Polarization dressed multi-order fluorescence of Pr\(^{3+}\):Y\(_2\)SiO\(_5\)

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We report polarization dressed second-, fourth- and sixth-order fluorescence processes in a Pr\(^{3+}\):Y\(_2\)SiO\(_5\) crystal. By changing the polarization states of dressing fields and generating fields, the fluorescence baselines, suppression and Autler–Townes splitting of emission peaks can be controlled. The polarization dependencies of fluorescence generated from two inequivalent crystallographic sites are compared. The experimental results agree with the dressing theoretical calculations well.

I. Introduction

In recent decades, quantum coherence excitation and coherence transfer have been thoroughly studied in atomic gases. These processes lead to many important physical phenomena, such as electromagnetically induced transparency (EIT),\(^1\) enhanced four-wave mixing (FWM)\(^2\) and the group velocity control of traveling light.\(^3,4\) Compared with atomic gases, solid materials are more appropriate for practical applications. Rare-earth ion doped crystals (like Pr\(^{3+}\):Y\(_2\)SiO\(_5\)) exhibit obvious advantages for coherent excitation, such as the absence of atomic diffusion, high density of atoms, compactness, long dephasing times and narrow homogeneous linewidths. The recent research on quantum coherence control in rare-earth ion doped crystals has been reported, including EIT in solid materials,\(^5,6\) enhanced FWM,\(^7\) stimulated Raman adiabatic passage,\(^8\) light velocity reduction and coherent storage,\(^9,10\) controllable erasing of optically stored information\(^11\) and all-optical routing.\(^12\) To realize practical applications, such processes are required to be controlled reliably.

Polarizations of the incident laser beams play important roles in atomic transitions when multi-Zeeman energy levels are involved in the atomic systems. The absorption and fluorescence spectra with different polarizations of probe and pump beams have been studied theoretically and experimentally.\(^13,14\) EIT processes can be controlled by selecting different transitions among Zeeman sublevels via the polarization states of the laser beams.\(^15\) The polarization rotation of an optical beam can also be controlled by another stronger laser beam based on the atomic coherence in multilevel EIT systems.\(^16\) We have investigated the polarization dependencies of FWM in atomic systems previously.\(^17,18\) The results show that the intensity and polarization characteristics of FWM signals can be effectively controlled by the polarization states of the pump and dressing beams.

The aim of this paper is to investigate the polarization dependencies of fluorescence in the Pr\(^{3+}\):Y\(_2\)SiO\(_5\) crystal. The two-level, V-type and \(\Lambda\)-type three-level, as well as N-type four-level systems are studied and compared. The polarization states of generating fields and dressing fields are modulated by a quarter-wave-plate (QWP) and a half-wave-plate (HWP). The dressing state theory is used to explain the experimental results. The paper is organized as follows: in Section II, we briefly introduce the experimental setup; in Section III, the theoretical model is discussed; in Section IV, we show the results and provide our explanation. Section V is our conclusion.

II. Experimental setup

Y\(_2\)SiO\(_5\) is a monoclinic biaxial crystal and belongs to the \(C\(_{2h}\)\) space group. When the Pr\(^{3+}\) ions substitute for the Y\(^{3+}\) ions in Y\(_2\)SiO\(_5\) crystals they occupy two inequivalent crystallographic sites (sites I and II, respectively).\(^19\) The \(\text{^3H}_4\), \(\text{^1D}_2\) levels of Pr\(^{3+}\):Y\(_2\)SiO\(_5\) are split by the crystal field into manifolds consisting of nine and five singlets, respectively. Fig. 1(a) shows the energy levels of the \(\text{^3H}_4\) ground-state multiplets and \(\text{^1D}_2\) excited-state multiplets for the two crystallographic sites of Pr\(^{3+}\) ions. These two crystallographic sites of Pr\(^{3+}\) ions have different Stark splittings, which can be identified by their fluorescence spectra (see Fig. 1(c)). The spectral lines FL1 and FL2 are attributed to the optical transitions from the lowest Stark level \(\gamma_0\) (\(\gamma_0^*\)) of \(\text{^1D}_2\)
to the lowest level $\delta_0$ of the ground-state $^3\text{H}_4$ for sites I and II, respectively. In crystals, each Stark level is further split into three hyperfine states ($\pm 1/2$, $\pm 3/2$, $\pm 5/2$) by the crystal field. The interaction between two centers with different electronic states can happen through phonon-assisted energy transfer. In this case the donor–acceptor pairs consisting of Pr\textsuperscript{3+}(i) and Pr\textsuperscript{3+}(ii) ions form. Moreover, the induced dipole-dipole interaction between Pr\textsuperscript{3+}(i) and Pr\textsuperscript{3+}(ii) ions also happens, so one can treat the two ions as heteronuclear-like molecules. Depending on the interaction between Pr\textsuperscript{3+} ions, the multi-level systems are obtained (see Fig. 1(b)).

