Multi-dressing time delayed fourth- and sixth-order fluorescence processes in Pr$^{3+}$:YSO

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We report the multi-dressing time delayed fluorescence processes in a Pr$^{3+}$:YSO crystal, which have been investigated both theoretically and experimentally. The time delay of fluorescence processes can be controlled by the dressing effect which can be adjusted by the power or detuning of laser fields. In addition, we investigate the spectrum of the fluorescence signal at four different stages in the time domain. Specifically, there exists competition between fluorescence and spontaneous parametric four-wave mixing (SP-FWM), which can be separated due to the dressing effect. The experimental results can be well explained by the theoretical model.

1. Introduction

Lots of studies focus on the quantum coherence excitation and coherence transfer in atomic gases. Theses processes lead to many important physical phenomena, such as electromagnetically induced transparency (EIT), four-wave mixing (FWM) and six-wave mixing (SWM) under EIT conditions. Compared with atomic gases, solid materials are more appropriate for practical applications. Rare-earth-ion-doped crystals, for example Pr$^{3+}$:Y$_2$SiO$_5$, have unique properties, such as long optical coherence times and optical controllability of the ionic states. Research areas like light coherent storage, all-optical routing, optical velocity reduction and reversible storage of double light pulses, and all-optically controlled higher-order nonlinear fluorescence (FL) have been realized with such crystals. Since the population and state transfer can be induced by an external electric field pulse, the lifetime of the state can be modified by the Stark shift in the atomic system. Similarly, the principal motivation of this work is how to control lifetime by using the dressing effect from the external laser field in a Pr:YSO crystal. Such results can find potential applications in optical information storage and processing on a photonic chip.

In this paper, we investigate the lifetime competition between SP-FWM and FL by gate control. At the zero delay stage there exist SP-FWM and FL signals together, and a strong SP-FWM signal can be demarcated from the FL signal in the same channel due to the dressing effect by utilizing the change of power and detuning of controlling fields. Moreover, for the long delay stage, there exists only the FL signal, so SP-FWM and FL signals can be separated in the time domain by gate control. The delay of FL signal lifetime depends on the splitting distance between the two dark states due to the dressing effect.

2. Experimental setup and basic theory

Y$_2$SiO$_3$ is a monoclinic crystal and belongs to the $C_{2h}^6$ space group with eight molecules per unit cell. Triply ionized rare-earth ions substitute for the Y$^{3+}$ ions and occupy two inequivalent crystallographic sites with eight molecules per unit cell. Therefore, Pr$^{3+}$:YSO:Y$_2$SiO$_3$ has been realized with such crystals. Since the population and state transfer can be induced by an external electric field pulse, the lifetime of the state can be modified by the Stark shift in the atomic system. Similarly, the principal motivation of this work is how to control lifetime by using the dressing effect from the external laser field in a Pr:YSO crystal. Such results can find potential applications in optical information storage and processing on a photonic chip.

In this paper, we investigate the lifetime competition between SP-FWM and FL by gate control. At the zero delay stage there exist SP-FWM and FL signals together, and a strong SP-FWM signal can be demarcated from the FL signal in the same channel due to the dressing effect by utilizing the change of power and detuning of controlling fields. Moreover, for the long delay stage, there exists only the FL signal, so SP-FWM and FL signals can be separated in the time domain by gate control. The delay of FL signal lifetime depends on the splitting distance between the two dark states due to the dressing effect.
coherence properties of such a system, the two FL processes can be unified together by the fourth-order coherence process as follows

\[
\rho_{FL,1}^{(4)} = \frac{-|G_1|^2}{\left(d_1 + |G_1|^2/\Gamma_{00} + |G_2|^2/\Gamma_{12} + |G_2|^2/\Gamma_{21}\right) \left(\Gamma_{00} + |G_1|^2/d_1 + |G_2|^2/d_2\right)} 
\times \frac{-|G_1|^2}{\left(d_1 + |G_1|^2/\Gamma_{00} + |G_2|^2/\Gamma_{12} + |G_2|^2/\Gamma_{21}\right) \left(\Gamma_{11} + |G_1|^2/d_1\right)}. 
\]

