Opening Four-Wave Mixing and Six-Wave Mixing Channels via Dual Electromagnetically Induced Transparency Windows

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Highly efficient four-wave mixing (FWM) and six-wave mixing (SWM) processes can coexist in a four-level $Y$-type atomic system due to atomic coherence. The simultaneously opened dual electromagnetically induced transparency windows in this four-level atomic system allow observation of these two nonlinear optical processes at the same time, which enables detailed studies of the interplay between the FWM and SWM processes. Three-photon and five-photon destructive interferences are also observed.

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Enhanced four-wave mixing (FWM) processes due to atomic coherence have been experimentally demonstrated in several multilevel atomic systems [1–5]. The keys to such enhanced nonlinear optical processes include the enhanced nonlinear susceptibility due to the induced atomic coherence and slowed laser beam propagation in the atomic medium, as well as greatly reduced linear absorption of the generated optical field due to electromagnetically induced transparency (EIT) [6,7]. Recently, the generation of six-wave mixing (SWM) has been reported in a closed-cycle $N$-type system in a cold atomic sample [8]. On the other hand, two-photon and three-photon destructive interferences have also been studied in various four-level $Y$-type [9], $N$-type [10], and double-Λ-type [2,5] atomic systems. These multiphoton interferences and light-induced atomic coherence are very important in nonlinear wave-mixing processes and might be used to open certain nonlinear optical processes in multilevel atomic systems that are otherwise closed due to high absorption [11]. Recently, there has been some theoretical interest in generating efficient $\chi^{(5)}$ processes in multilevel atomic systems for 3-qubit quantum computation [12] and liquid light condensate [13].

In this Letter, we report our experimental demonstration of generating highly efficient FWM and SWM processes simultaneously in an open-cycle $Y$-type atomic system, in which the dual-EIT windows are used to transmit the generated FWM and SWM signals, respectively. Several features in this work are distinctly different and advantageous over the previously reported SWM processes [8,14]. First, FWM and SWM processes can be observed simultaneously in this open-cycle $Y$-type system, which is not the case in the closed-cycle $N$-type system [8]. Such coexistence of FWM and SWM processes allows us to investigate the interplay between these two interesting nonlinear optical effects, and to obtain the beat signal between them to get the $\chi^{(5)}$ coefficient. Second, the generated FWM and SWM signals fall into two separate EIT windows in this four-level dual-EIT system, so the linear absorptions for the generated FWM and SWM signals are both greatly suppressed. By individually controlling (or tuning) the EIT windows, the generated FWM and SWM signals can be clearly separated and distinguished or pulled together (by frequency detunings) to observe interferences between them. Third, since the amplitude of the FWM signal can be controlled by the coupling beam (via dressed states), the relative strengths of the FWM and SWM can be adjusted easily. So, the SWM signal can be made to be in the same order as the FWM signal. Fourth, multiphoton destructive interference effects for both FWM (three-photon interference) and SWM (five-photon interference) are clearly observed in the experiment. Although double-resonance [9] and triple-resonance [10,15] EIT spectroscopies have been reported previously by detecting fluorescence, the current method is a coherent phenomenon. Finally, by designing the propagation directions of the (pump, coupling, and probe) laser beams, we can achieve Doppler-free configurations [7] for all the EIT subsystems in this $Y$-type atomic system. This makes the FWM and SWM processes very efficient even with relatively weak cw laser beams in an atomic vapor cell.

To the best of our knowledge, such a phenomenon of coexisting FWM and SWM channels via dual-EIT windows in multilevel atomic systems has not been reported, either experimentally or theoretically, in the literature. This specially designed experimental scheme to simultaneously generate different nonlinear wave-mixing processes opens a new research frontier in manipulating higher-order nonlinear optical processes with induced atomic coherence and quantum interference.

