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Simulating resonance-mediated two-photon absorption enhancement in rare-earth ions by a rectangle phase modulation*

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Improving the up-conversion luminescence efficiency of rare-earth ions via the multi-photon absorption process is crucial in several related application areas. In this work, we theoretically propose a feasible scheme to enhance the resonance-mediated two-photon absorption in Er^{3+} ions by shaping the femtosecond laser field with a rectangle phase modulation. Our theoretical results show that the resonance-mediated two-photon absorption can be decomposed into the on-resonant and near-resonant parts, and the on-resonant part mainly comes from the contribution of laser central frequency components, while the near-resonant part mainly results from the excitation of low and high laser frequency components. So, the rectangle phase modulation can induce a constructive interference between the two parts by properly designing the modulation depth and width, and finally realizes the resonance-mediated two-photon absorption enhancement. Moreover, our results also show that the enhancement efficiency of resonance-mediated two-photon absorption depends on the laser pulse width (or laser spectral bandwidth), final state transition frequency, and intermediate and final state absorption bandwidths. The enhancement efficiency modulation can be attributed to the relative weight manipulation of on-resonant and near-resonant two-photon absorption in the whole excitation process. This study presents a clear physical insight for the quantum control of resonance-mediated two-photon absorption in the rare-earth ions, and there will be an important significance for improving the up-conversion luminescence efficiency of rare-earth ions.

Keywords: coherent quantum control, femtosecond pulse shaping, two-photon absorption, rare-earth ions

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1. Introduction

The up-conversion luminescence of rare-earth ions is a nonlinear optical phenomenon, where two or multiple photons are simultaneously absorbed to generate a new photon. Therefore it was considered to be a well-established technique to realize the photon conversion from low to high frequency. So far, the up-conversion luminescence of rare-earth ions has attracted considerable attention due to its unique optical properties, such as photo-stability, narrow spectrum, near infrared excitation, large Stokes shift, long luminescence lifetime, and well-defined emission bands.[1,2] In recent years, the rare-earth ion doped luminescent materials have been successfully applied in many related areas, such as laser sources,[3,4] fiber optic communications,[5,6] color displays,[7,8] biolabeling, and biomedical sensing.[9–12]

Controlling the up-conversion luminescence of rare-earth ions is very important for further understanding its dynamical process and extending its applications. Recently, several research groups have devoted to the up-conversion luminescence manipulation in rare-earth ion doped luminescent materials, and proposed a lot of schemes to enhance, suppress, or tune the up-conversion luminescence intensity. Basically, these schemes can be divided into two types. One is to use the chemical method, such as controlling chemical composition,[13] crystal structure,[14] nanoparticle size,[15] and surface groups.[16] The other one is using the physical method, such as varying electric field,[17] magnetic field,[18] temperature,[19] laser wavelength,[20] laser pulse duration,[21] and laser repetition.[22] Moreover, we developed a femtosecond pulse shaping technique to effectively manipulate the up-conversion luminescence of rare-earth ions. For example, the red and green up-conversion luminescence in Er^{3+}-doped nanocrystals can be tuned by a square phase modulation.[23] The control efficiency of up-conversion luminescence by a π phase step modulation in Er^{3+}-doped glass will be affected by the laser repetition rate.[24] The up-conversion luminescence in Dy^{3+}-doped glass can be effectively suppressed by both the laser polarization and phase modulation,[25] and the laser po-

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larization will affect the control efficiency by the phase modulation.

The improvement of up-conversion luminescence efficiency is always an important topic for scientists. In this work, we theoretically propose a rectangle phase modulation scheme to enhance the resonance-mediated two-photon absorption in Er^{3+} ions. We show that, by properly designing the depth and width of rectangle phase modulation, the resonance-mediated two-photon absorption can be effectively enhanced, and the absorption enhancement efficiency depends on the laser pulse width (or spectral bandwidth), final state transition frequency, and intermediate and final state absorption bandwidths. Moreover, we also show that the absorption enhancement can be well explained by considering the constructive interference between on-resonant and near-resonant two-photon excitation pathways, and the enhancement efficiency modulation can be attributed to the relative weight manipulation of on-resonant and near-resonant two-photon absorption in the whole excitation process. Here, we establish a theoretical model of resonance-mediated two-photon absorption, and present the physical control mechanism of two-photon absorption enhancement in Er^{3+} ions, which is very helpful for improving the up-conversion luminescence efficiency of rare-earth ions in the future experimental design.

