Enhancement of six-wave mixing by atomic coherence in a four-level inverted Y system

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The authors have considered the coexisting dressed four-wave mixing (FWM) and six-wave mixing (SWM) in an open four-level inverted Y configuration. The authors also report an experimental observation of optical pumping-assisted FWM and electromagnetically induced transparency (EIT)-assisted SWM. The efficient SWM can be selected by EIT window and controlled by the coupling as well as dressed field detuning and power. Due to EIT and optical pumping assistance, the enhanced SWM signal is more than ten times larger than the coexisting FWM signal. © 2007 American Institute of Physics. [DOI: 10.1063/1.2713868]

Efficient higher-order multiwave mixing effects have been studied recently.\textsuperscript{1–5} Under electromagnetically induced transparency (EIT) conditions,\textsuperscript{6} not only the four-wave mixing (FWM) can be resonantly enhanced but also the generated FWM signals can be allowed to transmit through the atomic medium with little absorption.\textsuperscript{1–5} The six-wave mixing (SWM) has also been observed in a closed-cycle four-level cold atom\textsuperscript{6} and doubly excited autoionizing Rydberg states.\textsuperscript{7} The nonlinear signal decreases by several orders of magnitude with an increase in the order of nonlinearity of the interaction.\textsuperscript{3} On the contrary to the solid or liquid media (in which FWM and SWM signals can be generated at the same time),\textsuperscript{8} only either FWM processes or SWM processes were experimentally generated in typical closed-cycle multilevel atomic systems previously.\textsuperscript{1–7} depending on the arrangements of lasers and number of energy levels involved. There have also been some theoretical interests to generate efficient coexisting $\chi^{(3)}$ and $\chi^{(5)}$ nonlinear processes in multilevel atomic systems.\textsuperscript{11}

In this letter, we consider the interplay between the coexisting FWM and SWM processes and demonstrate such coexistence in one atomic system by employing a specially designed experimental scheme [Fig. 1(a)]. More importantly, we can optimize the SWM process via opened EIT window and optical pumping from an additional hyperfine energy level (3)). Investigations of such intermixing and interplay between different types of nonlinear wave-mixing processes will help us to understand and optimize the generated higher-order nonlinear optical signals.

The cw laser beams are aligned spatially in the pattern, as shown in Fig. 1(a), with four beams ($E_2, E_3, E_5,$ and $E' \text{c}$) propagating through the atomic medium in the same direction with small angles ($\sim 0.3^{\circ}$) between them in a square-box pattern. For a four-level inverted Y-type atomic system, as shown in Figs. 1(b)–1(g), if three strong laser beams (with either $E_2' \text{ or } E_3' \text{ beam blocked}$) drive the two transitions (1) to (2) and (1) to (3) and a weak laser beam (propagating in the opposite direction) probes the transition (1) to (1)), these configurations satisfy the two-photon Doppler-free condition for the |0⟩→|1⟩→|2⟩ ladder-type EIT subsystem (it is not the case for the |0⟩→|1⟩→|3⟩ A-type EIT subsystem).\textsuperscript{12}

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bative approach for such interaction can be described by following two pairs of coupled equations:

\[ \frac{d\rho^{(1)}}{dt} = -i\hbar^{(1)} \rho^{(1)} + iG_{F} e^{i\mathbf{k}_{F} \cdot \mathbf{r}} \rho^{(0)} + iG_{S} e^{-i\mathbf{k}_{S} \cdot \mathbf{r}} \rho_{20} \]

and \[ \frac{d\rho_{30}}{dt} = -d_{30} \rho_{30} + iG_{E} e^{i\mathbf{k}_{E} \cdot \mathbf{r}} \rho_{10}^{(1)} , \]

\[ \frac{d\rho^{(3)}}{dt} = -i\hbar^{(3)} \rho^{(3)} + iG_{F} e^{i\mathbf{k}_{F} \cdot \mathbf{r}} \rho_{20} + iG_{S} e^{i\mathbf{k}_{S} \cdot \mathbf{r}} \rho_{30}^{(2)} \]

