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Seeded spontaneous parametric four-wave mixing and fluorescence of Pr\(^{3+}\):YSO

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Abstract
We demonstrate the seeded spontaneous parametric four-wave mixing (SP-FWM) process associated with the fluorescence signals of Pr\(^{3+}\):YSO both theoretically and experimentally. For the first time, the dressed SP-FWM processes are observed by self-dressing and/or external-dressing in this crystal. Various possible amplification processes are discussed by selectively seeding various multi-wave mixing (MWM) signals. Two contributions of the dressing effect and the dipole–dipole interaction to the linewidths of the SP-FWM signal and the conjugate signal of the seeding MWM signal associated with the fluorescence signal have been distinguished. In addition, the Autler–Townes-like (or Autler–Townes) splitting effect of the fluorescence spectrum induced by itself (or external) field and the mutual interaction between two fluorescence signals are investigated which are well explained using the presented theoretical model. Such results can find potential applications in all-optical communication and optical information processing on a photonic chip.

Keywords: lifetime, dipole–dipole interaction, multi-wave mixing, dressing effect, Pr\(^{3+}\):YSO

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

1. Introduction

Atomic coherence is the result of interaction between light and materials, which leads to many important physical phenomena. Currently, most experimental researches on atomic coherence are reported in hot or cold atomic gas systems. Compared with atomic gases, atomic coherence-induced effects in solid materials have more practical applications. The recent research progresses related to atomic coherence in solid-state materials, including electromagnetically induced transparency (EIT) in solid materials [1–4], light velocity reduction and coherent storage [5–8], controllable erasing of optically stored information [9], all-optical routing based on optical storage [10], optical velocity reduction and reversible storage of double light pulses [11] and enhanced four-wave mixing (FWM) based on atomic coherence [12], 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 letter, we show the seeded spontaneous parametric FWM (SP-FWM) processes associated with the fluorescence signals of solid-state Pr\(^{3+}\):YSO both theoretically and experimentally. In such a system, for the first time, the dressed SP-FWM processes are observed by self-dressing and/or external-dressing in this crystal. Various possible amplification processes are discussed by selectively seeding various multi-wave mixing (MWM) signals. Two contributions of the dressing effect and the dipole–dipole interaction to the linewidths of the SP-FWM signal and the conjugate signal of the seeding MWM signal associated with the fluorescence signal have been distinguished. In addition, the Autler–Townes-like (or Autler–Townes) splitting effect of the fluorescence spectrum induced by itself (or external) field and the mutual interaction between two fluorescence signals are investigated which are well explained using the presented theoretical model. Such results can find potential applications in all-optical communication and optical information processing on a photonic chip.
corresponding orbit states (extrinsic property). In addition, the Autler–Townes-like (or Autler–Townes) splitting effect of the fluorescence spectrum induced by itself (or external) field and the mutual interaction between two fluorescence signals are investigated which are well explained using the presented theoretical model. Such results can find potential applications in all-optical communication and optical information processing on a photonic chip.

2. Basic theory

Figure 1(a) shows the relevant energy-level diagram of 0.05 at % rare-earth Pr3+ doped Y2SiO5 (Pr: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 Pr3+ ions. The unperturbed lifetime [13] of the excited state $^1D_2$ is $T_2 = 164 \mu s$. The degeneracy of the energy levels of the Pr3+ ion is removed completely by the crystal field of YSO since the point symmetry groups of both cation sites are $C_1$. Under the action of the YSO crystal field, the terms in $^3H_4$ and $^1D_2$ states are split into nine and five Stark components, respectively. The Pr3+ impurity ions occupy two nonequivalent cation sites (sites I and II, respectively) in the YSO crystal lattice. Considering the two nonequivalent cation sites, we can construct a simplified energy-level diagram for the Pr3+ ion associated with the energy bands for the YSO crystal (see figure 1(a)), where the energy level of site I is labeled by a Greek letter without an asterisk and the one for site II is labeled by the one with an asterisk. Here, the ground states of Pr3+ in both sites can be considered to be degenerate. In this work, the pump fields for the excitation processes of fluorescence signals are coupled into the transition between two degenerated states $\delta_0(\Omega) \leftrightarrow \delta_0(\Omega')$. Meanwhile, for MWM processes, two states [2] and [3] both with frequency detunings $\Delta_0$ with respect to the center energy level $\delta_0(\Omega)$ are involved. In addition, at $T = 77 \text{K}$, the phonons in the crystal matrix participate in the nonradiative energy transport between Pr3+ ions localized at different cation vacancies in the YSO crystal. Such a mechanism is known to exist [13, 14] and the transport probability is $\sim T^4$.

The sample (a 3 mm Pr:YSO crystal) is held at 77 K in a cryostat (CFM-102). Three tunable dye lasers (narrow scan with a 0.04 cm$^{-1}$ linewidth) pumped by an injection locked single-mode Nd:YAG laser (Continuum Powerlite DLS 9010, 100 Hz repetition rate, 5 ns pulse width) are used to generate the pumping fields $E_1(\omega_1, \Delta_1), E_2(\omega_2, \Delta_2)$, and $E_3(\omega_3, \Delta_3)$ with the frequency detuning of $\Delta_i = \omega_0 - \omega_i(i = 1, 2, 3)$, respectively, where $\omega_0$ denotes the corresponding atomic transition frequency. To make a description conveniently, we define several abbreviated symbols as follow: FL1 and FL2 mean the fluorescence signals radiated from the same level [1], but pumped by different fields $E_1$ and $E_2$, respectively.

