Experimental Studies of a Strongly Driven Rabi Transition

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The response of a strongly driven Rabi transition, which is associated with a strongly driven two-level atom, is studied. Such a driven Rabi transition exhibits Mollow splitting including gain features, and the Mollow spectrum associated with the original transition is also altered. We demonstrate that driving the Rabi transition produces a new transition at its own Rabi frequency, which can be driven to exhibit further Mollow splitting. Our experimental results are in good agreement with theory based on doubly dressed states. [S0031-9007(98)05703-2]

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A strongly driven two-level atom (TLA) has been extensively studied due to its fundamental importance in the understanding of light-atom interactions. The weak probe field absorption spectrum is significantly modified by a strong pump field with classic examples such as the Autler-Townes doublet when probing to a third level [1] and the Mollow spectrum when probing to the same level [2]. These spectra can be explained by the formalism of the *dressed states* [3], the eigenstates of the "atom + field" Hamiltonian.

A natural extension of this early work is to apply a second strong field to the TLA and study the new dynamics of the system. Doubly driven systems have been extensively studied theoretically [4], but there are limited experimental studies, and these all involve two near resonant fields. They include several studies examining the strong pump, strong probe situation [5], resonance fluorescence [6], and the Autler-Townes probe absorption spectrum [7]. The present Letter also reports experimental measurements of a doubly driven system, but the configuration is different from those studied previously. Only one strong field is near resonance, and the second saturating field is at very low frequencies, far from the transition frequency of the TLA. This low frequency field interacts with the standard dressed states for the first driving field. It is shown that the traditional probe absorption spectrum (Mollow spectrum) is dramatically modified by the second driving field. Additionally, the second low frequency resonance provides a new frequency regime to probe, in the region of the low frequency pump field. The absorption spectra in both the near resonance and low frequency regions can be readily observed, and all the features can be interpreted in terms of the doubly dressed eigenstates of the "atom + two fields" Hamiltonian. From this perspective the pumping scheme is particularly attractive as the polarization of the pumping beams are orthogonal, and this enables an elegant analytical analysis.

There is another aspect of this work in that the whole study can be taken to another evolution. To explain this idea one should be clear about the origin of the low frequency resonance. This corresponds to transitions within the dressed state doublets associated with the near resonance pump field. This low frequency transition is at the *generalized Rabi frequency* of the near resonance pump field, and is referred to as the *Rabi transition* for the remainder of the paper. The transition is allowed when the dipole moment transition matrix has diagonal terms [8]; i.e., the system has a permanent dipole moment. This is common for spin systems. The transition has been studied indirectly in early NMR studies [9]; however, it has only been observed directly once [10] prior to our studies [11]. In the current Letter, the Rabi transition is readily observed and is attractive for bichromatic studies because it displays the characteristics of a very simple TLA. We therefore treat the Rabi transition as a new "ideal" TLA, and study the effect of doubly driving this Rabi transition. The changes in the absorption spectra are analogous to those for the standard TLA. However, the spectra are obtained with unprecedented clarity, and this novel approach provides an even better illustration of the power of the dressed state formalism.

We begin with a brief introduction to the doubly dressed states theory used in this paper. A spin system with dipole moment $\vec{\mu}$ interacts with a field $\vec{B}$ via the dipole interaction ($H = -\vec{\mu} \cdot \vec{B}$), and the standard $\Delta S = 1$ transitions between the spin states are driven by $\hat{x}$ polarized fields, where the $\hat{z}$ axis is defined to lie along the spin axis. Consider then a TLA interacting with a $\hat{x}$- and a $\hat{z}$-polarized electromagnetic field, characterized by quantum numbers n and m, respectively, and so the Hamiltonian is

