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2017 Laser Phys. Lett. 14 045301

(http://iopscience.iop.org/1612-202X/14/4/045301)

IOP Publishing | Astro Ltd
Laser Physics Letters
Laser Phys. Lett. **14** (2017) 045301 (7pp)
doi:10.1088/1612-202X/aa5e3b

Letter

Polarization control efficiency manipulation in resonance-mediated two-photon absorption by femtosecond spectral frequency modulation

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Received 19 January 2017, revised 1 February 2017
Accepted for publication 1 February 2017
Published 20 February 2017

Abstract

The femtosecond laser polarization modulation is considered as a very simple and efficient method to control the multi-photon absorption process. In this work, we theoretically and experimentally show that the polarization control efficiency in the resonance-mediated two-photon absorption can be artificially manipulated by modulating the femtosecond spectral frequency components. We theoretically demonstrate that the on- and near-resonant parts in the resonance-mediated two-photon absorption process depend on the different femtosecond spectral frequency components, and therefore their contributions in the whole excitation process can be controlled by properly designing the femtosecond spectral frequency components. The near-resonant two-photon absorption is correlated with the femtosecond laser polarization while the on-resonant two-photon absorption is independent of it, and thus the polarization control efficiency in the resonance-mediated two-photon absorption can be manipulated by the femtosecond spectral frequency modulation. We experimentally verify these theoretical results by performing the laser polarization control experiment in the Dy^{3+}-doped glass sample under the modulated femtosecond spectral frequency components, and the experimental results show that the polarization control efficiency can be increased when the central spectral frequency components are cut off, while it is decreased when both the low and high spectral frequency components are cut off, which is in good agreement with the theoretical predictions. Our works can provide a feasible pathway to understand and control the resonance-mediated multi-photon absorption process under the femtosecond laser field excitation, and also may open a new opportunity to the related application areas.

Keywords: up-conversion luminescence, coherent control, femtosecond laser

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

1. Introduction

The multi-photon absorption in the matter is a nonlinear process, which has attracted a lot of attention because of its special advantages compared with one-photon absorption, such as the lower damage to sample, larger penetration depth, higher spatial resolution, and so on. So far, the multi-photon absorption has been widely applied in various related areas, such as the frequency up-conversion laser source [1–4], optical limiting [5, 6], optical reshaping [7], stimulated scattering [8], confocal microscopy [9–11], three-dimensional data storage [12–14], and nano/micro fabrication [15–17]. If the multi-photon absorption can be artificially manipulated, including the absorption enhancement or suppression and the state-selected absorption, the related application areas can be greatly extended. Therefore, the manipulation of the multi-photon absorption has become a meaningful research field, and has aroused the considerable interest of scientists in the field of optical science. Usually, the infrared femtosecond laser was used as the excitation source of the multi-photon absorption in the experiment because of the higher laser intensity. Recent studies have shown that the femtosecond pulse shaping technique by the phase and/or amplitude modulation in the frequency domain is a well-established method to control the multi-photon absorption, such as the non-resonant two photon absorption [18], (1 + 1) resonance-mediated two-photon absorption [19], (2 + 1) or (1 + 2) resonance-mediated three-photon absorption [20, 21], and so on. Moreover, the femtosecond pulse polarization modulation has also shown to be an available technique to manipulate the multi-photon absorption [22–25], and the related research results showed that the femtosecond laser polarization can vary the non-resonant multi-photon absorption between the two neighboring states but can't affect the single-photon absorption in the state transition process [23].

In our previous studies, we only considered the control efficiency by varying the femtosecond laser polarization [24, 25]. Here, we further study the polarization control efficiency manipulation by additionally modulating the femtosecond spectral frequency components. In this work, we theoretically and experimentally show that the polarization control efficiency in the resonance-mediated two-photon absorption can be increased or decreased by properly designing the femtosecond spectral frequency components. Our theoretical results show that the near-resonant excitation process in the resonance-mediated two-photon absorption will be affected by the femtosecond laser polarization, but the on-resonant excitation process is independent of it, and therefore the polarization control efficiency can be manipulated by varying the femtosecond spectral frequency components, because the on-resonant and near-resonant two-photon absorptions result from the different spectral frequency component excitations. Our experimental results show that the polarization control efficiency in the Dy^{3+}-doped glass sample can be increased when the central frequency components are cut off, while can be decreased when both the low and high frequency components are cut off, which are consistent with the theoretical simulations.