2.1. Experiment setup

$E_1$ pumps the sample and is reflected by the back surface of YSO crystal in its original path, which is named as $E_1'$. $E_2$ and $E_2'$ derived from the same dye laser with a small angle between them, counterpropagate with $E_1$ through the sample (see figure 1(b)), and are coupled to the corresponding transition by tuning the grating of dye laser, as shown in figure 1(a). $E_3$ counterpropagates with $E_2'$, which has a small angle $\theta$ with $E_1$. The diameters of all beams are 1 mm. The properties in the excitation spectrum of the Pr:YSO crystal are investigated by detecting the fluorescence signal with a photomultiplier tube (PMT) and a fast gated integrator. The insets of figure 1(a) are the measured FL signals in the $(a1)$ time domain and $(a2)$ frequency domain, respectively. The measured fluorescence lifetime is about 500 $\mu$s, which is larger than the one shown in [13]. Meanwhile, two fluorescence peaks are observed by scanning the pump filed $E_1$ in a wide frequency range, where the left peak ($\gamma_1$) belongs to site I at a wavelength of 605.97 nm and the right one ($\gamma_2$) is from site II at 607.96 nm. Both linewidths are about 245 GHz, which are broadened both by the homogeneous broadening caused by phonons (~150 GHz) introduced by the lattice heat vibration and the power broadening (~94 GHz) of the pumping field.

2.2. Theoretical model

The fluorescence signal can be used to monitor the interaction between the incident fields, and the intensity of this signal can be expressed by the diagonal elements of the density matrix. In the current system, there are two probable pathways [15, 16] $\rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{11}^{(2)}$ and $\rho_{00}^{(0)} \rightarrow \rho_{01}^{(0)} \rightarrow \rho_{11}^{(2)}$ (i.e. FL1 signal radiated from level $|1\rangle$). To highlight the mechanism of
the dressing effect and allow the reader to easily understand, we specially choose one transition channel of all perturbation chains to analyze the output signal. Considering the power dependence on $E_1$, the density-matrix element $\rho_{11}(t)$ is given by

$$
\rho_{11}(t) = \frac{|G_1|^2}{(d_1 + |G_1|^2 / \Gamma_0)} (T_1 + |G_1|^2 / d_1),
$$

where $d_1 = \Gamma_0 + i\Delta_1$, $G_1 = -\mu_E / h$ is the Rabi frequency, and $\Gamma_0$ is the dephasing rate. Note that, $\Gamma_0$ is the transverse dephasing rate of the ground state $|0\rangle$ broadened by the dipole–dipole interaction. By applying field $E_2$ to modify such a process, the output signal can be given as

$$
\rho_{11}(t) = \left[ |G_1|^2 \right. \\
\left. + \frac{(d_2 + |G_2|^2 / \Gamma_0) (T_2 + |G_2|^2 / d_2)}{(d_2 + |G_2|^2 / \Gamma_0) + (T_2 + |G_2|^2 / d_2)} \right],
$$

where $d_2 = \Gamma_0 + i\Delta_2$. From equation (2), one can see that the output signal is a combined signal of FL1 and FL2 excited by $E_2$ with mutual interaction between them.

In addition to the fluorescence spectrum, a spontaneous parametric (SP)-FWM spectrum will occur in the paraxial direction due to the so-called phase-matching FWM (PC-FWM) process (see figure 4(c)) occurring in a ‘double-Λ-type’ system, in which the strong pumping fields $E_1$ and $E_1'$ are mixed with two weak generated fields $E_2$ and $E_2'$ satisfying the phase-matching conditions (PMCs) $k_{22} = k_1 + k_1 - k_5$ and $k_5 = k_1 + k_1' - k_5$, respectively. For the PC-FWM process, two states [2] and [3] both with frequency detunings $\Delta_1 \delta_0$ with respect to the center energy level $\delta_0(|0\rangle)$ are involved. Both signals are detected by a pair of point symmetrical PMTs as shown in figure 1(b). Quantum mechanically, one can express such a coupling with the Hamiltonian

$$
H = \frac{\hbar}{\nu} \hat{a}^\dagger \hat{b}^\dagger + \hat{a} \hat{b},
$$

where $\hat{a}^\dagger (\hat{a})$ is the boson-creation (annihilation) operator acting on the electromagnetic excitation of the Stokes channel, whereas $\hat{b}^\dagger (\hat{b})$ acts on the anti-Stokes channel, $\nu$ is the group velocity of light in the nonlinear medium, and $g = \chi^{(3)} E_1 E_2 S$ is the pumping parameter of the amplifier, which depends on the nonlinear susceptibility $\chi^{(3)}$ and the pump-field amplitudes. Different from the case occurring in the nonlinear crystal, $\chi^{(3)}$ is a function of higher-order density-matrix elements which can be described by the perturbation chains [17].

