Resonance fluorescence in atomic coherent systems: Spectral features.

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Abstract

We study resonance fluorescence in a four level ladder system and illustrate some novel features due to quantum interference and atomic coherence effects. We find that under three photon resonant conditions, in some region of the parameter space of the rabi frequencies $\Omega_1, \Omega_2, \Omega_3$, emission is dominantly by the level 4 at the line center even though there is an almost equal distribution of populations in all the levels. As one increases $\Omega_3$ with $\Omega_1$ and $\Omega_2$ held fixed, the four level system 'dynamically collapses' to a two level system. The steady state populations and the the resonance fluorescence from all the levels provide adequate evidence to this effect.

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Four-level atomic systems with three photon resonant interaction, lead to very different phenomena than those due to two photon effects in three level systems [1], as revealed by the absorption characteristics in these systems [2]. Such characteristics have been attributed to quantum interference (QI) and atomic coherent (ACh) effects. The interplay amongst the driving field strengths and detunings control the nature of QI and ACh mechanisms which in turn form a powerful tool for controlling the spontaneous emission and fluorescence from various levels of the atomic system [3,4]. To recapitulate a few known results, while the occurrence of dark resonances [1] and electromagnetically induced transparency (EIT) [5] were typical effects expected in two photon processes, in 4-level systems interacting with three driving fields, narrow absorption at the line center was reported [2]. Other four level studies have revealed as varied a phenomena like photon switching [6], two photon absorption and inhibition [7] and other interference effects [8]. Effect of QI on the suppression of spontaneous emission [9], coherent control of polarization of light [10] have been demonstrated experimentally and the effect of relative phases of the lasers [11] has also been reported.

In this communication we study the resonance fluorescence from various levels of a 4-level atomic system in the parameter space spanned by the rabi frequencies of the driving fields, \((\Omega_1, \Omega_2, \Omega_3)\). Apart from the already known features like supression of emission, narrowing of emission line and Mollow splitting of lines at high driving intensities, we present novel features exhibited in certain regions in this parameter space. For low [12] ground state excitation energies, we find that the four level system 'dynamically collapses' to a two level system for certain values of \(\Omega_2\) and \(\Omega_3\). That is, two of the levels get 'dynamically decoupled' from the rest of the system. Further we identify the region in the parameter space where we can control the fluorescence from the 4th level. A look at the steady state populations in these regions further throws light on the nature of the interferences. This rich variety of phenomena may find possible applications in the areas of high resolution, control of fluorescence at definite frequencies, quantum computing [13] and population transfer [14].

As already mentioned, the model we consider is a 4-level ladder system driven by three resonant fields. Here (see fig 1), 1-2, 2-3 and 3-4 are the allowed dipole transitions. We
take the decay constants of the system to be very similar to that of the Rubidium system of reference [3]. The energy level separations are denoted by $\omega_{21}, \omega_{32}$ and $\omega_{43}$ respectively. We sketch the outlines of the calculation here, the details will be provided elsewhere. In setting up the Hamiltonian for the four level system, we treat the driving fields classically and write it in the standard form

$$H = \sum_{i=1}^{4} E_i a_i^\dagger a_i + \sum_{j=1}^{3} \hbar \omega_j (e^{-i\omega_j t} a_j^\dagger a_j + \text{h.c.})$$

(1)

where $\Omega_j = \frac{\mu_{j+1} E_i}{\hbar}$ is the Rabi frequency of the coupling fields, $a_i^\dagger, a_i$ describe the creation and annihilation of the electrons in the respective levels $i$, and $E_i$ are the energy levels of the atomic system in the absence of any external field. We follow the approach of Agarwal [15] and Narducci et al. [16], for obtaining the fluorescence spectrum. We employ the equation of motion of the density matrix of the atomic system in the interaction representation,

$$\frac{\partial \rho}{\partial t} = -i \frac{\hbar}{\mu} [H_I, \rho] + \mathcal{L}_{\text{irrev}}$$

(2)

where $H_I$ is the Hamiltonian in the interaction representation, and $\mathcal{L}_{\text{irrev}}$ is the irreversible part of the Liouville operator described by the master equation given in reference [17].

The density matrix equations (2) are rewritten in the rotating wave approximation as

