Saturation of biphoton generation near atomic resonance*

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We have numerically investigated the biphoton generation rate as a function of several parameters in the spontaneous four-wave mixing in cold atoms. It has been found that the biphoton generation rate can easily reach saturation with the intensity of the coupling laser increasing. The saturation intensity is mainly dependent on the dephasing rate of the ground states, unrelated to the pumping laser. It implies that though the biphoton waveform can be manipulated by the coupling laser, the generation rate of the biphoton cannot increase markedly after the saturation. The saturation effect also suggests that there is an optimal coupling laser for obtaining the largest biphoton generation rate with a sufficiently long coherence time.

Keywords: two-photon generation, four wave mixing, cold atoms

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1. Introduction

In recent years, the biphoton generated by spontaneous four wave mixing (SFWM) with electromagnetically induced transparency (EIT) has drawn much attention, as it has a narrow bandwidth (natural or even sub-natural), long coherence time (hundreds of ns), and a spectrum near the atomic resonance.[1−5] All these features make it have great potential in quantum communication and quantum computation, especially for long-distance quantum communication and linear optical quantum computation (LOQC).[6,7] This is because the above features not only allow the storage of photon entanglement in atomic ensembles, which are essential for quantum repeaters and efficient multiphoton entanglement in the LOQC, but also enable the interference of independent photon sources in a long time interval within the coherence time, which is the foundation for the multiphoton entanglement without synchronization.[8] By using the SFWM with EIT in cold atoms, a generation rate as high as 10^4 pairs per second for biphoton with a natural or even sub-natural bandwidth has been achieved.[3] It has also been demonstrated that the nondegenerate biphoton with both the time-frequency and the polarization entanglements can be generated by using this method.[9]

It has been shown that the Rabi frequency of the coupling laser ($\Omega_{\rm c}$) plays an important role in the waveform of the two-photon correlation in the SFWM.[9,10] The two-photon correlation has two characteristic regimes determined by the competition between the linear and the nonlinear optical responses, finally dependent on $\Omega_{\rm c}$ and the atomic optical depth $d_{\rm o}$. In the case of a small $\Omega_{\rm c}$ and a large atomic $d_{\rm o}$, the two-photon correlation has a rectangular shape similar to the conventional type-II spontaneous parametric down conversion (SPDC). While in the inverse case, the two-photon correlation presents a damped Rabi oscillation due to the interference of two four wave mixing (FWM) channels originating from the third-order susceptibility.

For practical applications, the generation rate of a biphoton source is a key parameter determining its usefulness. In this paper, we numerically investigate the biphoton generation rate as a function of several parameters in the SFWM. Taking a four-level double-Λ system for example, we give the functional relation between the biphoton generation rate and the pumping laser (its Rabi frequency $\Omega_{\rm p}$ and detuning $\Delta_{\rm p}$) and the coupling laser ($\Omega_{\rm c}$). We find there is a saturation

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effect in the biphoton generation rate even if the intensity of the coupling laser is very small. The saturation intensity of the coupling laser is mainly dependent on the dephasing rate of the ground states and the optical depth of the atomic ensembles, unrelated to the pumping laser. Although the two-photon waveform distinctly alters with Ω_c increasing, the biphoton generation rate only increases slightly after the saturation. Contrasted to the nonlinear parametric process in crystals under weak pumping, this saturation effect is clearly absent, since the nonlinear susceptibility in crystals is a constant.

Similar saturation phenomena have been widely reported in the EIT assisted four-wave mixing in cold or hot atoms.[11,12] But there is an essential difference, because the spectrum of the generated FWM signal field does not need to be considered in those cases. In the SFWM for the biphoton generation, the spectrum of paired photons must be considered in the two-photon correlation, and the generation rate of paired photons is an integral of the whole spectrum within the detection range.

This paper is organized as follows. In Section 2, we derive a simple explicit expression of the biphoton generation rate as a function of parameters Ω_p, Δ_p, Ω_c, and d_o in the SFWM. In Section 3, we numerically study the relation between the biphoton generation rate and these parameters. Analysis of the saturation effect is also given. Finally, we draw a conclusion in Section 4.

