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ORIGINAL PAPER

Intermediate state absorption enhancement in resonance-mediated (2+1) three-photon excitation process

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Abstract: In this paper, we theoretically study the control of intermediate state absorption in resonance-mediated (2+1) three-photon excitation process by shaping the femtosecond pulse with a π phase step modulation under weak laser field. Our results show that the intermediate state absorption can be enhanced, and the coherent enhancement increases with the increase of the pulse intensity and pulse duration. Our analysis indicates that the absorption enhancement results from the absorption reduction of the final state from the intermediate state in the shaped laser field. Furthermore, the effects of the population difference of the final and intermediate states in the unshaped laser field and the transition dipole moment from the intermediate state to the final state on the absorption enhancement are discussed and analyzed.

Keywords: Multi-photon absorption; Pulse shaping; Coherent control

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1. Introduction

With the development of the femtosecond pulse shaping technique, it is now possible to obtain such a pulse shape with an almost arbitrary temporal distribution by controlling the spectral phase or/and amplitude in the frequency domain [1]. Nowadays, the coherent control strategies by shaping the femtosecond pulse have been successfully applied for the control of various nonlinear optical processes [2–5]. The quantum control is dominant by the interference among different optical pathways connecting the initial and final states, and therefore the main challenge is how to manipulate the constructive or destructive interference among these different pathways by precisely controlling the pulse shape. The pre-designed pulse with a simple spectral phase or/and amplitude pattern is usually employed to control the simple model systems [6–18], which is useful for better understanding of the underlying physical mechanisms and further development of coherent control techniques and concepts. For those large and complex quantum systems, the feedback control strategies based on a learning algorithm have proven to be an efficient method to automatically optimize the laser field to achieve desired outcomes [19–28].

Recently, the femtosecond pulse shaping technique has been widely utilized to control the multiphoton absorption processes, such as non-resonant two-photon absorption [6–9], non-resonant three-photon absorption [10, 11], resonance-mediated two-photon absorption [12–14], resonance-mediate (2+1) three-photon absorption [15–17], and stimulated Raman process [18]. In these previous studies, the control of the final state was primary concern. However, here we focus on the control of the intermediate state. In actual experiments, the control of the target state is usually affected by its higher state, so it is necessary to study the control of the intermediate state. In this paper, we theoretically study the intermediate state absorption control in the resonance-mediated (2+1) three-photon excitation process under weak laser field. We show that the intermediate state absorption can be enhanced by shaping the femtosecond pulse with a π phase step modulation, and the coherent enhancement increases with the increase of the pulse intensity and pulse duration. It is indicated that the absorption reduction of the final state from the intermediate state in the shaped laser field is the essential contribution of the absorption enhancement. Finally, we discuss the effects of the population difference of the final and intermediate states

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*Corresponding author, E-mail: zrsun@phy.ecnu.edu.cn in the unshaped laser field and the transition dipole moment from the intermediate state to the final state on the absorption enhancement.

2. Theoretical model

We consider the interaction of a weak laser field E(t) with a three level system, as shown in Fig. 1(a). Here, |g>, |i> and |f> are the ground, intermediate and final states, respectively. |g> → |i> is coupled by the laser field E(t) with non-resonant two-photon excitation, and |i> → |f> is coupled via one-photon excitation. We assume that the pulse duration is much shorter than the lifetime of the excited states (i.e., the intermediate and final states), and the population is initially in the ground state |g>. The time-dependent probability amplitudes $C_n(t)$ in each state can be obtained by solving the time-dependent Schrödinger equation and given by [29]

$$i\hbar\frac{\partial C_n(t)}{\partial t}=H(t)C_n(t), \tag{1}$$

where $H(t)$ is the Hamiltonian of the three-level system interacting with the laser field $E(t)$. As shown in Fig. 1(a), the ground state |g> is excited to the intermediate state |i> via the state |m> that is off resonance. Thus, the time-dependent Hamiltonian can be written as

$$H(t)=\hbar\begin{bmatrix} E_g & \mu_{mg}E(t) & 0 & 0\\ \mu_{mg}E(t) & E_m & \mu_{im}E(t) & 0\\ 0 & \mu_{im}E(t) & E_i & \mu_{fi}E(t)\\ 0 & 0 & \mu_{fi}E(t) & E_f \end{bmatrix}, \tag{2}$$

