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Optical pumping and coherence effects in fluorescence from a four level system

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Abstract. An effective four-level system around the D2 line of ^{85}Rb at room temperature, is experimentally investigated by fluorescent studies under the action of two driving fields L1 and L2. This system exhibits unique features in fluorescence as a function of frequency separation between L1 and L2. In particular, at two-photon resonance, when the Rabi frequency of L1 exceeds that of L2, signatures of Electromagnetically Induced Transparency effect (EIT) arising from the three-level Λ sub-system is present as a sub-natural dip in fluorescence from the fourth level. At comparable strengths of L1 and L2 the fluorescence features indicate a regime, where the effects arising from optical pumping and EIT effect due to ground hyperfine level coherence coexist. We see in the coexistence regime, saturation effects arising from difference frequency crossing (DFC) resonances and optical pumping around the EIT window. At low strengths of L1, all signs of coherence vanishes from the system and the fluorescent features result from incoherent optical pumping through the Autler-Townes split states of the excited state hyperfine levels, which are split due to the stronger L2 laser. The dominant role of the L1 laser in creating a robust transparency signal even in the presence of an off-resonant excitation is brought out. The results are supported by density matrix calculations.

PACS. 32.80.Qk Coherent control of atomic interaction with photons – 33.80.Be Level crossing and optical pumping

1 Introduction

Quantum superposition of life-time broadened atomic states and resultant changes in absorption of a resonant probe light has been the subject matter of intense theoretical and experimental study in the past decade [1–3]. A few examples of the varied phenomena given rise by atomic coherence in three or multi-level systems are Lasing Without Inversion (LWI) [4–7], Electromagnetically Induced Transparency (EIT) [8,9] and enhancement of non-linear susceptibilities [10,11]. EIT has been observed in room temperature Rb vapour cells in three and four level configurations. Determination of hyperfine structures [12], ladder type EIT in Doppler broadened systems [9], enhancement of index of refraction [13], sub-natural fluorescence features [14] and width of EIT resonance [15] are a few of the many experiments done in rubidium system at room temperature.

In contrast, very few papers have addressed the nature of the transparency signal obtained in these measurements. Often, in the presence of a strong pump laser, a combination of destructive quantum interference effect and the ac-stark shifting of the excited state level contribute towards a decrease in the absorption of the probe [16]. The experiment performed in [16], separates effects of true quantum interference and that of level splitting. This, they achieve by keeping the Rabi frequency of the pumping laser well below the life-time broadened decay rates of the atomic states. For such low pumping strengths the ac Stark shift has a negligible contribution to the decrease in the absorption profile of the probe. Nevertheless, they show that even with such low pumping strengths a reduction of 15.5 percent takes place in the absorption of the probe which is purely due to the destructive quantum interference of the excitation pathways.

In [17], EIT effect in the Λ system of the D1 hyperfine manifold of levels of ^{87}Rb is studied. Here they examine the nature of transparency signal as a function of changing line-width of the coupling laser. The line-width is increased by introducing an external noise generator in the current driver. They show a gradual loss of transparency as a function of increasing line-width of the coupling laser.

However, a controlled loss of coherence and subsequent changes in the spectral features from a multi-level system have been studied only to a limited extent. It has been shown that the presence of close-lying excited levels in a hyperfine structure only slightly perturbs the EIT system [18]. It was also shown that by tuning to the "center of

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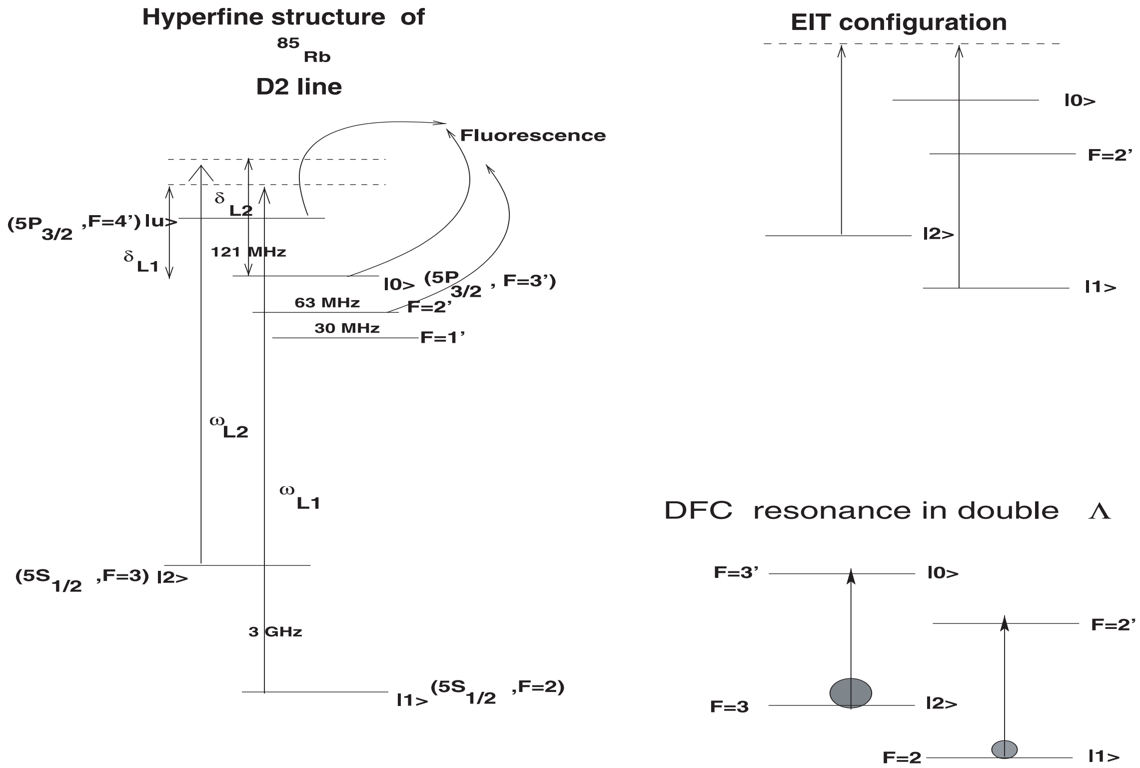

Fig. 1. The four-level scheme around the D2 line of ^{85}Rb.

gravity" of hyperfine levels, EIT can be achieved in atoms with a multi-level hyperfine structure [19].

In this paper we have looked for signatures of induced transparency and level splitting, in fluorescence, arising predominantly from an off-resonant transition in an effective four level system around the D2 line of ^{85}Rb. This experimental study in fluorescence is similar in spirit to the experiment of Wong et al. [20]. In this paper the authors study the several pump-probe resonance features present in absorption of a probe, arising from a four-level scheme around the D1 line of sodium, in a vapour cell at room temperature. They study these resonances for a copropagating geometry of the pump and probe field which eliminates Doppler broadening. The various spectral features are explained as those resulting from the underlying three level Λ system and V system. In particular, they show that for the Λ scheme possible in the D1 transition, a transmission peak ΛP due to EIT is present in absorption, at the two-photon resonance for the highest pump power. At lower pump powers they show that a dip ΛD due to optical pumping replaces the peak (Fig. 2 in [20]). They also show saturation effects due to Difference Frequency Crossing (DFC) accompanied by optical pumping from the underlying double Λ scheme.