$$
\rho^{(0)}_{00} \rightarrow \rho^{(1)}_{11} \rightarrow \rho^{(2)}_{22} \rightarrow \rho^{(3)}_{33} \rightarrow \rho^{(3)}_{33(\text{Stokes})}, \quad \rho^{(0)}_{00} \rightarrow \rho^{(1)}_{11} \rightarrow \rho^{(2)}_{22} \rightarrow \rho^{(3)}_{33(A)} \rightarrow \rho^{(3)}_{33(\text{anti-Stokes})}.
$$

Although there have been several perturbation chains for the generated nonlinear signal, it is reasonable to choose one of them to analyze the physical mechanism and fit the change rule by the modulus square of the corresponding component of such a perturbation chain. Taking the dressing effect of $E_1$ into account, we obtain

$$
\rho^{(3)}_{12(\text{Stokes})} = -i G_{\text{abs}}^* G_{12} |d_1 (d_2 + |G_2|^2 / d_2)|, \quad \rho^{(3)}_{12(\text{Stokes})} = -i G_{\text{abs}}^* G_{12} |d_1 (d_2 + |G_2|^2 / d_2)|,
$$

where $d_1 = \Gamma_1 + i(\Delta_1 - \Delta_0)$, $d_2 = \Gamma_2 + i(\Delta_2 - \Delta_0)$, and $d_3 = \Gamma_3 + i(\Delta_3 + \Delta_0)$; $\delta$ is the linewidth of Stokes and anti-Stokes signals. Thus, the intensities of output signals (without any seeding light) at these two channels are [18]

$$
\langle \hat{a}_{\text{Stokes}} \hat{a}_{\text{Stokes}} \rangle = 1/2 \left[ \cos \frac{2\pi A \sin \phi_1 + \phi_2}{2} \right]^2 \right] A, \quad \langle \hat{a}_{\text{anti-Stokes}} \hat{a}_{\text{anti-Stokes}} \rangle = 1/2 \left[ \cos \frac{2\pi B \sin \phi_1 + \phi_2}{2} \right]^2 \right] B, \quad \langle \hat{a}_{\text{Stokes}} \hat{a}_{\text{anti-Stokes}} \rangle = 1/2 \left[ \cos \frac{2\pi A \sin \phi_1 + \phi_2}{2} \right]^2 \right] A B, \quad \langle \hat{a}_{\text{Stokes}} \hat{a}_{\text{Stokes}} \rangle = 1/2 \left[ \cos \frac{2\pi B \sin \phi_1 + \phi_2}{2} \right]^2 \right] B A.
$$

where, for convenience, we set $\rho^{(3)}_{12(\text{Stokes})} = A e^{i\phi_1}$ and $\rho^{(3)}_{12(\text{anti-Stokes})} = B e^{i\phi_2}$ with $A$, $B$, $\phi_1$, and $\phi_2$ being the modulus and phase angles of $\rho^{(3)}_{12(\text{Stokes})}$ and $\rho^{(3)}_{12(\text{anti-Stokes})}$, respectively. Now, we focus on the signal’s lifetimes in the time domain. Generally, the dephasing rate ($\Gamma_1$) of the measured fluorescence signal radiated from level $|1\rangle$ is determined by the transverse dephasing time, ($T_2$), including the longitudinal dephasing time (spontaneous emission lifetime ($T_1$)) and the irreversible transverse dephasing time, ($T_2$), i.e. $I_{12}(2\pi T_1)^{-1} + (2\pi T_2)^{-1}$. Note that, $(2\pi T_1)^{-1}$ is the longitudinal dephasing rate (determined by the lifetime) belonging to the intrinsic property and $(2\pi T_2)^{-1}$ is the irreversible transverse dephasing rate which can be controlled by input (external) parameters. Considering the sample used in the current experiment associated with the interaction of coupling fields, the broadened linewidth [13] of the measured fluorescence signal can be described as $I_{12} = I_{\text{pop}} + I_{\text{ion-spin}} + I_{\text{ion-ion}} + I_{\text{phonon}}$, where $I_{\text{pop}} = (2\pi T_1)^{-1}$ depends on the location of the energy-level in phase-space, $I_{\text{ion-spin}}$ relates to the ion–spin coupling effect of the individual ion, $I_{\text{ion-ion}}$ is determined by the interaction among the rare earth ions which can be controlled by the power of the external field and the impurity concentration, and $I_{\text{phonon}}$ is related to the temperature of the sample. The last three terms are components of $(2\pi T_2)^{-1}$. However, in our case, because the starting points of the pulse laser and measure process are triggered simultaneously, the factor affecting the measured linewidth should include a coherence process between two levels $|0\rangle$ and $|1\rangle$, which can be described as a decoherence rate $\Gamma_{01}$ where $\Gamma_{01} = (\Gamma_{01} + \Gamma_{10})/2$ with $i \neq j (i, j = 0, 1, \ldots)$. Therefore, the intensity of the measured fluorescence signal can be described as

$$
I(t) = I_0 \exp \left( -\Gamma_{\text{FL}} t \right),
$$

where $I_{\text{FL}} = (\Gamma_{12} + \Gamma_{11})$, in which $\Gamma_{10} = (2\pi T_1)^{-1} + (2\pi T_2)^{-1}$ is the transverse dephasing rate of the ground state $|0\rangle$. In detail, by taking the controlling terms into account, one can get
Thus, the lifetime is modified since the perturbed state leads to a lower power (average power \( P_t \)) or a higher power of \( E_1 \) at various fixed detuning \( \Delta_1 \).

