Threshold nonlinear absorption in Zeeman transitions

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Abstract

We experimentally study the absorption spectroscopy from a collection of gaseous ⁸⁷Rb atoms at room temperature irradiated with three fields. Two of these fields are in a pump–probe saturation absorption configuration. The third field co-propagates with the pump field. The three fields address Zeeman degenerate transitions between hyperfine levels ⁵S₁/₂, F₁ and ⁵P₃/₂, F₀, F₁ around the D₂ line. We find a sub-natural absorption resonance in the counter-propagating probe field for equal detunings of all three fields. This absorption arises in conjunction with the appearance of increased transmission due to electromagnetically induced transparency in the co-propagating fields. The novel feature of this absorption is its onset only for the blue of ⁵P₃/₂, F₀, as the laser frequency is scanned through the excited states ⁵P₃/₂, F₀, F₁ and F₂. The absorption rapidly rises to near maximum values within a narrow band of frequency near ⁵P₃/₂, F₀. Our experimental results are compared with a dressed atom model. We find the threshold absorption to be a result of coherent interaction between the dressed states of our system.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The absorption spectrum of a driven degenerate two-level system (DTLS) is shown [1] to be radically different from the usual Mollow triplet corresponding to a pure two-level system. The atomic coherence induced by multi-photon interactions in the DTLS results in a competition between constructive or destructive quantum interference which gives rise to subnatural width resonances (SNWR) [2]. The resonance could manifest as electromagnetically induced absorption (EIA), electromagnetically induced transparency (EIT), or even gain, depending on the polarization, intensity and detunings of the driving fields. The line shapes are further influenced by whether the system is closed (cyclic) or open [5], velocity selection [7] and the presence of small polarization admixtures [6]. Akulshin et al [3] have extensively studied the DTLS involving various ground state and excited state hyperfine levels both theoretically and experimentally. They arrive at criteria [4] for the occurrence of EIA in the DTLS, namely (i) Fₑ = Fₚ + 1, (ii) the ground to excited state transition is closed and (iii) Fₑ > 0.

The occurrence of EIA has been attributed to the transfer of coherence from excited state to ground state [8] and also to the transfer of population [9]. Inhibition of EIA due to excited state decoherence has also been reported experimentally [10]. Most of these studies involve the interaction of the DTLS with two driving fields. Multiple driving fields interacting with the DTLS have been reported in the context of coherent hole burning [11] where the same transition is addressed by a saturating beam and a probe.

In this paper we experimentally study the occurrence of sub-natural absorption resonances in the counter probe during a three-field irradiation of gaseous, room temperature ⁸⁷Rb atoms. These three fields address Zeeman degenerate transitions in a DTLS configuration. These resonances arise as a result of dark resonances created by the co-propagating fields, inducing a higher order, off-resonant absorption to neighbouring hyperfine levels in the counter field. The absorption is studied as a function of detuning of the three driving fields. We find, as a function of this detuning, the existence of a sharp threshold where the absorption begins to show up. We compare our experimental results with a theoretical dressed state model of our system which shows...
from a single laser have the same laser frequency [7].

3 Excited hyperfine states are shown primed. The hyperfine levels around the D2 line of $^{87}$Rb are shown in figure 2. Experiment

we do not expect coherent population oscillations [12] to affect dressed states of the system. Since the intensity regime of the irradiate a room temperature sample of $^{87}$Rb atoms. The three beams L1, L2 and L3, derived from a single laser as shown in figure 1(a). Figure 1(b) shows the Zeeman sub-levels of the $F = 1$ ground hyperfine state and that of excited hyperfine states $F = 0', 1'$ relevant for the experiment described here.