In close analogy, we have studied the pump-probe resonances in ^{85}Rb *in fluorescence* at room temperature. Surprisingly, the fluorescence, though dominated by an off-resonant excitation of the four-level scheme still show the ΛD and ΛP features at the two-photon resonance. The fluorescence also carries signatures of optical pumping and saturation effects in the underlying double Λ scheme. We also get a coexistence regime in fluorescence similar to what was seen in absorption. We explore the dominant role played by the L1 laser in maintaining a robust transparency signal even in the presence of off-resonant excitations. In addition, we argue that the presence of a *dip in fluorescence* can only be ascribed to EIT in the underlying Λ subsystem. This makes the identification of transparency easier compared to earlier schemes. A detailed four-level density matrix calculation supports our experimental observations.

2 Level scheme

The response of ^{85}Rb vapour to the presence of two lasers L1 and L2 around the D2 line is studied. The level scheme of ^{85}Rb relevant for this experiment is given in Figure 1. The laser L1 is scanned around the levels $5S_{1/2}, F = 2$ to $5P_{3/2}, F = 1', 2', 3'$. The laser L2 is held fixed at a given detuning from the level $5S_{1/2}, F = 3$ to $5P_{3/2}, F = 3'$ (Hyperfine levels of $5P_{3/2}$ manifold are denoted primed). The laser L2 also off-resonantly excites the transition $5S_{1/2}, F = 3$ to $5P_{3/2}, F = 4'$. The three-level Λ system entering the EIT scheme are the ground hyperfine levels $5S_{1/2}, F = 2, 3$ and their common upper level $5P_{3/2}, F = 3'$. These are denoted respectively as $|1\rangle$, $|2\rangle$ and $|0\rangle$. The majority of fluorescence occurs from the "unconnected" fourth level $5P_{3/2}, F = 4'$ denoted as $|u\rangle$. The fluorescence from this level and from levels $|0\rangle$ and $F = 2'$ are studied as a function of the detuning of laser L1 (δ_{L1}) from the transition $5S_{1/2}, F = 2$ to $5P_{3/2}, F = 3'$ and for

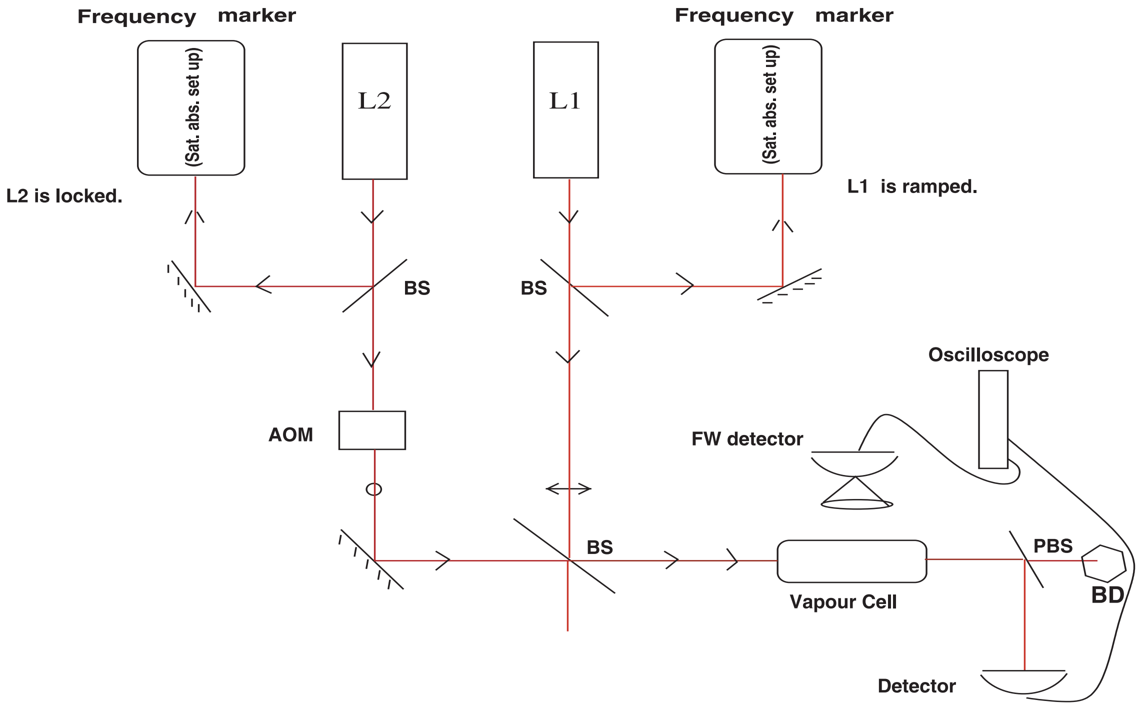

Fig. 2. Schematic of the experimental set-up.

a fixed δ_{L2}. For the purposes of later discussions we denote the Rabi frequencies of laser L1 by Ω_{L1}, of laser L2 addressing the $F = 3 - F = 3'$ transition by Ω_{L2} and the Rabi-frequency of the off-resonant transition $(3 - 4')$ by $\Omega^{'}$ [21]. Note that since the same laser L2 addresses the transitions $F = 3-3'$ and $F = 3-4'$ the ratio of their Rabi strengths $\Omega_{L2}/\Omega^{'}$ is a constant.

The D2 line of ^{85}Rb has other excited state hyperfine states, $5P_{3/2}$ $F = 1'$ and $2'$ which do not affect our essential results. This is because the $F = 1'$ level does not participate in the Λ system as it is not accessed by the L2 laser and the transparency due to $F = 2'$ level need not be considered separately from that produced by $F = 3'$ as we will show later. So this renders the system effectively as a four-level system.

3 Experimental set-up

The experimental arrangement is shown in Figure 2. The L1 and L2 lasers were external cavity diode lasers (ECDLs). The output power of L1 laser was typically 20 mW and that of L2 Laser was 130 mW. Both L1 and L2 lasers were single mode at 780 nm with linewidths less than a few MHz. The vapour cell at room temperature contains both ^{85}Rb and ^{87}Rb in their natural abundance ratios. The lasers L1 and L2 are copropagating and collinear and overlap along the entire length of the cell. The L1 laser is focussed with a beam diameter at the waist of about 1 mm. A tunable Acousto-Optic-Modulator(AOM) is used in the path of the L2 laser to shift its frequency in the range of 60–100 MHz. With the help of the AOM the laser L2 can be locked at detunings δ_{L2} from $5P_{3/2}, F = 3'$, ranging from +60 MHz to −180 MHz.

