Slow light via four-wave mixing in a hot rubidium vapour*

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In this work, we report on an off-resonant four-wave mixing experiment via a ladder-type configuration in a hot rubidium atomic vapour. We find for the first time, to the best of our knowledge, that the generated light is delayed compared with the reference. At the same time, the seeded signal beam is also delayed, though the delay time is not as so large as the one that the generated light has. Both delayed times can be adjusted experimentally by controlling the two-photon detuning. The experimental results are in good agreement with our theoretical predictions. Our results may be important for storing telecom-band photons.

Keywords: slowed light, four-wave mixing

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1. Introduction

Four-wave mixing (FWM) is a nonlinear-optical process in which three waves are combined coherently to generate the fourth wave in a medium. The frequencies and the momenta of these four waves should satisfy the requirements of conservation of energy and momentum, respectively. With the developments of quantum optics and quantum information, FWM in an atomic vapor has gradually attracted much attention because it has many applications in these two fields. For example, it can be used to produce a squeezed light,[1] to create an entangled photon pair,[2–6] and to realize a twin-beam squeezing.[7] In addition to the conservation of linear momentum and energy, angular momentum conservation should also be satisfied in FWM, which gives rise to a correlation in the orbital angular momentum among four beams.[8] One of these interesting subjects is to study the effect of delayed or stopped light in FWM process. In Ref. [9], the delayed generated field was discussed. The effect of stopping the generated light using a hot vapor cell was perfectly realized by Camacho *et al.*[10] Note that in most of the previously mentioned work a lambda-type or a double lambda-type atomic level configuration was used. Such a system benefits from long coherence time between the two stable low energy levels. However, there were few experiments in which other atomic level configurations were used. Recently, Hsu *et al.*[11] demonstrated the generation of a new field in a double-ladder system through resonant FWM in a hot atomic vapor cell, and this process was theoretically analyzed in detail in Refs. [12] and [13]. In these papers, two lasers each with a wavelength of 794.3 nm and a laser with a wavelength of 852.2 nm were used to generate a new field at 852.2 nm. Willis *et al.*[14] performed an FWM experiment in a diamond-type atomic level configuration in a warm vapor. In that experiment, three lasers with wavelengths of 780 nm, 795 nm, and 1324 nm were used to generate a new field at 1367 nm. Chanelière *et al.*[15] reported on the generation of an entangled photon pair in the telecommunications band through spontaneous FWM via a ladder-type atomic configuration in a cold atomic vapor. Very recently, we experimentally generated a new field at 780 nm by combining two fields each at 1529 nm and one field at 780 nm through FWM in the ladder-type configuration in a hot Rb atomic vapor,[16] and further, prepared a non-classical correlated photon pair in the telecom band.[17] Besides, we[16] also realized the transfer of an image between two fields with a large frequency difference via FWM in such a configuration. There are also some results about FWM process, such as dressed FWM spectroscopy,[18] suppressing and enhancing FWM via quantum interference.[19,20]

Although slow FWM light,[9] the stopped FWM light[10] in a lambda configuration and slow light in quantum dot photonic crystal waveguides[21] were obtained experimentally very recently, and also some theories about slow light in the ladder-type configuration were given,[22–24] there is no such experimental report on the ladder configuration, so we want to experimentally check whether the delayed FWM can occur in such a configuration. This is our main motivation. Such a work is definitely useful for further investigating the storage and the retrieval of a light in the ladder configuration. Our results may be important for storing telecom-band photons in an atomic ensemble, because the ability to store telecom-band photons is very important for quantum communications as is well known. In this work, we report on an off-resonant FWM in the ladder-type Rb atomic configuration. We find that the generated light is delayed compared with a reference. At the

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same time, the seeded signal beam is also delayed, though the delay time is not as large as the one that the generated light has. Both delay times can be adjusted experimentally by controlling the detuning of the pump lights. We also make a detail theoretical analysis, and the calculated results are in good agreement with the experimental results.

The rest of this paper is organized as follows. Having given a simple introduction in Section 1, we briefly introduce the atomic configuration to be used and our experimental setup in Section 2. In Section 3, we show our experimental results. Then, we give a detailed theoretical description in Section 4. Finally, we draw some conclusions from the present study in Section 5.

