Investigation of multi-bunching by generating multi-order fluorescence of NV center in diamond

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Fabricating electronics from solid-state quantum emitters is a promising strategy for the miniaturization and integration of electronic devices. However, the practical realization of solid-state quantum devices and circuits for signal transmission and processing at room temperature has remained challenging. Herein, we investigated the multi-bunching phenomenon by generating multi-order fluorescence from a pseudo-thermal source at room temperature using the nitrogen-vacancy (NV) center in diamond. We demonstrate the shift in time of multi-bunching by controlling the effect of dressing to realize logical gates and transistor switching operations. We also suggest the optimization of the time propagation delay (TPD) of the gate circuit by changing the boxcar gate position.

Introduction

Currently, research on solid-state quantum devices at room temperature has drawn much attention. The nitrogen-vacancy (NV) center in diamond is a solid-state system that combines desirable spin coherence with atomic-like optical transitions. It shows excellent characteristics such as a bright photostable optical transition, desirable long spin coherence time at room temperature and optical controllability of the ionic states. It can be considered an efficient pseudo-thermal light source for applications in the fields of quantum information, quantum optics, and metrology. For quantum information applications, quantum gates based on optics play an important role in quantum-information processing and have been thoroughly investigated in the recent years. For example, quantum gates are widely used to encode information in quantum cryptography, as such as quantum key distribution and quantum security direct communication. They are also used in entanglement generation, quantum dense coding, entanglement purification, remote state transmission, and so on. Quantum gates are also important elements of quantum computers and quantum algorithms.

In classical and quantum optics, multi-order fluorescence occurs due to differences in energy levels, while multi-bunching results from the multi-order fluorescence of constructive interference from indistinguishable photons. From the viewpoint of quantum mechanics, in explaining the photon bunching effect, Glauber attributed the two-photon bunching to the corresponding two-photon interference effect. Similarly three-photon bunching has also been interpreted as the result of three-photon interference.

In this paper, we report the novel experiments, in which multi-bunching was generated by multi-order fluorescence as a pseudo-thermal source from nitrogen-vacancy (NV) centers in diamond. Based on analytical and numerical simulation, the effect of quantum path interference among the multi-bunching phenomenon is discussed. Moreover, a NOR logical gate and a transistor switching model were developed based on the photon bunching shift with time caused by a dressing effect. Further, the time propagation delay (TPD) of the gate circuit can be optimized by changing the period of Rabbi Oscillations.

Experimental setup

The sample used in our experiment was a bulk NV center crystal, consisting of substitutional nitrogen-lattice vacancy pairs orientated along the [111] crystalline direction. The sample was doped with nitrogen at less than 5 parts per billion (ppb) and held in a container at room temperature. Fig. 1(a) presents the co-existing negative (NV−) and neutral (NV0) charge states and the energy level structure of the multi-order fluorescence signal emitted by the diamond NV center, which constructs a double V-type system. The identifying features of NV− and NV0 are their optical zero phonon lines (ZPLs) at 1.945 eV (637 nm) and 2.156 eV (575 nm), respectively, and the associated vibrionic
bands extending from their ZPLs to higher/lower energy in absorption/emission spectrum. The NV center with a triple ground state (\(^3\)A\(_2\)), a triplet excited state (\(^3\)E), and the intermediate singlet state (\(^1\)A and \(^1\)E) is treated as a three-level system. The ground energy level of site NV\(^0\) is labeled by \(^3\)E and the excited state by \(^3\)A\(_2\). It is known that the van der Waals force describes interactions between molecules; hence, in the system, we considered the ground states (\(^3\)E and \(^3\)A\(_2\)) of the NV\(^0\) and NV\(^-\) states, which compose a heteronuclear-like molecule energy level,\(^1,2,3\) to be within the valid range of van der Waals forces. This molecule energy level can be expressed as
\[
\Delta E = \sum (C_n/R_n^6)/\hbar (n = 6, 8, 10, 12, 13, and 14),
\]
where \(\sum (C_n/R_n^6)\) represent the van der Waals interactions of the \(^3\)E and \(^3\)A\(_2\) states.\(^4\) Therefore, the ground state levels \(^3\)E and \(^3\)A\(_2\) are equivalent to one-level as shown in the shaded region in Fig. 1(a).

