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Simulating the resonance-mediated (1 + 2)-three-photon absorption enhancement in Pr^{3+} ions by a rectangle phase modulation

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Enhancing the upconversion luminescence of rare earth ions is crucial for their applications in the laser sources, fiber optic communications, color displays, biolabeling, and biomedical sensors. In this paper, we theoretically study the resonance-mediated (1 + 2)-three-photon absorption in Pr^{3+} ions by a rectangle phase modulation. The results show that the resonance-mediated (1 + 2)-three-photon absorption can be effectively enhanced by properly designing the depth and width of the rectangle phase modulation, which can be attributed to the constructive interference between on-resonant and near-resonant three-photon excitation pathways. Further, the enhancement efficiency of resonance-mediated (1 + 2)-three-photon absorption can be affected by the pulse width (or spectral bandwidth) of femtosecond laser field, final state transition frequency, and absorption bandwidths. This research can provide a clear physical picture for understanding and controlling the multi-photon absorption in rare-earth ions, and also can provide theoretical guidance for improving the up-conversion luminescence.

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

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1. Introduction

The rare-earth ions doped materials have unique optical properties, and have been used in laser sources,[1] fiber optic communications,[2] color displays,[3] biolabeling and biomedical sensors.[4,5] The Pr^{3+} ion has relatively wide absorption band in the vis-NIR regions, and has been used as function photonic materials like fiber optical communication,[6,7] lasers,[8,9] LEDs,[10] field-emission display or photoluminescence devices.[11] The Pr^{3+} ion is an important activator with several meta-stable states, which offer stimulated emissions in blue, green, red and infrared regions.[12] In order to improve the luminescence efficiency of Pr^{3+} ions, many researchers have made great effects to propose some different schemes. Han *et al.* have increased fluorescence to 27-fold in comparison to $K_3Lu(PO_4)_2$: Pr^{3+} by introducing the Gd^{3+} ions into the lattice.[13] Gates *et al.* have enhanced visible-to-ultraviolet upconversion in the phosphor Y_2SiO_5:Pr^{3+} by cooping Li^+ ion.[14] The red emission in $NaGdTiO_4$:Pr^{3+} can be increased with the increasing temperature from 77 K to 350 K.[15] Moreover, Tian *et al.* have observed that ferroelectric remanent polarization remarkably enhances the photoluminescence intensity of Pr^{3+} -doped $(Bi_{0.5}Na_{0.5})TiO_3$ ceramic samples.[16] However, these methods for enhancing luminescence efficiency of rare-earth-doped materials are still limited.

The shaped femtosecond pulse is an effective method to manipulate the multi-photon absorption by modulating the constructive or destructive interference between different excitation pathways in atoms,[17,18] molecules,[19,20] and condensed state system.[21] For example, the shaped pulse with π phase modulation has been used to enhance or suppress the (1 + 1) resonance-mediated two-photon absorption in Rb atom and (2 + 1)-resonance-mediated three-photon absorption in Na atom.[18,22] Zhang *et al.* has demonstrated that the single-photon fluorescence of IR 125 can be modulated by the polarization modulation of femtosecond laser field.[23] Recently, with the development of the femtosecond pulse shaping technique, the femtosecond pulse has been changed as almost arbitrary temporal distribution by controlling the laser spectral phase, amplitude and polarization.[24] The upconversion luminescence of rare-earth ions is a nonlinear optical phenomenon, which convert two or multiple low energy photons into one high energy photon via two-photon or multi-photon absorption process. Researchers have proposed some pulse shaping methods to modulate upconversion luminescence of rare earth ions.[25–27] The upconversion luminescence of Dy^{3+} ion doped glass can be enhanced by the intermediated femtosecond laser field with a π phase modulation.[25] The π phase modulation has been used to enhance and suppress the single and two-photon fluorescence in Er^{3+} ions.[26] The green and red up-conversion fluorescence in Er^{3+}-doped $NaYF_4$ nanocrystals can be manipulated by

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square phase modulation.[27] These results illustrate that the shaped femtosecond pulse is an effective tool to modulate the up-conversion luminescence of rare-earth ions.