2.2. Experimental setup

Three beams L1, L2 and L3, derived from a single laser irradiate a room temperature sample of $^{87}$Rb atoms. The laser is an external cavity diode laser (ECDL) with a line width typically around 1 MHz operating around the D2 line at 780 nm. The laser frequency is scanned around the hyperfine manifold starting from the ground state $5S_{1/2}, F = 1$ to excited states $5P_{3/2}, F = 0'$ and $5P_{3/2}, F = 1'$. The laser frequency is monitored through a saturation absorption setup as indicated in figure 2. A phase-sensitive feedback mechanism is employed to lock the frequency of the laser to any one of the excited states. As shown in figure 2, L1 and L2 are collinear and co-propagating and L3 counter propagates to both L1 and L2. L1 is typically of higher intensity than L2 with intensity ranging between 0.1 $I_2$ and 0.5 $I_2$. where $I_2$ is the saturation intensity for the transition $5S_{1/2}, F = 1$ to $5P_{3/2}, F = 0'$. L2 has intensity in the range of 0.1 $I_2$–0.5 $I_2$. L3 has the least intensity ranging between 0.1 $I_2$ and 0.01 $I_2$. All three beams being derived from a single laser have the same laser frequency $\omega_L$. At any given $\omega_L$, the L2 beam can be ramped over a frequency range of $\pm 10$ MHz around $\omega_L$, by a combination of two acousto-optic modulators (AOMs in figure 2). Thus for a given detuning $\delta_{10}$, measured from $5P_{3/2}, F = 0'$ level, the L2 beam, through the AOMs can scan a width of $\pm 10$ MHz about $\delta_{10}$. Here $\delta_{10} = \omega_L - \omega_{10}$ where $\omega_{10}$ is the frequency difference between $5S_{1/2}, F = 1$ and $5P_{3/2}, F = 0'$. The experiment consists of recording the transmitted intensities of all three beams using fast photodiodes. The transmitted intensities show two kinds of variations. One of them is due to changing $\omega_L$. These variations mimic the standard saturation absorption variations in the transmission of counter-propagating L3 probe, for appropriate intensity ratios of L1 and L3. But because of the presence of L2 co-propagating with L1, at every $\omega_L$, the condition for a two-photon Raman resonance is satisfied between L1 and L2. This results in transmission changes in L2 and L3 which are very different from a standard saturation experiment. In fact, during the $\pm 10$ MHz frequency scan of L2 centred at $\omega_L$, we see enhanced transmission of L2 due to EIT as expected. As the AOM scan rate is faster than the scan of the laser frequency $\omega_L$, the spectral features are dominated by features during the faster frequency scan in the narrow bandwidth centred around $\delta_{10}$. The slower scan of the laser frequency $\omega_L$ results in a varying background.

The L1 and L2 beams are orthogonally polarized with the quantization axis as the direction of propagation and the L3 beam is polarized similar to the L1 beam. The vapour cell containing gaseous rubidium atoms in their natural isotopic abundance is covered with three layers of $\mu$-metal shield. The residual field is in the range of 10–20 milligauss inside the cell.

2.3. Level structure for multi-photon transitions

The three laser beams L1, L2 and L3 can drive Zeeman degenerate transitions as shown in figure 3. Figure 3(a) shows transitions between the Zeeman sublevels of $5S_{1/2}, F = 1$ and $5P_{3/2}, F = 0'$. The laser beams L1 (shown thick) and L2, which are oppositely circularly polarized, form a $\Lambda$ system with the magnetic hyperfine levels $m_F = \pm 1$ of the ground state $F = 1$ and the excited state level $F = 0'$. The beam L3 is counter propagating to L1 and L2 and hence there is a
frequency offset in the atomic rest frame due to the thermal motion. L3 connects the same set of levels as L1, shown here with dotted lines. Figure 3(b) shows transitions between the Zeeman sublevels of $5S_{1/2}, F = 1$ and $5P_{3/2}, F = 1'$. For oppositely circularly polarized L1 and L2 this gives rise to a Λ and a V configuration for EIT. Even here, because the L3 beam (shown with dotted lines) is counter to L1 and L2, it does not take part in the transparency effect.

Because of the presence of several velocity classes of atoms at room temperature, even when the laser is addressing resonantly the transition shown in figure 3(a), it is off-resonantly addressing transitions in the $F = 1'$ manifold and vice versa. This effect is most prominent for the L1 laser, which has a higher intensity than L2 and L3. So a realistic picture of possible laser transitions is shown in figure 3(c). Here the dot-dashed line indicates the transitions addressed by the L1 laser off-resonantly. If, in addition, we consider optical pumping by the strong L1 laser, we see that most of the population is in the ground state $m_F = -1$. Considering this, we ignore transitions starting from the ground state $m_F = 0$. Thus, an $N$ system as shown in figure 3(d) is the appropriate level configuration addressed by the fields L1, L2 and L3.