The L1 and L2 lasers are linearly polarized with orthogonal polarizations. After the cell the two beams are separated using a polarizing beam splitter and their transmission recorded on a photodiode. The main diagnostic of the processes in the cell is by fluorescence. The fluorescence from the cell is collected by a femto-watt detector which is insensitive to the frequency of light incident on it. Fluorescence is recorded at a given detuning δ_{L2} for various scanned values of δ_{L1}. The L1 laser is scanned at the rate of 20 MHz/s, which is sufficiently slow for adiabatic transfer of populations between the various hyperfine levels.

4 Results and discussions

Unique to this level scheme is the role of laser L1. Since $F = 2$ is the lower of the two hyperfine states, in the absence of L1, significant population transfer between the ground state hyper-fine levels do not take place. It is clear that for a fixed L2 the fluorescence from the vapour cell will be a constant due to constant population in $|2\rangle$ (the state $5S_{1/2}, F = 3$). This fluorescence is predominantly from the "cyclic" transition $5S_{1/2}, F = 3$ and $5P_{3/2}, F' = 4$ ($|u\rangle$).

In the presence of a slowly scanned L1, optical pumping redistributes population between the ground state hyperfine levels. The resultant features in fluorescence are dependent on two factors: one is the effective difference in frequencies of L1 and L2. These are conveniently expressed as differences in detunings δ_{L1} and δ_{L2} from $F = 3'$. The other is the ratio of Rabi frequencies of L1 and L2 i.e. the ratios of Ω_{L1} and Ω_{L2}. It is to be noted that in the absorption study of a probe connecting a ground state and an excited state, its detuning from the excited state and the population of the ground state decide the signal strength. Our fluorescence study, is from a system consisting of several excited states, under the action of two laser fields L1 and L2. The fluorescence is dominated by an off-resonant transition whose ground state population gets redistributed due to optical pumping. So in parameter regimes where the optical pumping effect is dominant, fluorescence maximum results, when an atom of a certain velocity $\mathbf{v}$ sees

1 L1 on resonance with $|0\rangle$ ($\delta_{L1} - \mathbf{k}\cdot\mathbf{v} = 0$) and L2 resonant with $|u\rangle$ ($\delta_{L2} - 121$ MHz $- \mathbf{k}\cdot\mathbf{v} = 0$). Here 121 MHz is the hyperfine separation between the excited levels $3'$ and $4'$. This will enable this atom with velocity $\mathbf{v}$ to get maximally optically pumped from $|1\rangle$ to $|2\rangle$ and also be in resonance with the dominant fluorescing transition $|2\rangle$ and $|u\rangle$. We expect to see maximal fluorescence in this case;
2 L1 on resonance with $|0\rangle$ ($\delta_{L1} - \mathbf{k}\cdot\mathbf{v} = 0$) and L2 non-resonant with $|u\rangle$. In this case, though the optical pumping mechanism is efficient, because the atom is not resonant with the dominant transition ($|2\rangle - |u\rangle$), the resulting fluorescent maximum will not be as large as [1]. Nevertheless, in this case, if the atoms see any of the other excited state ($|0\rangle$ or $F' = 2$) on resonance ($\delta_{L2} - \mathbf{k}\cdot\mathbf{v} = 0$ or $\delta_{L2} - \mathbf{k}\cdot\mathbf{v} = -63$ MHz), then the fluorescence will be enhanced. Here 63 MHz is the hyperfine separation between $F = 3'$ and $F = 2'$;
3 L1 not on resonance and L2 on resonance with $|u\rangle$. Because of inefficient optical pumping, an excess fluorescence over background is not seen. The background fluorescence is maintained by the average population in $|2\rangle$.

The peculiar features of such velocity selected transfer resulting in narrow width fluorescence peaks have been experimentally studied by us [25] and theoretically explained in detail [26]. It has been found that any change the levels $|0\rangle$ and $F' = 2$ may undergo due to a strong L2 and a weak L1 can be brought out through these fluorescence studies. In particular for co-propagating and collinear L1 and L2, the Autler-Townes (AT) split positions of $|0\rangle$ and $F' = 2$, due to L2 at δ_{L2}, can be brought out in fluorescence, with a slowly scanned L1. At appropriate laboratory detunings $\delta_{L1}\pm$, the AT split positions of $|0\rangle$, due to L2 at δ_2, as seen by an atom with a velocity $\mathbf{v}$ is given by [27]

$$\delta_{L1\pm} - \mathbf{k}\cdot\mathbf{v} = \frac{\delta_{L2} - \mathbf{k}\cdot\mathbf{v}}{2} \pm \frac{1}{2}\sqrt{(\delta_{L2} - \mathbf{k}\cdot\mathbf{v})^2 + \Omega_{L2}^2}. \quad (1)$$

For large detunings δ_{L2}, Ω_{L2} may be neglected in comparison and hence the simplified equations are

$$\delta_{L1+} \approx \delta_{L2} \quad (2)$$

$$\delta_{L1-} - \mathbf{k}\cdot\mathbf{v} \approx 0 \quad (3)$$

$$\delta_{L1-} \approx \mathbf{k}\cdot\mathbf{v}. \quad (4)$$

These are the AT split positions of $|0\rangle$ as seen by an atom with velocity $\mathbf{v}$. It is evident that atoms of all velocity class see the level at δ_{L1+} (Eq. (2)) to be at the same position whereas the level at δ_{L1-} (Eq. (4)) appears shifted by different amounts for atoms of different velocity. It is through this velocity non-selective and velocity selective AT split levels of $|0\rangle$, that optical pumping takes place to $F = 3$ and results in fluorescence peak features as brought out in [25]. Trace B of Figure 3 brings out these fluorescence features due to optical pumping very clearly. For a fixed detuning $\delta_{L2} = 13$ MHz, we expect fluorescence peaks at

1 $\mathbf{k}\cdot\mathbf{v} = \delta_{L1-} = \delta_{L2} - 121$ MHz $= -108$ MHz. As explained earlier fluorescence peak results whenever an atom of a particular velocity class $\mathbf{v}$ sees L1 in resonance with one of the the AT split level of $|0\rangle$ (i.e., at δ_{L1-}) and L2 at resonance with $|u\rangle$. In trace B of Figure 3, fluorescence due to this is shown as a huge peak labeled β_1;
2 $\delta_{L1-} = \mathbf{k}\cdot\mathbf{v} = \delta_{L2} - 184$ MHz $= -171$ MHz. (Here 184 MHz is the hyperfine separation between $|u\rangle$ and $F' = 2$.) At this detuning, an atom of a particular velocity class $\mathbf{v}$ sees L1 in resonance with one of the AT split level of $F' = 2$ and L2 at resonance with $|u\rangle$. This results in a peak at β_2 at -171 MHz;
3 $\delta_{L1+} = \delta_{L2} = 13$ MHz. At this detuning, as explained earlier optical pumping of all velocity class of atoms from $|1\rangle$ to $|2\rangle$ takes place. This results in a fluorescence peak labelled α in Figure 3. Though there is an excess number over average of atoms in $|2\rangle$, these appear at different detunings to upper levels $|0\rangle$ and $|u\rangle$ for different velocities of atoms. Hence even though this results in a fluorescence peak at α, the peak is not as prominent as β_1 due to inefficient excitation processes to the upper levels.