and \[ \frac{d\rho_{30}}{dt} = -d_{30} \rho_{30} + iG_{E} e^{-i\mathbf{k}_{E} \cdot \mathbf{r}} \rho_{10}^{(3)} , \]  

where \( d_{1} = \gamma_{20} + i(\Delta_{1} + \Delta_{2}) \). In the steady state, Eqs. (1) and (2) can be solved together with perturbation chain (F1) to give \( \rho_{F}^{(3)} = -2iG_{E} e^{i\mathbf{k}_{E} \cdot \mathbf{r}} d_{1} d_{2} \rho_{30} \). Second, one can easily calculate the susceptibility \( \chi^{(5)} \) for SWM from the Liouville pathways (S1) and (S2) for arbitrary \( G_{2} \) to be

\[ \chi^{(5)} = 2iG_{G} G_{F} e^{i\mathbf{k}_{F} \cdot \mathbf{r}} / (d_{1} d_{2} d_{3} + |G_{S}|^{2}) . \]

Under \( |G_{S}|^{2} \ll \Gamma_{10}^{10} \), \( P_{10}^{(3)} \) can be expanded to be

\[ P_{10}^{(3)} = -2iG_{E} G_{F} e^{i\mathbf{k}_{E} \cdot \mathbf{r}} / (d_{1} d_{2}) (1 - |G_{S}|^{2} / d_{1} d_{2}) \]

and the dressed FWM process converts to a coherent superposition of signals from FWM and SWM in the weak coupling field limit.\(^{13}\)

Similarly, if we block \( E'_{3} \) instead of \( E_{3} \) [Fig. 1(d)], we obtain the dressed FWM of chain (F2)

\[ \rho_{F}^{(3)} = -2iG_{E} G_{F} e^{i\mathbf{k}_{E} \cdot \mathbf{r}} / (d_{1} d_{2} d_{3} + |G_{S}|^{2}) \]

with \( k_{2} = k_{1} + k_{2} - k_{3} \). and the dressed chain \( \rho_{F}^{(3)} \) with the perturbed state [Fig. 1(e)]. Under \( |G_{S}|^{2} \ll \Gamma_{10}^{10} \), \( P_{F}^{(3)} \) can be expanded to be

\[ P_{F}^{(3)} = -2iG_{E} G_{F} e^{i\mathbf{k}_{E} \cdot \mathbf{r}} / (d_{1} d_{2}) (1 - |G_{S}|^{2} / d_{1} d_{2}) \]

and the dressed FWM process converts to a coherent superposition of signals from FWM and SWM in the weak coupling field limit.

The two relevant experimental schemes are shown in Figs. 1(b) and 1(d). Four energy levels from \(^{85}\)Rb atoms (in vapor cell) are involved in the experimental schemes. All involved laser beams are cw lights. In the first experiment [Fig. 1(d)], energy levels of |0\( \rangle \) (\( 5S_{1/2}, F = 3 \)) \( |1\) (\( 5P_{3/2} \)) and \( |2\) (\( 5D_{3/2} \)) form a cascade three-level atomic system. With coupling beam \( E_{2} \) or \( E'_{2} \) (connecting transition |1\) to |2\) propagating in the opposite direction of the weak probe field \( E_{1} \) (connecting transition |0\) to |1\), as shown in Fig. 1(a), two-photon Doppler-free EIT condition is satisfied\(^{12}\) and a narrow EIT window appears for the probe beam, which is well understood. As Fig. 1(a) indicates, \( E_{2} \), \( E_{2}' \), and \( E_{3} \) \( E_{3}' \) (blocked) propagate through the atomic cell with a small angle (about 0.3°) and \( E_{1} \) goes in the opposite direction also with a small angle. From the phase-matching condition, the generated FWM signal \( E_{F} \) (due to one probe photon from \( E_{1} \) and one photon each from \( E_{2} \) and \( E_{2}' \), respectively) falls in a slightly different direction [lower right corner in Fig. 1(a)]. When a dressing beam \( E_{3} \) connecting transition \( 5S_{1/2} (F = 2) \rightarrow 5P_{3/2} \) is added [Fig. 1(d)], the FWM signal is greatly modified due to the constructive or destructive interference between the two dressed FWM channels \( |P_{0}^{(1)} \rangle \rightarrow |P_{3}^{(3)} \rangle \), as shown in Figs. 2(a) and 2(b), which are measured at a frequency detuning of \( \Delta_{2} = 400 \) MHz. Such dressed FWM effect has the optimum efficiency region as \( P_{3} \)
reaches $\sim 20$ mW [Fig. 2(b)]. The generated FWM signal is inside the EIT window of the cascade system, so as the coupling or probe frequency detuning changes, the FWM signal will follow the EIT window to appear at different frequencies. As one can see, even with signal will follow the EIT window to appear at different is inside the EIT window of the cascade system, so as the \( \Delta_1 = -400 \) MHz, which is a result of three-photon destructive interference (i.e., three photon, with one probe photon plus two coupling photons, process interferes with the generated signal photon destructively, as was shown in the fluorescence detection \(^1\)).

