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Polarization control of intermediate state absorption in resonance-mediated multiphoton absorption process

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Abstract

We theoretically and experimentally demonstrate the control of the intermediate state absorption in an (n + m) resonance-mediated multi-photon absorption process by the polarization-modulated femtosecond laser pulse. An analytical solution of the intermediate state absorption in a resonance-mediated multi-photon absorption process is obtained based on the time-dependent perturbation theory. Our theoretical results show that the control efficiency of the intermediate state absorption by the polarization modulation is independent of the laser intensity when the transition from the intermediate state to the final state is coupled by the single-photon absorption, but will be affected by the laser intensity when this transition is coupled by the non-resonant multi-photon absorption. These theoretical results are experimentally confirmed via a twophoton fluorescence control in (2 + 1) resonance-mediated three-photon absorption of Coumarin 480 dye and a single-photon fluorescence control in (1 + 2) resonance-mediated three-photon absorption of IR 125 dye.

Keywords: polarization modulation, intermediate state absorption, control efficiency, laser intensity

(Some figures may appear in colour only in the online journal)

1. Introduction

Single- or multi-photon absorption or fluorescence in the molecular system has become a well-established tool that was widely applied on fluorescence correlation spectroscopy and fluorescence imaging spectroscopy and microscopy [1-4]. It is crucial to artificially enhance or suppress the molecular absorption or fluorescence to further extend its related applications. In recent years, a coherent control strategy by shaping the femtosecond laser pulse in the frequency domain, such as the laser phase, amplitude or polarization modulation, has opened a new opportunity to manipulate the molecular absorption or fluorescence, and several schemes have been proposed and experimentally realized [5–17]. For example,

the single- or two-photon fluorescence can be enhanced or suppressed by the laser spectral phase and/or amplitude modulation based on either an open- or closed-loop scheme [5-16], and the two-photon fluorescence can be suppressed by the laser polarization modulation [17].

Recently, the intermediate state absorption control in the resonance-mediated multi-photon absorption process has attracted considerable attention because the target state absorption control was usually affected by its higher state absorption in an actual experiment [18–21]. For example, the single-photon fluorescence in IR 125 or IR 144 dye can be enhanced by the laser spectral phase modulation or polarization modulation, which is attributed to the higher excited state absorption [19–21]. However, these previous works just

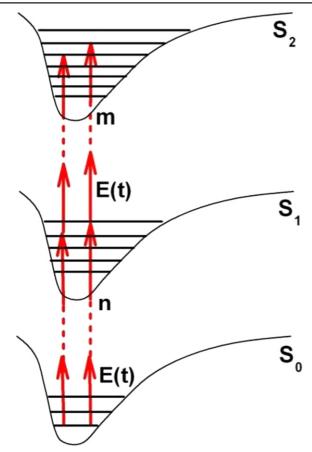


Figure 1. Schematic energy-level diagram of the (n + m) resonancemediated multi-photon absorption process.

qualitatively explained the intermediate state absorption control and did not give the theoretical simulation. In order to better understand and control the intermediate state absorption in the resonance-mediated multi-photon absorption process, in this paper we theoretically and experimentally study the intermediate state absorption control in the resonance-mediated multi-photon absorption process by the polarizationmodulated femtosecond laser pulse. Based on the timedependent perturbation theory, we establish a theoretical model to describe the polarization-controlled intermediate state absorption in an (n+m) resonance-mediated multi-photon absorption process and obtain the corresponding analytical solution. We theoretically show that the laser intensity does not affect the control efficiency of the intermediate state absorption by the polarization modulation when the transition from the intermediate state to the final state is coupled by the single-photon absorption, but will affect this control efficiency when this transition is coupled by the non-resonant multi-photon absorption. Finally, we confirm these theoretical predictions by performing these experiments of two-photon fluorescence control in (2+1) resonance-mediated threephoton absorption of Coumarin 480 dye and single-photon fluorescence control in (1+2) resonance-mediated threephoton absorption of IR 125 dye.