(c) Power dependence of the decay rate \( \Gamma_{FL} \) at \( \Delta_1 = 0 \) by changing the power of \( E_1 \). Solid curves are the theoretical predictions by equation (20). \( G_1 = 2x \times 590 \text{GHz} \) at 9 mW and \( G_1 = 2x \times 164 \text{GHz} \) at 2.5 mW.

\[
(2\pi T_1)_{\Delta} = 16P(u + \Delta v)^3 \beta^6 \eta^3 / 4\pi c^3, \quad (9)
\]

\[
(2\pi T_0)_{\Delta} = 0, \quad (10)
\]

\[
(2\pi T_2)_{\Delta} = P_t(t) + \gamma_0, \quad (11)
\]

\[
(2\pi T_3)_{\Delta} = P_t(t) + \gamma_0, \quad (12)
\]

where \([18, 19]\)

\[
P_t(t) = \exp \left[ -c_D \sum_{n=5,6,7} \left( A_{nD} / R_{nD} \right) \right], \quad (13)
\]

\[
P_0(t) = \exp \left[ -c_H \sum_{n=6,8,10,12,13,14} \left( A_{nH} / R_{nH} \right) \right], \quad (14)
\]

and \( \gamma_0 \) is the total effect of \( \Gamma_{\text{phonon}} \) and \( \Gamma_{\text{ion-spin}} \). In equations (9) and (10), the term \( \nu + \Delta v \) represents the location of the energy-level, which can be modulated by the coupling field. Thus, the lifetime is modified since the perturbed state leads to population redistribution. In equations (12) and (13), \( c_H \) and \( c_D \) represent the population densities at the triplet energy-level \( H_1 \) and singlet energy-level \( D_2 \), respectively, controlled by the pump power. \( \sum (A_{nH} / R_{nH}) \) and \( \sum (A_{nD} / R_{nD}) \) represent the dipole–dipole interactions \([18, 19]\) of states \( H-H \) and \( D-D \), respectively.

The same expression forms can also be used to describe the intensities of the PC-FWM signals \( E_1 \) and \( E_{\text{as}} \) as

\[
I_1(t) = I_{01S} \exp \left[ -\Gamma_{1S} t \right], \quad (15)
\]

\[
I_{\text{as}}(t) = I_{0\text{as}} \exp \left[ -\Gamma_{\text{as}S} t \right], \quad (16)
\]

where \( \Gamma_1 = 2\Gamma_{12} + \Gamma_{13}, \quad \Gamma_{\text{as}} = 2\Gamma_{13} + \Gamma_{12}, \quad I_{01S} \propto |\rho_{12S}^{(3)}|^2 \)

and \( I_{0\text{as}} \propto |\rho_{12\text{as}}^{(3)}|^2 \). Differing from the case of fluorescence signals, the PC-FWM signals are from the coherent processes, whose linewidths are determined by the atomic coherence time and, therefore, are much narrower. Note that, the above discussions do not include the broadened mechanism, which must be considered to fit the corresponding linewidth.

3. Experimental results and analysis

3.1. Fluorescence with induced dipole–dipole interaction

First, by scanning the pumping field \( E_1 \) only, one can easily observe the excitation fluorescence spectrum, FL1, by collecting the spontaneously emitted light at various average power levels of pumping field \( E_1 \). The profile (dashed curve) consisting of the baselines of different signals is the intensity of the non-resonant fluorescence signal changed by the power of pumping field \( E_1 \).

(a) Theoretical predictions by equation (13). (b) The measured FL1 lifetimes represented as the decay rate \( \Gamma_{FL}(G_1) \) at \( b1 \) a lower power or \( b2 \) a higher power of \( E_1 \) at various fixed detuning \( \Delta_1 \).

(b) The measured fluorescence signal changed by the power of pumping field \( E_1 \). The dip below the baseline is the dressing effect of \( E_1 \) on the non-resonant fluorescence signal \( (I_{11} + iG_1 I_{11}|I_{11} + \Delta_1|) \), see equation (1) at the resonant region \( (\Delta_1 \sim 0) \). Figure 2(a) presents the theoretical predictions according to equation (1), which agrees well with the experimental results.

