Abstract: We experimentally compare the probe transmission, four-wave mixing (FWM) and fluorescence signals under dressing effects for the first time. Especially, the interplay between two ladder subsystems is investigated in the $Y$-type atomic system. Moreover, the two-photon fluorescence signal with ultra-narrow linewidth is obtained, which is much narrower than the Doppler-free electromagnetically induced transparency window. Such fluorescence with very high coherence and monochromaticity can be potentially applied in metrology and quantum correlation.

Opening fluorescence and four-wave mixing via dual electromagnetically induced transparency windows

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Received: 9 May 2012, Revised: 29 May 2012, Accepted: 1 June 2012
Published online: xx xxxx 2012

Key words: fluorescence; four-wave mixing; electromagnetically induced transparency

1. Introduction

Enhanced four-wave mixing (FWM) process has been experimentally reported in multi-level atomic systems [1–3]. Therein, the Autler-Townes (AT) splitting has been investigated [4]. The keys in such enhanced nonlinear optical processes include greatly reduced linear absorption of the generated optical fields due to electromagnetically induced transparency (EIT). A great deal of attention has been paid to observe and understand EIT and related effects in multi-level atomic systems interacting with electromagnetic fields [5–8]. Under EIT conditions, not only can the FWM signals be allowed to transmit through the atomic medium but also the fluorescence induced by spontaneous emission can be generated [9–11]. In the fluorescence spectrum the AT splitting has also been reported in lithium molecules [9].

In this letter, we compare the probe transmission, FWM and fluorescence signals under dressing effects for the first time. The ultra-narrow two-photon fluorescence signal, which is sheared twice by EIT window, is obtained in ladder or $Y$-type atomic system. Such fluorescence with very high coherence and monochromaticity can be potentially applied in metrology long-distance quantum communication and quantum correlation. Also, we investigate the interaction effect between two ladder subsystems on the measured signals. Moreover, the amplitude of the signals can be effectively controlled by the incident beam intensity and frequency detuning.

2. Theoretical model and experimental scheme

The experiment is carried out in atomic vapor of $^{85}$Rb. The energy levels $5S_{1/2} (F = 3) (|0\rangle), 5P_{3/2} (|1\rangle), 5D_{5/2} (|2\rangle)$, and $5D_{3/2} (|3\rangle)$ form the four-level $Y$-type system
as shown in Fig. 1a. The vapor cell is set at $60^\circ$. A weak laser field $E_1 (\omega_1, k_1, \text{Rabi frequency } G_1, \text{wavelength } 780 \text{ nm})$ from an external cavity diode laser (ECDL) with horizontal polarization probes the lower transition $|0\rangle$ to $|1\rangle$. Two strong coupling fields, $E_2 (\omega_2, k_2, G_2)$, and $E'_2 (\omega_2, k'_2$ and $G'_2$), each with a vertical polarization and wavelength 775.98 nm, are split from a continuous wave (CW) Ti:Sapphire laser, driving the upper transition $|1\rangle$ to $|2\rangle$. Another two strong coupling fields, $E_3 (\omega_3, k_3$ and $G_3)$ and $E'_3 (\omega_3, k'_3$ and $G'_3$), each with a vertical polarization and wavelength 776.16 nm, are split from another ECDL laser, driving the upper transition $|1\rangle$ to $|3\rangle$.

The laser beams are aligned spatially in the square pattern (Fig. 1b). Four coupling beams propagate through Rb vapor in the same direction with small angles ($\sim 0.3^\circ$) between one another and the probe field $E_1$ propagates in the opposite direction from them. Thus, two ladder-type subsystems ($|0\rangle - |1\rangle - |2\rangle$ and $|0\rangle - |1\rangle - |3\rangle$) form and two EIT windows appear in the probe transmission spectrum [4], which is measured by a silicon photodiode. Also, two FWM singles $F1$ (satisfying the phase-matching condition $k_{F1} = k_1 + k_2 - k'_2$) and $F2$ (satisfying $k_{F2} = k_1 + k_3 - k'_3$) will generate within the two EIT windows and propagate in the same direction as shown in Fig. 1b, which are detected by an avalanche photodiode detector (APD). Besides, three types of fluorescence due to spontaneous emission are detected in our experiment: the decay of photons from $|1\rangle$ to $|0\rangle$ will generate single-photon fluorescence signal $R0$ (wavelength 780 nm), and the decay of photons from $|2\rangle$ and $|3\rangle$ to $|1\rangle$ will generate two-photon fluorescence signals $R1$ and $R2$ (wavelength 776 nm), as shown in Fig. 1a. Compared with FWM, the fluorescence signals are non-directional and detected by another photodiode. The two-photon fluorescence signals can also fall into the EIT window and form the Doppler-free sharp peak.