or

\[
\rho_{FL,2}^{(4)} = \frac{-|G_1|^2}{\left(d_2 + |G_2|^2/\Gamma_{00} + |G_1|^2/\Gamma_{12} + |G_2|^2/\Gamma_{21}\right) \left(\Gamma_{11} + |G_1|^2/d_1 + |G_2|^2/d_2\right)} 
\times \frac{-|G_2|^2}{\left(d_2 + |G_2|^2/\Gamma_{00} + |G_1|^2/\Gamma_{12} + |G_2|^2/\Gamma_{21}\right) \left(\Gamma_{22} + |G_2|^2/d_2\right)}. 
\]

where \(d_1 = \Gamma_{10} + i\Delta_1, d_2 = \Gamma_{20} + i\Delta_2, d_1 = \Gamma_{12} + i(\Delta_1 - \Delta_2)\) and \(d_2 = \Gamma_{21} + i(\Delta_2 - \Delta_1)\). \(G_i = -\mu_i\hbar\Gamma_i\) is the Rabi frequency, and \(\Gamma_i\) is the transverse decay rate.

Therefore, the intensity of the measured FL signal can be described as:

\[
I_1(t) = a_0 e^{-G_2 t} + \rho_{11}^{(4)} e^{-G_1 t} + a_1 e^{-\left(\gamma_1 T_2 + T_2\right)} + a_2 e^{-\left(\gamma_1 T_2 + T_2\right)} 
\]

where \(a_0 e^{-G_2 t}\) is the FL signal intensity at the photoexcitation stage, and \(G_2^2\) is the Rabi frequency of the input fields. \(\rho_{11}^{(4)} e^{-G_1 t}\) is the FL signal intensity at the zero delay stage, \(\Gamma_{11}\) is a decoherence rate; \(a_1 e^{-\left(\gamma_1 T_2 + T_2\right)}\) is the FL signal intensity at the adiabatic population transition stage, \(\gamma_1\) is the pulse width, and \(T_0\) is the delay time, which is mainly determined by phonon-assisted non-radiative transition which is mainly determined by acoustic phonons at low temperature. \(a_2 e^{-\left(\gamma_1 T_2 + T_2\right)}\) is the FL signal intensity at the spontaneous radiation stage, and \(a_0, a_1, a_2\) depend on the input laser field power.

Generally, the dephasing rate \(T_1(t)\) of the measured FL signal radiated from level \(|1\) is determined by the dephasing time, \(T_2(t)\), including the longitudinal dephasing time (spontaneous emission lifetime \(T_{11}\)) and the reversible transverse dephasing time, \(T_{12}\), i.e., \(T_1 = (2\pi T_1)^{-1} + 2\pi T_{12}^{-1}\). However, in our case, because the starting points of the pulse laser and measuring process are triggered simultaneously, the factor affecting the measured linewidth should include a coherence process between two levels \(|0\) and \(|1\) which can be described as the coherence rate \(\Gamma_{10}\), where \(\Gamma_{10} = (\Gamma_1 + \Gamma_0)/2, (i,j = 0, 1, 2)\). \(\Gamma_{FL} = T_{11} + T_{10}\) in which \(\Gamma_{00} = 2\pi T_{10}^{-1} + 2\pi T_{20}^{-1}\) is the dephasing rate of the ground state \(|0\). In detail, by taking the controlling terms into account, one can get

\[
(2\pi T_1)^{-1} = 16\pi(\nu + \Delta\nu)^3/\eta^2\hbar^3, 
\]

\[
(2\pi T_2)^{-1} = 0, (2\pi T_{10})^{-1} = P_1(t) + \gamma, (2\pi T_{20})^{-1} = P_0(t) + \gamma, 
\]

where \(\gamma\) is the total effect of \(\Gamma_{\text{phonon}}\) (is related to the temperature of the sample) and \(\Gamma_{\text{ion-spin}}\) (ion-spin relates to the ion–spin coupling effect of the individual ion), and
Thus, the lifetime is modified with \( \Delta t \), respectively, controlled by the pump power. \( \sum |A_{\alpha\beta}|^2 R_{\alpha\beta}^{(n)} \) and \( \sum |A_{\alpha\beta}|^2 R_{\alpha\beta}^{(n)} \) represent the induced dipole–dipole interactions of states H–H and D–D, respectively.

