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Competition between spontaneous parametric four-wave mixing and fluorescence in Pr\textsuperscript{3+}:YSO

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**Abstract**

We report the competition between the spontaneous parametric four-wave mixing and second- or fourth-order fluorescence (FL) signals in Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} crystal. By changing the powers of controlling fields and blocking different fields, the competitive two signals can be identified through the dressing effect. Meanwhile, the delay of fourth-order FL in the time domain results from splitting the energy level. Such results can find potential applications in optical information storage and processing on a photonic chip.

**Keywords:** multi-wave mixing, fluorescence, atomic coherence

(Some figures may appear in colour only in the online journal)

1. Introduction

Since rare-earth-ion-doped crystal, for example Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5}, has unique properties in comparison to atomic vapors, such as long coherence times for the optical and optical controllability of the ionic states [1], atomic coherence-induced effects in solid materials attract a lot of attention. Research like electromagnetically induced transparency (EIT [2–4]), light coherent storage [5–7], all-optical routing [8], optical velocity reduction and reversible storage of double light pulses [5, 9], enhanced four-wave mixing (FWM [10]), all-optically controlled higher-order fluorescence (FL [11]), has been realized with such crystal. Recently, based on the studies of narrow-bandwidth paired photon [12] and FWM [13], coexisting parametric amplification and cascaded nonlinearity processes [14] are reported.

In this letter, the spontaneous parametric FWM (SP-FWM) processes as well as the FL signals are investigated both theoretically and experimentally in Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} crystal. By changing the power of controlling fields and opening different dressing fields, a strong SP-FWM is demarcated from a second- or fourth-order FL signal in the same channel caused by the dressing effect. The two signals can be separated in the time domain, accompanied by a long delay time in the FL decay process, due to splitting the energy level.

2. Experimental setup and basic theory

2.1. Experimental setup

In the experiment, the sample is a thick Pr\textsuperscript{3+}:YSO crystal whose Pr\textsuperscript{3+} concentration is 0.05 at % and is kept at 77 K. The Pr\textsuperscript{3+} impurity ions occupy two nonequivalent cation sites (sites I and II, respectively) in the YSO crystal lattice.

Figure 1(a) shows an inverse N-type (RN) level system constructed by V-type (|0\rangle ↔ |1\rangle |2\rangle ) and A-type (|0\rangle |3\rangle ↔ |1\rangle ) systems. In the system, we consider two lower states |0\rangle (\delta_0 + \delta_n), which is a heteronuclear-like molecule energy level [11, 15], and |3\rangle (\delta_1 + \delta_l), and two excited states
\[ |2\rangle (\gamma_0 + \gamma_6) \] and \(|1\rangle (\gamma_0 + \gamma_6^*), \] where \(\delta_0\) and \(\gamma_0\) are from site I, and \(\delta_0^*\) and \(\gamma_0^*\) are from site II [11]. Five driving fields \(E_1\) (with frequency \(\omega_1\), wave vector \(k_1\), and the Rabi frequency \(G_1\)), \(E_2\) and \(E_2'\) (\(\omega_2, k_2\) and \(k_2', G_2\) and \(G_2'\)), \(E_3\) and \(E_3'\) (\(\omega_3, k_3\) and \(k_3', G_3\) and \(G_3'\)) are from three near-transform-dye lasers (repetition rate: 10Hz, pulse width: 5ns, line width: 0.04 cm^{-1}) pumped by an injection locked single-mode Nd:YAG laser with a frequency detuning of \(\Delta_1 = \omega_{\text{imu}} - \omega_i (i = 1, 2, 3)\) and Rabi frequency \(G_i = \mu_{\text{imu}}E_i/\hbar\) (\(G_i' = \mu_{\text{imu}}E_i/\hbar\)) with power \(P_i\), respectively, where \(\omega_{\text{imu}}\) and \(\mu_{\text{imu}}\) denote the transition frequency and dipole moment between \(|m\rangle\) and \(|p\rangle\), and \(E_i\) is the electrical field intensity of \(E_i\). These laser beams are in the same plane \(x-o-y\) as shown in figure 1(c). Vertically polarized \(E_2\) and \(E_3\) propagate in the opposite direction to \(E_1\). The propagation directions of horizontally polarized beams \(E_2'\) and \(E_3'\) are the opposite, and have a small angle (~0.5°) with \(E_2\) and \(E_1\). In figure 1(b1), two weak field anti-Stokes (\(E_{S2}\)) and Stokes (\(E_{S1}\)) of the SP-FWM are generated by a strong pumping field \(E_1\) and the reflected beam \(E_{1r}\) (\(\Delta_{1r} = \Delta_1\)) due to the so-called phase-conjugate SP-FWM process. Similarly, the SP-FWM signal can also be generated in a \(X\)-type level system with opening \(E_1\) and \(E_2\) (\(E_3\) as shown in figures 1(b2) and (b3)). Such a SP-FWM satisfies the phase-matching condition \(k_s + k_{S2} = k_1 + k_i (i = 1, 2, 3)\) in figure 1(d1). The emitted signals \(E_S\) and \(E_{S2}\) form a spatial conical alignment, as shown in figure 1(d2), but the FL is a non-coherent signal. Therefore, the pair of pure SP-FWM (\(E_S\) and \(E_{S2}\)) are only detected by two photomultiplier tubes (PMT1 and PMT2) along the direction of \(E_1'\) and \(E_{1r}'\), and a composite signal (including FL signal and SP-FWM) is monitored by another detector PMT3, as shown in figure 1(c).

