Bunching and antibunching in four wave mixing NV center in diamond

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Faizan Raza, Irfan Ahmed, Dan Zhang, Al Imran, Abubakkar Khan, Condon Lau, and Yanpeng Zhang

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The determination of classical and quantum states through photon bunching and antibunching phenomena may have potential applications in quantum information processing and long-distance quantum communications. We report the photon bunching and multi antibunching phenomena by generating multi-order fluorescence and four-wave mixing (FWM) at room temperature using the Nitrogen-vacancy (NV) center in diamond. We have implied FWM process to demonstrate the interference pattern emerging from NV of nano-crystals in classical, nonclassical and intermediate (classical and nonclassical) regimes. Intersystem crossing is controlled by the fluence of incident beams. The interference pattern from dominant ionization of NV$^-$ to NV$^0$ and NV$^0$ to NV$^-$ suggests the bunching and antibunching like phenomena of photons, respectively. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/1.5039979

I. INTRODUCTION

Second-order coherence known as two-photon bunching led the emergence of quantum optics. $^1$ The correlation and interference phenomena provide a solid foundation for development of quantum theory of photons. $^2$ Paul Dirac considered the single photon interference in which superposition only comes from the photon itself $^3$ However, in the case of the generation of twin/paired photons, a similar statement can be established for interference of two photons, in which superposition comes from the pair of photons jointly measured (analogous definition of Dirac) sharing the same energy level as in multi-wave mixing (MWM). $^4,5$ Recently, photon bunching in the case of polarized beam splitters (PBS) has been demonstrated using diamond NV defects. $^6$ The classical-to-quantum transition with FWM $^7$ and classical and nonclassical bunching control by nonlinear response of the media $^5,8$ can be devised using FWM.

In this regard, due to stable photon emission $^9$ with high quantum efficiency $^{10}$ of nitrogen-vacancy (NV) color center, the negatively charged NV center in diamond is configured here with FWM process to realize the intersystem crossing (ISC) by changing the fluence of lasers, which can modify the nonlinear optical response of the media $^5,8$ in nano-diamond (ND).

Multidressed suppression and enhancement of self-diffracted SP-FWM process is studied in hot atomic system including sodium hyperfine structures of the ground states. $^{11}$ In rare earth doped crystals (Pr$^{3+}$:YSO), AT-splitting caused by strong dressing effect is studied in both spectral and temporal signal in multi-level atomic systems. $^{12,13}$ Unlike Pr$^{3+}$:YSO and hot atomic systems, dressing

$^a$Faizan Raza and Irfan Ahmed contributed equally to this work
$^b$Electronic mail: condon.lau@cityu.edu.hk
$^c$Electronic mail: ypzhang@mail.xjtu.edu.cn
effect in NV center is very weak. We have studied competition between SP-FWM and multi-order fluorescence emission in hybrid signal in weak dressed atomic-like system.

In this paper, we show the classical bunching of indistinguishable photons in both two and three modes under the phonon induced mixing state of ISC in a three level V-type system with spontaneous emission of FWM by changing the fluence of the non-resonant beam. The setting of non-resonant beam to high fluence is termed as high gain regime to achieve bunching like effect. We also demonstrate the photon multi anti-bunching like effect in the low gain of spontaneous parametric FWM (SP-FWM) by fine fluence tuning of the resonant beam and fixing the non-resonant beam. The setting of resonant beam and fixing the non-resonant beam is termed as precise quantum threshold to achieve anti-bunching like effect. Furthermore, we observed intermediate (classical and nonclassical) state by exciting system in medium gain regime. For the first time, we observed multi antibunching like effect using slow light.

II. EXPERIMENTAL SETUP AND THEORETICAL MODEL

We used two 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) which are used to generate the pumping fields $E_1 (\omega_1, \Delta_1)$ and $E_2 (\omega_2, \Delta_2)$ with detuning $\Delta_i = \omega_{mn} - \omega_i$, where $\omega_{mn}$ denotes the corresponding atomic transition frequency and $\omega_i$ is the laser frequency. The sample used in our experiment was a bulk NV center crystal, consisting of substitutional nitrogen-lattice vacancy pairs orientated along the [100] crystalline direction. The sample used in our experiment contains 0.05% nitrogen per diamond crystal. The sample held in a container at 77 K in a cryostat. Acquisition time of the experiment is 100ms. The NV center is known to exist in negative (NV$^-$) and neutral (NV$^0$) charged states as shown in Fig. 1(e). The features of NV$^-$ and NV$^0$ are their optical zero phonon lines (ZPL) at 1.945eV (637nm) and 2.156eV (575nm) respectively. The NV$^-$ center is treated as a three-level system with a ground triplet state ($^3$A), a triplet excited state ($^3$E), and an intermediate singlet state ($^1$A). The ground and excited energy level of site NV$^0$ is labeled by $^3$E and $^3$A$_1$ respectively. In our experiment, with $E_1$ (575nm) and $E_2$ (637nm) excitations, the hybrid signals which include SP-FWM and fourth-order fluorescence (FL) signals, are generated simultaneously. Figure 1(c) shows hybrid signals detected by D3. The sharp peak shown in fig. 1(c) comes from direct transition from $^3$E, $m_s = 0$⟩ to $^3$A$_2$, $m_s = 0$⟩ with SP-FWM signal emission. The second peak relates to intersystem crossing (ISC) by phonon assisted transition and then decays to the ground state with fluorescence emission. In Fig. 1(c2), $t_1$-$t_4$ represents time position of boxcar gate of the hybrid signal.

