Switching suppression and enhancement of fluorescence and six-wave mixing by phase modulation

Zhiguo Wang, Peng Ying, Peiying Li, Dan Zhang, Heqing Huang, Hao Tian & Yanpeng Zhang

Key Laboratory for Physical Electronics and Devices of the Ministry of Education & Shaanxi Key Lab of Information Photonic Technique, Xi’an Jiaotong University, Xi’an 710049, China.

The conversion between enhancement and suppression in six-wave mixing (SWM) and fluorescence signals by phase modulation has demonstrated for the first time. It is observed in our experiment the suppression of SWM and fluorescence is transformed into enhancement in company with the switch from electromagnetically induced absorption (EIA) in the transmitted probe with the relative phase changed from 0 to π/2. Our research could be potentially applied in optical communication and quantum information processing.

Lots of studies focus on the electromagnetically induced transparency (EIT) and the higher-order nonlinear optical process comprising four-wave mixing (FWM) and six-wave mixing (SWM) under EIT condition. In the meantime, the fluorescence owing to spontaneous emission is also observed. Moreover, the enhancement and suppression of FWM (SWM) and fluorescence corresponding to electromagnetically induced absorption (EIA) and EIT has aroused much interest. The switch between bright state (EIA in the transmitted probe signal and enhancement in FWM and fluorescence signals) and dark state (EIT in the transmitted probe signal and suppression in FWM and fluorescence signals) can be realized by controlling phase difference between the two circularly polarized components of a single coherent field. In addition, the switch can also be obtained by manipulating the dressing field power and the probe detuning.

In this paper, we experimentally demonstrate the phase-modulated switch between enhancement and suppression of SWM and fluorescence signals for the first time. First, the phase modulated switch between bright and dark states is realized under self-dressing condition. Second, we study the phase modulated switch under multisdressing condition including the external-dressing. At last, we observe the dependence of the measured signals on relative phase.

Results

Our experiment is carried out in a 85Rb vapor cell. The energy levels 5S1/2(F = 3), 5S1/2(F = 2), 5P3/2, 5D3/2, and 5D5/2 constitute a five-level atomic system (Fig. 1(a)). The transition 5S1/2(F = 3) (|0⟩) - 5P3/2 (|1⟩) is probed by the laser beam E1 (frequency ω1, wave vector k1 and Rabi frequency G1). The transition 5P3/2 - 5S1/2(F = 2) (|3⟩) is driven by two coupling beams E2 (ω2, k3 and G3) and E5 (ω5, k5 and G5). Two dressing beams E2 (ω2, k2 and G2) and E4 (ω4, k4 and G4) respectively drive the upper transitions 5P3/2 - 5D5/2 (|2⟩) and 5P3/2 - 5D3/2 (|4⟩). In normal experiment, the five beams are placed in a square-box pattern (Fig. 1(b)). The beams E2, E4, E5 and E3 propagate through the cell in the same direction with tiny angles about 0.3° between any two. In the opposite direction of E2 there is the probe beam E1. However, in our experiment, the normal experimental configuration should be modified since we study the phase-controlled switch. The coupling beam E2 (E4) is deviated with an angle ± (β) from the normal position (Fig. 1(c) and 1(d)). In the experimental system, we use E3 and E5 to represent the SWM signals generated by E1, E2, E3 and E5 and by E1, E3, E5 and E3 respectively. Besides, the single-photon fluorescence caused by the photon decay from the level |1⟩ is called R0. The sign R1 (R2) denotes the fluorescence due to the photon decay from the level |2⟩ (|4⟩).

Generally, we can obtain the density matrix elements ρ(0) i, j (related with EIT), ρ(4) i, j (related with SWM signal), and ρ(1) i, j , ρ(2) i, j and ρ(3) i, j (related with fluorescence R0, R1 and R2) by solving the density-matrix equations. With Liouville pathway ρ(j) 00 → ρ(j) 11 and E4 blocked, we can obtain...
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\[ |G_1|^2 \sigma \Delta \Phi / d_2 \]

is the self-dressing term and external dressing (the external-dressing effect refer to that the field will dresses the involved energy levels to modify the signals, while it is not the participated field for the generation of these signals, for example, the term \(|G_2|^2 \sigma \Delta \Phi / d_2\) is the external dressing term) condition, respectively. For the fluorescence signals, via the Liouville pathway \(\rho_{10}^{(0)} \to \rho_{10}^{(1)} \to \rho_{11}^{(2)}\), one can obtain

\[ \rho_{11}^{(2)} = -|G_2|^2/|\Gamma_{11}|(d_1 + |G_2|^2 \sigma \Delta \Phi / d_2) \]  

or

\[ \rho_{11}^{(2)} = -|G_2|^2/|\Gamma_{11}|(d_1 + |G_2|^2 \sigma \Delta \Phi / d_2 + |G_4|^2 \sigma \Delta \Phi / d_3) \]