2.2. Basic theory

Theoretically, the coupled signals \(E_S\) and \(E_{S2}\) are generated in three different level systems. One case is open \(E_1\) (\(E_2\) or \(E_3\)). For instance, a pair SP-FWM is generated by \(E_1\) and its reflected beam \(E_{1r}\) between \(|0\rangle\) and \(|1\rangle\). Considering Liouville’s pathways \(\rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{20}^{(2)} \rightarrow \rho_{30}^{(3)}\) and \(\rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{20}^{(2)} \rightarrow \rho_{30}^{(3)}\) with self-dressing effect, the density matrix elements are written as

\[
\rho_{10}^{(3)} (S_1) = - i G_1 G_{S2}^* G_1 / [(d_1 + |G_1|^2 / \Gamma_{10}) (d_0 + |G_1|^2 / d)] \quad (1a)
\]

\[
\rho_{10}^{(3)} (S_2) = - i G_2 G_{S2}^* G_2 / [(d_2 + |G_2|^2 / \Gamma_{20}) (d_0 + |G_2|^2 / d)] \quad (1b)
\]

where \(d_1 = \Gamma_{10} + \Gamma_{11}\) and the frequency detuning \((\Delta_{10})\) of \(E_{1r}\) is equal to \(\Delta_1\). Another pair of SP-FWM is generated by \(E_1\) and \(E_2\) in the \(V\)-type energy level systems, the density matrix elements of \(E_{S2}\) and \(E_{S2}\) with external dressing effect of \(E_3\) can be written as

\[
\rho_{10}^{(3)} (S_2) = - i G_2 G_{S2}^* G_2 / [(d_2 + |G_2|^2 / \Gamma_{20}) (d_1 + |G_2|^2 / d_2)] \quad (2a)
\]

\[
\rho_{10}^{(3)} (S_3) = - i G_3 G_{S2}^* G_3 / [(d_3 + |G_3|^2 / \Gamma_{30}) (d_1 + |G_3|^2 / d_3)] \quad (2b)
\]

through the pathways \(\rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{20}^{(2)} \rightarrow \rho_{30}^{(3)}\) and \(\rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{20}^{(2)} \rightarrow \rho_{30}^{(3)}\), where \(d_{30} = \Gamma_{30} + i(\Delta_1 - \Delta_3)\), \(d_2 = \Gamma_{20} + i\Delta_2\). Similarly, in the \(\Lambda\)-type energy level systems, the density matrix elements of \(E_{S3}\) and \(E_{S3}\) with an external dressing effect of \(E_2\) can be described as

