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Electromagnetic field induced modification of branching ratios for emission in structured vacuum

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Abstract. We report a fundamental effect of the electromagnetic field induced modification of the branching ratios for emission into several final states. The modifications are especially significant if the vacuum into which the atom is radiating has a finite spectral width comparable with the separation of the final states. This is easily realizable in cavity QED. Further, our results are quite generic and are applicable to any system interacting with a structured reservoir.

Mollow discovered in 1969 how the spectral characteristics of the radiation emitted by a system are modified quite significantly due to a coherent field driving the system [1]. Such spectral modifications have been the subject of extensive experimental and theoretical studies [2] and were explained neatly in terms of the dressed state picture [3]. The work of Mollow was extended to the case of emission in a cavity [4, 5]. It was further found especially in the context of multilevel systems that the driving fields can produce well defined interference minimum in the spectrum [6]–[9]. Such minimum is usually interpreted in terms of the interferences produced by different dressed state emissions. Very often this interference is also referred to as the quenching of spontaneous emission [10]. A related question is—what is the effect of driving fields on branching ratios in emission to multiple states. In a different class of experiments Suckewer and co-workers [11] found definite evidence of external field induced changes in branching ratios. Their experimental finding has been rather difficult to explain due to complicated nature of the laser plasma used in the experiment. It is therefore desirable to look for simpler systems where one can analyze how external fields could affect branching ratios.

In this paper, we analyze a cavity QED system to highlight the field induced modification of the branching ratios. In view of the enormous progress made in the context of cavity QED

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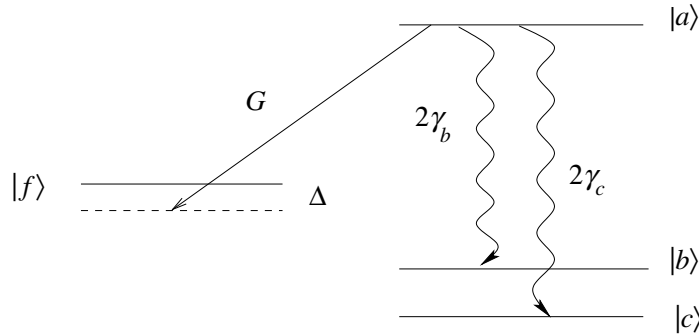


Figure 1. Schematic diagram of a four-level atom.

[12]–[14] such findings are within the reach of present experiments. Our analysis also suggests that changes in the branching ratios are notable if one works in a regime where separation between the two final states is more than the width of the vacuum into which the system is radiating. Thus some of the dispersive effects are also important. These conditions are easy to satisfy in the context of cavity QED systems. Clearly if the spectral width of the vacuum is very large as in free space then one would not expect any significant change in branching ratios. Although the results that we present are specifically in the context of QED, they can be generally applicable to a much wider class of systems. For example, we can consider the interaction of any system with a structured reservoir of finite width [15]–[18]. Further, the analysis would also be applicable to nano environments which lead to significant spectral modifications [19].

We start by illustrating the nature of the problem in the context of a simple four level model shown schematically in figure 1. Let us consider the decay of the excited state $|a\rangle$ to two lower levels $|b\rangle$ and $|c\rangle$. Let us also assume that the excited state $|a\rangle$ is connected to another level $|f\rangle$ by a laser field with Rabi frequency $2G$. We can write all the density matrix equations for such a system clearly the population of the states $|b\rangle$ and $|c\rangle$ changes according to

$$\dot{\rho}_{bb} = 2\gamma_b \rho_{aa}, \quad \dot{\rho}_{cc} = 2\gamma_c \rho_{aa} \quad (1)$$

and thus

$$\frac{\rho_{bb}(t \rightarrow \infty)}{\rho_{cc}(t \rightarrow \infty)} = \frac{\gamma_b}{\gamma_c}. \quad (2)$$

The ratio is independent of the external field applied on the transition $|a\rangle \rightarrow |f\rangle$. However, the spectral distribution itself would depend on the external field. We will see later that the result of equation (2) is intimately connected to the Markov approximation, which is used in deriving equation (1). The validity of the Markov approximation requires that the spectral width of the vacuum be much greater than the Rabi frequency of the field that drives the transition $|a\rangle \rightarrow |f\rangle$. This condition is satisfied in free space. Therefore the branching ratios can be affected by considering a vacuum whose width is comparable with the applied external field or even less. This is also very relevant to the question of the interaction of multilevel systems with engineered reservoirs [15]–[18]. Further, we know from the early work of Purcell [20] that the spontaneous emission in a cavity is considerably modified [21] because the spectral width of the available mode is much smaller than in free space. Thus the question of the modification of branching ratios can be settled by considering emission in a cavity.

