All-optically controlled fourth- and sixth-order fluorescence processes of Pr$^{3+}$:YSO

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(Received 11 November 2013; accepted 24 January 2014; published online xx xx xxxx)

We report all-optically controlled fourth- and sixth-order fluorescence processes with and without splitting, in a heteronuclear-like molecule system of Pr$^{3+}$:YSO both in theory and experiment. We construct the asymptote among Pr$^{3+}$ ions localized at different cation vacancies. By changing the frequency detuning and power of the controlling field, fluorescence signal can be controlled to switch from an enhanced peak to a suppressed dip, and vice versa. Such transition has a potential application to produce all-optical switches. The results can be well explained by a theory model based on the high-order coherent process. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4864397]

As well known, rare earth ion doped inorganic crystals can improve or overcome the ultra-fast decoherence processes in solids which prohibit successful implementation of coherent excitation. Compared with atomic gases, atomic coherence-induced effects in solid materials are very attractive for practical applications. Progresses related to atomic coherence in solid-state materials, including enhanced four-wave mixing (FWM) based on atomic coherence, optical velocity reduction and reversible storage of double light pulses, all-optical routing based on optical storage, controllable erasing of optically stored information, light velocity reduction and coherent storage, and electromagnetically induced transparency (EIT) in solid materials, have provided bases for potential applications. Meanwhile, the fluorescence spectrum offers an effective method to investigate the lifetime of population and the excitation process. To realize practical applications, such processes are required to be controlled reliably.

In this paper, we theoretically and experimentally show all-optically controlled fluorescence process in a heteronuclear-like molecule system of Pr$^{3+}$:YSO. Two sets of fluorescence signals, with and without splitting, are chosen to be controlled objects. By changing the detuning and power of the controlling field, desiredly modulated results of fluorescence signal can be obtained, such as the switch between the enhanced peak and suppressed dip, which has a potential application in all-optical switch fabrication. Considering the dressing effects of both coupling fields associating with their excitation competition on the particles of ground state, a theory model based on the high-order coherent process is developed to explain these results.

To implement current experiment, the sample is held at 77 K in a cryostat (CFM-102). Figure 1(a) shows the simplified energy-level diagram of 0.05% rare-earth Pr$^{3+}$ doped Y$_2$SiO$_5$ (Pr$^{3+}$:YSO) crystal. We confine ourselves to a detail analysis of the triplet energy-level $^3H_4$ and singlet energy-level $^1D_2$ in the current work, since it is easy to identify them reliably by investigating the optical spectrum of the Pr$^{3+}$ ions. The Pr$^{3+}$ impurity ions occupy two nonequivalent cation sites (sites I and II, respectively), in the YSO crystal lattice, which can be identified by their spectra (see Fig. 1(e)). The energy level of site I is labeled by a Greek letter without asterisk and the one for site II with an asterisk. However, under the action of crystal field of YSO, the interaction (i.e., the van der Waals interaction) between Pr$^{3+}$ ions localized at different cation vacancies can happen, so one can treat the two ions as a heteronuclear-like molecule.

The energy level shifts of the ground states ($\delta_0$ and $\delta_0'$) of site I and site II induced by the van der Waals interaction, can be given by $\Delta_0 = -\Sigma(C_{mn}/R^n)(n = 6, 8, 10, 12, 13, 14)$, respectively, where $\Sigma(C_{mn}/R^n)$ represent the van der Waals interactions of states H-H'. The expressions for states $D$-H have the similar forms. Note that the ground state $|0\rangle$ of such heteronuclear-like molecule is constituted by $\delta_0$ and $\delta_0'$, while $|1\rangle$ ($\gamma_0$ and $\gamma_0'$) from site I and $|2\rangle$ ($\gamma_0^\prime$ and $\gamma_0''$) from site II as shown in Fig. 1(b). Considering the two nonequivalent cation sites, we can construct a V-type three-level system ($|0\rangle \leftrightarrow |1\rangle \leftrightarrow |2\rangle$) (see Fig. 1(c)). Two tunable dye lasers (with a 0.04 cm$^{-1}$ linewidth) pumped by an injection locked single-mode Nd:YAG laser (Continuum Powerlite DLS 9010, 10 Hz repetition rate, 5 ns pulse width) are used to generate the pumping fields $E_1(\omega_1, \Delta_1)$ and $E_2(\omega_2, \Delta_2)$ with frequency detunings defined as $\Delta_1 = \Delta_0 + \Delta_E$ and $\Delta_2 = \Delta_0 + \Delta_E$, respectively, where $\omega_{\text{mon}} - \omega_i (i = 1, 2)$ and $\omega_{\text{mon}}$ denotes the corresponding transition frequency. $E_1$ drives the transition $|0\rangle \leftrightarrow |1\rangle$, and then $|1\rangle$ emits fluorescence signal FL1. $E_2$, that is used to be the control light, counterpropagates with $E_1$ through the sample (see Fig. 1(c)) and is coupled to $|0\rangle \leftrightarrow |2\rangle$ to generate fluorescence signal FL2, as shown in Fig. 1(a). The controlled results are monitored by two photomultiplier tubes with a fast gated integrator.