2. Biphoton generation rate

A schematic diagram of the biphoton generation by the SFWM with EIT in a four-level double-Λ system is shown in Fig. 1(a). In the presence of the pumping laser (ω_p) and the coupling laser (ω_c), paired photons (ω_s, ω_{as}) are spontaneously generated from the FWM parametric process. In the dressed state picture, the on-resonance coupling laser will split the excited state $|2\rangle$ into two, and consequently create two FWM channels in the biphoton generation, as shown in Fig. 1(b). When the nonlinear interaction dominates, due to the interference between these two FWM channels, the two-photon correlation leads to damped Rabi oscillation.[10] However, since the coupling laser also creates a transparent window for the propagation of the anti-Stokes photons with the slow light effect, the phase matching condition can wash out this interference mechanism when the linear propagation effect dominates.[9] In this condition, the two-photon correlation has a rectangular shape with very long coherence time.

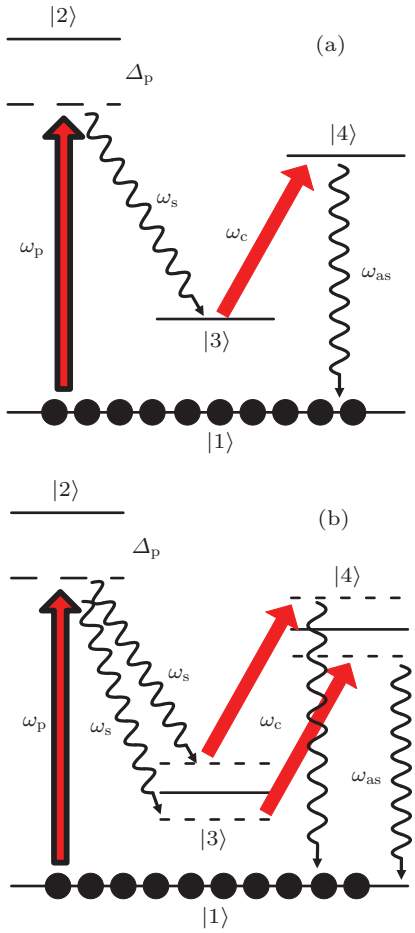


Fig. 1. (colour online) Schematic diagram of the biphoton generation via the SFWM in a four-level double-Λ system. The ground states $|1\rangle$, $|3\rangle$ are $|5S_{1/2}, F = 2\rangle$, $|5S_{1/2}, F = 3\rangle$, and the excited states $|2\rangle$, $|4\rangle$ are $|5P_{3/2}, F = 3\rangle$, $|5P_{1/2}, F = 3\rangle$ of ^{85}Rb, respectively. The atoms are prepared initially in the ground state $|1\rangle$. Panels (a) and (b) show the energy level diagram and the two FWM channels in the dressed state picture, respectively.

Now we turn to the discussion of the biphoton generation rate in the SFWM parametric process. The perturbation theory has been proved very suitable to account for the correlation of paired photons generated via the SFWM.[9] It gives that the two-photon wave function in the SFWM is the Fourier transform of the product of the nonlinear parametric coupling coefficient and the longitudinal detuning function, which can be expressed as

$$\psi(\tau) = \frac{l}{2\pi} \int \mathrm{d}\omega_{as} \kappa(\omega_{as}) \phi(\omega_{as}) \, \mathrm{e}^{-\mathrm{i}\omega_{as}\tau}, \qquad (1)$$

where the nonlinear parametric coupling coefficient $\kappa(\omega_{as})$ and the longitudinal detuning function $\phi(\omega_{as})$

respectively are

$$\kappa(\omega_{\rm as}) = -{\rm i}\,(\sqrt{\varpi_{\rm s}\varpi_{\rm as}}/2c)\,\chi^{(3)}_{\rm as}(\omega_{\rm as})E_{\rm p}E_{\rm c}\,,$$
$$\phi(\omega_{\rm as}) = {\rm sinc}(\Delta k l/2)\,{\rm e}^{{\rm i}(k_{\rm as}-k_{\rm s})l/2}. \quad (2)$$

In Eq. (1), we have ignored the constant phase, since it has no contribution to the two-photon correlation. The τ is the time interval between the detections of the anti-Stokes and the Stokes photons, $\Delta k = k_{\rm as} - k_{\rm s} - k_{\rm c} + k_{\rm p}$ is the phase mismatching for a usually backward generation configuration, $\chi^{(3)}_{\rm as}$ is the third-order susceptibility to the anti-Stokes photons, $\varpi_{\rm s}$ and $\varpi_{\rm as}$ are the central frequencies of the Stokes and the anti-Stokes photons, respectively, and l is the length of the atomic ensembles.