In this paper, we theoretically study the up-conversion luminescence control behaviors in Pr^{3+} ions by the rectangle phase modulation of the femtosecond laser field. The theoretical results show that the resonance-mediated $(1+2)$-three-photon absorption in Pr^{3+} ions can be effectively enhanced by the rectangle phase modulation, and the absorption enhancement efficiency is correlated with the laser pulse width, final state transition frequency and absorption bandwidths. The absorption enhancement efficiency has been decreased under the lager laser pulse width and energy level absorption bandwidths, while the absorption enhancement efficiency first increases and then decreases with the increase of the final state transition frequency. These research can provide a clear physical picture for understanding and controlling the multi-photon absorption in rare-earth ions, and can provide theoretical guidance for the related experimental works.

2. Theoretical model

Figure 1(a) show the up-conversion luminescence processes of Pr^{3+} ions via the resonance-mediated $(1+2)$-three-photon absorption. We use the ground state $|g\rangle$, intermediate state $|i\rangle$ and final state $|f\rangle$ to represent these states $^3H_4$, $^3F_4$, and $^3P_0$, respectively. For the femtosecond laser field, its spectral bandwidth is always larger than absorption bandwidth of intermediate state $|i\rangle$. The electrons in ground state $|g\rangle$ can be pumped to the final state $|f\rangle$ via the intermediate state $|i\rangle$, which is called the resonance-mediated $(1+2)$-three-photon absorption process. The resonance-mediated $(1+2)$-three-photon absorption includes both on-resonant and near-resonant three-photon absorption. For on-resonant part, the ground state electrons are first pumped to the intermediate state $|i\rangle$ by absorbing one photon with the frequency of $|g\rangle \rightarrow |i\rangle$, and then is further pumped to the final state $|f\rangle$ by absorbing two photons with the photon frequency sum of $|i\rangle \rightarrow |f\rangle$. For near-resonant part, the ground state electrons can absorb three photons and reach the final state $|f\rangle$ without through the intermediate state $|i\rangle$, where the frequency sum of these three photons is equal to the transition frequency of $|g\rangle \rightarrow |f\rangle$. Then, the electrons in the final state $|f\rangle$ can relax to the lower states $^3H_6$, $^3H_5$, and $^3H_4$, and further generate the up-conversion fluorescence via the transition processes of $^3P_0 \rightarrow {}^3H_6$, $^3P_0 \rightarrow {}^3H_5$, and $^3P_0 \rightarrow {}^3H_4$.

Based on theoretical research method of the resonance-mediated $(2+1)$-three-photon absorption in Na atom,[22] we construct the third order perturbation theory to study the resonance-mediated $(1+2)$-three-photon absorption of Pr^{3+} ion under the low femtosecond laser field excitation. The resonance-mediated $(1+2)$-three-photon absorption includes the contribution of on-resonant term $A_{\rm on}$ and near-resonant term $A_{\rm near}$, and can be described in the frequency domain as

$$P_f \propto \int_{-\infty}^{+\infty} \mathrm{d}\omega_f A\left(\omega_{fg}\right) \left|A_{\rm on} + A_{\rm near}\right|^2, \tag{1}$$

with

$$A_{\rm on} \propto \mathrm{i}\pi \int_{-\infty}^{+\infty} \mathrm{d}\omega_{ig} A\left(\omega_{ig}\right) E(\omega_{ig}) A^{(2)}\left(\omega_{fi}\right), \tag{2}$$

$$A_{\rm near} \propto -\wp \int_{-\infty}^{+\infty} \frac{E\left(\omega_{ig}-\delta\right) A^{(2)}\left(\omega_{fi}+\delta\right)}{\delta} \mathrm{d}\delta, \tag{3}$$

where $A^{(2)}(\Omega)$ can be defined as

$$A^{(2)}(\Omega) = \int_{-\infty}^{\infty} E(\omega) E(\Omega-\omega) \mathrm{d}\omega, \tag{4}$$