\[
\rho_{10}^{(3)} (S_3) = - i G_3 G_{S3}^* G_3 / [(d_3 + |G_3|^2 / \Gamma_{30}) (d_1 + |G_3|^2 / d_3)] \quad (3a)
\]

\[
\rho_{10}^{(3)} (S_3) = - i G_3 G_{S3}^* G_3 / [(d_1 + |G_3|^2 / \Gamma_{30}) (d_3 + |G_3|^2 / d_3)] \quad (3b)
\]
determined by the pathways $\rho_{12}^{(0)} = \rho_{12}^{(1)} = \rho_{12}^{(2)} = \rho_{12}^{(3)}$ and $\rho_{13}^{(0)} = \rho_{13}^{(1)} = \rho_{13}^{(2)}$, where, $d_3 = \gamma_{13} + i \gamma_{13}, \gamma_{13} = \Gamma_{13} + i \Delta$.

The coupling SP-FWM process can be expressed by Hamiltonian [16] $H = \{\hat{a}_{\text{s}}^{\dagger}(\Delta_{\text{s}}) + \hat{a}_{\text{s}}(\Delta_{\text{s}})\} \gamma / \nu$, where $\hat{a}_{\text{s}}(\Delta_{\text{s}})$ is the Boson-creation (-annihilation) operator acting on the electromagnetic excitation of the Stokes channel, whereas $\hat{a}_{\text{s}}^{\dagger}(\Delta_{\text{s}})$ acts on the anti-Stokes channel. $\nu$ is the group velocity of light in the nonlinear medium. The nonlinear coupling coefficient $g = -\gamma_{\text{s}} \gamma_{\text{s}}^{\dagger} / 2\pi$ relies on the nonlinear susceptibilities $\chi^{(2)}_{\text{s}}$ and the pump-field amplitude, where $\chi^{(2)}_{\text{s}} = \left(\gamma_{\text{s}}^{(2)} E_{\text{s}} / 2 \nu \right. \left. \text{s} \right)$.

Then, when $E_{\text{s}}$ and $E_{\text{s}}$ signals propagate in the nonlinear medium, the intensity of the output signals are proportional to the photon numbers. The photon numbers can be expressed as

$$N_{\text{s}} = \left\{ \hat{a}_{\text{s}}^{\dagger} \hat{a}_{\text{s}} \right\} = \left\{ \text{cosh} \left[ 2t \sqrt{g_{\text{s}} g_{\text{s}}^{\dagger}} \cos \varphi + \sin \left[ 2t \sqrt{g_{\text{s}} g_{\text{s}}^{\dagger}} \sin \varphi \right] \right\} g_{\text{s}} / 2 g_{\text{s}2}$$

$$N_{\text{s}} = \left\{ \hat{a}_{\text{s}}^{\dagger} \hat{a}_{\text{s}} \right\} = \left\{ \text{cosh} \left[ 2t \sqrt{g_{\text{s}} g_{\text{s}}^{\dagger}} \cos \varphi + \sin \left[ 2t \sqrt{g_{\text{s}} g_{\text{s}}^{\dagger}} \sin \varphi \right] \right\} g_{\text{s}} / 2 g_{\text{s}}$$

(4a)

(4b)

where $\varphi = (\varphi_{\text{s}} + \varphi_{\text{s}}) / 2$, $\varphi_{\text{s}}$ and $\varphi_{\text{s}}$ are phase angles of $E_{\text{s}}$ and $E_{\text{s}}$.

The FL signal is generated accompanying the SP-FWM process. The intensity is proportional to the diagonal density matrix elements theoretically. In this system, with opening $E_1$ (E2 or E3), via two mutually conjugate Liouville pathways $\rho_{10}^{(0)} = \rho_{10}^{(1)} = \rho_{10}^{(2)} = \rho_{10}^{(3)}$, and $\rho_{12}^{(0)} = \rho_{12}^{(1)} = \rho_{12}^{(2)} = \rho_{12}^{(3)}$, the corresponding density matrix elements $\rho_{11}^{(0)} = \rho_{11}^{(1)} = \rho_{11}^{(2)} = \rho_{11}^{(3)}$ of second-order FL are expressed as

$$\rho_{11}^{(0)} = \rho_{11}^{(1)} = \rho_{11}^{(2)} = \rho_{11}^{(3)}$$

(5a)

$$\rho_{12}^{(0)} = \rho_{12}^{(1)} = \rho_{12}^{(2)} = \rho_{12}^{(3)}$$

(5b)

$$\rho_{13}^{(0)} = \rho_{13}^{(1)} = \rho_{13}^{(2)} = \rho_{13}^{(3)}$$

(5c)

