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Emission spectrum of coherently driven three-level atoms

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Analytical expressions for the emission spectrum and intensity correlation functions are derived for a V-type three-level atom driven by two strong, resonant coherent fields. It is found that coherent mixing of the atomic states by the strong fields modifies the atomic decay dynamics, which results in dramatical narrowing of spectral features predicted by Narducci, Oppo, and Scully [Opt. Commun. **75**, 111 (1990)].

Spontaneous decay of excited atoms can be changed by the modification of the electromagnetic vacuum reservoir. For example, spontaneous decay of excited atoms confined in an optical cavity can be enhanced if the cavity is tuned into resonance with the atomic transition frequency, or inhibited if the cavity is tuned off resonance.[1] In the strong atom-cavity coupling limit, exchange of energy between the atom and cavity field occurs, and the single peaked free-space atomic emission spectrum is modified so as to display two spectral features with a linewidth that can be up to a factor of 2 smaller than the atomic-decay linewidth.[2–4]

Quite differently, atomic-decay dynamics can also be modified by the strong field dressing of atoms. Such modification is manifest in the atomic fluorescence spectrum. Recently, Narducci, Oppo, and Scully[5] have studied the fluorescence spectrum scattered by a strong atomic transition sharing the same ground state with a weak transition in a V-type, three-level atom driven by two intense, near-resonant coherent fields.[6,7] Their numerical calculations show that the linewidths of the fluorescence spectrum scattered by the strong atomic transition can be less than the natural linewidth when the weak atomic transition is strongly saturated. This new effect has been experimentally demonstrated very recently.[8] In this paper, we show that the spectral narrowing can be motivated by an analysis based on the dressed-state picture.[7,9] We derive analytical expressions for the linewidths and intensities of scattered spectrum, and the intensity correlation functions in the strong field regime. We show that coherent mixing of the atomic states by the intense driving fields modifies the spectral content of the quantum fluctuations in the atomic dipole moment, and leads to the dramatical narrowing of spectral features.

Consider a V-configuration, three-level atom with the ground state $|0\rangle$, and excited states $|1\rangle$ and $|2\rangle$ as depicted in Fig. 1. The transition $|0\rangle \to |1\rangle$ of frequency ω_{01} is driven by a resonant coherent field of Rabi frequency Ω_1. The transition $|0\rangle \to |2\rangle$ of frequency ω_{02} is driven by another resonant coherent field of Rabi frequency Ω_2. γ_1 (γ_2) is the radiative decay rate of the excited state $|1\rangle$ ($|2\rangle$). Under the rotating-wave approximation, and in the interaction representation, the Hamiltonian is

$$H = \frac{\Omega_1}{2}\left[a_1 S_{10} + a_1^\dagger S_{01}\right] + \frac{\Omega_2}{2}\left[a_2 S_{20} + a_2^\dagger S_{02}\right] . \tag{1}$$

Here a_i ($a_i^\dagger$) is annihilation (creation) operator for the field i and $S_{ij} = |i\rangle\langle j|$.

The atom-field product states form manifolds according to their energy as shown in Fig. 2(a). For the manifold with energy $E(n_1,n_2) = n_1\omega_{01} + n_2\omega_{02}$, the three degenerate atom-field product states are $|0\rangle|n_1\rangle|n_2\rangle$, $|1\rangle|n_1-1\rangle|n_2\rangle$, and $|2\rangle|n_1\rangle|n_2-1\rangle$. Diagonalizing the Hamiltonian H in the basis set for the manifold N, we obtain three eigenvalues: $E = \Omega/2$, 0, and $-\Omega/2$ where $\Omega \equiv (\Omega_1^2 + \Omega_2^2)^{1/2}$. The corresponding eigenstates (dressed states) are

$$|1,n_1,n_2\rangle = \left[\frac{1}{2}\right]^{1/2}\left[|0\rangle|n_1\rangle|n_2\rangle - \frac{\Omega_1}{\Omega}|1\rangle|n_1-1\rangle|n_2\rangle - \frac{\Omega_2}{\Omega}|2\rangle|n_1\rangle|n_2-1\rangle\right] , \tag{2a}$$

$$|2,n_1,n_2\rangle = \frac{\Omega_2}{\Omega}|1\rangle n_1-1\rangle|n_2\rangle - \frac{\Omega_1}{\Omega}|2\rangle|n_1\rangle|n_2-1\rangle , \tag{2b}$$

$$|3,n_1,n_2\rangle = \left[\frac{1}{2}\right]^{1/2}\left[|0\rangle|n_1\rangle|n_2\rangle + \frac{\Omega_1}{\Omega}|1\rangle|n_1-1\rangle|n_2\rangle + \frac{\Omega_2}{\Omega}|2\rangle|n_1\rangle|n_2-1\rangle\right] . \tag{2c}$$