To be specific, we consider the case of a Rydberg atom in a cavity which has a frequency ω_c , which we can tune somewhere between the levels $|b\rangle$ and $|c\rangle$. The transition $|a\rangle \rightarrow |f\rangle$ is not resonant with the cavity and is driven by the laser field of frequency ω_l . We now present a first principle calculation of the branching ratios. The Hamiltonian for the system in the interaction picture is given by,

$$\hat{\mathbf{H}} = G|a\rangle\langle f|e^{-i\Delta t} + \sum_{\omega} g_{b\omega}|a\rangle\langle b|e^{-i\omega_c t+i\omega_{ab}t}\hat{a}_{\omega} + \sum_{\omega} g_{c\omega}|a\rangle\langle c|e^{-i\omega_c t+i\omega_{ac}t}\hat{a}_{\omega} + \text{h.c.}, \quad (3)$$

$$\Delta = \omega_l - \omega_{af}.$$

Here, ω_{ab} and ω_{ac} represent, respectively, the frequencies of the two transitions. The cavity field is represented by the annihilation and creation operators \hat{a}_{ω} and $\hat{a}_{\omega}^{\dagger}$. The coupling constants are denoted by G , $g_{b\omega}$ and $g_{c\omega}$. The sum over ω in equation (3) would be converted into an integral over the spectral width of the single mode cavity. The wavefunction of the system of the cavity field and the atom can be written as,

$$|\psi\rangle = \alpha|a, 0\rangle + \beta|f, 0\rangle + \sum_{\omega} b_{\omega}|b, \omega\rangle + \sum_{\omega} c_{\omega}|c, \omega\rangle. \quad (4)$$

Here, ω denotes the state of the cavity with one photon at the frequency ω . Various amplitudes can be obtained by substituting equations (3) and (4) into the Schrödinger equation. We work with Laplace transforms. The transform $\hat{\alpha}(z)$ of the excited state amplitude $\alpha(t)$ is given by,

$$\left\{ z + \frac{G^2}{z+i\Delta} + \sum_{\omega} \frac{|g_{b\omega}|^2}{(z+i\omega-i\omega_{ab})} + \sum_{\omega} \frac{|g_{c\omega}|^2}{(z+i\omega-i\omega_{ac})} \right\} \hat{\alpha}(z) = 1. \quad (5)$$

Further, the amplitudes of the final states $|b\rangle$ and $|c\rangle$ are found to be

$$\hat{b}_{\omega}(z) = -i \frac{g_{b\omega}^*}{z} \hat{\alpha}(z+i\omega_{ab}-i\omega), \quad (6)$$

$$\hat{c}_{\omega}(z) = -i \frac{g_{c\omega}^*}{z} \hat{\alpha}(z+i\omega_{ac}-i\omega). \quad (7)$$

We now convert the sums in equation (5) to integrals using,

$$\sum_{\omega} \frac{|g_{b\omega}|^2}{(z+i\omega-i\omega_{ab})} \rightarrow \int d\omega \left[\frac{\kappa/\pi}{(\omega-\omega_c)^2 + \kappa^2} \right] \frac{|g_b|^2}{(z+i\omega-i\omega_{ab})} = \frac{|g_b|^2}{(z+\kappa-i\delta_b)}, \quad (8)$$

where $\delta_b = \omega_{ab} - \omega_c$. Finally, one can prove that the populations in the states $|b\rangle$ and $|c\rangle$ would be given by,

$$P_i = |g_i|^2 \int d\omega \frac{\kappa/\pi}{(\kappa^2 + \omega^2)} |\hat{\alpha}(-i(\omega - \delta_i))|^2, \quad i = b, c. \quad (9)$$

Note that we can identify $2|g_i|^2/\kappa$ with the decay $2\gamma_i$ of the state $|a\rangle$ to the state $|i\rangle$ in a resonant cavity. This is the constant first calculated by Purcell [20] and observed later by Goy *et al* [21]. The coefficient α given by equation (5) can now be written in a more transparent form,

$$\hat{\alpha}(z) = \left\{ z + \frac{G^2}{z+i\Delta} + \frac{\kappa\gamma_b}{z+\kappa-i\delta_b} + \frac{\kappa\gamma_c}{z+\kappa-i\delta_c} \right\}^{-1}, \quad (10)$$