Figure 2(b) depicts the deduced decay rate \( \Gamma_{FL}(G_1) \) from the measured lifetime of the fluorescence signal FL1 at \( b1 \) a lower power (average power \( P_1 = 2.5 \text{ mW} \)) or \( b2 \) a higher power (average power \( P_1 = 9.0 \text{ mW} \)) of pump beam \( E_1 \) versus the frequency detuning \( \Delta_1 \). Such a lifetime, represented by the slope value obtained by the logarithmic transformation \( \left( \gamma_{FL}(G_1) = -\ln(n_{FL}^{(1)}(t)) \right) \) of the delay curve (shown in the inset of figure 1), can be controlled by the dipole–dipole interaction \([19] \propto R^{-6} \) among the rare earth ions as expressed in equations (9)–(14). Figure 2(b) shows the evolution of decay rate, \( \Gamma_{FL} \), with respect to the detuning \( \Delta_1 \) at a low power regime, which shows a minimum value for the FL1 lifetime at resonance due to the strong interaction among ions. A different phenomenon is observed at higher excitation power as shown in figure 2(b2), in which the resonant FL1 lifetime is significantly modified by the saturation absorption effect and the dipole–dipole interaction. According to equations (9) and (10) in which the interval \( \Delta v \) \((\Delta v = \Gamma_{FL}) \) is introduced, the cubic wavelength dependence \((\nu + \Delta v)^3 \) is used to fit the decay rate, which shows a good agreement. To seek the underlying physical mechanism, the dependence of the fluorescence lifetime at \( \Delta_1 = 0 \) on the power of pumping field \( E_1 \) is investigated as shown in figure 2(c). Initially (the region labeled by I), the saturation absorption effect is too weak to be observed, and the modulated lifetime with increasing power is mainly due to the dipole–dipole interaction of \( H \rightarrow H \) states, i.e. the term \( P_\text{as} \) shown in equation (14). Then, the Autler–Townes-like
splitting appears (the region labeled by II), revealing that the saturation absorption effect described by equation (9) appears. Therefore, the enhanced lifetime is caused by the combination of the saturation absorption effect and the dipole–dipole interaction (in turn, H–H states and D–D states, respectively).

However, the lifetime becomes larger (the region labeled by III) as the power further increases, in which the saturation absorption effect is dominant. Considering the above analysis, the theoretical predictions agree quite well with the experimental data as shown in the corresponding panels (solid lines).

Next, by applying a dressing field \( E_2 \), such a fluorescence signal FL1 can be controlled by varying the frequency detuning \( \Delta_2 \) and power \( P_2 \). Two sets of FL1 signal are chosen to study the dressed effects. The first one is the FL1 spectrum without splitting (i.e. at a lower \( P_1 \) power as shown in figures 3(a) and (c)) and the second one presents the splitting (i.e. at a higher \( P_1 \) power as shown in figures 3(b) and (d)). We first show the modulated results of the FL1 signal by changing the detuning (\( \Delta_2 \)) of the strong dressing field \( E_2 \). Then, the dependence of the FL1 signal is investigated by changing the \( E_2 \) power.

Figure 3(a1) shows the dressing effect of \( E_2 \) on the fluorescence signal FL1 (without the splitting) at various fixed detuning values \( \Delta_2 \). The power of \( E_2 \) is fixed at a relatively strong level (average power with 9.0 mW corresponding to the Rabi frequency \( G_2 = 2\pi \times 590 \) GHz). The profile (dashed curves in figure 3(a1)), connecting the baselines of the signals with different \( \Delta_2 \) detunings, is the fluorescence signal FL2 excited by the pump beam \( E_2 \), where the curve shows the Autler–Townes-like splitting due to the saturation absorption effect of \( E_2 \). For the case shown in figure 3(a1), when the detuning \( \Delta_2 \) is far away from the resonant region, the fluorescence signal FL1 is not affected by \( E_2 \). As the detuning \( \Delta_2 \) becomes closer to the resonant point, the baseline of the fluorescence signal FL1 raises gradually due to the competing excitations of the particles by \( E_1 \) and \( E_2 \) beams, and the intensity of the fluorescence signal FL1 is reduced. At two peaks of FL2, as shown in figure 3(e), the \( E_2 \) dominates the competition and the dressing effect of \( E_2 \) on the fluorescence signal FL1 (the suppressed dip) begins to appear. At the near-resonant region, the competitiveness of \( E_2 \) on particles decreases gradually until \( \Delta_2 \approx 0 \) due to the saturation absorption effect of the fluorescence signal FL2. However, the dressing effect of \( E_2 \) on the fluorescence signal FL1 increases gradually and reaches the maximum at \( \Delta_2 \approx 0 \). Such a mutual interaction between fluorescence signals FL1 and FL2 can be well interpreted by equation (2). Due to the low \( E_1 \) power, the dressing effect of \( E_1 \) (\( |G_1|^2/|I_{10} + i\Delta_2| \) and \( |G_1|^2/|I_{00} \)) can be neglected in equation (2). Therefore, the output signals are controlled via the competing action of \( E_2 \) on the particles of the ground state (\( |G_2|^2/|I_{00} \)) and the dressing effect on energy-level position (\( |G_2|^2/|I_{10} + i\Delta_2| \)). Different from the case shown in figures 3(a1) and (b1) presents the dressing effect of \( E_2 \) on the fluorescence signal FL1 with the splitting at various fixed detuning values of \( \Delta_2 \). At the region being far away from the resonance (\( |\Delta_2| >> 0 \), one can observe a pure fluorescence signal FL1 with the splitting. In addition, comparing with the case in figure 3(a), the baselines of such signals are raised due to the enhancement of non-resonant fluorescence signal FL1. Tuning \( \Delta_2 \) from off-resonance to resonance gradually, the same behaviors occur as discussed above for figure 3(a), where the external-dressing splitting of the FL1 signal overlaps with the self-dressing splitting (\( \pm \Delta \)) as shown in figure 3(e). Note that the dressing effect of \( E_1 \) (\( |G_1|^2/|I_{10} + i\Delta_1| \) and \( |G_1|^2/|I_{00} \)) cannot be neglected in equation (2).