In general, the expression of the density-matrix element $\rho_{10}^{(3)}$ related to the FWM processes and density-matrix elements $\rho_{11}^{(1)}, \rho_{22}^{(2)},$ and $\rho_{33}^{(4)}$ related to the fluorescence processes can be obtained by solving the coupled density-matrix equations. First, with $E_1$, $E_2$, and $E'_2$ on $(E_3$ and $E'_3$ blocked), the simple FWM process $F1$ via the Liouville pathway

$$\rho_{10}^{(0)} \xrightarrow{i\Delta_1} \rho_{10}^{(1)} \xrightarrow{i\Delta_2} \rho_{10}^{(2)} \xrightarrow{i\Delta_1} \rho_{10}^{(3)}$$

gives

$$\rho_{F1}^{(3)} = -iG_a \exp \{i[k_{F1} \cdot \mathbf{r}] \},$$

where

$$G_a = G_1 G_2(G'_2)^*, \quad d_1 = \Gamma_{10} + i\Delta_1,$$

and

$$d_2 = \Gamma_{20} + i(\Delta_1 + \Delta_2)$$

with frequency detuning $\Delta_1 = \Omega_i - \omega_1$ ($\Omega_i$ is the atomic resonance frequency for the corresponding transition), and $\Gamma_{ij}$ is the transverse relaxation rate between states $|i\rangle$ and $|j\rangle$. Considering the self-dressing effect of $E_2$ and $E'_2$, the energy level $|1\rangle$ could be split into two dressed states $|+\rangle$ and $|-\rangle$, which can be described via the Liouville pathway

$$\rho_{00}^{(0)} \xrightarrow{i\Delta_1} \rho_{\pm 0}^{(1)} \xrightarrow{i\Delta_2} \rho_{\pm 0}^{(2)} \xrightarrow{i\Delta_1} \rho_{\pm 0}^{(3)}$$

and the expression of $\rho_{F1}^{(3)}$ can be modified as

$$\rho_{F1SD}^{(3)} = -iG_a \exp \{i[k_{F1} \cdot \mathbf{r}] \} \left( d_1 + \frac{|G|^2}{d_2} \right)^2 d_2.$$
Next, when the coupling field $E_3$ is turned on, its dressing effect should also be considered, resulting in the double dressed FWM process as

$$P_{1DD}^{(3)} = -iG_a \exp \left\{ i k F_1 \cdot r \right\} \frac{d_1}{d_1 + \frac{|G_2|^2}{d_2}} \frac{d_2}{d_2 + \frac{|G_3|^2}{d_3}},$$

where $d_1 = \Gamma_{30} + i(\Delta_1 + \Delta_3)$. Similarly, with $E_1$, $E_3$, and $E'_3$ on ($E_2$ and $E'_2$ blocked), the other FWM process $F_2$ via the Liouville pathway

$$\rho_{10}^{(0)} \rightarrow \rho_{11}^{(1)} \rightarrow \rho_{10}^{(2)} \rightarrow \rho_{10}^{(3)}$$

gives

$$P_{2DD}^{(3)} = -iG_b \exp \left\{ i k F_2 \cdot r \right\} \frac{d_1}{d_1 + \frac{|G_2|^2}{d_3}}$$

with $G_b = G_1G_3(G_3')^*$, which can also be singly or doubly dressed.

For the fluorescence, the fluorescence signal $R_0$ is described by

$$\rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{30}^{(2)} \rightarrow \rho_{10}^{(3)} \rightarrow \rho_{10}^{(4)}$$

By solving the coupled density-matrix equations, the expression of the density-matrix element $\rho_{11}^{(0)}$ can be obtain as

$$\rho_{11}^{(2)} = \frac{-|G_1|^2}{d_1 I_{11}},$$

the amplitude squared of which is proportional to the intensity of $R_0$. When the beams $E_2$ and $E_3$ are turned on, the fluorescence process $R_0$ is also described, by the Liouville pathway

$$\rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{30}^{(2)} \rightarrow \rho_{10}^{(3)} \rightarrow \rho_{10}^{(4)}$$

Therefore the expression of $\rho_{11}^{(2)}$ can be modified as

$$\rho_{11}^{(2DD)} = \frac{-|G_1|^2}{I_{11} \left( d_1 + \frac{|G_2|^2}{d_2} + \frac{|G_3|^2}{d_3} \right)}.$$  