2. Theoretical model

The schematic diagram of resonance-mediated two-photon absorption is shown in figure 1(a). Here, the three levels $|\mathrm{g}\rangle$, $|\mathrm{i}\rangle$ and $|\mathrm{f}\rangle$ represent the ground state, the intermediate state and the final state, respectively. In our theoretical simulation, the transition frequencies of $|\mathrm{g}\rangle \rightarrow |\mathrm{i}\rangle$ and $|\mathrm{i}\rangle \rightarrow |\mathrm{f}\rangle$ are considered to be approximately equal. When such a three-level quantum system is excited by the femtosecond laser field, the state transition of $|\mathrm{g}\rangle \rightarrow |\mathrm{f}\rangle$ is induced by the resonance-mediated two-photon absorption through the intermediate state $|\mathrm{i}\rangle$. If the femtosecond laser spectral bandwidth is larger than the absorption bandwidth of the intermediate state $|\mathrm{i}\rangle$, the resonance-mediated two-photon absorption can be decomposed into two parts, one is the on-resonant excitation process, and the other one is the near-resonant excitation process, as shown in figure 1(a). The on-resonant excitation process is that the population in the ground state $|\mathrm{g}\rangle$ is pumped to the intermediate state $|\mathrm{i}\rangle$ by absorbing one photon and then further excited to the final state $|\mathrm{f}\rangle$ by absorbing the other one, where the energies of the two photons must match with the two transition frequencies of $|\mathrm{g}\rangle \rightarrow |\mathrm{i}\rangle$ and $|\mathrm{i}\rangle \rightarrow |\mathrm{f}\rangle$, respectively. However, the near-resonant excitation process is that the population in the ground state $|\mathrm{g}\rangle$ is directly pumped to the final state $|\mathrm{f}\rangle$ by simultaneously absorbing two photons. Here, the excitation process does not need to go through the intermediate state $|\mathrm{i}\rangle$, as long as the frequency sum of the two photons is equal to transition frequency of $|\mathrm{g}\rangle \rightarrow |\mathrm{f}\rangle$, the near-resonant two-photon absorption can occur. Under the femtosecond laser field excitation, the resonance-mediated two-photon transition probability in the three-level quantum system can be expressed as [26, 27]

$$S \propto \int_{-\infty}^{+\infty} \mathrm{d}\omega_{\mathrm{f}} A(\omega_{\mathrm{f}}) \left| \int_{-\infty}^{+\infty} A(\omega_{\mathrm{i}}) \int_{-\infty}^{+\infty} E(t_1) \exp\left[\mathrm{i}(\omega_{\mathrm{f}} - \omega_{\mathrm{i}})t_1\right] \times \int_{-\infty}^{t_1} E(t_2) \exp(\mathrm{i}\omega_i t_2) \mathrm{d}t_2 \mathrm{d}t_1 \mathrm{d}\omega_{\mathrm{i}} \right|^2, \quad (1)$$

where $E(t)$ is the intensity distribution of the femtosecond laser field in the time domain, $A(\omega_{\mathrm{i}})$ and $A(\omega_{\mathrm{f}})$ are the absorption line-shape functions of the intermediate state $|\mathrm{i}\rangle$ and final state $|\mathrm{f}\rangle$, and ω_{i} and ω_{f} are the resonant transition frequencies

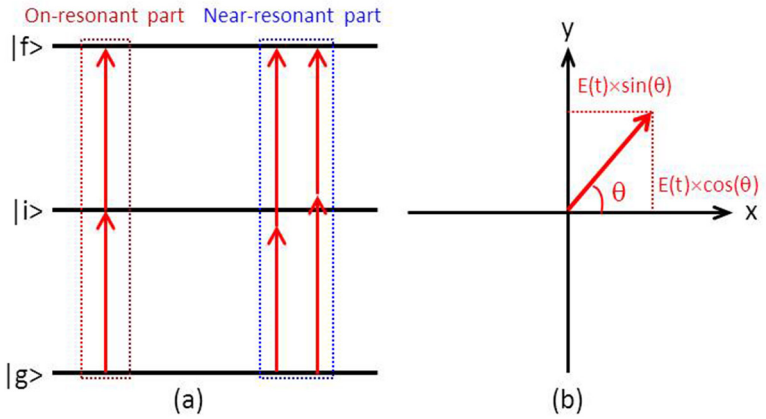

Figure 1. The schematic diagrams of resonance-mediated two-photon absorption (a) and the polarization modulated laser field (b).