3. Results and Discussion

Figure 4 shows the typical transmitted intensities of L2 and L3 beams for various detunings $\delta_{10}$. As stated before, L1, L2 and L3 beams have the same detuning $\delta_{10}$ as they are derived from the same laser. The L2 beam scans, at every detuning $\delta_{10}$, a frequency range of $\pm 10$ MHz centred around $\delta_{10}$. Trace B of figure 4(a) shows the ramp voltage scan applied to the AOMs in the path of L2 resulting in a frequency scan of $\pm 10$ MHz of L2, centred around a given $\delta_{10}$. On either side of these AOM frequency scans are shown traces A and C. Traces A and C show the transmitted intensity profiles of L2 and L3 respectively, during the scan of L2 centred around a given value of $\delta_{10}$. As can be seen from trace A, L2 shows increased transmission over the background, whenever L1 and the scanned L2 are at the same detuning, satisfying the two-photon resonance condition for EIT. The EIT window is typically of 1–2 MHz width. On the other hand, the transmitted intensity of L3 given in trace C shows a sharp absorption resonance, at the very position at which L2 shows EIT transmission resonances. The width of this absorption is sub-natural ranging between 300 kHz and 800 kHz. This width, however, is larger than that predicted by the ground state Zeeman coherence. This is mainly due to the presence of gradients in the residual magnetic field in our setup. The graph of trace C has been amplified six times for clear representation in this combined graph. Since this absorption in L3 occurs whenever the co-propagating L1 and L2 satisfy the two-photon EIT condition, it implies a higher order absorption mechanism for L3 absorption. In fact, a related three-photon absorption was seen by us in an $N$-level scheme [13] for similar geometry of beams. The novelty of such a feature here, for Zeeman degenerate transitions, is its onset [14] as shown in figure 5.

Shown in figure 5 are traces A and C from the same experimental run as for figure 4 but in a different region of $\delta_{10}$ around $F = 0'$. As can be seen from this graph, while trace A remains qualitatively unchanged even in this region, trace C shows disappearance of the absorption feature below $\delta_{10} < 12$ MHz marked by the vertical line in the figure. For all values of $\delta_{10}$ below this value, there is no narrow absorption seen in trace C. The background absorption of L3 in this region shows changes that are not correlated with the appearance of
the EIT feature in the L2 field (trace A). We have estimated for this graph that the narrow absorption disappears within a 5 MHz change of $\delta_{10}$ to the left of the vertical line. Conversely, we can say that the nonlinear absorption in L3 has a detuning-dependent onset around $\delta_{10} = 12$ MHz.

For the sake of clarity we have shown in figures 6(a) and (b), the entire region of $\delta_{10}$ values extending up to $-35$ MHz. The narrow absorption feature in L3 (trace C) is totally absent in this region. Only beyond the vertical line do we see the onset of this absorption. In contrast, trace A of this figure, representing L2 transmission, continues to show the EIT feature for all values of $\delta_{10}$. Figure 6(b) shows that the narrow absorption feature in trace C continues for positive $\delta_{10}$ at least up to 35 MHz.

We have repeated the experiment with various intensities of the L2 field keeping the L3 intensity the same. Every time we see that there is an onset of EIT correlated absorption in L3 occurring in the vicinity of $\delta_{10}$. The value of $\delta_{10}$ at which this occurs is not strongly dependent on the intensity of the L1 beam. However, the width of the absorption feature and the EIT transmission feature increases at higher intensities of L2 as expected.

We remark here that there are dispersion-like features below $\delta_{10} = 12$ MHz in figure 5. These features, which are correlated with the occurrence of EIT, disappear for smaller powers of L1, L2 and L3. In this low-intensity regime, the absorption resonance disappears within a narrower span of the laser frequency scan of 2–3 MHz.

Plotted in figure 7 is the mean absorption contrast of the narrow absorption in L3 as a function of $\delta_{10}$. The contrast is defined as the ratio of the maximum value of absorption to the background value. The error bars indicate maximum...
deviations from the mean value of the contrast for similar runs of the experiment. We see from this figure that the absorption rises to its maximum value in a narrow range of $\delta_{10}$.

