Highly efficient four-wave mixing induced by quantum constructive interference in rubidium vapour*

Wang Gang(王 刚)[a], Xue Yan(薛 艳)[a]†, Cui Cui-Li(崔淬砺)[a], Qu Yi(曲 艺)[b], and Gao Jin-Yue(高锦岳)[a]

[a] College of Physics, Jilin University, Changchun 130023, China

[b] Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, China

(Received 14 March 2011; revised manuscript received 10 August 2011)

We demonstrate efficient four-wave mixing with an intensity conversion efficiency of nearly 100% in theory without considering the Doppler-broadening effect in a four-level double-Λ system of hot ^{87}Rb gas. The corresponding experimental value of about 73% was reported in our earlier work under the same conditions. This dramatic efficiency is critically dependent on the constructive interference between two four-wave mixing processes relevant to the internally generated four-wave mixing signal.

Keywords: four-wave mixing, quantum constructive interference

PACS: 42.50.Gy, 42.65.Ky, 42.50.Hz **DOI:** 10.1088/1674-1056/21/3/034205

1. Introduction

In quantum optics, four-wave mixing (FWM) is a nonlinear process attracting broad interest and has been studied theoretically and experimentally in many atomic models with a variety of laser-coupling schemes in recent years.[1−16] Many interesting proposals regarding the applications of FWM have been carried out, such as frequency conversion,[17] quantum entanglement[18,19] and stopped light.[20] To our knowledge, most of these studies have been done with pulsed light to achieve a high conversion efficiency, and a 100% photon conversion efficiency has been reported in theory and experiment.[21,22] Whereas the research within the continuous wave (cw) field regime is relatively few with a low efficiency ($\sim 10\%$[23]) due to the limited medium length of the cold atom. In this sense, low efficiency remains a problem to be solved. The efficiency $I_{\rm f}(z=L)/I_{\rm p}(z=0)$ of about 73% has been reported experimentally in our previous work.[24] Here we give a theoretical discussion in detail and point out that the value could be as high as 100% without the consideration of the Doppler effect. This value is comparable to that observed through pulsed lasers and proves to be critically relevant to quantum constructive interference between two four-wave mixing processes. We believe that this result is significant for the nonlinear process with cw fields.

2. Theory

Consider a four-level ^{87}Rb atom system that interacts with two strong cw lasers Ω_1 and Ω_2 and a weak probe cw laser $\Omega_{\rm p}$, as depicted in Fig. 1, where δ_1, δ_2, $\delta_{\rm p}$ and $\delta_{\rm f}$ are the detunings of the input fields Ω_1, Ω_2, $\Omega_{\rm p}$, and the generated FWM field $\Omega_{\rm f}$ from the corresponding transition, respectively. In the interaction picture, the interaction Hamiltonian is expressed with the rotational wave and the electric dipole approximations as follows ($\hbar = 1$):

$$\begin{aligned} H_{\rm I} = & -\Delta\omega_1|2\rangle\langle 2| - \Delta\omega_{\rm p}|3\rangle\langle 3| - \Delta\omega_2|4\rangle\langle 4| \\ & - \Omega_{\rm p}|3\rangle\langle 1| - \Omega_1|3\rangle\langle 2| - \Omega_2|4\rangle\langle 2| \\ & - \Omega_{\rm f}|4\rangle\langle 1| + {\rm H.c.}, \end{aligned} \tag{1}$$

where $\Delta\omega_{\rm p} = \delta_{\rm p}$ is the detuning from the single-photon resonance between $|1\rangle$ and $|3\rangle$, $\Delta\omega_1 = \delta_{\rm p} - \delta_1$ is the detuning from the two-photon resonance between $|1\rangle$ and $|2\rangle$, and $\Delta\omega_2 = \delta_{\rm p} - \delta_1 + \delta_2$ is the detuning from

*Project supported by the National Natural Science Foundation of China (Grant Nos. 10904047 and 11104111), the National Basic Research Program of China (Grant No. 2011CB921603), and the Basic Research Foundation of Jilin University, China.