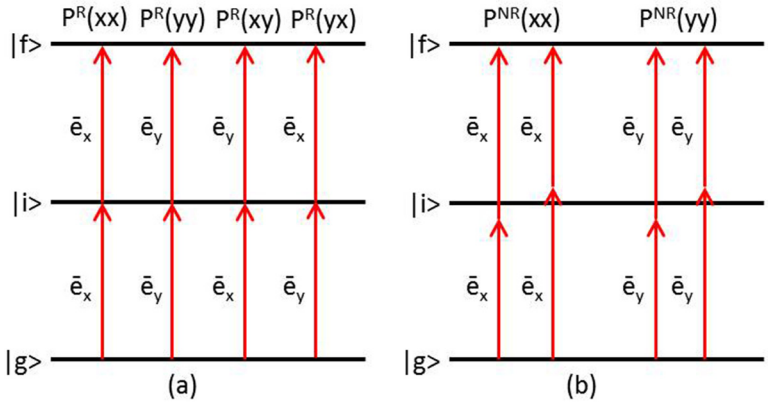

Figure 2. The possible excitation pathways of on-resonant (a) and near-resonant (b) two-photon absorption by the polarization modulated femtosecond laser field.

of $|\mathrm{g}\rangle \rightarrow |\mathrm{i}\rangle$ and $|\mathrm{g}\rangle \rightarrow |\mathrm{f}\rangle$. By transforming equation (1) into the frequency domain, the resonance-mediated two-photon transition probability can be further written as

$$S \propto \int_{-\infty}^{+\infty} \mathrm{d}\omega_{\mathrm{f}} A(\omega_{\mathrm{f}}) |P_{\text{On-Res.}} + P_{\text{Near-Res.}}|^2, \tag{2}$$

with

$$P_{\text{On-Res.}} \propto \mathrm{i}\pi \int_{-\infty}^{+\infty} \mathrm{d}\omega_{\mathrm{i}} A(\omega_{\mathrm{i}}) E_0(\omega_{\mathrm{f}} - \omega_{\mathrm{i}}) E_0(\omega_{\mathrm{i}}) \mathrm{e}^{\mathrm{i}[\Phi(\omega_{\mathrm{f}}-\omega_{\mathrm{i}})+\Phi(\omega_{\mathrm{i}})]}, \tag{3}$$

$$P_{\text{Near-Res.}} \propto \wp \int_{-\infty}^{+\infty} \mathrm{d}\omega E_0(\omega_{\mathrm{f}} - \omega) E_0(\omega) \mathrm{e}^{\mathrm{i}[\Phi(\omega_{\mathrm{f}}-\omega)+\Phi(\omega)]} / (\omega_{\mathrm{i}} - \omega), \tag{4}$$

where $\wp$ is the Cauchy's principal value, $E(\omega)$ is the Fourier transform of $E(t)$ with $E(\omega) = E_0(\omega) \times \exp[\mathrm{i}\Phi(\omega)]$, and $E_0(\omega)$ and $\Phi(\omega)$ are the spectral amplitude and phase in the frequency domain, respectively. As can be seen, the on-resonant term $P_{\text{On-Res.}}$ in equation (3) represents the interferences of all on-resonant two-photon excitation pathways via the intermediate state $|\mathrm{i}\rangle$ with the transition frequencies of ω_{i} and ω_{f}–ω_{i}, while the near-resonant term $P_{\text{Near-Res.}}$ in equation (4) includes all other near-resonant two-photon excitation pathways with the transition frequencies of ω and ω_{f}–ω. In this theoretical model, the resonant term is excluded from the near-resonant term by the Cauchy's principal value operator $\wp$. Moreover, it can be found from equations (3) and (4) that the on-resonant and near-resonant two-photon absorptions should result from the excitations of different spectral frequency components.

