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Laser polarization and phase control of up-conversion fluorescence in rare-earth ions

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We theoretically and experimentally demonstrate the up-conversion fluorescence control via resonance-mediated two-photon absorption in rare-earth ions by varying both the laser polarization and phase. We show that both the laser polarization and phase can control the up-conversion fluorescence, and the up-conversion fluorescence intensity is decreased when the laser polarization changes from linear through elliptical to circular. We also show that the laser polarization will affect the control efficiency of the up-conversion fluorescence by varying the laser phase, and the circular polarization will reduce the control efficiency. Furthermore, we suggest that the control efficiency by varying the laser polarization and the effect of the laser polarization on the control efficiency by varying the laser phase can be artificially manipulated by controlling the laser spectral bandwidth. This optical control method opens a new opportunity to control the up-conversion fluorescence of rare-earth ions, which may have significant impact on the related applications of rare-earth ions.

The luminescent materials doped with rare-earth ions have attracted considerable interest due to their unique optical and chemical properties, such as intense luminescence, good photostability and low toxicity. Recently, the luminescent lanthanide materials have been widely applied in laser sources[1,2], fiber optic communications[3,4], light-emitting diodes[5], color displays[6–9], and biolabels[10–12]. Especially, these luminescent materials have been developed as a new class of luminescent optical labels that have become promising alternatives to organic fluorophores and quantum dots for applications in biological assays and medical imaging[13,14]. Here, the lanthanide luminescent probes can offer low autofluorescence background, large Stokes shift, high resistance to photobleaching and high penetration depth, and so have shown to be an attractive detecting method with high sensitivity and selectivity. Consequently, in recent years the optical properties of rare-earth ions and their related materials have become an important research subject.

The up-conversion fluorescence in rare-earth ions by converting low frequency photon to high frequency emission via two-photon or multi-photon absorption has attracted a lot of attention because of its potential applications in various related fields[15–17]. The ability to control the up-conversion fluorescence suppression, enhancement or tuning is very important for further extending its applications. Nowadays, two main methods are proposed to control the up-conversion fluorescence. One is varying the material property, such as controlling dopant-host combination[18,19], nanoparticle size[20–22], or dopant concentration[23–25]. The other one is varying the laser parameters, such as controlling the laser spectral phase[26,27], or utilizing the two-color laser fields[28]. Recently, we experimentally and theoretically proved that the femtosecond pulse shaping technique can provide an effective method to control the up/down-conversion fluorescence, and showed that the up/down-conversion fluorescence via single- and two-photon absorption in Er^{3+} ions can be enhanced, suppressed or tuned by a π or square phase modulation[26,27]. In this letter, we further study the up-conversion fluorescence control via resonance-mediated two-photon absorption in rare-earth ions by varying both the laser polarization and phase. Our theoretical results indicate that the resonance-mediated two-photon absorption can be controlled by both the laser polarization and phase, and the control efficiency by varying the laser phase will be affected by the laser polarization. To demonstrate the laser polarization and phase control, we perform the experiment in Dy^{3+} ions by the polarization- and phase-shaped femtosecond laser pulse, and show that the up-conversion fluorescence intensity is decreased as the laser polarization is changed from linear to circular, and the control efficiency by

a π phase modulation is reduced for the circular polarization. Finally, we propose a scheme to artificially manipulate the control efficiency by varying the laser polarization and the effect of the laser polarization on the control efficiency by varying the laser phase with the controllable laser spectral bandwidth.

Considering the nonlinear interaction between a linearly polarized femtosecond laser field E(t) and a quantum system with broad absorption line, the multi-photon absorption can be considered as a sum of each individual transition. On the basis of the theoretical model in the atom system with narrow absorption line limit[29], the resonance-mediated two-photon absorption $S^{(1+1)}$ in rare-earth ions can be approximated as

$$S^{(1+1)} \propto \int_{-\infty}^{+\infty} d\omega_f A(\omega_f) \left| \int_{-\infty}^{+\infty} A(\omega_i) \int_{-\infty}^{+\infty} E(t_1) \exp\left[i(\omega_f - \omega_i)t_1\right] \times \int_{-\infty}^{t_1} E(t_2) \exp(i\omega_i t_2) dt_2 dt_1 d\omega_i \right|^2, \quad (1)$$