Fig. 1 (d) shows the schematic diagram of the experimental arrangement. Our experiments are carried out in a 0.05 at% Pr\textsuperscript{3+} doped Y\textsubscript{2}SiO\textsubscript{5} crystal. The sample (a 3 mm Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} crystal) is held at 77 K in a cryostat (CMF-102). Three tunable dye lasers (narrow scan with a 0.04 cm\textsuperscript{-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 $E_a (\omega_a, A_a), E_b (\omega_b, A_b)$, and $E_c (\omega_c, A_c)$ with the frequency detuning of $\Delta_i = \omega_{\text{mm}} - \omega_i$ ($i = a, b, c$), where $\omega_{\text{mm}}$ denotes the corresponding atomic transition frequency. $E_a$ drives the transition $|a\rangle \leftrightarrow |b\rangle$, $E_b (E_c)$ couples to the transition $|a\rangle \leftrightarrow |c\rangle$ ($|b\rangle \leftrightarrow |d\rangle$). The two-level system is formed when only one pumping field is on. With two pumping fields $E_a$ and $E_b$ (or $E_c$) are on, V-type three-level $|a\rangle \leftrightarrow |b\rangle \leftrightarrow |c\rangle$ (or \Lambda-type three-level system $|a\rangle \leftrightarrow |b\rangle \leftrightarrow |d\rangle$) is set up. When $E_a$ and $E_b$ (or $E_c$) are on, the N-type four-level system $|a\rangle \leftrightarrow |b\rangle \leftrightarrow |c\rangle \leftrightarrow |d\rangle$ is excited. The fluorescence signals are monitored by a photomultiplier tube (PMT) with a fast gated integrator. The HWP, QWP and polarized beam splitter (PBS) are used to control the polarization states of pumping fields.

$$P_{FL1(x)}^{(2)} = \rho_{bb(x,y)}^{(2)} + \rho_{bb(yy,x)}^{(2)} \equiv \frac{-2c_xc_y|G_a|^2\cos 2\theta \sin 2\theta}{d_1 + |G_a|^2(c_x^2 \cos 2\theta + c_y^2 \sin^2 2\theta) + |G_a|^2(c_x^2 \cos 2\theta + c_y^2 \sin^2 2\theta)}/d_1$$

III. Theoretical model

A. Effective nonlinear susceptibilities

Classically, the fluorescence signal intensity is proportional to the square of the polarization induced in the medium. For the second-order fluorescence process, the polarization is $P_k^{(2)} = \rho_{k} \sum_{i,j} |E_i||E_j|$. The Y\textsubscript{2}SiO\textsubscript{5} crystal belongs to the $C_{2v}$ space group and considering that all the incident beams are transverse wave, only four tensor elements are nonzero which are denoted as $\chi_{xyx}^{(2)}, \chi_{yx\overline{y}}^{(2)}, \chi_{x\overline{y}x}^{(2)}$ and $\chi_{y\overline{y}y}^{(2)}$. When HWP\textsubscript{s} are used to change the polarization directions of linear polarized incident beams, the effective nonlinear susceptibilities can be defined as

$$\chi_x = (\chi_{xyx}^{(2)} + \chi_{yx\overline{y}}^{(2)})\cos 2\theta \sin 2\theta,$$

$$\chi_y = (\chi_{yx\overline{y}}^{(2)} + \chi_{y\overline{y}y}^{(2)})\sin^2 2\theta,$$