The femtosecond laser polarization technique has shown to be a well-established tool to control the multi-photon absorption [22–25]. In the real experiment, the quarter-wave ($\lambda/4$) plate was usually utilized to change the laser polarization from linear into elliptical or circular. Mathematically, the polarization modulated femtosecond laser field in the space can be decomposed into two orthogonal directions (x and y), as shown in figure 1(b). Thus, the polarization shaped femtosecond laser field can be expressed as

$$\overline{E}_p(t) = E(t)\cos(\theta)\bar{e}_x + E(t)\sin(\theta)\bar{e}_y, \tag{5}$$

where θ is the angle between the input laser polarization direction and the $\lambda/4$ wave plate optical axis. As can be seen in equation (5), the output laser is linear polarization for $\theta = m\pi/2$ ($m = 0, 1, 2\ldots$), circular polarization for $\theta = (2m+1)\pi/4$, and elliptical polarization for other rotation angle. Figure 2 shows the possible excitation pathways of on-resonant (a) and near-resonant (b) two-photon absorptions by the polarization modulated femtosecond laser field. One can see that the two photons via the on-resonant absorption can come from the same polarization direction (i.e. $P^{\mathrm{R}}(xx)$, and $P^{\mathrm{R}}(yy)$) or different polarization directions (i.e. $P^{\mathrm{R}}(xy)$ and $P^{\mathrm{R}}(yx)$), as

shown in figure 2(a), but the two photons via the near-resonant absorption can only come from the same polarization direction (i.e. $P^{\mathrm{NR}}(xx)$ and $P^{\mathrm{NR}}(yy)$), as shown in figure 2(b). Thus, the on-resonant part contribution $S_{\text{On-Res.}}$ and near-resonant part contribution $S_{\text{Near-Res.}}$ induced by the polarization modulated femtosecond laser field can be respectively written as

$$\begin{aligned} S_{\text{On-Res.}} &= [\cos^4(\theta) + \sin^4(\theta) + 2\cos^2(\theta)\sin^2(\theta)] \\ &\quad \times \int_{-\infty}^{+\infty} \mathrm{d}\omega_{\mathrm{f}} A(\omega_{\mathrm{f}}) \left|P_{\text{On-Res.}}\right|^2 \\ &= \int_{-\infty}^{+\infty} \mathrm{d}\omega_{\mathrm{f}} A(\omega_{\mathrm{f}}) \left|P_{\text{On-Res.}}\right|^2 , \end{aligned} \tag{6}$$

and

$$S_{\text{Near-Res.}} = [\cos^4(\theta) + \sin^4(\theta)] \int_{-\infty}^{+\infty} \mathrm{d}\omega_{\mathrm{f}} A(\omega_{\mathrm{f}}) \left|P_{\text{Near-Res.}}\right|^2 . \tag{7}$$

As can be seen, the on-resonant contribution $S_{\text{On-Res.}}$ is independent of the $\lambda/4$ plate angle θ (see equation (6)). However, the near-resonant term $S_{\text{Near-Res.}}$ is correlated with the $\lambda/4$ plate angle θ (see equation (7)), which is maximal value for $\theta = m\pi/2$ (linear polarization) and minimal value for $\theta = (2m+1)\pi/4$ (circular polarization). That is to say, the femtosecond laser polarization modulation can suppress the near-resonant two-photon absorption but does not affect the on-resonant two-photon absorption.

Figure 3 shows the normalized resonance-mediated two-photon absorption by varying the $\lambda/4$ plate angle θ based on equation (2) (red solid line), together with the normalized on-resonant (blue dashed line) and near-resonant (black dotted line) part contributions according to equations (6) and (7). As can be seen, when the femtosecond laser polarization is changed from linear through elliptical to circular, the near-resonant two-photon absorption is suppressed, while the on-resonant two-photon absorption keeps unchanged, which is consistent with above analysis. One can see that the polarization control efficiency of the near-resonant two-photon absorption can be up to 50%. Here, the polarization control efficiency is defined as $\eta = (I_{\max} - I_{\min})/I_{\max}$, where $I_{\max}$ and $I_{\min}$ are the maximal and minimal absorption. Since the resonance-intermediated two-photon absorption includes both the contributions of on-resonant and near-resonant parts, and the total polarization control efficiency should be between 0% and 50%, which depends on the relative weight of the on-resonant and near-resonant parts in the whole excitation process.

