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The ultrafast coherent control of fine green emission in Er^{3+} ion system by the shaped ultra-short laser pulses

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ABSTRACT

In this paper, we theoretically demonstrate that the fine green emission in Er^{3+} ion system can be effectively controlled by the shaped ultra-short laser pulses with a π phase modulation. Our results show that the two photon transition probabilities of two excited states are effectively tuned by the single π phase modulation of 800 nm or 1600 nm laser pulses. By combining π phase modulation of the two exciting pulses, one excited state can be populated at maximum value, while the other state population can be widely tuned from zero to the maximum value. Furthermore, we explain the physical control mechanism of fine green emission by considering the second-order power spectrum of the shaped pulses. These schemes can be applied to realize the selective population of multiple excited states.

1. Introduction

The rare earth ions with abundant energy levels and unique transition properties are widely used in many fields such as optical communication, solid-state laser, solar cells, light emitting displays, biological labeling [1–2]. Recently, due to the development of nanotechnology and biotechnology, especially, the new methods of the materials synthesis, many works focus on the synthesis of rare earth doped nano materials, optical properties and biological applications [3–5]. The energy level structure of rare earth doped material in nano size, spectral properties, and the control of rare earth luminescence have become the hot research topics in the field of rare earth luminescent. The ability to tune luminescence peaks and relative intensities of rare earth can greatly expand the applications in optical biolabeling, light emitting displays, and so on. The emission colors can be determined and effectively tuned by several factors, such as the doped ions and concentrations [6–8], Nanoparticles' size [9], phase [10], and surface ligands [11]. The inner filter effect and energy-transfer processes can be also utilized to tune the color of emissions. For example, Wolfbeis used covalently conjugating organic dyes with high extinction coefficients to tune the upconversion emission of $NaYF_4$:Yb, Er or $NaYF_4$:Yb, Tm nanoparticles [12]. To date, organic dyes, heavy-metal complexes, semiconductor quantum dots, and Au nanoparticles have been used to tune the multicolor emissions by utilizing energy-transfer processes. Recently, Renren Deng experimentally realized tunable emission with arbitrary colors in core–shell upconversion nanocrystals by adjusting the pulse width of infrared laser beams [13]. However, for given composition rare earth nanoparticles, new effective methods are still limited to tune the color emission. Therefore, it is an important task for scientific workers to seek new flexible and effective methods to control the rare earth emissions.

Quantum coherent control based on femtosecond pulse shaping can provide a new idea for the color modulation of rare earth ions. The quantum interference of different excitation pathways connecting the initial and final states can be manipulated by properly designing specific laser field. At present, quantum coherent control has been successfully used to control various optical processes in different fields, such as multiphoton absorption [14–17], multiphoton-ionization photoelectron spectroscopy [18], Stokes Raman scattering [19], high harmonic generation [20],

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energy transfer [21], biomedical imaging [22]. The initial results of tuning color emissions of rare earth ions have been achieved by coherent control based on femtosecond pulse shaping. The green emission in Er^{3+}-doped glass can be enhanced and tuned by π-phase-shaped 800 nm femtosecond laser pulses [23]. Fine tunable red–green upconversion luminescence of glass ceramic containing 5%Er^{3+}: $NaYF_4$ nanocrystals can be realized by two near infrared femtosecond lasers [24]. Shian Zhang experimentally reported the control of the single and two-photon fluorescence in Er^{3+} ion by a π or square phase shaped femtosecond laser pulse [25]. Nevertheless, the development of tuning color emission based on coherent control is still in initial stage. Farther researches should be done to make clear the correlation between the color emissions modulation and pulse phase shaping. Therefore, different femtosecond pulse shaping methods should be researched, and the factors and physical mechanism influencing luminescence modulation should be explored to guide experiment.

When the glass ceramic containing 5%Er^{3+}: $NaYF_4$ nanocrystals excited simultaneously by 800 nm and 1490 nm femtsecond laser, the bright green light centered 550 nm and 522 nm can be observed [24]. In this paper, we establish a theoretical model and use the femtosecond laser with π phase modulation to finely tune the green light in Er^{3+} ion system. We theoretically show that the two photon transition probabilitis of two excited states are widely tuned by the single π phase modulation of 800 nm or 1600 nm femtosecond laser. Besides, by combining π phase modulation of the two exciting pulses, one excited state is maximally populated, while the population of the other state is widely tunable from zero to a maximum value. Furthermore, the physical control mechanism of fine green emission can be explained by the second-order power spectrum (SOPS) of the shaped pulses.

2. Theoretical method

When the glass ceramic containing 5%Er^{3+}: $NaYF_4$ nanocrystals was excited by 800 nm or 1490 nm femtsecond laser, weak green emission was detected. However, when it is excited simultaneously by 800 nm and 1490 nm femtsecond laser, bright green light can be observed [24]. Here, we ignore the weak green emission excited by single 800 nm or 1490 nm femtsecond laser, and only consider the bright green light excited simultaneously by 800 nm and 1490 nm femtsecond laser. We establish a relatively simple model and consider two photon processes to calculate the transition probability of the green emission.