As a result, with all beams on, the total intensity of the two-order FL signal can be described as $I_{\text{FL}} = \rho_{11}^{(1)} + \rho_{12}^{(1)} + \rho_{13}^{(1)}$.

Next, when $E_1, E_2$ and $E_3$ open, considering the interplays among $\rho_{1i}^{(n)}(i = 1, 2, 3)$ [11] and the dressing effects, three types of fourth-order FL are obtained, which can be expressed as

$$\rho_{11}^{(4)} = \left\{ \left[ G_{11}^{(2)} + \left( d_2 + |G_{12}^{(2)}| f_{11}^{(1)} + |G_{13}^{(2)}| f_{12}^{(1)} \right) \right] \right\}$$

$$\times \left\{ \left[ f_{10}^{(0)} + \left( d_1 + |G_{11}^{(0)}| f_{10}^{(1)} + |G_{12}^{(0)}| f_{10}^{(2)} \right) \right] \right\}$$

$$\times \left\{ \left[ f_{10}^{(0)} + \left( d_1 + |G_{11}^{(0)}| f_{10}^{(1)} + |G_{12}^{(0)}| f_{10}^{(2)} \right) \right] \right\} + \text{H.c.}$$

(6a)

$$\rho_{12}^{(4)} = \left\{ \left[ G_{11}^{(2)} + \left( d_2 + |G_{12}^{(2)}| f_{12}^{(1)} + |G_{13}^{(2)}| f_{12}^{(1)} \right) \right] \right\}$$

$$\times \left\{ \left[ f_{10}^{(0)} + \left( d_1 + |G_{11}^{(0)}| f_{10}^{(1)} + |G_{12}^{(0)}| f_{10}^{(2)} \right) \right] \right\}$$

$$\times \left\{ \left[ f_{10}^{(0)} + \left( d_1 + |G_{11}^{(0)}| f_{10}^{(1)} + |G_{12}^{(0)}| f_{10}^{(2)} \right) \right] \right\} + \text{H.c.}$$

(6b)

$$\rho_{13}^{(4)} = \left\{ \left[ G_{11}^{(2)} + \left( d_2 + |G_{13}^{(2)}| f_{13}^{(1)} + |G_{13}^{(2)}| f_{13}^{(1)} \right) \right] \right\}$$

$$\times \left\{ \left[ f_{10}^{(0)} + \left( d_1 + |G_{11}^{(0)}| f_{10}^{(1)} + |G_{13}^{(0)}| f_{10}^{(2)} \right) \right] \right\}$$

$$\times \left\{ \left[ f_{10}^{(0)} + \left( d_1 + |G_{11}^{(0)}| f_{10}^{(1)} + |G_{13}^{(0)}| f_{10}^{(2)} \right) \right] \right\} + \text{H.c.}$$

(6c)

described by the pathways $\rho_{10}^{(0)} = \rho_{10}^{(1)} = \rho_{10}^{(2)} = \rho_{10}^{(3)}$, and $\rho_{22}^{(0)} = \rho_{22}^{(1)} = \rho_{22}^{(2)} = \rho_{22}^{(3)}$, and $\rho_{33}^{(0)} = \rho_{33}^{(1)} = \rho_{33}^{(2)} = \rho_{33}^{(3)}$, and $\rho_{12}^{(0)} = \rho_{12}^{(1)} = \rho_{12}^{(2)} = \rho_{12}^{(3)}$, and $\rho_{13}^{(0)} = \rho_{13}^{(1)} = \rho_{13}^{(2)} = \rho_{13}^{(3)}$.