As discussed above, the polarization control efficiency of the resonance-intermediated two-photon absorption is decided by both the contributions of the on-resonant and near-resonant two-photon absorptions, and the on-resonant and near-resonant parts depend on the different spectral frequency components. It is easy to verify that the on-resonant part mainly comes from the contribution of the central spectral frequency components, while the near-resonant part mainly results from the excitation of the low and high spectral frequency components (see figure 1(a)), and therefore the polarization control efficiency can be artificially manipulated by properly designing

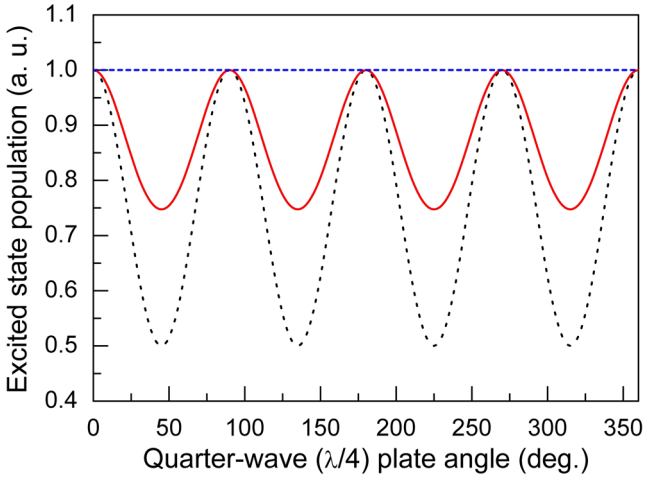

Figure 3. The theoretical simulation of the normalized resonance-mediated two-photon absorption in the final excited state by varying the $\lambda/4$ plate angle θ (red solid line), together with the normalized on-resonant (blue dashed line) and near-resonant (black dotted line) part contributions.

the femtosecond spectral frequency components. One simple way is to cut off the central spectral frequency components to reduce the contribution of the on-resonant part while keep the near-resonant part excitation, and thus the polarization control efficiency can be increased. The other simple way is to cut off both the low and high spectral frequency components to eliminate the excitation of the near-resonant part while keep the on-resonant part contribution, and therefore the polarization control efficiency can be decreased. Obviously, the femtosecond spectral frequency modulation can provide a simple and efficient method to increase or decrease the polarization control efficiency of the resonance-mediated two-photon absorption.

3. Experimental confirm

To confirm that the polarization control efficiency of the resonance-mediated two-photon absorption can be artificially tuned by the femtosecond laser spectral frequency modulation, we perform the polarization control experiment in the Dy^{3+}-doped glass sample, and the experimental arrangement is shown in figure 4. Here, a Ti-sapphire mode-locked regenerative amplifier (Spectra-physics, Spitfire) is used as the femtosecond laser excitation source with the pulse width of about 60 fs, central wavelength of 804 nm and repetition rate of 1 kHz. A programmable 4-f configuration zero-dispersion pulse shaper is used to modulate the spectral components in the frequency domain, which is composed of a pair of diffraction gratings with 1200 lines mm^{-1}, a pair of concave mirrors with focal length of 200 mm and an one-dimension liquid-crystal spatial light modulator (SLM-S320d, JENOPTIK), and the SLM is placed at the Fourier plane and used to modulate the femtosecond spectral amplitude by selectively filtering out the portion of the frequency components. An achromatic quarter-wave ($\lambda/4$) plate is used to vary the femtosecond laser