†Corresponding author. E-mail: xy4610@jlu.edu.cn

© 2012 Chinese Physical Society and IOP Publishing Ltd

the three-photon resonance between $|1\rangle$ and $|4\rangle$, and $\Omega_k = \mu_{ij}E_k/2\hbar$ ($k = 1, 2, \mathrm{p}, \mathrm{f}$) denote the one-half Rabi frequencies for the respective transitions, with μ_{ij} and E_k being the dipole moment and the amplitude of the field for the transition between levels $|i\rangle$ and $|j\rangle$, respectively.

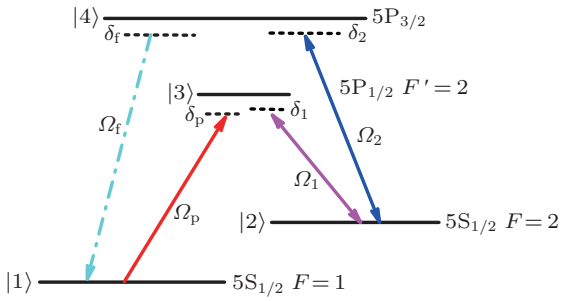


Fig. 1. (colour online) Diagram of a four-level double-Λ ^{87}Rb atom system with the relevant laser couplings.

The atomic responses of the medium to the applied fields can be expressed by the equations of motion for the probability amplitude elements as

$$\frac{\partial a_2}{\partial t} = \mathrm{i}\,\Omega_1^* a_3 + \mathrm{i}\,\Omega_2^* a_4 + \mathrm{i}\left(\Delta\omega_1 + \mathrm{i}\,\frac{\gamma_2}{2}\right)a_2, \quad (2a)$$

$$\frac{\partial a_3}{\partial t} = \mathrm{i}\,\Omega_\mathrm{p} + \mathrm{i}\,\Omega_1 a_2 + \mathrm{i}\left(\Delta\omega_\mathrm{p} + \mathrm{i}\,\frac{\gamma_3}{2}\right)a_3, \quad (2b)$$

$$\frac{\partial a_4}{\partial t} = \mathrm{i}\,\Omega_\mathrm{f} + \mathrm{i}\,\Omega_2 a_2 + \mathrm{i}\left(\Delta\omega_2 + \mathrm{i}\,\frac{\gamma_4}{2}\right)a_4, \quad (2c)$$

where the first-order weak field approximation $a_1 \simeq 1$ is assumed, which is reasonable in the experiment because the probe and the FWM fields are much weaker than the coupling and the pump fields. The probe field Ω_p and the generated FWM field Ω_f obey Maxwell's equations in the slowly varying amplitude and phase approximation, and are described by

$$\frac{\partial\Omega_\mathrm{p}}{\partial z} + \frac{1}{c}\frac{\partial\Omega_\mathrm{p}}{\partial t} = \mathrm{i}\,\kappa_{13}a_3, \quad (3a)$$

$$\frac{\partial\Omega_\mathrm{f}}{\partial z} + \frac{1}{c}\frac{\partial\Omega_\mathrm{f}}{\partial t} = \mathrm{i}\,\kappa_{14}a_4, \quad (3b)$$

where $\kappa_{13(14)} = 2\pi\omega_{\mathrm{p(f)}}N|\mu_{13(14)}|^2/(\hbar c)$, with N denoting the atomic concentration. The steady-state solution obtained from Eqs. (2a)–(2c) can be written as

$$a_2 = \frac{D_2\Omega_1^*}{\Delta}\Omega_\mathrm{p} + \frac{D_\mathrm{p}\Omega_2^*}{\Delta}\Omega_\mathrm{f}, \quad (4a)$$

$$a_3 = \frac{|\Omega_2|^2 - D_1D_2}{\Delta}\Omega_\mathrm{p} - \frac{\Omega_1\Omega_2^*}{\Delta}\Omega_\mathrm{f}, \quad (4b)$$

$$a_4 = \frac{|\Omega_1|^2 - D_1D_\mathrm{p}}{\Delta}\Omega_\mathrm{f} - \frac{\Omega_1^*\Omega_2}{\Delta}\Omega_\mathrm{p}, \quad (4c)$$

where

$$D_1 = \Delta\omega_1 + \mathrm{i}\,\gamma_2/2, \quad (5a)$$

$$D_\mathrm{p} = \Delta\omega_\mathrm{p} + \mathrm{i}\,\gamma_3/2, \quad (5b)$$

$$D_2 = \Delta\omega_2 + \mathrm{i}\,\gamma_4/2, \quad (5c)$$

$$\Delta = D_1D_2D_\mathrm{p} - D_2|\Omega_1|^2 - D_\mathrm{p}|\Omega_2|^2. \quad (5d)$$