Due to electronic spin–spin interaction, the two triplet states \(^3\)A\(_2\) and \(^3\)E are further split into three spin fine structures as \(m_s = 0\) and \(m_s = \pm 1\). The energy difference between \(m_s = 0\) and \(m_s = \pm 1\) for a triplet ground state is \(D = 2.8\) GHz, while for a triplet excited state, it is \(D = 1.42\) GHz, where \(D\) is also called zero field splitting. The fine structure in the triplet excited state (\(^3\)E) further splits into four hyperfine structures (see Fig. 1(a): \(A\(_1\), \(A\(_2\), \(E\(_x\), and \(E\(_z\))\) under the effect of electronic spin–spin interaction and spin–orbit interaction or both.\(^5\) Fig. 1(b) shows the schematic of our experimental setup. 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) were used to generate the pumping fields \(E\(_1\) (637.1 nm) \(E\(_2\) (575 nm)\) and the coupling field \(E\(_1\) (637.5 nm)\), which in turn, were used to drive the transitions. These transitions generated the multi-order fluorescence signals detected by three individual photodetectors denoted as D1, D2, and D3. After detection, the output pulses from D1, D2 and D3 are coupled to a three-fold coincidence counting circuit. Although the fluorescence signals were detected at different detectors, we could not predict which photon was detected at which detector. This detection and generation process made these photons indistinguishable and entangled in the classical regime.

### Theoretical model

With the introduction of a single strong pumping field \(E\(_1\) (637.5 nm), the two-level system \(|0\rangle \leftrightarrow |1\rangle\) (see Fig. 1(a)) came under operation and second-order fluorescence was generated. The second-order fluorescence signal (FL1) can be generated through the perturbation chain \(\rho^{(0)}_{00} \rightarrow \rho^{(1)}_{01} \rightarrow \rho^{(2)}_{11}\) and the signal intensity can be described from the diagonal density matrix elements as \(\rho^{(2)}_{11} = -|G_1|^2 / (\langle \Gamma_{10} + i\Delta_1 \rangle \Gamma_{11})\). Later, by injecting another pumping field \(E\(_2\) (575 nm), a V-type three-level system \(|0\rangle \leftrightarrow |1\rangle \leftrightarrow |2\rangle\) (see Fig. 1(a)) came under operation and generated a fourth-order fluorescence signal. The fourth-order fluorescence signal (FL2) process can be described by the perturbation chain \(\rho^{(0)}_{00} \rightarrow \rho^{(1)}_{01} \rightarrow \rho^{(2)}_{02} \rightarrow \rho^{(3)}_{03} \rightarrow \rho^{(4)}_{04}\) and \(\rho^{(5)}_{30} \rightarrow \rho^{(6)}_{33}\), whose diagonal density matrix element is given as
\[
\rho^{(2)}_{11} = -|G_2|^2 |G_1|^2 / \langle (\Gamma_{10} + i\Delta_1) \Gamma_{11} \rangle \langle \Gamma_{12} \Gamma_{20} \rangle.
\]

In the process of fluorescence propagation, since \(e_{0AFL1}E_{FL1} = N_{FL1}\rho^{(0)}_{00}, e_{0AFL2}E_{FL2} = N_{FL2}\rho^{(0)}_{00}\) and \(e_{0AFL3}E_{FL3} = N_{FL3}\rho^{(0)}_{00}\), we can write down the susceptibilities of the FL1, FL2 and FL3, respectively.\(^5\) Furthermore, the group velocities of the FL1 (\(v_{FL1}\)), FL2 (\(v_{FL2}\)) and FL3 (\(v_{FL3}\)) pulses are given by
\[
v_{FL1(2,3)}(\omega, \gamma) = \frac{c}{1 + \left(\frac{\partial \omega_{FL1(2,3)}}{\partial \omega_{FL1(2,3)}}\right)}
\]

When the three detectors D1, D2, and D3 are positioned at equal distance \(S\) to monitor the photon counting at each individual detector, we can obtain time \(t_i = S/v_{FLi} (i = 1, 2, 3)\) when the detectors are triggered.

As shown in Fig. 1(b), after detection, the output pulses from D1, D2 and D3 are sent to a three-fold coincidence counting circuit, which gives a three-photon counting histogram as a function of \(t_1 - t_2, t_1 - t_3\) and \(t_2 - t_3\), where \(t_j (j = 1, 2, 3)\) is the registration time of the photo-detection event at D1, D2 and D3, respectively. The third order correlation function of pseudo-thermal light comes from a coherent superposition of six different indistinguishable probable amplitudes and can be calculated by using the density operator of the field.\(^6\)
In such cases, the Glauber’s correlation function\textsuperscript{17} for pseudo-thermal light can be normalized as

\[
g^{(3)} (t_1, t_2, t_3) = 1 + \sin^2 \left( \frac{\Delta \Omega t_{12}}{2 \pi} \right) + \sin^2 \left( \frac{\Delta \Omega t_{13}}{2 \pi} \right) + \sin^2 \left( \frac{\Delta \Omega t_{23}}{2 \pi} \right) + 2 \sin \left( \frac{\Delta \Omega t_{12}}{2 \pi} \right) \sin \left( \frac{\Delta \Omega t_{13}}{2 \pi} \right) \sin \left( \frac{\Delta \Omega t_{23}}{2 \pi} \right)
\]

