COMPETITION BETWEEN POLARIZATION-DRESSING AND PHONON EFFECTS IN FOURTH-ORDER FLUORESCENCE OF Pr3+:Y2SiO5

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ABSTRACT: We report modulating spontaneous four-wave mixing, fourth-order fluorescence, and intensity noise correlation by polarization-dressing effect at different temperatures in a Pr3+:Y2SiO5 crystal. The delay time, Autler–Townes splitting, and shape of correlation curve are changed observably with temperature and polarization state of dressing field. These results demonstrate competition between polarization-dressing and phonon effects. The impact of temperature on nonlinear response is very important for the stability of quantum optical devices.

I. INTRODUCTION

Rare-earth ion-doped crystal has been widely studied because of its advantages for coherent excitation and because it is more appropriate for practical applications. Recent researches on quantum coherence effect in rare-earth ion have been reported, including electromagnetically induced transparency (EIT),1,2 stimulated Raman adiabatic passage,3 light velocity reduction, coherent storage,4,5 and all-optical routing.6 A Pr3+-doped Y2SiO5 crystal has long coherence time and narrow spectral width as compared to traditional nonlinear crystals. So, the atomic coherence can be induced easily in Pr3+:Y2SiO5. Rare-earth ion-doped crystal can find potential applications in all-optical communication and optical information processing on the photonic chip.7

In practical applications, coherent excitation processes are required to be controlled reliably. Polarization-dressing effect offers a method to modulate the coherent transition. Because different polarization states of pumping field correspond to different transition ways in Zeeman levels, the EIT processes can be controlled via polarization-dressing effect.7,8 We investigated polarization-dressed multi-order fluorescence (FL) and spontaneous four-wave mixing (SFWM) in the Pr3+:Y2SiO5 crystal at low temperature.9,10 We can control dressing signals by changing power, detuning, and polarization field. The correlation and anticorrelation have been observed at different temperatures.11 Such polarization-dressed signals have potential applications such as all-optical switch and route devices.12 Recently, hybrid correlation between different types of states was identified as a useful resource for quantum optical information processing and quantum computing.13,14 Hybrid correlation between SFWM and multi-order fluorescence has also been obtained in the Pr3+:Y2SiO5 crystal.15

In our previous works, all experiments were carried out at low temperature, so the phonon effect has never been considered. In practical applications, temperature plays an important role in device stability. Phonon effect can significantly affect the nonlinear response of quantum optical devices with increasing temperature. In this paper, we investigate the polarization-dressing effect of fourth-order fluorescence at different temperatures. In this case, phonon effect must be considered. Our results indicate that the polarization-dressing effect competed with phonon effect. We also modulate the hybrid correlation shape by cross-phase modulation. Research about competition between polarization-dressing and phonon effects is very important for obtaining a hybrid signal resource, controlling quantum optical devices, and improving the stability of all-optical devices.

II. THEORETICAL MODEL

In a V-type three-level system with two pumping fields E1 and E3 opening, the four-wave mixing and fluorescence signals are generated simultaneously. The intensity of FWM is proportional to the third-order nonlinear density matrix elements. The third-order nonlinear density matrix elements for Stokes E5 and anti-Stokes E5 signals can be written as

\[ \rho_{S1}^{(3)} = \frac{-iG_5 G_{S1} G_{G1}}{\Gamma_{00} + \frac{(|G_5|^2)}{|d_1|^2} + \frac{(|G_{S1}|^2)}{|d_i|^2} + \frac{(|G_{G1}|^2)}{|d_i|^2}} \left( d_2 + \frac{(|G_5|^2)}{|d_1|^2} d_i + \frac{(|G_{S1}|^2)}{|d_i|^2} d_i + \frac{(|G_{G1}|^2)}{|d_i|^2} d_i \right) \]  

\[ \rho_{S2}^{(3)} = \frac{-iG_5 G_{S2} G_{G1}}{\Gamma_{00} + \frac{(|G_5|^2)}{|d_1|^2} + \frac{(|G_{S1}|^2)}{|d_i|^2} + \frac{(|G_{G1}|^2)}{|d_i|^2}} \left( d_2 + \frac{(|G_5|^2)}{|d_1|^2} d_i + \frac{(|G_{S1}|^2)}{|d_i|^2} d_i + \frac{(|G_{G1}|^2)}{|d_i|^2} d_i \right) \]  