2. Experimental setup

The ladder-type configuration used in our experiment is shown in Fig. 1. It consists of one ground state $|1\rangle$, two degenerated intermediate states $|2\rangle$ and $|3\rangle$, and one upper state $|4\rangle$. The transition frequency between the ground state and the intermediate state matches the D_2 line (780 nm) of ^{85}Rb, and the transition between the intermediate state and the upper state can be driven by a laser at 1529.4 nm. We experimentally generate a new field at 780 nm using the combinations of a laser at 780 nm and two lasers at 1529 nm via a non-collinear FWM configuration.

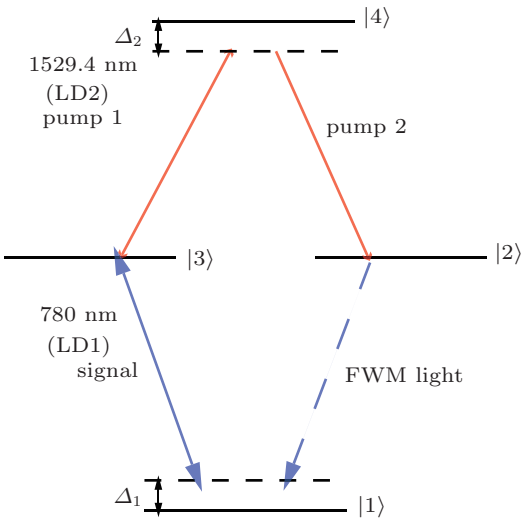


Fig. 1. (color online) Energy levels of ^{85}Rb and the applied fields in FWM experiment.

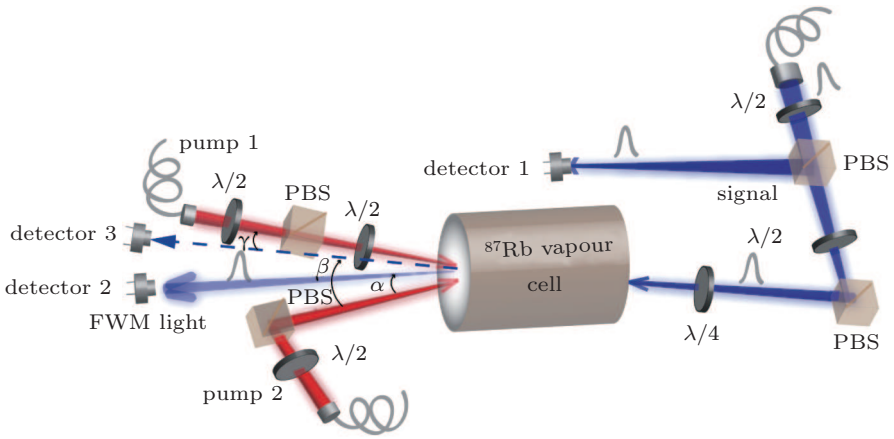


Fig. 2. (color online) Simplified experimental set-up.

The simplified experimental setup is shown in Fig. 2. The states $|1\rangle$ and $|4\rangle$ correspond to levels $5S_{1/2}$ ($F = 3$) and $4D_{5/2}$ of ^{85}Rb atoms, and states $|2\rangle$ and $|3\rangle$ are sub-levels of $5P_{3/2}$. A continuous wave (cw) laser at 780 nm from an external cavity diode (LD1, DL100, Toptica) was frequency-detuned Δ_1 to the transition of $5S_{1/2}$ ($F = 3$)–$5P_{3/2}$ ($F' = 3$), (we define that the blue shift detuning is represented with positive sign in the text) and was pulsed using an acoustic-optical modulator (AOM). A small portion of this pulsed laser, which was separated using a 1/2 wave plate and a polarization beam splitter, was used as a reference signal, and detected by detector 1. The remaining output from LD1 was attenuated and used as an input signal to a 5-cm-long cell filled with ^{85}Rb. A cw laser at 1529 nm from another external cavity diode (LD2, DL 100, Prodesign, Toptica) was divided into two pump beams by a beam splitter: pump 1 and pump 2. These two beams had parallel polarization directions and nearly co-propagated through the Rb cell. The angle $\gamma+\beta$ between these two beams was about 1.05°. LD2 was frequency-detuned $\Delta 2$ to the transition of $5P_{3/2}$ ($F' = 3$)–$4D_{5/2}$ ($F'' = 4$). The signal beam had circular polarization and nearly counter-propagated with pump 1. The angle γ between these two beams was about 0.35°. The generated FWM light, which had a small angle α of 0.35° with respect to pump 2, was detected by detector 2. The signal beam was detected by detector 3. All signals from detectors were used as the inputs to an oscilloscope. All detectors are