2. Theoretical model

The up-conversion excitation process via the resonance-mediated two-photon absorption in Er^{3+} ions is shown in Fig. 1(a), where these states $^4I_{15/2}$, $^4I_{11/2}$, and $^4F_{7/2}$ represent the ground state $|g\rangle$, intermediate state $|i\rangle$, and final state $|f\rangle$, respectively. Under the femtosecond laser field excitation, the population in the ground state $|g\rangle$ can be pumped to the final state $|f\rangle$ via the intermediate state $|i\rangle$, i.e., a resonance-mediated two-photon absorption. Finally, the population in the final state $|f\rangle$ can relax to the three lower states $^2H_{11/2}$, $^4S_{3/2}$, and $^4F_{9/2}$, and emit the up-conversion fluorescence signals via the three transition processes of $^2H_{11/2} \rightarrow {}^4I_{15/2}$, $^4S_{3/2} \rightarrow {}^4I_{15/2}$, and $^4F_{9/2} \rightarrow {}^4I_{15/2}$. If the femtosecond laser spectral bandwidth is larger than the intermediate state absorption bandwidth, the resonance-mediated two-photon absorption can be decomposed into two parts, i.e., on-resonant and near-resonant two-photon absorption. In the on-resonant two-photon absorption process, the population in the ground state $|g\rangle$ is pumped to the intermediate state $|i\rangle$ by absorbing one photon, and then is further pumped to the final state $|f\rangle$ by absorbing the other one photon. However, in the near-resonant two-photon absorption process, the population in the ground state $|g\rangle$ is directly pumped to the final state $|f\rangle$ by simultaneously absorbing two photons without via the intermediate state $|i\rangle$. Considering the low femtosecond laser field excitation with the perturbation region, the resonance-mediated two-photon transition probability based on the second order perturbation theory can be approximated as[26]

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

where $A(\omega_\mathrm{i})$ and $A(\omega_\mathrm{f})$ are the absorption line-shape functions of immediate state $|i\rangle$ and final state $|f\rangle$, and ω_i and ω_f are the resonant transition frequencies of $|g\rangle \rightarrow |i\rangle$ and $|g\rangle \rightarrow |f\rangle$. By transforming Eq. (1), the two-photon transition probability in the frequency domain can be further written as

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

with

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

and

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

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 $\Phi(\omega)$ represent the spectral amplitude and phase, respectively. Here, $E_0(\omega)$ is defined by the function of $E_0(\omega) = \exp[-\ln 4(\omega\omega)^2/(\omega)^2]$, where ω is the laser central frequency and ω is the laser spectral width. It is obvious that the on-resonant term $A_{\mathrm{On-Res.}}$ in Eq. (3) shows the interference of all resonant two-photon excitation pathways with the transition frequencies of ω_i and $\omega_\mathrm{f}\omega_\mathrm{i}$. However, the near-resonant term $A_{\mathrm{Near-Res.}}$ in Eq. (4) represents the interference of all other two-photon excitation pathways with the transition frequencies of ω and $\omega_\mathrm{f}\omega$.

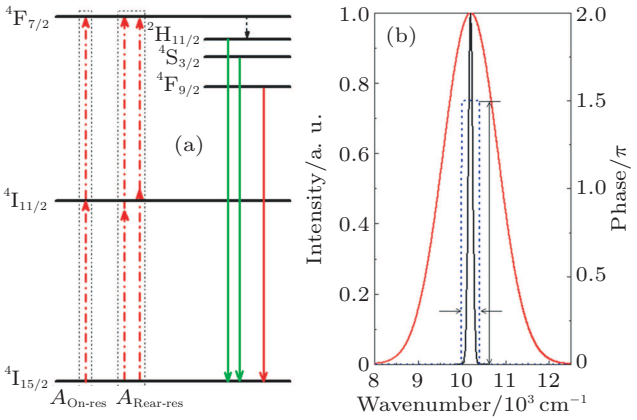


Fig. 1. (color online) (a) The energy level diagram of Er^{3+} ions and up-conversion excitation processes by on-resonant and near-resonant two-photon absorption. (b) The femtosecond laser spectrum (red line) modulated by a rectangle phase modulation, together with the absorption spectrum of intermediated state (black line).