Thus, if one were to see fluorescence as a function of detuning δ_{L1} from doubly driven scheme of Figure 1, with a weak L1 and a strong L2 one will see fluorescence *peaks* at $\delta_{L1+} = \delta_{L2}$, $\delta_{L1-} = \delta_{L2} - 121$ MHz and $\delta_{L1-} = \delta_{L2} - 184$ MHz due to Autler-Townes splitting of upper levels $|0\rangle$ and $F' = 2$. Incidentally, it has been pointed out in literature [27], that sub-Doppler and subnatural linewidth features are indeed possible for large detunings in the asymmetric AT level splitting case for the level at $\delta_{L1+} = \delta_{L2}$ and recently it has been experimentally verified [28]. It should be noted however that this reduction, is a reduction in the width of the *peak* in fluorescence and is an incoherent reduction and does not involve quantum cancellation of probability amplitudes as is the case with EIT.

Thus we see that for $\Omega_{L2} \gg \Omega_{L1}$, optical pumping is the dominant effect resulting in *peaks* in fluorescence. We will now discuss situations where the optical pumping effect is inhibited and the resultant signatures in fluorescence.

It is very well-known that destructive interference between dressed states in a three level system leads to cancellation of absorption at resonance and coherent population trapping (CPT) [1]. The resonance referred to here is the two-photon resonance and for the Λ sub-system of

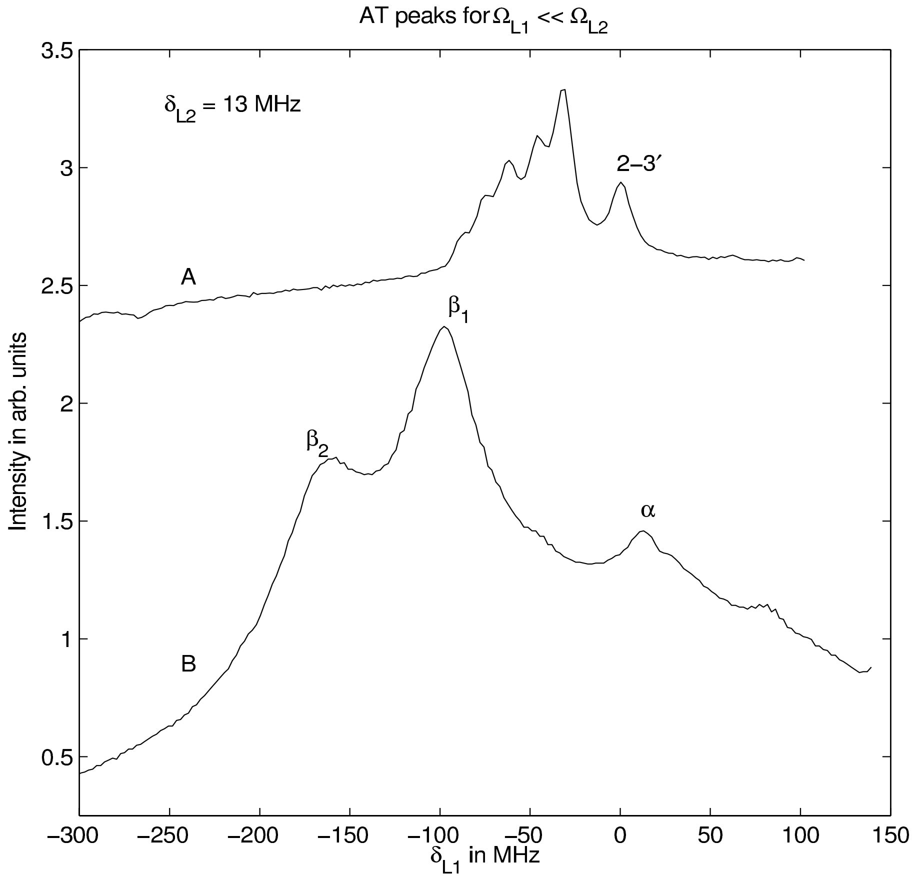

Fig. 3. Trace A gives the saturation absorption spectrum of L1 laser showing its instantaneous frequency. Trace B is a typical fluorescence spectrum from the cell when $\Omega_{L1} \ll \Omega_{L2}$. The peaks α, β_1 and β_2 are at 13 MHz, −108 MHz and −171 MHz respectively.

Figure 1, it occurs at $\delta_{L1} = \delta_{L2}$. This means, for negligible decay rates Γ_0 of the upper level $|0\rangle$ and Γ_2 of level $|2\rangle$ and for comparable strengths Ω_{L1} and Ω_{L2} the population gets locked in the dressed non-coupled state ($|NC\rangle$). Here $|NC\rangle$ is given by

$$|NC\rangle = \frac{\Omega_{L2}}{\sqrt{|\Omega_{L1}|^2 + |\Omega_{L2}|^2}}|1\rangle - \frac{\Omega_{L1}}{\sqrt{|\Omega_{L1}|^2 + |\Omega_{L2}|^2}}|2\rangle. \tag{5}$$

This results in quantum cancellation of absorption amplitudes to $|0\rangle$ from both the lower levels $|1\rangle$ and $|2\rangle$ resulting in transparency.

On closer examination of our system, we see that the two-photon resonance condition for transparency discussed above, for the three level system comprising the lower hyperfine levels $|1\rangle$ and $|2\rangle$ and the upper level $|0\rangle$, occurs at the same detuning $\delta_{L1+} = \delta_{L2}$, at which we got a peak in fluorescence due to optical pumping. For appropriate Ω_{L1} and Ω_{L2}, at this two-photon detuning, the three level Λ sub-system must enter in a coherent population trapped state (CPT) in which neither L1 nor L2 will be absorbed. In the absence of L1 absorption optical pumping of atoms do not take place and in the absence of L2 absorption to $|0\rangle$, fluorescence contribution from this level is decreased. However the off-resonant excitation by L2 to the unconnected level $|u\rangle$ does take place. We experimentally find that for $\Omega_{L1} > \Omega_{L2}$ this off-resonant excitation does not totally destroy the CPT state and the clear signature of transparency in the form of a sub-natural *dip* in fluorescence is seen. Figure 4 brings out these features for the same δ_{L2} as in Figure 3 but with $\Omega_{L1} > \Omega_{L2}$.

