<table>
<thead>
<tr>
<th>項目</th>
<th>内容</th>
</tr>
</thead>
<tbody>
<tr>
<td>タイトル</td>
<td>電磁誘導透過と二段階励起の干涉</td>
</tr>
<tr>
<td>著者</td>
<td>早矢 望之; 藤澤 秀彦; 木戸 博明; 高橋 啓一; 深永 正弘</td>
</tr>
<tr>
<td>引用</td>
<td>Journal of the Optical Society of America B: Optical Physics, 27(8): 1645-1650</td>
</tr>
<tr>
<td>発行日</td>
<td>2010-07-26</td>
</tr>
<tr>
<td>種類</td>
<td>Journal Article</td>
</tr>
<tr>
<td>URL</td>
<td><a href="http://hdl.handle.net/2298/19641">http://hdl.handle.net/2298/19641</a></td>
</tr>
<tr>
<td>右側</td>
<td></td>
</tr>
</tbody>
</table>
Interference between electromagnetically induced transparency and two-step excitation in three-level ladder systems

Nobuhito Hayashi, Akihiko Fujisawa, Hiroaki Kido, Ken-ichi Takahashi, and Masaharu Mitsunaga

Graduate School of Science and Technology, Kumamoto University, 2-39-1 Kurokami, Kumamoto-shi, Kumamoto 860-8555, Japan

*Corresponding author: mitunaga@sci.kumamoto-u.ac.jp

Received April 21, 2010; revised June 9, 2010; accepted June 27, 2010; posted June 29, 2010 (Doc. ID 127199); published xx xx, xxxx

We have performed electromagnetically induced transparency (EIT) and two-photon absorption experiments in ladder-type three-level systems in a hot sodium atomic vapor, using the $3S_{1/2}-3P_{3/2}4D_{3/2}$, $3S_{1/2}-3P_{3/2}4D_{5/2}$, $3S_{1/2}-3P_{1/2}5S_{1/2}$, and $3S_{1/2}-3P_{3/2}5S_{1/2}$ transitions. In particular, in the most pronounced $3S_{1/2}-3P_{3/2}4D_{3/2}$ system, we have observed quite unique spectral line shapes that are superpositions of sharp dips and peaks, in contrast to ordinary EIT spectra. The peaks and dips have apparently different physical origins and the line shape can be interpreted as the interference between EIT and two-step excitation. © 2010 Optical Society of America

1. INTRODUCTION

Quantum interference phenomena in three-level ladder systems have been quite extensively studied since the first report in lead vapor [1]. Electromagnetically induced transparency (EIT) signals have been observed in ladder systems, for example, using Rb vapor [2–5] and Cs atoms [6], where the absorption of a weak probe beam (frequency $\omega_p$, and wave number $k_p$) resonant to the lower ladder transition is reduced in the presence of a strong coupling beam ($\omega_c$ and $k_c$) resonant to the higher ladder transition. The EIT theory and the signal line shapes for the ladder systems, so far, have been very similar to the Λ-type systems, with the main difference being that the two-photon coherence lifetime is much shorter for the ladder system and mainly given by the highest excited state lifetime. Interesting interference effects between four- and six-wave mixings in a ladder system have been studied [7–10]. Also, topics such as detection of Rydberg states [11], slow light [12], and optical polarization control [13,14] have all been studied using ladder-type systems.

Although the above-mentioned works have dealt with EIT in ladder systems, there is another important elementary process, two-step excitation (TSE), which is an incoherent part of two-photon absorption (TPA) [15–17], as a competing process against EIT. In TSE, the atoms are once transferred to level 2 (the intermediate level) by the probe beam and then to level 3 (the highest level) by the coupling beam (see Fig. 1 for the level configuration). In a Doppler broadened medium with a counterpropagating configuration, and if the two-photon resonance condition $\omega_p + \omega_c = \omega_{21}$ is satisfied, there is always a group of atoms with velocity $v$ which are single-photon resonant to both the probe wave ($\omega_{21} = \omega_p - k_p v$), and the coupling wave ($\omega_{32} = \omega_c + k_c v$), assuming that $k_p = k_c$. In this case the probe is further absorbed by this TSE process.

