Observation of two multiwave mixing processes via dual optical pumping channels

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Electromagnetically induced transparency (EIT) has been studied well in the last two decades[1]. Because the generated signals can be transmitted through the resonant atomic medium with little absorption under the EIT condition, multiwave mixing (MWM)[2–4] processes can be investigated. Using one EIT window, the generated four-wave mixing (FWM) and six-wave mixing (SWM) signals are able to coexist in an open-cycle atomic system. Moreover, the competition and interference[5,6] between these FWM and SWM processes have been studied via atomic coherence in a four-level atomic system.

In Fig. 1(a1), the weak probe field $E_1$ (with tunable frequency $\omega_1$, wave vector $k_1$, and Rabi frequency $G_1$) drives the transition from $|0\rangle$ to $|1\rangle$ with resonant wavelength of 780.245 nm. The frequency detuning is $\Delta_1$. The coupling field $E_2$ ($\omega_2$, $k_2$, $\Delta_2$, and $G_2$) drives the transition between $|1\rangle$–$|2\rangle$ with the resonant wavelength of 775.978 nm; two pump beams, $E_3$ ($\omega_3$, $k_3$, $\Delta_3$, and $G_3$) and $E_4$ ($\omega_4$, $k_4$, $\Delta_4$, and $G_4$), from the same laser drive the transition between $|3\rangle$–$|4\rangle$ with the resonant wavelength of 780.234 nm, and another pump beam $E_5$ ($\omega_5$, $k_5$, $\Delta_5$, and $G_5$) drives the transition between $|1\rangle$–$|4\rangle$ with the resonant wavelength of 780.235 nm. Using four external cavity diode lasers (ECDL) with line width less than or equal to 1 MHz, these relevant transitions can be effectively driven in $^{85}$Rb atoms system. The powers of beams, $E_1$, $E_2$, $E_3$, $E_5$, and $E_4$, are 3.7, 50, 20, 20, and 48 mW, respectively. Laser beams $E_2$, $E_3$, and $E_4$, propagating in the same direction with small angles of $\sim 0.3^\circ$ and overlap at one spot inside a rubidium atom vapor cell of 7.5-cm length. A typical atomic density in the cell is $2 \times 10^{14}$ cm$^{-3}$. The probe beam $E_1$ and pump beam $E_2$ propagate in the same direction (Fig. 1(b)).

According to the phase-matching conditions, one FWM and two SWM processes can be generated simultaneously in the same direction. Firstly, by blocking the coupling beams $E_2$ and $E_4$, the FWM process with the phase-matching condition $k_f = k_1 + k_3 - k'_3$ can be regarded as a scattering of the probe field $E_1$ over the small-angle static grating formed by the conjugate field pair $E_3$ and $E_5$. This FWM process can be further analyzed by Liouville pathway. In Fig. 1(a1), $|0\rangle$ and $|1\rangle$ are hyperfine structures of the ground state; therefore the Liouville pathway of the FWM process is $\rho_{00}^{(0)} \xrightarrow{\omega_3} \rho_{00}^{(1)} \xrightarrow{\omega_2} \rho_{10}^{(2)} \xrightarrow{\omega_1} \rho_{10}^{(3)}$, which results in the density matrix element $\rho_{10}^{(3)} = -G_A \exp(ikr)/(d_1 d_3 F_{00})$, where $G_A = iG_1 G_3 G_5^* d_1 = i\Delta_1 + \Gamma_{10}$, $d_3 = i\Delta_3 + \Gamma_{13}$, and $\Gamma_{ij}$ are the transverse relaxation rates between states $|i\rangle$ and $|j\rangle$. The atomic coherence can affect the MWM process significantly. The fields related to the generation of FWM signal can affect the FWM signal due to its dressing effect; these are called self-dressing fields[7]. Correspondingly, the effect brought by the coupling beams $E_2$