For signal $R_1$, via Liouville pathway

$$\rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{20}^{(2)} \rightarrow \rho_{21}^{(3)} \rightarrow \rho_{22}^{(4)}$$

we can obtain the density-matrix element $\rho_{22}^{(4)}$ as

$$\rho_{22}^{(4)} = \frac{|G_1|^2|G_2|^2}{I_{22}d_1d_2d_4},$$

where $d_4 = I_{21} + i\Delta_2$, the amplitude squared of which is proportional to the intensity of $R_1$. Considering the self-dressing effect of $E_2$ and $E'_2$, the expression of $\rho_{22}^{(4)}$ can be modified as

$$\rho_{22}^{(4)} = \frac{|G_1|^2|G_2|^2}{I_{22}d_1d_4 \left( d_4 + \frac{|G_2|^2}{d_1} \right)}.$$  

When the beam $E_3$ is turned on, its dressing effect should be considered and the doubly dressed fluorescence process $R_1$ is given as

$$\rho_{22}^{(4DD)} = \frac{|G_1|^2|G_2|^2}{I_{22}d_1d_2d_4 \left( d_4 + \frac{|G_2|^2}{d_1} \right)}.$$  

Correspondingly, the fluorescence signal $R_2$ is related with the density-matrix element

$$\rho_{33}^{(4)} = \frac{|G_1|^2|G_3|^2}{I_{33}d_1d_2d_5},$$

and doubly dressed R2 process is given as

$$\rho_{33}^{(4DD)} = \frac{|G_1|^2|G_3|^2}{I_{33}d_1d_2d_5 \left( d_5 + \frac{|G_3|^2}{d_1} \right)}.$$  

3. Fluorescence and FWM via EIT windows

Fig. 2 presents the measured FWM and fluorescence signals opened via the EIT windows versus the probe detuning $\Delta_1$. First, with all five beams on and $P_2 = 1$ mW, the measured curves under different $\Delta_2$ are depicted in Fig. 2a1 – Fig. 2a4. In the probe transmission signals, two EIT windows arise at $\Delta_2 = -\Delta_2$ and $\Delta_1 = -\Delta_3$ (labeled as P1 and P2) within the Doppler absorption background. The FWM signals F1 and F2 fall into the two EIT windows respectively. As $\Delta_2$ changes, the EIT window P1 and FWM signal F1 shift from left to right, and overlap with the fixed EIT window P2 and FWM signal F2 at $\Delta_2 = \Delta_3 = 0$ (Fig. 2a3). For the fluorescence signals, the big background curve represents the 780 nm fluorescence R0 ($\rho_{11}^{(2)}$). The other two small sharp peaks on it are the 776 nm fluorescence R1 ($\rho_{22}^{(4)}$) and R2 ($\rho_{33}^{(4)}$) falling into the EIT windows. The intensity of fluorescence R1 reaches its maximum at the resonant point ($\Delta_2 = 0$ MHz, Fig. 2a3) and decreases gradually as $\Delta_2$ is set farther from resonance (from Fig. 2a3 to Fig. 2a1), due to the effect of single-photon term $d_1$ and $d_4$ in $\rho_{22}^{(4)}$. Next, when $P_1$ increases to 6 mW, the dressed signals are shown in Fig. 2b1 – Fig. 2b4 where $E'_3$ is blocked. Here, the FWM signal F2 disappears due to the absence of $E'_3$ and the fixed FWM signal F1 shows AT splitting from the self-dressing effect of $E_3 (E'_2)$, denoting by the dressing term $|G_2|^2 / d_4$ in $\rho_{11}^{(2DD)}$. When the EIT windows P1 and P2 overlap in Fig. 2b3, F1 is suppressed obviously due to the dressing effect of $E_3$ denoting by the dressing term $|G_3|^2 / d_5$. In