of exactly the same type. The signal beam had approximately Gaussian profile with a waist of 0.5 mm in the center of the cell, and its pulse width was about 200 ns (full width at half maximum). The beam waists of the other two beams in the center of the cell were 1 mm and 0.5 mm respectively. The pulse energy of each signal pulse was about 5 nJ. The powers of the two pump beams were 9.0 mW and 9.4 mW respectively. The cell was heated to about 160 °C corresponding to a density of atoms 2.1×10^{14} cm^{-3} (or 2.1×10^{20} m^{-3}).

3. Experimental results

First, we performed the FWM experiment in the cw case. In this case, all three input fields are in cw modes. During the experiment, we found that FWM conversion efficiency is strongly dependent on several parameters, such as the detunings of pump beams and the signal beam, the polarizations of the signal beam and pump beams, etc. The maximal conversion efficiency achieved in our experiment is about 54%. Here, we only show the experimental data related to the time delay. Having obtained the generated FWM light at 780 nm, we switched the system into the pulsed mode. In this case, an AOM was used to pulse the input laser at 780 nm. The other two beams at 1529.4 nm still operated in cw mode. The generated FWM signal was detected by detector 2. Simultaneously, we detected the reference and the signal beam by detectors 1 and 3 respectively. We found that the generated light is delayed compared with the reference. At the same time, the seeded signal beam is also delayed, though the delay time is not as large as the one that the generated light has. Both delay times can be adjusted by controlling the two-photon detuning. The experimental data are shown in Figs. 3 and 4(a). One set of experimental results obtained under the condition of $\Delta_1 = 1.96$ GHz and $\Delta_2 = -1.89$ GHz is shown in Fig. 3(a). It can be clearly seen that there is a time delay of about 58 ns (26 ns) between the reference and the generated FWM pulse (signal beam). If we change the two-photon detuning, both delay times change too, which are shown in Fig. 4(a). Under the condition of $\Delta_1 = 1.96$ GHz and $\Delta_2 = -1.68$ GHz, there is no obvious delay between the FWM pulse and the reference nor between the reference and the signal beam. The experimental results in this case are shown in Fig. 3(b). (The reason why the large detuning of Δ_1 and Δ_2 are chosen is that the FWM conversion efficiency is maximal in this case. This is because the large single-photon detuning can significantly reduce the resonant absorptions of the Rb to the infrared light and the generated FWM signal).

What we want to mention is that although a slow FWM signal was reported in a lambda-type configuration in Ref. [9], our experiment shows some big differences in contrast to that work: except that the configurations of atomic levels are different, nearly degenerated FWM is performed in that experiment, all beams are near co-propagated through the atoms, therefore Doppler broadening is quite small in Ref. [9]. On the contrary, in our experiment, the non-degenerated FWM is considered, all beams are counter propagated through atoms, therefore the Doppler effect must be taken in account.