The biphoton generation rate is an integral of the modulus square of the two-photon wave function in the area of $\tau \in (-\infty, \infty)$, which can be written as

$$R = \int |\psi(\tau)|^2 {\rm d}\tau = \frac{1}{2\pi}\int l^2\,|\kappa(\omega_{\rm as})\phi(\omega_{\rm as})|^2 {\rm d}\omega_{\rm as}, \quad (3)$$

where the Parseval's theorem has been used.

Assuming that the atomic population remains in the ground state, we can give the linear and the nonlinear susceptibilities to the Stokes and the anti-Stokes photons in the SFWM[9,13]

$$\chi_{\rm s}(-\delta) = \frac{N|\mu_{12}|^2\Omega_{\rm p}^2}{\varepsilon_0\hbar\Delta_{\rm p}^2}\frac{(\delta + {\rm i}\gamma_{14})}{d(\delta)},$$
$$\chi_{\rm as}(\delta) = \frac{4N|\mu_{14}|^2}{\varepsilon_0\hbar}\frac{(\delta + {\rm i}\gamma_{13})}{d(\delta)},$$
$$\chi^{(3)}_{\rm as}(\delta) = (\chi^{(3)}_{\rm s}(-\delta))^* = \frac{N\mu_{12}\mu_{23}\mu_{34}\mu_{41}}{\varepsilon_0\hbar^3\Delta_{\rm p}d(\delta)}, \quad (4)$$

where $d(\delta) = \Omega_{\rm c}^2 - 4(\delta + {\rm i}\gamma_{13})(\delta + {\rm i}\gamma_{14})$, $\delta = \omega_{\rm as} - \varpi_{\rm as}$, N is the density of the atomic ensembles, μ_{ij} are the electric dipole matrix elements, γ_{ij} are the dephasing rates, $\Delta_{\rm p}$ is the pumping laser detuning, and $\Omega_{\rm p} = \mu_{12}E_{\rm p}/\hbar$ and $\Omega_{\rm c} = \mu_{14}E_{\rm p}/\hbar$ are the Rabi frequencies of the pumping and the coupling lasers, respectively. Taking into account that the relation of wavenumbers k_i and the linear susceptibility is $k_{\rm s} = \frac{\omega_i}{c}\sqrt{1+\chi_i}$, the phase mismatching Δk can be simplified as

$$\Delta k = \frac{2\omega_{31}}{c} + \frac{2d_{\rm o}\gamma_{14}(\delta + {\rm i}\gamma_{13})}{l d(\delta)}$$
$$\approx \frac{2d_{\rm o}\gamma_{14}(\delta + {\rm i}\gamma_{13})}{l d(\delta)}, \quad (5)$$

where $d_{\rm o}$ is the on-resonance optical depth of the atomic ensembles for the anti-Stokes photons, which equals to $\frac{Nl|\mu_{14}|^2\varpi_{\rm as}}{c\varepsilon_0\hbar\gamma_{14}}$,[14] ω_{31} is the frequency difference of two ground states $|3\rangle$ and $|1\rangle$, and is about 3 GHz for ^{85}Rb. In Eq. (5), we have used the approximations $\chi_{\rm s} \approx 0$ and $k_{\rm s} \approx \varpi_{\rm s}/c$, since $\Delta_{\rm p}$ is far larger than $\Omega_{\rm p}$ required by the ground state approximation.

By substituting Eqs. (2), (4), and (5) into Eq. (3), the expression of the biphoton generation rate can be further simplified as

$$R = \frac{1}{2\pi}\int {\rm d}\delta \left| \frac{B\Omega_{\rm p}\Omega_{\rm c}}{2\Delta_{\rm p}} \frac{\sin\left(\frac{d_{\rm o}\gamma_{14}(\delta + i\gamma_{13})}{d(\delta)}\right)}{(\delta + {\rm i}\gamma_{13})} \right|^2$$
$$\times \exp\left(-\frac{2d_{\rm o}\gamma_{14}}{\Omega_{\rm c}^2 + 4\gamma_{13}\gamma_{14}}\right), \quad (6)$$

where B is a constant equaling to $\sqrt{\frac{\varpi_{\rm s}}{\varpi_{\rm as}}}\frac{\mu_{23}}{\mu_{14}}$. Equation (6) is an explicit expression of parameters $\Omega_{\rm p}$, $\Delta_{\rm p}$, $\Omega_{\rm c}$, and $d_{\rm o}$. From Eq. (6), the biphoton generation rate is directly proportional to the square of $\Omega_{\rm p}/\Delta_{\rm p}$, while its dependence on parameters $\Omega_{\rm c}$ and $d_{\rm o}$ is not readily worked out. Therefore, we use the numerical simulation to examine the relation of the biphton generation rate and parameters $\Omega_{\rm c}$ and $d_{\rm o}$.