where \( d_i = \Gamma_{10} + i\Delta_1 \) and \( d_3 = \Gamma_{20} + i\Delta_2 \). \( \Gamma_{ij} \) is the transverse decay rate. \( G_i = \mu E_i / h \) is the Rabi frequency of the incident
field \( \mathbf{E}_1 \). In eqs 1 and 2, we consider the self-dressing effect of \( \mathbf{E}_1 \) and the external-dressing effect of \( \mathbf{E}_2 \) simultaneously. In this system, the detected signal intensity is the sum of two fluorescence signals, FL1 and FL2. The density matrix elements of fourth-order fluorescence are

\[
\rho_{\text{FL1}}^{(4)} = \frac{|G_1|^2}{d_2 + \frac{|G_1|^2}{\Gamma_0} + \frac{|G_1|^2}{d_1}} \left( \Gamma_0 + \frac{\Gamma_0}{d_1} + \frac{\Gamma_0}{d_2} \right) \times \left( \frac{1}{d_4 + \frac{|G_1|^2}{\Gamma_0} + \frac{|G_1|^2}{d_2}} \right) \left( \Gamma_1 + \frac{\Gamma_1}{d_4} + \frac{\Gamma_1}{d_2} \right)
\]

\[ (3) \]

\[
\rho_{\text{FL2}}^{(4)} = \frac{|G_1|^2}{d_2 + \frac{|G_1|^2}{\Gamma_0} + \frac{|G_1|^2}{d_1}} \left( \Gamma_0 + \frac{\Gamma_0}{d_1} + \frac{\Gamma_0}{d_2} \right) \times \left( \frac{1}{d_4 + \frac{|G_1|^2}{\Gamma_0} + \frac{|G_1|^2}{d_2}} \right) \left( \Gamma_2 + \frac{\Gamma_2}{d_4} + \frac{\Gamma_2}{d_2} \right)
\]

\[ (4) \]

where \( d_{12} = \Gamma_{12} + i(\Delta_1 - \Delta_2) \) and \( d_{13} = \Gamma_{13} + i(\Delta_1 - \Delta_3) \). Similarly, in a \( \Lambda \)-type three-level system, the third-order nonlinear density matrix elements for Stokes and anti-Stokes signals \( E_{\text{S1}} \) and \( E_{\text{ASS}} \) are given by

\[
\rho_{\text{S1}}^{(3)} = \frac{-iG_4 G_{\text{ASS}} G_3}{d_1 + \frac{|G_3|^2}{\Gamma_{13}} + \frac{|G_3|^2}{d_{13}}} \left( d_{13} + \frac{|G_3|^2}{\Gamma_{13}} \right) \times \left( \frac{1}{\Gamma_{03} + \frac{|G_3|^2}{d_{13}} + \frac{|G_3|^2}{\Gamma_{13}}} \right)
\]

\[ (5) \]

\[
\rho_{\text{ASS}}^{(3)} = \frac{-iG_4 G_{\text{ASS}} G_3}{d_1 + \frac{|G_3|^2}{\Gamma_{13}} + \frac{|G_3|^2}{d_{13}}} \left( d_{13} + \frac{|G_3|^2}{\Gamma_{13}} \right) \times \left( \frac{1}{\Gamma_{30} + \frac{|G_3|^2}{d_{13}} + \frac{|G_3|^2}{\Gamma_{13}}} \right)
\]

\[ (6) \]

The density matrix elements of FL1 are

\[
\rho_{11}^{(4)} = \frac{|G_1|^2}{d_1 + \frac{|G_1|^2}{\Gamma_{11}} + \frac{|G_1|^2}{d_{11}}} \left( d_{11} + \frac{|G_1|^2}{\Gamma_{11}} \right) \times \left( \frac{1}{d_4 + \frac{|G_1|^2}{\Gamma_{11}} + \frac{|G_1|^2}{d_{11}}} \right) \left( \Gamma_{11} + \frac{\Gamma_{11}}{d_4} + \frac{\Gamma_{11}}{d_{11}} \right)^2
\]

\[ (7) \]

where \( d_1 = \Gamma_{11} + i\Delta_\nu, d_{10} = \Gamma_{01} + i(\Delta_1 - \Delta_0), d_{13} = \Gamma_{13} + i\Delta_\nu, \) and \( d_{01} = \Gamma_{01} - i\Delta_\nu \).