The power dependences of the fluorescence signal FL1 ((c1) without and (d1) with the splitting, respectively) on various fixed power values \( P_2 \) of \( E_2 \) \( (\Delta_2 = 0) \) are shown in figure 3. We first focus on the case shown in figure 3(c1). Apparently, at the lowest \( E_2 \) intensity, the dressing effect on the fluorescence signal FL1 is too weak to be observed. Then, the enhanced peaks of the fluorescence signal FL1 can be obtained at
intermediate power levels as the competition between $E_1$ and $E_2$ excitations. As a demonstration, one can find that the profile consisting of the baselines (i.e. the intensity of the fluorescence signal FL2) of spectra at various power levels of $E_2$ increases gradually as the power of $E_2$ increases. At a higher power level, the strong coupling between the field and particles leads to the generation of a dark state, and thus the suppressed dip on the fluorescence signal FL1 appears. Note that, in this case, although the enhancement of the fluorescence signal FL2 by $E_2$ still exists, the dressing effect is the dominant one. Figure 3(c) gives the theoretical prediction according to equation (2), which agrees well with the experimental data. Finally, the baseline (the intensity of the nonresonant FL1 signal) of the signal reaches saturation when $E_2$ power further increases, and then declines due to the dressing effect in the case shown in figure 3(d). Meanwhile, the profile of the fluorescence signal FL1 switches from splitting shape to a pure suppressed dip. So, the line shape switch of the fluorescence signal FL1 can be controlled by various parameters used in the current experiment, which can have potential applications in all-optical communication. In addition, the modulated decay rate $\Gamma_{f1}(G_1, G_2)$ corresponding to the case of figure 3(c) is studied as shown in figure 3(f). Similar to the above discussion, the region labeled I of figure 3(f) can be attributed to the dipole–dipole interaction of $H – H$ states, and the region labeled III is dominated by the dressing effect of $E_2$ as the pure dip shown in figure 3(c). Region II is from a combined action.

3.2. Seeded narrow-band spontaneous parametric four-wave mixing process

Nonlinear processes can occur easily in the paraxial direction of the incident laser beam. In the current experiment, the spontaneous parametric (SP)-FWM process occurs due to the reflection of $E_1$ by the back surface of the YSO crystal. Figure 4(a) depicts the power dependences of the measured (a1) Stokes signal and (a2) anti-Stokes signal of the SP-FWM process on various powers of $E_1$ versus $\Delta_1$. The baseline of each signal is the non-resonant fluorescence signal, and the peaks correspond to SP-FWM signals. Therefore, by increasing the $E_1$ power, the intensity of the non-resonant fluorescence signal FL1 increases gradually (see the dashed line of figure 4(a)). The intensity of each peak, corresponding to the measured (a1) Stokes and (a2) anti-Stokes signals of the SP-FWM process, respectively, also increases gradually with the increasing power. To prevent the ‘double-$\Lambda$’ type system shown in figure 4(c) in which two states [2] and [3] both with frequency detunings $\Delta \omega$ with respect to the center energy level $\delta_0(0)$ are involved, such a process can be well interpreted by the phase conjugate (PC)-FWM process at high excitation power. The experimental results agree well with the theoretical predictions, as shown in equations (6) and (7). The self-dressing effect of $E_1$ appears at a high power level as predicted by the theoretical equations. Figure 4(b) gives the dressed signals of the SP-FWM process by varying the power of the applied $E_2$ field. Different from the above case, the profile (dashed line in figure 4(b)) consisting of the baselines of these signal curves represents the SP-FWM signal generated by $E_2$.

![Figure 4](image)

**Figure 4.** (a) Power dependences of the (a1) Stokes and (a2) anti-Stokes signals of the SP-FWM process on the generating field $E_1$. (b) Power dependences of the (b1) Stokes and (b2) anti-Stokes signals of the SP-FWM process on the dressing field $E_2$. Insets are the theoretical predictions. $G_1 = 2 \pi \times 590 \text{GHz}$. (c) The ‘double-$\Lambda$’ configuration with the corresponding phase-matching condition and the spatial alignment for the PC-FWM process. The width of the ground state represents the broadened degenerate state. (d) The dephasing rates of Stokes light corresponding to cases (a) and (b).

The total signal becomes larger gradually when the power of $E_1$ increases. Such a phenomenon can be attributed to the new SP-FWM process introduced by applying the external dressing field $E_2$. Insets are the theoretical predictions with consideration of the above situations, which agree well with the experimental data. The decay rates of the Stokes light corresponding to cases (a) and (b) versus the powers of $E_1$ and $E_2$, respectively, are investigated as shown in figure 4(d), both of which increase with increasing power. The observed features can be explained by the term $\Gamma_{\text{reg}} = 2F_{12} + F_{10}$ in equation (15) (shown in the methods section) relating to the power, so one can deduce that the reduced lifetime of the output signal is caused by the increased power of the pumping beam.