### 3.1. Dressed state model

We have derived the dressed states of our system with only the strong laser $L_1$ and where $L_1$ addresses both the excited states $F = 0', 1'$ and the ground state $F = 1, m_F = 1$, at any given $\delta_{10}$. The dressed states are then probed by $L_2$ and $L_3$ lasers. The Hamiltonian for this is given by

$$\mathcal{H} = \hbar \begin{pmatrix} \delta_{10} & 0 & \Omega_{L_1}^{\delta_{10}} \\ 0 & \delta_{10} - \Delta_{bhf} & \Omega_{L_1}^{\delta_{10}} \\ \Omega_{L_1}^{\delta_{10}} & \Omega_{L_1}^{\delta_{10}} & 0 \end{pmatrix}.$$  

(1)

The dressed eigenstates are given below in the regime of $\delta_{10}$, where the effect of the off-resonant coupling by $L_1$ can be treated to first order:

$$|+\rangle = \frac{1}{\Omega_0} \left[ \Omega_{L_1} |1'\rangle + f^+ (|1, m_F = 1\rangle + \frac{\Omega_{L_1}^\delta}{(\delta_{10} - f^-)} |0'\rangle) \right]$$

$$|+\rangle = \frac{1}{\Omega_0} \left[ f^+ |1'\rangle + \Omega_{L_1}^\delta (|1, m_F = 1\rangle + \frac{\Omega_{L_1}^\delta}{(\delta_{10} - f^-)} |0\rangle) \right]$$

$$|+\rangle = |0'\rangle - \frac{\Omega_{L_1}^\delta \Omega_{L_1}^\delta}{g^2} |1'\rangle$$

where $\Omega_0, f^\pm$ and $g^2$ are defined as

$$\Omega_0^2 = \Omega_{L_1}^2 + \left( (\delta_{10} - \Delta_{bhf})/2 + \sqrt{\Omega_{L_1}^2 + (\delta_{10} - \Delta_{bhf})^2/4} \right)^2,$$

$$f^+ = \frac{\Delta_{bhf} - \delta_{10}}{2} \pm \left( \sqrt{\Omega_{L_1}^2 + (\delta_{10} - \Delta_{bhf})^2/4} \right),$$

$$g^2 = \Omega_{L_1}^2 - [(\delta_{10})(2\delta_{10} - \Delta_{bhf})].$$

In these equations (1)–(7), we represent the Rabi strength of the off-resonant transition from $5S_{1/2}, F = 1, m_F = 1 (|1, m_F = 1\rangle)$ to $5P_{3/2}, F = 1, m_F = 1 (|1'\rangle)$ as $\Omega_{L_1}$, and that of $5S_{1/2}, F = 1, m_F = 1 (|1, m_F = 1\rangle)$ to $5P_{3/2}, F = 0 (|0'\rangle)$ transition as $\Omega_{L_1}^\delta$, $\Delta_{bhf}$ denotes the hyperfine separation between $|1'\rangle$ and $|0'\rangle$ which is 72 MHz. $\delta_{10}$ is the detuning of all three beams from the $5S_{1/2}, F = 1$ transition.

We plot in figure 8 the dressed energy values of the dressed states created by $L_1$ which connect $|1, m_F = 1\rangle$ with both the excited states $|0'\rangle$ and $|1'\rangle$, for an atom at rest as a function of the laboratory detuning $\delta_{10}$. We see in this figure that the two branches of the dressed energies of $|0'\rangle$ and $|1'\rangle$ lie very close to each other between values of $\delta_{10}$ varying between 10 and 60 MHz. In the figure this region is highlighted by a rectangle. These two branches are in the linear regime of energy eigenvalues of the dressed states and hence they are predominantly made of only excited states $|0'\rangle$ and $|1'\rangle$ respectively. Thus the absolute energy values are in between the energies of the excited states $|0'\rangle$ and $|1'\rangle$. Furthermore, we see that the energy separation is within a typical excited state line width (6 MHz). Thus transitions from the ground state to these dressed states will result in interference of pathways. We believe that this interference is the origin of our narrow absorption in L3. More importantly, we see that these dressed state energies diverge below $\delta_{10} = 0$. The spectral position

![Figure 6](image1.png)

**Figure 6.** L2 EIT transmission and corresponding L3 absorption shown as a function of $\delta_{10}$, showing regions where the L3 absorption alone is absent. (a) The absence of L3 absorption resonance to the left of the vertical line (trace C) for values extending up to $\delta_{10} = -35$ MHz. (b) The presence of L3 absorption resonance (trace C) beyond the vertical line extending values of $\delta_{10} = 35$ MHz. Trace A in both the figures represents L2 beam transmission showing EIT for all values.