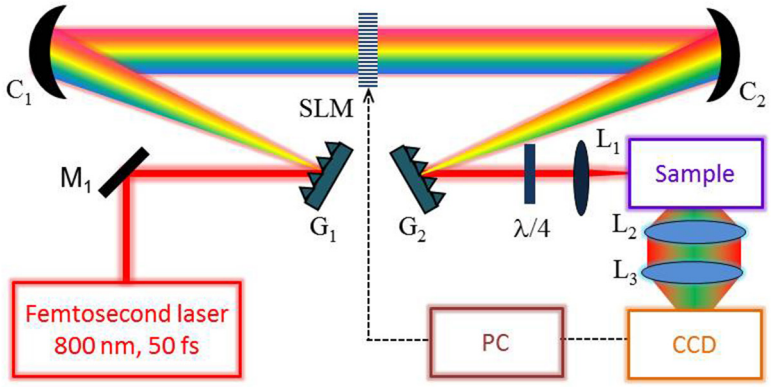

Figure 4. Schematic diagram of the experimental arrangement for the polarization control of up-conversion luminescence in the Dy^{3+}-doped glass sample by the femtosecond spectral frequency modulation. Here, a spatial light modulator (SLM) is used to realize the laser spectral frequency modulation, and a $\lambda/4$ wave plate is used to vary the laser polarization.

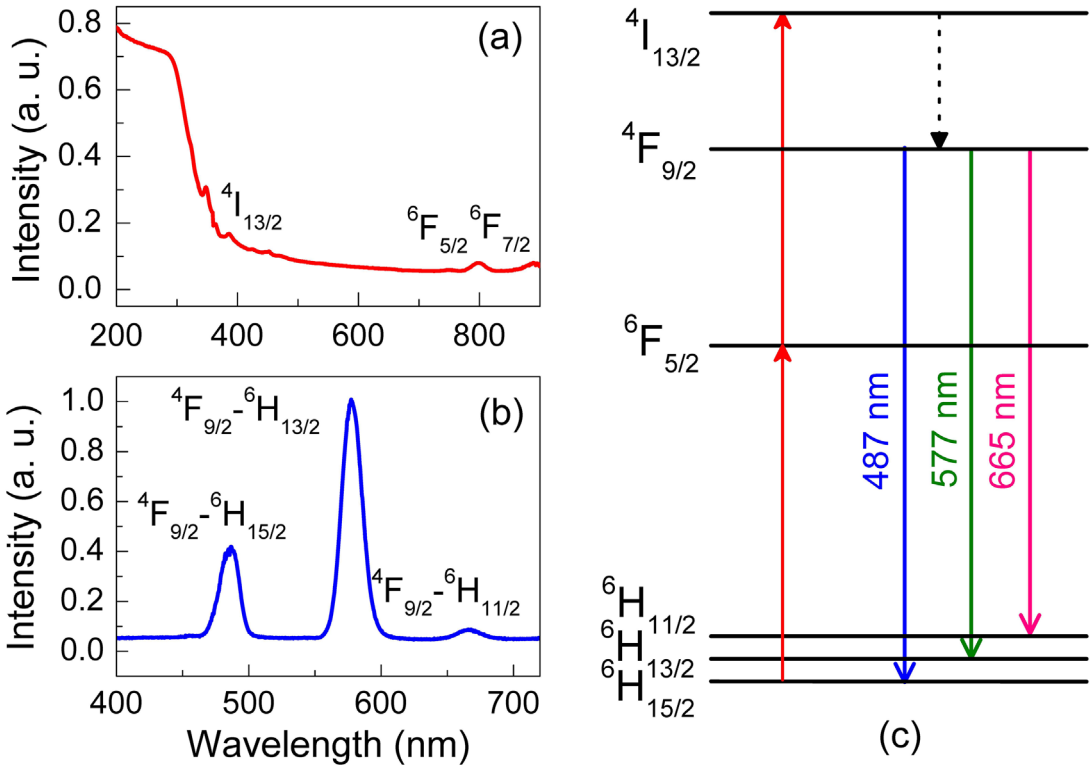

Figure 5. The UV–VIS–NIR absorption spectrum of Dy^{3+}-doped glass sample (a), the up-conversion luminescence spectrum in the visible light region under the 800 nm femtosecond laser pulse excitation (b), and the energy level diagram of Dy^{3+} ion and the up-conversion luminescent processes (c).

