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Coherent enhancement of resonance-mediated multiphoton absorption

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Abstract

In this paper, we theoretically investigate the coherent enhancement of resonance-mediated (2+2) four-photon absorption. It is found that by shaping the spectral phase with a π phase step, the resonance-mediated (2+2) four-photon transition probability can be enhanced. Furthermore, the coherent enhancement dependences on the detuning between the two two-photon absorptions, laser spectral bandwidth and laser centre frequency are explicitly discussed and analysed. We believe these theoretical results may play an important role in enhancing more complex resonance-mediated multiphoton absorption processes.

1. Introduction

The multiphoton absorption processes have attracted widespread interest because of their potential applications in biology, biophotonics and medicine by fluorescence spectroscopy, three-dimensional fluorescence imaging or photodynamic therapy [1–4]. Recently, the coherent control of the multiphoton absorption processes making use of the shaped femtosecond laser pulse has been widely studied in atomic and molecular systems, involving non-resonant two-photon absorption [5–8], non-resonant three-photon absorption [9, 10], resonance-mediated (1+1) two-photon absorption [11–13] and resonance-mediated (2+1) three-photon absorption [14–16]. In the weak-field regime, for non-resonant multiphoton absorption processes (i.e. a transition without the intermediate states), the transition probability is maximal with the transform-limited (TL) pulse, and can be reduced and even completely eliminated by manipulating the spectral phase, but cannot be enhanced [6]. However, for resonance-mediated multiphoton absorption processes (i.e. a transition with the intermediate states), the transition probability can be enhanced by properly designing the spectral phase distribution [11, 14–16].

Previous studies were on the resonance-mediated (1+1) two-photon absorption or (2+1) three-photon absorption, which is a process with the coherent combination of the linear and linear absorption or the nonlinear and linear absorption. As an extension, in this paper we study the resonance-mediated multiphoton absorption processes with the coherent combination of the nonlinear and nonlinear absorption. Studying the model system is important in the control of photo-induced dynamics, where dynamical processes can be understood on the basis of theoretical evaluation, and thus larger or more complex quantum systems with more freedom degrees may become controllable and understandable. So, here we consider the simplest case of a resonance-mediated (2+2) four-photon absorption process. We theoretically find that by designing the spectral phase distribution with a π phase step, the resonance-mediated (2+2) four-photon transition probability can be enhanced. Moreover, we also explicitly discuss the influences of the detuning between the two two-photon absorptions, the laser spectral bandwidth and the laser centre frequency on the coherent enhancement.

2. Theoretical model

Figure 1 schematically shows the resonance-mediated (2+2) four-photon absorption process, where $|g\rangle$, $|r\rangle$ and $|f\rangle$ represent the ground state, the intermediate excited state and the final excited state, respectively. The transitions for $|g\rangle \rightarrow |r\rangle$ and $|r\rangle \rightarrow |f\rangle$ are both coupled by a laser field $E_p(t)$ with non-resonant two-photon excitation. In the weak laser field, the resonance-mediated (2+2) four-photon transition amplitude can be approximated by fourth-order perturbation theory as [11, 16]

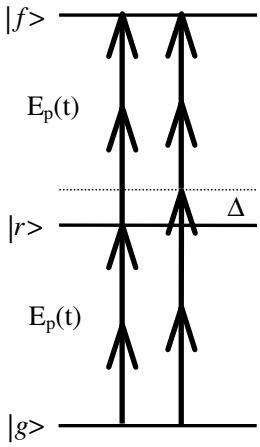

Figure 1. Schematic diagram of the resonance-mediated (2+2) four-photon absorption process. Transitions for $|g\rangle \rightarrow |r\rangle$ and $|r\rangle \rightarrow |f\rangle$ are both coupled by the laser field $E_p(t)$ with non-resonant two-photon excitation.