Theoretically, the intensity of the fluorescence signal is proportional to the diagonal density matrix elements. In current system, with $E_1$ ($E_2$) on, via two mutually conjugate...
pathways $\rho_{00}^{(2)} E_{10}^{(1)} (E_{01}^{(1)})^*$ and $\rho_{00}^{(0)} (E_{10}^{(0)})^* \rho_{11}^{(1)}$, we can obtain $\rho_{11}^{(2)} = |G_1|^2 / (\Gamma_1 d_1)$ and $\rho_{11}^{(12)} = -|G_1|^2 / (\Gamma_1 d_1 d_2)$, where $d_1 = |i\Delta_1 - \Delta_2, d_1^* = \Gamma_0 - i\Delta_1, d_2 = \Gamma_0 + i\Delta_2$, and $d_2^* = \Gamma_2 - i\Delta_2, G_i = -\mu_j, \mu_j$, is the Rabi frequency of $E_i$ with $\mu_j$ the electric dipole moment between levels $|i\rangle$ and $|j\rangle$, and $\Gamma_j$ is the transverse decay rate. Thus, with all beams on, the corresponding intensity of the total fluorescence signals is $I_{FL} = \rho_{11}^{(2)} + \rho_{11}^{(12)} + \rho_{22}^{(2)} + \rho_{22}^{(12)}$.

Here, in order to analyze the physical mechanism of the fluorescence spectrum conveniently, we just focus on one of the two perturbation chains. Considering the dressing effect of $E_1$ ($E_2$), the diagonal element $\rho_{11}^{(2)}$ ($\rho_{22}^{(2)}$) corresponding to the fluorescence signal FL1 (FL2) is given by:

$$\rho_{11}^{(2)} = -|G_1|^2 / [(d_1 + |G_1|^2 / \Gamma_0)] \Gamma_1/[|G_1|^2 / d_1]$$, (1a)

$$\rho_{22}^{(2)} = -|G_1|^2 / [d_2 + |G_1|^2 / \Gamma_0] \Gamma_2/[|G_1|^2 / d_2]$$, (1b)

When $E_1$ and $E_2$ are on, processes $\rho_{11}^{(2)}$ and $\rho_{22}^{(2)}$ can interplay with each other. Considering the coherence property of the system as mentioned above, the two fluorescence processes can be unified by the fourth-order coherence process as following:

$$\rho_{11}^{(4)} = \frac{|G_1|^2}{[d_1 + |G_1|^2 / \Gamma_0 + |G_2|^2 / d_2] \Gamma_1/[|G_1|^2 / d_1 + |G_2|^2 / d_2]} \times \frac{|G_2|^2}{[d_2 + |G_2|^2 / \Gamma_0 + |G_1|^2 / d_2] \Gamma_2/[|G_2|^2 / d_2]}$$, (2a)

FIG. 1. (a) Simplified energy-level diagram of Pr$^{3+}$ ions in a YSO crystal. (b) Molecular energy level constituted by Pr$^{3+}$ ions. (c) V-type three-level system $(|0\rangle \leftrightarrow |1\rangle \leftrightarrow |2\rangle)$ of a heteronuclear-like molecule system of Pr$^{3+}$-YSO for the interaction between two fluorescence signals. (d) Experimental setup scheme. FL: fluorescence, PMT: photomultiplier tube, PBS: polarized beam splitter, and LI: lens. (e) Typical fluorescence signals in frequency domain at lower excitation power, whereas the higher excitation power case is shown in Fig. 2(c) with $\Delta_2$ far away from the resonance. (f) Fluorescence signals FL1 versus $\Delta_2$ in which solid only turn $E_1$ on, dash turn $E_1$ and $E_2$ on, and dot turn $E_1$ and $E_2$ on, respectively. (g) Two-level diagram of Pr$^{3+}$ ions in a YSO crystal. (h) and (i) Dressed-state pictures for two-level and V-type three-level system, respectively.
fluorescence background is reflected by $\Gamma_{11}^{\ast} + |G_{1}|^{2}/d_{1} + |G_{2}|^{2}/d_{2}$. To demonstrate the validity of the V-type three-level configuration, the modulated FL1 signal is investigated by tuning the frequency of $E_{2}$ into the transition between $|0\rangle \leftrightarrow |2\rangle$. The curve with upper triangles in Fig. 1(f) shows the modulated result when $E_{2}$ is open, which has the similar profile as discussed above with two-level system. That is, the raised baseline of FL1 is due to the fluorescence signal radiated from level $|2\rangle$ excited by $E_{2}$, and the peak of FL1 is split by the dressing effect of $E_{2}$ on level $|0\rangle$ as shown in Fig. 1(i). These spectral features can be understood in the picture of the dressed states in V-type three-level system and are well reproduced by the simulation based on Eqs. (2a) and (2b). Therefore, we can conclude that the levels of two sites are coupled by van der Waals interaction via Feshbach resonance and can use the V-type three-level configuration to analyze the observed phenomena.