4. Discussion

So far, we have presented the SP-FWM process dressed by self- or external-dressing field. As an extension to the above discussions, such a process can amplify the seeded signals propagating along with the corresponding output channels [17]. We make the generated fields obtained by MWM processes as the seeded signals, which can occur by turning on/off the incident beams selectively in the current experiment. First, by opening $E_1, E_2$ and $E_3$ (a) a FWM1 (FWM2) signal with the phase-matching condition $k_0 = k_1 + k_2 - k_3$ ($k_{03} = k_2 - k_1$) will be generated, which propagates along the hypotenuse of the emission cone of the Stokes (anti-Stokes) beam as shown in figure 4(c). By $\rho^{(0)}_{22} \xrightarrow{E_1} \rho^{(1)}_{21} \xrightarrow{(E_3)} \rho^{(2)}_{22} \xrightarrow{E_2} \rho^{(3)}_{21}$ and taking the dressing effects of $E_1$ and $E_2$ (or $E_1, E_2$ and $E_3$) into consideration [15, 16], the corresponding density-matrix element $\rho^{(3)}_{11} (\rho^{(3)}_{22})$ and the FWM1’s (FWM2’s) intensity are given by
\( \rho_{F1}^{(3)} = \rho_{12}^{(3)} = \frac{-iG_1|G_2|^2}{(d_{12} + |G_1|^2 / \Gamma_{22} + |G_2|^2 / d_3) d_3}, \) \hspace{1cm} (17)

\( \rho_{F2}^{(3)} = \rho_{13}^{(3)} = \frac{-iG_1G_2G_3}{(d_{13} + |G_1|^2 / \Gamma_{33} + |G_2|^2 / d_4 + |G_3|^2 / d_5) d_5} \)

\[ \times [d_7 + |G_4|^2 / d_6 + |G_5|^2 / d_3 + |G_6|^2 / d_4], \] \hspace{1cm} (18)

\[ I_{F1}(t) = I_{0,F1}(t) \exp \left[-\Gamma_{F1} t \right], \] \hspace{1cm} (19)

\[ I_{F2}(t) = I_{0,F2}(t) \exp \left[-\Gamma_{F2} t \right], \] \hspace{1cm} (20)

where \( d_1 = \Gamma_{22} + i(\Delta_1 - \Delta_2), d_2 = \Gamma_{33} + i(\Delta_1 + \Delta_2), d_3 = \Gamma_{33} + i(\Delta_1 - \Delta_3), d_4 = \Gamma_{13} + i(\Delta_1 + \Delta_2 - \Delta_3 - \Delta_4), \)

\[ \Gamma_{F1} = 3\Gamma_{12}, \] \( I_{0,F1} \propto |\rho_{F1}^{(3)}|^2, \quad \Gamma_{F2} = \Gamma_{13}, \quad \text{and} \quad I_{0,F2} \propto |\rho_{F2}^{(3)}|^2. \]

\( \Gamma_{12}(\Gamma_{13}) \) corresponds to the term \( \Gamma_{12}(\Gamma_{13}) \) in equation (17) (equation (18)) modulated by the dipole–dipole interaction and dressing effect. Therefore, the dephasing rate \( \Gamma_{F1}(\Gamma_{F2}) \) of the FWM1 (FWM2) signal is controlled by the \( E_1 \) power. Seeded by the FWM1 or FWM2 signal, the output intensities at the two SP-FWM channels become

\[ \hat{\alpha}_{out}^{+}\hat{\alpha}_{out} = \frac{1}{2} \left[ \cos \left( 2\sqrt{\lambda} \sin \frac{\varphi_1 + \varphi_2}{2} \right) + \cos \left( 2\sqrt{\lambda} \cos \frac{\varphi_1 + \varphi_2}{2} \right) \right] \left[ |\alpha|^2 + \frac{A}{B} \right], \] \hspace{1cm} (21)

\[ \hat{\beta}_{out}^{+}\hat{\beta}_{out} = \frac{1}{2} \left[ \cos \left( 2\sqrt{\lambda} \sin \frac{\varphi_1 + \varphi_2}{2} \right) + \cos \left( 2\sqrt{\lambda} \cos \frac{\varphi_1 + \varphi_2}{2} \right) \right] \left[ |\beta|^2 + \frac{B}{A} \right], \] \hspace{1cm} (22)

\[ \hat{\beta}_{out}^{+}\hat{\beta}_{out} = \frac{1}{2} \left[ \cos \left( 2\sqrt{\lambda} \sin \frac{\varphi_1 + \varphi_2}{2} \right) + \cos \left( 2\sqrt{\lambda} \cos \frac{\varphi_1 + \varphi_2}{2} \right) \right] \left[ \frac{B}{A} |\beta|^2 + \frac{B}{A} \right], \] \hspace{1cm} (23)