$$A_f^{(2+2)} = A_f^{(2+2)\mathrm{on-res}} + A_f^{(2+2)\mathrm{near-res}} \tag{1}$$

with

$$A_f^{(2+2)\mathrm{on-res}} = \frac{i\pi}{\hbar^4}\mu_{rg}^2\mu_{fr}^2 A^{(2)}(\omega_{rg})A^{(2)}(\omega_{fr}) \tag{2}$$

$$A_f^{(2+2)\mathrm{near-res}} = -\frac{\rho}{\hbar^4}\mu_{rg}^2\mu_{fr}^2 \times \int_{-\infty}^{\infty}\frac{1}{\Delta}A^{(2)}(\omega_{rg}-\Delta)A^{(2)}(\omega_{fr}+\Delta)d\Delta \tag{3}$$

where $A^{(2)}(\Omega)$ is the non-resonant two-photon transition amplitude with $A^{(2)}(\Omega) = \int_{-\infty}^{\infty} E(\Omega-\omega)E(\omega)\,\mathrm{d}\omega$, ρ is the Cauchy principal value, and μ_{rg}^2 and μ_{fr}^2 are effective non-resonant two-photon dipole coupling for the transitions $|g\rangle \rightarrow |r\rangle$ and $|r\rangle \rightarrow |f\rangle$, respectively. The term $A_f^{(2+2)\mathrm{on-res}}$ is the resonant contribution, which interferes all the on-resonant four-photon excitation pathways (i.e. $\Delta = 0$). The term $A_f^{(2+2)\mathrm{near-res}}$ is the non-resonant contribution, which interferes all the near-resonant four-photon excitation pathways (i.e. $\Delta \neq 0$) by the weighting factor $1/\Delta$. So, the total transition amplitude $A_f^{(2+2)}$ depends on the intra- and intergroup interferences involving on- and near-resonant four-photon excitation pathways. The resonance-mediated (2+2) four-photon transition probability is given by $P_f^{(2+2)} = \left|A_f^{(2+2)}\right|^2$, and the on- and near-resonant contributions can be written as $P_f^{(2+2)\mathrm{on-res}} = \left|A_f^{(2+2)\mathrm{on-res}}\right|^2$ and $P_f^{(2+2)\mathrm{near-res}} = \left|A_f^{(2+2)\mathrm{near-res}}\right|^2$, respectively.

Silberberg *et al* previously showed that by manipulating the laser spectral phase, the non-resonant two-photon transition amplitude $A^{(2)}(\Omega)$ in the weak-field regime can be reduced, but cannot be enhanced [6]. Thus, it is easy to verify that $P_f^{(2+2)\mathrm{on-res}}$ is maximal for the TL pulse, and the other phase distribution can reduce and even eliminate it. However, $P_f^{(2+2)\mathrm{near-res}}$ integrates over both negative and positive contributions, so the TL pulse induces destructive interference among the four-photon excitation pathways; thus it can be enhanced by properly designing the laser pulse that induces constructive interference instead of destructive interference. If $P_f^{(2+2)\mathrm{near-res}}$ is much larger than $P_f^{(2+2)\mathrm{on-res}}$, $P_f^{(2+2)}$ can be enhanced.