(7)

(8)

to describe the profile of fluorescence \(R_0\) under single-dressing or double-dressing case, respectively. With the pathway \(\rho_{33}^{(0)} \to \rho_{13}^{(1)} \to \rho_{11}^{(2)}\), one can acquire

\[ \rho_{11}^{(4)} = -|G_2|^2/|\Gamma_{11}|(d_1 + |G_2|^2 \sigma \Delta \Phi / d_2 + |G_4|^2 \sigma \Delta \Phi / d_3) \]

(9)

to describe the DC background of \(R_0\). Consequently, the fluorescence \(R_0\) is proportional to \(\rho_{10}^{(1)} + \rho_{12}^{(2)}\). For two-photon fluorescence \(R_1\), via \(\rho_{20}^{(0)} \to \rho_{10}^{(1)} \to \rho_{21}^{(2)} \to \rho_{21}^{(4)} \to \rho_{22}^{(4)}\), we have

\[ \rho_{22}^{(4)} = |G_2|^2 |G_2|^2/|\Gamma_{22}|d_1d_1(d_2 + |G_2|^2 \sigma \Delta \Phi / d_1) \]

(10)

with \(d_0 = \Gamma_{21} + i\Delta_2\). Similarly, the other two-photon fluorescence \(R_2\) is described as

\[ \rho_{44}^{(4)} = |G_1|^2 |G_4|^2/|\Gamma_{44}|d_1d_1(d_2 + |G_2|^2 \sigma \Delta \Phi / d_1) \]

(11)

with \(d_0 = \Gamma_{41} + i\Delta_4\).

First, we consider the phase-modulated switch of self-dressed signals as shown in Fig. 2. In the experiment, we turn on \(E_1, E_2, E_3, E_4\) and block \(E_3\) and the signals are obtained by scanning \(\Delta_1\) at different discrete designated \(\Delta_1\) with \(\Delta \Phi_1 = 0\) (Fig. 2(a), viewed as the reference point at the normal configuration), \(\Delta \Phi_1 = -\pi/5\) (Fig. 2(b)) and \(\Delta \Phi_1 = \pi/5\) (Fig. 2(c)). In Fig. 2(a1), the EIT caused by \(E_2\), meeting \(\Delta_1 + \Delta_2 = 0\), emerges in the larger range of the probe detuning. The EIA, satisfying \(\Delta_1 + \Delta_2 = |G_2|^2/\Delta_3\), only appears at the large probe detuning, such as \(\Delta_1 = \pm 400\) MHz. In Fig. 2(b1), we can find the probe transmission signals present as EIT at negative detunings \(\Delta_1\) while change from strong EIT to partial-EIT-partial-EIT and lastly to weak EIA with \(\Delta_1\) increasing at positive detunings \(\Delta_1\). In Fig. 2(c1), with the probe detuning transformed from negative to positive, the signals turn from EIA to partial-EIT-partial-EIT and lastly to EIT. Obviously, the variations of the probe transmission signals are quite the contrary in above three figures. This is caused by the modulation of the relative phase \(\Delta \Phi_1\) in the dressing term \(|G_2|^2 \sigma \Delta \Phi / d_2\) in \(\rho_{10}^{(1)}\). Thus, at a certain detuning \(\Delta_1\), we can switch EIT and EIA by adjusting the relative phase \(\Delta \Phi_1\). Similarly, the dressing effect on the SWM signal caused by \(|G_2|^2 \sigma \Delta \Phi / d_2\) is also regulated by \(\Delta \Phi_1\). According to \(\rho_{31}^{(0)}\), the intensity of SWM has inverse correlation with \(\cos(\Delta \Phi_1 - \theta)\) where \(\theta = \arctan(\Delta_1/\Gamma_{10})\).