Figures 2(a) and 2(c) show two sets of the measured fluorescence signal FL1 versus $\Delta_{1}$ at different $\Delta_{2}$. The profiles with Lorentzian lineshapes (dash curves in Figs. 2(a) and 2(c)), consisted of the baselines of different signals, are the fluorescence signals FL2 versus $\Delta_{2}$. The Autler-Towns (AT)-like splitting phenomena do not appear on the FL2 profiles (dash curves), because the dressing effect of $E_{2}$ can be neglected due to its low power. However, the ability of excitation competition on the particles of ground state still occurs. For the first case (FL1 pumped with Lorentzian lineshape pumped by lower power of $P_{1}$) shown in Fig. 2(a), when $\Delta_{2}$ is far away from the resonant region, the FL1 signal is not affected. If $\Delta_{2}$ is close to the resonant point, the baseline of the FL1 signal is raised gradually due to the excitation competition on the particles of ground state between $E_{1}$ and $E_{2}$, and the intensity of FL1 signal is reduced. If there is only second-order FL signals, FL2 is the baseline when $\Delta_{1}$ is scanned with different $\Delta_{2}$, and the FL2 will become stronger with $\Delta_{2}$ approaching the resonant point. As $E_{1}$ does not change, the dressing effect on FL2 will not change. However, according to the experiment results in Fig. 2(a), we can see that the dressing effect indeed changes. The reason is that there is a fourth-order FL signal in the system, and the FL2 signal is dressed by $E_{2}$. And the mutual interactions mentioned above can be well explained by Eq. (2).

Due to the small powers of $E_{1}$ and $E_{2}$, the dressing effects of $E_{1}$ (i.e., the terms $|G_{1}|^{2}/d_{1}$ and $|G_{1}|^{2}/d_{2}$) and $E_{2}$ ($|G_{2}|^{2}/d_{2}$ and $|G_{2}|^{2}/d_{12}$) can be neglected in Eq. (2). However, the pump probability of $E_{2}$ ($|G_{2}|^{2}/T_{00}$) becomes larger and larger as $\Delta_{2}$ tunes to the resonant point. Therefore, the pump processes are switched between $E_{1}$ and $E_{2}$. Figure 2(c) shows the modulated results of FL1 signal at different $\Delta_{2}$ in which the profile of FL1 shows the AT-like splitting caused by the higher $P_{1}$. Similarly, the dressing effect of $E_{2}$ can be neglected, whereas that of $E_{1}$ must be considered. Associating with the excitation competition, one can explain the controlling process well. Based on above analysis, the corresponding theoretical predictions are shown in Figs. 2(b) and 2(d), respectively, which agree well with experimental data.

Next, the control field $E_{2}$ is at a higher power level shown in Figs. 2(e)–2(g) in contrast to the case shown in Figs. 2(a)–2(d). Except for the power of $E_{2}$, all the other experimental conditions are identical. However, modulated results of the FL1 signal shown in Figs. 2(e) and 2(g) display the difference from the above case. First of all, the spectrum profiles of FL2 (dashed line) exhibit AT-like splitting caused by $E_{2}$, whereas the case in lower excitation power of $E_{2}$ shows the pure Lorentzian profile (see dashed line in Fig. 2(a)). Therefore, the dressing effect of $E_{2}$ described as $|G_{2}|^{2}/d_{12}$ in Eq. (2) cannot be neglected any longer. Second, if $\Delta_{2}$ is close to the resonant point, the lineshape of the FL1 signal shown in Fig. 2(e) is switched from an emission peak to a suppressed dip, which can attribute to the dressing effect of $E_{2}$ described as $|G_{2}|^{2}/d_{12}$ in Eq. (2). Meanwhile, the baseline of the FL1 signal is changed gradually due to the excitation competition on the particles of ground state between $E_{1}$ and $E_{2}$. $E_{2}$ and $E_{1}$ to $E_{2}$ associating with the excitation competition between $E_{1}$ and $E_{2}$. For experimental results shown in Figs. 2(e) and 2(g), the corresponding

![Figure 2](image-url)
theoretical results are shown in Figs. 2(f) and 2(h), respectively. It is clear to see that experiment and theory agree well with each other.