For a simple four-level $Y$-type atomic system as shown in Fig. 1(b), if two strong laser beams drive the two upper transitions ($|1\rangle$ to $|2\rangle$ and $|1\rangle$ to $|3\rangle$, respectively) and a weak laser beam probes the lower transition ($|0\rangle$ to $|1\rangle$), two ladder-type EIT subsystems will form and two EIT windows appear [9]. Depending on the frequency detunings of the two coupling beams, these two EIT windows can either overlap or be separated in frequency on the probe beam transmission signal. Now, if two pump fields, $E_3$ ($\omega_3$, $k_3$, and Rabi frequency $G_3$) and $E'_3$ ($\omega_3$, $k'_3$, and Rabi frequency $G'_3$), drive the upper transition $|1\rangle$ to $|3\rangle$ and one strong field, $E_2$ ($\omega_2$, $k_2$, and Rabi frequency $G_2$),
will only consider the diffracted FWM and SWM signals relevant to our experimental measurements. By setting the propagation direction of the probe beam $E_1$ as indicated in Fig. 1(a) and blocking $E_3$, the diffracted FWM ($E_j$) and SWM ($E_j$) signal beams will be in the same direction determined by the phase-matching conditions: $k_j = k_1 + k_3 - k'_3$ and $k_j = k_1 + k_2 - k_3 - k'_3$, respectively. The total FWM process can be considered as due to the constructive or destructive interference between the two dressed FWM channels. We can write down the two SWM processes [indicated in Figs. 1(e) and 1(f)] as (II) \[ \rho_{00}^{(0)} \omega_1 \rho_{10}^{(1)} \omega_2 \rho_{20}^{(2)} \omega_3 \rho_{30}^{(3)} \omega_4 \rho_{40}^{(4)} \omega_5 \rho_{50}^{(5)} \] and (III) \[ \rho_{00}^{(0)} \omega_1 \rho_{11}^{(1)} \omega_2 \rho_{21}^{(2)} \omega_3 \rho_{31}^{(3)} \omega_4 \rho_{41}^{(4)} \omega_5 \rho_{51}^{(5)} \] respectively.

In general for arbitrary field strengths of $E_2$, $E_3$, and $E'_3$, one needs to solve the 11 coupled density-matrix equations to higher orders to obtain $\rho_{10}^{(3)}$ for the FWM and $\rho_{10}^{(5)}$ for the SWM processes, which we have done in simulating the experimental results later on. In order to see the relation and interplay between these FWM and SWM processes, we calculate these nonlinear susceptibilities via appropriate perturbation chains here for simplicity. When both $E_2$ and $E'_3$ are blocked, the simple FWM via chain (I) gives $\rho_{10}^{(3)} = -iG_{a} e^{i k_3 \cdot \vec{r}} / (d_1^2 d_3)$, where $G_{a} = G_{1}G_{3}/G_{j}^2$, $d_{1} = \Gamma_{10} + i \Delta_{1}$, $d_{3} = \Gamma_{30} + i (\Delta_{1} + \Delta_{3})$ with $\Delta_{j} = \Omega_{j} - \omega_{j}$, and $\Gamma_{ij}$ is the transverse relaxation rate between states $| i \rangle$ and $| j \rangle$. Next, when the coupling field $E_3$ is turned on, the above simple FWM process will be dressed and a perturbative approach for such interaction can be described by the following coupled equations:

\[
\begin{align*}
\frac{d\rho_{10}^{(1)}}{dt} &= -d_{1} \rho_{10}^{(1)} + iG_{1} e^{i k_{1} \cdot \vec{r}} \rho_{00}^{(0)} + iG_{2} e^{-i k_{3} \cdot \vec{r}} \rho_{20}^{(2)} \\
\frac{d\rho_{20}^{(2)}}{dt} &= -d_{2} \rho_{20}^{(2)} + iG_{2} e^{i k_{2} \cdot \vec{r}} \rho_{10}^{(1)},
\end{align*}
\]

\[
\begin{align*}
\frac{d\rho_{10}^{(3)}}{dt} &= -d_{1} \rho_{10}^{(3)} + iG_{1} e^{i k_{1} \cdot \vec{r}} \rho_{00}^{(0)} + iG_{2} e^{-i k_{3} \cdot \vec{r}} \rho_{20}^{(2)} \\
\frac{d\rho_{20}^{(3)}}{dt} &= -d_{2} \rho_{20}^{(3)} + iG_{2} e^{i k_{2} \cdot \vec{r}} \rho_{10}^{(1)},
\end{align*}
\]

where $d_2 = \Gamma_{20} + i (\Delta_{1} + \Delta_{2})$. Equations (1) and (2) ($\rho_{00}^{(0)} = 1$) can be solved together with chain (I) to give $\rho_{10}^{(n)} = -2iG_{a} e^{i k_{3} \cdot \vec{r}} / [d_{1} d_{3} (d_{1} d_{2} + |G_{a}|^2)]$.