3. Numerical simulation

We consider a cloud of cold ^{85}Rb atoms prepared in a two-dimensional (2D) magneto-optical trap (MOT). The shape of the cold ^{85}Rb atomic cloud is cylinderical in the pumping and detection direction. The advantages of the ^{85}Rb 2D MOT for biphoton generation are that it not only can trap more atoms than the ordinary three-dimensional (3D) MOT, but also has a lower ground state dephasing rate.[2,3,5,15] These merits are important for the performance of the EIT and long two-photon coherence time.

Figure 2 shows the relation of the biphoton generation rate and the four parameters $\Omega_{\rm p}$, $\Delta_{\rm p}$, $\Omega_{\rm c}$, and $d_{\rm o}$. For each simulation, only one parameter changes. All parameters used in the simulation are available at the present experimental condition.[3,16] Figures 2(a) and 2(b) show that the biphoton generation rate scales squarely with $\Omega_{\rm p}/\Delta_{\rm p}$. However, due to the spontaneous radiation of the excited state $|2\rangle$, the background count scales with $(\Omega_{\rm p}/\Delta_{\rm p})^4$. Though the biphoton generation rate monotonously increases with the increasing ratio $\Omega_{\rm p}/\Delta_{\rm p}$, the background count increases more remarkablely. The overall effect decreases the factor of the violation of the Cauchy–Schwarz inequality of photon pairs. Figure 2(c) shows that the biphoton generation rate becomes saturated

at a very low coupling Rabi frequency, only requiring several γ_{14}. It's very instructive. First, for the FWM and the spontaneous parametric down conversion (SPDC) in crystals under weak pumping, this saturation effect can not happen, because the nonlinear susceptibility is a constant. Second, this saturation effect shows that the biphoton generation rate cannot increase remarkablely after the saturation. A similar saturation effect has been observed in the EIT assisted FWM. In that case, the Ω_c has an optimal value for the highest FWM conversion efficiency. Over the optimal value, the FWM conversion efficiency decreases gradually.[12] Figure 2(d) shows that the biphoton generation rate nearly scales linearly with d_o. The relevant experimental result was reported in a ^{85}Rb 2D MOT, but the dependence in the Rabi oscillation regime has not been considered.[3]

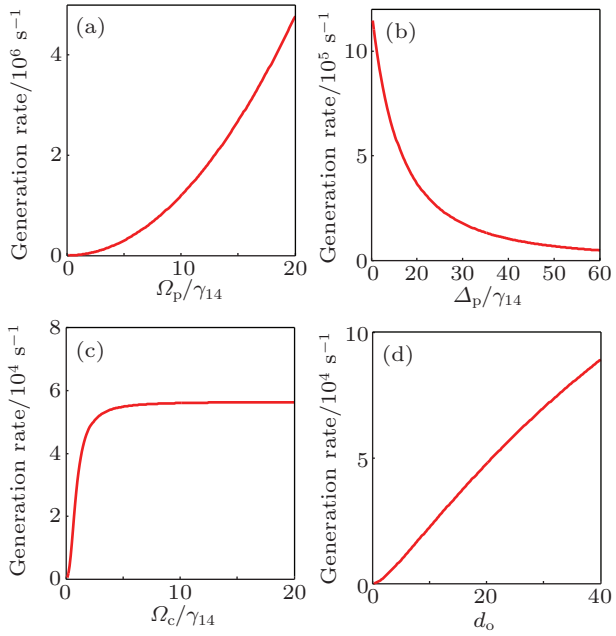


Fig. 2. (colour online) The biphoton generation rate as a function of parameters (a) Ω_p, (b) Δ_p, (c) Ω_c, and (d) d_o. If not specified, the parameters are $\gamma_{14} = 2\pi \times 10^6$ Hz, $\gamma_{13} = 0.02\gamma_{14}$, $\Omega_p = 2\gamma_{14}$, $\Delta_p = 50\gamma_{14}$, $\Omega_c = 2\gamma_{14}$, and $d_o = 20$.