Trace A of this figure is the saturation absorption spectrum of L1, which gives the frequency of L1 at any instant during the scan. Figure 4B shows the fluorescence profile from the cell when laser L2 is blocked. We see that in the absence of laser L2, the fluorescence does not show any distinctive feature. For our experiment, this is taken as the background level. Trace 4C is a typical fluorescence profile in the presence of both the lasers L1 and L2 with $\Omega_{L1} > \Omega_{L2} > \Omega'$. There is a prominent dip in fluorescence around $\delta_{L1} = 13$ MHz. This dip is at the two-photon resonance condition

$$\delta_{L1} - \delta_{L2} = 0 \tag{6}$$

for the occurrence of EIT, in the Λ sub-system $|1\rangle$, $|2\rangle$ and $|0\rangle$. The dip has a sub-natural width FWHM = 5.6 MHz. This occurs at the same position α, where for the same detuning, a peak in fluorescence appeared (trace B of Fig. 3) due to optical pumping. We wish to point out that this is the same effect seen in absorption, in the four-level Λ scheme in the hyperfine manifold around the D1 line of sodium [20] (Fig. 2 in [20], trace (b) and trace (c)), at

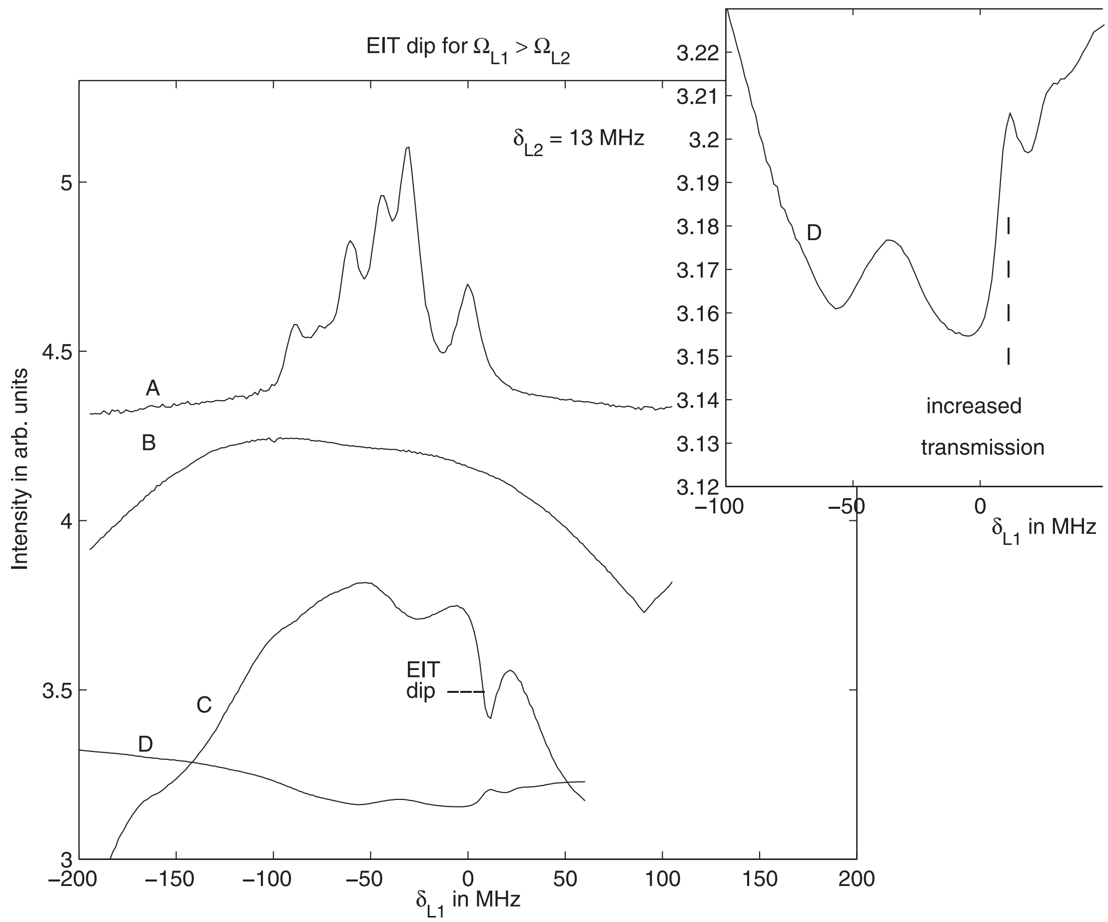

Fig. 4. Induced transparency effect in the three level Λ subsystem resulting in sub-natural fluorescence dip features. Trace A is the saturation absorption spectrum of L1 which serves as its instantaneous frequency marker. Trace B is the fluorescence signal in the absence of L2. Trace C is the signal in the presence of both L1 and L2. The only prominent feature in the signal is the sub-natural transparency dip of width 5.6 MHz. Trace D is the probe transmission of L1 which shows a corresponding increase at transparency (shown enlarged in the inset).

the two photon resonance detuning. In their paper also, they see a dip in probe transmission going to a peak in transmission as a function of increasing pump power.

In our experiment, accompanying this dip, we also see a loss of peaks at positions of β_1 and β_2. This is a very intriguing result because at these detunings we are far away from the two-photon resonance. We are tempted to argue that this is indeed a signature of adiabatic transfer of population from the ground state directly to the $|NC\rangle$ state.

The width of the dip is comparable to the natural width of the excited state in several of the experimental realisation. We attribute this to the lifting of Zeeman degeneracy due to earth's field. Nevertheless we argue that the mere presence of a dip in fluorescence can only point to existence of an underlying CPT state. The reasons are as follows.

- At the two-photon resonance, the transparency condition is satisfied both for $|1\rangle$, $|2\rangle$ and $|0\rangle$ subsystem and for $|1\rangle$, $|2\rangle$ and $F' = 2$ subsystem simultaneously. So even though there are 5 levels ($F = 2$, 3 and $F' = 2'$, $3'$ and $4'$, see Fig. 1) addressed by L1 and L2 together, at the two photon resonance, under transparency conditions there are no free levels available for absorption of L1. The $F' = 4$ level cannot absorb L1 due to selection rule violation. Even if the cyclic transition from $F = 2$ to $F = 1'$ exists, this does not cause redistribution of atoms between the ground hyperfine states. Because the system exhibits transparency for the two excited states $|0\rangle$ and $F' = 2$ simultaneously at the same two-photon resonance detuning, all the absorption pathways that transfer population through L1 are inhibited.
- During transparency the L2 laser is also not absorbed by $|0\rangle$ and $F' = 2$. The absorption of L2 by the off-resonant transition between $F' = 4$ and $F = 3$ (between $|u\rangle$ and $|2\rangle$) does not give rise to population redistribution (due to absence of absorption to $|0\rangle$). Thus this gives rise to a uniform feature in the entire transparency window since L2 is held fixed at a fixed detuning δ_{L2}.
- The transparency signal is also present when we look at the fluorescence profile at $\Omega_{L1} = \Omega_{L2}$ (Fig. 5). Here the EIT dip feature at α of sub-natural width 4.5 MHz