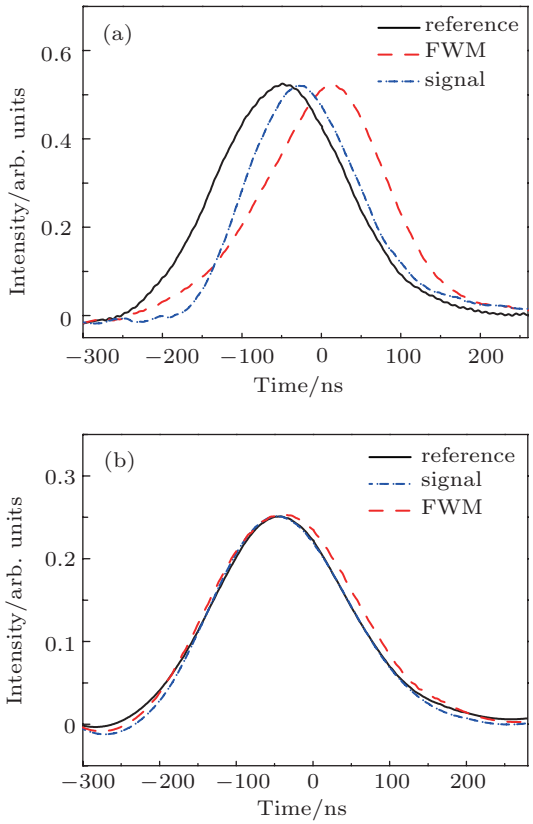


Fig. 3. (color online) (a) The signal beam (short dashed line) is delayed about 26 ns compared with the reference under the condition of $\Delta_1 = 1.96$ GHz and $\Delta_2 = -1.89$ GHz; the generated FWM beam (dashed line) is delayed about 58 ns under the same conditions; (b) both FWM beam and signal beam are not delayed under the conditions of $\Delta_1 = 1.96$ GHz and $\Delta_2 = -1.68$ GHz. The black line is the reference.

We believe that the time delay is caused by the coupling effect between the generated FWM beam and the signal beam in the FWM process, and such coupling effect brings the steep dispersion and small group velocity to the generated light and signal beam in the Rb cell. The coupling effect exists in the ladder-type configuration consisting of three energy levels: $5S_{1/2}$ ($F = 2$), $5P_{3/2}$, and $4D_{5/2}$. Both generated field and the signal beam can be derived from the coupled propagation equations. The generated FWM light (also signal beam) is delayed if the two-photon resonance is satisfied. In Fig. 3(a), the two-photon detuning is $\delta = \Delta_1 + \Delta_2 = 70$ MHz, satisfying the two-photon resonance if the Doppler effect is considered. (The Doppler width is about $(\omega_s - \omega_p)u/c = \pm 180$ MHz, which is calculated through the atomic most probable velocity $u = 290$ m/s at a temperature of 160 °C). In this case, there are two coupling coefficients k_1 and k_3 (calculated and shown in next section). Both k_1 and k_3 contribute to the delay of the generated field, only k_1 contributes to the delay of the signal beam.

The dispersion in the atom is steep at the two-photon detuning $\delta = 70$ MHz, and small group velocity for FWM light (also for signal beam) is easily obtained. On the contrary, the two-photon detuning of $\delta = \Delta_1 + \Delta_2 = 280$ MHz, like in Fig. 3(b), is beyond the width of the Doppler effect, and the dispersion from the first-order coefficient is flat, so there is no time delay.

4. Theoretical calculation

In order to support our point shown before, we make a detailed theoretical analysis in the following. The effective interaction Hamiltonian for this system is

$$\begin{aligned} H_{\rm int} &= \hbar(\Delta_1+\Delta_2)\,|4\rangle\langle 4| + \hbar\Delta_1(|2\rangle\langle 2| + |3\rangle\langle 3|) \\ &\quad -\hbar(\Omega_{\rm F}\,|2\rangle\langle 1| + \Omega_{\rm S}\,|3\rangle\langle 1| \\ &\quad +\Omega_{\rm P1}\,|4\rangle\langle 3| + \Omega_{\rm P2}\,|4\rangle\langle 2| + {\rm H.c}), \end{aligned} \tag{1}$$

where, $\Omega_{\rm P1}$, $\Omega_{\rm P2}$, $\Omega_{\rm S}$, and $\Omega_{\rm F}$ are Rabi frequencies of Pump 1, Pump 2, Signal and the generated FWM fields; Δ_1 and Δ_2 are the frequency detunings of Signal and Pump 1, the generated FWM light fields respectively. The density matrix equation of motion is

$$\dot{\rho}_{ij} = -\frac{\rm i}{\hbar}\sum_k (H_{ik}\rho_{kj} - \rho_{ik}H_{kj}) - \frac{1}{2}\sum_k (\Gamma_{ik}\rho_{kj} + \rho_{ik}\Gamma_{kj}), \tag{2}$$

where Γ is a relaxation matrix. We assume $\Omega_{\rm P1}$, $\Omega_{\rm P2} \gg \Omega_{\rm S}$, $\Omega_{\rm F}$. According to Refs. [13] and [21], the solutions of off-diagonal elements ρ_{21} and ρ_{31} for steady-state (high-order terms are ignored) can be calculated as