![Fig. 1. (a) Five-level atomic system of the $^{85}$Rb atom; (b) spatial beam geometry used in the experiment; (c) hyperfine structures of the $^{85}$Rb atom corresponding to Fig. 1(a1).](image-url)
and $E_4$, as external fields on this FWM process, is denoted as external dressing effect [8]. Thus, the Liouville pathway of such external dressed FWM process can be modified as $\rho_{00} \overset{\omega_1}{\rightarrow} \rho_{30} \overset{\omega_2}{\rightarrow} \rho_{52}G_1G_2 + \rho_{52}$. The above expression becomes $\rho_{10}^{(3)} = -G_A \exp(\mathbf{i} \mathbf{kr})/[d_1 + |G_1|^2/d_2 + |G_4|^2/d_4] d_1 \Gamma_{00}$, where $d_2 = i(\Delta_1 - \Delta_2) + \Gamma_20$ and $d_3 = i(\Delta_1 - \Delta_3) + \Gamma_30$. When all incident beams are active except for the pump beam $E_4$, one can observe the SWMI process satisfying the phase-matching condition $k_{\mathbf{s}} = k_1 + k_2 - k_2'$ with one photon each from the beams $E_1, E_3, E_1'$ and two photons from the beam $E_2$. This generated SWMI signal falls into an EIT window in a ladder-type three-level subsystem (|0⟩ - |1⟩ - |2⟩) (Fig. 2). Similar to the case of FWM, the Liouville pathway of SWMI process can be obtained as $\rho_{00} \overset{\omega_1}{\rightarrow} \rho_{30} \overset{\omega_2}{\rightarrow} \rho_{52}G_1G_2 + \rho_{52}$, $\rho_{20}^{(4)} \rightarrow \rho_{10}^{(5)}$. The density matrix element related to such SWMI process is $\rho_{10}^{(5)} = G_B \exp(\mathbf{i} \mathbf{kr})/[d_1 + |G_1|^2/d_2 + |G_4|^2/d_4] d_1 \Gamma_{00}$, with $G_B = iG_4G_3G_2^*G_2^*$. When the self-dressing effect of $E_2$ and external dressing effect of $E_4$ are considered, the modified Liouville pathway of the SWMI becomes $\rho_{00} \overset{\omega_1}{\rightarrow} \rho_{30} \overset{\omega_2}{\rightarrow} \rho_{52}G_1G_2 + \rho_{52}$, and the density matrix element becomes $\rho_{10}^{(5)} = G_B \exp(\mathbf{i} \mathbf{kr})/[d_1 + |G_1|^2/d_2 + |G_4|^2/d_4] d_1 \Gamma_{00}$, with $d_5 = i(\Delta_1 - \Delta_5) + \Gamma_{50}$. Finally, with all incident active except for the coupling beam $E_2$, the second pathway of SWMI process (SWMI2) with phase-matching condition $k_2 = k_1 + k_3'$ and $k_4' = k_4$ is generated by $E_1, E_3, E_1'$ and $E_4$. Similarly, the Liouville pathway $\rho_{00} \overset{\omega_1}{\rightarrow} \rho_{30} \overset{\omega_2}{\rightarrow} \rho_{52}G_1G_2 + \rho_{52}$, and the expression $\rho_{10}^{(5)} = G_C \exp(\mathbf{i} \mathbf{kr})/[d_4 + |G_2|^2/d_2 + |G_4|^2/d_4] d_4 \Gamma_{00}$ can be obtained without considering the dressing effect, where $G_C = iG_1G_2G_3G_4 + i(\Delta_1 - \Delta_4 - \Delta_0) + \Gamma_{40}$. The modified Liouville pathway and density matrix element considering the self-dressing effect of $E_2$ and external dressing effect of $E_2$, respectively, are $\rho_{00} \overset{\omega_1}{\rightarrow} \rho_{30} \overset{\omega_2}{\rightarrow} \rho_{52}G_1G_2 + \rho_{52}$, and $\rho_{10}^{(5)} = G_C \exp(\mathbf{i} \mathbf{kr})/[d_4 + |G_2|^2/d_2 + |G_4|^2/d_4] d_4 \Gamma_{00}$, respectively, where $d_4 = i(\Delta_1 - \Delta_4 - \Delta_0) + \Gamma_{40}$. The MWM signals $E_F$, $E_{51}$, and $E_{52}$ can be detected along the same direction by an avalanche photodiode detector (APD).