In the current V-type level system, there exists a second-order FL signal FL3 (FL4) via the pathway \( \rho_{00} \rightarrow \rho_{11} \rightarrow \rho_{22} \) and considering the self- or external-dressing effect, the diagonal element \( \rho_{22}^{(2)} \) for the fluorescence signal FL3 (FL4) is given by

\[
\rho_{22}^{(2)} = \frac{-|G_1|^2}{d_1 + |G_1|^2/d_1} \left( T_{11} + |G_1|^2/d_1 \right).
\]

In addition to the FL spectrum, a SP-FWM spectrum will occur in the paraxial direction due to the so-called phase-matching FWM (PC-FWM) process (see Fig. 1(d)) occurring in the V-type system, in which the strong pumping fields \( E_1 \) and \( E_2 \) are mixed with two weakly generated fields \( E_a \) and \( E_{as1} \), satisfying the phase-matching conditions (PMCs) \( k_{as1} = k_1 + k_2 - k_{as1} \) and \( k_{as1} = k_2 - k_1 - k_{as1} \), respectively. Both signals are detected by a pair of point symmetrical PMTs as shown in Fig. 1(b). The density matrix elements can be obtained by the perturbation chains \( \rho_{00} \rightarrow \rho_{11} \rightarrow \rho_{22} \rightarrow \rho_{33} \) (Stokes signal) and \( \rho_{00} \rightarrow \rho_{22} \rightarrow \rho_{33} \) (anti-Stokes signal) together with the dressing effect of \( E_2 \)

\[
\rho_{33}^{(3)} = \frac{-iG_{as1}^* G_1 G_2}{d_1 + |G_1|^2/d_1} \left( T_{11} + |G_1|^2/d_1 \right).
\]

The intensities of the PC-FWM signals \( (E_{as1} \text{ and } E_{as3}) \) are described as

\[
I_{3S}(t) = I_{0(3S)} \exp[-T_{3S}t].
\]
where \( G_{\text{s1}} = (I_{10}) + (I_{10}) + (I_{20}) + (I_{20}) + (I_{20}) + (I_{20}) \), \( I_{\text{g(s1)}} \approx \left| \rho_{(g(s1))}^{(0)} \right|^{2} \) and \( I_{\text{g(s1)}} \approx \left| \rho_{(g(s1))}^{(2)} \right|^{2} \). Differing from the case of FL signals, the PC-FWM signals are from the coherent processes. The linewidths are determined by the atomic coherence time and are much narrower.

When \( E_{1} \), \( E_{2} \) and \( E_{3} \) in \( |0\rangle \leftrightarrow |1\rangle \leftrightarrow |2\rangle \leftrightarrow |3\rangle \) are on simultaneously, a multi-dressed FL5 signal can be obtained as shown in Fig. 1(c). Such a process can be described by the sixth-order coherence process as

\[
I_{\text{FL5}}(t) = I_{\text{g(s1)}} \exp[-\Gamma_{\text{g(s1)}} t].
\]  

(12)

Similarly, one can obtain the same results at \( \Delta_{1} = 0 \) as shown in Fig. 2(c2). The observed features can be explained by the decoherence rate \( \Gamma_{\text{s1}} \) in eqn (13) relating to the power of \( E_{2} \). The induced dipole–dipole interaction increases with the increased power of \( E_{2} \), so one can deduce that the increased \( \Gamma_{\text{s1}} \) will result in a reduced lifetime of the output signal.