$$\rho_{21} = A_{21}^{(1)}\Omega_{\rm F} + A_{21}^{(3)}\Omega_{\rm S}, \tag{3}$$

$$\rho_{31} = A_{31}^{(1)}\Omega_{\rm S} + A_{31}^{(3)}\Omega_{\rm F}, \tag{4}$$

where

$$\begin{aligned} A_{21}^{(1)} &= \frac{{\rm i}(|\Omega_{\rm P1}|^2 + \Gamma_{21}\Gamma_{41})}{\Gamma_{21}(|\Omega_{\rm P1}|^2 + |\Omega_{\rm P2}|^2 + \Gamma_{21}\Gamma_{41})}, \\ A_{21}^{(3)} &= \frac{-{\rm i}\Omega_{\rm P2}^*\Omega_{\rm P1}}{\Gamma_{21}(|\Omega_{\rm P1}|^2 + |\Omega_{\rm P2}|^2 + \Gamma_{21}\Gamma_{41})}, \\ A_{31}^{(1)} &= \frac{{\rm i}(|\Omega_{\rm P2}|^2 + \Gamma_{21}\Gamma_{41})}{\Gamma_{21}(|\Omega_{\rm P1}|^2 + |\Omega_{\rm P2}|^2 + \Gamma_{21}\Gamma_{41})}, \\ A_{31}^{(3)} &= \frac{-{\rm i}\Omega_{\rm P2}\Omega_{\rm P1}^*}{\Gamma_{21}(|\Omega_{\rm P1}|^2 + |\Omega_{\rm P2}|^2 + \Gamma_{21}\Gamma_{41})}, \end{aligned}$$

where Γ_{21} and Γ_{41} are complex relaxation rates defined as $\Gamma_{21} = \gamma' + {\rm i}\Delta_1$ and $\Gamma_{41} = \Gamma + \gamma' + {\rm i}\delta$, with $\delta = \Delta_1 + \Delta_2$ being the two-photon detuning, and $2\gamma'$ and 2Γ being the spontaneous emission rates of the level $|2\rangle$ (or $|3\rangle$) and the level $|4\rangle$ respectively; $A_{21}^{(1)}$ and $A_{21}^{(3)}$ are related to χ^1-type linear susceptibility and χ^3-type nonlinear susceptibility respectively. A similar explanation is applicable to $A_{31}^{(1)}$ and $A_{31}^{(3)}$. The polarizations of the signal beam and the generated FWM light are $P_{\rm S} = Nu_{31}\rho_{31}$ and $P_{\rm F} = Nu_{21}\rho_{21}$. The evolutions of signal beam and FWM beam can be described by Maxwell's equation as follows:

$$\left(\frac{1}{c}\frac{\partial}{\partial t} + \frac{\partial}{\partial z}\right)\Omega_{\rm (F,S)} = {\rm i}\frac{u_{(21,31)}k_{(F,S)}}{2\hbar\varepsilon_0}P_{\rm (F,S)}. \tag{5}$$

We define coefficient $\xi = Nu^2K_{\rm S}/2\hbar\varepsilon_0 = Nu^2K_{\rm F}/2\hbar\varepsilon_0 = Nu^2K/2\hbar\varepsilon_0$, where $K_{\rm F}$ and $K_{\rm S}$ are the wave vectors and $K_{\rm F} = K_{\rm S}$ in our experimental system, $u_{(21)}$ and $u_{(31)}$ are dipole moments and $u = u_{21} = u_{31}$, $\omega_{\rm S}$ and $\omega_{\rm F}$ are frequencies and $\omega = \omega_{\rm S} = \omega_{\rm F}$ in our experiment, N is the active density of atoms. The signal and the generated FWM beams are co-propagating in our system. The evolutions of signal beam and FWM beam are described by the following equations:

$$\left(\frac{1}{c}\frac{\partial}{\partial t} + \frac{\partial}{\partial z}\right)\Omega_{\rm S} = {\rm i}\xi(A_{31}^{(1)}\Omega_{\rm S} + A_{31}^{(3)}\Omega_{\rm F}), \tag{6}$$

$$\left(\frac{1}{c}\frac{\partial}{\partial t} + \frac{\partial}{\partial z}\right)\Omega_{\rm F} = {\rm i}\xi(A_{21}^{(1)}\Omega_{\rm F} + A_{21}^{(3)}\Omega_{\rm S}). \tag{7}$$