In the experiment, the scan range of $\Delta_1$ is 5 GHz; thus, the information of both $F = 3$ (Fig. 1(a1)) and $F = 2$ (Fig. 1(a2)) can be obtained. In Fig. 1(a2), by changing the current or temperature of the corresponding ECDL, the wavelength of the $E_1$ tunes to 780 - 244 nm and the wavelength of the $E_2$ beam to 780 - 244 nm to achieve different optical pumping process. Similar to the system shown in Fig. 1(a1), MWM processes ($E_1, E_3, E_4$) in Fig. 1(a2) also satisfy the phase-matching conditions $k_1 = k_1 + k_1 - k_1', k_1' = k_1 + k_1 - k_1' + k_4 - k_4$, and $k_2' = k_1 + k_3 - k_1' + k_4 - k_4$, respectively. Based on the method in the first atomic system, the following detailed theoretical analyses are obtained as

(1) Undressed FWM process:

$$\rho_{00} \overset{\omega_1}{\rightarrow} \rho_{30} \overset{\omega_2}{\rightarrow} \rho_{52}G_1G_2 + \rho_{52}$$

$$\rho_{30} \rightarrow \rho_{10}^{(3)} = -G_A \exp(\mathbf{i} \mathbf{kr})/(d_4d_5\Gamma_{00})$$

where $d_4 = \Gamma_{30} + i\Delta_1$, $d_5 = \Gamma_{10} + i\Delta_3$. Dressed FWM process:

$$\rho_{00} \overset{\omega_1}{\rightarrow} \rho_{30} \overset{\omega_2}{\rightarrow} \rho_{52}G_1G_2 + \rho_{52}$$

$$\rho_{30} \rightarrow \rho_{10}^{(3)} = -G_A \exp(\mathbf{i} \mathbf{kr})/[d_4 + |G_2|^2/d_2 + |G_4|^2/d_4] d_4 \Gamma_{00}$$

(2) Undressed SWMI process:

$$\rho_{00} \overset{\omega_1}{\rightarrow} \rho_{30} \overset{\omega_2}{\rightarrow} \rho_{52}G_1G_2 + \rho_{52}$$

$$\rho_{30} \rightarrow \rho_{10}^{(3)} = G_B \exp(\mathbf{i} \mathbf{kr})/[d_4 + |G_2|^2/d_2 + |G_4|^2/d_4] d_4 \Gamma_{00}$$

Dressed SWMI process:

$$\rho_{00} \overset{\omega_1}{\rightarrow} \rho_{30} \overset{\omega_2}{\rightarrow} \rho_{52}G_1G_2 + \rho_{52}$$

$$\rho_{30} \rightarrow \rho_{10}^{(3)} = G_B \exp(\mathbf{i} \mathbf{kr})/[d_4 + |G_2|^2/d_2 + |G_4|^2/d_4] d_4 \Gamma_{00}$$

(3) SWMI2 process:

With the main one in six alternative pathways, the undressed-SWMI2 process can be addressed as

$$\rho_{14} \overset{\omega_1}{\rightarrow} \rho_{14} \overset{\omega_2}{\rightarrow} \rho_{14} \overset{\omega_3}{\rightarrow} \rho_{14} \overset{\omega_4}{\rightarrow} \rho_{30}$$

$$\rho_{30} = G_C \exp(\mathbf{i} \mathbf{kr})/[d_1d_2(d_1d_2)\Gamma_{44}]$$

where $d_{12} = i(\Delta_1 - \Delta_3) + \Gamma_{14}$, $d_{13} = i(\Delta_2 - \Delta_3) + \Gamma_{14}$, and $d_{14} = i(\Delta_4 + \Gamma_{14})$. The dressed SWMI2 process can be addressed as

$$\rho_{14} \overset{\omega_1}{\rightarrow} \rho_{14} \overset{\omega_2}{\rightarrow} \rho_{14} \overset{\omega_3}{\rightarrow} \rho_{14} \overset{\omega_4}{\rightarrow} \rho_{30}$$

$$\rho_{30} = G_C \exp(\mathbf{i} \mathbf{kr})/[d_1d_2(\Gamma_{44})^2\Gamma_{44}]$$

where $d_{15} = i(\Delta_1 + \Delta_2) + \Gamma_{24}$. From the expressions listed above, clearly, although the structures of the two energy levels are similar, the influences the coupled fields $k_3$ and $k_4$ produce on the different SWMI signals are quite different.