where $\theta$ is the rotated angle of the HWP\textsubscript{s}' axis from the x axis. If the polarization states are changed by QWP, the effective nonlinear susceptibilities are

$$\chi_x = (\chi_{xyx}^{(2)} + \chi_{yx\overline{y}}^{(2)})\sqrt{\cos^4 \theta + \sin^4 \theta + \frac{1}{2}(\sin^2 \theta \cos^2 \theta)},$$

$$\chi_y = (\chi_{yx\overline{y}}^{(2)} + \chi_{y\overline{y}y}^{(2)})\sin^2 2\theta.$$

B. Density matrix element

In quantum theory, the fluorescence signal intensity can also be described by the diagonal density matrix elements. In the two-level system, with a strong pumping field $E_a (E_b)$ on, the fluorescence signal FL1 (FL2) is generated through the perturbation chains $\rho_{aa}^{(0)} \to \rho_{aa}^{(1)} \to \rho_{bb}^{(2)}$ and $\rho_{aa}^{(0)} \to \rho_{ab}^{(1)} \to \rho_{bb}^{(2)} \to \rho_{bb}^{(2)} \to \rho_{bb}^{(2)} \to \rho_{bb}^{(2)} \to \rho_{bb}^{(2)}$. Considering the self-dressing effect of $E_a (E_b)$, the diagonal density matrix element $\rho_{bb}^{(2)}$ is given by

$$\rho_{bb}^{(2)} = |G_a|^2/[d_1 + |G_a|^2/G_{bb}(G_{bb} + |G_a|^2/d_1)],$$

$$\rho_{bb}^{(2)} = |G_b|^2/[d_2 + |G_b|^2/G_{bb}(G_{bb} + |G_b|^2/d_2)],$$

where $d_1 = G_{bb} + iA_{ba}, d_2 = G_{bb} + iA_{pb}, G_i = -\mu_i E_i/h$ is the Rabi frequency of $E_i$ with the electric dipole moment $\mu_i$ between levels $|i\rangle$ and $|j\rangle$, and $G_{ij}$ is the transverse decay rate. When the polarization direction of $E_a$ is changed by a HWP, the diagonal density matrix element $\rho_{bb}^{(2)}$ can be described as

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where $c_{xy}$ is the anisotropic factor denoting the different susceptibilities $\chi^{(2)}_{xy}$, $\chi^{(2)}_{yx}$ in different directions.

In the V-type three-level system, when two pumping fields $E_a$ and $E_b$ are both on, two fluorescence signals FL1 and FL2 are generated simultaneously. The intensity of the total fluorescence signals is the sum of two signals. Moreover, these two processes can interact with each other. Considering the coherence of the system, such a process can be described by the fourth-order coherence process as

$$
\rho^{(4)}_{bb} = \frac{-|G_{bb}|^2}{(d_2 + |G_b|^2/G_{bb} + |G_a|^2/d_2)} \left( \Gamma_{bb} + |G_a|^2/d_1 + |G_b|^2/d_2 \right)
\times \frac{-|G_a|^2}{(d_1 + |G_a|^2/\Gamma_{bb} + |G_b|^2/d_2)} \left( \Gamma_{bb} + |G_a|^2/d_1 \right).
$$

The intensity of the total fluorescence signals is the summing contribution of each transition path. When $E_a$ is linearly polarized, only $|a\rangle \leftrightarrow |b\rangle$ ($j = \pm 1/2, \pm 3/2, \pm 5/2$) vertical transition pathways are allowed. Although there are several perturbation chains for the generated nonlinear signal, it is reasonable to choose one of them to analyze the physical mechanism. For example, the diagonal density matrix element for the $|a_{21}\rangle \leftrightarrow |b_{32}\rangle$ transition pathway can be written as

$$
\rho^{(4)}_{bb} = \frac{-|G_{bb}|^2}{(d_2 + |G_b|^2/G_{bb} + |G_a|^2/d_2)} \left( \Gamma_{bb} + |G_a|^2/d_1 + |G_b|^2/d_2 \right)
\times \frac{-|G_a|^2}{(d_1 + |G_a|^2/\Gamma_{bb} + |G_b|^2/d_2)} \left( \Gamma_{bb} + |G_a|^2/d_1 \right).
$$