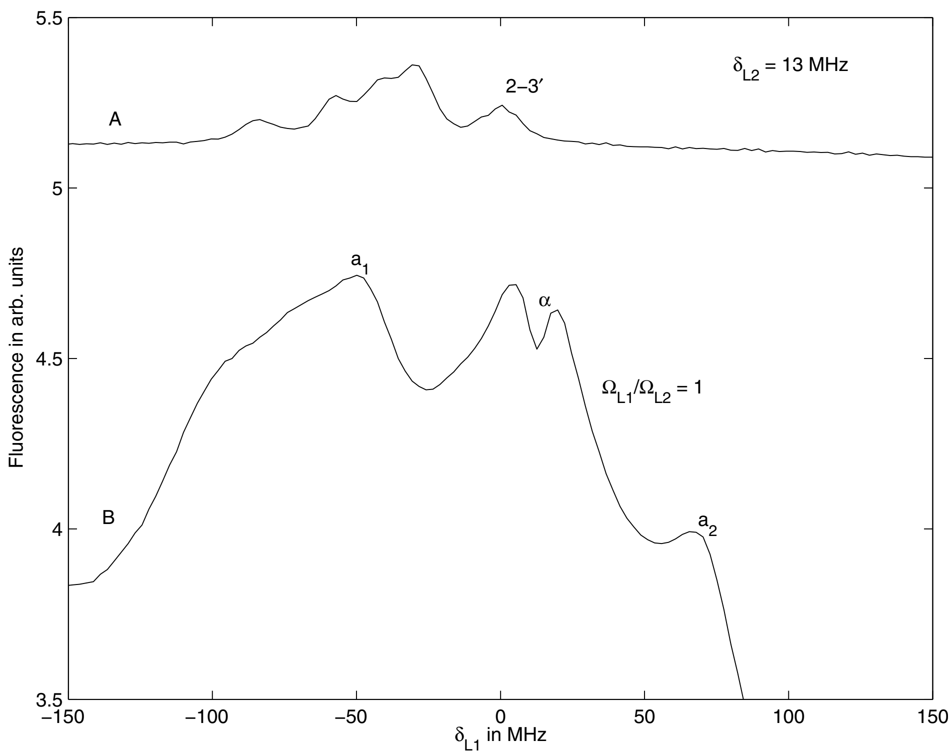

Fig. 5. Trace A denotes the saturation absorption spectrum of L1. Trace B shows the co-existence of transparency and optical pumping effects for $\Omega_{L1} = \Omega_{L2}$.

is centered in a peak of width 30 MHz. The central broad peak around α occurs due to optical pumping. As the intensity of Ω_1 is increased we see this feature washing out (trace C of Fig. 4). This is similar to what has been observed in absorption by [20] for comparable pump and probe field strengths. Most interestingly the satellite peaks at a_1 and a_2 occur due to the double Λ scheme shown in Figure 1. In the double Λ configuration neither L1 nor L2 address the same excited state. Instead they address two different excited states, $F = 3'$ (addressed by the fixed L2 laser) and $F = 2'$ (addressed by the scanned L1 laser). When the detunings of each of the lasers with respect to the different hyperfine level they address match, that is when δ_{L2} is fixed at 13 MHz from $F = 3'$, the detuning of L1 from $F = 2'$ matches that of δ_{L2} whenever $\delta_{L1} = 63 \pm 13$ MHz, then the same velocity class of atoms is addressed by both L1 and L2. In our case the laser L1 causes saturation of this velocity class for transitions $F = 2$ to $F = 2'$. Thus this velocity class becomes depleted in the ground hyperfine state $F = 2$. This is shown by a small circle if Figure 1. Since spontaneous emission from $F = 2'$ to $F = 3$ is permitted, the very same velocity class gets accumulated in $F = 3$. This is shown with a relatively bigger circle in the figure. This causes an increased absorption of L2 and hence an increased fluorescence at these frequencies. In literature this resonance is referred to as the cross-transition resonances [20]. This is also called as the difference frequency crossing resonance because for the case of degenerate ground hyperfine levels, this occurs whenever the difference between the laser frequencies ω_{L1} and ω_{L2} matches the hyperfine separation of the excited states. In our case, because of the non-degeneracy of ground hyperfine states, this occurs whenever $|\omega_{L1} - \omega_{L2}| \approx 3036 \pm 63$ MHz, where 3036 MHz is the ground hyperfine separation and 63 MHz is the hyperfine separation between $F = 2'$ and $F = 3'$. Thus it is seen on either side of the two-photon resonance where $|\omega_{L1} - \omega_{L2}| = 3036$ MHz. As L2 is held fixed at 13 MHz where our transparency dip α occurs, the DFC features appear approximately at -50 MHz (a_1) and 76 MHz (a_2) respectively.

Thus we see that, we have a pure transparency effect present for $\Omega_{L1} > \Omega_{L2}$. This appears in the form of a sub-natural dip in the fluorescence feature. Furthermore this is the only prominent feature in the entire spectrum (trace C of Fig. 4). The absence of any other feature even away from the two-photon resonance may be indicative of transfer of population directly from the ground state to the $|NC\rangle$ state.

A combination of optical pumping and transparency occurs for $\Omega_{L1} = \Omega_{L2}$. Here the system retains the coherence of the ground hyperfine levels which gives rise to the sub-natural transparency dip. However, the dephasing of the ground state coherence permits optical pumping even slightly away from the transparency window. This explains the peak feature centered on the transparency dip in Figure 5. In addition, the system also exhibits DFC resonances away from the two-photon resonance at a_1 and a_2.

We have seen earlier (Fig. 3) that when $\Omega_{L1} < \Omega_{L2}$ all trace of coherence vanishes. The system just has peaks at β_1, β_2 and α due to optical pumping through the incoherent level splitting of upper hyperfine levels $|0\rangle$ and $F' = 2$.

Thus in our experiment we see a transition from a coherent EIT effect, to coexistence of EIT and optical pumping effect, to ultimate destruction of coherence in the form of absence of transparency and saturation effects, and presence of optical pumping effects due to incoherent AT splitting. *It is clear from these arguments that a dip in fluorescence, if present, can only signify partial or complete coherence of the ground state hyperfine levels.*

It has been pointed out earlier that [18] the presence of off-resonant transitions only slightly decoheres the EIT signal. The presence of Zeeman sublevel due to earth's field has resulted in broadening of the peaks as is the case in [20].

We emphasise the crucial role played by the laser L1, as compared to the role played by L2. Since L2 is held fixed its non-absorption to the common excited states $|0\rangle$ and $F = 2'$ can at best cause a reduction in over-all DC fluorescence. But, on the other hand, it is the total inhibition of optical pumping pathways to L1 at two-photon resonance that causes maximal reduction in fluorescence from the off-resonant transition. This is clear when one compares Figures 3, 4 and 5. A clear requirement of $\Omega_{L1} > \Omega_{L2}$ for transparency (Fig. 4) and the reverse effect of the total absence of transparency (Fig. 3) with $\Omega_{L2} > \Omega_{L1}$ clearly indicates the dominant role played by L1 laser. Since L1 does not participate in the off-resonant excitation the dephasing due to this transition does not open up any optical pumping pathways to L1. Thus the transparency is robust even in the presence of these excitations. It might be argued that this is a generic requirement for transparency in such effective four-level systems.