where E_g, E_m, E_i and E_f are the eigenvalues of the states |g>, |m>, |i> and |f>, and μ_{mg}, μ_{im} and μ_{fi} are the transition dipole moments of the transitions |g> → |m>, |m> → |i> and |i> → |f>, respectively. Finally, the population in each state after the laser field P_n is given by the absolute squares of $C_n(t \to \infty)$ as follows

$$P_n=|C_n(t\to\infty)|^2. \tag{3}$$

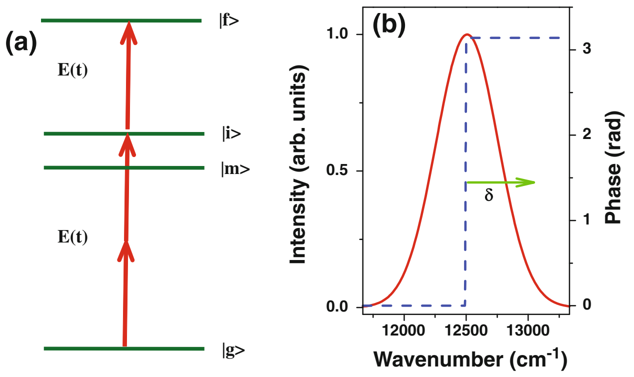

Fig. 1 (Color online) **a** The schematic diagram of the resonance-mediated (2+1) three-photon excitation. Here, |g> → |i> is coupled by the laser field E(t) with non-resonant two-photon excitation, and |i> → |f> is coupled via one-photon excitation. **b** The schematic diagram of the π phase step modulation applied on the laser spectrum

Multiphoton absorption process can be manipulated by shaping the femtosecond pulse. A π phase step modulation has shown to be an efficient method to suppress or enhance the multiphoton absorption, such as non-resonant two-photon absorption [7], non-resonant three-photon absorption [11], and resonance-mediated (2+1) three-photon absorption [15–17]. Here, we employ the π phase step modulation to control the intermediate state absorption in resonance-mediated (2+1) three-photon excitation process. Fig. 1b shows the schematic diagram of the π phase step modulation applied on the laser spectrum. The π phase step modulation can be defined by the function of $\Phi(\omega)=\pi\delta(\omega-\omega_{\mathrm{step}})/2$, where $\delta(\omega-\omega_{\mathrm{step}})$ denotes the signum function which takes the values of -1 and $+1$ for $\omega\le\omega_{\mathrm{step}}$ and $\omega\ge\omega_{\mathrm{step}}$, and thus $\Phi(\omega)$ is characterized by a phase jump from $-\pi/2$ to $\pi/2$ at the step position ω_{step}. Thus, the modulated laser field in frequency domain $E_{\mathrm{mod}}(\omega)$ is given by $E_{\mathrm{mod}}(\omega)=E(\omega)\times\exp[i\pi\delta(\omega-\omega_{\mathrm{step}})/2]$, where $E(\omega)$ is the Fourier transform of the unmodulated laser field E(t), and the modulated laser field in time domain $E_{\mathrm{mod}}(t)$ is given by the convolution of the unmodulated laser field E(t) with $\exp[i\omega_{\mathrm{step}}t)/(\pi t)$, i.e., $E_{\mathrm{mod}}(t)=E(t)\otimes\exp[i\omega_{\mathrm{step}}t)/(\pi t)$. By substituting $E_{\mathrm{mod}}(t)$ into Eqs. (1)–(3), the population in each state P_n induced by the π phase step modulation is obtained.

3. Results and discussion

In our simulation, the time-dependent Schrödinger equation in Eq. (1) is numerically solved by Runge–Kutta method. The parameters of the quantum system in Fig. 1(a) are set as follows. The transition frequencies for |g> → |m>, |m> → |i> and |i> → |f> (i.e., ω_{mg}, ω_{im}, and ω_{fi}) are 17000, 8100, and 12400 cm^{-1} respectively. Thus the transition frequency of the two-photon absorption |g> → |i> (i.e., ω_{ig}) is 25,100 cm^{-1}. We assume that the ratio of the transition dipole moments μ_{mg}, μ_{im} and μ_{fi} is 1:1:1.5. The central frequency of the laser field is set to be $\omega_0=\omega_{\mathrm{fg}}/3=$ 12,500 cm^{-1}, where ω_{fg} is the transition frequency from the ground state |g> to the final state |f> with $\omega_{\mathrm{fg}}=\omega_{\mathrm{ig}}+\omega_{\mathrm{fi}}=$ 37,500 cm^{-1}.