2. Theoretical method

Figure 1 shows the schematic energy-level diagram of the (n+m) resonance-mediated multi-photon absorption process in the isotropic molecular system exited by the femtosecond laser pulse $\vec{E}(t)$, where S_0 , S_1 and S_2 represent the ground state, the intermediate state and the final state, respectively. The transition from the S_0 state to S_1 state is coupled by *n*photon absorption and the transition from the S_1 state to S_2 state is coupled by *m*-photon absorption. In our simulation, we consider the perturbative regime and that the interaction between the femtosecond laser field and the molecular system can be well described by time-dependent perturbation theory. In this case, the intermediate S_1 state population P_{S1} in the (n+m) resonance-mediated multi-photon absorption process can be approximated as

$$P_{S1} = A_{S1} - A_{S2}, \tag{1}$$

where A_{S1} represents the *n*-photon absorption in the S_1 state, which causes the population of the S_1 state, and A_{S2} represents the (n+m) resonance-mediated multi-photon absorption in the S_2 state, which causes the depopulation of the S_1 state. In the molecular system with a broad absorption line, the multi-photon absorption can be considered as a sum of each individual transition. On the basis of the theoretical model of the atomic system with a narrow absorption line limit [22, 23], A_{S1} and A_{S2} can be written as

$$A_{S1} = \frac{1}{\hbar^{2n}} \int_{-\infty}^{+\infty} G(\omega_{S1}) \left| \int_{-\infty}^{+\infty} \mu_{S0 \to S1} \vec{E}^n(t) \right| \times \exp(i\omega_{S1}t) dt^2 d\omega_{S1}, \qquad (2)$$

and

$$A_{S2} = \frac{1}{\hbar^{2(n+m)}} \int_{-\infty}^{+\infty} G(\omega_{S2}) \left| \int_{-\infty}^{+\infty} G(\omega_{S1}) \mu_{S0 \to S1} \mu_{S1 \to S2} \right|$$
$$\times \int_{-\infty}^{+\infty} \int_{-\infty}^{t_1} \vec{E}^m(t_1) \vec{E}^n(t_2) \exp[i(\omega_{S2} - \omega_{S1}) t_1]$$
$$\times \exp(i\omega_{S1} t_2) dt_2 dt_1 d\omega_{S1} \right|^2 d\omega_{S2},$$
(3)

where \hbar is the Planck constant, $G(\omega_{S1})$ and $G(\omega_{S2})$ are the molecular absorption line-shape function in the S_1 state and S_2 state, $\mu_{S0\to S1}$ and $\mu_{S1\to S2}$ are the effective dipole coupling for the state transitions $S_0 \to S_1$ and $S_1 \to S_2$, and ω_{S1} and ω_{S2} are the frequency for the state transitions $S_0 \to S_1$ and $S_0 \to S_1$ and $S_0 \to S_2$.

When the femtosecond laser field with linear polarization, i.e. $\vec{E}(t) = E_0(t) \cos(\omega t) \vec{e}_x$, is modulated by a quarter wave plate ($\lambda/4$ wave plate), the output laser field can be written as

$$\vec{E}_{\lambda/4}(t) = E_0(t) \Big[\cos(\varphi) \cos(\omega t) \vec{e}_x \\ + \sin(\varphi) \cos(\omega t + \pi/2) \vec{e}_y \Big],$$
(4)

where φ is the angle between the polarization direction of the input laser field and the optical axis of the $\lambda/4$ wave plate.