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_x + \mathcal{H}_z,$$

where the parts of the Hamiltonian characterize the noninteracting atom + fields system $\mathcal{H}_0$, the interaction with the near resonant $\hat{x}$ polarized field under the rotating wave approximation $\mathcal{H}_x$, and the interaction with the low frequency $\hat{z}$ polarized field $\mathcal{H}_z$. The experimental TLA studied consists of a $m_s = 0$ and $m_s = -1$ pair of spin states in a dc magnetic field aligned along the

$\hat{z}$ axis. Defining the $|-1\rangle$ and $|0\rangle$ spin states as $|e\rangle$ and $|g\rangle$ with energies $\hbar\omega_0$ and 0, respectively, the Hamiltonian components become

$$\mathcal{H}_0 = \hbar\omega_0|e\rangle\langle e| + \hbar\omega_x\left(aa^\dagger + \frac{1}{2}\right) + \hbar\omega_z\left(bb^\dagger + \frac{1}{2}\right),$$

$$\mathcal{H}_x = \hbar g(|e\rangle\langle g|a + |g\rangle\langle e|a^\dagger),$$

$$\mathcal{H}_z = \hbar k(|e\rangle\langle e|)(b + b^\dagger),$$

where $a(b)$ and $a^\dagger(b^\dagger)$ are the annihilation and creation operators for the $\hat{x}(\hat{z})$ field and $g(k)$ characterizes the interaction strength of the TLA with the $\hat{x}(\hat{z})$ polarized field. $\mathcal{H}_0 + \mathcal{H}_x$ can be diagonalized by the standard dressed states [3], given by

$$|1,N,m\rangle = \cos\theta|e,n-1,m\rangle + \sin\theta|g,n,m\rangle,$$

$$|2,N,m\rangle = -\sin\theta|e,n-1,m\rangle + \cos\theta|g,n,m\rangle,$$

with $\tan 2\theta = \chi_x/\delta_x$, $\delta_x = \omega_0 - \omega_x$, $\chi_x = -2g\sqrt{n}$, and where the *generalized Rabi frequency* is given by $\Omega_x = \sqrt{\delta_x^2 + \chi_x^2}$. The energy levels form an infinite ladder of doublets, with adjacent doublets separated by $\hbar\omega_x$ and levels within the same doublet separated by $\hbar\Omega_x$ [see Fig. 1(b)].

$$(\mathcal{H}_0 + \mathcal{H}_x)|i,N,m\rangle = \left[\frac{1}{2}\omega_0 + N\omega_x + \left(m + \frac{1}{2}\right)\omega_z + (-1)^i\frac{1}{2}\Omega_x\right]|i,N,m\rangle.$$

We see that the permanent transition moment, μ_z, allows transitions between the dressed states within the same doublet as $\langle 2,N,m|\mu_z|1,N,m\rangle = -\cos\theta\sin\theta\,\mu_{ee}$ in this case, whereas the transition is forbidden for the standard dipole moment μ_x ($\langle 2,N,m|\mu_x|1,N,m\rangle = 0$). Hence, the $\hat{z}$ polarized field drives transitions between the dressed states within the same doublet, with transition frequency Ω_x. The orthogonal nature of the two pump fields allows us to diagonalize the total Hamiltonian $\mathcal{H}$ with the *doubly dressed states*

$$|\alpha,N,M\rangle = \cos\phi|2,N,m-1\rangle + \sin\phi|1,N,m\rangle,$$

$$|\beta,N,M\rangle = -\sin\phi|2,N,m-1\rangle + \cos\phi|1,N,m\rangle,$$

where $\tan 2\phi = \Lambda/\delta_z$, $\delta_z = \Omega_x - \omega_z$, $\Lambda = -(\chi_x/\Omega_x)k\sqrt{m}$, and the new generalized Rabi frequency $\Omega_z = \sqrt{\delta_z^2 + \Lambda^2}$. Each of the energy level doublets in the singly dressed states are now "dressed" giving a new infinite ladder of energy level doublets, with adjacent doublets separated by $\hbar\omega_z$ and levels within the same doublet separated by $\hbar\Omega_z$ [see Fig. 1(c)]. The energies of the doubly dressed states are given by

$$E_{\alpha(\beta),N,M} = \left[\frac{1}{2}\omega_0 + N\omega_x + M\omega_z - (+)\frac{1}{2}\Omega_z\right].$$