Both EIT and TSE take place only when the two-photon resonance is satisfied, but the behaviors are exactly the opposite in the probe transmission spectra. While EIT tends to increase the probe transmission and the signals are observed as upward peaks, TSEs should further increase the probe absorption and they are observed as downward dips. Then the problem arises as to which process is dominant and what line shapes we should expect when both processes coexist. This problem has been left unnoticed so far.

In this paper we report for the first time, to the best of our knowledge, the observation of ladder-type EIT in a hot sodium vapor. We have mainly studied the $3S_{1/2}-3P_{3/2}4D_{3/2}$ ($D1-4D$) transition. The probe transmission spectra had a quite unique feature showing anomalous resonance line shapes within the Doppler broadened background. Such line shapes have never been observed before. Experimental findings strongly suggest that this anomalous line shape can be attributed to the interference of TSE and EIT. Simultaneous measurements of the $3P$ and $4D$ fluorescence spectra supported this idea. We have also studied $3S_{1/2}-3P_{3/2}4D_{3/2,5/2}$ ($D2-4D$), $3S_{1/2}-3P_{3/2}5S_{1/2}$ ($D1-5S$), and $3S_{1/2}-3P_{3/2}5S_{1/2}$ ($D2-5S$) systems. In the case of $D2-4D$, a similar type of interference pattern was observed. For $D1-5S$ and $D2-5S$ systems, however, two-photon resonance spectra were much simpler and no interference effects were observed.

As a similar type of works, inhibition of TPA by using EIT has been extensively studied [18–20], but they needed a third laser beam to connect the intermediate level to a fourth level. In our case the physical mechanism is totally different and no third beam is necessary, and the
interference can be observed with simple three-level ladder systems using only two input beams. Also, a phenomenon called electromagnetically induced absorption (EIA) [21–23] exhibits a sharp extra absorption at the two-photon resonance point. But it is observed only for Zeeman sublevels within the same hyperfine level and the physical mechanism is again different from our case. In particular, EIT and EIA do not coexist. We will first show our experimental results in the next section, which is followed by the theoretical explanation and discussion of our observations.

2. EXPERIMENT

A. Setup

Our experimental setup is illustrated in Fig. 2 [24,25]. In a typical ladder-type experimental setup, one needs two independently tunable single frequency dye lasers (RDLs). One (RDL 1, Coherent Radiation CR899-21) was tuned to the $3S_{1/2}-3P_{1/2}$ (D1 line, 589.6 nm) or $3S_{1/2}-3P_{3/2}$ (D2 line, 589.0 nm) transition as the probe beam, and the other (RDL 2, Coherent Radiation CR699-21) was tuned to the $3P_{1/2}-4D_{3/2}$ (568.3 nm), $3P_{3/2}-4D_{3/2,5/2}$ (568.8 nm), $3P_{1/2}-5S_{1/2}$ (615.4 nm), or $3P_{3/2}-5S_{1/2}$ (616.0 nm) transition (see Fig. 1 for the related energy-level diagrams) as the coupling beam. These two beams collinearly counterpropagated to each other in a glass cell of length 7.5 cm containing Na atoms without any buffer gas so that the two-photon resonance condition is quasi-Doppler free and the residual Doppler broadening is estimated to be 60 MHz. (When the two beams copropagate, we did not observe any EIT or TSE signals.) In the configuration of Fig. 2, the probe and coupling polarizations are linear and orthogonal due to polarizing beam splitters (PBSs). We also tried a noncollinear configuration without using a PBS and, in this case, the choice of polarizations for probe and coupling is arbitrary. The transverse beam qualities of the two beams were improved by employing a spatial filter for the coupling beam and a single mode fiber for the probe beam. The probe transmission signal and the fluorescence signal from the 4D or 3P state were simultaneously monitored by using a photodetector and a photomultiplier tube (PMT), respectively. Interference filters for 586, 589, and 620 nm were placed in front of the PMT for the fluorescence detection from 4D, 3P, and 5S levels, respectively. Typical coupling and probe powers were 300 and 1 mW, respectively, unless it is specified.