Next, Fig. 2(d) shows the evolution of the SP-FWM at different locations of the boxcar integrator gate pumped by different powers of \( E_{2} \) as scanning \( \Delta_{2} \) at fixed \( \Delta_{3} = 0 \). At first, at the photoexcitation stage, the SP-FWM shows a Lorentzian lineshape. Next, at the decay stage, the intensity of the SP-FWM signal reaches a maximum. The population decreases as the boxcar integrator gate moves backward along the time axis, which leads to a decrease in the intensity of the SP-FWM signal as shown in Fig. 2(d). The reason is that the population decreases as the boxcar integrator gate moves backward along the time axis, which leads to a decrease in the intensity of the SP-FWM signal as shown in Fig. 2(d). At last, the intensities of SP-FWM signals increase with the increased power of \( E_{2} \). However, the spectra of the SP-FWM signals do not show AT splitting, which is insensitive for the dressing effect due to the increased power of \( E_{2} \) as shown in Fig. 2(d1)-(d3). So, the decay process of a strong SP-FWM signal is observed at zero delay. The baseline (the dashed curve in Fig. 2(d)) shows the SP-FWM signal at different locations of the boxcar integrator gate in the time domain as \( \Delta_{1} = -350 \) GHz. The population decreases as decay increases, so the intensity of SP-FWM signal decreases. Similarly, the intensity of the SP-FWM signal also decreases at \( \Delta_{1} = 0 \) (the solid curve in Fig. 2(d)).

We next investigate the lifetime delay of the FL signal by nonradiative relaxation, and the AT splitting spectra. Fig. 3(a) and (b) show the delay time of the FL signal at different powers of \( E_{2} \). There exist two peaks, which correspond to the locations of the zero delay time and a longer delay time. The boxcar integrator gate moves backward along the time axis, which leads to a decrease in the intensity of the SP-FWM signal as shown in Fig. 2(d). At last, the intensities of SP-FWM signals increase with the increased power of \( E_{2} \). However, the spectra of the SP-FWM signals do not show AT splitting, which is insensitive for the dressing effect due to the increased power of \( E_{2} \) as shown in Fig. 2(d1)-(d3). So, the decay process of a strong SP-FWM signal is observed at zero delay. The baseline (the dashed curve in Fig. 2(d)) shows the SP-FWM signal at different locations of the boxcar integrator gate in the time domain as \( \Delta_{1} = -350 \) GHz. The population decreases as decay increases, so the intensity of SP-FWM signal decreases. Similarly, the intensity of the SP-FWM signal also decreases at \( \Delta_{1} = 0 \) (the solid curve in Fig. 2(d)).

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3. Experimental results and discussion

Fig. 2(a) and (b) show two sets of measured lifetimes of SP-FWMS at different values of \( \Delta_{1} \). The decoherence rate \( (\Gamma_{\text{s1}}) \) increases, and the lifetime of the SP-FWM signal reduces with increasing power of \( E_{2} \) at \( \Delta_{1} = -350 \) GHz as shown in Fig. 2(c1). The lifetime and intensity of the SP-FWM signals are detected with a digitizing oscilloscope and are averaged with a fast gated integrator (gate width of 10 \( \mu \)s).
into $|G_{2+}\rangle$ if $\Delta_2 < 0$. The Hamiltonian can be written as

$$H' = -h \begin{bmatrix} \Delta_1 & G_2 \\ G_2 & 0 \end{bmatrix},$$

where $\Delta_1 = \Delta_2 - (1/2)\lambda_{\pm}$. From the relation

$$H'(G_{2+\pm}) = \lambda_{\pm}|G_{2+\pm}\rangle,$$

we can obtain

$$\lambda_{\pm} = [(-1)^{1/2} \Delta_1 \pm \sqrt{\Delta_2^2 + 4|G_2|^2}/2].$$

The obvious delay of the right peak in Fig. 3(a1) is caused by the residual particles in $|G_{2+}\rangle$ transferring to $|G_{-}\rangle$ through a phonon-assisted non-radiative transition which is mainly determined by acoustic phonons at low temperature. Therefore, the delay of the FL decay process depends on the splitting gap between $|G_{2+}\rangle$ and $|G_{-}\rangle$. We can obtain the expression of the splitting distance