By opening $E_1$ and $E_2$ fields, the Stokes $E_s$ and anti-Stokes $E_{as}$, are generated with phase-matching condition $k_1 + k_2 = k_{as} + k_s$. Both $E_s$ and $E_{as}$ are detected pair of photomultiplier tube

FIG. 1. (a) and (b) three level system in NV center exited by two and three laser beams, respectively. (c) Time domain signal from the NV center, where $t_1$-$t_4$ are different positions of boxcar gate. (d) Experimental setup, (e) The NV$^0$ and NV$^-$ charge state inter-conversion mechanism.
PMT1 and PMT3, respectively as depicted in Fig. 1(d). The FL signal generated along with SP-FWM is also detected by PMT2. The perturbation chain of $E_{\text{as}}$ in a V-type three level system is $\rho_{00}^{(0)} \rightarrow \rho_{20}^{(1)} \rightarrow \rho_{00}^{(2)} \rightarrow \rho_{100}^{(3)}$. The density matrix for anti-Stokes in a V-type system can be written as
\[
\rho_{\text{as}}^{(3)} = \frac{-iG_1 G_2 G_2}{(\Gamma_{20} + i\Delta_2)(\Gamma_{00} + i(\Delta_2 - \Delta_1) + i(\Delta_1 - \Delta_3) + d_1)}
\]
where $d_1 = |G_2|^2/[(\Gamma_{12} + i(\Delta_1 - \Delta_3))]$ is the dressing effect. $G_i = \mu_i E_i/h$ is the Rabi frequency of $E_i$ with the electric dipole moment ($\mu_i$) of levels $|i\rangle$ and $|j\rangle$. $\Gamma_i$ is the transverse decay rate. Lifetimes of anti-Stokes signal can be written as $\Gamma = \Gamma_{10} + \Gamma_{00} + \Gamma_{20}$. The intensities of the $E_S$ and $E_{\text{as}}$ are described as
\[
\delta\tilde{I}_{S/\text{as}}(t) = I_{0(S/\text{as})} e^{-\Gamma_{S/\text{as}}t}
\]
where $\delta\tilde{I}_{0(S)} \propto |\rho_{100}^{(3)}|^2$ and $\delta\tilde{I}_{0(\text{as})} \propto |\rho_{100}^{(3)}|^2$. Now we discuss FL signal in V-type system generated via perturbation chain $\rho_{00}^{(0)} \rightarrow \rho_{10}^{(1)} \rightarrow \rho_{00}^{(2)} \rightarrow \rho_{20}^{(3)} \rightarrow \rho_{00}^{(4)} \rightarrow \rho_{30}^{(5)} \rightarrow \rho_{33}^{(6)}$ can be written as
\[
\rho_{33}^{(6)} = \frac{|G_1|^2 |G_2|^2 |G_3|^2}{(\Gamma_{20} + i\Delta_1)(\Gamma_{20} + i\Delta_2 + d_1^*) (\Gamma_{00} + d_2^*) (\Gamma_{00} + i\Delta_3) \Gamma_{00} \Gamma_{22}}
\]
where $d_1^* = |G_2|^2/[(\Gamma_{10} + |G_1|^2/|\Gamma_{20} + i\Delta_1|)]$ and $d_2^* = |G_3|^2/[(\Gamma_{10} + i\Delta_3)]$ are the nested and cascaded dressing effect caused the fields, respectively. The third order correlation function can be normalized as
\[
G_{\tau_1, \tau_2, \tau_3}^{(3)}(t_1, t_2, t_3) \propto 1 + \sin^2 \left( \frac{\Delta \omega(t_1 - t_2)}{2} \right) + \sin^2 \left( \frac{\Delta \omega(t_2 - t_3)}{2} \right) + \sin^2 \left( \frac{\Delta \omega(t_3 - t_1)}{2} \right)
\]
\[
+ 2 \sin \left( \frac{\Delta \omega(t_1 - t_2)}{2} \right) \sin \left( \frac{\Delta \omega(t_2 - t_3)}{2} \right) \sin \left( \frac{\Delta \omega(t_3 - t_1)}{2} \right)
\]
\[
G_{\tau_1, \tau_2}^{(3)}(t_1, t_2) = 4(1 + \alpha^2) S_1 e^{-\Gamma_{10} t_1 + \Gamma_{10} t_2} + 2 \alpha^2 S_2 [\cos(\Delta \tau_1 \tau_2 + \phi) e^{-\Gamma_{10} t_1 + \Gamma_{20} t_2}] + \Gamma_{10} R_{22} R_{33}
\]
where $\tau_1 \neq \tau_2 \neq \tau_3, \tau_1 = t_1 - t_2, \tau_2 = t_2 - t_3$ and $\tau_3 = t_3 - t_1$.