![Figure 7](image2.png)

**Figure 7.** Mean contrast of L3 absorption resonance as a function of the detuning $\delta_{10}$ from $F = 0$, showing the threshold value and the steep slope. The circles denote the mean values for various runs and the error bars indicate the extent of maximum and minimum deviation from the mean value. The solid line is drawn as a guide to the eye connecting the mean values.
at which the divergence between these energy values goes beyond 6 MHz defines our threshold. At this position and beyond the dressed states become distinguishable, thus killing the interference phenomenon and hence the absorption in L3.

It is important to note that for absorption from L3 to happen the energy of an L3 photon should be comparable to the dressed energies. The energy of an L3 photon is given by the detuning $\delta_{10}$. So in the linear regime, the dressed states are also coincident with the energy of an L3 photon, thus facilitating absorption.

Figure 9 shows the variation of dressed energies of $|0\rangle$ and $|1\rangle$ very close to the rectangular region highlighted in figure 8. We see that the energy difference shows a steep variation close to $\delta_{10} = 0$. This should be compared with the sharp threshold we see in our experimental curve (figure 7).

### 3.2. Threshold nonlinear resonances with off-resonant interactions

It is well known that for a pure $\Lambda$ or a double $\Lambda$ system, in the absence of any perturbing mechanism introducing light shifts to a dark state, the system exhibits no higher order nonlinearity [17]. In our system, this perturbation is introduced by the off-resonant transitions addressed by the strong L1 laser. This laser makes the transition probability for transitions of suitable velocity class of atoms, to the $F = 1'$, non-negligible. Our system, due to this off-resonant interaction, can be seen as a modified $N$ system as shown in figure 3(d). So, for a suitable velocity class of atoms, there is a suppression of linear susceptibility due to EIT and a simultaneous enhancement of absorption due to higher order nonlinearity to the near hyperfine level. This absorption due to higher order nonlinearity is distinctly seen in the L3 beam due to its counter geometry. In effect, this feature is very similar to the higher order nonlinear absorption seen in $N$ systems with subnatural widths [18–20]. The difference in the present case is its appearance at a specific detuning and the rapid rise in contrast, within a narrow frequency span. Both the double $\Lambda$ and tripod systems have been shown to have many properties [21–23] that aid specific quantum engineering of superposition states [24]. Our system, which is a modified double-$\Lambda$ system, exhibits a threshold nonlinear absorption of sub-natural width at the same frequency where transparency is seen due to ground state Zeeman coherence. The separation of absorptive and transmittive features in L2 and L3 beams is achieved by geometry of beam propagation. Such a feature is desirable from the viewpoint of monitoring the fidelity of the dark states. Passive monitoring of counter-propagating probe should reveal the nature of CPT coherence in the forward beams. Also, by using the real part of the nonlinear susceptibility, one can design phase gates which work only in a specific bandwidth. In this sense, the threshold phenomenon seen here can be used as a frequency filter.

### 4. Conclusions

We report in this paper an experimental observation of a subnatural absorption feature in a modified $N$ system around Zeeman degenerate transitions in the D2 manifold of $^{87}$Rb. This feature is seen when the gaseous Rb atoms are irradiated with three fields all derived from the same laser. The novelty of this absorption resonance is its frequency-dependent onset beyond a certain detuning of the laser. We show that this absorption feature in one of the beams arises due to off-resonant absorption to the neighbouring hyperfine excited states through a higher order absorptive nonlinearity. We have modelled our system as a dressed state formalism. This shows that the threshold nature of this absorption is due to interference between the dressed states of the excited states. This feature and its distinct appearance in only one of the three fields provides a powerful tool for frequency filtering in wave guides.
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