The left-circularly and right-circularly polarized beams drive the transitions $|a_{j+1}\rangle \leftrightarrow |b_j\rangle$ and $|a_j\rangle \leftrightarrow |b_{j+1}\rangle$, respectively. The density matrix elements corresponding to $|a_{3/2}\rangle \leftrightarrow |b_{3/2}\rangle$ and $|a_{-3/2}\rangle \leftrightarrow |b_{-3/2}\rangle$ are

$$
\rho^{(4)}_{bb} = \frac{-|G_{bb}|^2}{(d_2 + |G_b|^2/G_{bb} + |G_a|^2/d_2)} \left( \Gamma_{bb} + |G_a|^2/d_1 + |G_b|^2/d_2 \right)
\times \frac{-|G_a|^2}{(d_1 + |G_a|^2/\Gamma_{bb} + |G_b|^2/d_2)} \left( \Gamma_{bb} + |G_a|^2/d_1 \right).
$$
where $G_i^0$, $G_i^r$ and $G_i^i$ are the Rabi frequencies for linearly, left- and right-circularly polarized beams, respectively. So it will show different transition strengths induced by different Rabi frequencies in Zeeman sublevels.

In the N-type three-level system, when the polarization direction of $E_a$ is changed by HWP, the modulated fluorescence signal FL1 is generated. The density matrix elements can also be described by the fourth-order coherence process.

First, we investigate the fluorescence signal in the two-level system. In the N-type four-level system with three pumping fields frequencies in Zeeman sublevels.

IV. Results and discussion

First, we investigate the fluorescence signal in the two-level system. Fig. 3(a) and (b) show the signals FL1 ($|a\rangle \leftrightarrow |b\rangle$) and FL2 ($|a\rangle \leftrightarrow |c\rangle$) at different polarization directions with one pumping field $E_a$, respectively. Each fluorescence curve shows the Autler–Townes-like (AT) splitting that was caused by the saturation absorption effect of the strong beam $E_a$. The self-dressing effect of $E_a$ on spectral splitting is reflected by $|Ga|/d_1$ (see eqn (3)). In Fig. 3(a), with the rotation angle of the HWP changing from 0° to 45° (the dressing term changes from $c_2^2|Ga|^2/d_1$ to $c_2^2|Ga|^2/d_1$ in eqn (4b)), the depth and width of the suppressed dip reduce, and the fluorescence emission peaks increase gradually. It means that the dressing effect of $E_a$ is greater at 0° (horizontal polarization) than that at 45° (vertical polarization). The baseline of each signal which is the non-resonant fluorescence signal also reduces with an increase in the rotation angle. It can be interpreted by the change in the numerator from $c_2^2|Ga|^2$ to $c_2^2|Ga|^2$ in eqn (4b). In this case, the strong field $E_a$ acts as the dressing field and the generating field simultaneously. For FL2 (Fig. 3(b)), the evolution of the baseline is the same as FL1, but the variation of the AT splitting is unapparent at different polarization directions. It indicates that...
the level splitting of $\gamma^*_c$ (sites II) is insensitive to the polarization direction of the dressing field. Sites I and II are two inequivalent crystallographic sites. Fig. 3(c) and (d) are the signals FL1 and FL2 with two pumping fields, respectively. The dressing field $E_0$ is resonant with relevant transitions (satisfies the suppression condition $A_b = 0$), while the frequency detuning of the generating field $E_0$ is scanned. The polarization direction of $E_0$ is changed by HWP. One can see that the fluorescence baselines for both FL1 and FL2 increase with the rotation angle $\theta_1$ of HWP. Compared with the case pumped by one field (Fig. 3(b)), the suppressed dip of FL2 with two pumping fields becomes more obvious and the fluorescence emission peaks disappear. This attributes to the external-dressing effect of $E_0$. The coupling between the strong field $E_0$ and particles leads to the generation of a dark state, and the external-dressing splitting $(|G_b|^2/d_{02})$ overlaps with the self-dressing splitting $(|G_a|^2/d_4)$. In this case, eqn (4b) is rewritten as