Figures 2(a) and (b) show the results of blocking different coupling beams in the first five-level atomic system shown in Fig. 1(a1). The upper curve of Fig. 2(a1) shows the FWM signal when the beams $E_1, E_2$, and $E_3$ are active; and the detuning of $E_3$ is $\Delta_3 = 500$ MHz. The FWM signal is not conspicuous because the EIT window is absent due to the Doppler effect in the lower curve of Fig. 2(a1). Hence, the optical pumping is very weak. In Fig. 2(a2), when $E_2$ is turned on and $E_4$ is blocked, SWMI signal shown by the upper curve is generated and falls into the |0⟩ - |1⟩ - |2⟩ ladder-type EIT window shown by the lower curve. Figure 2(a3) shows the experimental results when $E_4$ is activated and $E_2$ is blocked. Here, the SWMI ($k_{e2}$) signal
shown by the upper curve is generated and falls into the \(|0⟩−|1⟩−|4⟩\) \(Λ\)-type EIT window, which is shown by the lower curve. More importantly, two sharply enhanced absorption dips are near the SWM signal peak shown in the upper curve of Figs. 2(a3) or (b3), this can be well interpreted by introducing the hyperfine structure in the upper curve of Figs. 2(a3) or (b3), this can be well interpreted by introducing the hyperfine structure of \(^{85}\text{Rb}\) [5S\(_{1/2}\), \(F = 2\)] dip (the upper curve of Fig. 2(a3)) is due to the leakage of the probe transmission, which have contributions from \(E_3\), \(E_3'\), \(E_4\) optical pumping of population from the ground-state [5S\(_{1/2}\), \(F = 2\)], as shown in Fig. 2(a3). Hence, the distance between the two dips is 121 MHz, corresponding to that between \([5P_{3/2}, F = 3]\) and \([5P_{3/2}, F = 4]\). When all beams are active, SWM1 (\(k_3\)), SWM2 (\(k_2\)), and FWM (\(k_1\)) are generated simultaneously with corresponding EIT windows, as shown in Fig. 2(a4). Similarly, two sharply enhanced absorption dips are apparent in Fig. 2(a4).

The experimental results when the detuning is set at \(\Delta_3 = 700\) MHz are shown in Figs. 2(b1)–(b4). The meaning of the signal curves in Figs. 2(b1)–(b4) is the same as in Figs. 2(a1)–(a4). Similarly, FWM signal (\(k_1\)) in Fig. 2(b1) is not conspicuous because there no EIT window exists. Because of the optical pumping effect corresponding to the transitions from the hyperfine levels \(^{85}\text{Rb}\) [5S\(_{1/2}\), \(F = 2\)] to \([5P_{3/2}, F = 1, 3]\), the generated SWM1 signal (\(k_{11}\)) (\(\Delta_3 = 500\) MHz is slightly stronger than the SWM1 signal (\(k_{11}\)) (\(\Delta_3 = 700\) MHz). This phenomenon can be identified in the upper curves of Figs. 2(a2) and (b2). When SWM2 is generated, similarly, two sharply enhanced absorption dips are present because of the effect of optical pumping in Figs. 2(b3) and (b4).

Next, we move from the first five-level atomic system shown in Fig. 1(a1) to the second similar five-level atomic system by tuning the wavelength of beams \(E_1\) and \(E_2\), as shown in Fig. 1(a2). Figures 2(c) and (d) show the results of blocking different coupling beams in the second system, as well as the significant differences from those in Figs. 2(a) and (b). When the beams \(E_1\), \(E_3\), and \(E_3'\) are all active, the upper curve of Fig. 2(c1) shows that the FWM signal (\(k_1'\)) is clearly Doppler broadened due to the strong optical pumping (\(\Delta_3 = 61.8\) MHz). In Fig. 2(c2), when \(E_2\) is activated, the SWM1 (\(k_3'\)) shown by the upper curve is generated and falls into the \([4⟩−[3⟩−[2⟩\) ladder-type EIT windows shown by the lower curve. Figure 2(c3) shows the experimental results when \(E_4\) is activated and \(E_3\) is blocked. Here, the generated SWM2 signal (\(k_2'\)) shown by the upper curve falls into the \([4⟩−[3⟩−[1⟩\) V-type EIT window shown by the lower curve. However, the SWM2 process comprises six detailed Liouville pathways. Meanwhile, the absorption increases in the \(^{85}\text{Rb}\) [5S\(_{1/2}\), \(F = 3\)] dip, which has contributions from the \(E_3\) and \(E_3'\) optical pumping of population from [5S\(_{1/2}\), \(F = 3\)]. Absorption decreases in the \(^{85}\text{Rb}\) [5S\(_{1/2}\), \(F = 2\)] dip also appear because of the leakage of probe transmission, which has a contribution from the \(E_4\) optical pumping of population from the ground-state [5S\(_{1/2}\), \(F = 2\)]. Hence, these contributions from the optical pumping of pump beams \(E_3\), \(E_3'\), and \(E_4\) can come to a balance, and there no absorption dips are produced (Figs. 2(c3)–(c4)).