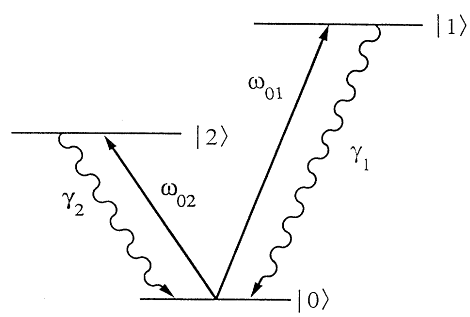

FIG. 1. Energy-level structures of a three-level atom. γ_1 (γ_2) is the spontaneous decay rate of state $|1\rangle$ ($|2\rangle$). ω_{01} (ω_{02}) is the $|0\rangle \to |1\rangle$ ($|0\rangle \to |2\rangle$) transition frequency.

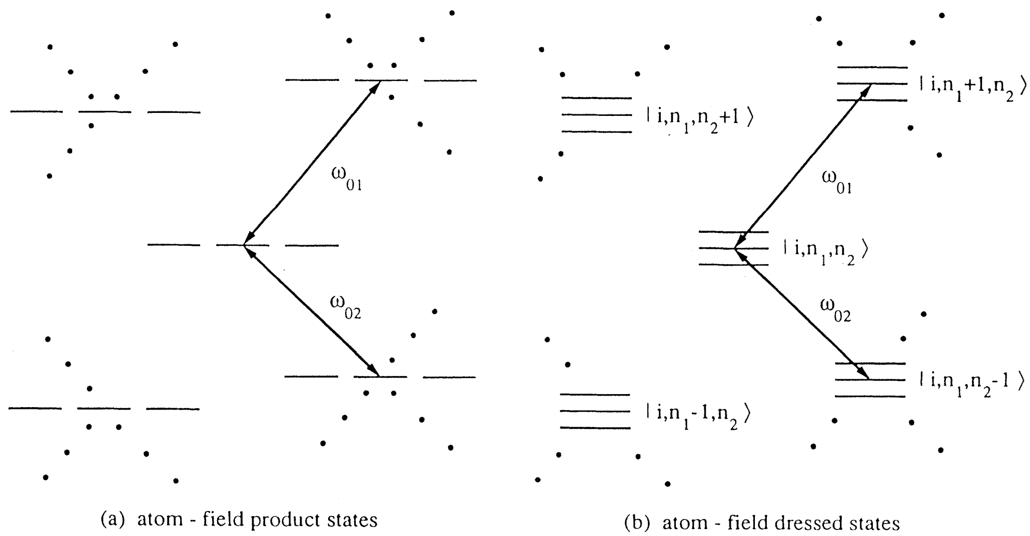

FIG. 2. Energy level structures of (a) atom-field product states with threefold degeneracy, and (b) atom-field dressed states. In (b), the degeneracy has been removed. Within each manifold, the spacing of neighboring levels is $\Omega/2$. There are two sets of transitions, centered at frequencies ω_{01} and ω_{02}, respectively.

The energy-level structures of the dressed states are depicted in Fig. 2(b). Using the technique developed by Cohen-Tannoudji and Raynaud,[9] we can write down the density matrix equations in the dressed-state representation. For the diagonal density-matrix element $\sigma_{ii}^0(n_1,n_2,t)$, which represents the population probability on the dressed-state $|i,n_1,n_2\rangle$, the equation of motion in the strong field regime, $\Omega \gg \gamma_1,\ \gamma_2$, is given by

$$\frac{d\sigma_{ii}^0(n_1,n_2,t)}{dt} = -\sum_j (\Gamma_{ij}+\Gamma'_{ij})\sigma_{ii}^0(n_1,n_2,t) + \sum_j \Gamma_{ji}\sigma_{jj}^0(n_1+1,n_2,t) + \sum_j \Gamma'_{ji}\sigma_{jj}^0(n_1,n_2+1,t) \ . \tag{3}$$

Here $\sigma_{ii}^0 = \langle i,n_1,n_2|\sigma(t)|i,n_1,n_2\rangle$ $(i,j=1,2,3)$. Γ_{ij} (Γ'_{ij}) is the spontaneous decay rate from the dressed-state $|i,n_1,n_2\rangle$ to $|j,n_1-1,n_2\rangle$ $(|j,n_1,n_2-1\rangle)$, and can be calculated by Fermi's golden rules.