When these equations are used in Eqs. (3a) and (3b), we can obtain

$$\frac{\partial\Omega_\mathrm{p}}{\partial z} = \mathrm{i}\,\kappa_{13}\frac{|\Omega_2|^2 - D_1D_2}{\Delta}\Omega_\mathrm{p} - \mathrm{i}\,\kappa_{13}\frac{\Omega_1\Omega_2^*}{\Delta}\Omega_\mathrm{f}, \quad (6a)$$

$$\frac{\partial\Omega_\mathrm{f}}{\partial z} = \mathrm{i}\,\kappa_{14}\frac{|\Omega_1|^2 - D_1D_\mathrm{p}}{\Delta}\Omega_\mathrm{f} - \mathrm{i}\,\kappa_{14}\frac{\Omega_1^*\Omega_2}{\Delta}\Omega_\mathrm{p}. \quad (6b)$$

For given $\Omega_\mathrm{p}(0)$ and with $\Omega_\mathrm{f}(0) = 0$, equations (6a) and (6b) can be solved analytically, yielding

$$\Omega_\mathrm{p}(z) = \frac{\Omega_\mathrm{p}(0)}{2\Lambda}\left[\left(\Lambda + \frac{K_2 - K_3}{2}\right)\mathrm{e}^{\mathrm{i}(D+\Lambda)z} + \left(\Lambda - \frac{K_2 - K_3}{2}\right)\mathrm{e}^{\mathrm{i}(D-\Lambda)z}\right], \quad (7a)$$

$$\Omega_\mathrm{f}(z) = \frac{\Omega_\mathrm{p}(0)}{2\Lambda}S_3[\mathrm{e}^{\mathrm{i}(D+\Lambda)z} - \mathrm{e}^{\mathrm{i}(D-\Lambda)z}], \quad (7b)$$

where we have defined the new parameters as

$$\Lambda = \sqrt{\left(\frac{K_2 - K_3}{2}\right)^2 + S_2S_3}, \quad (8a)$$

$$D = \frac{K_2 + K_3}{2}, \quad (8b)$$

$$K_2 = \kappa_{13}\frac{|\Omega_2|^2 - D_1D_2}{\Delta}, \quad (8c)$$

$$K_3 = \kappa_{14}\frac{|\Omega_1|^2 - D_1D_\mathrm{p}}{\Delta}, \quad (8d)$$

$$S_2 = -\kappa_{13}\frac{\Omega_1\Omega_2^*}{\Delta}, \quad (8e)$$

$$S_3 = -\kappa_{14}\frac{\Omega_1^*\Omega_2}{\Delta}. \quad (8f)$$

Consider the limiting case of $|\Omega_1|, |\Omega_2| \gg |\Delta\omega_1|, |\Delta\omega_\mathrm{p}|, \gamma_2, \gamma_3, \gamma_4$. In fact, as a result of the electromagnetically induced transparency (EIT) effect, the condition $\delta_\mathrm{p} = \delta_1 \simeq 0$ is used in our experiment to enhance the nonlinear coefficient $\chi^{(3)}$ associated with the nonlinear-optical generation process. Within this limit, and with the assumption that $|\Omega_1|^2, |\Omega_2|^2 \gg |D_1D_2|, |D_1D_\mathrm{p}|$, we can expand Λ and D as

$$\Lambda \simeq \frac{\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2}{2\Delta} - \frac{D_1\kappa_{13}\kappa_{14}}{2(\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2)}, \quad (9a)$$

$$D \simeq \frac{\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2}{2\Delta}. \quad (9b)$$

So,

$$D + \Lambda \simeq \frac{\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2}{\Delta}, \quad (10a)$$

$$D - \Lambda = \frac{D_1\kappa_{13}\kappa_{14}}{2(\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2)}. \quad (10b)$$