where $A(\omega_i)$ and $A(\omega_f)$ are the absorption line-shape function in the immediate state $|i\rangle$ and final state $|f\rangle$, and ω_i and ω_f are the resonant frequencies of the $|i\rangle$ and $|f\rangle$ states. By transforming Eq. (1) into the frequency domain, $S^{(1+1)}$ can be written as[26]

$$S^{(1+1)} \propto \int_{-\infty}^{+\infty} d\omega_f A(\omega_f) \left| P_{\text{Res.}}^{(1+1)} + P_{Near-\text{Res.}}^{(1+1)} \right|^2, \quad (2)$$

with

$$P_{\text{Res.}}^{(1+1)} \propto i\pi \int_{-\infty}^{+\infty} d\omega_i A(\omega_i) E_0(\omega_f - \omega_i) E_0(\omega_i) e^{i\left[\Phi(\omega_f - \omega_i) + \Phi(\omega_i)\right]}, \quad (3)$$

and

$$P_{Near-\text{Res.}}^{(1+1)} \propto \wp \int_{-\infty}^{+\infty} d\omega E_0(\omega_f - \omega) E_0(\omega) e^{i\left[\Phi(\omega_f - \omega) + \Phi(\omega)\right]} \Big/ (\omega_i - \omega), \quad (4)$$

where $E(\omega)$ is the Fourier transform of $E(t)$ with $E(\omega) = E_0(\omega)\exp[i\Phi(\omega)]$, and $E_0(\omega)$ and $\Phi(\omega)$ are the spectral amplitude and phase, respectively. Eq. (3) is the on-resonant term that interferes all on-resonant two-photon excitation pathways with the frequencies of ω_i and ω_f-ω_i, while Eq. (4) is the near-resonant term that interferes all other near-resonant two-photon excitation pathways with the frequencies of ω and ω_f-ω (see Fig. 1(a)). The on-resonant term is excluded from the near-resonant term by Cauchy's principal value operator $\wp$. When the linearly polarized laser field $\vec{E}(t) = E_0(t)\cos(\omega t)\vec{e}_x$ propagates through a quarter wave plate (λ/4 wave plate), the output laser field can be decomposed to two orthogonal polarization directions $\vec{e}_x$ and $\vec{e}_y$ and is expressed by

$$\vec{E}_{\lambda/4}(t) = E_0(t)\cos(\theta)\cos(\omega t)\vec{e}_x + E_0(t)\sin(\theta)\cos(\omega t)\vec{e}_y, \quad (5)$$

where θ is the angle of the input laser polarization direction and the λ/4 wave plate optical axis. It is easy to verify that the output laser is linear polarization for θ = mπ/2 (m = 0, 1, 2…), circular polarization for θ = (2m + 1)π/4, and elliptical polarization for other rotation angle θ. In the resonance-mediated two-photon absorption process, the two photons in the on-resonant term $P_{\text{Res.}}^{(1+1)}$ can come from the same polarization direction (i.e., $\vec{e}_x\vec{e}_x$ and $\vec{e}_y\vec{e}_y$) or different polarization directions (i.e., $\vec{e}_x\vec{e}_y$ and $\vec{e}_y\vec{e}_x$), whereas the two photons in the near-resonant term $P_{Near-\text{Res.}}^{(1+1)}$ only can come from the same polarization direction (i.e., $\vec{e}_x\vec{e}_x$ and $\vec{e}_y\vec{e}_y$). Thus, the on-resonant term $P_{\text{Res.}}^{(1+1)}$ and the near-resonant term $P_{Near-\text{Res.}}^{(1+1)}$ can be further written as

$$P_{\text{Res.}}^{(1+1)} \propto i\pi\left[\cos(\theta)^4 + \sin(\theta)^4 + 2\cos(\theta)^2\sin(\theta)^2\right] \times \int_{-\infty}^{+\infty} d\omega_i A(\omega_i) E_0(\omega_f - \omega_i) E_0(\omega_i) e^{i\left[\Phi(\omega_f - \omega_i) + \Phi(\omega_i)\right]} \propto i\pi \int_{-\infty}^{+\infty} d\omega_i A(\omega_i) E_0(\omega_f - \omega_i) E_0(\omega_i) e^{i\left[\Phi(\omega_f - \omega_i) + \Phi(\omega_i)\right]}, \quad (6)$$