where the $\wp$ is the Cauchy's principal value, and ω_{ig} and ω_{fg} are the resonant transition frequencies of $|g\rangle \rightarrow |i\rangle$ and $|g\rangle \rightarrow |f\rangle$. $A(\omega_{ig})$ and $A(\omega_{fg})$ represent the absorption functions of the immediate state and final state, which are Gaussian shape. Moreover, $\delta = \omega_{ig} - \omega$ is the detuning from the excited state $|i\rangle$. The femtosecond laser field in frequency domain can be expressed as $E(\omega) = E_0(\omega) \times \exp[\mathrm{i}\Phi(\omega)]$, where $E_0(\omega)$ and $\Phi(\omega)$ represent the spectral amplitude and phase, respectively. Substituting $E(\omega) = E_0(\omega) \times \exp[\mathrm{i}\Phi(\omega)]$ into Eq. (3) and (4), we can obtain

$$A_{\rm on} \propto \mathrm{i}\pi \int_{-\infty}^{+\infty} \mathrm{d}\omega_{ig} A\left(\omega_{ig}\right) E_0(\omega_{ig}) \mathrm{e}^{\mathrm{i}\phi\left(\omega_{ig}\right)} A^{(2)}\left(\omega_{fi}\right), \tag{5}$$

$$A_{\rm near} \propto -\wp \int_{-\infty}^{+\infty} \frac{E_0\left(\omega_{ig}-\delta\right) \mathrm{e}^{\mathrm{i}\phi\left(\omega_{ig}-\delta\right)} A^{(2)}\left(\omega_{fi}+\delta\right)}{\omega_{ig}-\omega} \mathrm{d}\delta, \tag{6}$$

$$A^{(2)}(\Omega) = \int_{-\infty}^{\infty} E_0(\omega) E_0(\Omega-\omega) \mathrm{e}^{\mathrm{i}\phi(\omega)+\mathrm{i}\phi(\Omega-\omega)} \mathrm{d}\omega. \tag{7}$$

In addition, $E_0(\omega)$ can be expressed by the function of $E_0(\omega) = \exp[-\ln 4(\omega-\omega_0)^2/(\Delta\omega)^2]$, ω and $\Delta\omega$ are the central frequency and spectral width of femtosecond laser field.

The phase modulation of femtosecond laser field is an effective method to manipulate the destructive or constructive interference between different excitation pathways.[25–27] Here, a rectangle phase modulation scheme has been proposed to enhance the resonance-mediated $(1+2)$-three-photon absorption in Pr^{3+} ions. Figure 1(b) show the femtosecond laser spectrum (red solid line), the intermediated state absorption spectrum (green dashed line), and the rectangle phase modulation (blue dotted line). As shown in Fig. 1(b), α is depth of the rectangle phase modulation, which can be changed from 0 to 2π, and β is the width of the rectangle phase modulation. When the femtosecond laser pulse is modulated by the rectangle phase modulation, the phase of the laser spectrum under the modulation width β is added the modulation depth α. Based on the experimental reports,[28,29] the Pr^{3+} ions parameters are set as follows in the theoretical calculation. The transition frequency of intermediate state is $\omega_{ig} = 6.97 \times 10^3$ cm^{-1}, and that of final state is $\omega_{fg} = 2.07 \times 10^4$ cm^{-1}. The absorption bandwidth (FWHM) of immediate state and final state

are both 100 cm^{-1}. Moreover, the femtosecond laser field in theoretical calculation is Gaussian shape with the center frequency 6.97×10^3 cm^{-1}, and the laser pulse width τ is 20 fs. The spectral bandwidth $\Delta\omega$ and the pulse width τ can satisfy Fourier transformation: $\tau\Delta\omega = 4\ln 2$. It is obvious that the larger pulse width is corresponding the smaller laser spectral bandwidth.