$$\Delta_\pm = \sqrt{\Delta_2^2 + 4|G_2|^2},$$

when the dressing effect of field $E_2$ can be neglected at the lower power of field $E_2$. The decay process of FL is delayed $\sim 280$ μs, as shown in Fig. 3(b1). Moreover, at the high power of field $E_2$ $(P_2 = 9$ mW), the splitting distance between $|G_{2+}\rangle$ and $|G_{-}\rangle$ is $\Delta_0 = 2\sqrt{|G_1|^2 + |G_2|^2}$, as $\Delta_1 = \Delta_2 = 0$. The splitting distance increases with the increased power $E_2$, the delay process of FL is delayed $\sim 540$ μs, as shown in Fig. 3(b3). In addition, at the rising edge of the right peak in the time domain, there exists the adiabatic population transition due to the dark states between $|G_{2+}\rangle$, and the time width depends on the pulse width. Finally, at the descending edge of the right peak, all the population is spontaneously radiated to the low energy level, as shown in Fig. 3(b). The corresponding theoretical predictions are shown in Fig. 3(c), which agree well with the experimental results.

Fig. 3(d) shows the intensity evolution of the FL signal. At first, there exists competition between SP-FWM and FL. The second curve in Fig. 3(d1) shows that FL is suppressed, and the SP-FWM is a little. The intensity of the FL signal decreases and the intensity of SP-FWM signal increases with increasing power $E_2$. Specially, the FL signal vanishes at maximum power $E_2$, as shown by the second or third curve in Fig. 3(d3). Secondly, at the rising edge of the right peak in the time domain, the relative width of AT-splitting of FL increases, as the power of $E_2$ increases. Finally, in the decay stage of the right peak in the time domain, the spectra of FL shows a Lorentzian lineshape as shown by the seventh curve in Fig. 3(d1)–(d3). To explain the above experimental results, we turn to the density matrix element related to the FL and SP-FWM signals. At the zero delay stage, the intensity of the SP-FWM signal increases with increasing power $E_2$. However, the intensity of the FL signal decreases, and even vanishes due to the dressing effect. In eqn (1), the term $|G_1|^2/d_1$ can determine the self-dressing effect when $\Delta_1$ is scanned. Due to the low $E_2$ power, the dressing effect of $E_2$ ($|G_1|^2/T_{00}$ and $|G_2|^2/d_1$ in eqn (1)) can be neglected. So the intensity of FL is suppressed, and the SP-FWM is generated (the second curve in Fig. 3(c1)). The pump probability of $E_2$ ($|G_2|^2/T_{00}$) becomes larger as $P_2$ increases and the dressing effect of $E_2$ ($|G_1|^2/d_1$ and $|G_2|^2/d_1$) should be considered. Therefore, the second curve in Fig. 3(d1) shows the profile of FL due to the self-dressing caused by $E_1$. Furthermore, the second curve in Fig. 3(d3) shows that the intensity of FL is fully suppressed by $E_1$ and $E_2$ synchronously, and leaves the SP-FWM signal only. Next, at the rising edge of the right peak in the time domain, there exists an adiabatic population transition between the dark states ($|G_{2+}\rangle$). The FL signal shows AT splitting spectra due to the dressing effect of $E_1$ at low powers of $E_2$, and we can obtain the expression of the splitting distance $\Delta_\pm$ shown by the sixth curve in Fig. 3(d1). The splitting distance will become larger when $P_2$ increases as shown by the sixth curve in Fig. 3(d3). Finally, at the delay stage, the FL signal is from state $|$ state spontaneous radiation, so the spectra of the FL signal shows a Lorentzian lineshape.