Now, we focus on the P2 power dependences of two sets of the FL1 signals (versus Δ1) on the dressing field E2 (Δ2 = 0) as shown in Figs. 3(a)–3(d). Figure 3(a) shows the power control on the FL1 signal pumped by lower power P1. As P2 increasing, the intensity of FL1 decreases gradually and even to zero due to the excitation competition by E2 shown by the baselines of FL1. One can find that the FL1 lineshape does not change initially which reveals that the dressing effect of E2 ((|G2|2/d21)2) can be neglected. And then the dressing effect of E2 leads to the AT-like splitting on the FL1 lineshape. Finally, the FL2 signal is dominant and suppressed by the dressing effect of E1 ((|G1|2/d2i$)$). Therefore, all-optically controlled process can be used as a high contrast switching. For the higher power P1 case shown in Fig. 3(c), the similar process can be obtained and well explained by considering the dressing effect of E1. Accordingly, the correspondingly controlled processes in theory are shown in Figs. 3(b) and 3(d), respectively.

Finally, different from above cases, the modulated results of FL1 signal are obtained by scanning Δ3 at different P2 as shown in Figs. 3(e)–3(h), which can better pick up the controlled information of FL1. At low power P1, the normalized FL1 signal is smaller than one (see Fig. 3(e)). On one hand, as P2 increasing, the power level of FL1 decreases gradually until the dressing effect of E2 appears. On the other hand, the emission peak of FL2 appears on the FL1 spectrum and increases gradually. Once the dressing effect of E2 appears, the ability of E2 excitation competition on particles is weakened, and then the intensity of FL1 recovers. At this moment, the suppressed dip on FL1 can be observed clearly. Therefore, the switch between enhanced peak and suppressed dip is achieved. However, in the case of high P1, such switch cannot be obtained as shown in Fig. 3(g) in which only the suppressed dip occurs. Such phenomena can be attributed to the influence from both E1 and E2, and the theoretical results shown in Fig. 3(h) agree with the experimental data well.

So far, we have shown the all-optically controlled fluorescence process in a heteronuclear-like molecule system of P23+;YSO both theoretically and experimentally. Such controlled process can be extended to any multi-level system. Figure 4(a) shows the modulated FL1 signals by opening (a1) self-dressing field E1 in |0⟩ ↔ |1⟩, (a2) E1 and external-dressing field E2 in |0⟩ ↔ |1⟩ ↔ |2⟩, (a3) E1 and external-dressing field E3 in |0⟩ ↔ |1⟩ ↔ |3⟩, and (a4) E1, E2, and E3 in |0⟩ ↔ |1⟩ ↔ |2⟩ ↔ |3⟩, respectively, in a N-type four-level system shown as Fig. 4(b). In such case, the dressing effect is weaker than the one in V-type, which is due to the fact that dressing d3 = |G3|2/d30 in Eq. (4). When E1, E2, and E3 in |0⟩ ↔ |1⟩ ↔ |2⟩ ↔ |3⟩ are on simultaneously, a multi-dressed FL1 signal can be obtained as shown in Fig. 4(a4). According to the analysis method used for Fig. 2(a), there must be a six-order FL signal in this four-level atomic system. Such sixth-order coherence process can be written as

$$
\rho_{11}^{(4)} = |G_1|^2/[(d_1 + |G_3|^2/\Gamma_{31} + |G_1|^2/d_{30}) \\
\times (d_1 + |G_1|^2/\Gamma_{00} + |G_1|^2/d_{30})] \\
\times |G_1|^2/(\Gamma_{11} + |G_3|^2/d_3 + |G_1|^2/(d_1 + |G_3|^2/d_{30}))^2,
$$

where

$$
d_1 = \Gamma_{13} + i\Delta_3, d_2 = \Gamma_{30} + i(\Delta_3 - \Delta_1),
$$
d_1 = \Gamma_{03} + i(\Delta_3 - \Delta_1).