The master equation is used to calculate the populations of these states and the transition moments between the doubly dressed states in the standard fashion [3]. For example, in the isotropic relaxation case ($T_1 = T_2$), $w = (\delta_X/\Omega_X)W_{\rm eq}$, where w is the steady state dressed state population difference between the levels $|2,N,m\rangle$ and $|1,N,m\rangle$, and $W_{\rm eq}$ is the equilibrium population difference in the bare states. It can be seen then that in the case where $\delta_X < 0$ and $W_{\rm eq} < 0$ (as is the case reported in this Letter) w becomes positive and we have a population inversion in the dressed state basis. A qualitative representation of the population results are given in Fig. 1(b), where the thicker line indicates the more highly populated energy level. It can then be inferred whether a particular transition is emissive or absorptive.

The $m_s = 0$ and $m_s = -1$ spin levels involved in our experiments are part of the spin substates of the ^3A orbital ground state of the nitrogen vacancy (N-V) center in diamond, and the magnetic transition is detected with the Raman heterodyne technique, details of which are found elsewhere [11]. We studied both absorption and dispersion probe responses, but only absorption results are shown. Figure 2(i) shows the absorption of a weak $\hat{x}$ polarized probe, in the absence of saturating fields. This corresponds to the original "bare" transition with the dominant component at ~66 MHz. The transition is driven by a $\hat{x}$ field slightly off resonance; $\omega_x \sim 69$ MHz. The results are shown in Fig. 2(ii), where it can be seen that the probe response exhibits the well known Mollow profile with absorption and amplification features at $\omega_x \pm \Omega_x$.

Now, a second driving field is applied near the Rabi transition $\Omega_x \sim 10.5$ MHz. This $\hat{z}$ polarized field

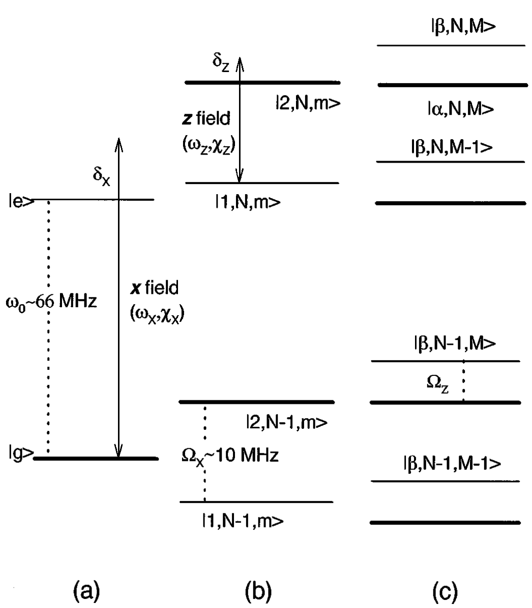

FIG. 1. Representative energy level diagram for (a) the bare atom + noninteracting field, (b) the dressed states due to $\hat{x}$ pump, and (c) the doubly dressed states due to the additional $\hat{z}$ pump. The bold levels indicate the more heavily populated state.