Fig. 4(a) and (b) show lifetimes of the FL signal at different $\Delta_2$. It is obvious that the lifetime of the FL signal decays as shown in Fig. 4(b), compared with the lifetime in Fig. 4(a). At $\Delta_2 = -300$ GHz, since the dressing effect of $E_2$ can be neglected, the splitting distance between $|G_{1+}\rangle$ is $\Delta_+ = \sqrt{\Delta_2^2 + 4|G_1|^2}$. However, as $\Delta_2$ is close to the resonant point, the splitting distance between $|G_{2+}\rangle$ and $|G_{-}\rangle$ is $\Delta_0 = 2\sqrt{|G_1|^2 + |G_2|^2}$ due to the dressing effect of $E_1$ and $E_2$. So the delay of the FL signal is longer at $\Delta_2 = 0$, as shown in Fig. 4(b). For the FL signal spectra, first of all, at the zero delay stage, the spectrum of the FL signal is suppressed by $E_1$, and the dressing effect of $E_2$ can be neglected as $\Delta_2$ is far away from the resonant point, as shown by the first curve in Fig. 4(c1) and (c5). If $\Delta_2$ is tuned close to the resonant point, the lineshape of the FL signal is switched from the suppressed dip (the first curve in Fig. 4(c1)) to the emission peak of the SP-FWM signal (the first curve in Fig. 4(c3)), due to the dressing effect of $E_2$ ($|G_1|^2/d_1$ in eqn (1)). Secondly, at the adiabatic population transition stage, the spectrum of the FL signal shows AT like splitting due to the self-dressing effect of $E_1$ ($|G_1|^2/d_1$ and $|G_1|^2/d_2$ in eqn (1)), as shown by the fourth curve in Fig. 4(c3), when $\Delta_2$ is far away from the resonant region. However, at $\Delta_2 = 0$, the FL signal is completely suppressed due to the dressing effect of $E_1$ ($|G_1|^2/d_1$ and $|G_1|^2/d_2$ in eqn (1)) and $E_2$ ($|G_2|^2/d_2$ and $|G_2|^2/d_2$ in eqn (1) together, as shown by the fourth curve in Fig. 4(c3). Thirdly, at the spontaneous radiation stage, the pump probability of $E_2$ ($|G_2|^2/T_{00}$) becomes larger if $\Delta_2$ is tuned to the resonant point. However, there exists a dressing effect as $\Delta_2 = 0$ so the intensity of FL signal decreases as shown in the sixth curve in Fig. 4(c3).

So far, we have shown that the delay of the fluorescence lifetime process of $Pr^{3+}$:YSO in a V-type three level system can be all-optically controlled. Such controllable processes can be extended to any multi-level system. Fig. 5(a) shows the delay of the FL5 lifetime in an N-type level system. The delay of the FL5 lifetime is obviously decayed if $\Delta_1$ is tuned to the resonant point, as shown in Fig. 5(a2), compared with the FL4 lifetime in Fig. 5(a1) at $\Delta_1 = -300$ GHz. The reason is that, due to the dressing effect of $E_2$ and $E_3$, the FL5 delay is $\sim 550$ μs, as shown in Fig. 5(a1). While if $\Delta_1$ is tuned to the resonant point, the dressing effect will be increased and the delay of the FL5 signal becomes $\sim 800$ μs. At the zero delay stage, there exists only the SP-FWM signal due to the strong dressing effect of $E_1$, $E_2$ and $E_3$, and the intensity of the FL signal is completely suppressed as shown by the third and fourth curves in Fig. 5(b). The slope of the lifetime curve of the SP-FWM signal is larger at $\Delta_1 = 0$ than the slope at $\Delta_1 = 300$ GHz, as shown in Fig. 5(a). At the other
stage, the FL5 signal shows the same characteristics as those in Fig. 3 or 4.

4. Conclusion

In summary, we have shown the multi-dressing time delayed FL processes in a Pr\textsuperscript{3+}:YSO crystal, which have been investigated both theoretically and experimentally. The decay process of a strong SP-FWM signal is observed at the zero delay, while the FL signal decays after a longer delay time. For FL processes in the time domain, there are four stages, e.g., the photoexcitation stage, the zero delay stage, the adiabatic population transition stage and the spontaneous radiation stage. The time delay of FL processes depends on the distance of AT-splitting between dark states due to the dressing effect. The splitting distance increases if laser power increases, and the time delay will become larger and larger. Specially, there exists competition between FL and SP-FWM at the zero delay stage, which can be controlled by utilizing dressing effects through adjusting the power or detuning of pump fields. The FL signal is delayed and the SP-FWM can be distinguished from the composite channel in the time domain. Such research can be applied in optical information storage and processing on photonic chips in the future.

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References