One can see that the output laser is linearly polarized for $\varphi = 0$, $\pi/2$, π and $3\pi/2$, circularly polarized for $\varphi = \pi/4$, $3\pi/4$, $5\pi/4$ and $7\pi/4$, and elliptically polarized for the other rotation angle φ . In the (n+m) resonance-mediated multi-photon absorption process, all the *n* or *m* photons can only come from the same polarization direction, but between these *n* and *m* photons can be either the same polarization direction or different polarization directions. Thus, A_{S1} and A_{S2} in equations (2) and (3) can be further expressed as

$$A_{S1} = \left[\cos^{2n}(\varphi) + \sin^{2n}(\varphi)\right] \frac{1}{\hbar^{2n}} \int_{-\infty}^{+\infty} G(\omega_{S1})$$
$$\times \left| \int_{-\infty}^{+\infty} \mu_{S0 \to S1} \vec{E}^{n}(t) \exp(i\omega_{S1}t) dt \right|^{2} d\omega_{S1}, \quad (5)$$

and

$$A_{S2} = \left\{ \left[\cos^{2n}(\varphi) + \sin^{2n}(\varphi) \right] \times \left[\cos^{2m}(\varphi) + \sin^{2m}(\varphi) \right] \right\} \times \frac{1}{\hbar^{2(n+m)}} \int_{-\infty}^{+\infty} G(\omega_{S2})$$
$$\times \left| \int_{-\infty}^{+\infty} G(\omega_{S1}) \mu_{S0 \to S1} \mu_{S1 \to S2} \right.$$
$$\times \int_{-\infty}^{+\infty} \int_{-\infty}^{t_1} \vec{E}^m(t_1) \vec{E}^n(t_2) \exp\left[i(\omega_{S2} - \omega_{S1}) t_1 \right]$$
$$\times \left. \exp\left(i\omega_{S1} t_2 \right) dt_2 dt_1 d\omega_{S1} \right|^2 d\omega_{S2}.$$

(6)

By normalizing equations (5) and (6) with $\vec{E}^{n(m)}(t) = \vec{E}^{n(m)}(t) / E_0^{n(m)}$, where E₀ is the peak spectral amplitude, P_{S1} can be further simplified as

$$P_{S1} = \frac{\mu_{S0 \to S1}^2 E_0^{2n}}{\hbar^{2n}} \Big[\cos^{2n}(\varphi) + \sin^{2n}(\varphi) \Big] \\ \times \left\{ \int_{-\infty}^{+\infty} G(\omega_{S1}) \left| \int_{-\infty}^{+\infty} \tilde{E}^n(t) \exp(i\omega_{S1}t) dt \right|^2 d\omega_{S1} \right. \\ \left. - \frac{\mu_{S1 \to S2}^2 E_0^{2m}}{\hbar^{2m}} \Big[\cos^{2m}(\varphi) + \sin^{2m}(\varphi) \Big] \\ \times \int_{-\infty}^{+\infty} G(\omega_{S2}) \left| \int_{-\infty}^{+\infty} G(\omega_{S1}) \int_{-\infty}^{+\infty} \int_{-\infty}^{t_1} \tilde{E}^m(t_1) \right. \\ \left. \times \tilde{E}^n(t_2) \exp[i(\omega_{S2} - \omega_{S1})t_1] \\ \times \exp(i\omega_{S1}t_2) dt_2 dt_1 d\omega_{S1} \Big|^2 d\omega_{S2} \Big\}.$$

$$(7)$$

As can be seen from equations (5)–(7), both A_{S1} and A_{S2} depend on the laser polarization (i.e. $\lambda/4$ wave plate rotation angle φ). A_{S1} is proportional to $\begin{bmatrix} \cos^{2n}(\varphi) + \sin^{2n}(\varphi) \end{bmatrix}$ and A_{S2} is proportional to $\begin{bmatrix} \cos^{2n}(\varphi) + \sin^{2n}(\varphi) \end{bmatrix}$ and $\begin{bmatrix} \cos^{2n}(\varphi) + \sin^{2n}(\varphi) \end{bmatrix} \times \begin{bmatrix} \cos^{2m}(\varphi) + \sin^{2m}(\varphi) \end{bmatrix}$, and therefore P_{S1} can be controlled by varying the laser polarization, but the control efficiency is correlated with the absorbed photon number *n* and *m*. Here, the control efficiency is defined by the function of $\eta = (P_{S1_Max} - P_{S1_Min})/P_{S1_Lin}$, where P_{S1_Max} and P_{S1_Min} are, respectively, the maximum