Figure 3 gives the waveform of the two-photon correlation as a function of parameters Ω_c and d_o. As shown by Fig. 3(a), the increase of the Rabi frequency of the coupling laser can dramaticly alter the correlation waveform of the two-photon. In Fig. 3(b), it is obvious that with the rising of d_o, the amplitude of the two-photon correlation gradually increases, and the coherence time also improves. Comparing Fig. 3(a) with Fig. 2(c), we find that though the waveform of the two-photon correlation varies with Ω_c, the total generation rate changes slightly after the saturation.

Since Eq. (6) has shown that the saturation effect is independent of Ω_p and Δ_p, it is necessary to investigate the relation between the saturation effect and parameters γ_{13} and d_o. Generally in experiment, the dephasing rate of the ground state γ_{13} is not a constant, which is influenced by the specific experimental conditions. For example, a very large dephasing rate has been demonstrated in a three-level system for the biphoton generation.[5]

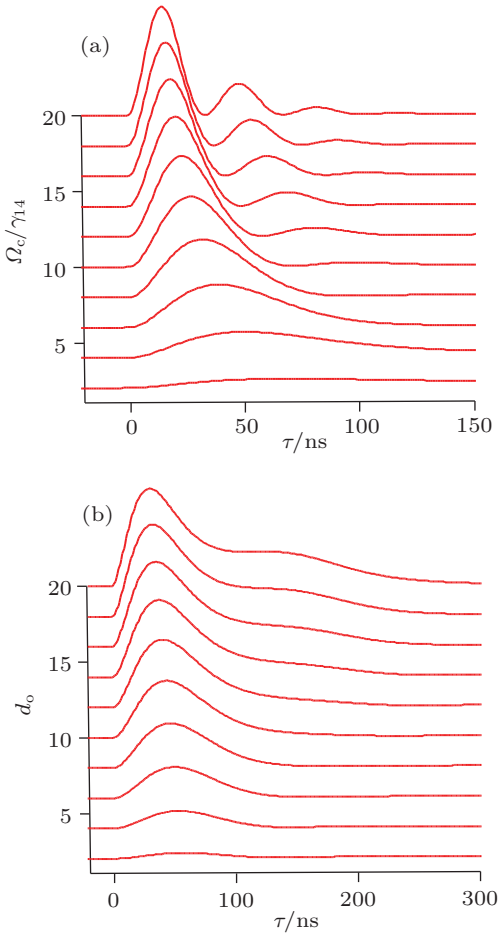


Fig. 3. (colour online) The waveform of the two-photon correlation versus (a) Ω_c and (d) d_o. If not specified, the parameters are $\gamma_{14} = 2\pi \times 10^6$ Hz, $\gamma_{13} = 0.02\gamma_{14}$, $\Omega_p = 2\gamma_{14}$, $\Delta_p = 50\gamma_{14}$, $d_o = 10$, and $\Omega_c = 5\gamma_{14}$.

Figure 4 exhibits the dependences of the saturation effect on γ_{13} and d_o. From Fig. 4(a), we can see that the larger ground state dephasing rate γ_{13} requires a higher Ω_c to get saturated. Because the larger γ_{13} can cause more absorption of the anti-Stokes photons, the biphoton generation rate is in negative correlation to γ_{13}. For the relation between the saturation effect and d_o, figure 4(b) shows that near the saturation, Ω_c increases slightly with d_o. This suggests that in a practical experiment, the saturation intensity of the coupling laser in the SFWM for biphoton genera-

tion is nearly fixed. Because the coherence time of the biphoton is inversely proportional to Ω_c, there exists an optimal Ω_c if one wants to produce the largest rate of biphoton with sufficiently long coherence time.