and

$$P_{Near-\text{Res.}}^{(1+1)} \propto \wp\left[\cos(\theta)^4 + \sin(\theta)^4\right] \times \int_{-\infty}^{+\infty} d\omega E_0(\omega_f - \omega) E_0(\omega) e^{i\left[\Phi(\omega_f - \omega) + \Phi(\omega)\right]} \Big/ (\omega_i - \omega). \quad (7)$$

As can be seen from Eqs. (6) and (7), the on-resonant term $P_{\text{Res.}}^{(1+1)}$ depends on the laser phase while is independent of the laser polarization, and the near-resonant term $P_{Near-\text{Res.}}^{(1+1)}$ depends on both the laser phase and polarization, thus the resonance-mediated two-photon absorption $S^{(1+1)}$ can be controlled by varying the laser phase or/and polarization. It is easy to verify that $S^{(1+1)}$ is maximal value for θ = mπ/2 (the linear polarization) and minimal value for θ = (2m + 1)π/4 (the circular polarization). Therefore, when the laser polarization changes from linear through elliptical to circular, $S^{(1+1)}$ is decreased. Furthermore, it can be seen that the laser polarization will affect the control efficiency of $S^{(1+1)}$ by varying the laser phase, and the circular polarization will reduce this control efficiency.

In order to demonstrate the laser polarization and phase control of the up-conversion fluorescence in rare-earth ions, we perform the experiment in the glass sample $60SiO_2$-$20Al_2O_3$-$20CaF_2$ doped with Dy^{3+} ions, and the excitation and detection scheme is shown in Fig. 1(a), which involves the $^6H_{15/2}$ state as the ground state $|g\rangle$, the $^6F_{5/2}$ state as the intermediate state $|i\rangle$, and the $^4I_{13/2}$ state as the final state $|f\rangle$. The transition frequency of the $^6F_{5/2}$ state is $\omega_i = \omega_{^6F_{5/2}} = 12432\ \text{cm}^{-1}$, corresponding to the wavelength of 804 nm, and the transition frequency of the $^4I_{13/2}$ state is $\omega_f = \omega_{^4I_{13/2}} = 25919\ \text{cm}^{-1}$, corresponding to the wavelength of 386 nm. The population in the $^4I_{13/2}$ state via resonance-mediated two-photon absorption can spontaneously decay to the $^6H_{15/2}$,$^6H_{13/2}$ and $^6H_{11/2}$ states by the lower $^6F_{9/2}$ state.

Our experimental arrangement is shown in Fig. 2. A Ti-sapphire mode-locked regenerative amplifier (Spectra-physics, Spitfire) is used as the excitation source with the pulse width of about 50 fs, the center wavelength of 803 nm and the repetition rate of 1 kHz. A programmable 4-f configuration zero-dispersion pulse shaper is used to vary the laser phase in the frequency domain, which is composed of a pair of diffraction gratings with 1200 lines/mm, 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 control the spectral phase and/or amplitude. A λ/4 wave plate is used to vary the laser polarization from linear through elliptical to circular and vice-versa. The polarization- and phase-shaped laser pulse is focused into the glass sample doped with Dy^{3+} ions with a lens of 50-mm focal length, and the laser intensity at the focus is estimated to be $\sim 4 \times 10^{12}$ W/cm^2. The up-conversion fluorescence signal is perpendicularly collected by a telescope system and measured by a spectrometer with charge-coupled device (CCD).