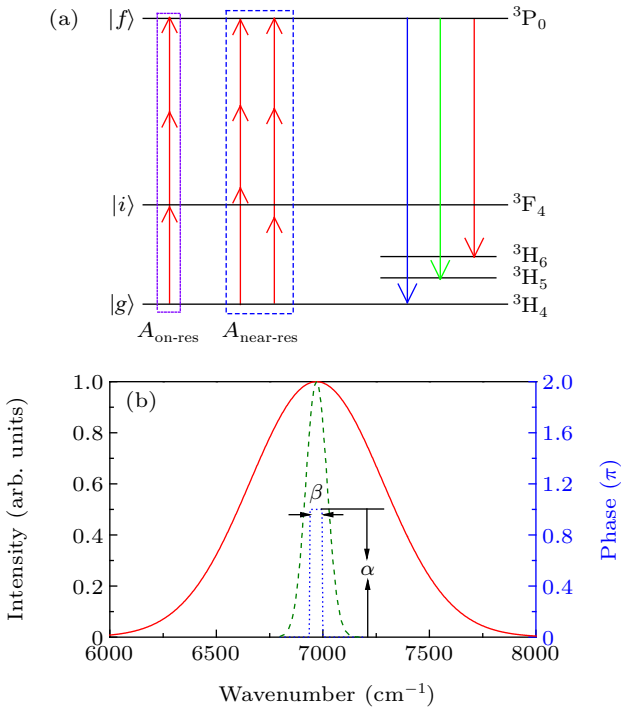


Fig. 1. (a) The resonance-mediated (1+2)-three-photon absorption processes with on-resonant and near-resonant excitation pathways and luminescence processes in Pr^{3+} ions, (b) the femtosecond laser field (red solid line) modulated by a rectangle phase (blue dotted line) with modulation depth α and modulation width β and the absorption spectrum of intermediate state (green dashed line) in Pr^{3+} ions.

As can be seen in Eqs. (5)–(7), the near-resonant term A_{near} is not only related to the opposite sign of $1/(\omega_{ig} - \omega)$ for different laser spectrum components, but also related to the phase terms $e^{i\phi(\omega_{ig}-\delta)}$ and $e^{i\phi(\omega)+i\phi(\Omega-\omega)}$ for different laser frequencies. For the transform-limited (TL) pulse with the phase term $(\omega) = 0$, the on-resonant term A_{on} is an imaginary number, while the near-resonant term A_{near} is a real number. It is clear that the coherent interference cannot occur between the on-resonant and near-resonant transition pathways. Moreover, as can be seen in Eq. (6), the detuning $\delta = \omega_{ig} - \omega$ is positive or negative for the laser frequency composition $\omega < \omega_{ig}$ or $\omega > \omega_{ig}$, and the destructive interference can happen between near-resonant transition pathways. However, for the shaped pulse, the phase can play an important role for the final state population, and the rectangle phase modulation can be used to enhance the final population. In the actual experiment, the matrix material composition and preparation method always affect the absorption bandwidth and energy levels of rare earth ions by the crystal field, spin–orbit coupling and host lattice.[30,31] The energy levels of Ln^{3+} are affected by both Coulomb force and spin–orbit coupling, and the splitting of the energy levels can be induced by the crystal field. Therefore, we discuss the influence of transition frequency and absorption bandwidth of middle state and final state. In addition, the laser parameters also affect the maximal absorption enhancement, including the laser pulse width, the laser intensity and polarization. The middle and strong laser intensity always induce high-order multi-photon process, and different polarizations of the laser field can generate the complex excitation pathways.[32,33] In this paper, we primary focus on resonance-mediated $(1+2)$-three-photon absorption in Pr^{3+} ions, and discuss the effect of the laser pulse width or spectral bandwidth on the maximal absorption enhancement. Therefore, based on different situations in the actual experiment, we further analyze and discuss the influence of the laser parameters (laser pulse width or spectral bandwidth) and energy level structure (final state transition frequency, final state and intermediate state absorption bandwidth) on the maximal absorption enhancement.