The four-order FL in the V-type level system, $E_3$ is the external dressing field and $E_2$ is the external dressing field for fourth-order FL (equation 6c) in the A-type level system.

According to equations (1)–(6), we can see that the SP-FWM and FL signals can be controlled by atomic coherence (dressing effect). But the dressing effect in SP-FWM is less sensitive than that in the FL signal [14].

3. Experimental results and discussions

Firstly, we investigated competition between SP-FWM and second-order FL signals in two different sites of Pr3+:Y2SiO5 crystal.

Figure 2 shows pure SP-FWM ($E_3$ and $E_3$) and the second-order FL signal with $E_1$ on only and being scanned. In the first case, the beam is arranged to drive the levels $|0\rangle \leftrightarrow |1\rangle$. In figures 2(a) and (b), when $P_1$ changes from 0.5 to 5.5 mW, the emission peaks (the profiles are indicated by dashed curves) of pure $|E_3\rangle$ and $|E_3\rangle$ increase monotonously, due to the enhanced intensity of pumping field $E_1$, which determines the intensities of generation signals (equations (1a) and (1b)). In the composite channel (figure 2(c), the baseline (short-dashed curve), the non-resonant FL signal $G_{11}$ in equation (5a), increases gradually with increasing $P_1$. When $P_1$ is increased, the FL signal evolves from Autler–Townes (AT)-like splitting to pure suppression, caused by the saturation absorption effect of the strong beam $E_1$ and reflected beam $E_3\gamma$, and the self-dressing effect of $E_1 (|G_{11}|^{2} |G_{11}|)$ and $|G_{11}|^{2} d_{2}$ in equation (5a)). The profile of the suppression dips ($\lambda = 0$) is described by the dashed curve in figure 2(c). When $P_1$ is large enough, the suppression dip tends to be invariant and approaches the background
of the first curve because FL is suppressed to minimum at \( \Delta_1 = 0 \) by the dressing effect of \( E_1 \). In particular, a small emission peak—the SP-FWM \( (E_S) \)—is present in the suppression dip of FL (figure 2(h1), the zooming in of the last curve in figure 2(c)). The reason is that the coherent radiation process is strengthened, accompanied by a strong suppression of the FL signal. In order to demonstrate the phenomena more clearly, the corresponding theoretical simulations of composite signals are presented in figure 2(g). Theoretically, when \( P_1 \) increases, the FL signal is suppressed and the SP-FWM in the spatial emission cone is enhanced gradually. So, the composite signal in figure 2(g3) is composed of the FL (figure 2(g1)) and SP-FWM (figure 2(g2)). Zooming in the last curve in figure 2(g3) specifies the emission in the dip, shown as in figure 2(h2). By comparison, the SP-FWM (figures 2(a) and (b)) with narrow line-width is loaded into the suppression dip of FL (figure 2(c)) due to the lesser dressing effect in SP-FWM.

In the second case, when \( E_1 \) drives \( \{0\} \leftrightarrow \{1\} \), the signals are shown in figures 2(d)–(f) with similar conditions used in figures 2(a)–(c). The emission peaks of pure \( E_S \) and \( E_{\text{AS}} \) signals generated in \( \{0\} \leftrightarrow \{2\} \) (figures 2(d) and (e)) and the background signal (short-dashed curve in figure 2(f)) of second-order FL from \( \{2\} \) to \( \{0\} \) will rise gradually with increasing \( P_1 \) for the same reason as in figures 2(a)–(c). However, as \( P_1 \) increases, the scanning curves of second-order FL in figure 2(f) changes from the emission peak to AT-like splitting, and then to a pure suppression dip (the profile of the resonant point is indicated by the dashed curve), which is different from that in figure 2(c). The reason is that the dipole moment \( \mu_{01} \) is four times larger than \( \mu_{02} \), so the Rabi frequency \( G_1 \) in \( \{0\} \leftrightarrow \{1\} \) is larger than that in \( \{0\} \leftrightarrow \{2\} \) with the same power \( P_1 \), which can lead to larger suppression of the FL signal and an enhanced atomic coherence effect.