Figures 1(b) and 1(c) show the absorption and up-conversion fluorescence spectra of the glass sample doped with Dy^{3+} ions, respectively. As shown in Fig. 1(b), there are two obvious absorption peaks around the wavelength of 800 and 388 nm, which are corresponding to the $^6F_{5/2}$ and $^4I_{13/2}$ states. As shown in Fig. 1(c), three up-conversion fluorescence signals can be observed at the wavelengths of 487, 577 and 665 nm, which result from these state transitions

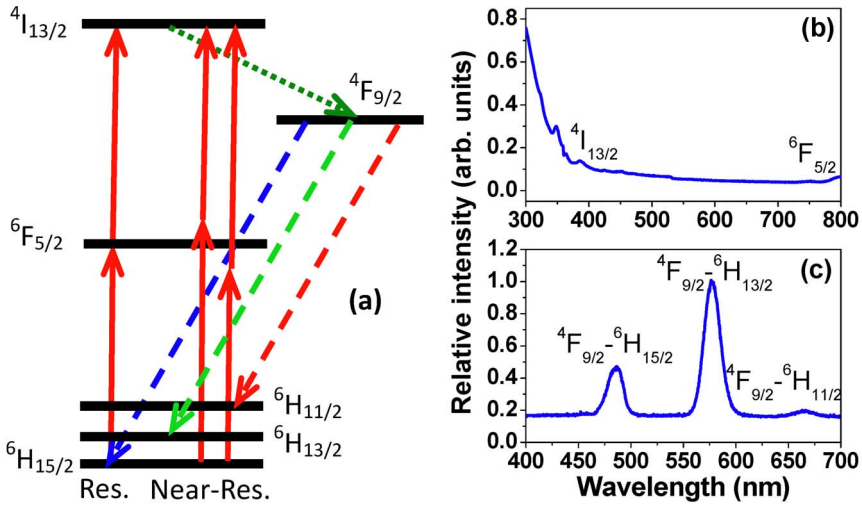

Figure 1 | The energy level diagram of the two-photon absorption and up-conversion fluorescence detected schemes in the glass sample $60SiO_2$-$20Al_2O_3$-$20CaF_2$ doped with Dy^{3+} ions (a), and its absorption (b) and fluorescence (c) spectra.

$^4F_{9/2} \rightarrow {}^6H_{15/2}$, $^4F_{9/2} \rightarrow {}^6H_{13/2}$ and $^4F_{9/2} \rightarrow {}^6H_{11/2}$. Here, we utilize a π phase step modulation to control the up-conversion fluorescence, and this spectral phase modulation is schematically shown in the inset of Fig. 2, where δ represents the phase step position. The π phase step modulation is characterized by a phase jump from 0 to π at the phase step position δ. The shaped femtosecond laser pulse with the π phase step modulation has been proved to be a well-established tool in quantum coherent control because it can induce a constructive or destructive interference between different excitation pathways, and therefore it has been widely employed to manipulate various multiphoton absorption process in atomic and molecular systems and played an important role for developing the quantum control concepts and techniques[30–32].

We first demonstrate the up-conversion fluorescence control by varying the laser polarization. Figure 3 presents the experimental up-conversion fluorescence intensity as the function of the λ/4 wave plate angle, together with the theoretical calculation. As expected, the up-conversion fluorescence intensity can be controlled, which is decreased as the laser polarization is changed from linear through elliptical to circular, but the control efficiency is relatively low (only about 8%). The low control efficiency can be analyzed by the theoretical description formulated in Eqs. (6) and (7). The near-resonant term $P^{(1+1)}_{Near-Res.}$ depends on the laser polarization while the on-resonant term $P^{(1+1)}_{Res.}$ is independent of the laser polarization, and the near-resonant term $P^{(1+1)}_{Near-Res.}$ is much smaller than the on-resonant term $P^{(1+1)}_{Res.}$, thus the control efficiency by varying the laser polarization is low since the weight of the near-resonant term $P^{(1+1)}_{Near-Res.}$ is small.