In such case, the dressing effect is weaker than the one in V-type, which is due to the fact that dressing d3 in Eq. (4). When E1, E2, and E3 in |0⟩ ↔ |1⟩ ↔ |2⟩ ↔ |3⟩ are on simultaneously, a multi-dressed FL1 signal can be obtained as shown in Fig. 4(a4). According to the analysis method used for Fig. 2(a), there must be a six-order FL signal in this four-level atomic system. Such sixth-order coherence process can be written as

![Fig. 3. Power dependences of FL1 signal versus Δ1 pumped by (a) a low power or (c) a high power of E1 at different P2. (b) and (d) Corresponding theoretical results. FL1 signal at fixed detuning (Δ2 = 0) pumped by (e) a low power or (g) a high power of E1 is modulated by scanning Δ3 at different P2. (f) and (h) Corresponding theoretical predictions.](image)

![Fig. 4. (a) From left to right, the FL1 signals modulate by opening (a1) self-dressing field E1 in |0⟩ ↔ |1⟩, (a2) E1 and external-dressing field E2 in |0⟩ ↔ |1⟩ ↔ |2⟩, (a3) E1 and external-dressing field E3 in |0⟩ ↔ |1⟩ ↔ |3⟩, and (a4) E1, E2, and E3 in |0⟩ ↔ |1⟩ ↔ |2⟩ ↔ |3⟩, respectively, in N-type four-level system. (b) N-type four-level system for (a).](image)
three-level system, the validity of the latter system we constructed is demonstrated. Two sets of $F_{11}$ signals, with and without splitting dressed by its pump field, are chosen to be controlled objects. By changing the detuning and power of the controlling field, desirably modulated results of $F_{11}$ signal can be obtained, such as the switch between the enhanced peak and suppressed dip, which has a potential application to all-optical switch device. Considering the dressing effects of both coupling fields associating with the excitation competition between them on the particles of ground state, a theory model based on the high-order coherent process is developed to explain these results, which agree with the experimental data well.

In summary, we have presented the all-optically controlled fourth- and sixth-order fluorescence processes of $Pr^{3+}:YSO$ in a heteronuclear-like molecule system both in theory and experiment. By the comparison between two-level and $A$-type three-level system, the validity of the heteronuclear-like molecule system is demonstrated. Two sets of $F_{11}$ signals, with and without splitting dressed by its pump field, are chosen to be controlled objects. By changing the detuning and power of the controlling field, desirably modulated results of $F_{11}$ signal can be obtained, such as the switch between the enhanced peak and suppressed dip, which has a potential application to all-optical switch device. Considering the dressing effects of both coupling fields associating with the excitation competition between them on the particles of ground state, a theory model based on the high-order coherent process is developed to explain these results, which agree with the experimental data well.

This work was supported by the 973 Program (2012CB921804), NSFCs (11104214, 61308015, 11104216, 61078002, 61078020, 61108017, and 61205112).

$$\rho_{11}^{(6)} = \frac{-|G_3|^2|G_1|^2|G_2|^2}{|d_1 + |G_3|^2/d_{33}|(\Gamma_{11} + |G_3|^2/d_{13} + A)} \times \frac{|d_2 + |G_2|^2/d_{21}|(\Gamma_{22} + |G_2|^2/d_{22})}{(d_1 + |G_1|^2/d_{10} + |G_2|^2/d_{12}) |\Gamma_{00} + |G_1|^2/d_{03} + |G_2|^2/d_{12}|}$$

(5)

where $d_{23} = \Gamma_{23} + i(\Delta_2 - \Delta_1 + \Delta_2)$ and $A = |G_1|^2/d_1 + |G_3|^2/d_{30}$. By comparing the $A$-type and $V$-type three-level systems, the validity of the latter system we constructed is demonstrated.

In summary, we have presented the all-optically controlled fourth- and sixth-order fluorescence processes of $Pr^{3+}:YSO$ in a heteronuclear-like molecule system both in theory and experiment. By the comparison between two-level and $A$-type three-level system, the validity of the heteronuclear-like molecule system is demonstrated. Two sets of $F_{11}$ signals, with and without splitting dressed by its pump field, are chosen to be controlled objects. By changing the detuning and power of the controlling field, desirably modulated results of $F_{11}$ signal can be obtained, such as the switch between the enhanced peak and suppressed dip, which has a potential application to all-optical switch device. Considering the dressing effects of both coupling fields associating with the excitation competition between them on the particles of ground state, a theory model based on the high-order coherent process is developed to explain these results, which agree with the experimental data well.

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