### III. RESULTS AND DISCUSSION

Herein, we investigated two-photon bunching and antibunching like phenomenon by changing fluence of off resonant $E_1$ beam from 89 (mJ/cm$^2$) to 12 (mJ/cm$^2$). For measuring and calculating two-photon bunching, we blocked the PMT3 and kept the rest of the PMTs on scanning mode. The second order correlation function can be described as $G^{(2)}(\tau_1) = \langle \delta I(t_1) \delta I(t_2) \rangle$. Where $\tau_1$ is the selected time delay and $\delta I(t_{1/2})$ are intensity fluctuations. By simplifying $G^{(2)}(\tau_1)$, two photon bunching and anti-bunching can be written as $G^{(2)}(\tau_1) \propto 1 + \sin^2(\Delta \omega(t_1 - t_2)/2)$ and $G^{(2)}(\tau_1) = |R_{33} R_{E}|^2 (e^{-2\Gamma_{10} t_1} + e^{-2\Gamma_{10} t_2} - 2\cos(\Delta \tau_1 \tau_2) e^{-(\Gamma_{10} + \Gamma_{20}) t_{12}})$. Where $\Gamma_{10}$ and $\Gamma_{20}$ are constants. By opening $E_1$ and $E_2$ fields, $E_s$ and $E_{\text{as}}$ are generated along with FL emission in a composite channel. The competition between SP-FWM and FL composite signal determines the sign of correlation function to be positive or negative (Fig. 2(a1)--(a6)). By changing fluence of $E_1$ from 89 (mJ/cm$^2$) to 12 (mJ/cm$^2$), photon bunching is switched to photon antibunching like phenomenon. In order to understand switch between classical (bunching) and nonclassical (antibunching), we use double dressing effect of $E_1$ and $E_2$ beams. Dressing effect can be used to control precise emission of FL and SP-FWM emission in a composite signal. In case of single dressing field, FL signal $\rho_{11}^{(2)} = |G_1|^2/|\Gamma_{11}| + |G_2|^2/|\Gamma_{11}|$ increases gradually with increasing fluence of $E_1$, when the fluence is further increased the FL signal evolves from dressing state to suppression, caused by the dressing effect $|G_1|^2/|\Gamma_{11}|$ of $E_1$. With further suppression of FL signal the SP-FWM emission is enhanced gradually at high fluence. However, in double dressing scenario, dressing effect dependence on fluence is slightly reversed. When $E_1$ is at 89 (mJ/cm$^2$), strong FL $\rho_{11}^{(4)} = |G_1|^2 |G_2|^2 / (\Gamma_{10} + \Gamma_{12} + d_{11}) \Gamma_{11} \Gamma_{00}$.
comes from strong dressing effect of composite channel behaves as pure SP-FWM channel due to significant increase in SP-FWM, which

\[ E^2 \text{ beam is set to 1 (mJ/cm}^2 \text{)} \]

antibunching like phenomena is observed as shown in Figs. 2(b2)–2(b3). As fluence of the resonant emission decreases, the interference between the Stokes and anti-Stokes emission becomes dominant, and antibunching like phenomena is observed as shown in Eq. (1). As fluence of \( E^2 \) is reduced, the interference between the Stokes and anti-Stokes emission becomes dominant, and antibunching like phenomena is observed as shown in Figs. 2(b2)–2(b3). As fluence of the resonant beam is set to 1 (mJ/cm²), the anti-bunching like effect becomes more prominent (Fig. 2(b3)) due to strong dressing effect of \( E^1 \).