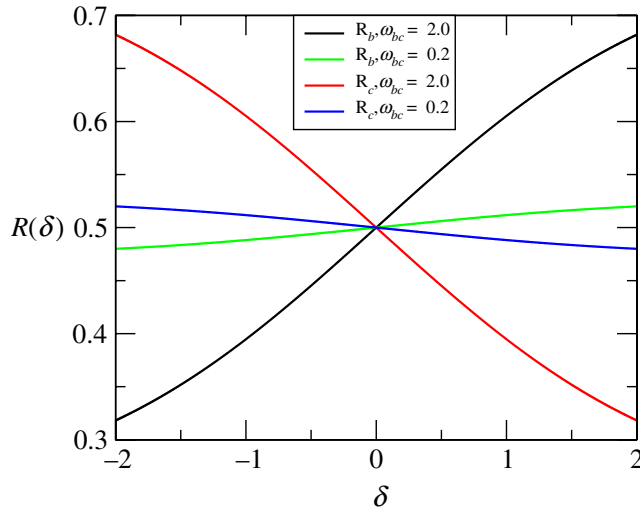


Figure 2. The populations R_b and R_c defined by $R_b = P_{b,\gamma_b=1,\gamma_c=1}/P_{b,\gamma_b=1,\gamma_c=0}$ and $R_c = P_{c,\gamma_b=1,\gamma_c=1}/P_{c,\gamma_b=0,\gamma_c=1}$ plotted as a function of detuning $\delta = (\omega_{ab} + \omega_{ac}) - 2\omega_c$ of the cavity in absence of any driving field. Here, ω_{bc} is the separation between the states $|b\rangle$ and $|c\rangle$. All parameters are normalized with respect to κ .

where Δ is the detuning of the coherent drive as shown in figure 1. The results given by equations (9) and (10) are the basic results of this paper. These are exact—no approximation on the coupling constant has been made. Similarly, no approximation on the strength of the coherent drive field has been used. Further all dispersive effects are included through the complex Lorentzians in equation (10) and therefore no Markov approximation is used. It may be noted that the poles in $\hat{\alpha}(z)$ lead to spectral modifications due to both coherent drive as well as due to strong coupling effects [4], [12]–[14]. The exact location of such poles would depend on various detunings; field strength and the coupling constants g . We do not discuss the issue of spectral modifications in this paper.

The branching ratio is given by P_b/P_c . In order to highlight the effect of the external fields on the branching ratios we consider some special cases first. Let us assume that $\gamma_b = \gamma_c = 1$. Further, assuming $\delta_b = -\delta_c$ and $\Delta = 0$ one can prove that $P_b = P_c$. Thus, we do not find any dependence of the branching ratio on the coherent drive. In order to obtain asymmetric branching ratios we can consider a cavity which is asymmetrically detuned from the two transition frequencies. Even in the absence of any coherent drive the asymmetric tuning can lead to $P_b \neq P_c$. We show this in figure 2 for a fixed separation between the states $|b\rangle$ and $|c\rangle$, and for varying tuning of the cavity. The effect gets more pronounced as the separation between the two states increases. Thus in order to obtain specifically the effect of the driving field we consider the symmetric situation $\delta_b = -\delta_c$. In figure 3 we show how the branching ratio depends on the strength of the coherent drive. Note that $G/\kappa > 1$ corresponds to the case when the Rabi frequency of the external field exceeds the spectral width of the cavity vacuum. This is typical of the situation when Markov approximation does not hold. The effect of overlapping resonances and quantum interferences is also very much evident in the time dependence of the population of the excited state as shown in figure 3(b). In figures 4 and 5, we show how the branching ratios