Next, we consider a different laser configuration with \( E_3 \) and \( E_3' \) coupling laser beams [connecting transition \( 55S_{1/2}(F = 2) \rightarrow 5P_{3/2} \) and one dressing laser beam \( E_2 \) (in the transition from \( 5P_{3/2} \) to \( 5D_{3/2} \)), as shown in Fig. 1(b). In this configuration, both FWM and SWM processes can coexist and be generated in the same direction satisfying \( k_{F2} \) and \( k_{E1} \), respectively. The dressed FWM and two SWM processes can be understood in simple pictures as given in Figs. 1(c) and Figs. 1(f) and 1(g), respectively. The FWM signal \( E_F \) is generated by the interaction of three fields \( E_1, E_3, \) and \( E'_3 \) [Fig. 1(b)], which can be considered as scattering of \( E_1 \) field over the small-angle single grating formed by the coupling field pair of \( E_3 \) and \( E'_3 \). Since the probe field \( E_1 \) propagates in the opposite direction of the coupling fields \( E_3 \) and \( E'_3 \), such generated FWM signal is Doppler broadened and is identified to be the broad peak in the bottom curve of Figs. 3(a) and 3(b). This FWM signal is determined by the phase-matching condition, the frequency detunings, and by blocking of each of the beams \( E_1, E_3, \) and \( E'_3 \), respectively. Similarly, SWM process (signal \( E_2 \)) can also exist at the same time with one photon each from \( E_1, E_3, \) and \( E'_3 \), and two photons from the same dressing beam \( E_2 \), as shown in Figs. 1(f) and 1(g). The generated SWM signal falls in the same EIT window of the cascade system, as shown in Fig. 3(b) (the narrow peak in the middle of the top curve). Again, this SWM signal can be identified by blocking each of the participating laser beams, the phase-matching condition, and the frequency detunings of the participating laser beams. Due to EIT and optical pumping, the enhanced SWM signal gets more than ten times larger than the coexisting FWM signal. The dip in the middle of the SWM signal is due to five-photon destructive interference (i.e., between five-photon \( \omega_1 + \omega_2 - \omega_3 + \omega_4 \) or \( \text{between five-photon} \omega_1 + \omega_2 + \omega_3 - \omega_4 \) and \( \omega_5 \), which is clearly observed in this coherent signal detection. Also, there exists an interference between coexisting weak broad FWM and the strong narrow SWM in Fig. 3 [curve (b)]. Figure 4 presents the evolutions of the EIT-assisted SWM amplitude versus the dressing and the coupling fields powers, respectively. There obviously exists an optimal SWM efficiency as \( P_3 \) reaches \( \sim 120 \) mW [Fig. 4(b)], which may correspond to the slow group velocity region of the SWM signal or the atomic coherence matching in this system.\(^2\) The measurements agree quantitatively with the theoretical calculations.\(^3\)

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