In our previous studies, we have shown that shaping the laser spectral phase can provide a very important method to control the up-conversion fluorescence in rare-earth ions[26,27]. To verify the effect of the laser polarization on the control efficiency of the up-conversion fluorescence by shaping the laser spectral phase, we

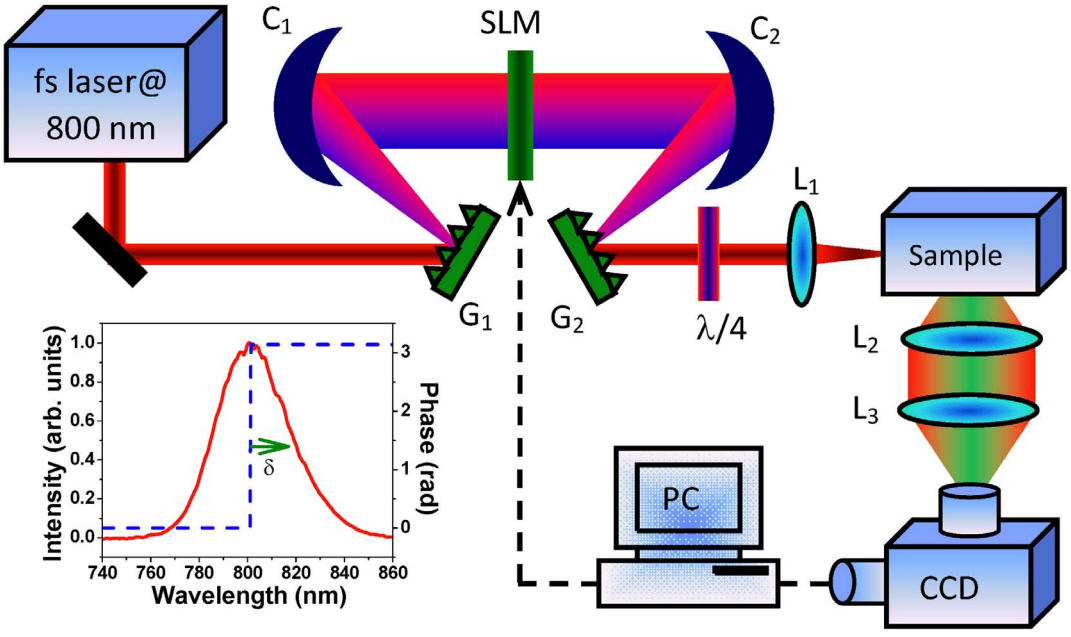

Figure 2 | Schematic diagram of the experimental arrangement for the laser polarization and phase control of up-conversion fluorescence in rare-earth ions. Here, a spatial light modulator (SLM) is used to modulate laser spectral phase and a λ/4 wave plate is employed to vary the laser polarization. Inset shows the shaped laser spectrum by a π phase step modulation.

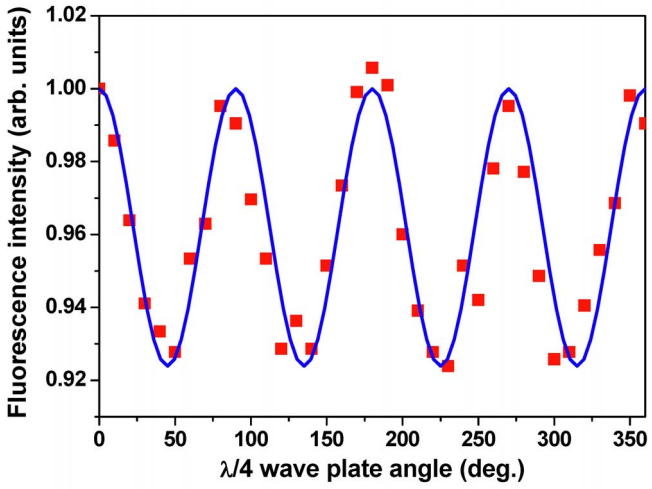

Figure 3 | The experimental (red squares) and theoretical (blue line) up-conversion fluorescence intensity by varying the λ/4 wave plate angle.

experimentally measure the up-conversion fluorescence intensity as the function of the π phase step position for the linearly and circularly polarized femtosecond laser pulses, as shown in Fig. 4, and the theoretical calculation results are also given. As can be seen, the up-conversion fluorescence can be effectively suppressed by the π phase step modulation, but the control efficiency will be affected by the laser polarization, which is decreased as the laser polarization is changed from linear to circular. Similarly, the effect of the laser polarization on the control efficiency by varying the laser phase can also be explained by considering the contributions of the on-resonant term $P_{Res.}^{(1+1)}$ and the near-resonant term $P_{Near-Res.}^{(1+1)}$ in Eqs. (6) and (7). The near-resonant term $P_{Near-Res.}^{(1+1)}$ is correlated with the laser polarization, and the circular polarization will yield the minimum contribution of the near-resonant term $P_{Near-Res.}^{(1+1)}$, thus the control efficiency by varying the laser phase is decreased when the laser polarization changes from linear to circular.