polarization from linear through elliptical to circular and vice-versa. The shaped femtosecond laser pulse is focused into the Dy^{3+}-doped glass sample with a lens of 50 mm focal length, and the laser intensity at the focus is estimated to be about 8×10^{12} W cm^{-2}. All the up-conversion luminescence signals are perpendicularly collected by a telescope system and measured by a spectrometer with charge-coupled device (CCD). In our experiment, the glass sample is prepared with the composition in the mol% of $60SiO_2/20Al_2O_3/20CaF_2/1DyF_3$. These high purity raw materials are mixed homogeneously and melted at 1400 °C for 30 min in a covered corundum crucible in air. The melted products are poured onto a cold brass plate, and then pressed by another plate. Finally, the synthetic glass sample is polished for optical measurement after cooling down [28].

The UV–VIS–NIR absorption spectrum of the Dy^{3+}-doped glass sample is shown in figure 5(a). Obviously, two absorption peaks can be observed at the wavelengths of 389 and 800 nm, which are corresponding to the state transitions of $^6H_{15/2} \rightarrow {}^4I_{13/2}$ and $^6H_{15/2} \rightarrow {}^6F_{5/2}$. The up-conversion luminescence spectrum in the visible light region under the femtosecond laser pulse excitation is shown in figure 5(b). One can see that there are three main up-conversion luminescence signals around the wavelengths of 487, 577 and 665 nm, which can be attributed to the transition processes of $^4F_{9/2} \rightarrow {}^6H_{15/2}$, $^4F_{9/2} \rightarrow {}^6H_{13/2}$ and $^4F_{9/2} \rightarrow {}^6H_{11/2}$. On the basis of the

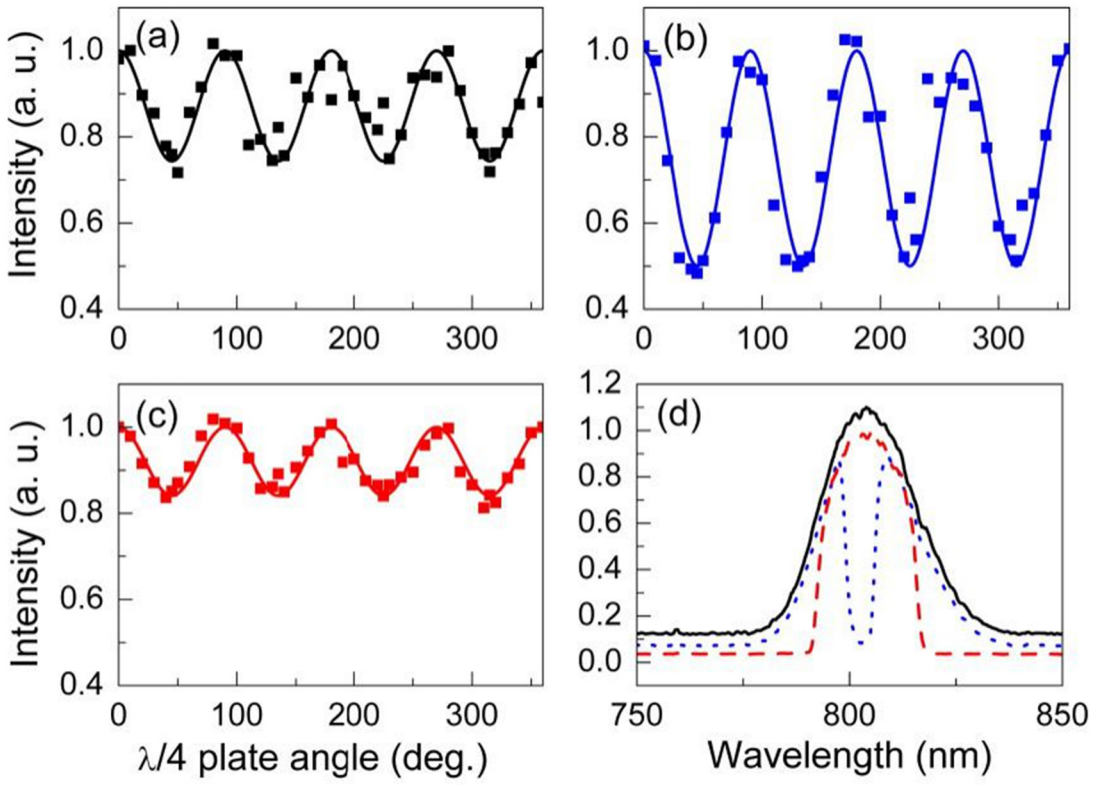

Figure 6. The up-conversion luminescence intensity by varying the $\lambda/4$ plate angle θ for the unmodulated spectrum (a), the modulated spectrum with the central frequency components being cut off (b) and the modulated spectrum with both the low and high frequency components being cut off (c), together with the above three corresponding laser spectra (d).