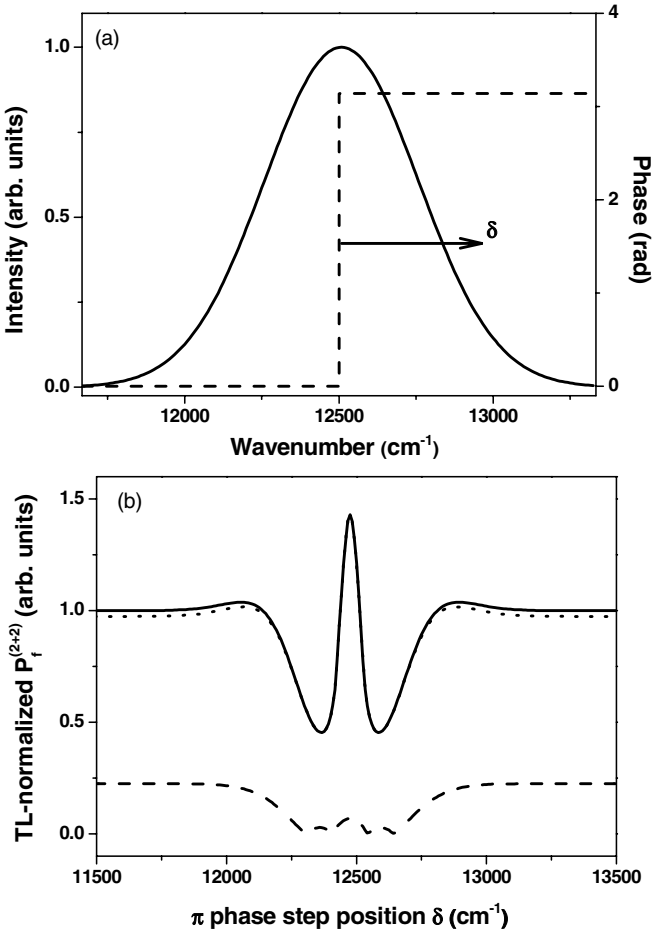

Figure 2. (a) Schematic diagram of a π phase step modulation applied on the laser spectrum. (b) The TL-normalized $P_f^{(2+2)}$ (solid line) as a function of the π phase step position δ, together with $P_f^{(2+2)\mathrm{on-res}}$ (dashed line) and $P_f^{(2+2)\mathrm{near-res}}$ (dotted line).

3. Results and discussion

The shaped pulse with a π phase step is a well-established method in quantum control because it is convenient to obtain the antisymmetric phase distribution or the controllable two-subpulse sequences, and therefore has been widely employed to manipulate various multiphoton absorption processes [5, 6, 10, 14–16]. In this paper, we also utilize the π phase step to enhance the resonance-mediated (2+2) four-photon transition probability $P_f^{(2+2)}$, and this simple spectral phase modulation is presented in figure 2(a). In our simulation, these parameters of the quantum system are set as follows. The laser pulse has a Gaussian profile with the centre frequency ω_0 of 12 500 cm^{-1} and the spectral bandwidth Δ_p (a full width at half maximum (FWHM)) of 600 cm^{-1}. The transition frequencies for $|g\rangle \rightarrow |r\rangle$ and $|r\rangle \rightarrow |f\rangle$ are 25 050 cm^{-1} and 24 850 cm^{-1}, i.e. $\omega_{rg} = 250\,50\ \mathrm{cm}^{-1}$ and $\omega_{fr} = 24\,850\ \mathrm{cm}^{-1}$, respectively. Figure 2(b) shows the resonance-mediated (2+2) four-photon

transition probability $P_f^{(2+2)}$ as a function of the π phase step position δ, together with the on-resonant component $P_f^{(2+2)\mathrm{on-res}}$ (dashed line) and the near-resonant component $P_f^{(2+2)\mathrm{near-res}}$ (dotted line). All data are normalized by $P_f^{(2+2)}$ induced by the TL pulse (hereafter the same method is used). As can be seen, $P_f^{(2+2)\mathrm{on-res}}$ cannot be enhanced, while the $P_f^{(2+2)\mathrm{near-res}}$ can be enhanced and achieves maximal enhancement at the π phase step position of 12 475 cm^{-1}, which is equal to $(\omega_{rg}+\omega_{fr})/4$. Since $P_f^{(2+2)\mathrm{near-res}}$ is much larger than $P_f^{(2+2)\mathrm{on-res}}$, $P_f^{(2+2)}$ is also maximally enhanced at the π phase step position of $(\omega_{rg}+\omega_{fr})/4$.