Since only the near-resonant term $P_{Near-Res.}^{(1+1)}$ in the resonance-mediated two-photon absorption process is correlated with the laser polarization (see Eqs. (6) and (7)), one simple way to artificially manipulate the control efficiency by varying the laser polarization and the effect of the laser polarization on the control efficiency by varying the laser phase is controlling the laser spectral bandwidth. With the increase of the laser spectral bandwidth, the contribution of the near-resonant term $P_{Near-Res.}^{(1+1)}$ in the resonance-mediated two-photon absorption process will increase, and therefore both the control efficiency by varying the laser polarization and the effect of the laser polarization on the control efficiency by varying the laser phase will increase. Consequently, one can choose the laser spectral bandwidth according to the experimental requirements. Increasing the laser spectral bandwidth can improve the control efficiency by varying the laser polarization, and decreasing the laser bandwidth can suppress the effect of the laser polarization on the control efficiency by varying the laser phase.

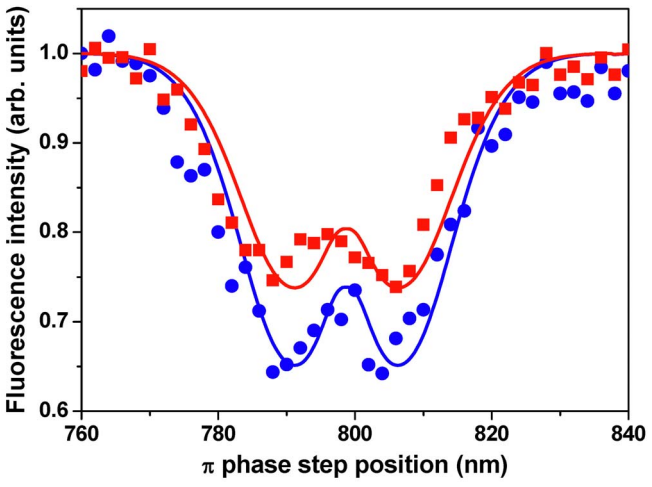

Figure 4 | The experimental (scatter) and theoretical (line) up-conversion fluorescence intensity as the function of the π phase step position for the linearly (circles) and circularly (squares) polarized laser pulse.

In summary, we have experimentally and theoretically demonstrated that the up-conversion fluorescence via resonance-mediated two-photon absorption in rare-earth ions can be controlled by varying the polarization and phase of the femtosecond laser pulse. It was shown that the up-conversion fluorescence in Dy^{3+} ions can be controlled by both the laser polarization and phase; when the laser polarization changes from linear to circular, the up-conversion fluorescence intensity is decreased; the laser polarization will affect the control efficiency by the π phase step modulation, and the circular polarization will reduce the control efficiency. Additionally, a feasible scheme by controlling the laser spectral bandwidth was proposed to artificially manipulate the control efficiency by varying the laser polarization and the effect of the laser polarization on the control efficiency by varying the laser phase. We believe that these theoretical and experimental results are very useful for further understanding and controlling the up-conversion fluorescence via multi-photon absorption in various rare-earth ions. Since the up-conversion fluorescence in the rare-earth-doped glass can be well controlled by the ultrafast pulse shaping method, the higher control efficiency in the rare-earth-doped crystal should be obtained due to its better optical property. Furthermore, the up-conversion fluorescence control of rare-earth ions can be obtained under the low femtosecond laser field, that is to say, such a laser field does not damage the biomolecules, and therefore the optical control method can also be applied in biosystem.

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Author contributions

Y.Y. wrote the main manuscript text and prepared figures; H.Z. and Y.Y. measured the experimental data; S.Z. performed the theoretical calculation; J.Q. provided the sample materials and S.Z., T.J., J.D. and Z.S. revised the manuscript text. All authors reviewed the manuscript.

Additional information

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