3. Results and discussion

First, we study the influence of the femtosecond laser pulse width on the resonance-mediated $(1+2)$-three-photon absorption enhancement in Pr^{3+} ion. For the shaped femtosecond pulse with different pulse widths, the TL-normalized maximal populations of final state $P_{\text{f}}^{\max}$ (black squares), on-resonant (blue triangles), and near-resonant (red circles) absorption are shown in Fig. 2(a), and the corresponding optimal modulation depth α (black circles) and width β (blue squares) are shown in Fig. 2(b). Here, the TL-normalized maximal population $P_{\text{f}}^{\max}$ can be obtained by varying both rectangle phase depth α and width β, and is normalized by the TL pulse excitation. Therefore, the resonance-mediated $(1+2)$-three-photon absorption enhancement efficiency can be reflected by the TL-normalized maximal population $P_{\text{f}}^{\max}$. Moreover, the on-resonant and near-resonant absorption in Fig. 2(a) are also normalized by the TL pulse excitation. Hereafter, the same method is performed. As can be seen in Fig. 2(a), the maximal population $P_{\text{f}}^{\max}$ can be effectively enhance to 8.539 with the laser pulse width of 10 fs. These can be explained as follows. As can be seen in Eqs. (5)–(7), the near-resonant term A_{near} is related to both the opposite sign of $1/(\omega_{ig} - \omega)$ and the phase terms $e^{i\phi(\omega_{ig}-\delta)}$ and $e^{i\phi(\omega)+i\phi(\Omega-\omega)}$ for different laser frequencies. For the TL pulse with the phase $\Phi(\omega) = 0$, the detuning $\delta = \omega_{ig} - \omega$ in Eq. (3) is positive for the laser frequency composition $\omega < \omega_{ig}$ and negative for the laser frequency composition $\omega > \omega_{ig}$, and the destructive interference occur between near-resonant three-photon transition pathways. However, for the shaped pulse with the rectangle phase modulation, the phase $\Phi(\omega)$ is not equal to 0. The rectangle phase

modulation with proper modulation depth and width can realize the sign inversion of $e^{i\phi(\omega_{ig}-\delta)}$ and $e^{i\phi(\omega)+i\phi(\Omega-\omega)}$ in Eqs. (6) and (7), and induce the constructive interference between near-resonant transition pathways, which results in the enhancement of the final population. Moreover, the maximal population P_f^{max} decreases from 8.539 to 1.226 with the laser pulse width increasing from 10 fs to 50 fs in Fig. 2(a). The resonance-mediated $(1+2)$-three-photon absorption includes the contribution of on-resonant and near-resonant parts, and the relative weigh of near-resonant part in the whole excitation process is larger than the on-resonant part. When the laser pulse width increases, the laser spectral bandwidth decreases, which induces the effective near-resonant three-photon transition pathways decrease. The maximal population P_f^{max} decrease with the decrease of the effective near-resonant three-photon absorption. Furthermore, as can be seen in Fig. 2(a), the maximal population P_f^{max} (black squares) is larger than the direct sum of no-resonant (blue triangles) and near-resonant (red circles) absorption. It illustrates that the constructive interference occurs between the on-resonant and near-resonant excitation pathways. In addition, as shown in Fig. 2(b), with the increase of the laser pulse width, the optimal modulation width decreases, and the optimal modulation depth have not greatly changed.

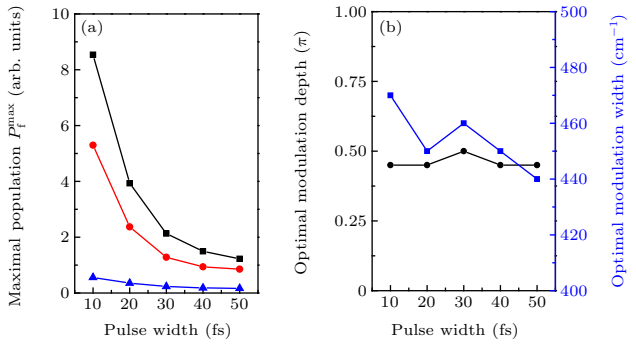


Fig. 2. (a) The TL-normalized maximal population of final state P_f^{max} (black squares), on-resonant (blue triangles) and near-resonant (red circles) absorption with the increase of the laser pulse width, (b) the corresponding optimal depth α (black circles) and width β (blue squares) of the rectangle phase modulation.