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the fluorescence signals, two suppression dips lower than the background curve containing sharp peaks R1 and R2 can be observed. These suppression dips are induced by the dressing effects of $E_2$ ($E_2'$) and $E_3$ individually, described by the terms $|G_{22}|^2/d_2$ and $|G_{33}|^2/d_3$ in $\rho_{11DD}$. Such dressing effects can be modulated by $E_1$ according to $\rho_{11DD}$, therefore when $P_1$ is small (Fig. 2a) the dips are invisible. When $\Delta_3$ moved far from resonance, the dip at $\Delta_1 = -\Delta_3$ gradually becomes shallower, corresponding with the weakened EIT. On the other hand, R2 peak get slightly higher with $\Delta_3$ increasing (from Fig. 2b2 to Fig. 2b1), which is entirely different from the case in Fig. 2a where R1 peak weakens with $\Delta_2$ increasing. This is for the reason that as the power of $E_1$ increases, its dressing effect on the two-photon term $d_3$ in $\rho_{33}$ should be considered, expressed as $d_3 + |G_{33}|^2/d_3$. Hence the fluorescence peaks R1 and R2 are suppressed around the resonant point. More importantly, the fluorescence peak R2 within the dip can be seen with ultra-narrow linewidth (about 10 MHz), which is much narrower than the EIT windows (about 50 MHz). Such high-resolution fluorescence is generated because it has been sheared twice by the EIT window P2. First, due to the two-photon dressing term $|G_{33}|^2/d_3$, the single-photon term $d_1$ of $\rho_{11DD}$ is clipped out, resulting in the suppression dip on R0, which is of the same width as the EIT window P2. Further, such clipped single-photon term, as a factor of two-photon term $d_2$, participates in the process of two-photon fluorescence R2 ($\rho_{33}$) which also stays in the EIT window P2. Therefore the fluorescence is sheared for the second time to an ultra-narrow peak. Similarly, in the $|0\rangle - |1\rangle - |2\rangle$ subsystem fluorescence R1 can also be sheared twice by the EIT window P1.

In the following, we observe the dressing effect and the interplay between two ladder subsystems by scanning coupling detuning $\Delta_2$. When $E_1$, $E_2$, and $E_3$ are turned on, we first study the singly dressed signals in $|0\rangle - |1\rangle - |2\rangle$ subsystem as depicted in Fig. 3a1 – Fig. 3a3 with different $\Delta_1$ values. In the probe transmission signals, the heights of baselines (horizontal background) represent Doppler absorption background at corresponding $\Delta_1$. EIT peaks higher than baselines appear at $\Delta_1 + \Delta_2 = 0$ and electro-
magnetically induced absorption (EIA) dips lower than baselines satisfying \( \Delta_1 + \Delta_2 = |G_2|^2/\Delta_1 \) can be observed at large detuning (Fig. 3a1 and Fig. 3a3). The FWM signal F1 with double-peak structure can be observed in Fig. 3a2 due to the synthesis of the self-dressing effect and the two-photon emission feature. Such double-peak structure becomes unobvious in Fig. 3a1 and Fig. 3a3, since the dressing effect weakens at the large frequency detunings. For the detected fluorescence signal, the baselines with suppression dips represent fluorescence R0, and the peaks within the dips are fluorescence R1. With \( \Delta_1 \) set far from resonance, the dip gradually becomes shallower, and eventually almost invisible at large detuning (Fig. 3a1 and Fig. 3a3), as corresponds to the weakening process of EIT. On the contrary, the peak gets stronger with \( \Delta_1 \) increasing, for R1 is suppressed around the resonant point according to the dressed term \( d_2 + |G_2|^2/d_1 \) in \( \rho^{(4)}_{22SD} \). Moreover, the suppression dips just fall into the EIT windows satisfying \( \Delta_1 + \Delta_2 = 0 \), and fluorescence peaks at large detuning are in alignment with EIA satisfying \( \Delta_1 + \Delta_2 = |G_2|^2/\Delta_1 \). In order to demonstrate the phenomena more clearly, we present the corresponding calculations of fluorescence signals below the experimental curves. Especially, the calculated R0 (the dash lines) and R1 (the dash-dot lines) are displayed separately. Such theoretical calculations confirm our experimental analysis stated above.

When \( E_3 \) is also turned on, the \( |0\rangle - |1\rangle - |2\rangle \) and \( |0\rangle - |1\rangle - |3\rangle \) subsystems will interplay with each other, resulting in some interesting phenomena as shown in Fig. 3b1–Fig. 3b3. In the probe transmission, the profile of baselines reveals the EIT induced by \( E_3 \) at \( \Delta_3 = -\Delta_1 = 0 \) and the peaks over each baseline are EIT induced by \( E_2(E_2') \). It is obvious that the EIT induced by \( E_2(E_2') \) is smaller at \( \Delta_3 = 0 \) (Fig. 3b2) than \( \Delta_3 \) is detuned (Fig. 3b1 and Fig. 3b3). This is the result of the strong cascade-dressing interaction between \( E_2(E_2') \) and \( E_3 \) near \( \Delta_1 = 0 \) according to the doubly dressed element

\[
\rho^{(1)}_{10DD} = \frac{iG_1}{d_1 + |G_2|^2/d_2 + |G_2'|^2/d_3}.
\]