When equations (8) and (10) are used in Eqs. (7a) and (7b), we obtain

$$\Omega_{\rm p}(z) = \frac{\kappa_{14}|\Omega_1|^2}{\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2}\left(\Omega_{\rm p}(0)\,{\rm e}^{{\rm i}\,Qz} + \frac{\kappa_{13}|\Omega_2|^2}{\kappa_{14}|\Omega_1|^2}\Omega_{\rm p}(0)\,{\rm e}^{-{\rm i}\,Pz}\right), \quad (11a)$$

$$\Omega_{\rm f}(z) = \frac{\kappa_{14}\Omega_1^*\Omega_2}{\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2} \times (\Omega_{\rm p}(0)\,{\rm e}^{{\rm i}\,Qz} - \Omega_{\rm p}(0)\,{\rm e}^{-{\rm i}\,Pz}), \quad (11b)$$

where

$$P = \frac{\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2}{D_2|\Omega_1|^2 + D_{\rm p}|\Omega_2|^2}, \quad (12a)$$

$$Q = \frac{\Delta\omega_1\kappa_{13}\kappa_{14}}{2(\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2)}. \quad (12b)$$

In particular, in the near resonance case of δ_1, δ_2, $\delta_{\rm p} \simeq 0$, we assume that z is large enough to make $|\,{\rm e}^{-{\rm i}\,Pz}| \simeq 0$. Then equations (11a) and (11b) can be simplified as

$$\Omega_{\rm p}(z) = \frac{\kappa_{14}|\Omega_1|^2}{\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2}\Omega_{\rm p}(0), \quad (13a)$$

$$\Omega_{\rm f}(z) = \frac{\kappa_{14}\Omega_1^*\Omega_2}{\kappa_{14}|\Omega_1|^2 + \kappa_{13}|\Omega_2|^2}\Omega_{\rm p}(0). \quad (13b)$$

It is easy to derive the relationship $\Omega_{\rm f}(z)/\Omega_{\rm p}(z) = \Omega_2/\Omega_1$ from Eqs. (13a) and (13b). This relationship depicts the well known Rabi-frequency matching in the double-Λ system. And the maximum frequency conversion efficiency $\Omega_{\rm f}(z)/\Omega_{\rm p}(0) \simeq 1/2$ accords to Eqs. (13a) and (13b) with the limited condition $\kappa_{14}|\Omega_1|^2 = \kappa_{13}|\Omega_2|^2$.

Equations (11a) and (11b) indicate that there are two contributions to the growth of the FWM field $\Omega_{\rm f}$. The first contribution originates from a cw field with amplitude $\Omega_{\rm p}(0)$ and wave vector Q, whereas the second term is from another cw field with the same amplitude $\Omega_{\rm p}(0)$ and wave vector P. Since these two parts have the same frequency, the two parts will interfere with each other constructively or destructively depending on parameters Q and P. For simplicity, we assume that the coupling field Ω_1 and the probe field $\Omega_{\rm p}$ are at resonance with the respective transitions, i.e. $\Delta\omega_1 = 0, \Delta\omega_{\rm p} = 0$, and the coupling field Ω_1 and the pump field Ω_2 satisfy the condition $\kappa_{14}|\Omega_1|^2 = \kappa_{13}|\Omega_2|^2$. Thus we obtain the following expressions from Eqs. (11a) and (11b):

$$\Omega_{\rm p}(z) \simeq \frac{1}{2}\Omega_{\rm p}(0)\left(1 + {\rm e}^{-{\rm i}\,\frac{2\kappa_{14}}{D_2}z}\right), \quad (14a)$$

$$\Omega_{\rm f}(z) \simeq \frac{1}{2}\Omega_{\rm p}(0)\left(1 - {\rm e}^{-{\rm i}\,\frac{2\kappa_{14}}{D_2}z}\right). \quad (14b)$$

It is easy to identify that the probe field $\Omega_{\rm p}$ and the generated FWM field $\Omega_{\rm f}$ oscillate periodically when they propagate, as shown in Fig. 2. As usual, we can obtain the largest FWM signal when $Pz = (2n+1)\pi$ and the smallest signal when $Pz = 2n\pi$, which can be achieved by adjusting pump detuning δ_2 or distance z. When Pz is tuned from $2n\pi$ to $(2n+1)\pi$, the probe field $\Omega_{\rm p}$ is expended on the generation of the FWM field $\Omega_{\rm f}$, which forms an intuitive FWM process. The FWM field $\Omega_{\rm f}$ is absorbed and converted into the probe field $\Omega_{\rm p}$ in the range $(2n+1)\pi < Pz < (2n+2)\pi$, which is a counter intuitive FWM process. In the ideal case, the maximum frequency conversion efficiency can reach 100% due to quantum constructive interference. The quantum constructive and destructive interferences given in Eqs. (11a) and (11b) can also be proved via adjusting the pump detuning δ_2 as shown in Fig. 3. The