Next, the competition of SP-FWM and fourth-order FL is investigated by opening different dressing fields in the RN-type level system (figure 3) and changing the power of \( P_1 \). Figure 3(a) shows the measured signal versus \( \Delta_1 \) from the composite channel by opening \( (a1) \ E_1, (a2) \ E_1 \) and \( E_2 \) \( (a3) \ E_1 \) and \( E_2 \), \( (a4) \ E_1 \) and \( E_2 \), \( (a5) \ E_1 \) and \( E_2 \), and \( (a6) \) all the laser beams in the RN level system, respectively. All curves share the same scale. (c1)–(c3) are corresponding to \( (a1) \), \( (a3) \) and \( (a6) \), respectively. Inset is the gate width of boxcar integrator in (c4). (d) Dressed-state pictures for the (d1) V-type and (d2) A-type level system, respectively.

![Figure 2](image1.png)

**Figure 2.** Evolutions of (a) pure \( E_0 \), (b) pure \( E_{\text{AS}} \) and (c) composite signal (SP-FWM and FL) by scanning \( E_1 \) in \( \{0\} \leftrightarrow \{1\} \) with \( P_1 \) being 0.5, 1.5, 2.5, 3.5, 4.5 and 5.5 mW from left to right. (d)–(f) Same as (a)–(c), but scanning \( E_1 \) in \( \{0\} \leftrightarrow \{2\} \). (g) The simulations of signals (1) FL, (2) SP-FWM and (3) the total signal, respectively. The condition is the same as in (c). (h1) and (h2) Zooming in on the last curve in (b) and the corresponding theoretical curve, respectively.

![Figure 3](image2.png)

**Figure 3.** (a) Composite signal versus \( \Delta_1 \) modulated by opening \((a1) \ E_1, (a2) \ E_1 \) and \( E_2 \), \((a3) \ E_1 \) and \( E_2 \), \((a4) \ E_1 \) and \( E_2 \), \((a5) \ E_1 \) and \( E_2 \), \((a6) \) all the laser beams in the RN level system, respectively. All curves share the same scale. (b) Corresponding theoretical results with the same condition as in (a) at fixed detuning \( \Delta_2 = \Delta_3 = 0 \). (c) Intensity of the composite signal at \( \Delta_1 = 0 \) in the time domain. (c1)–(c3) are corresponding to \((a1) \), \((a3) \) and \((a6) \), respectively. Inset is the gate width of boxcar integrator in (c4). (d) Dressed-state pictures for the (d1) V-type and (d2) A-type level system, respectively.
FL signal in figure 3(a1) has been discussed in figure 2(c). When \( E_1 \) and \( E_2' \) are open, an obvious AT splitting appears in the emission peak of the fourth-order FL signal (figure 3(a2)) due to the self-dressing effect of \( E_2' \) which splits \([0] \) into \([G_1 \pm] \) as shown in figure 3(d1) (determined by \( |G_3\rangle^2d_{d2} \) and \( |G_2\rangle^2d_{d2} \) in equation (6a)). In figure 3(d1), the splitting energy levels \([\pm] \) each include three sublevels and are from three hyperfine energy levels of \( P_1 \). The three sublevels can be viewed as one fine level, inasmuch as the distance between \([+\) and \([−) \) is much larger than that among the sublevels. Similarly, as \([1] \) is split into \([G_1 \pm] \) (figure 3(d2)) by \( E_3' \) (\( |G_3\rangle^2d_{d2} \) and \( |G_2\rangle^2d_{d2} \) in equation (6c)), a suppression dip in the fourth-order FL signal (figure 3(a3)) appears by opening \( E_1 \) and \( E_3' \). When another strong beam \( E_2 (E_3) \) is open, the fourth-order FL signal undergoes stronger suppression and a narrow line-width SP-FWM is generated, as shown in figures 3(a4) and (a5). The strongest SP-FWM signal (including the generation signals \( E_{S2} \) and \( E_{S3} \)) in figure 3(a6) will appear in the composite channel, as the fourth-order FL signals in the V- and A-type level systems are suppressed more strongly if all beams are on. The simulations in figure 3(b) make the process of the competition between FL and SP-FWM reappear.