Figure 2 shows the population of the intermediate state |i> P_i as the function of the π phase step position ω_{step} with different pulse intensities (left-hand-column panel) and pulse durations (right-hand-column panel). Left-hand-column panel shows the TL-normalized P_i with the pulse duration of 100 fs for the pulse intensity of 2×10^9 (a − 1),

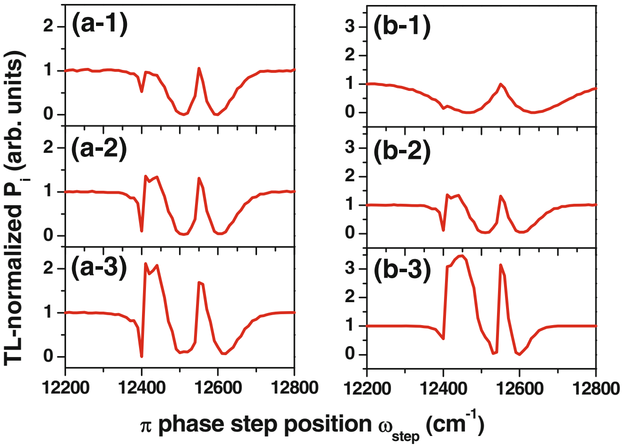

Fig. 2 (Color online) The TL-normalized population of the intermediate state |i> p_i in the resonance-mediated (2+1) three-photon excitation process as the function of the π phase step position ω_{step} with different pulse intensities (*left-hand-column panel*) and the pulse durations (*right-hand-column panel*). *Left-hand-column panel* shows the TL-normalized P_i with the pulse duration of 100 fs for the pulse intensity of 2 × 10^9 (*a-1*), 5 × 10^9 (*a-2*) and 8 × 10^9 W/cm^2 (*a-3*), and right-hand-column panel shows the TL-normalized P_i with the pulse intensity of 5 × 10^9 W/cm^2 for the pulse duration of 50 (*b-1*), 100 (*b-2*) and 150 fs (*b-3*)

5 × 10^9 (a − 2) and 8 × 10^9 W/cm^2 (a − 3), and right-hand-column panel shows the TL-normalized P_i with the pulse intensity of 5 × 10^9 W/cm^2 for the pulse duration of 50 (b − 1), 100 (b − 2) and 150 fs (b − 3). All traces are normalized by P_i induced by the transform-limited (TL) pulse. As can be seen, P_i can increase and also decrease to zero by the simple spectral phase modulation. That is to say, the intermediate state absorption in the resonance-mediated (2+1) three-photon excitation process can be enhanced and also completely suppressed. The enhancement greatly depends on the pulse intensity and pulse duration, which increases with the increase of the pulse intensity and pulse duration. However, the suppression is independent of the pulse intensity and pulse duration.

To demonstrate that the intermediate state absorption enhancement in the resonance-mediated (2+1) three-photon excitation process is not due to the higher nonlinear optical effect, we consider the case of the two-photon absorption from the ground state |g> to the intermediate state |i> without the final state |f>, and the calculated result is shown in Fig. 3 with the pulse intensity of 8 × 10^9 W/cm^2 and the pulse duration of 100 fs. One can see that no absorption enhancement is observed, and the phenomenon is the same as that obtained by the perturbation theory under weak laser field [7]. Therefore, it can be concluded that the higher nonlinear optical process does not occur in the range of the pulse intensity that is shown in Fig. 2, and its effect can be excluded.