B. D1-4D System

First we show in Fig. 3 a typical probe transmission spectrum along with 3P and 4D fluorescence spectra for the D1-4D system as a function of the probe detuning frequency. Two sharp signals corresponding to $3S_{1/2}-3P_{1/2}-4D_{3/2}$ (D1-4D) system or $3S_{1/2}-3P_{3/2}-4D_{3/2,5/2}$ (D2-4D) system. (b) $3S_{1/2}-3P_{3/2}-5S_{1/2}$ (D1-5S) system or $3S_{1/2}-3P_{3/2}-5S_{1/2}$ (D2-5S) system.

![Fig. 1. (Color online) Related energy-level schemes for the ladder-type EIT in sodium vapor.](image1)

![Fig. 2. (Color online) Schematic of the experimental setup: RDL, ring dye laser; PBS, polarizing beam splitter; HWP, half-wave plate; SMF, single mode fiber; PD, photodetector; PMT, photomultiplier tube; IF, interference filter; OSC, oscilloscope.](image2)

![Fig. 3. Typical probe transmission spectrum (top trace) for D1-4D system, fluorescence spectrum from the $3P_{1/2}$ level (middle trace), and fluorescence spectrum from the $4D_{3/2}$ level (bottom trace) as a function of probe detuning frequency in the presence of coupling beam. Coupling detuning is 0 GHz.](image3)
The main question of the results in Fig. 3 is of course the anomalous resonance line shapes. Is it a single process or a combination of two or more independent processes? In order to investigate the signal behavior in more detail, the coupling power $I_c$ dependence with the probe power $I_p$ fixed at 0.98 mW was studied and the results are shown in Fig. 4. Clearly, when the coupling power is low (17 mW), only the downward dip signals are observed. As $I_c$ is increased, the central peaks grow up and become quite pronounced when $I_c$ = 358 mW. In this way the dips and the peaks have different $I_c$ dependences, indicating that they are different elementary processes, and now it is reasonable to attribute dips to TSE and peaks to EIT and the signals are simply superpositions of TSE and EIT.

We have also investigated probe power $I_p$ dependence of the spectra with $I_c$ fixed at 364 mW as shown in Fig. 5. In contrast to Fig. 4, this time only EIT components are observed for low probe powers and the TSE components grow up as the probe power is increased. This behavior is explained by the fact that EIT is a linear process and TSE is a nonlinear process with respect to probe amplitude, and so in the limit of weak probe amplitude, only the EIT component survives.

As will be discussed in the next section, the TSE signals are incoherent and always show simple Lorentzian line shapes. On the other hand, the coherent EIT line shape depends on the spectral positions within the Doppler profile and it is Lorentzian in the middle and is dispersion type in the wing. These behaviors are experimentally checked and Fig. 6 shows the results of the coupling frequency dependence of the probe transmission spectra. As can be seen in Fig. 6, symmetrical EIT-TSE combined line shapes could be observed only when the coupling detuning frequency was in the middle of the Doppler broadened profile ($\delta_0 = 0$). When $\delta_0$ was shifted to the higher or lower frequency the EIT line shapes became dispersion type. This finding once again supports the idea that the signals are due to interference between TSE and EIT.

![Fig. 4.](image-url)

**Fig. 4.** Probe transmission spectra for D1-4D system when the coupling power is varied as 358, 197, 132, 75, and 17 mW.

![Fig. 5.](image-url)

**Fig. 5.** Probe transmission spectra for D1-4D system when the probe power is varied as 3.1, 2.1, 1.1, 0.55, and 0.23 mW.

### C. D2-4D System

Next we show the results of the D2-4D system in Fig. 7. This time we have four peaks because both $4D_{3/2}$ and $4D_{5/2}$ levels, separated by 1028 MHz, are two-photon allowed and so the four peaks correspond to $3S_{1/2}(F = 2)$-to-$4D_{3/2}$, $3S_{1/2}(F = 2)$-to-$4D_{5/2}$, $3S_{1/2}(F = 1)$-to-$4D_{5/2}$, and $3S_{1/2}(F = 1)$-to-$4D_{3/2}$ two-photon transitions. The signal line shape for the $F = 1$-to-$4D_{5/2}$ transition is similar to the D1-4D case, but the $F = 2$-to-$4D_{5/2}$ case is more complicated, having further lobes in the wings. The other two peaks for $4D_{3/2}$ levels are not symmetrical. But this is simply because of the spectral position, and when the coupling frequency was adjusted such that they were in the middle, then these two peaks gave symmetrical interference signals and in turn the signals for $4D_{5/2}$ became asymmetrical dispersive type. Therefore the interference could be observed for all the four peaks.