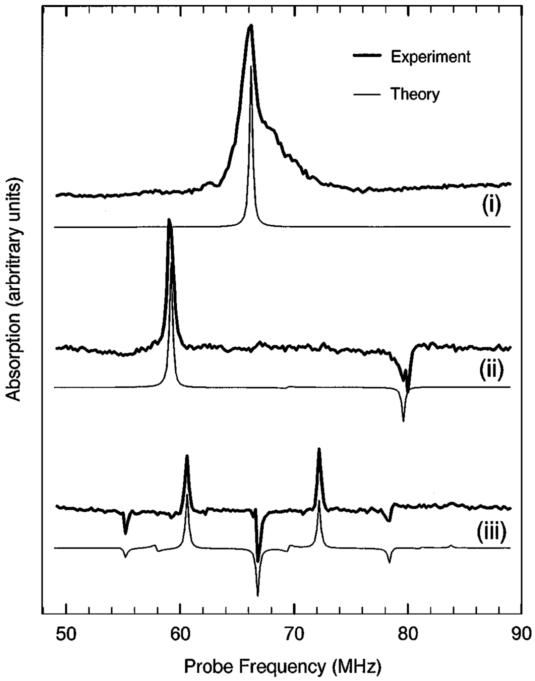

FIG. 2. The EPR $\hat{x}$ polarized probe absorption spectrum: (i) with no $\hat{x}$ or $\hat{z}$ pump fields where the shoulder on the high frequency side of the major resonance at 66 MHz is due to weaker hyperfine features at 68 and 70 MHz, (ii) with a $\hat{x}$ pump ($\delta_x \sim -3$ MHz, $\Omega_x \sim 10.5$ MHz) midway between the two weak hyperfine lines, and (iii) $\hat{x}$- plus $\hat{z}$-polarized pump ($\delta_z \sim -1$ MHz, $\Omega_z \sim 2.7$ MHz). The thick line gives the experimental results, and the thin line gives the theoretical results using the doubly dressed states for a single, homogeneously broadened EPR line. The extra structure in some of the features of the driven spectra is due to the neglected hyperfine lines.

($\omega_z \sim 11.5$ MHz, $\Omega_z \sim 2.7$ MHz) drives the Rabi transition off resonantly, producing the doubly dressed states. Figure 2(iii) shows how this second field dramatically changes the probe absorption spectrum in the region of the original transition. The absorption/amplification components from Fig. 2(ii) are split into four new components at $\omega_x \pm \omega_z \pm \Omega_z$, which are emissive or absorptive depending on the populations of the doubly dressed states, represented in Fig. 1(c). These features are readily seen to be associated with the high and low Mollow components. The features are due to transitions between the doubly dressed states, with the feature at $\omega_x - \omega_z - \Omega_z$ representing the transition between $|\alpha, N, M - 1\rangle$ and $|\beta, N - 1, M\rangle$, for instance. However, two new lines have appeared at $\omega_x \pm \Omega_z$ where there were no features prior to the application of the $\hat{z}$ field. The reason for this is that in the singly dressed atom, the population of the $|i, N, m\rangle$ and the $|i, N - 1, m\rangle$ ($i = 1, 2$) states are equal, and so there is no net absorption at ω_x. However, the application of the second driving field redistributes these populations, allowing transitions between the $|\beta, N, M\rangle$ and the $|\alpha, N - 1, M\rangle$ states, for example, giving rise to these new features. The thin line shows the theoretical results displaying excellent agreement with the experimental results.

As mentioned earlier, the emergence of the Rabi transition allows the study of a new frequency regime. A weak $\hat{z}$ polarized field can be used to probe the Rabi transition itself. In a separate experiment, the original transition is driven by the first pump at $\sim$69 MHz with $\Omega_x \sim 10$ MHz, but this time a $\hat{z}$ polarized probe scans over the region about Ω_x. Figure 3(i) shows the probe absorption response, and a clear transition at the generalized Rabi frequency is observed. It is emissive because of the population inversion in the dressed states caused by the first negatively detuned pump field. Next, the $\hat{z}$ polarized pump field may be applied, the results being shown in Fig. 3(ii). Here, the Rabi transition ($\sim$10 MHz) has been driven by a $\hat{z}$ polarized field, slightly detuned ($\sim$10.5 MHz). An ac Stark split spectrum results with features at $\omega_z \pm \Omega_z$, completely analogous to the results for the singly driven original transition [cf. Fig. 2(ii)]. In the Rabi transition, the overall sign of the spectrum is reversed as the Rabi transition was emissive to begin with.