Expression $\rho_{10}^{(1)}$ shows an interesting interplay between the FWM and SWM processes [16]. With coupling field $E_2$ on, the intermediate energy level $| 1 \rangle$ is dressed to become two split levels $| + \rangle$ and $| - \rangle$ with induced coherence between them. The FWM signals will have quantum interference via these two different split intermediate levels $| + \rangle$ and $| - \rangle$, which can either enhance or suppress the total observed FWM signal. When the coupling field $G_{2}$ is very strong [$G_{2} \gg G_{j} (G_{3} \gg G_{j})$], there exists a maximum suppression of the FWM at the condition $\Delta_{1} = -\Delta_{2} = -\Delta_{3}$. Also, one can easily calculate the susceptibility $\rho_{10}^{(5)}$ for SWM from pathways (II) and (III) [as shown in
Figs. 1(e) and 1(f)] directly to be $\rho_{10}^{(5)} = \rho_{10}^{(II)} + \rho_{10}^{(III)} = 2iG_1^2|G_2|^2e^{iky+d_1d_2d_3}$.

The two EIT windows are generated by the double-ladder EIT subsystems in the $Y$-type four-level system with both pump fields (between $|1\rangle$ and $|3\rangle$) and coupling field (between $|1\rangle$ and $|2\rangle$) stronger than the probe beam (between $|0\rangle$ and $|1\rangle$), as shown in Fig. 1(c). Since the generated SWM signal falls into one ladder-type EIT window ($|0\rangle - |1\rangle - |2\rangle$ branch), the SWM processes can be very efficient, especially when the FWM signal is suppressed. For finite frequency detunings $\Delta_2$ and $\Delta_3$, the two EIT windows in the $Y$-type system will be separated, and the generated FWM and SWM signals in these two EIT windows are easily distinguishable. There is one three-photon interference pathway (i.e., interference between $\chi^{(1)}(\omega_1 + \omega_3 - \omega_2)$ and $\omega_1$) for the FWM process and two five-photon interference pathways (i.e., between five-photon $\omega_1 + \omega_2 - \omega_3 + \omega_4 - \omega_5$ and $\omega_1$, and between five-photon $\omega_1 + \omega_2 - \omega_3 - \omega_4 + \omega_5$ and $\omega_1$) for the SWM processes. In this system, the three-photon and five-photon interferences are destructive ones, in which $\chi^{(1)}$ and $\chi^{(5)}$ are zeros at the line centers. Coexisting SWM and FWM signal efficiencies and the amount of suppression of the FWM signal are most prominent under the multiple-EIT condition of $\Delta_1 = -\Delta_2 = -\Delta_3$ and $G_2 \gg G_3 (G'_1) \gg G_1$ in Fig. 1(c) and $G_3 \gg G_2 (G'_1) \gg G_1$ in Fig. 1(d).

The experimental demonstrations of coexisting FWM and SWM processes, as well as controllable FWM and SWM processes, were carried out in atomic vapor of $^{87}$Rb. The energy levels of $5s_{1/2}(F = 2), 5p_{3/2}, 5d_{3/2},$ and $5d_{5/2}$ form the four-level $Y$-type system as shown in Fig. 1(b). The laser beams were carefully aligned as indicated in Fig. 1(a) (without $E'_2$). The vapor cell temperature is set at 60 $^\circ$C. The probe laser beam $E_1$ with wavelength of 780 nm from an external cavity diode laser (ECDL), connecting transition $5s_{1/2} - 5p_{3/2}$ is horizontally polarized and has a power of about $P_1 = 3.5$ mW. The pump laser beams $E_3$ and $E'_3$ (wavelength 775.98 nm connecting transition $5p_{3/2} - 5d_{5/2}$) are split from a cw Ti:sapphire laser with equal power ($P_3 = P'_3$), each with a vertical polarization. The coupling laser beam $E_2$ (with power $P_2$, and wavelength 776.16 nm) connecting transition $5p_{3/2} - 5d_{3/2}$ is from another ECDL and is vertically polarized. Great care was taken in aligning the laser beams with spatial overlaps and wave vector phase-matching conditions with small angles ($\sim 0.3^\circ$) between them. The diameters at the vapor cell center for the pump and coupling beams are about 0.5 mm, respectively, and the diameter of the probe beam ($E_1$) is about 0.3 mm. The diffracted FWM and SWM signals ($k_f$ and $k_s$) with horizontal polarization are in the direction of $E_f$ and $E_s$ (at the lower right-hand corner of Fig. 1(a)) and are detected by an avalanche photodiode detector (APD). The transmitted probe beam is simultaneously detected by a silicon photodiode.