For intense coherent driving fields, we have $\sigma_{ii}^0(n_1,n_2,t) \cong \sigma_{ii}^0(n_1+1,n_2,t) \cong \sigma_{ii}^0(n_1,n_2+1,t)$.[9] Solving Eq. (3) in the steady state, we obtain

$$\sum_{n_1,n_2} \sigma_{11}^0(n_1,n_2) = \sum_{n_1,n_2} \sigma_{33}^0(n_1,n_2) = \tfrac{1}{2} \ , \qquad \sum_{n_1,n_2} \sigma_{22}^0(n_1,n_2) = 0 \ , \tag{4}$$

where the normalization condition $[\sum_{(i,n_1,n_2)} \sigma_{ii}^0(n_1,n_2) = 1]$ has been assumed. It is seen that in the steady state, the atomic population is equally distributed among the dressed states $|1,n_1,n_2\rangle$ and $|3,n_1,n_2\rangle$, but there is no population in the dressed states $|2,n_1,n_2\rangle$. This is not surprising if one notes Eq. (2b) that $|2,n_1,n_2\rangle$ contains the superpositions of excited atomic states $|1\rangle$ and $|2\rangle$ only. In the dressed-state picture, spontaneous decay corresponds to transitions from the dressed states of manifold N to the dressed states of manifold $N-1$. For nonvanishing transition matrix elements, the atom must change its state from $|1\rangle$ or $|2\rangle$ to $|0\rangle$ with no accompanying change in the number of photons for the two driving coherent fields. Such spontaneous transitions from the upper dressed states to $|2,n_1,n_2\rangle$ are forbidden. So any initial population in the dressed states $|2,n_1,n_2\rangle$ decays away quickly, and in the steady state, there is no population in the dressed states $|2,n_1,n_2\rangle$. Projecting the dressed state population into the atom-field product states, we find that in the steady state, the population distributions are

$$P_0 = \tfrac{1}{2}, \quad P_1 = \frac{\Omega_1^2}{2\Omega^2} ,$$

and

$$P_2 = \frac{\Omega_2^2}{2\Omega^2} , \tag{5}$$

where P_j is the population probability on the atomic state $|j\rangle$. Equation (5) shows that when $\Omega_1 \gg \Omega_2$, the population probability for the state $|2\rangle$ is negligible. The simple physical reason for such population trapping is due to ac Stark shift of the ground state $|0\rangle$ induced by the field of large Rabi frequency Ω_1. The large detuning Ω_1 for the Autler-Townes doublet transition prevents the atom from reaching the state $|2\rangle$. For $\Omega_2 \gg \Omega_1$, the situation is reversed.

We next consider the off-diagonal matrix elements which are related to the transition dipole moments. There are two sets of transitions; $|i,n_1,n_2\rangle \rightarrow |j,n_1-1,n_2\rangle$, and $|i,n_1,n_2\rangle \rightarrow |j,n_1,n_2-1\rangle$. Each set contains three-frequency components located at $\omega_{01}, \omega_{01} \pm \Omega$, and $\omega_{02}, \omega_{02} \pm \Omega$, respectively. For the transitions at frequencies ω_{01} and $\omega_{01} \pm \Omega$, the corresponding density-matrix elements are

$$\sigma_{ij}(n_1, n_2, t) = \langle i, n_1, n_2 | \sigma | j, n_1 - 1, n_2 \rangle, \quad i,j = 1,3 . \tag{6}$$