![Fig. 6.](image-url)

**Fig. 6.** Probe transmission spectra for D1-4D system when the coupling detuning frequency is varied as 1.891, 1.271, 0, −1.071, and −1.887 GHz.
D. 1-5S System

In the case of the 5S_{1/2} final level, the spectra were simpler but very interesting. Figure 8 shows the results of the D1-5S system. In this case we have four peaks corresponding to the \( F = 2 \) to \( F' = 1 \), \( F = 2 \) to \( F' = 2 \), \( F = 1 \) to \( F' = 1 \), and \( F = 1 \) to \( F' = 2 \) two-photon transitions with the initial 3S_{1/2} and final 5S_{1/2} levels. (The hyperfine splitting frequency of the 5S_{1/2} level is 150 MHz.) In this case the peaks are simple Lorentzian dips, implying that only TSE is observed. Polarization dependence was checked by setting (probe and coupling) as (V and H), (H and H), (\( \sigma_+ \) and \( \sigma_+ \)), and (\( \sigma_+ \) and \( \sigma_- \)), but no apparent change was observed for the four cases. In the case of coherent 3S-5S TPA [15,16], only the polarization combinations (H and H) and (\( \sigma_+ \) and \( \sigma_+ \)) satisfying the selection rules \( \Delta m = 0 \) gave the absorption signals. The fact that we observe signals in every case strongly suggests that this is an incoherent stepwise excitation to the 5S_{1/2} level.

E. D2-5S System

Finally we show the results of the D2-5S system in Fig. 9. Similarly to the previous case, we have four signals but this time they are upward peaks in contrast to downward dips in Fig. 8. The signals, however, sometimes become dips depending on the polarization conditions. [Especially, the \( F = 1 \) to \( F' = 1 \) signals become dips when (probe and coupling) polarizations are (H and H) and (\( \sigma_+ \) and \( \sigma_- \)).]

The physical mechanism of which (peak or dip) is dominant for each particular system is still unclear and should be investigated further. Anyway, with the 5S level as the final level of the ladder system, we did not observe any evidence of interference pattern as we did with the 4D levels.

3. THEORY AND DISCUSSION

Anomalous two-photon resonance line shapes observed in Figs. 3–7 cannot be explained within the framework of an ordinary EIT theory, which gives much simpler line shapes. The EIT theory assumes the probe field \( \mathcal{E}_p \) to be weak, and so only the linear susceptibility with respect to \( \mathcal{E}_p \) is calculated. On the other hand, the TPA process requires higher order terms for \( \mathcal{E}_p \) in order to populate the highest level. In order to calculate the absorption coefficient \( \alpha \), then, we will have to obtain the higher order solutions up to the fifth order (up to the terms with \( |\mathcal{E}_p|^2 |\mathcal{E}_p|^2 \mathcal{E}_p \) ) by the perturbative approach.

The Liouville equations governing the three-level ladder-type system as shown in Fig. 1 are written as

\[
\dot{\rho}_{33} = \frac{i}{2} \Omega_3 \rho_{23} - \frac{i}{2} \Omega_3 \rho_{32} - \Gamma_3 \rho_{33},
\]
where $\Omega_p$ ($\Omega_c$) is the Rabi frequency of probe (coupling), $\gamma_{ij}$ is the population decay rate from level $i$ to level $j$, and $\Gamma_j=\sum\gamma_{ij}$. $\gamma_{ij}$ is the dephasing rate between levels $i$ and $j$, and $\delta_i=\omega_{21}-\omega_p$, $\delta_1=\omega_{12}-\omega_c$, and $\delta_0=\delta_1+\delta_2$ is the two-photon detuning.