absorption and luminescence spectra, the excitation and up-conversion luminescence processes are shown in figure 5(c), together with the energy level diagram of Dy^{3+} ions. Under the femtosecond laser field excitation, the population in the ground state $^6H_{15/2}$ is pumped to the final excited state $^4I_{13/2}$ via the intermediate state $^6F_{5/2}$. That is to say, the excitation process is the resonance-mediated two-photon absorption. The population in the excited state $^4I_{13/2}$ can spontaneously relax to the lower excited state $^4F_{9/2}$, and emits the up-conversion fluorescence via the decay to the three states $^6H_{15/2}$, $^6H_{13/2}$ and $^6H_{11/2}$ (see figure 5(b)). Since these up-conversion luminescence signals result from the population of the final excited state $^4I_{13/2}$, the up-conversion luminescence intensity can reflect the transition probability of the resonance-mediated two-photon absorption in the excited state $^4I_{13/2}$.

Figure 6 shows the polarization control behaviors of the up-conversion luminescence intensity in Dy^{3+}-doped glass sample under the excitations of three different modulated femtosecond spectral frequency components. Here, the three different femtosecond spectral frequency components are given in figure 6(d), including the unmodulated laser spectrum and the modulated laser spectrum with the central frequency components or both the low and high frequency components being cut off. Here, the central frequency components are cut off with wavelength range of 797–809 nm, and both the low and high frequency components are cut off with the wavelengths larger than 814 nm and smaller than 793 nm. As can be seen, the polarization control efficiency is about 24% with the excitation of unmodulated femtosecond laser spectrum (see figure 6(a)). However, the polarization control efficiency can be increased to about 48% with the central spectral frequency components being cut off, (see figure 6(b)), while is decrease to about 18% with both the low and high spectral frequency components being cut off (see figure 6(c)). On the basis of above theoretical analysis, the experimental observations can be intuitively explained as follows. When the central spectral frequency components are blocked, the two-photon absorption mainly comes from the contribution of the near-resonant part, and the polarization control efficiency will increase. However, the two-photon absorption mainly results from the excitation of the on-resonant part when both the low and high spectral frequency components are blocked, and thus the polarization control efficiency will decrease. It is obvious that the experimental results are consistent with the theoretical predictions. Therefore, one can make such a conclusion that combining the laser polarization modulation and spectral frequency modulation can provide an efficient method to manipulate the control efficiency of the resonance-mediated two-photon absorption.

4. Conclusions

In conclusion, we have proposed a new method to artificially manipulate the polarization control efficiency of resonance-mediated two-photon absorption by varying the femtosecond spectral frequency components. Our theoretical and experimental results showed that the on-resonant part in the

resonance-mediated two-photon absorption mainly depends on the excitation of the central spectral frequency components, which is independent of the laser polarization, while the near-resonant part mainly come from the contribution of the low and high spectral frequency components, which will be affected by the laser polarization, and therefore the polarization control efficiency of the resonance-mediate two-photon absorption can be increased or decreased by properly designing the femtosecond spectral frequency components. These theoretical and experimental studies present a clear physical insight for the polarization control efficiency manipulation in the resonance-mediated two-photon absorption, which are very meaningful for further controlling and understanding the resonance-mediated multi-photon absorption process, and also can open a new way to various related application areas.

Acknowledgments

This work was partly supported by Program of Introducing Talents of Discipline to Universities (B12024), National Natural Science Foundation of China (No. 11474096), Science and Technology Commission of Shanghai Municipality (No. 14JC1401500 and No. 16520721200), and Higher Education Key Program of He'nan Province of China (No. 17A140025).

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