In order to demonstrate the dependence of the resonance-mediated $(1+2)$-three-photon absorption enhancement on final state transition frequency, we show the TL-normalized maximal population P_f^{max}, on-resonant and near-resonant absorption by varying the final state transition frequency in Fig. 3(a), together with the optimal modulation depth α (black circles) and width β (blue squares) in Fig. 3(b). As can been seen in Fig. 3(a), when the final state transition frequency increases, the maximal population P_f^{max} first increases and then decreases, and near-resonant absorption mainly shows a decline, while on-resonant absorption continues increasing. In the theoretical calculation, the transition frequencies ω_{ig} and ω_{fg} are 6.97×10^3 cm^{-1} and 2.07×10^4 cm^{-1}. The laser center frequency is 6.97×10^3 cm^{-1}, and the corresponding third-harmonic frequency is 2.091×10^4 cm^{-1}. Therefore, the contribution of on-resonant part in the whole excitation process can increase with the increase of the final state transition frequency, and that of near-resonant part decrease. When the final state transition frequency is close to 2.055×10^4 cm^{-1}, the final state transition frequency have a relatively large deviation to the laser third-harmonic frequency 2.091×10^4 cm^{-1}. The on-resonant absorption is relatively small compared with the near-resonant absorption, the constructive interferences between near-resonant and on-resonant transition pathways is not very effective due to the large numerical difference. With the final state transition frequency increase, the constructive interference between near-resonant and on-resonant transition pathways become more effective, and the maximal population P_{fmax} increases. However, when the final state transition frequency is further increased and close to 2.091×10^4 cm^{-1}, the near-resonant absorption rapidly decreases with slow increase of the on-resonant absorption, which induces the decrease maximal population P_{fmax}. Furthermore, as can be seen in Fig. 3(b), the optimal modulation depth and width monotonously decrease with the increase of the final state transition frequency. Similarly, in the actual experiment, the intermediate state transition frequency may also change, but the laser central frequency can be set to be equal to the intermediate transition frequency ω_{ig} in the experiment and theoretical calculation. Therefore, the maximal population P_f^{max} by varying the intermediate transition frequency should have similar behaviors with that by varying the final state transition frequency, and we will not discuss it again in this paper.

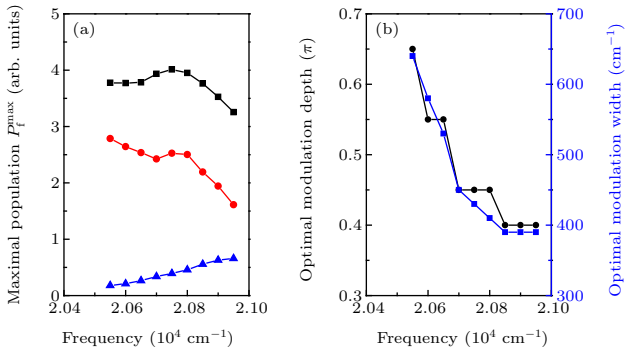


Fig. 3. (a) The TL-normalized maximal population of final state P_f^{max} (black squares) by changing the final state transition frequency, and together with the on-resonant (blue triangles) and near-resonant (red circles) absorption; (b) the corresponding optimal depth α (black circles) and width β (blue squares) of the rectangle phase modulation.

Finally, we further demonstrate the influence of the final state absorption bandwidth on resonance-mediated $(1+2)$-three-photon absorption enhancement. Figure 4(a) shows the TL-normalized maximal population P_f^{max}, on-resonant and near-resonant absorption by varying the final state absorption bandwidth. Figure 4(b) shows the corresponding optimal modulation depth α (black circles) and width β (blue