The femtosecond laser phase modulation strategy has shown to be an effective method to manipulate the destructive or constructive interference of different excitation pathways in atomic or molecular system.[27,28] For example, Gandman *et al.* utilized a π phase modulation to realize the enhancement of resonance-mediated $(2+1)$ three-photon absorption[29,30] Here, we propose a rectangle phase modulation scheme to enhance the resonance-mediated two-photon absorption in Er^{3+} ions, and this simple phase modulation is schematically shown in Fig. 1(b), together with the femtosecond laser spectrum and intermediated state absorption spectrum. Here, the two parameters α and β represent the depth and width of rectangle phase modulation, respectively. In this phase modulation, the phase of those frequency components inside the modulation width β is set to be the modulation depth α, where α can be tuned from 0 to 2π. On the basis of Eqs. (3) and (4), it can be seen that the rectangle modulation depth α and width β will change the phase term of $\Phi(\omega_f-\omega)+\Phi(\omega)$, and finally affect the two-photon transition probability S. Based on the previous experimental reports,[31,32] the modeling parameters in our theoretical calculation are set as follows. The transition frequencies of intermediate and final states are $\omega_i = 10200\ \mathrm{cm}^{-1}$ (corresponding to 980 nm) and $\omega_f = 20700\ \mathrm{cm}^{-1}$ (corresponding to 483 nm), and both the two state absorption bandwidths (FWHM) are $\Delta(\omega_i) = \Delta(\omega_f) = 100\ \mathrm{cm}^{-1}$. Therefore, the central frequency of femtosecond laser field is also set to be $\omega_0 = 10200\ \mathrm{cm}^{-1}$, corresponding to the transition frequency ω_i, and the laser pulse width (FWHM) is $\tau = 10$ fs. Here, in order to avoid other up-conversion excitation process, the laser repetition rate is set to be less than 1 kHz, thus the laser pulse separation of 1 ms is far smaller than the excited state lifetime of Er^{3+} ions in the range of microseconds. In addition, the central position of the rectangle shape is fixed at $\omega_0 = \omega_i = 10200\ \mathrm{cm}^{-1}$ during all the simulations. For the femtosecond laser field with Gaussian shape, the laser spectral bandwidth $\Delta\omega$ and the laser pulse width τ meet such a relationship of $\tau\Delta\omega = 4\ln 2$. Obviously, the smaller laser pulse width is corresponding to the larger laser spectral bandwidth. In the following discussion, if not specified, these parameters are considered to be the default.

3. Results and discussion

It can be seen from Eqs. (3) and (4) that, for the transform-limited (TL) femtosecond laser field (i.e., $\Phi(\omega) = 0$), the on-resonant term $A_{\mathrm{On-Res.}}$ is an imaginary number (see Eq. (3)), while the near-resonant term $A_{\mathrm{Near-Res.}}$ is a real number (see Eq. (4)). Therefore the on-resonant and near-resonant terms will not occur the destructive or constructive interference. However, the on-resonant term $A_{\mathrm{On-Res.}}$ mainly comes from the contribution of the laser central frequency components, while the near-resonance term $A_{\mathrm{Near-Res.}}$ mainly results from the excitation of low and high laser frequency components. Thus, the rectangle phase modulation can create the constructive interference between the on-resonant and near-resonant two-photon excitation pathways by properly designing the modulation depth α and width β, and finally realize the resonance-mediated two-photon absorption enhancement. But the absorption enhancement efficiency depends on the relative weight of on-resonant and near-resonant two-photon absorption in the whole excitation process. Typically, the smaller relative weight difference will yield the larger enhancement efficiency. In the rare-earth ion doped luminescent materials, the state absorption bandwidth and transition frequency of rare-earth ions are usually affected by the matrix material composition and preparation method, such as crystal field, spin–orbit coupling, and host lattice.[33] Considering the various situations in the future experiment, next we mainly discuss and analyze the effects of energy level structure of Er^{3+} ions and femtosecond laser parameters on the maximal absorption enhancement, including the femtosecond laser pulse width (or spectral bandwidth), final state absorption bandwidth and transition frequency, and intermediate state absorption bandwidth.