\[ \text{Intensity of FL } \rho_{FL} = \rho_{FL}^{(3)} \text{ dominates overall SP-FWM } \rho_{SP-FWM} = \rho_{SP-FWM}^{(3)} \text{ in composite signal i-e } \rho = \rho_{FL} + \rho_{SP-FWM} \text{ due to strong dressing effect of } E^1 \text{ i-e } |G_2|^2/|G_1|^2 < 1 \text{ as seen from the variable } d_{11}. \]

When \( P_1 \text{ of } E^1 \) is gradually decreased, amplitude of photon bunching decreases as fluence of \( E^1 \) is reduced as shown in Fig. 2(a3). The photon bunching calculated in Figs. 2(a1)–2(a3) is the result of the dominant classical emission of FL photons due to the fluence of the \( E^1 \) beam under high gain regime under double dressed state, and hence said to be classical bunching. Figure 2(c1) shows the calculated two photon bunching which corresponds to Figs. 2(a1).

In order to switch pure FL emission (classical) to pure SP-FWM emission (nonclassical) double dressing effect is used. Next, we set \( E^1 \) to low threshold (12 (mJ/cm²)) and change the fluence of \( E^2 \) (637 nm) from 89 (mJ/cm²) to 12 (mJ/cm²), to obtain quantum threshold to excite spin state \( |A_2, ms=0 \rangle \) to \( |E, ms=0 \rangle \) (NV⁺). The curves obtained with the said experimental conditions are calculated using \( G_2^{(3)}(\tau_1) \) and presented in Figs. 2(b1)–2(b3). At first, \( E^1 \) is set at 89 (mJ/cm²) to achieve the ionization from NV⁰ to NV⁺. When \( E^2 \) is at high fluence, population transfer happens from spin state \( |A_2, ms=0 \rangle \) to \( |E, ms=0 \rangle \) (NV⁻) which leads to the prominent emission of SP-FWM. Composite channel behaves as pure SP-FWM channel due to significant increase in SP-FWM, which comes from strong dressing effect of \( E^2 \) \((|G_2|^2/|G_1|^2) \) mentioned in Eq. (1). As fluence of \( E^2 \) is reduced, the interference between the Stokes and anti-Stokes emission becomes dominant, and antibunching like phenomena is observed as shown in Figs. 2(b2)–2(b3). As fluence of the resonant beam is set to 1 (mJ/cm²), the anti-bunching like effect becomes more prominent (Fig. 2(b3)) due to decrease in dressing effect of \( E^2 \). The bunching count at \( \tau=0 \) is less than the side peaks and also less than zero. All results shown in Fig. 2(b) suggests antibunching like effect. This phenomenon precisely meets that theoretical results of antibunching like effect depicted in Fig. 2(c2).

By varying fluence of \( E^1 \) from 63 (mJ/cm²) to 25 (mJ/cm²) (medium gain regime), composite signal that includes prominent FL and SP-FWM emission can be obtained. Unlike Fig. 2, we cannot observe switch between pure photon bunching to antibunching like effect in Fig. (3). The three photon bunching calculated in Figs. 3(a1)–3(a4) are obtained using Eq. (3), which shows the direct relationship of off-resonant field with indistinguishable photon bunching. When \( P_1 \text{ of } E^1 \) is 63 (mJ/cm²), dominant FL emission \( \rho_{FL}^{(3)} \) and weak SP-FWM emission is observed as shown in Fig. 3(a1). As the \( P_1 \text{ of } E^1 \) decreases, the photon bunching count decreases, leading to the reduced amplitude of \( G_2^{(3)}(\tau_1, \tau_2, \tau_3) \) in a positive scale. One must note that this is the minimum count achieved with \( E^1 \) is set at medium fluence and \( E^2 \) at 2 (mJ/cm²). To achieve upper quantum threshold under SP-FWM configuration and emission from NV⁺, we fix the fluence of \( E^1 \) at 12 mJ/cm² and change \( E^2 \) from 63 (mJ/cm²) to 25 (mJ/cm²). When \( E^2 \) is at 63 (mJ/cm²), SP-FWM emission is enhanced in