squares). When the final state absorption bandwidth changes from 10 cm^{-1} to 300 cm^{-1}, the maximal population P_f^{max} monotonically decreases. This can be explained as follows. The relative weight of near-resonant part is still larger than that of on-resonant part in the whole excitation processes. However, the coherent interference between near-resonant transition pathways is attenuated with the increase of the final absorption bandwidth, and thus the maximal population P_f^{max} has been greatly suppressed. Moreover, as shown in Fig. 4(b), the corresponding optimal modulation width and depth almost remain unchanged with the increase of the final state absorption bandwidth. In addition, with the increasing of the intermediate state absorption bandwidth, the contribution of on-resonant part in the whole excitation processes increases, the relative weigh of near-resonant for the final state population decreases. The contribution of near-resonant part in the whole excitation processes still plays a main role for the final state population, and the maximal population P_f^{max} decreases with the increasing of the intermediate state absorption bandwidth. Therefore, the TL-normalized maximal population P_f^{max} has the same evolution behavior by increasing the absorption bandwidth of intermediate state or final state.

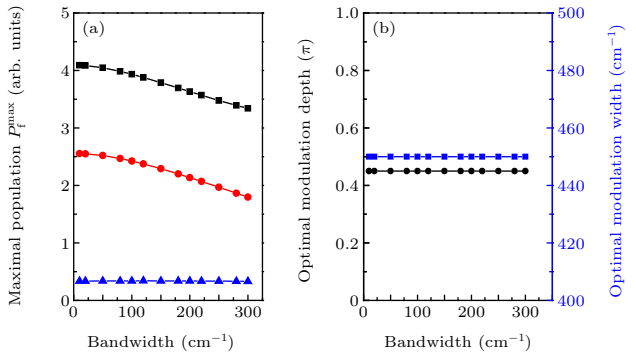


Fig. 4. (a) The TL-normalized maximal population P_f^{max} (black squares) as a function of the final state absorption bandwidth, and together with on-resonant (blue triangles) and near-resonant (red circles) absorption, (b) the corresponding optimal depth α (black circles) and width β (blue squares) of the rectangle phase modulation.

Obviously, femtosecond laser pulse width, final state transition frequency, and absorption bandwidth can affect the maximal population P_f^{max} of the resonance-mediated $(1+2)$-three-photon absorption. These results can provide us with new solutions to improve the enhancement efficiency of the resonance-mediated $(1+2)$-three-photon absorption in Pr^{3+} ion-doped material. One can design the matrix material structure and property to control the state transition frequency and absorption bandwidth, and also can use different femtosecond laser devices to select the proper laser parameters. Moreover, the phase modulation is unusual routes to tune or enhance the up-conversion luminescence of rare-earth ions in the real-time, dynamical and reversible manners, which is beneficial for many widespread applications of rare-earth ions, especially for biomedical applications. Based the requirements of the practical applications, the upconversion luminescence intensity of rare-earth ions can be manipulated flexibly and quickly by the phase modulation. In addition, the peak intensity of the shaped pulses is weaker than the TL-pulse, which has less photo-damage to living organisms and samples.

4. Conclusion

In this paper, we have theoretically used a rectangle phase modulation to enhance the resonance-mediated $(1+2)$-three-photon absorption in the Pr^{3+} ions. The results show that the resonance-mediated $(1+2)$-three-photon absorption has been effectively enhanced by the rectangle phase modulation due to the constructive interference between on-resonant and near-resonant absorption pathways. The TL-normalized maximal population P_f^{max} is related to the laser pulse width (or laser spectral bandwidth), final state transition frequency, and absorption bandwidths. These studies not only can provide a clear physical picture for understanding and controlling the multi-photon absorption in rare-earth ions, but also can provide theoretical guidance for the related experimental works.