In the experiment, generated FWM signals are reflected by a polarized beam splitter, and the vertically polarized component of FWM signals is detected. So, the effective density matrix elements can be written as \( \rho_{(1)}^{(3)} = \rho_{(1\nu)}^{(3)} + \rho_{(1\upsilon)}^{(3)} \). We use a half-wave plate (HWP) to modulate the polarization direction of incident field \( \mathbf{E}_0 \) and the dependence of Rabi frequency on polarization angle can be described as \( \zeta_\nu^2|G_1|^2\cos^2 2\theta \) for \( \rho_{(1\nu)}^{(3)} \) and \( \zeta_\upsilon^2|G_1|^2\sin^2 2\theta \) for \( \rho_{(1\upsilon)}^{(3)} \). When a quarter-wave plate (QWP) is used to change the polarization state of incident field, the Rabi frequency is \( \zeta_\nu^2|G_1|^2 \cos^2 \theta + \sin^2 \theta \) for \( \rho_{(1\nu)}^{(3)} \) and \( \zeta_\upsilon^2|G_1|^2 \sin^2 \theta \cos^2 \theta \) for \( \rho_{(1\upsilon)}^{(3)} \), respectively, where \( \theta \) is defined as the rotated angle of the WP’s axis from the \( x \) axis and \( \zeta_{\nu,\upsilon} \) is the anisotropic factor in different directions of crystal. \( \Gamma^p_q(p = -1, 1) \) is the Rabi frequency of a pumping field with left circular, linear, and right circular polarization, respectively. Different laser polarization states have different Clebsch–Gordan (CG) coefficients.

When strong pumping fields couple with energy levels, energy levels will split as induced by the dressing effect. In the V-type three-level system, ground state \( |0 \rangle \) is split into \( |\pm \rangle \) dressing levels by \( \mathbf{E}_2 \). Considering the excitation of pumping field \( \mathbf{E}_1 \) and dressing effect of \( \mathbf{E}_2 \), there are two spontaneous emissions from \( |2 \rangle \) to \( |\pm \rangle \) dressing levels. The wavelength of two spontaneous emissions can be written as \( \lambda = \left| (-1)^{\Delta_1} \pm \sqrt{\Delta_1^2 + 4|G_1|^2} / 2 \right| \). Therefore, the splitting distance between \( |+ \rangle \) and \( |- \rangle \) is \( \Delta_1 = \sqrt{\Delta_1^2 + 4|G_1|^2} \). Moreover, considering the self-dressing effect of \( \mathbf{E}_1 \), the splitting distance can be expressed as \( \Delta_1 = 2\sqrt{|G_1|^2 + |G_1|^2} \) when \( \Delta_1 = \Delta_3 = 0 \). Similarly, in the \( \Lambda \)-type three-level system, excited state \( |1 \rangle \) is split into \( |\pm \rangle \) dressing levels by \( \mathbf{E}_1 \) and \( \mathbf{E}_1 \) with distance \( \Delta_1 = 2\sqrt{|G_1|^2 + |G_1|^2} \).

The linewidth (\( \Gamma \)) of the measured fluorescence signal is determined by the longitudinal relaxation time (\( T_1 \)) and transverse relaxation time (\( T_2 \)), that is, \( \Gamma = (2\pi T_1)^{-1} + (2\pi T_2)^{-1} \), where \( T_1 \) is determined by the width of energy level and \( T_2 \) by the dephasing rate. Considering the dressing level, the broadened linewidth of the fluorescence signal can be further described as \( \Gamma = \Gamma_{\text{pop}} - \Gamma_{\text{ion-egg}} - \Gamma_{\text{ion-egg}} - \Gamma_{\text{phonon-egg}} \), where \( \Gamma_{\text{pop}} = (2\pi T_1)^{-1} \) depends on the location of the energy level in phase space, \( \Gamma_{\text{ion-egg}} \) relates to the ion-egg coupling effect of the individual ion, and \( \Gamma_{\text{ion-egg}} \) is determined by the interaction among the rare-earth ions, which can be controlled by the power of external field and impurity concentration. Phonon relaxation rate is given by \( \Gamma_{\text{phonon}} \propto 2\pi|n(\omega, T) + 1|/h \), where \( n(\omega, T) = 1/\exp(\omega/k_B T) - 1 \) is the thermal occupation of phonon mode. So, \( \Gamma_{\text{phonon}} \) related to the temperature of the sample. \( \Gamma(\Delta_{1}) \) represents the location of the energy level induced by dressing fields.