Interestingly, the $\hat{z}$-polarized probe reveals an absorptive response of the system at the frequency given by $\Omega_z \sim 1.8$ MHz, interpreted as being the "Rabi of the

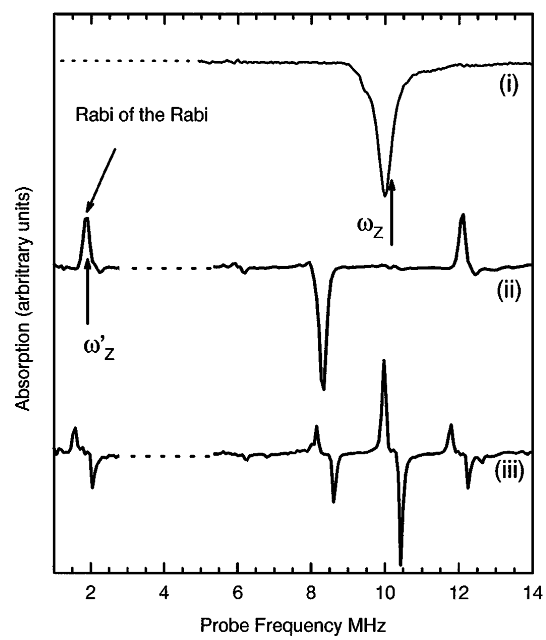

FIG. 3. The Rabi transition, probed with a $\hat{z}$ polarized weak field: (i) The undriven Rabi transition at $\sim$10 MHz, where the dotted line indicated there are no spectral features of interest; (ii) the Rabi transition is driven by a $\hat{z}$ pump ($\delta_z \sim -0.5$ MHz, $\Omega_z \sim 1.8$ MHz), with its frequency indicated by an arrow in (i). The Rabi of the Rabi is highlighted at $\sim$1.8 MHz, and (iii) the Rabi of the Rabi is driven with a second $\hat{z}$ pump ($\delta_z' \sim 100$ kHz, $\Omega_z' \sim 200$ kHz), with its frequency indicated by an arrow in (ii).

Rabi" [Fig. 3(ii)]. It demonstrates transitions within the doubly dressed state doublets, analogous to the situation for the Rabi transition (where the transition is within the singly dressed state doublets). In an effort to examine this apparent symmetry, a second strong $\hat{z}$-polarized field can be applied to the Rabi of the Rabi, and the results of this are shown in Fig. 3(iii). Here, the second $\hat{z}$ field is applied at ~1.9 MHz, slightly detuned from the Rabi of the Rabi, and the resultant splitting of the Rabi of the Rabi into a Mollow type spectrum is observed. Furthermore, the spectrum in the region of the Rabi transition is dramatically changed by the application of this field to the Rabi of the Rabi, again showing the sidebands being split, and two large central features appearing [cf. Fig. 2(iii)]. This gives beautifully clear evidence of the production of "triply dressed states," showing for the first time a triply driven TLA with three fields of different strengths and frequencies, and demonstrates the underlying symmetry of the system.

In this Letter we report the fascinating and elegant physics arising from driving a TLA with multiple fields and probing to the same transition. Driving the EPR transition gives rise to a $\hat{z}$ allowed Rabi transition. As the new transition has significantly decreased hyperfine structure and inhomogeneous broadening, it behaves as a new nearly ideal two-level system. Driving this transition produces its own Mollow spectrum as well as significantly altering the near resonance absorption spectrum. Driving the Rabi transition gives rise to its own Rabi resonance, and driving the Rabi of the Rabi gives further splittings of the spectra. This series of experiments of driving a TLA, producing a Rabi transition, and then treating the new Rabi transition as a new TLA can theoretically be continued indefinitely. All signals are obtained with good signal to noise. The doubly driven results are well explained with the doubly dressed state formalism, and it is anticipated that all the features associated with the three pump field case can be readily interpreted in terms of the triply dressed states.

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