We first study the dependence of TL-normalized maximal population of final state $P_f^{\max}$ on the femtosecond laser pulse width, and the calculated result is shown in Fig. 2, together with the corresponding optimal modulation depth α and width β. Here, the TL-normalized maximal population $P_f^{\max}$ is obtained by changing both the rectangle modulation depth α and width β, and therefore the TL-normalized maximal population can directly reflect the two-photon absorption enhancement efficiency. Hereafter, the same method is performed. One can see from Fig. 2(a) that, with the increase of laser pulse width from 10 to 80 fs, the maximal population $P_f^{\max}$ will decrease, and finally approach 1, i.e., no absorption enhancement. As shown in Eqs. (2)–(4), the resonance-mediated two-photon absorption involves two parts, i.e., on-resonant and near-resonant two-photon absorption, and the relative weight of two parts in the whole excitation process depends on the laser spectral bandwidth (i.e., laser pulse width). If the laser spectral bandwidth is smaller than the intermediate state absorption bandwidth, the whole excitation process is dominated by the on-resonant two-photon absorption. In this case, the two-photon absorption cannot be enhanced by this rectangle phase modulation. However, when the laser spectral bandwidth is larger than the intermediate state absorption bandwidth, the near-resonant two-photon absorption will occur, and its weight in the whole excitation process will increase with the increase of the laser spectral bandwidth. Thus, the two-photon absorption can be enhanced due to the constructive interference between the on-resonant and near-resonant excitation pathways by controlling the modulation depth α and width β, and the absorption enhancement efficiency will increase with the in-

crease of laser spectral bandwidth due to the weight increase of near-resonant two-photon absorption. Moreover, as shown in Fig. 2(b), with the increase of the laser pulse width, the optimal modulation depth α and width β show an opposite evolution behavior. The larger (or smaller) modulation depth combining with the smaller (or larger) modulation width can induce the maximal absorption enhancement (i.e., maximal population).

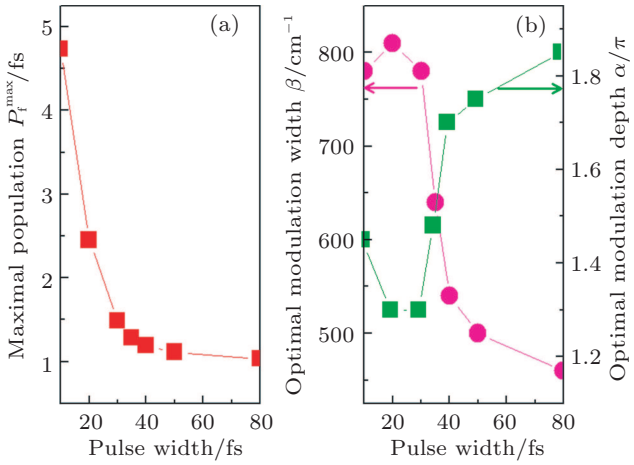


Fig. 2. (color online) (a) The TL-normalized maximal population P_f^{max} as the function of the laser pulse width, and (b) the corresponding optimal modulation depth α and width β.