In our experiment, the FWM and fourth-order FL signals were illuminated by the same pumping fields and were generated simultaneously, so the correlation exists between them. The intensity noise correlation function \( G^{(2)}(\tau) \) can be expressed as

\[
G^{(2)}(\tau) = \frac{\langle \delta\rho_{\text{FL}}^{(4)}(t_\tau)|\langle \delta\rho_{\text{S}}^{(3)}(t_\nu) \rangle \rangle}{\langle \langle |\delta\rho_{\text{FL}}^{(4)}(t_\nu) |^2 \rangle \rangle \langle \langle |\delta\rho_{\text{S}}^{(3)}(t_\nu) |^2 \rangle \rangle} \cos(\Delta \varphi)
\]

\[ (8) \]

where \( \Delta \varphi = \varphi_{\text{S/AS}} - \varphi_{\text{FL}}, \varphi_{\text{S/AS}} \) is the initial phase of FWM, and \( \varphi_{\text{FL}} \) is the dressed phase of fourth-order FL. So, the correlation functions change with the dressing nonlinear phase via cross-Kerr effect.

III. RESULTS AND DISCUSSION

Figure 1 shows FL signals in a time domain detected by photomultiplier tube D3 in \( \Lambda \)-type (Figure 1a–c) and V-type (Figure 1d–f) three-level systems. There are two peaks in these hybrid signals. The right peak is attributed to the adiabatic population transfer between dressed states, whereas the left peak is the contribution of SFWM without adiabatic population transfer. In the \( \Lambda \)-type three-level system, excited
state |G⟩ is split into |G⟩ and |G⟩ caused by E. Dressing level |G⟩ is further split into |G⟩ induced by E. The transfer of population can occur between dressing levels through phonon-assisted nonradioactive transition. We attribute the delay of right peak to the transfer of residual particles between |G⟩ and |G⟩. The delay time of right peak is determined by the distance between |G⟩ and |G⟩. One can see that the delay time decreases with the increase of temperature both in Λ-type and V-type three-level systems. This effect results from the competition between dressing and phonon effects. Phonon relaxation rate $\Gamma_{\text{phonon}} \propto 2\pi[n(n,T) + 1]/h$ includes pure dephasing contributions from temperature-dependent phonon scattering. At high temperature, dressed state levels are broadened by the phonon effect, which causes the splitting distance of dressed state to decrease. Considering the phonon broadening, we can write the expression of the splitting distance as $\Delta = 2\sqrt{|G|^2 + |G|^2} - (\Gamma_{\text{phonon}})^2$. At room temperature, the dressing effect is balanced by the phonon effect, so two peaks combine into one peak. At low temperature, phonon relaxation rate $\Gamma_{\text{phonon}}$ decreases, which causes the transition distance between dressing levels to increase. As a result, the right peak presents obvious delay. In addition, the proportion of FWM and FL in hybrid signals also changes with temperature.

Next, we investigate the polarization-dressing effect on delay time when the dressing field E or E is changed by QWP. When the polarization state of dressing field E or E changes from circular to linear, one can see that the delay time of right peak increases. In this case, the splitting distance is replaced by $\Delta = 2\sqrt{|G|^2 + |G|^2(\cos^4\theta + \sin^4\theta)}$. Because the Rabi frequency of a linearly polarized state ($\Delta_{\text{Lin}}$) is greater than that of a circularly polarized state ($\Delta_{\text{Cir}}$), the delay time of the linearly polarized state is longer than that of the circularly polarized state. With the temperature increasing, polarization dependence of delay time becomes not obvious because of competition between dressing and phonon effects. Such splitting and delay hybrid signals can be exploited for all-optical transistor switching applications.14,19

Figure 2 shows FL signals in a frequency domain detected at different temperatures and polarization states. (a) FL signals in a frequency domain detected at different temperatures and polarization states. (b) FL signals in a frequency domain detected at different temperatures and polarization states. (c) FL signals in a frequency domain detected at different temperatures and polarization states. (d) FL signals in a frequency domain detected at different temperatures and polarization states.