We solve the above coupled equations with boundary conditions of $\Omega_{\rm S}(0) = \Omega_{\rm S0}$ and $\Omega_{\rm F}(0) = 0$, and obtain the following expressions:

$$\begin{aligned} \Omega_{\rm F}(z) &= \frac{2k_4\Omega_{S0}}{\sqrt{(k_1-k_3)^2 + 4k_2k_4}}\exp\left[\frac{1}{2}(k_1+k_3)z\right] \\ &\quad \times\sinh\left(\frac{1}{2}\sqrt{(k_1-k_3)^2 + 4k_2k_4}z\right), \end{aligned} \tag{8}$$

$$\begin{aligned} \Omega_{\rm S}(z) &= 2\Omega_{S0}\exp\left[\frac{1}{2}(k_1+k_3)z\right] \\ &\quad \times\left[\cosh\left(\frac{1}{2}\sqrt{(k_1-k_3)^2 + 4k_2k_4}z\right)\right. \\ &\quad +\frac{k_1-k_3}{\sqrt{(k_1-k_3)^2 + 4k_2k_4}} \\ &\quad \left.\times\sinh\left(\frac{1}{2}\sqrt{(k_1-k_3)^2 + 4k_2k_4}z\right)\right], \end{aligned} \tag{9}$$

where

$$\begin{aligned} k_1 &= {\rm i}\xi A_{31}^{(1)} - {\rm i}\frac{\omega}{c}, \quad k_2 = {\rm i}\xi A_{31}^{(3)}, \\ k_3 &= {\rm i}\xi A_{21}^{(1)} - {\rm i}\frac{\omega}{c}, \ \text{and} \ k_4 = {\rm i}\xi A_{21}^{(3)}. \end{aligned}$$

Equations (8) and (9) are similar to the solutions obtained in Refs. [25] and [26]. Equation (8) shows that the generated FWM beam accumulates a phase across the atoms, which is represented by the exponential term. If the two-photon detuning of δ falls in the range of Doppler band, δ would be approximate to 0 for atoms which are in resonance transition. The delay of the generated FWM beam can be calculated through

$$\frac{1}{2}\frac{{\rm d}\,{\rm Im}[(k_1+k_3)z]}{{\rm d}\omega}\bigg|_{\delta=0}.$$

It is obvious to know that the contribution to the delay of the FWM beam is from two coefficients k_1 and k_3 together. (The efficiency of FWM light is affected by coefficients k_1, k_2, k_3,

and k_4 together.) Under the condition of $\Delta_1 \gg \gamma'$, the delay of the generated FWM beam can be calculated as

$$\tau(\delta) = \frac{\mathrm{d}\,|\mathrm{Im}[(k_1+k_3)z]|}{2\mathrm{d}\omega} \approx \frac{\mathrm{d}\{\xi\,\mathrm{Re}[(\mathrm{i}/\Gamma_{21}+\mathrm{i}\Gamma_{41}/(\Omega^2+\Gamma_{21}\Gamma_{41}))]z\}}{2\mathrm{d}\delta} + \frac{z}{2c}, \quad (10)$$

where Rabi frequency $\Omega = \sqrt{|\Omega_{\mathrm{p1}}|^2 + |\Omega_{\mathrm{p2}}|^2}$, $\Gamma_{21} = \gamma' + \mathrm{i}\Delta_1$, and $\Gamma_{41} = \Gamma + \gamma' + \mathrm{i}\delta$. Detuning δ strongly affects the delay of the generated FWM light. When the parameter k_2 is small compared with k_1, k_3, k_4, equation (9) becomes $\Omega_{\mathrm{S}}(z) = 2\Omega_{\mathrm{S0}}\exp(k_1 z)$. The delay of the signal beam can be calculated as $\mathrm{d}\,|\mathrm{Im}[(k_1)z]|/\mathrm{d}\omega$, therefore it has no relation with the nonlinear susceptibility. When the δ is beyond the range of Doppler band, the above formulas are not suitable for calculating the delays of the signal and the generated FWM light.