Using the secular approximation and including the cascade interference contributions,[9] the equations of motion for the components of the dipole moment, $\langle D(\omega_{01}) \rangle$ $[\langle D(\omega_{01} \pm \Omega) \rangle]$, evolving at frequency ω_{01} $(\omega_{01} \pm \Omega)$ are given by

$$\frac{d\langle D(\omega_{01}) \rangle}{dt} = -\left[i\omega_{01} + \frac{\Gamma}{2} \right] \langle D(\omega_{01}) \rangle , \tag{7a}$$

$$\frac{d\langle D(\omega_{01} \pm \Omega) \rangle}{dt} = -\left[i(\omega_{01} \pm \Omega) + \frac{3\Gamma}{4} \right] \langle D(\omega_{01} \pm \Omega) \rangle , \tag{7b}$$

where

$$\Gamma = \frac{\Omega_1^2 \gamma_1 + \Omega_2^2 \gamma_2}{\Omega^2} . \tag{8}$$

The equations of motion for the components of dipole moments evolving at frequencies $\omega_{02} \pm \Omega$ (ω_{02}) are found to be the same as Eq. (7b) [7(a)]. Equation (7) shows that the dipole moments decay exponentially, and the corresponding spectral profiles are Lorentzian. The fluorescence intensity at frequency ω_{01} is given by

$$I(\omega_{01}) = T\Gamma'_{11} \sum_{n_1, n_2} [\sigma_{11}(n_1, n_2) + \sigma_{33}(n_1, n_2)] = \frac{T\Omega_1^2 \gamma_1}{4\Omega^2} , \tag{9}$$

where T is the signal average time. Similar calculations can be performed to determine the fluorescence intensities at frequencies $\omega_{01} \pm \Omega$, ω_{02}, and $\omega_{02} \pm \Omega$. The final expression for the spectral distribution $S(\omega - \omega_{01})$ of the fluorescence light near ω_{01} is

$$\frac{S(\omega - \omega_{01})}{T} = \frac{\Omega_1^2 \gamma_1}{4\Omega^2} \left[\frac{\Gamma}{(\omega - \omega_{01})^2 + \Gamma^2/4} + \frac{3\Gamma/2}{(\omega - \omega_{01} - \Omega)^2 + (3\Gamma/4)^2} + \frac{3\Gamma/2}{(\omega - \omega_{01} + \Omega)^2 + (3\Gamma/4)^2} \right] , \tag{10a}$$

and the spectral distribution $S(\omega - \omega_{02})$ near ω_{02} is

$$\frac{S(\omega - \omega_{02})}{T} = \frac{\Omega_2^2 \gamma_2}{4\Omega^2} \left[\frac{\Gamma}{(\omega - \omega_{02})^2 + \Gamma^2/4} + \frac{3\Gamma/2}{(\omega - \omega_{02} - \Omega)^2 + (3\Gamma/4)^2} + \frac{3\Gamma/2}{(\omega - \omega_{02} + \Omega)^2 + (3\Gamma/4)^2} \right] . \tag{10b}$$

For on-resonance excitation and in the strong field regime, the fluorescence spectrum of a V-configuration, three-level atom driven by two coherent fields consists of two triplet spectral features centered at frequencies ω_{01} and ω_{02} with sidebands separated from the central peak by an effective Rabi frequency Ω. For each triplet, the height for the central peak is three times that of the sideband peaks, and the linewidth for the sideband peaks is 1.5 times broader than the central peak linewidth Γ which is the weighted average of the decay rates γ_1 and γ_2. This is quite similar to the fluorescence spectrum of a two-level atom driven by an intense monochromatic field.[10-13] For the special case of $\gamma_1 \gg \gamma_2$, $|0\rangle \rightarrow |1\rangle$ is a strong transition, while $|0\rangle \rightarrow |2\rangle$ is a weak transition. Since $\gamma_1 \geq \Gamma \geq \gamma_2$, the spectrum, $S(\omega - \omega_{01})$, scattered by the strong transition demonstrates subnatural linewidth behavior while the spectrum, $S(\omega - \omega_{02})$, scattered by the weak transition demonstrates supernatural linewidth behavior. This indicates that spectral distributions of quantum fluctuations of the atomic dipole moment have been modified. The quantum fluctuations near frequency ω_{01} have been reduced at the expenses of increased quantum fluctuations near frequency ω_{02}. When $\Omega_2 \gg \Omega_1$, the fluorescent light scattered by the strong transition is dramatically reduced, and the linewidth approaches that of the weak transition. The spectrum scattered by the strong transition collapses. This is consistent with the prediction of Narducci, Oppo, and Scully.[5] Inspecting the content of the dressed states expressed by Eq. (2), it is clear that the reduction of the spectral linewidth is due to the Rabi-frequency-dependent weight of the slow-decay state $|2\rangle$ in the equally occupied dressed states $|1, n_1, n_2\rangle$ and $|3, n_1, n_2\rangle$.