$\Gamma_{\text{phonon}}$ is dominant at room temperature, so the linewidth of the FL curve increases. Moreover, the splitting distance of dressed state level $\Delta = 2\sqrt{|G|^2 + |G|^2} - (\Gamma_{\text{phonon}})^2$ is decreased because of the phonon broadening effect, so AT splitting disappears. When the temperature is below 110K, the phonon effect $\Gamma_{\text{phonon}}$ decreases, and the dressing effect is dominant, which result in obvious AT splitting.

Now, we compare the polarization-dressing effect on AT splitting when the polarization state of dressing field E or E is changed by QWP. With the rotation angle of QWP changing from 0 to 45°, the depth of the suppressed dip reduces. The external-dressing effect of E or E on spectra splitting is reflected by $|G|^2/d_{33}$ (in eqs 3 and 7). Because of different CG coefficients, the dressing effect of the linearly polarized field $|G|^2/d_{33}$ is stronger than that of the circularly polarized field $|G|^2/d_{33}$, which leads to the decrease of AT splitting depth from 0 to 45°.

At room temperature, AT splitting disappears due to phonon effect. The fluorescence emission peaks increase gradually from linear to circular polarization in the Λ-type system. On the contrary, they decrease from linear to circular polarization in the V-type system. In the Λ-type system, there is only one signal FL1 from site I, and E acts as an external-dressing field. The suppression dressing effect of linear polarization is stronger than circular polarization, so fluorescence emission peak increases from linear to circular polarization. In the V-type system, there are two signals, FL1 and FL2, from sites I and II, respectively. E acts as the external-dressing field for FL1 and the generating field for FL2 simultaneously. The generating effect becomes weak from linear to circular polarization, which causes the intensity of fluorescence emission peak to decrease.

Next, we modulate the fourth-order FL signal by changing the frequency detuning $\Delta$, of dressing field (as shown in Figure 3). At 77 K, the FL peak evolves from AT splitting to pure suppression dip, with $\Delta = \Delta$, changing from large detuning to resonance. When $\Delta = \Delta$, the dip becomes close to the resonant point, the dip term $|G|^2/d_{33}$ and $|G|^2/d_{13}$ in eqs 3 and 4 increase. Because of the satisfaction of suppression condition $\Delta = \Delta = 0$ in the experiment, the suppression effect and AT splitting decrease. Because the temperature increased to 110 K, the evolution of FL peak with frequency detuning $\Delta$ was less obvious. This is attributed to the dressing phonon interaction term $|G|^2/\Gamma_{20}$, which increases because of the phonon broadening effect, which counteracts the dressing effect. As a result, the spectrum is less sensitive to the frequency detuning of dressing field at high
temperature. Such controllable splitting and suppression signals have potential applications in all-optical router and switching devices.

Finally, we investigate the intensity noise correlation of hybrid signals. Correlation properties of hybrid signals can be used to reduce the noise level of quantum optical devices. Figure 4a,b shows the intensity noise correlation function for SFWM and FL composite signals is determined by assisted FL process. The shape of the hybrid correlation curve follows the intensity of the composite signal and the competition between SFWM and FL signals. At 77 K, the phonon effect decreases, and the proportion of SFWM in composite signal is large, so the sharp peak of correlation curve is more obvious. With the temperature increase, the phonon-assisted FL process is enhanced, and the proportion of SFWM is reduced. Therefore, the sharp peak of correlation curve is small.

Figure 5 shows the correlation functions of $E_S$-FL and $E_S$-E$_{AS}$ at 110 K when a QWP is used to change the polarization state of $E_S$. In Figure 5a, we find that the maximum correlation between $E_S$ and FL is obtained at the linearly polarized state ($\theta = 0^\circ$). When the polarization state of dressing field $E_d$ changes from linearly polarized to right circularly polarized ($\theta = 45^\circ$), the correlation peaks decrease at first and then switch from correlation to anticorrelation. This result may be explained by the cross-phase modulation (XPM) induced by dressing field $E_d$. In eq 8, the nonlinear phase shift between $E_S$ and FL can be characterized as $\Delta \varphi = 2(k_2n_2^2 - k_{FL}n_{FL}^2)|E_d|^2e^{-2z/\kappa_{11}}$, where $n_2 = Re \chi^{(3)}/\epsilon_0\epsilon_1$ is the nonlinear refractive index of the Kerr medium, $r$ is the beam radius of dressing field, and $z$ is the length of the YSO crystal. When the polarization of the dressing field $E_d$ is changed from linearly polarized to right circularly polarized, the effective Rabi frequency and third-order effective susceptibility can be modulated. Therefore, the nonlinear phase shift between $E_S$ and FL is changed. Correspondingly, the correlation value switches from positive to negative.