The π phase step modulation provides a very effective way to enhance the resonance-mediated (2+2) four-photon absorption, as shown in figure 2(b). Now we study the influence of the detuning between the two two-photon absorptions Δ_d (i.e. $\Delta_\mathrm{d} = \omega_{fr} - \omega_{rg}$) on the coherent enhancement of $P_f^{(2+2)}$, here we fix ω_{rg} while scan ω_{fr}. Figure 3(a) shows the TL-normalized $P_f^{(2+2)}$ at the π phase step position of $(\omega_{rg}+\omega_{fr})/4$ as a function of Δ_d. With the increase of Δ_d, the coherent enhancement decreases, which is due to the decrease of the enhancement of $P_f^{(2+2)\mathrm{near-res}}$. However, it is noted that the coherent enhancement also decreases when Δ_d is close to zero. To demonstrate the physical origin, we calculate $P_f^{(2+2)}$, $P_f^{(2+2)\mathrm{on-res}}$ and $P_f^{(2+2)\mathrm{near-res}}$ as a function of the π phase step position δ for $\Delta_\mathrm{d} = 10$ cm^{-1}, and the calculated results are presented in figure 3(b). It can be found that $P_f^{(2+2)\mathrm{on-res}}$ is much larger that $P_f^{(2+2)\mathrm{near-res}}$. Thus, the above-mentioned phenomenon can be intuitively understood as follows. With the decrease of Δ_d, $P_f^{(2+2)\mathrm{on-res}}$ increases but $P_f^{(2+2)\mathrm{near-res}}$ decreases. When Δ_d approaches zero, $P_f^{(2+2)\mathrm{on-res}}$ is much larger than $P_f^{(2+2)\mathrm{near-res}}$. Since $P_f^{(2+2)\mathrm{on-res}}$ cannot be enhanced, the coherent enhancement of $P_f^{(2+2)}$ will decrease.

Next we demonstrate the dependence of the coherent enhancement of $P_f^{(2+2)}$ on the laser spectral bandwidth Δ_p. Figure 4 shows the TL-normalized $P_f^{(2+2)}$ at the π phase step position of $(\omega_{rg}+\omega_{fr})/4$ as a function of Δ_p for $\Delta_\mathrm{d} = 50$ cm^{-1}. As can be seen, the coherent enhancement increases when the Δ_p increases, while it will decrease if Δ_p is far larger than Δ_d. With the increase of Δ_p, $P_f^{(2+2)\mathrm{on-res}}$ increases but $P_f^{(2+2)\mathrm{near-res}}$ decreases, and so the decrease of the coherent enhancement in the broad-band laser spectrum is also attributed to $P_f^{(2+2)\mathrm{on-res}}$ being much larger than $P_f^{(2+2)\mathrm{near-res}}$.

By varying the detuning between the two two-photon absorptions Δ_d or the laser spectral bandwidth Δ_p, the coherent enhancement of $P_f^{(2+2)}$ can be effectively manipulated, as shown in figures 3(a) and 4. Actually, the coherent enhancement should depend on the ratio of Δ_p to Δ_d (i.e. Δ_p/Δ_d). If Δ_p/Δ_d is very small, $P_f^{(2+2)\mathrm{near-res}}$ is much larger than $P_f^{(2+2)\mathrm{on-res}}$, but the enhancement of $P_f^{(2+2)\mathrm{near-res}}$ decreases, and therefore the enhancement of $P_f^{(2+2)}$ will decrease. However, when Δ_p/Δ_d is very large, although the enhancement of $P_f^{(2+2)\mathrm{near-res}}$ increases, $P_f^{(2+2)\mathrm{on-res}}$ is much larger than $P_f^{(2+2)\mathrm{near-res}}$, and thus the enhancement of $P_f^{(2+2)}$