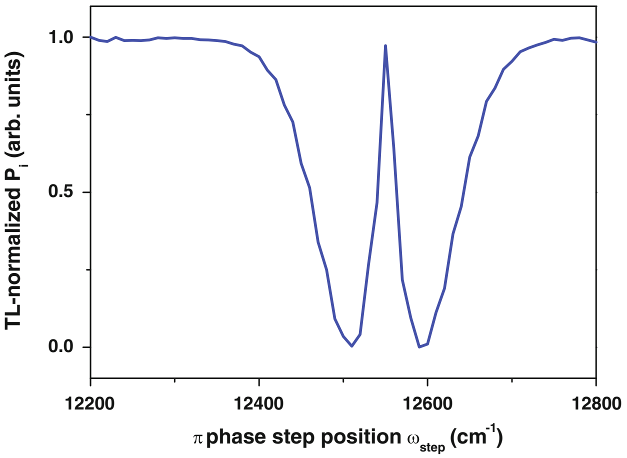

Fig. 3 (Color online) The TL-normalized population of the intermediate state |i> P_i without the final state |f> as the function of the π phase step position ω_{step} with the pulse intensity of 8 × 10^9 W/cm^2 and the pulse duration of 100 fs

We present the population of the ground state |g> P_g (blue dotted line), the intermediate state |i> P_i (red solid line) and the final state |f> P_f (green dashed line) as the function of the π phase step position ω_{step} with the pulse intensity of 5 × 10^9 W/cm^2 and the pulse duration of 100 fs, as shown in Fig. 4. As can be seen, P_f tremendously decreases in those positions that P_i increases, but P_g does not. Consequently, we can conclude that the increase of P_i should be attributed to the decrease of P_f. That is to say, the absorption reduction of the final state |f> from the intermediate state |i> in the shaped laser field leads to the absorption enhancement of the intermediate state |i>.

Since the absorption of the final state |f> from the intermediate state |i> affects the control of the intermediate state |i>, next we study the effect of the population difference of the final state |f> P_f and the intermediate state

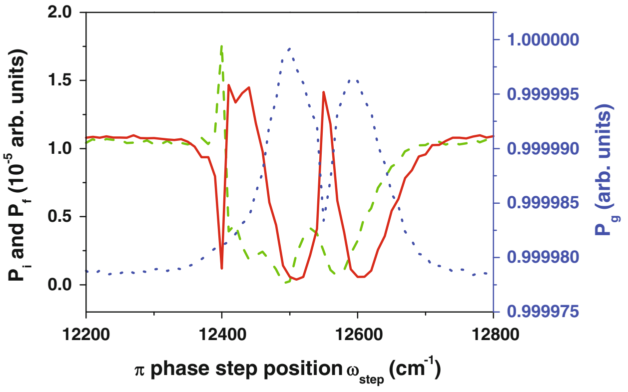

Fig. 4 (Color online) The population of the ground state |g> P_g (*blue dotted line*), the intermediate state |i> P_i (*red solid line*), and the final state |f> P_f (*green dashed line*) as the function of the π phase step position ω_{step} with the pulse intensity of 5 × 10^9 W/cm^2 and the pulse duration of 100 fs

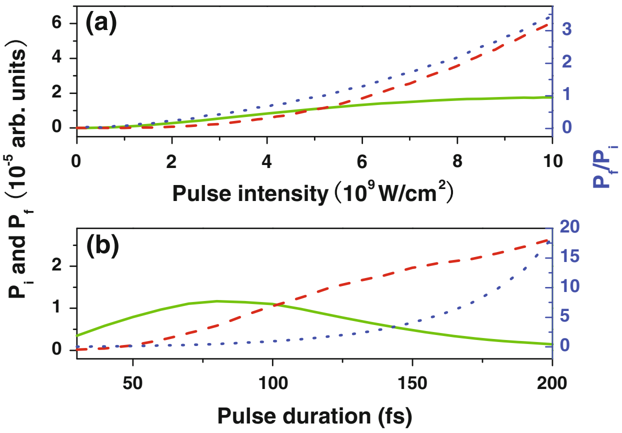

Fig. 5 (Color online) The population of the intermediate state |i> P_i (*green solid lines*) and final state |f> P_f (*red dashed lines*) in the unshaped laser field as the function of the pulse intensity for the pulse duration of 100 fs (**a**) and the pulse duration for the pulse intensity of 5×10^9 W/cm^2 (**b**), together with their ratio P_f/P_i (*blue dotted lines*)