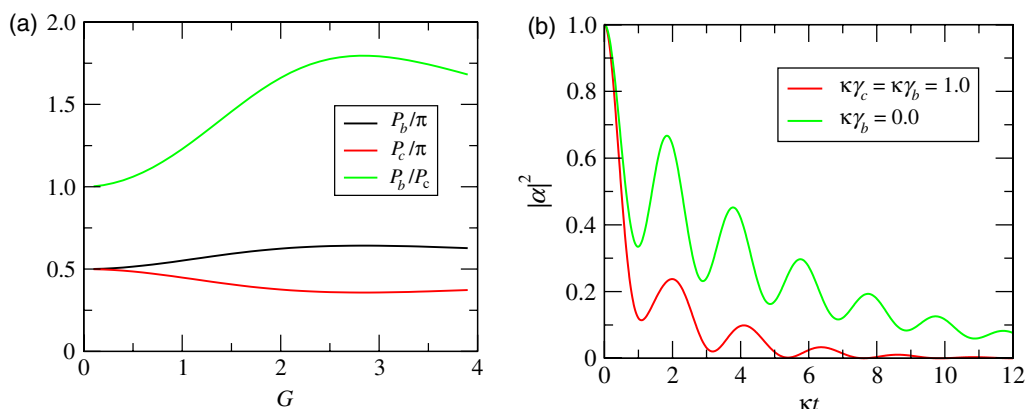


Figure 3. (a) Populations of states $|b\rangle$ and $|c\rangle$ and the branching ratio P_b/P_c as a function of the driving field, for cavity detuning of $\delta_b = -\delta_c = 2.0$ and fixed drive detuning of $\Delta = 2.0$. (b) Comparison between the population of the excited state as a function of time in the case of both lower states available and only one lower state available ($\gamma_b = 0$) for $G = 1.0$, $\delta_c = -2.0$ and $\Delta = 2.0$. All parameters are normalized with respect to κ .

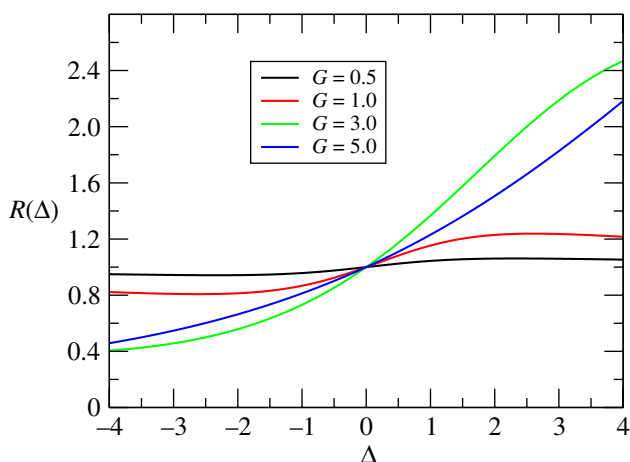


Figure 4. Branching ratios $R = P_b/P_c$ plotted as a function of the detuning Δ of the coherent drive for different values of the driving field. The cavity detuning is kept fixed at $\delta_b = -\delta_c = 2.0$. All parameters are normalized with respect to κ .

change with a change in the detuning of the coherent drive. Figure 5 is for the case when the two transitions $|a\rangle \leftrightarrow |b\rangle$ and $|a\rangle \leftrightarrow |c\rangle$ are within the spectral width of the cavity.

In conclusion, we have shown how branching ratios can depend on external electromagnetic fields. Our calculations show that the effects are especially pronounced if the vacuum of the electromagnetic field has a bandwidth comparable to the strength of the field. These kinds of fundamental modifications are expected to occur generally in any system–bath interaction.

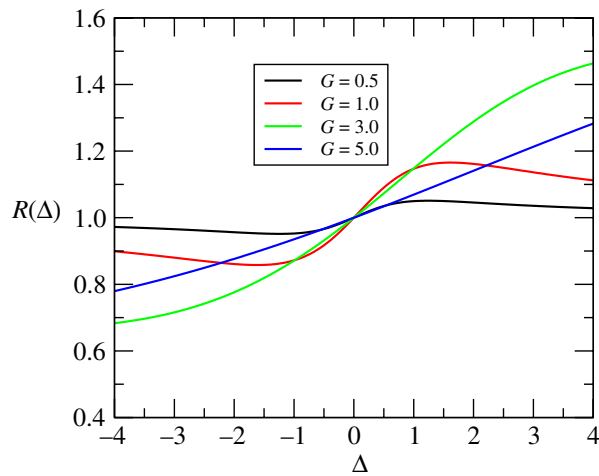


Figure 5. Branching ratios $R = P_b/P_c$ plotted as a function of the detuning of the coherent drive as in figure 4 but now for a cavity detuning of $\delta_b = -\delta_c = 0.5$. All parameters are normalized with respect to κ .

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