The transparency effect and the absence of optical pumping has close analogies with systems described in [21] and [24]. In contrast to [21] we do not see here suppression of simultaneous absorption of two photons. The peak due to level splitting arises from a sequential absorption by the optically pumped atom into level $|2\rangle$ from level $|1\rangle$.

5 Numerical modelling

Using density matrix formalism we have numerically modelled the four-level system of Figure 1 with two copropagating laser beams L1 and L2. The analysis is very much on the lines of [26]. The four levels under consideration are: the two ground hyperfine levels $F = 2$, 3 ($|1\rangle$ and $|2\rangle$) and the two excited levels $F' = 3'$, $4'$ ($|0\rangle$ and $|u\rangle$). For simplicity, we have considered an one-dimensional situation, where the two driving fields are in the $\pm z$-direction and the atom is moving along the z-direction with a velocity $\mathbf{v}$. The detuning of the L1 laser taking the Doppler effect into account is $\Delta_{L1} = \delta_{L1} - \mathbf{k}\cdot\mathbf{v}$ and the L2 laser's detuning is $\Delta_{L2} = \delta_{L2} - \mathbf{k}\cdot\mathbf{v}$. The total Hamiltonian for the system consisting of the atom and the light fields is written in the interaction picture as

$$H = H_0 + H_I \tag{7}$$

where H_0 is the Hamiltonian for the bare atom and H_I is the atom-light interaction Hamiltonian. They are given as

$$H_0 = \hbar\omega_1|1\rangle\langle 1| + \hbar\omega_2|2\rangle\langle 2| + \hbar\omega_0|0\rangle\langle 0| + \hbar\omega_u|u\rangle\langle u|$$

and

$$\begin{aligned} H_I = -\frac{\hbar}{2}[&\Omega'|2\rangle\langle u| \exp(-i\omega_{L2}t) \\ &+\Omega_{L2}|2\rangle\langle 0| \exp(-i\omega_{L2}t) \\ &+\Omega_{L1} \exp(-i\omega_{L1}t)|1\rangle\langle 0| + H.C]. \end{aligned}$$

The dynamics of the system described by this Hamiltonian can be studied using the density matrix $\rho = \sum \rho_{ij}|i\rangle\langle j|$. The time evolution of the density matrix ρ is given by the Liouville equation

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[H, \rho] - \frac{1}{2}\{\Gamma, \rho\} \tag{8}$$

with Γ representing the relevant decay rates from various excited states. The rate equations of the four levels for an atom moving with a velocity $\mathbf{v}$ are derived under the rotating wave approximation [26]. For each velocity $\mathbf{v}$, steady state values of ρ_{ij} are obtained by numerically solving the 15 rate equations for the four levels, for various values of δ_{L1} and δ_{L2}, subject to the constraint $\rho_{11} + \rho_{22} + \rho_{00} + \rho_{uu} = 1$. Thus for an atom with a velocity $\mathbf{v}$ we obtain the population of each level and coherence between various levels, for a given value of δ_{L1} and δ_{L2}. The fluorescence emitted by atoms with a velocity $\mathbf{v}$ is given by

$$Fluorescence(\Delta_{L1}, \Delta_{L2}) = \Gamma_{uu}\rho_{uu} + \Gamma_{01}\rho_{00} + \Gamma_{02}\rho_{00}. \tag{9}$$

As the detector collects fluorescence from atoms which are in thermal motion, we take the average of each ρ_{ij} value over the range of velocities, weighted by the one dimensional Maxwellian velocity distribution. Thus, the Maxwell weighted averaged fluorescence, is calculated for a fixed δ_{L2} and for various scanned values of δ_{L1}. Figure 6 reproduces the essential experimental features. Trace A is the saturation absorption signal of L1. Trace B shows the computed fluorescence signal with $\Omega_{L1} > \Omega_{L2}$. Trace C is for $\Omega_{L1} = \Omega_{L2}$ and trace D for $\Omega_{L1} < \Omega_{L2}$. We see a nice transition from the coherent EIT effect for $\Omega_{L1} > \Omega_{L2}$ to the incoherent optical pumping effect at $\Omega_{L1} < \Omega_{L2}$ passing through a coexistence regime at $\Omega_{L1} = \Omega_{L2}$. Since this simulation does not involve the excited level $F' = 2$, peaks associated with this level are absent. Thus the peak at β_2 and the DFC peaks a_1 and a_2 are absent. A dressed state analysis of the full five level system is underway and will be discussed elsewhere.

The simulation also studied the role of off-resonant level $|u\rangle$. (The fluorescent contribution of this level from that of the level $|0\rangle$ cannot be distinguished in the experiment). Seen in Figure 7 is the total fluorescence calculated from the system (from levels $|0\rangle$ and $|u\rangle$) when

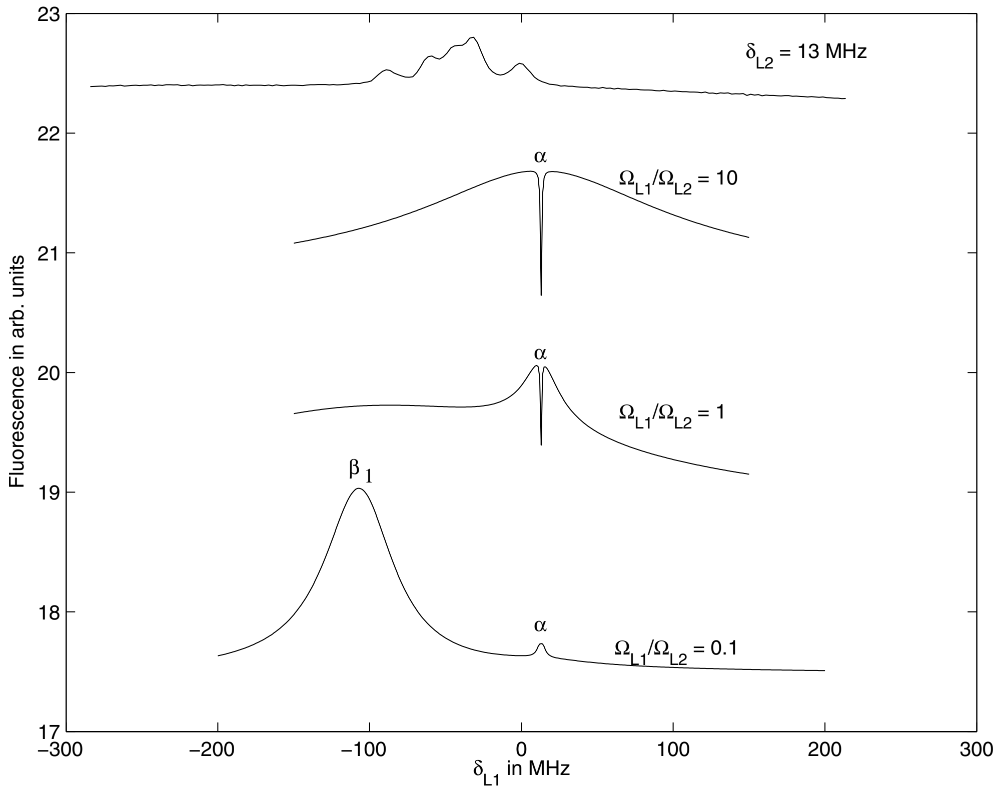

Fig. 6. Trace A is the saturation absorption spectrum of L1. Traces B, C, D are the numerically evaluated fluorescence from the four-level system for $\Omega_{L1}/\Omega_{L2} = 10$, 1, 0.1 respectively.