Figure 2(d) shows the results with the detuning of \(\Delta_3 = 700\) MHz. The FWM signal (\(k_1'\)) in upper curve of Fig. 2(d1) is less obvious because no EIT window is present in the lower curve. A comparison of the coexistent FWM signals (\(k_1'\)) and SWM signals (\(k_3'\), \(k_2'\)) in Figs. 2(d1)–(d4) with those in Figs. 2(c1)–(c4) shows that the intensities of FWM (\(k_1'\)) and SWM signals (\(k_3'\), \(k_2'\)) at \(\Delta_3 = 700\) MHz are smaller than those at \(\Delta_3 = 61.8\) MHz. No absorption dips are produced for the same reason as that in Figs. 2(c).

Now, we focus on the interplay between the two coexistent SWM signals by varying the frequency detuning of the coupling field \(E_2\) when all five laser beams (\(E_1\), \(E_2\), \(E_3\), \(E_3'\), and \(E_4\)) are active. The experimental results with the two EIT windows overlapping or separating under four different situations and corresponding to those in previous experiments are shown in Fig. 3. For the first five-level atomic system shown in Fig. 1(a1), a ladder-type EIT window and \(Λ\)-type EIT window can coexist. The FWM (\(k_1\)) and SWM signals (\(k_{11}\), \(k_{22}\)) can be controlled to overlap or separate by varying the frequency detuning of the coupling beam \(E_2\). The upper curves in Fig. 3 show the interplay and competition between two coexistent SWM processes. In Figs. 3(a1)–(a5) and 3(b1)–(b5), two generated SWM signals can be observed by blocking different laser beams and scanning the probe detuning \(\Delta_1\). In Figs. 3(a1)–(a5), when the frequency of \(E_2\) is changed from \(-192\) to \(176\) MHz, two EIT windows begin to overlap, whereas two SWM signals (\(k_{11}\), \(k_{22}\)) begin to compete. When the SWM2 signal (\(k_{22}\)) and the right peak of SWM1 signal (\(k_{11}\)) overlap, an enhancement for the SWM2 signal (\(k_{22}\)) at the resonance \(\Delta_1 + \Delta_2 - G_1 = 0\) (from the dressing field \(E_2\)) and a suppression for the SWM1 signal (\(k_{11}\)) at the resonance.
Fig. 3. (Color online) (a) and (b) Evolution of the interplay between two coexistent MWM signals of the atomic system shown in Fig. 1(a1); (c) and (d) evolution of the interplay between two coexistent MWM signals of the system shown in Fig. 1(a2). In each panel, the upper curves show the MWM signals and the lower curves show the probe transmission signals. The experimental condition is the same as that in Figs. 2- (a1)–(a4): $\Delta_2 = -192, -52, 10, 47, 176$ MHz, gradually. (b1)–(b4): $\Delta_2 = -123, -23, -15, 33, 141$ MHz, gradually. (c1)–(c4): $\Delta_2 = -30, -15, -10, 10, 28$ MHz, gradually. (d1)– (d4): $\Delta_2 = -96, -49, -41, -34, 0$ MHz, gradually. Other parameters are $G_1 = 4.6$ MHz, $G_2 = 50$ MHz, $G_3 = G_4 = 20$ MHz, and $G_4 = 30$ MHz.