Figure 3(c) shows the variation of composite signal intensity at \( \Delta t = 0 \) in the time domain by opening (c1) \( E_1 \), (c2) \( E_1 \) and \( E_2' \), and (c3) all beams. Figure 3(c4) is the gate of the boxcar integrator and width \( \Delta t = 10 \mu s \) (inset in figure 3(c)). Compared with figures 3(c2) and (c3), the signal intensity in figure 3(c1) is the strongest at the location indicated by the gate, which is in accordance with the intensity at the resonant point in figures 3(a1), (a3) and (a6). Here, the intensity of FL and \( E_5 \) can be expressed by \( I_{E_5}(t) = I_{E_5\text{exp}}[-\Gamma_{E_5}(t-t_0)] \) and \( I_S(t) = I_{S\text{exp}}[-\Gamma_S(t-t_0)] \), where \( I_{E_5\text{exp}} \propto \rho_{E_5}^{(1)}(k-2.4) \), \( I_{S\text{exp}} \propto \rho_{S}^{(3)}(k) \). \( \Gamma_{E_5,S} \) and \( t_0 \) are the transverse decay rate and delay time, respectively. The delay time is mainly determined by the non-radiative relaxation time when pulse width (5 ns) is neglected. Compared to only opening one beam in figure 3(c1), the fourth-order FL signals decay after a longer delay time (~540 μs) while the other decay process of a strong SP-FWM signal is observed at zero delay when all beams open in figure 3(c3). The reason is that particles are excited to \([1] \) which is split into \([\pm] \) caused by both \( E_3 \) and \( E_1 \), the contribution of the radiative process from \([\pm] \) particles to FL which is suppressed strongly at zero delay, and the SP-FWM with insensitive dressing effect can be observed (left peak in figure 3(c3)) simultaneously. The obvious delay of the right peak in figure 3(c3) is caused by the residual particles in \([+\) transferring to \([−) \) through phonon-assisted nonradiative transition, which is mainly determined by acoustic phonons at low temperature. The decay process of fourth-order FL in the V-type level system is delayed a short time ~242 μs in figure 3(c2) due to the smaller splitting between \([G_1 \pm] \) and \([G_1 \mp] \) only with a strong dressing field \( E_5' \). The second-order FL is not delayed in figure 3(c1) with the smallest splitting due to a weak dressing effect of \( E_1 \). Therefore, the delay of the FL decay process depends on the splitting gap between \([\pm] \) (including three group sublevels of splitting hyperfine energy levels).

In order to deeply research the competition process of SP-FWM and fourth-order FL, we change the powers of \( E_3 (E_1) \) and \( E_2 (E_1) \) in A- and V-type level systems. The intensity of composite signal in the time domain is similar to that shown in figure 3(c3) with a large splitting gap between \([\pm] \).

Figure 4(a1) shows pure \( E_5 \) intensity dependence by scanning \( E_1 \) and increasing \( P_3 \). When \( P_3 \) increases, the increasing SP-FWM of \( E_5 \) and its reflected beam \( E_{S3} \) contributes to the background (square), which rises gradually. Although the total intensity of \( E_5 \) increases (circle), the peak intensity which is the SP-FWM \( E_{S3} \) reduces (upper triangle) because of the enhanced self-dressing effect of \( E_5 \) with increasing \( P_3 \) (\( |G_3\rangle^2/|\Gamma_1| \) and \( |G_3\rangle^2/|\Gamma_1| \) in equation (3)). Similarly, as \( P_1 \) increases, the background and the total intensity of \( E_5 \) both increase in figure 4(a2) because of the contribution of \( E_{S3} \), but the emission peak keeps invariant because \( E_{S3} \) suffers from the increasing dressing effect of \( E_1 \) (\( |G_3\rangle^2/|\Gamma_1| \) in equation (3)). In the V-type system, the \( E_5 \) signal is obtained as shown in figure 4(c1) with increasing \( P_3 \) and figure 4(c2) with increasing \( P_1 \). The intensity of \( E_{S2} \) (triangle) in figure 2(c1) reduces because the self-dressing effect of \( E_2 \) (determined by the terms \( |G_3\rangle^2/|\Gamma_2| \) and \( |G_3\rangle^2/|\Gamma_1| \) in equation (2)) is enhanced. The background (square) and total (circle) signals in figure 4(c1) increase due to the contribution.
of SP-FWM (generated by $E_2$ and its reflected beam $E_{2r}$) increases gradually. In figure 4(c2), the background (square) increases but the total signal (circle) decreases when the $P_1$ changes from 0.5 to 2.5 mW, and then increases. The reason is that the dressing effect of $E_1$ ($|G_2|^2d_{11}$ and $|G_1|^2d_1$ in equation (2)) is dominant for $E_{S2}$ in the weak-power region and then the generation of $E_{S2}$ balances the dressing effect when $P_1$ is strong. As a result, $E_{S2}$ (triangle) keeps invariant after the decreases in figure 4(c2).