$$P_{FL1(y)}^{(2)} = \rho_{bb}^{(2)}(xxy) + \rho_{bb}^{(2)}(yy) = \frac{-|G_a|^2}{\left(d_{1} + |G_a|^2/\Gamma_{bb} + c^2|G_b|^2\cos^22\theta_2/d_{02}\right) \left(\Gamma_{bb} + |G_a|^2/d_{1} + c^2|G_b|^2\cos^22\theta_2/d_{02}\right)}$$

where $d_{02} = \Gamma_{bb} + iA_b$ and $d_{02} = \Gamma_{bb} + i(A_a - A_b)$. Because the polarization direction of the external-dressing field is changed, the intensity of the fluorescence baseline depends on the dressing term $c^2|G_b|^2\cos^22\theta_2/d_{02}$ of $E_0$. We can see that the suppression effect of $E_0$ on the fluorescence baseline is greater at 0° ($c^2|G_b|^2/d_{02}^2$) than that at 45° ($c^2|G_b|^2/d_{02}^2$). For FL1, the baseline is suppressed even to zero at 0°, but the suppression effect on the baseline of FL2 is smaller than FL1. We can draw the conclusion that polarization dependencies for FL1 and FL2 are different. Such a difference results from the crystallographic anisotropy of the $Y_2SiO_5$ crystal.

Now, we investigate the fluorescence signal in the V-type three-level system (Fig. 4(a) and (b)). In this case, the intensity of the fluorescence baseline is the sum of two signals FL1 and FL2, so the measured results are the combinative action of dressing and excitation effects. The AT splitting results from the dressing effects of $E_0 (|G_a|^2/d_{41})$ and $E_b (|G_b|^2/d_{02})$ on the level $|a\rangle$. For FL1 (Fig. 4(a)), the fluorescence baseline raises as the dressing field $E_0$ is changed from horizontally to vertically polarized, which can be interpreted by different dressing effects of $E_0$ (changing from $c^2|G_b|^2/\Gamma_{cc}$ to $c^2|G_b|^2/\Gamma_{cc}$). Compared with the two-level system, the fluorescence baseline is not suppressed to zero at 0°. It is due to the generation of fluorescence FL2 from level $|c\rangle$. The width of the suppressed dip reduces gradually with $\theta_2$ but the depth does not change. For FL2 (Fig. 4(b)), the suppression effect of dressing field $E_0$ at 0° is greater than that in the two-level system, and the depth of the suppressed dip reduces gradually with $\theta_1$. This is because the dressing effect of $E_0$ is sensitive to the polarization.

Fig. 4(c) shows the signal FL1 in the $\Lambda$-type three-level system and the polarization direction of the dressing field $E_0$ is changed by HWP. It is different from the V-type three-level system that the baseline reduces gradually with $\theta_3$, and the fluorescence emission peaks become obvious. As described in eqn (7), these phenomena can be interpreted by the cooperation...
of the numerator \( \varepsilon^2 |G_a| \cos^2 \theta \) and the dressing term \( \varepsilon^2 |G_a| \cos^2 \theta_3/\Gamma_{bb} \). The AT splitting of emission peaks is attributed to the dressing effects of \( |E_a|/|d_1| \) and \( |E_b| \varepsilon^2 |G_a| \cos^2 \theta_3/\Gamma_{cb} \) on level |b\>. When three fields \( E_a, E_b \) and \( E_c \) are all on, a N-type four-level system is set up. We keep the dressing fields \( E_b \) and \( E_c \) close to the resonant point and scan the detuning of the generating field \( E_a \). One can see in Fig. 4(d) that the baseline is greater than that in the \( \Lambda \)-type three-level. It is due to the generation of fluorescence FL2 by \( E_b \). There is excitation competition on the particles of the ground state between \( E_a \) and \( E_b \). Because of the further dressing by \( E_b \), the emission peaks of FL1 reduce and the suppressed dips become deeper.