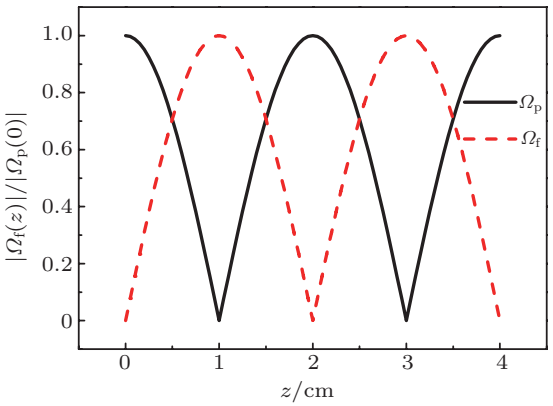


Fig. 2. (colour online) Numerical simulations of the periodical oscillations of probe and FWM fields, each as a function of distance z according to Eqs. (14a) and (14b). Here $D_2 = 2\kappa_{14}/\pi$ is assumed.

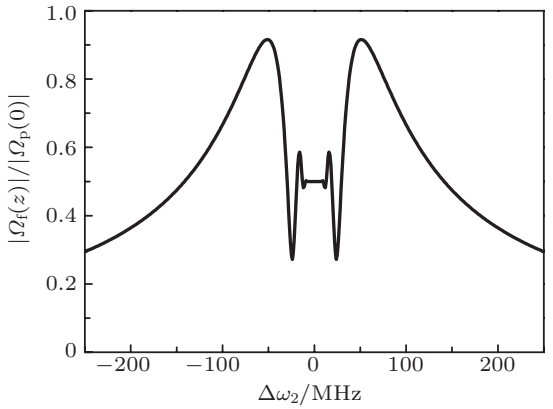


Fig. 3. Numerical simulation of the FWM field as a function of three-photon detuning $\Delta\omega_2$ according to Eqs. (14a) and (14b). Here $2\kappa_{14}z = 150$ MHz and $\gamma_4 = 6$ MHz are assumed only for simplifying this simulation.

intensity of the FWM field oscillates damply when the pump field Ω_2 is tuned from off resonance to near resonance.

Until now, it has been clear that the generation of the FWM field is due to quantum constructive interference between the two cw fields, which are denoted as the two terms on the right-hand sides of Eqs. (11a) and (11b). Furthermore, we discuss the origins of the two cw fields described in Eqs. (11a) and (11b). Now we pay more attention to Maxwell's Eqs. (6a) and (6b). The first terms on the right-hand sides of Eqs. (6a) and (6b) denote the probe (signal) absorption and dispersion, and the second terms denote the parametric gain. Equivalently, the first term represents a one-photon process $|1\rangle - |3\rangle$ for Eq. (6a) and $|1\rangle - |4\rangle$ for Eq. (6b), the second term represents a three-photon process $|1\rangle - |4\rangle - |2\rangle - |3\rangle$ for Eq. (6a) and $|1\rangle - |3\rangle - |2\rangle - |4\rangle$ for Eq. (6b), and they interfere with each other destructively or constructively as discussed by Payne and Deng,[22] and Deng and Payne.[25] So, in this investigated system, two FWM processes exist. With the pumps of the two strong fields Ω_1 and Ω_2, one process results from the conversion of the probe field Ω_p into the FWM field Ω_f, and the other one results from the conversion of the FWM field Ω_f into the probe field Ω_p, as shown in Fig. 2. Additionally, according to Eq. (6b), the FWM field Ω_f is enhanced because of the FWM process $|1\rangle - |3\rangle - |2\rangle - |4\rangle - |1\rangle$, whereas it is reduced because of the signal absorption that yields another FWM process $|1\rangle - |4\rangle - |2\rangle - |3\rangle - |1\rangle$. As a result, on the right-hand side of Eq. (11b), the first term denotes a cw field generated by the FWM process $\Omega_\mathrm{f} \rightarrow \Omega_\mathrm{p}$, and the second term denotes a cw field used to generate the FWM process $\Omega_\mathrm{p} \rightarrow \Omega_\mathrm{f}$. Thus, we can attribute the generation of the FWM field Ω_f to the quantum constructive interference between the two FWM processes $|1\rangle - |3\rangle - |2\rangle - |4\rangle - |1\rangle$ and $|1\rangle - |4\rangle - |2\rangle - |3\rangle - |1\rangle$, which is quite different from the one in classical nonlinear optics. In other words, we can also attribute the generation of the FWM field to the four-photon Rabi oscillation due to quantum interference, whereas it is a different statement about the same physical nature.