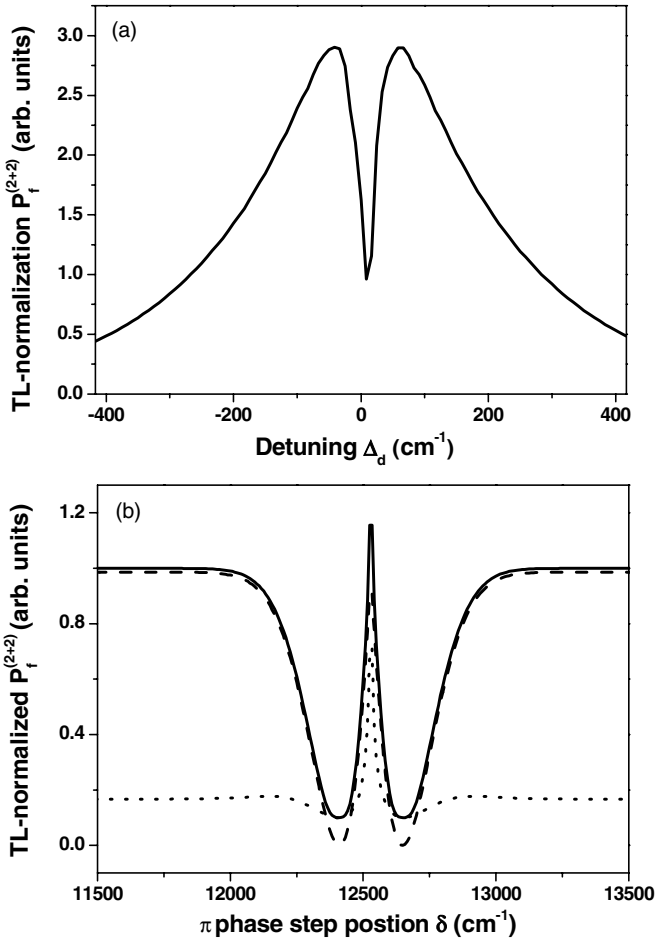

Figure 3. (a) The TL-normalized $P_f^{(2+2)}$ at the π phase step position of $(\omega_{rg}+\omega_{fr})/4$ as a function of the detuning between the two two-photon absorptions Δ_d. (b) The TL-normalized $P_f^{(2+2)}$ (solid line) as a function of the π phase step position δ for the detuning $\Delta_\mathrm{d} = 10$ cm^{-1}, together with $P_f^{(2+2)\mathrm{on-res}}$ (dashed line) and $P_f^{(2+2)\mathrm{near-res}}$ (dotted line).

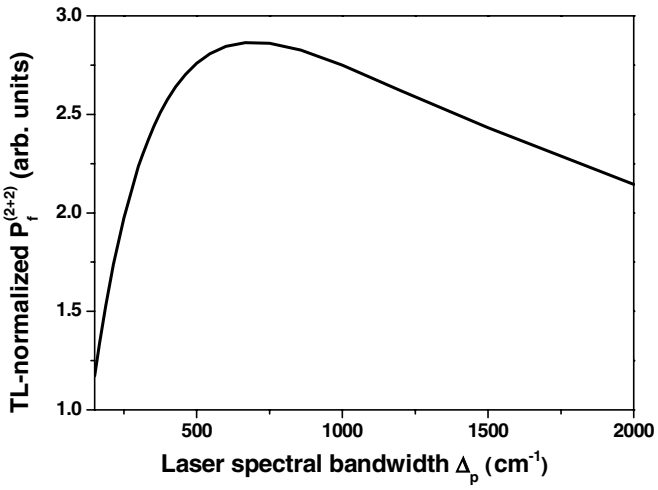

Figure 4. The TL-normalized $P_f^{(2+2)}$ at the π phase step position of $(\omega_{rg}+\omega_{fr})/4$ as a function of the laser spectral bandwidth Δ_p for the detuning $\Delta_\mathrm{d} = 50$ cm^{-1}.