In the SWM signals, with the relative phase \(\Delta \Phi_1\) changed from 0 (Fig. 2(a2)) to \(-\pi/5\) (Fig. 2(b2)), the intensity of SWM is obviously enhanced at \(\Delta_1 \approx 0\) since \(\cos(-\pi/5 - \theta) < \cos(0 - \theta)\) in the region while is suppressed at \(\Delta_1 < 0\) for \(\cos(-\pi/5 - \theta) > \cos(0 - \theta)\) here. One can also see the difference between the SWM signals with \(\Delta \Phi_1 = 0\) (Fig. 2(a2)) and those with \(\Delta \Phi_1 = \pi/5\) (Fig. 2(c2)). Compared with the SWM in Fig. 2(a2), the SWM signal in Fig. 2(c2) is suppressed in
Figure 2 | In each sub-curve, measured probe transmission signals (a1), (b1) and (c1)), SWM signals ((a2), (b2) and (c2)) and fluorescence signals (a3), (b3) and (c3)) versus $\Delta_2$. $\Delta_2$ is scanned around $-\Delta_1$ from $-90$ MHz to $90$ MHz with $E_2$ blocked. Each sub-curve corresponds to different fixed $\Delta_1$. For (a1)–(a3) $\Delta p_1 = 0$, for (b1)–(b3) $\Delta p_1 = -\pi/5$ and for (c1)–(c3) $\Delta p_1 = 3\pi/5$. The other parameters are $\Delta_2 = 0$, $G_1 = 10.85$ MHz, $G_3 = 19.46$ MHz, $G_5 = 16.66$ MHz and $G_7 = 14.4$ MHz. (d) Theoretical calculations for probe, SWM and fluorescence signals versus $\Delta p_1$ by scanning $\Delta_2$ at three typical detunings $\Delta_1$.

Figure 3 | In each sub-curve, measured probe transmission signals (a), fluorescence signals (b), and SWM signals (c)–(d) versus $\Delta_2$ ($\Delta_2$ is scanned around $-\Delta_1$ from $-90$ MHz to $90$ MHz) with the fixed $\Delta_1$. Each sub-curve corresponds to different discrete fixed $\Delta_1$, (a1), (b1) and (c) Signals obtained with $\Delta p_2 = 0$, (a2), (b2) and (d) Signals obtained with $\Delta p_2 = \pi/2$. (a), (b), (c) and (d) Signals obtained with all beams turned on. (c2) and (d2) Signals obtained with $E_2$ blocked. (c3) Signals obtained by subtracting the signals in (c2) from the signals in (c1). (d3) Signals obtained by subtracting the signals in (d2) from the signals in (d1). Other parameters are $\Delta_1 = 100$ MHz, $\Delta_3 = 0$, $\Delta p_1 = 0$, $G_1 = 10.72$ MHz, $G_5 = 21.16$ MHz, $G_3 = 18.87$ MHz, $G_7 = 14.4$ MHz and $G_4 = 14.16$ MHz.

caused by the dressing field $E_2$ and the peak in each dip represents the two-photon fluorescence $R_2$. The profile dip consisting of baselines reveals $R_0$ suppressed by $E_2$ owing to the dressing term $|G_2|^2/d_2$ in $\rho_2^{(2)}$, and the profile peak represents the two-photon fluorescence $R_1$. With $\Delta p_2$ changed from 0 (Fig. 3(b1)) to $\pi/2$ (Fig. 3(b2)), the height of the peak in each sub-curve gets much high because the dressing term $|G_2|^2e^{i\Delta p_2}/d_2$ in $\rho_2^{(2)}$ has generated the enhancement effect on $R_0$ at $\Delta p_2 = \pi/2$. In this process, the dip in each sub-curve becomes shallow due to the weakened dressing effect of $E_2$ on $R_0$ caused by the modulation of $\Delta p_2$.

In Fig. 3(c1), the global profile (dashed curve) consisting of all baselines reveals the SWM signal $E_{S1}$. In the $E_{S2}$ signal, one can find the AT splitting which is because the self-dressing term $|G_1|^2/d_2$ acts on the two-photon term $d_2$ in $\rho_2^{(2)}$. The sub-curve at any $\Delta_1$ means the compound signal which includes two components: $I_{D1}$ and $I_{D2}$. $I_{D1}$ denotes the enhancement or suppression intensity of $E_{S1}$ (arising from the external-dressing field $E_2$ according to $\rho_2^{(2)}$) and $I_{D2}$ signifies the SWM $E_{S2}$ intensity with the suppression of $E_2$. Figure 3(c2) shows the SWM $E_{S2}$ intensity without the suppression of $E_2$ which is denoted by $I_{S2}$. The signals in Fig. 3(c3) are obtained by subtracting the signals ($I_{S2}$) in Fig. 3(c2) from the signals ($I_{D1} + I_{D2}$) in Fig. 3(c1). Therefore, the signals in Fig. 3(c3) represent two dressing results at different $\Delta_1$: one is $I_{D1}$, the other is $I_{D2} - I_{S2}$ which is the suppressed intensity caused by $E_2$ with regard to the SWM $E_{S2}$. When we alter the relative phase $\Delta p_2$, the experiment results similar to Fig. 3(c) are obtained, as shown in Fig. 3(d1)–3(d3). The modulation of signals caused by relative phase can be observed clearly, as shown in Fig. 3(c3) and Fig. 3(d3). For instance, with the relative phase switched from 0 to $\pi/2$, the original suppression signals (at $\Delta_1 + \Delta_2 = -45, -30$ and $-15$ MHz) transform into partial-suppression-partial-enhancement signals and the intensities of original partial-suppression-partial-enhancement signals (at $\Delta_1 + \Delta_2 = 0, 15, 30, 45$ and $60$ MHz) are strengthened significantly in correspondence with