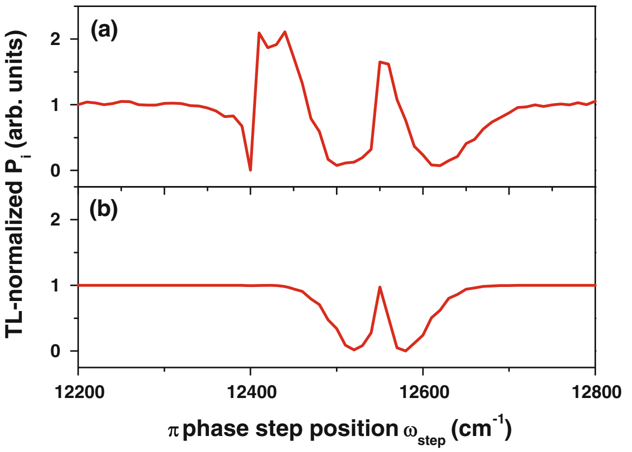

Fig. 6 (Color online) The TL-normalized population of the intermediate state |i> P_i as the function of the π phase step position ω_{step} (*a*) with the pulse intensity of 2×10^9 W/cm^2 and the pulse duration 100 fs for large μ_{fi} with μ_{mg}:μ_{im}:μ_{fi} = 1:1:3 and (*b*) with the pulse intensity of 8×10^9 W/cm^2 and the pulse duration 150 fs for small μ_{fi} with μ_{mg}:μ_{im}:μ_{fi} = 1:1:0.1

|i> P_i in the unshaped laser field on the absorption enhancement of the intermediate state |i>. Figure 5 shows the population of the intermediate state |i> P_i (green solid lines) and the final state |f> P_f (red dashed lines) induced by the unshaped pulse as the function of the pulse intensity for the pulse duration of 100 fs (a) and the pulse duration for the pulse intensity of 5×10^9 W/cm^2 (b), together with their ratio P_f/P_i (blue dotted lines). It is seen that P_f is smaller than P_i in low pulse intensity or short pulse duration, but is larger in high pulse intensity or long pulse duration. Thus, the absorption enhancement of the intermediate state |i> can be intuitively understood as follows. When P_f is much smaller than P_i, the resonance-mediated (2+1) three-photon absorption process is approximated as the non-resonant two-photon absorption, and so no absorption enhancement is observed. However, when P_f is close to P_i or much larger than it, the modulation of P_f in the shaped laser field has great effect on P_i, which leads to the increase of P_i. The ratio P_f/P_i monotonously increases with the increase of the pulse intensity and pulse duration, and this evolution behavior is the same as the absorption enhancement of the intermediate state |i> (Fig. 2). That is to say, the larger P_f/P_i yields the larger absorption enhancement. This further confirms that the absorption enhancement of the intermediate state |i> is due to the effect of the final state |f>.

The transition dipole moment from the intermediate state |i> to the final state |f> μ_{fi} decide the absorption of the final state |f>, and therefore the control efficiency of the intermediate state |i>. Finally, we discuss the influence of μ_{fi} on the absorption enhancement of the intermediate state |i>. Figure 6 shows the TL-normalized population of the intermediate state |i> P_i as the function of the π phase step position ω_{step} with the pulse intensity of 2×10^9 W/cm^2 and the pulse duration 100 fs for large μ_{fi} with μ_{mg}:μ_{im}:μ_{fi} = 1:1:3 (a) and with the pulse intensity of 8×10^9 W/cm^2 and the pulse duration 150 fs for small μ_{fi} with μ_{mg}:μ_{im}:μ_{fi} = 1:1:0.1 (b). It can be seen that when μ_{fi} is enough large, the absorption of the intermediated state |i> is enhanced at low pulse intensity and short pulse duration as shown in Fig. 6(a). However, when μ_{fi} is too small, the absorption enhancement cannot be obtained even at higher pulse intensity and longer pulse duration as shown in Fig. 6(b). Therefore, in order to obtain the absorption enhancement of the intermediated state |i> under weak laser field, the use of the large μ_{fi} is necessary.

4. Conclusions

We present a theoretical study on the intermediate state absorption control in the resonance-mediated (2+1) three-photon excitation process by shaping the femtosecond pulse with a π phase step modulation under weak laser field, and show that the intermediate state absorption can be enhanced, and the enhancement increases with the increase of the pulse intensity and pulse duration. We prove that the coherent enhancement is due to the absorption reduction of the final state from the intermediate state in the shaped laser field. In addition, we show that the population difference of the final and intermediate states in the unshaped laser field and the transition dipole moment from the intermediate state to the final state greatly affects the absorption enhancement. We believe that these results can provide a theoretical basis for experimental study and can be further extended to the control of the intermediate state in various resonance-mediated multiphoton absorption processes.

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