We plot the delayed times of the generated FWM beam and the signal beam versus two-photon detuning δ in Fig. 4(a) with the consideration of the Doppler-effect. In Fig. 4(a), we can see clearly that both the generated FWM beam and the signal beam have large time delays compared with the reference at the small value of δ. The experimental data for the signal beam are in good agreement with our theoretical predictions. On the contrary, there is a little difference between the experimental data and the theoretical predictions for the generated FWM beam when the two-photon detuning δ is close to and larger than Doppler limit. There is a negative value of delay for the generated FWM beam (signal beam) when $\delta > 165$ (180) MHz in Fig. 4(a), this corresponds to the case in which δ is beyond the range of Doppler band, equation (10) cannot be used to give a theoretical prediction in this case. This causes a little difference between the theoretical plot and the experimental data.

When we eliminate the pump 2 field, the delay time of signal field can also be observed to be 25 ns as shown in Fig. 4(b). In this process, the nonlinear process does not occur, because the delay of the signal field does not mainly rely on the susceptibility $\chi^{(3)}$, but only on the susceptibility $\chi^{(1)}$ in our experimental system when k_1, k_3, and k_4 are all higher than k_2. It is easy to explain this phenomenon: in Eqs. (6) and (7), the susceptibility $\chi^{(3)}$ mainly contributes to the gain in the field propagation, and the susceptibility $\chi^{(1)}$ mainly contributes to the dispersion and loss in propagation. In the nonlinear process, the FWM field is generated due to the nonlinear susceptibility $\chi^{(3)}$, then the generated field is delayed due to the larger linear susceptibility $\chi^{(1)}$ in propagation.

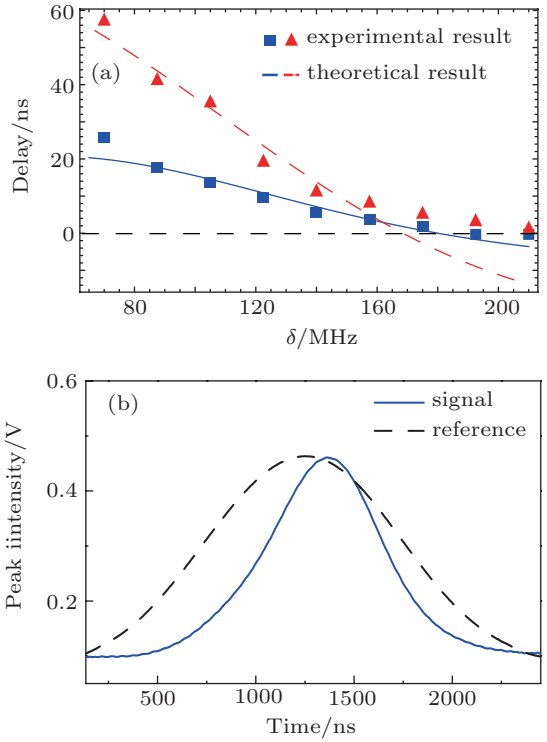


Fig. 4. (color online) (a) Delay times of the generated beam and the signal beam versus two-photon detuning δ under the conditions of $\gamma' = 3\times 2\pi\times 10^6$ s^{-1}, $\Gamma = 0.5\times 2\pi\times 10^6$ s^{-1}, $\Delta_1 = 1.96\times 2\pi\times 10^9$ s^{-1}, $\Omega_{\mathrm{p2}} = 4.0\times 2\pi\times 10^7$ s^{-1}, and $\Omega_{\mathrm{p1}} = 1.5\times 2\pi\times 10^7$ s^{-1}, $K = 4.15\times 10^6$, $N = 3.9\times 10^{18}$ m^{-3}. The solid triangles and dashed line correspond to the generated FWM beam, solid squares and solid lines correspond to the signal beam. The triangles and the squares are experimental data and the lines are theoretical fits. (b) The delay of signal beam without the pump 2 field completely.

5. Conclusion

In this paper, we perform the first experimental observation of slow FWM light in a ladder-type atomic structure, and find that the time delay can be adjusted experimentally by controlling the detuning of pump light. Such results are definitely useful for further study on the storage and the retrieval of a telecom-band light in a ladder configuration.

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