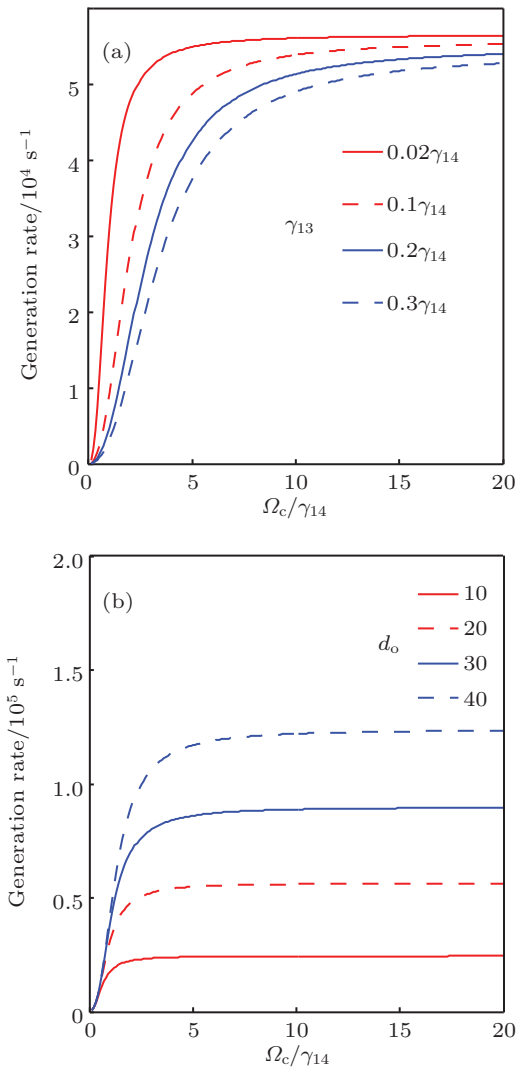


Fig. 4. (colour online) The saturation effect versus (a) γ_{13} and (b) d_o. If not specified, the parameters are $\gamma_{14} = 2\pi \times 10^6$ Hz, $\Omega_p = 2\gamma_{14}$, $\Delta_p = 50\gamma_{14}$, $d_o = 20$, and $\gamma_{13} = 0.02\gamma_{14}$.

Next let us study the biphoton generation rate in the two characteristic regimes. In the Rabi oscillation regime, the phase matching condition brought by the linear response can be ignored, thus the biphoton generation rate can be rewritten as

$$\begin{aligned} R &= \int |\psi(\tau)|^2 \mathrm{d}\tau \\ &\approx \int \mathrm{d}\tau \left| \frac{B\gamma_{14} d_o \Omega_p}{4\Delta_p} \mathrm{e}^{-\frac{\gamma_{14}+\gamma_{13}}{2}|\tau|} \sin\left(\frac{\Omega_c}{2}\tau\right) \right|^2 \\ &= \left(\frac{B\gamma_{14} d_o \Omega_p}{4\Delta_p}\right)^2 \\ &\quad \times \frac{\Omega_c^2}{(\gamma_{14}+\gamma_{13})((\gamma_{14}+\gamma_{13})^2 + \Omega_c^2)}. \end{aligned} \tag{7}$$

Equation (7) shows that in the Rabi oscillation regime, the biphoton generation rate has begun to saturate if $\Omega_c \gg (\gamma_{14} + \gamma_{13})$ is satisfied. It also gives that the biphoton generation rate scales squarely with d_o. Because the atomic optical depth in this regime is very small, this quadrate relation is soon replaced by a linear relation, as shown in Fig. 2(d). In the group delay regime, the nonlinear response can be viewed as a constant, and if d_o is very large, the biphoton generation rate is approximated to

$$R \approx \left(\frac{B\Omega_p}{2\Delta_p}\right)^2 \frac{\gamma_{14} d_o \Omega_c^2}{2(\Omega_c^2 + 4\gamma_{13}\gamma_{14})}. \tag{8}$$

From Eq. (8), the biphoton generation rate is proportional to the atomic optical depth in this regime, and the saturation condition is $\Omega_c^2 \gg 4\gamma_{13}\gamma_{14}$, which is the same as that for an observable EIT effect. Compared to the saturation condition in the Rabi oscillation regime (Eq. (7)), the requirement for saturation in the group delay regime is relaxed. Since Fig. 4(b) has shown that with the increase of d_o, the requirement of Ω_c for the saturation only slightly increases, it suggests that the saturation effect of the biphton generation rate is mainly determined by the linear response. The intensity dependence of $\chi^3(\delta)$ also contributes to the saturation effect (see Eq. (4)).

4. Conclusion

In this paper, we have studied the relation between the biphoton generation rate and several parameters in the SFWM process in cold atoms. We show that the biphoton generation rate is easily saturated with Ω_c increasing. The saturation intensity of the coupling laser is mainly dependent on the ground dephasing rate γ_{13}. By the analysis in the two characteristic regimes, the saturation condition is found to be nearly the same as that for an observable EIT effect. The existence of the saturation effect shows that though Ω_c plays an important role in the manipulation of the two-photon waveform, it cannot further significantly increase the total generation rate after the saturation. Because the coherence time of the biphoton is inversely proportional to Ω_c in the group delay regime, the saturation effect also suggests that there exists an optimal Ω_c for the largest biphoton generation rate with sufficiently long coherence time. This

saturation effect will be an obvious limitation to the biphoton generation rate via the method of SFWM in cold atoms.

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