The FWM signal F1 shows doublepeak structure induced by \( E_2(E_2') \), and is additionally suppressed by external dressing field \( E_3 \) when \( \Delta_3 = -\Delta_1 \) (Fig. 3b2). The fluorescence R0 is also suppressed by \( E_3 \) as depicted by the lower fluorescence baseline in Fig. 3b2, in addition to suppression effect of \( E_2 (E_2') \). Corresponding to the EIT window, the suppression dip induced by \( E_2(E_2') \) is shallower at \( \Delta_3 = 0 \) (Fig. 3b2) than \( \Delta_3 \) is detuned (Fig. 3b1 and Fig. 3b3). On the other hand, the fluorescence peak R1 in Fig. 3b2 is slightly stronger than the ones in Fig. 3b1 and Fig. 3b3 resulting from the enhancement effect of \( E_3 \) around \( \Delta_1 = -\Delta_1 \) by considering the nest-dressing term \( |G_2|^2/(d_1 + |G_3|^2/d_3) \) in \( \rho^{(4)}_{22DD} \). The corresponding calculations of fluorescence are also present in Fig. 3b3 which are in agreement with the experimental results.

Finally, we concentrate on the signal intensity depending on the power of laser beams by scanning \( \Delta_2 \) with \( \Delta_1 = \Delta_3 = 0 \). First, when the power of the beam \( E_2 \) changes from small to large, we arrange the experimental curves from bottom to top in Fig. 4a1–Fig. 4a3. In this case, beam \( E_2' \) is blocked and other beams are turned on, so that the signals in Fig. 4a2 represent the suppression degree induced by \( E_2 \) on the FWM signal F2. In accordance with expectation, both the height of EIT peak (Fig. 4a1) and the suppression degree of F2 (Fig. 4a2) get larger with \( P_2 \) increasing, for the reason that the function of the two-photon dressing term \( |G_2|^2/d_2 \) in \( \rho^{(1)}_{10DD} \) and \( \rho^{(3)}_{22DD} \) becomes stronger as \( P_2 \) increases. For the fluorescence signal (Fig. 4a3), on the one hand, the suppression dip deepens as \( P_2 \) increases, also due to the increasing dressing effect of \( E_3 \) on \( \rho^{(2)}_{11} \). On the other hand, the fluorescence peak R1 which is mainly dependent on the beam \( E_2 \) intensity according to \( \rho^{(4)}_{22} \), get greatly larger with \( P_2 \) increasing.

Next, when the power of the other coupling beam \( E_3 \) is changed with \( E_2' \) blocked, the results are strikingly dif-
different (Fig. 4b1 – Fig. 4b3). As $P_3$ increases, we find the EIT peak induced by $E_2$ ($E'_2$) decreases as shown from bottom to top in Fig. 4b1, which is contrary to the case of changing the power of self-dressing field $P_2$, where the EIT peak increases from bottom to top in Fig. 4a1. This is for the reason that the interaction between the dressing effect of $E_2$ ($E'_2$) and $E_3$ on $\rho_1^{(1)}$, which has been explained in Fig. 3b, becomes stronger with $P_3$ increasing. The FWM signal $F_1$ in Fig. 4b2 also weaken as $P_3$ increased, due to the stronger suppression effect of $E_3$ denoting by the dressing term $|G_3|^2/d_3$ in $\rho_1^{(3)}$. When we turn to the measured fluorescence signals (Fig. 4b3), we find the suppression dip related with $\rho_1^{(2)}$ become shallower as $P_3$ increases, also due to the stronger interaction between the dressing term $|G_3|^2/d_3$ and $|G_2|^2/d_2$ in $\rho_1^{(2)DD}$. In contrast, the peaks within the dips become slightly larger from bottom to top in Fig. 4b3, since the enhancement effect of $E_3$ on $R_1$ get stronger according to the nest-dressing term

\[
\frac{|G_3|^2}{d_1} + \frac{|G_2|^2}{d_3} \quad \text{in } \rho_1^{(4)DD}.
\]

4. Conclusion

We compare the probe transmission, FWM and fluorescence signals with dressing effects for the first time. Also, the interplay between two ladder subsystems is investigated in the $Y$-type system. The ultra-narrow two-photon fluorescence signal is obtained in ladder or $Y$-type atomic system, which is sheared twice by EIT window. Such fluorescence with very high coherence and monochromaticity can be potentially applied in metrology, long-distance quantum communication and quantum correlation. Moreover, the amplitude of the signals can be effectively controlled by the incident beam intensity.

Acknowledgements This work was supported by the 973 Program (2012CB921804), NSFC (10974151, 61078002, 61078020, 11104214, 61108017, and 11104216).

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