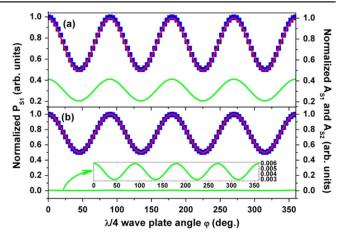


Figure 2. The intermediate state population P_{S1} (blue squares, left coordinate) in the (2+1) resonance-mediated three-photon absorption process as a function of the $\lambda/4$ wave plate rotation angle φ with the laser intensities of (a) 3.6×10^{12} and (b) 4.3×10^{11} W cm⁻², together with the two-photon absorption A_{S1} (red lines, right coordinate) and the resonance-mediated three-photon absorption A_{S2} (green lines, right coordinate). Inset in figure 2(b) shows the enlarged diagram of A_{S2} .

and minimum values of P_{S1} , and P_{S1_Lin} is the value of P_{S1} by the linearly polarized laser excitation (i.e. $\varphi = 0$). In addition, A_{S1} is proportional to the *n*th power of the laser intensity (i.e. E_0^{2n}) while A_{S2} is proportional to the (n+m)th power of the laser intensity (i.e. $E_0^{2(n+m)}$), and thus the control efficiency of P_{S1}, by varying the laser polarization, will be affected by the laser intensity that depends on the absorbed photon number *m*. When the transition from the intermediate S_1 state to the final S_2 state is coupled by the single-photon absorption (i.e. m=1), the polarization modulation has the same effect on both A_{S1} and A_{S2} (i.e. $\cos^{2n}(\varphi) + \sin^{2n}(\varphi)$), and thus the control efficiency of P_{S1} by the polarization modulation is independent of the laser intensity. However, when the transition from the intermediate S_1 state to the final S_2 state is coupled by the non-resonant multi-photon absorption (i.e. $m \ge 2$), the polarization modulation produces a different effect on A_{S1} and A_{S2} , and therefore the control efficiency of P_{S1} will be affected by the laser intensity.

3. Theoretical results

To demonstrate the above theoretical predictions, we theoretically simulate the control of P_{S1} by varying the laser polarization for the relatively high and low laser intensities, and the simulation results are presented in figures 2 and 3. Here, we consider two simple and representative cases of the intermediate state absorption control in (1+2) and (2+1) resonance-mediated three-photon absorption processes. In our simulation, the femtosecond laser pulse has the Gaussian shape with the central frequency of $\omega = 12500 \text{ cm}^{-1}$ and the pulse duration $\tau = 50 \text{ fs}$, and the molecular absorption in the S₁ or S₂ state (i.e. $G(\omega_{S1})$ or $G(\omega_{S2})$) has also the Gaussian

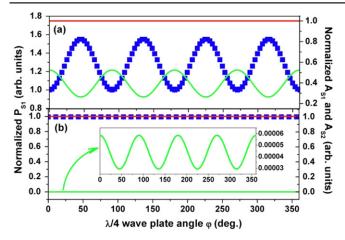


Figure 3. The intermediate state population P_{S1} (blue squares, left coordinate) in the (1+2) resonance-mediated three-photon absorption process as a function of the $\lambda/4$ wave plate rotation angle φ with the laser intensities of (a) 3.9×10^{12} and (b) 4.0×10^{10} W cm⁻², and the single-photon absorption A_{S1} (red lines, right coordinate) and the resonance-mediated three-photon absorption A_{S2} (green lines, right coordinate) are also given together. Inset in figure 3(b) shows the enlarged diagram of A_{S2} .