The signal detected from the composite channel (figures 4(b) and (d)) is worth studying. Theoretical simulations of figures 4(b1) and (d1) are shown in figure 4(e). By changing $P_3$ from weak to strong, the background indicated by the short-dashed curve in figure 4(b1) is the non-resonant FL of $\rho_{11}^{(4)}$ (equation (6c)) and the decreases caused by the enhanced self-dressing terms $|G_2|^2d_{11}$, $|G_3|^2d_1$ and $|G_2|^2d_30$ in equation (6c).

However, the background signal indicated by the short-dashed curve in figure 4(b2) rises (BC section) after falling (AB section). The AB section is the fourth-order FL signal dressed by $E_1$ ($|G_2|^2d_{11}$ and $|G_1|^2d_1$ in equation (6c)). Particularly, the signal of fourth-order FL (equation (6c)) indicated by the fourth curve in figure 4(b2) has been suppressed to the weakest. Then the background (BC section of the dashed curve) increases, which is SP-FWM ($E_{S3}$) generated by $E_1$ and $E_3$. The differences between the background in figure 4(b2) and that in figure 4(b1) demonstrate that the enhanced dressing effect root in larger Rabi frequency, which is related to the dipole moment $\mu$ between $|m\rangle$ and $|n\rangle$. For the gradually stronger suppression of fourth-order FL by increasing $P_1$, the signal in figure 4(b2) alters from suppression dip to emission peak with narrow line-width, which is SP-FWM ($E_{S3}$) generated by $E_1$ and $E_3$. The emission peak of the SP-FWM signals in the composite channel are similar in figures 4(b1) and (b2), both of which become stronger due to the increasing $E_{S3}$ when $P_3$ and $P_1$ increase.

In the V-type level system, the variations of the signal in the composite channel in figure 4(d) are similar to those in figure 4(b2). When we scan $E_1$ and change the power of $E_2$ in the V-type level system, the experimental result in figure 4(d1) is different from that in the A-type level system (figure 4(b1)) although $P_3$ and $P_2$ are equal. The SP-FWM in figure 4(b1) is stronger than that in figure 4(d1) due to the coherence of an individual ion being better than that of a heteronuclear-like molecule. Despite these differences, a SP-FWM process appears in the suppression dip of the fourth-order FL signal both in A-type and V-type level systems with the enhancing dressing effect when the power of the dressing field increases.

### 4. Conclusion

In summary, we have reported the competition between SP-FWM and second (fourth)-order FL in different level systems both theoretically and experimentally. Changing the power of different dressing fields and opening different fields, the SP-FWM will appear in the FL channel with strong dressing effects. Comparing the phenomena between A- and V-type level systems, we find that the competition process between SP-FWM and fourth-order FL is realized more easily in an individual ion system than that in a heteronuclear-like molecule system. Therefore, we can utilize dressing effects to control this competition process in different level systems. In addition, because dressing effect leads to energy level splitting, the FL decay process 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 a photonic chip in the future.

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### References