Fig. 1 shows the diagram of the two photon absorption processes in the Er^{3+} ion system by the 800 nm and 1600 nm femtosecond laser pulse with full width at half maximum (FWHM)

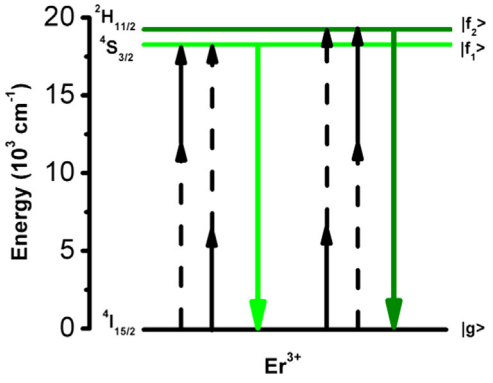

Fig. 1. The diagram of two photon absorption processes in Er^{3+} ion system. Here, $|g> \rightarrow |f_1>$ or $|f_2>$ is coupled by the 800 nm (dashed lines) and 1600 nm (solid lines) femtosecond laser.

600 cm^{-1}, where $|g>$, $|f_1>$ and $|f_2>$ are the ground state, the first excited state and the second excited state. The transition from $|g>$ to $|f_1>$ or $|f_2>$ can be coupled by the laser field $E_1(t)$ and $E_2(t)$, which are centered at 800 nm and 1600 nm. In our simulation, the transition frequencies for $|g>$ to $|f_1>$ and $|f_2>$ are 18350 cm^{-1} and 19150 cm^{-1}.

Assuming only the ground state $|g>$ is initially populated, the two photon transition probability of the excited state $|f_1>$ or $|f_2>$ can be approximated by the second-order time-dependent perturbation theory as [14,15]

$$P_f(t) \propto \left| \int_{-\infty}^{t} \int_{-\infty}^{t_1} E_1(t_1) E_2(t_2) \times \exp\left(i\omega_{fi} t_1 + i\omega_{ig} t_2\right) dt_2 dt_1 \right|^2, \quad (1)$$

and in the frequency domain is

$$P_f(\omega) \propto \left| \int_{-\infty}^{\infty} E_1(\omega) E_2(\omega_{fg} - \omega) d\omega \right|^2, \quad (2)$$

$$P_f(\omega) \propto \left| \int_{-\infty}^{\infty} A_1(\omega) A_2(\omega_{fg} - \omega) \times \exp\{i[\Phi_1(\omega) + \Phi_2(\omega_{fg} - \omega)]\} d\omega \right|^2, \quad (3)$$

where $E(\omega) = A(\omega)\exp[i\phi(\omega)]$ is the Fourier transform of $E(t)$, $A(\omega)$ and $\phi(\omega)$ are the spectral amplitude and phase. Eq. (3) shows that two photon transitions can occur to all photon pairs satisfying $\omega_{fi} + \omega_{ig} = \omega_{fg}$. The two photon transition probability can be tuned by tailoring the spectral phase $\phi(\omega)$.

3. Results and discussion

The π phase modulation can be defined by the function of $\phi(\omega) = \pi \times \theta(\omega - \omega_{step})$, where ω_{step} is the phase step position, and $\theta(\omega - \omega_{step})$ is Heaviside step function. Fig. 2(a) shows the laser spectrum of 800 nm femtosecond laser with π phase modulation. The spectral phase is set as zero for all wave numbers smaller than the phase step position ω_{step}, and is set as π for all larger ones. The 1600 nm femtosecond laser is transform-limited (TL) pulse having the minimum time duration with $\phi(\omega)=0$. Temporal intensity distribution of the TL pulse and the shaped pulses with ω_{step} = 12300 cm^{-1}, 12500 cm^{-1} are shown in Fig. 2(b). As can be seen the 800 nm TL pulse is the Gaussian shape, and the shaped pulses with the π phase modulation are the double humped pulses.

The two photon transition probability of two excited states can be widely tuned by the single π phase modulation of 800 nm femtosecond laser. Fig. 2(c) shows the population of excited states $|f_1>$ and $|f_2>$ as a function of the π phase step position. All the data is normalized by the probability excited by the TL pulse. It can be seen that both the population of excited states $|f_1>$ and $|f_2>$ can be strongly modulated, and are widely tunable from zero to a maximum value. We note that the population of excited states $|f_1>$ achieves a minimum of zero at ω_{step} = 12300 cm^{-1}. That is to say, the shaped pulses with ω_{step} = 12300 cm^{-1} induce the two photon excitation pathways from $|g> \rightarrow |f_1>$ with a destructive interference. The shaped pulses with ω_{step} = 12700 cm^{-1} induce the zero population of excited states $|f_2>$, which corresponds to the dark pulse that induces a destructive interference in different two photon pathways from $|g> \rightarrow |f_2>$. The shaped pulses with ω_{step} = 12500 cm^{-1} induce the same population of excited states $|f_1>$ and $|f_2>$.

The SOPS of the shaped pulses can be used as an excellent tool to explain the physical control mechanism of non-resonant two photon absorption [15,26]. Fig. 2(d) shows the SOPS of the TL pulse and the shaped pulses with ω_{step} = 12300, 12500 and 12700 cm^{-1}. As can be seen the SOPS intensities of the TL pulse are of maximum value and equal at 18350 and 19150 cm^{-1}. The 800 nm and