shape with the central frequency of $\omega_{S1} = n\omega$ or $\omega_{S2} = m\omega$ and the absorption bandwidth of $\Delta \omega_{S1} = \Delta \omega_{S2} = 400 \text{ cm}^{-1}$. Figure 2 shows the intermediate state population P_{S1} in a (2+1) resonance-mediated three-photon absorption process as a function of the $\lambda/4$ wave plate rotation angle φ with the laser intensities of (a) 3.6×10^{12} and (b) 4.3×10^{11} W cm⁻² (blue squares, left coordinate), together with the two-photon absorption in the S1 state AS1 (red lines, right coordinate) and the resonance-mediated three-photon absorption in the S₂ state A_{S2} (green lines, right coordinate). Here, these data of P_{S1} are normalized by the linearly polarized laser excitation, while these data of A_{S1} and A_{S2} are normalized by A_{S1} with the linearly polarized laser excitation, and hereafter the same method is utilized. One can see from figure 2 that both A_{S1} and A_{S2} decrease and have the same control efficiency when the laser polarization changes from linear to circular, and thus the laser polarization will induce the same control efficiency of P_{S1} for the high and low laser intensities (see equation (7)). So, one can conclude that the laser intensity will not affect the control efficiency of P_{S1} by the laser polarization modulation when the transition from the intermediate S_1 state to the final S_2 state is coupled by the single-photon absorption (i.e. m = 1).

Figure 3 shows the intermediate state population P_{S1} in the (1+2) resonance-mediated three-photon absorption process as a function of the $\lambda/4$ wave plate rotation angle φ with the laser intensities of (a) 3.9×10^{12} and (b) 4.0×10^{10} W cm⁻² (blue squares, left coordinate), and the single-photon absorption in the S₁ state A_{S1} (red lines, right coordinate) and the resonance-mediated three-photon absorption in the S₂ state A_{S2} (green lines, right coordinate) are also presented. When the laser polarization changes from linear to circular, A_{S1} remains constant while A_{S2} decreases, and therefore P_{S1} increases. Since the laser polarization has a different effect on A_{S1} and A_{S2} , the laser intensity will affect the control

efficiency of P_{S1} by the laser polarization modulation, and the higher laser intensity yields the larger control efficiency. Consequently, it can be further deduced that the control efficiency of the intermediate state population will be affected by the laser intensity when the transition from the intermediate S_1 state to the final S_2 state is coupled by the multiphoton absorption (i.e. $m \ge 2$).

4. Experimental confirmation

To confirm the above theoretical results of the intermediate state absorption control by varying the polarization modulation, we perform the experiment in Coumarin 480 dye and IR 125 dye with different laser intensities. In our experiment, we utilize a Ti-sapphire mode-locked regenerative amplifier (Spectra-Physics, Spitfire) as the excitation source with the pulse width (Full Width at Half Maximum) of about 50 fs, the central wavelength of 800 nm and the repetition rate of 1 kHz. An attenuator is used to vary the laser intensity, and a quarter wave plate ($\lambda/4$ wave plate) is employed to change the laser polarization from linear through elliptical to circular and vice versa. The polarization-shaped femtosecond laser pulse is focused into a quartz cuvette containing fluorescent dye with a lens of 200 mm focal length. The Coumarin 480 and IR 125 dyes are purchased from Exciton and used without further purification, which are dissolved in methanol solution with a concentration of 1.0×10^{-4} mol L⁻¹. The fluorescence signal is perpendicularly collected and measured by a spectrometer with a charge-coupled device.