Next, we further demonstrate the effect of final state transition frequency on the maximal two-photon absorption enhancement. Figure 3 shows the TL-normalized maximal population P_f^{max} by varying the final state transition frequency, and the optimal modulation depth α and width β are also given. Considering that the final state transition frequency of Er^{3+} ions will not significantly change in the actual experiment, here we consider the transition frequencies from 20400 to 21000 cm^{-1}. As shown in Fig. 3(a), with the increase of transition frequency, the maximal population P_f^{max} increases. Here, the laser frequency is 10200 cm^{-1}, and thus the two-photon absorption frequency is 20400 cm^{-1}. With the increase of final state transition frequency from 20400 to 21000 cm^{-1}, the detuning between the two-photon absorption frequency and final state transition frequency will increase, and thus the weight of near-resonant two-photon absorption in the whole excitation process will increase. When the weight of near-resonant two-photon absorption is further close to that of on-resonant two-photon absorption, the two-photon absorption enhancement efficiency will increase due to the more complete constructive interference between on-resonant and near-resonant two-photon excitation pathways. The optimal modulation depth α and width β are shown in Fig. 3(b). One can see that, with the increase of final state transition frequency, the optimal modulation width β monotonously increases, but the optimal modulation depth α shows a rapid increase and then slow decrease process. Actually, the intermediate state transition frequency will also change in the actual experiment, which is the same as the final state, but here we set the laser central frequency equal to the intermediate transition frequency. In this case, the maximal population modulation behavior should be the same as that by varying the final state transition frequency, and therefore here we do not repeat this discussion.

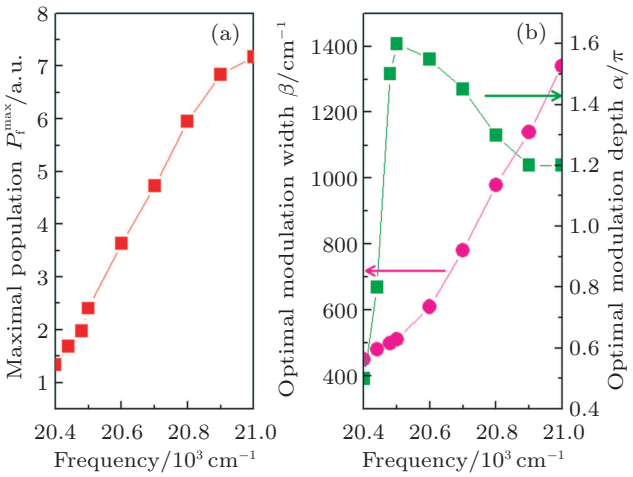


Fig. 3. (color online) (a) The TL-normalized maximal population P_f^{max} by varying the final state transition frequency, and (b) the corresponding optimal modulation depth α and width β.

Finally, we analyze the influence of final or intermediate state absorption bandwidth on the maximal absorption enhancement of final state. Figure 4 shows the TL-normalized maximal population P_f^{max} with the increase of final state absorption bandwidth, together with the optimal modulation depth α and width β. It can be seen that the maximal population P_f^{max} decreases with the increase of final state absorption bandwidth from 50 to 300 cm^{-1}. The control behavior can also be explained by considering the constructive interference of on-resonant and near-resonant two-photon excitation pathways. In the smaller absorption bandwidth of 50 cm^{-1}, the weight of on-resonant two-photon absorption in the whole excitation process is still larger than that of near-resonant two-photon absorption. When the final state absorption bandwidth increases from 50 to 300 cm^{-1}, the weight of on-resonant two-photon absorption will further increase. In the larger absorption bandwidth of 300 cm^{-1}, the on-resonant two-photon absorption may almost dominate the whole excitation process, and thus the enhancement efficiency of two-photon absorption will be greatly suppressed. Here, with the increase of final state absorption bandwidth, the optimal modulation width β increases, but the optimal modulation depth α almost keeps a constant, which is around 1.4π, as shown in Fig. 4(b). The similar evolution behavior can be found by varying the intermediate state absorption bandwidth, including the maximal population P_f^{max} and optimal modulation depth α and width β, as shown in Fig. 5. Similarly, the decrease of absorption enhancement efficiency is also due to the further increase in the weight of on-resonant two-photon absorption in the whole ex-

citation process when the intermediate state absorption bandwidth increases.