![FIG. 2. Measured second-order temporal correlation function (a1)-(a3) by changing the fluence of \( E^1 \) (575 nm) from 89 (mJ/cm²) to 12 (mJ/cm²) and fixing \( E^2 \) (637 nm) at 12 (mJ/cm²). (b1)-(b3) by changing the fluence of \( E^2 \) (637 nm) from 89 (mJ/cm²) to 12 (mJ/cm²) and fixing \( E^1 \) (575 nm) at 12 (mJ/cm²). (c1) and (c2) Simulated second-order temporal correlation functions corresponding to (a1) and (b1), respectively.](attachment://image.png)
FIG. 3. (A) Third-order correlation function versus delayed time by changing fluence of \( E_1 \) (a1) 63 (mJ/cm\(^2\)) (a2) 50 (a3) 3, (a4) 25 (mJ/cm\(^2\)) and by fixing \( E_2 \) at 25 (mJ/cm\(^2\)). (B) third-order correlation function versus delayed time with changing fluence of \( E_2 \) (b1) 63 (mJ/cm\(^2\)), (b2) 50 (mJ/cm\(^2\)) (b3) 38 (mJ/cm\(^2\)), (b4) 25 (mJ/cm\(^2\)) and by fixing \( E_1 \) at 12 (mJ/cm\(^2\)).

composite channel but still FL emission is observed. The dressing effect of \( E_1 \) and \( E_2 \) beams does not cancel each other as they both have significant contributions at medium fluence. In Figs. 3(b1)–3(b4), a small dip is observed along with dominant peaks, which can be explained from strong FL emission in composite signal when \( E_1 \) is set at medium fluence. In Figs. 3(b1)–3(b4), when \( E_2 \) is set at medium fluence, dip becomes more prominent than peak which suggests that antibunching-like effect. Although, SP-FWM \( \rho^{(3)}_{20} \) is dominant but FL emission cannot be ignored which is evident small peak in Figs. 3(b1)–3(b4).

In Fig. 4, by introducing \( E_3 \), photon bunching and multi antibunching like effect is observed in double V-type system by varying boxcar gate position. The anti-Stokes signal can be written via perturbation chain \( \rho^{(0)}_{00} \rightarrow \rho^{(1)}_{10} \rightarrow \rho^{(2)}_{20} \rightarrow \rho^{(3)}_{30} \rightarrow \rho^{(4)}_{40} \rightarrow \rho^{(5)}_{50(\alpha_1)} \) as \( \rho^{(5)}_{\alpha_1} = G_{1}G_{2}G_{3}G_{3}/d_{0}d_{1}d_{2}d_{3}d_{4} \), which will be parametrically amplified\(^{13,16} \) where \( d_{1} \) is the dressing effect. In Fig. 4(a), the experimental conditions are same as that of Fig. (2a) besides the introduction of \( E_3 \). At high fluence of \( E_1 \), pure photon bunching is observed which can be explained from strong dressing effect of \( E_1 \) \((|G_2|^2/|G_1|^2 < 1) \) mentioned in Eq. (2) and shown in Fig. 4(a1). One can notice that as boxcar gate position is changed from \( t_1 \) to \( t_4 \) (Fig. 1(c)), the SP-FWM emission increases gradually in comparison with the FL. SP-FWM emission is enhanced when gate position is at \( t_4 \). Next in order to demonstrate the multi antibunching like phenomenon, \( E_1 \) beam is set at 1 (mJ/cm\(^2\)). At prècised quantum threshold the influence of \( E_3 \) beam becomes strong. Switch between FL emission (Fig. 4(A)) and SP-FWM (Fig. 4(B)) can be explained by nested dressing condition \( |G_2|^2/|G_1|^2 > 1 \) (Eq. (3)). In Figs. 4(b1)–4(b4), multi antibunching like effect is observed which can be explained from slow light effect of \( E_3 \). By introducing \( E_3 \) beam under the quantum threshold, \( \rho^{(5)}_{\alpha_1} \) emission along with group

FIG. 4. (A) third-order temporal correlation function detected by fixing the fluence of \( E_1 \) at 89 (mJ/cm\(^2\)) and by moving the position of the boxcar gate (t1-t4). (B) third-order temporal correlation function detected by fixing the fluence of \( E_1 \) at 12 (mJ/cm\(^2\)) and by moving the position of the boxcar gate (t1-t4).
velocity $v_g = -\left(\partial \rho_{10}^{(1)} / \partial \Delta \lambda_{ij}\right)_{\Delta \lambda = 0}$ and the trigger time $t_1 = S/v_g$ of PMT1 will change, which is very much similar to the effect of an internal delay $\tau_1$ of Eq. (4) caused by $\rho_{as}^{(5)}$.

IV. CONCLUSION

In conclusion, second order and third order photon counting has been demonstrated experimentally and theoretically. The four-wave mixing process was configured to demonstrate the interference pattern. The intersystem crossing and phonon-induced mixing by changing the fluence of laser beams suggested the photon bunching and antibunching like effect. Moreover, the shift from the classical (bunching) to the quantum (antibunching) state, has been demonstrated by controlling the quantum threshold of SP-FWM configuration. These results have applications in designing logic gates and solid state quantum computation.

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