Further, we turn on all five laser beams including $E_2$ and observe the phase controlled switch of the multi-dressed (the multi-dressing effect is the case where two or more fields including self- and external-dressing fields dress the energy levels to modify the signals) signals by scanning $\Delta_2$ at different discrete fixed $\Delta_1$. The global profile generated from all baselines means the EIT induced by $E_2$. The global profile from all baselines means the EIT induced by $E_2$, meeting the condition $\Delta_1 + \Delta_2 = 0$. When we change the related phase $\Delta p_2$ from 0 (Fig. 3(a1)) to $\pi/2$ (Fig. 3(a2)), the original EITs are mostly switched to EIA. In Fig. 3(b1), the dip in each sub-curve shows the suppression on $R_0$.
the EIT-EIA switch in Fig. 3(a). It is worth mentioning that the
dressing result of $E_2$ is basically invariable since the relative phase
$\Delta \Phi_2$ is only related to the dressing effect of $E_2$.

Discussion
In order to clearly compare the variations of signals with the relative
phase, we discuss the relative phase dependence of the measured
signals by scanning $\Delta \alpha$ at the fixed detuning $\Delta \nu$ under the above
mentioned two cases of self-dressing and multi-dressing effects.
First, with $E_2$ blocked (Fig. 4(a), the self-dressing case), EIT in
Fig. 4(a1) can be switched to EIA along with the change of $\Delta \Phi_2$ from
bottom to top. During this process, the strongest EIT and EIA sepa-
rately appear at $\Delta \Phi_2 = -\pi/6$ and $\Delta \Phi_2 = 3\pi/4$. Depending on
whether $\Delta \Phi_2$ is greater than or less than $\pi/4$, the transmitted probe
signal behaves mainly EIA or EIT. In the phase matching range
where $\Delta \Phi_2$ is altered from $-\pi/6$ to $\pi/2$, compared with the signals
at the reference phase $\Delta \Phi_2 = 0$, the SWM signal $E_{S2}$ (Fig. 4(a2))
and fluorescence signal $R_2$ (peaks in Fig. 4(a3)) get large when $\Delta \Phi_2 > 0$
and become small while $\Delta \Phi_2 < 0$ due to the switch of the dressing
effect of $E_2$ induced by $\Delta \Phi_2$. The largest intensities of the SWM and
fluorescence signals appear at partial-EIT-partial-EIA ($\Delta \Phi_2 = \pi/4$)
and EIA ($\Delta \Phi_2 = \pi/2$), respectively. Along with continuing to
increase $\Delta \Phi_2$ from $\pi/2$ to $\pi$ or to decrease $\Delta \Phi_2$ from $-\pi/6$ to $-\pi/3$,
which correspond to increasing the deflection angle $\beta$ under the
abnormal configuration (Fig. 1(d)), the SWM and fluorescence signal
intensities decrease. This results from the classical effect with the

Figure 4 | The transmitted probe ((a1), (b1)), SWM ((a2), (b2)) and
fluorescence ((a3), (b3)) versus $\Delta \alpha$ at different relative phase $\Delta \Phi_2 = -\pi/3$, $-\pi/6$, $0$, $\pi/4$, $\pi/2$, $3\pi/4$, and $\pi$ from the bottom to top.(a1),(a2) and (a3)
Signals obtained with $E_2$ blocked. (b1),(b2) and (b3) Signals obtained with
all beams turned on. (a4) and (b4) the relative phase dependences of the
fluorescence peak (dash line with circles) and the fluorescence dip (solid
line with squares) in (a3) and (b3), respectively. The other parameters are
$\Delta \nu_1 = 0$, $\Delta \nu_2 = -120$ MHz, $\Delta \nu_3 = 150$ MHz, $\Delta \nu_4 = 0$, $G_1 = 11.84$ MHz, $G_2$ = 18.34 MHz, $G_3$ = 17 MHz, $G_4$ = 26.11 MHz and $G_4$ = 18 MHz.

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Author contributions
Z.G.W. and Y.P.Z. provided the idea and main contributions to the theoretical and experimental analysis of this work. P.Y., P.Y.L., D.Z., H.Q.H. and H.T. contributed to the presentation and execution of the work. All authors discussed the results and contributed to writing the manuscript.

Additional information
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