$$\begin{align}
\frac{\partial \rho_{12}}{\partial t} &= (i\Delta_1 - \Gamma_2/2)\rho_{12} - i\Omega_1 (\rho_{22} - \rho_{11}) + i\Omega_2 \rho_{13} \\
\frac{\partial \rho_{23}}{\partial t} &= (i\Delta_2 - (\Gamma_2 + \Gamma_3)/2)\rho_{23} - i\Omega_1 \rho_{13} - i\Omega_2 (\rho_{33} - \rho_{22}) + i\Omega_3 \rho_{24} \\
\frac{\partial \rho_{34}}{\partial t} &= (i\Delta_3 - (\Gamma_3 + \Gamma_4)/2)\rho_{34} - i\Omega_2 \rho_{24} - i\Omega_3 (\rho_{44} - \rho_{33}) \\
\frac{\partial \rho_{13}}{\partial t} &= (i(\Delta_1 + \Delta_2) - \Gamma_3/2)\rho_{13} - i\Omega_1 \rho_{23} - i\Omega_2 \rho_{12} + i\Omega_3 \rho_{14} \\
\frac{\partial \rho_{14}}{\partial t} &= (i(\Delta_1 + \Delta_2 + \Delta_3) - \Gamma_4/2)\rho_{14} - i\Omega_1 \rho_{24} + i\Omega_3 \rho_{13} \\
\frac{\partial \rho_{24}}{\partial t} &= (i(\Delta_2 + \Delta_3) - (\Gamma_2 + \Gamma_4)/2)\rho_{24} - i\Omega_1 \rho_{14} - i\Omega_2 \rho_{34} + i\Omega_3 \rho_{23} \\
\frac{\partial \rho_{22}}{\partial t} &= -\Gamma_2 \rho_{22} + i\Omega_1 (\rho_{21} - \rho_{12}) + i\Omega_2 (\rho_{23} - \rho_{32}) + \gamma_{23} \rho_{33} + \gamma_{24} \rho_{44} \\
\frac{\partial \rho_{33}}{\partial t} &= -\Gamma_3 \rho_{33} + i\Omega_3 (\rho_{34} - \rho_{43}) - i\Omega_2 (\rho_{23} - \rho_{32}) + \gamma_{34} \rho_{44} \\
\frac{\partial \rho_{44}}{\partial t} &= -\Gamma_4 \rho_{44} - i\Omega_4 (\rho_{34} - \rho_{43})
\end{align}$$

(3)
where \( \gamma_{ik} \) are the transition rates from the k to ith levels, \( \Gamma_i \) are the decay constants of the levels i and \( \Delta_i = \omega_{i,i+1} - \omega_i \) are the laser detunings (\( \bar{\rho}_{ij} = \bar{\rho}_{ji} \) and \( \text{Tr}\bar{\rho} = 1 \)). The level shifts and pure phase relaxations have been ignored in our analysis of the spectrum. We also ignore the contribution from \( \gamma_{ik} \) for \( i > k \) which corresponds to an excitation to a higher level. In other words we assume that the energy level separations to be far greater than the thermal excitation energy. Since it is easier to evaluate the solutions in the Laplace space, we recast (3) in a more compact form as

\[
\frac{d\psi}{dt} = M\psi + C \tag{4}
\]

where we identify \( \psi_1 = \bar{\rho}_{12}, \psi_2 = \bar{\rho}_{23}, \psi_3 = \bar{\rho}_{34}, \psi_4 = \bar{\rho}_{13}, \psi_5 = \bar{\rho}_{14}, \psi_6 = \bar{\rho}_{24}, \psi_7 = \bar{\rho}_{22}, \psi_8 = \bar{\rho}_{33}, \psi_9 = \bar{\rho}_{44}, \) and \( \psi_{i+9} = \psi_i^* \). M is a \((15 \times 15)\) matrix, and \( C \) is the inhomogenous part with the elements \( C_1 = C_{10}^* = -i\Omega_1 \) and the rest being zero. By taking the Laplace transform of the above equation one obtains the solution in the simple form

\[
\bar{\psi}_i(z) = \sum_j M_{ij}(z) \psi_j(\tau_0) + \frac{1}{z} \sum_j M_{ij}(z) C_j. \tag{5}
\]

where \( \bar{\psi}_i(z) \) is the Laplace transform of \( \psi(t) \) and \( M = (z - M)^{-1} \). The steady state solution is obtained by taking the limit \( z \to 0 (t \to \infty) \) in the above equation.

The fluorescence spectrum is proportional to the steady state correlation function of the scattered field, \( \lim_{t \to \infty} < E^-(r,t+\tau)E^+(r,t) > \), where \( E^-, E^+ \) correspond to the positive and negative frequency part of the scattered radiation. Since the source field operators and the atomic polarization operators are directly proportional to each other \([18]\), we can express the fluorescence spectrum in terms of the atomic correlation functions as

\[
\Gamma^1(z) = \int_0^\infty d\tau_1 \lim_{t \to \infty} < \mathcal{P}^\dagger(t + \tau_1)\mathcal{P}(t) > e^{-i\omega t}. \tag{6}
\]

where the atomic polarization operator \( \mathcal{P} \) is given by

\[
\mathcal{P}^\dagger = \sum_{i=1}^3 \mu_{i,i+1} \hat{a}_{i+1}^\dagger \hat{a}_i \tag{7}.
\]

where \( \mu_{ij} \) are modulus of the induced dipole moments.
Applying the quantum regression theorem the steady state spectrum is obtained, after some algebra, as

\[
< \mathcal{P}^\dagger(z)\mathcal{P}(\infty) > = \sum_{i=1}^{3} (\mu_{i,i+1}^2 (\mathcal{M}_{ii}(z_i)\psi_{i+6}(\infty)) + \sum_j \frac{1}{z_i} \mathcal{M}_{ij} C_j \psi_i^*(\infty)) \tag{8}
\]

where we have dropped all the rapidly oscillating terms. Here \( z_i = z - i\omega_i, z = i\omega \). Thus the spectrum has three distinct parts corresponding to the center frequencies at \( \omega_i, i = 1, 3 \).