(2)

where \( t_{12} = t_1 - t_2 \), \( t_{13} = t_1 - t_3 \), and \( t_{23} = t_2 - t_3 \); \( \Delta \Omega \) represents the bandwidth of the pseudo-thermal light source.\textsuperscript{14} The first term constitutes background noise, while the other four terms involve two- or three-photon bunching (multi-phonon bunching). With regard to N-order imaging with pseudo-thermal light, the maximum image visibility can be defined as \( V_{\text{max}} = \frac{g_{N\text{max}}^N - g_{N\text{min}}^N}{g_{N\text{max}}^N + g_{N\text{min}}^N} \).\textsuperscript{18} From eqn (2), we can observe that when \( t_1 = t_2 = t_3 \), \( g^{(3)}(t_1, t_2, t_3) = 6 \), and the \( g^{(3)} \) function achieves a maximum contrast of 6 to 1 (visibility \( V_{\text{max}} \approx 71\% \)), which is the result of the coexistence of three two- and three-photon bunching in the intensity noise correlation. If we switch-off D3, the second order temporal correlation (“two-photon bunching”) between the detector D1 and D2 can be obtained as

\[
g^{(2)} (t_1, t_2) = 1 + \sin^2 \left( \frac{\Delta \Omega t_1}{2 \pi} \right)
\]

(3)

From eqn (3), we can see that when \( t_1 = t_2 \), \( g^{(2)}(t_1, t_2) = 2 \), and the \( g^{(2)} \) function achieves a maximum contrast of 3 to 1 (visibility \( V_{\text{max}} \approx 33\% \)). Comparing eqn (2) and (3), we can predict the enhancement of the image resolution in third-order imaging. The resolution of the second-order imaging is determined by the last term of eqn (3), which is the product of two sinc functions. The resolution of third-order imaging is influenced by the last term of eqn (2), which is the product of three sinc functions.

### Experimental results and discussion

Herein, we investigated two-photon bunching by treating multi-order fluorescence as a pseudo-thermal light source, which is determined by eqn (3), in which the resolution of the second-order imaging is the product of two sinc functions as determined by the last term of eqn (3). The multi-order fluorescence signals are generated by two off-resonant beams \( E_1 \) and \( E_2 \) and a pumping beam \( E_3 \) in a double V-type level system (see Fig. 1(a)).

For measuring and calculating two-photon bunching, we blocked the detector D3 and kept the rest of the detectors on scanning mode. Fig. 2(a1–a3) show the experimental results of two-photon bunching by moving the position of the boxcar gate (shown in Fig. 2(c)) when D3 is turned off and both D1 and D2 are fixed in the same distance of the transverse plane. The corresponding simulation shown in Fig. 2(b1–b3) replicates the same pattern of experimental data. We can predict that by moving the position of the boxcar gate, the period of Rabi Oscillations can be controlled. In order to explain this phenomenon, we resorted to the self-Kerr effect by introducing the phase factor \( e^{i\omega t} \) into the Rabi frequency \( \Omega_0 = \mu_0 E_0 / \hbar \); hence, the effective Rabi frequency \( \Omega = \sqrt{\Delta \omega^2 + 2 |G_0| e^{i\omega t}} \) will also be changed. Further, from eqn (3), \( \Delta \omega = \Omega_0^2 \Gamma_1 / \Omega_2^2 + G_0^2 \Gamma_1 / \Omega_2^2 \) represents the bandwidth of the pseudo-thermal light source and \( \Gamma_1 \) and \( \Gamma_2 \) represent the decoherence rate of the \( i \) and \( j \) levels, respectively.\textsuperscript{19} This explains precisely the moving of the position of the boxcar gate, which changes the period of Rabi Oscillations.