In order to gain more physical insight into this problem, we next analyze the intensity correlation functions in the strong-field regime. It is convenient to calculate the intensity correlation functions[14] if we transform three atomic states into a new basis set $|0\rangle$, $|a\rangle = (\Omega_1/\Omega)|1\rangle + (\Omega_2/\Omega|2\rangle$, and $|b\rangle = (\Omega_2/\Omega|1\rangle - (\Omega_1/\Omega)|2\rangle$ as shown in Eq. (2). It is easy to check that the ground state $|0\rangle$ is coupled to $|a\rangle$ only. The state $|b\rangle$ is decoupled from $|0\rangle$ and $|a\rangle$, and cannot trap population because it is initially unpopulated and remains inaccessible. In the strong-field regime, we know from Eq. (10) that the effective decay rate for the excited state $|a\rangle$ is Γ. So the atom is cycling between the states $|0\rangle$ and $|a\rangle$ with an effective two-level Rabi frequency Ω, and damped at a rate Γ. We can immediately write down the intensity correlation function for this effective two-level system:[15]

$$g_{aa}(t)=1-\exp(3\Gamma\tau/4)\cos(\Omega\tau)\ . \quad (11)$$

Here $g_{aa}(\tau)$ is proportional to the probability per unit time of detecting a photon from the $|1\rangle\rightarrow|0\rangle$ or $|2\rangle\rightarrow|0\rangle$ transition at a later time τ after such a photon has been emitted at $\tau=0$. Let $g_{ij}(\tau)$ $(i,j=1,2)$ represent the probability per unit time of detecting a photon from the $|i\rangle\rightarrow|0\rangle$ transition at a later time τ after a photon from the $|j\rangle\rightarrow|0\rangle$ transition has been emitted. Projecting $g_{aa}(\tau)$ into the basis states $|1\rangle$ and $|2\rangle$, we found that

$$g_{11}(\tau)=g_{12}(\tau)=\frac{\Omega_1^2}{\Omega^2}[1-\exp(3\Gamma\tau/4)\cos\Omega\tau]\ , \quad (12a)$$

$$g_{22}(\tau)=g_{21}(\tau)=\frac{\Omega_2^2}{\Omega^2}[1-\exp(3\Gamma\tau/4)\cos\Omega\tau]\ . \quad (12b)$$

As expected, Eqs. (11), (12a), and (12b) demonstrate antibunching behavior. When $\gamma_1 \gg \gamma_2$, and $\Omega_1 \gg \Omega_2$, from Eq. (7) we have $\Gamma\cong\gamma_1$, and $g_{11}(\tau)\cong g_{aa}(\tau)$, $g_{22}(\tau)\cong 0$. The atom emits photons spontaneously with a rate Γ through the $|1\rangle\rightarrow|0\rangle$ transition with very little chance of being interrupted by a transition to the state $|2\rangle$ due to the large detuning Ω_1 induced by the ac Stark shift. However, as Ω_2 increases, the transition probability from $|0\rangle\rightarrow|2\rangle$ becomes larger. The fluorescence from $|1\rangle\rightarrow|0\rangle$ will be interrupted. This is consistent with the study of quantum jumps.[16–20] When $\Omega_2 \gg \Omega_1$, then $\Gamma\cong\gamma_2$, and $g_{11}(\tau)\cong 0, g_{22}(\tau)\cong g_{aa}(\tau)$, the atom cycles between $|0\rangle$ and $|2\rangle$, and is rarely interrupted by a transition to the state $|1\rangle$. Due to the reduction of the effective decay rate Γ, the emission of the fluorescence photons is dramatically reduced and the fluorescence spectrum collapses.

In conclusion, we have derived analytical expressions for the fluorescence spectrum and intensity correlation functions of a V-configuration, three-level atom driven by two intense coherent fields. Our results demonstrate that mixing of the atomic states by the intense coherent fields modifies the atomic decay dynamics, and leads to dramatical spectral narrowing in the emission spectrum.

Note added After submission of this manuscript, a paper by Narducci *et al.* appeared in Phys. Rev. A **42**, 1630 (1990) in which they gave a more elaborate analysis by solving density-matrix equations.

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