The steady-state solutions of Eq. (1) are obtained in a perturbative manner from the zeroth to the fifth order. Among them the third order term gives EIT, and the fifth order terms give TPA. The TPA terms are divided basically into two: coherent and incoherent terms. The incoherent term does not require two-photon coherence and is called TSE. We take TSE as the dominant TPA process, because it is the only term that gives the broad absorption spectrum and the other terms give sharp line shapes determined by the two-photon coherence $\gamma_{12}$. Taking into account the Doppler broadening, the probe absorption coefficient $\alpha(\delta_p)$ as a function of the probe detuning $\delta_p=\delta_0+\delta_2+\delta_3$, and $\delta_3=k_gH$ is the Doppler shift) can be written as

$$\alpha(\delta_p)=\alpha_0\int D(\delta_p)\left[\int L_p+\left(\frac{\Omega_p^2}{\gamma_{12}}\frac{\Omega_p^2}{\gamma_{12}^2}\right)^2 L_c^2\right],$$

(2)

where $\alpha_0$ is the peak absorption coefficient, $D(\delta_p)$ is the normalized Doppler distribution, $L_p=\gamma_{12}/(\gamma_{12}^2+\delta_p^2)$, $L_c=\gamma_{23}/(\gamma_{23}^2+\delta_c^2)$, and $\delta_0=\omega_r-\omega_{21}$. Here we assumed that probe and coupling are counterpropagating so that the Doppler shifts have opposite signs.

The physical origins of the three terms appearing in Eq. (2) are very clear. The first term simply shows the linear absorption and gives the Doppler broadened background. The second term, having the same sign as the first, represents further absorption when $\delta_0+\delta_2=0$ and is simultaneously satisfied and gives TSE contribution. The third term, on the other hand, shows the reduction in absorption, indicating EIT. Having $\Omega_p^2$ dependence, TSE becomes negligible when the probe power is infinitely small; meanwhile EIT has no probe power dependence.

This is consistent with what we found in Fig. 5. While the linewidth of TSE is given by $\gamma_{12}$ and $\gamma_{23}$, that of EIT is basically given by $\gamma_{13}$, which is usually smaller than $\gamma_{12}$ or $\gamma_{23}$ due to a rather long lifetime of the highest level.

(The actual values for the D1-4D system are $\gamma_{13}/2\pi=0.8$ MHz, $\gamma_{12}/2\pi=5$ MHz, and $\gamma_{23}/2\pi=5.8$ MHz [12].)

Numerically, however, the two laser linewidths (~1 MHz each) should be added to each constant.) Thus we naturally expect that the two-photon resonance signal consists of a broad TSE dip and a narrow EIT peak in the Doppler broadened background. Figure 10 shows a typical example of numerical simulation based on Eq. (2) with appropriate parameters given in the caption and agrees very well with the experimental data. We should also notice that, while the TSE term gives always Lorentzian-type spectra regardless of the spectral position, the EIT term changes its line shape from a Lorentzian in the center of the Doppler profile to a dispersion type in the wing. Experimental results shown in Fig. 6 are a clear manifestation of this behavior. Although Eq. (2) can qualitatively explain very well the experimental findings, it is a perturbative approach and it does not work for strong coupling beams. A more elaborate theory including the effect of coupling saturation will be needed.

4. SUMMARY

We have, for the first time, reported three-level ladder EIT and TPA experiments in sodium vapor. The two-photon resonance signals within the Doppler broadened profiles are observed in all the systems we have studied, i.e., D1-4D, D2-4D, D1-5S, and D2-5S. As the most pronounced example, we have investigated the D1-4D system in detail. In contrast to ordinary ladder EIT spectra, the two-photon resonance line shapes are quite anomalous having sharp peaks and broader dips, strongly suggesting that the strange line shapes are attributable to interference between the two elementary processes: EIT and TSE. Theoretical analyses and numerical simulation qualitatively support this idea. A detailed comparison between theory and experimental results will be reported in a separate paper.
REFERENCES


#1 AU: One author must be designated as the corresponding author. Please check that this is the one you want to use.