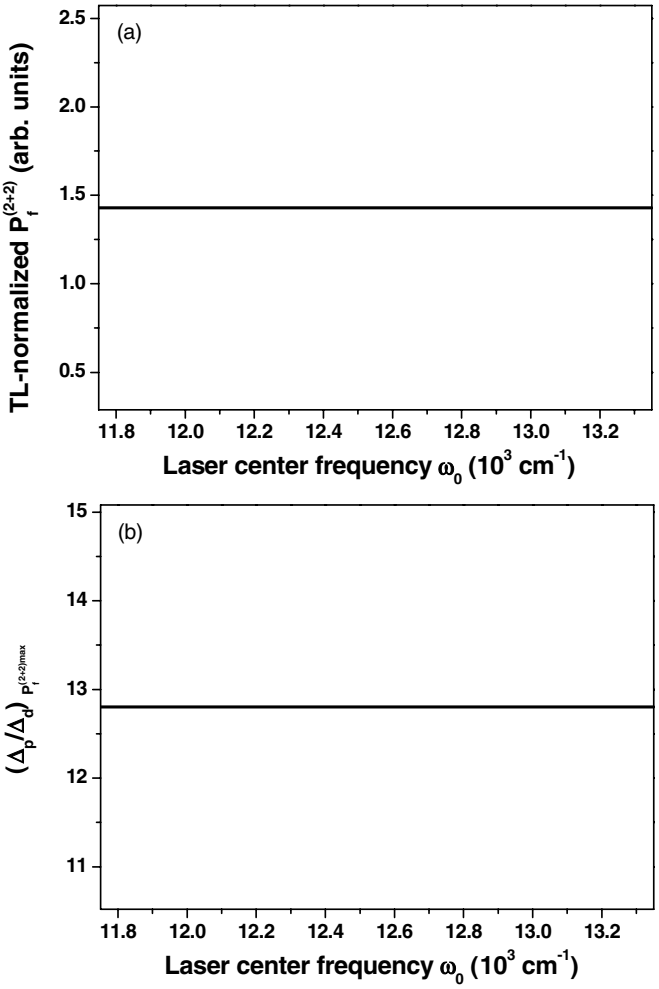

Figure 5. The TL-normalized $P_f^{(2+2)}$ at the π phase step position of $(\omega_{rg}+\omega_{fr})/4$ (a) and Δ_p/Δ_d for the maximal enhancement of $P_f^{(2+2)}$ (i.e. $(\Delta_p/\Delta_d)_{P_f^{(2+2)\max}}$) (b) as a function of the laser centre frequency ω_0.

will also decrease. Accordingly, it is crucial to select a correct Δ_p/Δ_d to achieve the maximal enhancement of $P_f^{(2+2)}$. In our simulation, the coherent enhancement of $P_f^{(2+2)}$ is maximal with Δ_p/Δ_d of about 12.8 (see figures 3(a) and 4).

The laser centre frequency ω_0 is an important parameter in studying the control of photo-induced atomic dynamics. Finally, we investigate the effect of ω_0 on the coherent enhancement of $P_f^{(2+2)}$ and the ratio Δ_p/Δ_d for the maximal enhancement of $P_f^{(2+2)}$ (i.e. $(\Delta_p/\Delta_d)_{P_f^{(2+2)\max}}$). Figure 5 shows the TL-normalized $P_f^{(2+2)}$ at the π phase step position of $(\omega_{rg}+\omega_{fr})/4$ (a) and $(\Delta_p/\Delta_d)_{P_f^{(2+2)\max}}$ (b) as a function of ω_0. It can be seen that with the increase of ω_0, both $P_f^{(2+2)}$ and $(\Delta_p/\Delta_d)_{P_f^{(2+2)\max}}$ remain constant. These results indicate that ω_0 does not affect the coherent enhancement of $P_f^{(2+2)}$ and $(\Delta_p/\Delta_d)_{P_f^{(2+2)\max}}$. Therefore, we can conclude that ω_0 can change $P_f^{(2+2)}$ but not affect its enhancement.

4. Conclusion

In summary, we have demonstrated that the resonance-mediated (2+2) four-photon absorption can be enhanced by shaping the laser pulse with the π phase step. Our theoretical results show that the coherent enhancement is achieved by the intra- and intergroup interferences involving the on- and near-resonant four-photon excitation pathways, which is independent of the laser centre frequency; while it greatly depends on the ratio of the laser spectral bandwidth to the detuning between the two two-photon absorptions, both very small and large ratio may lead to the decrease of the coherent enhancement. If more complex resonance-mediated multiphoton absorption processes are involved, our scheme can be iteratively applied once the excited states are identified. By correctly selecting the laser wavelength and spectral bandwidth, the coherent enhancement can be obtained.

Acknowledgments

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