where \( G_{F1} \propto \sqrt{2E_{0}\greek{h} N_\mu^2 \rho_{F1}^{(3)}} \) and \( G_{F2} \propto \sqrt{2F_{0}\greek{h} N_\mu^2 \rho_{F2}^{(3)}} \), \( \lambda \alpha = (\pm E_{0}\greek{h}\zeta_{2}\alpha)(r_1 G_{F1}/(\mu t_{12})^2) \) and \( |\beta|^2 = (\pm E_{0}\greek{h}\zeta_{2}\alpha)(r_2 G_{F2}/(\mu t_{12})^2) \) denote the intensities of the seeded fields \( E_{01} \) and \( E_{02}; r_1 \) and \( r_2 \) are radii of \( E_{01} \) and \( E_{02} \) beams. In addition, the corresponding dephasing rate of the anti-Stokes (Stokes) signal is given by \( I_{\Delta5} = \Gamma_{F1} + \Gamma_{F2} + \Gamma_{12}(\Gamma_{F2} + \Gamma_{12} + \Gamma_{13})). \)

Similarly, one can use the generated six-wave mixing (SWM) signal as seeding light by opening all \( E_1, E_2, E_2' \) and \( E_3 \) beams. By using the perturbation chain

\[ \rho_{13}^{(3)} \rightarrow \rho_{13}^{(3)} \rightarrow \rho_{13}^{(5)} \rightarrow \rho_{13}^{(5)} \rightarrow \rho_{13}^{(5)} \rightarrow \rho_{13}^{(5)}, \]

one gets the density-matrix element corresponding to the SWM signal and its intensity as

\[ \rho_{SWM}^{(5)} = \rho_{13}^{(5)} = \frac{-iG_1|G_2|^2}{(d_{13} + |G_1|^2 / \Gamma_{33} + |G_2|^2 / d_4 + |G_3|^2 / d_5) d_5} \]

\[ \times [d_7 + |G_4|^2 / d_6 + |G_5|^2 / d_3 + |G_6|^2 / d_4], \] \hspace{1cm} (24)

where \( G_{SWM} \propto \sqrt{2E_{0}\greek{h} N_\mu^2 \rho_{SWM}^{(5)}} \) and \( |\beta|^2 = (\pm E_{0}\greek{h}\zeta_{2}\alpha)(r_2 G_{SWM}/(\mu t_{12})^2) \) denote the intensity of the seeding field and \( r_2 \) is the radius of laser beam \( E_{SWM} \). The dephasing rate of the corresponding anti-Stokes signal is \( I_{\Delta5} = \Gamma_{13} + I_{SWM} + \Gamma_{12}. \)

Figures 5(a1) and (a2) present the measured output signals (Stokes and anti-Stokes, respectively) with no seeding light.
and seeded with FWM1 signal in Stokes channel, FWM2 signal in anti-Stokes channel and multi-MWM signals from left to right, respectively. Insets are the theoretical predictions. Once the Stokes (anti-Stokes) channel is seeded by the generated FWM1 (FWM2) beam, the output signal of this channel and the corresponding conjugate signal are both amplified. The efficiency of the non-degenerate FWM2 process is higher than the generated FWM1 process. By opening all laser beams, the SWM signal generated in the ‘N-type’ configuration provides seeding for the Stokes channel. In this case, the FWM1 and FWM2 signals coexist with the SWM signal simultaneously due to the wide linewidths of incident lasers [17]. Next, we consider lifetimes of the conjugate signals at various seeding cases as shown in figure 5(a3). The decay rate ($\Gamma_j$) versus the laser power is used to represent the lifetime. For three seeding cases (FWM1, FWM2 and SWM, respectively), the trends of change versus increasing laser power are increase, invariant, and decrease, respectively, which can be attributed to different dressing results caused by different configurations as shown in figures 5(b)–(d), which are well explained by the theoretical models given in equations (17), (18) and (25). Such results reveal that the linewidths (i.e. $\Gamma_{FS} = \Gamma_{F1} + \Gamma_{F2} + \Gamma_{F3}$ and $\Gamma_{AS} = \Gamma_{F1} + \Gamma_{SWM} + \Gamma_{F2}$) can be modulated by the dipole–dipole interaction and the dark state included in those equations.

5. Conclusion

In summary, we have presented the SP-FWM process with the fluorescence signals of Pr:YSO both theoretically and experimentally. The linewidths of SP-FWM signals and the conjugate signals of the seeding MWM beams associated with the fluorescence signals are measured, which have been shown to be controllable by the dressing effects on energy levels (intrinsic property) as well as the dipole–dipole interaction of the corresponding orbital states (extrinsic property). In such a system, the dressed SP-FWM process is observed by self-dressing and/or external-dressing configurations. Various amplification processes are discussed by selectively seeding with various generated MWM signals. In addition, the splitting of the fluorescence spectrum induced by itself and/or external field, as well as the mutual interaction between two fluorescence signals, are investigated, which are well explained with the presented theoretical models. Such controlled fluorescence and SP-FWM signals, and seeded amplification processes with generated MWM signal beams, in such a solid-state system can find potential applications in all-optical communication and optical information processing on the photonic chip.

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References