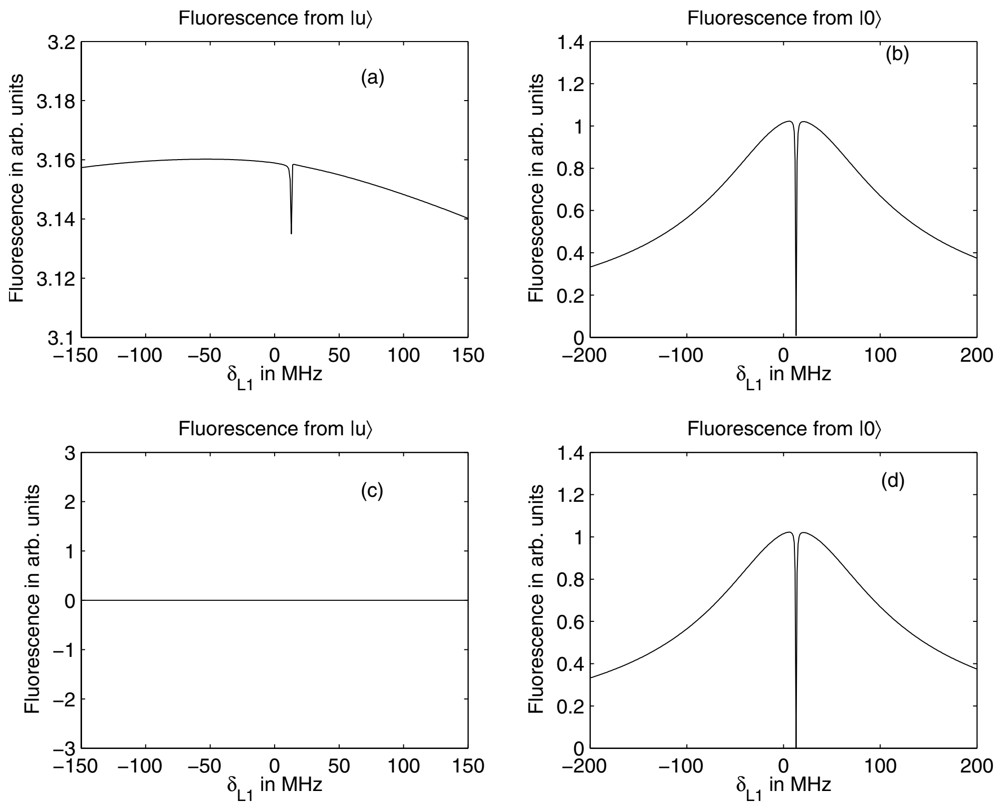

Fig. 7. Figures (a), (b), (c) and (d) show computed fluorescence spectra from $|u\rangle$ and from $|0\rangle$. The specific conditions are discussed in the text.

$\delta_{L2} = 13$ MHz. At the two-photon resonance condition $\delta_{L1} = \delta_{L2} = 13$ MHz, we see the transparency effect in the form of a dip in fluorescence from $|u\rangle$ AND in the form of a 100 percent dip in fluorescence, from $|0\rangle$ (Figs. 7a and 7b). A decrease in fluorescence from $|0\rangle$, is directly due to the transparency effect which results in CPT of atoms in the lower levels $|1\rangle$ and $|2\rangle$. A decrease in fluorescence from $|u\rangle$, is due to inhibition of optical pumping of atoms to the level $|2\rangle$ because of transparency related absence of absorption of L1. Unlike the dip in Figure 7b, the transparency dip from $|u\rangle$ in Figure 7a does not reach to zero because it is riding on the DC signal produced by the transition $|2\rangle - |u\rangle$. Had we frequency discriminated our fluorescence, this is what we would have seen from $|u\rangle$ and from $|0\rangle$. However, Figures 7a and 7b clearly bring out the fact that, the frequency insensitive fluorescence we see, is predominantly from $|u\rangle$. The fluorescence from $|0\rangle$, as shown in Figure 7b, is two times less than that from $|u\rangle$.

Figures 7c and 7d give the fluorescence from $|u\rangle$ and $|0\rangle$ respectively, when the off-resonant transition is switched off. We see the transparency signal from $|0\rangle$ to be the same as that of Figure 7b. It is therefore clear that the presence or absence of the non-resonant transition does not affect the underlying EIT effect provided $\Omega_{L1} > \Omega_{L2}$. Thus we see that the fluorescence dip is a good and reliable marker for the transparency effect.

6 Conclusions

An effective four level system around the D2 line of ^{85}Rb, is studied in fluorescence, under the action of two co-propagating and collinear fields L1 and L2. It was found that at two-photon resonance, the signature of the EIT effect in the underlying three level Λ subsystem is also present as a sub-natural dip in fluorescence, from an off-resonant fourth level. This effect is prominent when the strength of L1 (Ω_{L1}) exceeds that of L2 (Ω_{L2}) i.e., when $\Omega_{L1} > \Omega_{L2}$. We ascribe this to a complete loss of optical pumping pathways to L1 at transparency, combined with the selection rule non-availability of the off-resonant level to L1. At $\Omega_{L1} = \Omega_{L2}$, we get a coexistence regime, where we see that the dip in fluorescence due to transparency, being centered on a broader peak due to optical pumping. We also see in this regime, DFC resonances from the double Λ system present in our hyperfine manifold. When $\Omega_{L1} < \Omega_{L2}$, the transparency dip along with the DFC peaks vanish and the system losses all signs of coherent and cooperative effects. Instead the fluorescence now shows prominent peaks corresponding to the Autler-Townes split states of the excited state hyperfine levels, split due to the stronger laser L2. Thus we see, as a function of Ω_{L1}, a gradual transition from a purely coherent regime, which exhibits transparency, to the coexistence regime where the coherent transparency effect, the cooperative DFC effect and the incoherent optical pumping effect coexist, to a regime of total incoherence in the form of AT split states of excited levels. We emphasise the dominant role played by the laser L1 in maintaining a robust transparency signal. We argue that, the mere presence of a dip in fluorescence from such generic four-level or multi-level systems must signify transparency effects in the underlying three level subsystems. This, makes the identification of transparency easier. We reproduce the essential features of our experiment with a four-level density matrix simulation.

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