The dual-EIT windows of the $Y$-type system are measured by setting the frequency detunings at $\Delta_2 = -112$ MHz and $\Delta_3 = 0$, with three laser beams ($E_2$, $E_3$, and $E'_3$) on. These two modified EIT windows from the two ladder-type EIT subsystems (at $\Delta_1 = -\Delta_2$ and $\Delta_1 = -\Delta_3$ EIT positions in probe transmission trace) are depicted in Fig. 2 [peaks 4 and 5 of curve (b)]. Meanwhile, as the probe detuning $\Delta_1$ is scanned, several generated wave-mixing signals are observed on the APD [curve (a) of Fig. 2]. We identify peak 2 as a combination of the FWM signal ($k_f$) and a small amount of SWM signal ($k_s$). Peak 3 is the SWM signal ($k_s$) and peak 1 is another FWM signal ($k_f$) outside the EIT windows. Since the FWM and SWM signals are diffracted in the same spatial direction, we identify them by selectively blocking different laser beams and detuning different laser frequencies. We intentionally set large frequency detunings to separate the generated FWM and SWM signals in Fig. 2 for clarity. When the difference between $\Delta_2$ and $\Delta_3$ is reduced, these two EIT windows start to merge and the FWM and SWM signals begin to interfere. The dip in the middle of the FWM signal [peak 2 of curve (a) in Fig. 2] is due to three-photon destructive interference.

Figure 3(a) shows the FWM signal as a function of coupling power $P_2$, corresponding to peak 2 in Fig. 2. The top curve is for $P_2 = 0$, which is a simple FWM signal. The dip in the middle is due to three-photon destructive interference. As the coupling field $E_2$ is turned on, SWM signals within EIT windows (in peak 2 and peak 3 in Fig. 2) start to emerge and increase as $P_2$ increases. Such increases of SWM signals are at the cost of the FWM process. As shown in Figs. 3(a) and 3(b), as $P_2$ increases, the FWM signal decreases substantially (about 3 times in the $P_2$ power range shown) due to the destructive interference between two dressed FWM channels. In such a way, the relative strengths between FWM and SWM signals in the two EIT windows can be adjusted by simply tuning the coupling beam power $P_2$, which is very important for the

FIG. 2. Curve (a): Measured SWM (peak 3) and FWM (peaks 1 and 2) signals. Curve (b): Probe beam transmission (peaks 4 and 5: two ladder-type EIT windows) versus the probe detuning $\Delta_1$. The experimental parameters are $P_1 = 3.6$ mW, $P'_3 = 0$, $P_2 = 33$ mW, $P_3 = P'_3 = 130$ mW, $\Delta_2 = -112$ MHz, and $\Delta_3 = 0$. 

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interplay and control between FWM and SWM processes in this system. Maximal enhancement of SWM signal and the largest suppression of FWM signal can be achieved at the multi-EIT condition of $\Delta_1 = -\Delta_2 = -\Delta_3$ with a large value of $P_2$.

Figure 4 presents the changes of the SWM signal (corresponding to peak 3 in Fig. 2) as a function of the coupling field frequency detuning. It is seen from Fig. 4(a) that, as the coupling frequency detuning $\Delta_2$ changes, the generated SWM signal changes from symmetric to asymmetric, which is due to two-photon [7] or three-photon [10] resonant emission enhancement. Such asymmetric SWM spectra have been simulated by numerically solving the 11 coupled density-matrix equations for the system at the steady state and are plotted in Fig. 4(b). The dips at the line center of the SWM spectra are due to five-photon (one probe photon plus four pump and coupling photons) destructive interference with the generated signal photon, which looks like a multiphoton EIT phenomenon but is actually a suppression of generating SWM due to multiphoton destructive interference at the exact resonance.

The maximal FWM and SWM efficiencies in this system are quite high (measured to be about 10% and 1%, respectively). The coexistence of these two nonlinear wave-mixing processes and the generated signal photon, which is due to two-photon [7] or three-photon [10] resonant emission enhancement, can have important applications in many areas of physics. Understanding and optimizing higher-order nonlinear optical processes can have important applications in many areas of physics. We acknowledge funding support from the National Science Foundation.

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