Subsequently, we discuss the coexistence of two- and three-photon bunching in the intensity noise correlation formed by fixing the boxcar gate in the position as shown in Fig. 2(a2). This phenomenon is determined by eqn (2), in which the resolution of third-order imaging is the product of three sinc functions, influenced by the last term of eqn (2). The experimental results (Fig. 3(a1–a3)) of the third-order temporal correlation function of pseudo-thermal light, detected by changing the power of \( E_2 \) (575 nm), shows that one dominant peak is surrounded by three small peaks. The dominant peak can be assessed by the superposition of the three two-photon bunching and three-photon bunching. The other three smaller peaks are primarily caused by quantum path interference between the three two-photon bunching and a smaller contribution of the one three-photon bunching. Furthermore, by changing the power of \( E_2 \) (575 nm) from 1 mW to 7 mW, the dominant peak is subjected to shift with time, while the other three smaller peaks only change their position with time. In order to better explain this phenomenon, we introduced the effect of external dressing of \( E_3 \) (575 nm), so the polarization equations can be written as

\[

\rho_{10}^{(1)} = -|G_1|^2 / \left( \Gamma_{10} + i \Delta_1 + (G_2^2 / \Gamma_{12} - i (A_1 - A_2)) \right),

\rho_{20}^{(1)} = -|G_2|^2 / (\Gamma_{20} + i A_2) \quad \text{and} \quad \rho_{03}^{(1)} = -|G_3|^2 / \left( \Gamma_{30} + i A_3 + (G_2^2 / \Gamma_{32} - i (A_3 - A_2)) \right).

\]

By changing the power of \( E_2 \) (575 nm) from 1 mW to 7 mW, the susceptibilities of second-order fluorescence FL1 \( (j_{11,1}) \) and sixth-order fluorescence FL3 \( (j_{22,2}) \) will change. Moreover, according to the group velocity equation (shown in eqn (2)) the detector trigger time \( t_1 \) and \( t_3 \) will change, which is similar to the effect of an internal delay. We defined \( t_{12} = t_1 - t_2 \), \( t_{13} = t_1 - t_3 \), and \( t_{23} = t_2 - t_3 \). By scanning \( t_{12} \) and \( t_{23} \) and fixing \( t_{13} \), we can obtain the simulation results (shown in Fig. 3(b1–b3)), which are in good agreement with the experimental results.
When the power of $E_2$ (575 nm) is adequately high, the intensity of three smaller peaks will be enhanced and will have different intensities due to the effect of quantum path interference among three-photon bunching and three two-photon bunching. However, the intensity of photon bunching is suppressed as shown in Fig. 3(a4). This suppression suggests that the visibility of third-order thermal light imaging is reduced and the experimental results are in good agreement with the theoretical results. Fig. 3(c1–c3) shows the transistor-switching model. Controlled by the dressing effect, the bunching shifts with time controlled by the dressing effect. (d) NOR bunching logic gate model for a NOR logic gate consisting of three positions (A, B, Y shown in (c4)) and the corresponding truth table for the NOR gate. In this device, the position of the bunching labeled as A and B were defined as the two-input terminal and the center position labeled as Y is defined as device output.

Finally, as shown in Fig. 4, we investigated the two- and three-photon bunching in the intensity noise correlation by fixing the boxcar gate in the position as shown in Fig. 2(a3). By changing the power of $E_2$ (575 nm) from 1 mW to 7 mW, the dominant peak shifted and the other smaller peaks changed their position with time. This phenomenon can be described by the effect of the external dressing of $E_2$ (575 nm), which is similar to the theoretical interpretation of the internal-delay-like event caused by the effect of the external dressing. Comparing the results of Fig. 3, the number and the intensity of small peaks around the dominant peak increases and the intensity of the photon bunching is intensely suppressed due to conservation of energy. The switching contrast drops rapidly (from 71% to 35%) due to power changes as shown by comparing Fig. 4 with the switching contrast (from 71% to 60%) shown in Fig. 3. The stability of the logic gate also becomes a matter of consideration. This suggests that the position of the boxcar gate, as shown in Fig. 2(a3), is more sensitive to the dressing effect; the effect of the internal-delay-like event is more evident, which suggests that the time propagation delay can be optimized. Hence, the balance between the switching contrast $c = (V_{on} - V_{off})/(V_{on} + V_{off})$ and the time propagation delay, which are important measures of the speed of the gate circuit, will be significantly optimized. Compared with conventional logic gates, the potential speed and energy requirement can also reach the microsecond and pJ order of magnitude, respectively. However, in terms of the device stability, the proposed logic gate design is advantageous because the NV center is a potential single-photon source with a long coherence time. This makes the device a potential option for future quantum computing and quantum chips.

**Conclusion**

In conclusion, we investigated multi-bunching in the intensity noise correlation by using multi-order fluorescence in a diamond NV center as the pseudo-thermal light source. On the one hand,
due to the effect of dressing, the bunching peak can shift with time and the other smaller interference peaks also change their position with time, which can thus realize a transistor switching and a photon bunching NOR logic gate model. On the other hand, the time propagation delay of the gate circuit can be optimized by changing the period of Rabi oscillations.

Conflicts of interest
There are no conflicts to declare.

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