Subtracting the delta function contributions which corresponds to the coherent part of the spectrum, we get the real part of the incoherent contribution to be

\[
\Gamma_{\text{incoh}}^{(1)} = \sum_{i=1}^{3} (\mu_{i,i+1}^2 (\mathcal{M}_{ii}(z_i)\psi_{i+6}(\infty)) + \sum_j \frac{1}{z_i} \mathcal{N}_{ij}(z_i) C_j \psi_i^*(\infty)) \tag{9}
\]

where \( \mathcal{N} = M^{-1} \mathcal{M} \). With this expression for the correlation function, we proceed to numerically analyse the spectral features in the various domains of the parameter space.

We shall assume, for the sake of convenience, that the induced dipole moments of all the three transitions are equal and look at the emission spectrum at three photon resonance \( \Delta_i = 0, i = 1, 3 \). Further we write all the parameters \( \Delta_i, \Gamma_i, \gamma_{ik} \) and \( \Omega_i \) in terms of \( \gamma \) and set \( \gamma = 1 \). The emission spectrum of the 4 level system is presented in two regions of the parameter space centered around the points, \((7, 4, 1), (7, 4, 50)\) (fig2(a)) and \((7, 4, 50)\) (fig2(b)). They show some new and interesting features. We find in the former region that, at the line center, emission is dominated by the 4 \( \rightarrow \) 3 transition and the other two emissions are relatively small. A look at the variation of the peak intensities as one transits from low to high \( \Omega_3 \) indicates (fig4) that the emission of the 4 \( \rightarrow \) 3 transition peaks around \( 2\gamma \) and thereafter falls rapidly. Emission for the 2 \( \rightarrow \) 1 and 3 \( \rightarrow \) 2 is rather weak in this region. A look at the population distributions (fig3(a)), at this peak value, however reveals that at zero detuning , \( \rho_{22}(\infty) \approx \rho_{44}(\infty) \), and \( \rho_{33}(\infty) \) is larger by 10 %. This implies that in this region, the spectral features are controlled more by the atomic coherent effects than the population dynamics.

We now move over to the other region around \((7, 4, 50)\) in the parameter space and the features are given in fig3. The fluorescence in this region is dominated by level 2 and there...
is a total suppression of emission from levels 3 and 4 (see fig2(b) ). The populations in the levels (fig 3(b)) also seem to indicate that the system, now, effectively behaves like a two level system driven by a single resonant field. This is further substantiated by results in fig4 which shows the saturation of the peak intensity of emission from level 2 with increase in \( \Omega_3 \), while the peak intensities corresponding to levels 3 and 4 show a rapid fall and approaches zero. This can be understood heuristically by arguing that as \( \Omega_3 \) is increased, the two photon induced coherence between the levels 2 and 4 creates an 'EIT' like situation, which inhibits absorption from level 2 and the population is forced to reside in the level 2.

To conclude, we have seen that various competing atomic coherent and quantum interference effects are responsible for the control of fluorescence from various levels which might provide a powerful tool for such switching mechanisms. We have also seen the possibility of a four level system behaving effectively like a two level system. This feature might possibly be exploited in suppressing multi-photon processes when there is a need to work with high ground state excitation energies. A study of the photon statistics and collective effects in such systems, which will be dealt with elsewhere, should reveal any difference in the usual two level system and the 'dynamical' two level system.
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Figure Captions

Figure 1:

Schematic energy level diagram of the four level system. \(\omega_1, \omega_2\) and \(\omega_3\) are the frequencies of the driving fields.

Figure 2:

Power spectrum of the radiation emitted in the regions of driving field strengths a) (7,4,1) and b) (7,4,50). The parameters are \(\Gamma_2 = 6.0, \Gamma_3 = \Gamma_4 = 1.0, \gamma_{23} = \gamma_{34} = 1.0,\) and \(\gamma_{24} = 0.0,\) \(\Delta_1 = \Delta_2 = \Delta_3 = 0.\) For easy comparison we have superposed the centers of all the emission lines.

Figure 3:

Steady state populations in the levels 2,3 and 4 corresponding to the regions in the parameter space a) (7,4,1) and b) (7,4,50) as a function of \(\Delta_1.\) The rest of the parameters are the same as in fig2.

Figure 4:

Peak intensity of the emitted field corresponding to the transitions 4 \(\rightarrow\) 3 , 3 \(\rightarrow\) 2 and 2 \(\rightarrow\) 1 as a function of the driving field strength \(\Omega_3.\) The rest of the parameters same as in fig2(a).
power spectrum

fig 2a
Power spectrum

fig2b
steady state populations

\[ \Delta \text{fig3a} \]
steady state populations

\[ \Delta_{\text{fig3b}} \]