Figure 4(a) shows the absorption (red line) and fluorescence (green line) spectra of Coumarin 480 dye. According to the position of the absorption and fluorescence peaks, it can be determined that the excitation process in Coumarin 480 dye is the (2+1) resonance-mediated three-photon absorption, as shown in figure 4(c). Figure 4(b) presents the normalized two-photon fluorescence intensity as a function of the $\lambda/4$ wave plate rotation angle φ with the laser intensities of 3.6×10^{12} (red squares) and 4.3×10^{11} W cm⁻² (green circles), and the theoretical simulations are also given together (blue line). As expected, the laser intensity does not affect the control efficiency of the two-photon fluorescence intensity by varying the laser polarization. It can be found that this experimental result is consistent with the theoretical expectation. Similarly, on the basis of the absorption and fluorescence spectra of IR 125 dye in figure 5(a), the excitation process in IR 125 dye can be assigned as the (1+2) resonance-mediated three-photon absorption, as shown in figure 5(c). Figure 5(b) presents the normalized single-photon fluorescence intensity as a function of the $\lambda/4$ wave plate rotation angle φ with the laser intensities of 3.9×10^{12} (red squares) and 4.0×10^{10} W cm⁻² (green circles), together with the corresponding theoretical simulations (blue lines). One can see that the control efficiency of the single-photon fluorescence intensity, by varying the laser polarization, decreases when the laser intensity decreases. Obviously, such an experimental result is also in good agreement with the theoretical predictions.

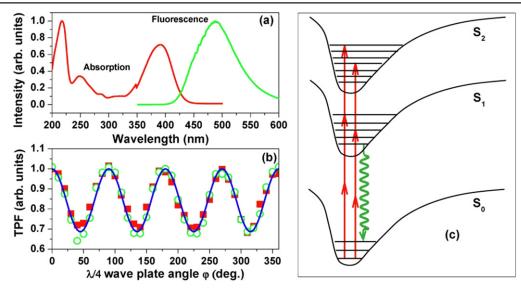


Figure 4. (a) The absorption (red line) and fluorescence (green line) spectra of Coumarin 480 dye. (b) The normalized two-photon fluorescence intensity as a function of the $\lambda/4$ wave plate rotation angle φ with the laser intensities of 3.6×10^{12} (red squares) and 4.3×10^{11} W cm⁻² (green circles), together with the theoretical simulations (blue line). (c) The schematic energy-level diagram of (2+1) resonance-mediated three-photon absorption process in Coumarin 480 dye.

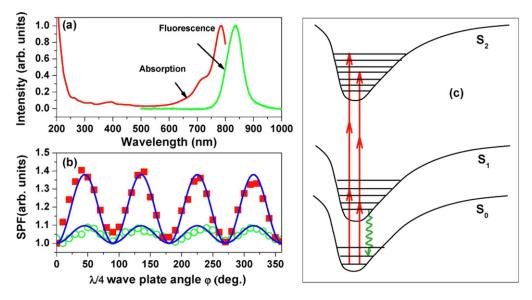


Figure 5. (a) The absorption (red line) and fluorescence (green line) spectra of IR 125 dye. (b) The normalized single-photon fluorescence intensity as a function of the $\lambda/4$ wave plate rotation angle φ with the laser intensities of 3.9×10^{12} (red squares) and 4.0×10^{10} W cm⁻² (green circles), together with the corresponding theoretical simulations (blue lines). (c) The schematic energy-level diagram of (1 + 2) resonance-mediated three-photon absorption process in IR 125 dye.

5. Conclusions

In conclusion, we have theoretically and experimentally shown that the intermediate state absorption in the (n+m)resonance-mediated multi-photon absorption process can be controlled by varying the laser polarization. A theoretical model was proposed to study the polarization-controlled intermediate state absorption based on the time-dependent perturbation theory, and an analytical solution was obtained. Our theoretical results showed that the laser intensity will not affect the control efficiency of the intermediate state absorption when the transition from the intermediate state to the final state is coupled by the single-photon absorption, which will affect it when this transition is coupled by the nonresonant multi-photon absorption. Furthermore, the twophoton fluorescence control in (2+1) resonance-mediated three-photon absorption of Coumarin 480 dye and singlephoton fluorescence control in (1+2) resonance-mediated three-photon absorption of IR 125 dye were experimentally performed to validate these theoretical results. We believe these theoretical and experimental results are very helpful to further understand and control the intermediate state absorption in various resonance-mediated multi-photon absorption processes.

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