$\Delta_1 - \Delta_4 = 0$ (from another dressing field $E_4$) occur, respectively. The total influence between these two SWM signals ($k_{s1}, k_{s2}$) depends on the competition between the enhancement and suppression. As Fig. 3(a2) shown, because the suppression for SWM1 ($k_{s1}$) is stronger than the enhancement caused by SWM2 ($k_{s2}$), the final influence is suppression. When the SWM2 signal ($k_{s2}$) and the middle dip of SWM1 signal ($k_{s1}$) overlap, these two modified SWM signal intensities show the suppression at exact resonances $\Delta_1 = \Delta_4 = 0$ (from the dressing field $E_4$) and $\Delta_1 + \Delta_4 = 0$ (from another dressing field $E_2$), respectively; the final influence is suppression (Fig. 3(a3)). When the SWM2 signal ($k_{s2}$) and the left peak of SWM1 signal ($k_{s1}$) overlap, an enhancement for the SWM2 signal ($k_{s2}$) at the resonance $\Delta_1 + \Delta_2 + G_2 = 0$ (from the dressing field $E_2$) and a suppression for the SWM1 signal ($k_{s1}$) at the resonance $\Delta_1 - \Delta_4 = 0$ (from another dressing field $E_4$) occur, respectively. As Fig. 3(a4) shown, the total influence brought by these two dressed SWM signals is enhancement. When the two EIT windows begin to separate, the interaction between these two SWM signals ($k_{s1}, k_{s2}$) diminishes. Figures 3(b1)–(b5) ($\Delta_3 = -700$ MHz), where the frequency of $E_2$ is changed from $-123$ to 141 MHz, show that when SWM2 signal ($k_{s2}$) and the right or left peak of SWM1 signal ($k_{s1}$) overlap, a phenomenon similar to that in Figs. 3(a2) and (a4) occurs. When the SWM2 signal ($k_{s2}$) and the middle dip of SWM1 signal ($k_{s1}$) overlap, the condition of suppression is the same as that in Fig. 3(a3) for both two SWM signals ($k_{s1}, k_{s2}$). Here, the suppression in Fig. 3(b3) is weaker than that in Fig. 3(a3).

Figures 3(c1)–(c4) and (d1)–(d4) show the interplay between SWM signals ($k_{s1}', k_{s2}'$) in another energy level system shown in Fig. 1(a2). Figures 3(c1)–(c4) ($\Delta_3 = 61.8$ MHz) show that, because there is only one peak in the SWM signals ($k_{s1}', k_{s2}'$), by changing the frequency of $E_2$ from $-30$ to 28 MHz, when SWM2 signal ($k_{s2}'$) and the left or right side of SWM1 signal peak ($k_{s1}'$) overlap, there is a suppression for SWM1 signal ($k_{s1}'$) at exact resonance $\Delta_1 - \Delta_4 = 0$ (from the dressing field $E_4$) and an enhancement for SWM2 signal ($k_{s2}'$) at exact resonances $\Delta_1 + \Delta_2 = 0$ (from another dressing field $E_2$). As shown in Figs. 3(c2) and (c4), the total influence between these two SWM signals is suppression. However, when SWM2 signal ($k_{s2}'$) and SWM1 signal ($k_{s1}'$) overlap completely, there exist suppressions for both the SWM signals ($k_{s1}', k_{s2}'$) [Fig. 3(c3)] at exact resonances $\Delta_1 - \Delta_4 = 0$ (from the dressing field $E_4$) and $\Delta_1 + \Delta_4 = 0$ (from another dressing field $E_2$). Here, because optical pumping is stronger, the final influence is weak suppression. In Figs. 3(d1)–(d4) ($\Delta_3 = -700$ MHz), the frequency of $E_2$ is changed from $-96$ to 0 MHz. When SWM2 signal ($k_{s2}'$) and the right or left side of SWM1 signal ($k_{s1}'$) overlap, a phenomenon similar to that shown in Figs. 3(c2) and (c4) is obtained. When the SWM2 signal ($k_{s2}'$) and SWM1 signals ($k_{s1}'$) overlap completely, two suppressions for both the SWM signals ($k_{s1}', k_{s2}'$) at the exact resonances $\Delta_1 - \Delta_4 = 0$ (from the dressing field $E_4$) and $\Delta_1 + \Delta_4 = 0$ (from the dressing field $E_2$), respectively, exist. As shown in Fig. 3(d3), the final influence is suppression.

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