Acknowledgments

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References

[1] Nilsson J, Clarkson W A, Selvas R, Sahu J K, Turner P W, Alam S U and Grudinin A B 2004 *Opt. Fiber. Technol.* **10** 5
[2] Tessler N, Medvedev V, Kazes M, Kan S and Banin U 2002 *Science* **295** 1506
[3] Li Y, Zhang J, Luo Y, Zhang X and Hao Z 2011 *J. Mater. Chem.* **21** 2895
[4] Nyk M, Kumar R, Ohulchanskyy T Y, Bergey E J and Prasad P N 2008 *Nano. Lett.* **8** 3834
[5] Gai S, Li C, Yang P and Lin J 2014 *Chem. Rev.* **114** 2343
[6] Chen H, Xia K, Liu Z, Wang X, Zhang X, Xu Y and Dai S 2019 *Chin. Phys. B* **28** 024209
[7] Simons D R, Faber A J and Waal H 1995 *J. Non-Cryst. Solids.* **185** 283
[8] Nikl M, Ren G, Ding D, Mihokova E, Jary V and Feng H 2010 *Chem. Phys. Lett.* **493** 72
[9] Pidol L, Viena B, Kahn-Harari A, Bessiere A and Dorenbos P 2005 *Nucl. Instrum. Methods Phys. Res.* **537** 125
[10] Birkhahn R, Garter M and Steckl A 1999 *Appl. Phys. Lett.* **74** 2161
[11] Okamoto S, Kobayashi H and Yamamoto H 1999 *J. Appl. Phys.* **86** 5594
[12] Kaminskii A A 1990 *Laser Crystals*, 2nd edn., (Berlin: Springer-Verlag)
[13] Han X, Wu J, Hu C, Yang Q, Xin C, Wang S and Ye Z 2021 *Inorg. Chem.* **60** 14978
[14] Cates E, Wilkinson A and Kim J 2012 *J. Phys. Chem. C* **116** 12772
[15] Zhang S, Liang H and Liu C 2013 *J. Phys. Chem. C* **117** 2216
[16] Tian X, Wu Z, Jia Y, Chen J, Zheng R K, Zhang Y and Luo H 2013 *Appl. Phys. Lett.* **102** 42907
[17] Meshulach D and Silberberg Y 1999 *Phys. Rev. A* **60** 1287
[18] Dudovich N, Dayan B, Faeder S M G and Silberberg Y 2001 *Phys. Rev. Lett.* **86** 47

[19] Lozovoy V V, Pastirk I, Walowicz K A and Dantus M 2003 *J. Chem. Phys.* **118** 3187
[20] Pastirk I, Dela Cruz J M, Walowicz K A, Lozovoy V V and Dantus M 2003 *Opt. Express* **11** 1695
[21] Walowicz K A, Pastirk I, Lozovoy V V and Dantus M 2002 *J. Phys. Chem. A* **106** 9369
[22] Gandman A, Chuntonov L, Rybak L and Amitay Z 2007 *Phys. Rev. A* **75** 031401
[23] Zhang S, Zhang H, Lu C, Jia T, Wang Z and Sun Z 2010 *J. Chem. Phys.* **133** 214504
[24] Weiner A M 2000 *Rev. Sci. Instrum.* **71** 1929
[25] Liu P, Cheng W, Yao Y, Xu C, Zheng Y, Deng L, Jia T, Qiu J, Sun Z and Zhang S 2017 *Laser. Phys. Lett.* **14** 115301
[26] Zhang S, Xu S, Ding J, Lu C, Jia T, Qiu J and Zhang S 2014 *Appl. Phys. Lett.* **104** 014101
[27] Zhang S, Yao Y, Xu S, Liu P, Ding J, Jia T, Qiu J and Sun Z 2015 *Sci. Rep.* **5** 13337
[28] Hull R, Parisi J, Osgood J, Warlimont H, Liu G and Jacquier B 2005 *Spectroscopic Properties of Rare Earths in Optical Materials* (Berlin: Springer) pp. 1–94
[29] Garnall W T, Goodman G L, Rajnak K and Rana R S 1989 *J. Chem. Phys.* **90** 3443
[30] Nadort A, Zhao J and Goldys E 2016 *Nanoscale* **8** 13099
[31] Qin X, Liu X, Huang W, Bettinelli M and Liu X 2017 *Chem. Rev.* **117** 4488
[32] Cheng W, Jia T, Shang X, Zhang S, Ma J, Feng D, Sun Z, and Qiu J 2014 *Phys. Rev. A* **89** 023401
[33] Zheng Y, Cheng W, Yao Y, Xu C, Feng D, Jia T, Qiu J, Sun Z and Zhang S 2017 *RSC. Adv.* **7** 13444