Figure 5b shows the correlation functions between $E_S$ and E$_{AS}$ signals. When the polarization state of $E_S$ changes from linearly polarized to circularly polarized, the correlation value $G^{(2)}(0)$ decreases first and then increases at $45^\circ$. $G^{(2)}(0)$ achieves its minimum value at an elliptically polarized state. The variation of $G^{(2)}(0)$ results from the competition between gain and dressing effect of $E_d$. Moreover, anticorrelation effect was not observed between $E_S$ and E$_{AS}$. For the SFWM process, it is insensitive to dressing effect, and the phase difference between $E_S$ and E$_{AS}$ is smaller because of the coherent nature as compared to the incoherent FL signal. So, all values of $G^{(2)}(0)$ are positive.

IV. CONCLUSIONS

In summary, we have investigated hybrid signals including fourth-order fluorescence and SFWM in both time and frequency domains at different temperatures and modulated by different polarization-dressing fields. By changing the experiment temperature and polarization states of dressing fields, the delay time, AT splitting, and proportion of SFWM and FL in hybrid signals are modulated desirably. Our experiment data are in good agreement with the theoretical results of competition between polarization-dressing and phonon effects.
In addition, the polarization dependences of hybrid correlation are compared at different temperatures. It is found that the competition between SFWM and FL signals determines the correlation shape and can be modulated by dressing and phonon effects. The switches between correlation and anticorrelation of hybrid signals are observed at different polarization states and attributed to cross-phase modulation induced by the dressing field. Our results demonstrate that phonon effect can significantly affect the nonlinear response of quantum optical devices. The dressing phonon interaction plays a vital role in determining the controlling parameters of quantum optical devices.

V. EXPERIMENTAL SETUP

Our experiments are carried out in a rare-earth Pr$^{3+}$-doped Y$_2$SiO$_5$ crystal, the concentration of Pr$^{3+}$ ion is about 0.05 atom %. The energy levels $^3H_4$ and $^1D_2$ of Pr$^{3+}$ ions are selected to couple with the pumping field. There are two crystallographic sites (denoted as sites I and II) of Pr$^{3+}$ ions in the Y$_2$SiO$_5$ crystal; Figure 6a shows their energy levels. Because two crystallographic sites have different Stark splitting, we can detect two fluorescence signals, FL1 ($\gamma_0$ to $\delta_0$) and FL2 ($\gamma_6^*$ to $\delta_0^*$). Because of the dipole–dipole interactions, we treat ions Pr$^{3+}$(I) and Pr$^{3+}$(II) as heteronuclear molecules. Therefore, we can construct the A-type three-level (0) $\leftrightarrow$ (1) $\leftrightarrow$ (3) (Figure 6c) and V-type three-level (0) $\leftrightarrow$ (1) $\leftrightarrow$ (2) systems.

The experimental setup is shown in Figure 6b. As pumping fields, we used 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, 10 Hz repetition rate, 5 ns pulse width). The pumping fields $E_1$, $E_3$, and $E_2$ (or $E_4$), counter-propagate through the YSO crystal, and the generated forward Stokes $E_5$ and backward anti-Stokes $E_3$ signals satisfy the phase-matching condition $k_1 + k_2 = k_3 + k_{AS}$. Where $k_1$ is the wavevector of the pumping fields and $k_{AS}$ is the wavevector of generated $E_3$ and $E_{AS}$ beams. The FL signal can be detected at any direction, but it diminishes quickly with distance. To detect pure SFWM signals, two photomultiplier tubes (D1 and D2) are placed at far position, where fluorescence almost vanished. Another PMT (D3) placed at near position is used to collect the hybrid signal (FL + SFWM). The polarization state of pumping fields is changed by quarter-wave plate (QWP) and half-wave plate (HWP).

**Figure 6.** (a) Simplified energy-level diagram of Pr$^{3+}$ ions in a YSO crystal. (b) Experimental setup. D: photomultiplier tube; PBS: polarized beam splitter; WP: wave plate; L: lens. (c, d) A-Type and V-type three-level systems in Pr$^{3+}$:Y$_2$SiO$_5$.

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