{1c}\langle(|a_1\rangle\langle c|)^H(t+\tau)\rangle\\&\quad+\vec{\mu}_{2c}\langle(|a_2\rangle\langle c|)^H(t+\tau)\rangle\\&=\vec{\mu}_{1c}\rho^S_{ca_1}(t+\tau)+\vec{\mu}_{2c}\rho^S_{ca_2}(t+\tau)\\&=(\vec{\mu}_{1c}\psi_2+\vec{\mu}_{2c}\psi_1)e^{i(\nu_0+\omega_{bc})(t+\tau)}.\end{aligned} \tag{B12}$$

Superscripts H and S stand here for the Heisenberg and Schrödinger picture, respectively. Now in order to find Eq. (B5) we need to rewrite this expectation value in terms of the system operators $(|i\rangle\langle j|)^H$ and carry out the replacement

$$\langle(|i\rangle\langle j|)^H(t)\rangle\rightarrow\langle(|i\rangle\langle j|\mathbf{P}^{(+)})^H(t)\rangle. \tag{B13}$$

Taking the Fourier transform of the result, in the limit $t\rightarrow\infty$, we find the spontaneous emission spectrum in the form [6]

$$S(\omega)=\mathrm{Re}\hat{\Gamma}^{(1)}(\alpha)|_{\alpha=i\omega}, \tag{B14}$$

where

$$\begin{aligned}\hat{\Gamma}^{(1)}(\alpha)=\mu_{2c}^2\Big(&M_{11}(\alpha')\rho_{a_2a_2}(\infty)+M_{12}(\alpha')\rho_{a_2a_1}(\infty)+M_{13}(\alpha')\rho_{a_2b}(\infty)+\sum_j N_{1j}(\alpha')C_j\rho_{a_2c}(\infty)\Big)\\+p\mu_{2c}\mu_{1c}\Big(&M_{11}(\alpha')\rho_{a_1a_2}(\infty)+M_{12}(\alpha')\rho_{a_1a_1}(\infty)+M_{13}(\alpha')\rho_{a_1b}(\infty)+\sum_j N_{1j}(\alpha')C_j\rho_{a_1c}(\infty)\Big)\\+p\mu_{1c}\mu_{2c}\Big(&M_{21}(\alpha')\rho_{a_2a_2}(\infty)+M_{22}(\alpha')\rho_{a_2a_1}(\infty)+M_{23}(\alpha')\rho_{a_2b}(\infty)+\sum_j N_{2j}(\alpha')C_j\rho_{a_2c}(\infty)\Big)\\+\mu_{1c}^2\Big(&M_{21}(\alpha')\rho_{a_1a_2}(\infty)+M_{22}(\alpha')\rho_{a_1a_1}(\infty)+M_{23}(\alpha')\rho_{a_1b}(\infty)+\sum_j N_{2j}(\alpha')C_j\rho_{a_1c}(\infty)\Big),\end{aligned} \tag{B15}$$

with $\alpha'=\alpha-i(\nu_0+\omega_{bc})$. The matrices M and N are defined as

$$M(\alpha)=(\alpha I-L)^{-1},\quad N(\alpha)=L^{-1}(\alpha I-L)^{-1}, \tag{B16}$$

and I is a 15×15 unit matrix.

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