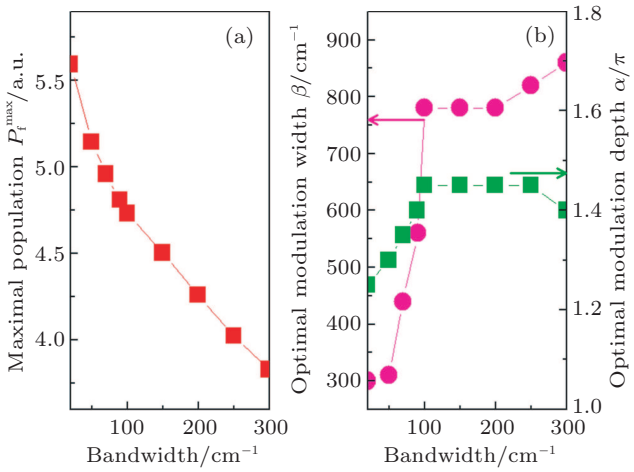


Fig. 4. (color online) (a) The TL-normalized maximal population P_f^{max} with the increase of final state absorption bandwidth, and (b) the corresponding optimal modulation depth α and width β.

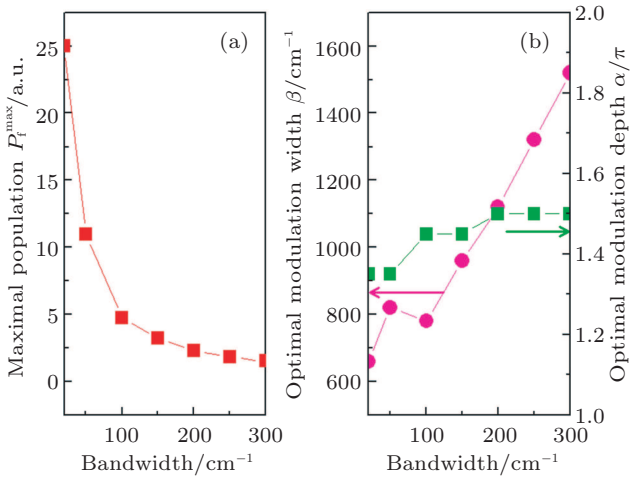


Fig. 5. (color online) (a) The TL-normalized maximal population P_f^{max} with the increase of intermediate state absorption bandwidth, and (b) the corresponding optimal modulation depth α and width β.

As shown above, the enhancement efficiency of resonance-mediated two-photon absorption will be affected by the final state absorption bandwidth and transition frequency, intermediate state absorption bandwidth, and femtosecond laser pulse width (or laser spectral bandwidth), but all these physical control mechanisms can be attributed to the relative weight manipulation of on-resonant and near-resonant two-photon absorption in the whole excitation process. In the future study, one can design the experiment from two aspects to improve the enhancement efficiency of resonance-mediated two-photon absorption in Er^{3+}-ion doped luminescent materials. One is to control the state transition frequency and absorption bandwidth of Er^{3+} ions by designing the matrix material structure and property, and the other one is to change the laser parameters by properly selecting the femtosecond laser device.

4. Conclusion

We have theoretically proposed a scheme to enhance the resonance-mediated two-photon absorption in Er^{3+} ions by shaping the femtosecond laser field with a rectangle phase modulation. The results showed that, by properly designing the rectangle phase modulation depth and width, the two-photon absorption can be enhanced due to the constructive interference of on-resonant and near-resonant two-photon excitation pathways, and the enhancement efficiency depends on the laser pulse width (or laser spectral bandwidth), final state transition frequency, and intermediate and final state absorption bandwidths. Furthermore, the results also showed that the two-photon absorption enhancement efficiency modulation can be attributed to the relative weight manipulation of on-resonant and near-resonant two-photon absorption in the whole excitation process. In this work, the rectangle phase modulation has shown to be a well-established method to enhance the resonance-mediated two-photon absorption in Er^{3+} ions. Similarly, this rectangle phase modulation scheme can also be further extended to realize the resonance-mediated multi-photon absorption enhancement in various rare-earth ions. Furthermore, the phase modulation method employed here may be extended to other related research fields, including coherent population trapping (CPT) and stimulated Raman adiabatic passage (STIRAP). Therefore, we believe that this study can open a new opportunity for the improvement of up-conversion luminescence efficiency in the rare-earth ion doped luminescent materials.

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