3. Conclusion

As discussed above, we theoretically studied efficient cw frequency conversion. The generated FWM field is enhanced by the quantum constructive interference between two internal FWM processes. We show that the frequency conversion efficiency is predicted to be 100% in theory. The corresponding experimental value of 73% (or 85.4% for the photon conversion efficiency) was reported in our earlier work, and is lower than the predicated value due to the Doppler effect and decoherence in the hot atom. This is an interesting study and may be useful for many potential applications.

References

[1] Fleischhauer M, Imamoglu A and Marangos J P 2005 *Rev. Mod. Phys.* **77** 633

[2] Deng L, Payne M G and Garrett W R 2006 *Phys. Rep.* **429** 123

[3] Deng L and Payne M G 2003 *Phys. Rev. Lett.* **91** 243902

[4] Kang H, Hernandez G and Zhu Y F 2004 *Phys. Rev. Lett.* **93** 073601

[5] Braje D A, Balić V, Goda S, Yin G Y and Harris S E 2004 *Phys. Rev. Lett.* **93** 183601

[6] Deng L, Kozuma M, Hagley E W and Payne M G 2002 *Phys. Rev. Lett.* **88** 143902

[7] Lu B, Burkett W H and Xiao M 1998 *Opt. Lett.* **23** 804

[8] Lukin M D, Matsko A B, Fleischhauer M and Scully M O 1999 *Phys. Rev. Lett.* **82** 1847

[9] Zibrov A S, Lukin M D and Scully M O 1999 *Phys. Rev. Lett.* **83** 4049

[10] Zibrov A S, Matsko A B, Kocharovskaya O, Rostovtsev Y V, Welch G R and Scully M O 2002 *Phys. Rev. Lett.* **88** 103601

[11] Jiang K J, Deng L and Payne M G 2007 *Phys. Rev. Lett.* **98** 083604

[12] Boyer V, McCormick C F, Arimondo E and Lett P D 2007 *Phys. Rev. Lett.* **99** 143601

[13] Zhang Y, Brown A W and Xiao M 2007 *Phys. Rev. Lett.* **99** 123603

[14] Zuo Z C, Sun J, Liu X, Mi X, Yu Z H, Jiang Q, Fu P M and Wu L A 2007 *Chin. Phys.* **16** 1042

[15] Li Y Y, Li L, Zhang Y P and Bi S W 2010 *Chin. Phys.* B **19** 090702

[16] Xue Y, Wang G, Wu J H, Zhang B and Gao J Y 2011 *Chin. Phys.* B **20** 033403

[17] Jain M, Xia H, Yin G Y, Merrian A J and Harris S E 1996 *Phys. Rev. Lett.* **77** 4326

[18] Payne M G and Deng L 2003 *Phys. Rev. Lett.* **91** 123602

[19] Yuan S H and Hu X M 2010 *Chin. Phys.* B **19** 074210

[20] Camacho R M, Vudyasetu P K and Howell J C 2009 *Nature Photonics* **3** 103

[21] Merriam A J, Sharpe S J, Shverdin M, Manuszak D, Yin G Y and Harris S E 2000 *Phys. Rev. Lett.* **84** 5308

[22] Payne M G and Deng L 2002 *Phys. Rev.* A **65** 063806

[23] Kang H, Hernandez G and Zhu Y F 2005 *J. Mod. Opt.* **52** 2391

[24] Wang G, Xue Y, Wu J H, Kang Z H, Jiang Y, Liu S S and Gao J Y 2010 *Opt. Lett.* **35** 3778

[25] Deng L and Payne M G 2003 *Phys. Rev.* A **68** 051801(R)