Next, we use a QWP to modulate the polarization states of pumping fields. Fig. 5(a) and (b) illustrate the fluorescence signals FL1 and FL2 in a V-type three-level system when the field \( E_a \) is changed by QWP, respectively. For FL1, when the polarization state of \( E_a \) changes from left-circularly polarized to linearly polarized, and then to right-circularly polarized (the QWP rotating from \(-45^\circ \) to \(0^\circ \) and then to \(45^\circ \)), we can see both fluorescence baseline and AT splitting evolve from weak to strong and then to weak. Fig. 2 shows transition paths and CG coefficients at different laser polarization states. Although there are some transition paths for the generated fluorescence signal, considering the population of each level, the optical transition from the lowest crystal-field level \( |\pm 5/2 \rangle \) is the dominant one. Since \( E_a \) is not only the dressing field but also the generating field for FL1, we should simultaneously consider the self-dressing role and the generating role. Because the Rabi frequency \( G_a^{25/12} \) for the linearly polarized state is greater than \( G_a^{25/2} \) \( (G_a^{15/2} \) for the left-(right-) circularly polarized state, the baseline (decided by \( |G_a^{25/12}|^2 \) in the numerator of eqn 6(a)) and AT splitting (decided by \( |G_a^{15/2}|^2/\Gamma_{bb} \) are stronger at \(0^\circ \) than that at \( \pm 45^\circ \). For FL2, \( E_a \) mainly acts as a dressing field. The suppressed effect of the linearly polarized field \( |G_a^{25/12}|^2/\Gamma_{bb} \) is stronger than left-(|\( G_a^{25/2}|^2/\Gamma_{bb}^{2b5/2} \) and right-circularly \( |G_a^{15/2}|^2/\Gamma_{b=2b5/2} \) polarized fields, so the lowest fluorescence baseline occurs at \( \theta_1 = 0^\circ \).

Fig. 5(c)–(h) show the fluorescence signals in N-type four-level systems. When the polarization state of \( E_a \) is modulated (shown in Fig. 5(c) and (d)), the evolution of the fluorescence baseline and suppressed dips is as similar as that in the V-type three-level, but the signals are further suppressed by the multi-dressing effect of \( |E_a|/\Gamma_{aa} \) and \( E_b \) \( (|G_b|/\Gamma_{bc} \) when the polarization state of \( E_b \) is modulated by QWP, both fluorescence signals FL1 (Fig. 5(e)) and FL2 (Fig. 5(f)) show a little change in the rotation angle \( \theta_2 \). This means that the level of site II is insensitive to the polarization state of the pumping field. Fig. 5(g) and (h) show signals FL1 and FL2, respectively, as the dressing field \( E_c \) is modulated by QWP. According to eqn (8), the dressing effect of \( E_c \) on level |b\> of FL1 is determined by \( |G_c|/\Gamma_{cc} \). The Rabi frequency of the linearly polarized field is greater than the circularly polarized field, so the AT splitting at \(0^\circ \) is stronger than that at \( \pm 45^\circ \). On the other hand, \( E_c \) also acts as a generating field for FL1, so the baseline excited by \( G_c^{25/2} \) at \(0^\circ \) is greater than that excited by \( G_c^{15/2} \) at \( \pm 45^\circ \). In Fig. 5(h), one can see that the fluorescence signal FL2 doesn’t change with the rotation angle \( \theta_3 \). This is because \( E_c \) does not dress the level |a\> and |c\> of FL2. For FL2, it is mainly dressed by \( E_a \).

V. Conclusion

In summary, we have reported the polarization dependencies of fluorescence signals in two-level, V-type and \( \Lambda \)-type three-level, as well as N-type four-level systems of the Pr\(^{3+}\):Y\(_2\)SiO\(_5\) crystal. The suppressed dips, emission peaks and fluorescence baselines show different evolvements as the polarization states of generating fields and dressing fields are changed by QWP or HWP. For the level of site I, the dressing effects of horizontal and linearly polarized fields are greater than that of vertical and circularly polarized fields. However, the dressing effect on the level of site II is insensitive to the polarization state. Our experimental data are in good agreement with the theoretical results from dressing effect analysis. Furthermore, our results provide an effective method to control high-order fluorescence processes.

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