M1 INTERNSHIP REPORT

SIMULATION OF SINGLE PHOTON EMISSION IN DOT-IN-ROD NANOCRYSTAL

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Introduction

Laboratoire Kastler Brossel

The Kastler Brossel Laboratory (LKB) is one of the main actor in the field of fundamental physics of quantum systems. The laboratory was funded in 1951 by Alfred Kastler and Jean Brossel. This lab is attached to three agencies including the National Centre for Scientific Research(CNRS), the École Normale Supérieure(ENS) and the Université Pierre et Marie Curie(UPMC). Many new themes have appeared recently in this field, such as quantum entanglement or Bose-Einstein Condensation in gases, which leads to a constant renewal of the research carried out in the laboratory.
Presently its activity focus on several themes: cold atoms (bosonic and fermionics systems), atom lasers, quantum fluids, atoms in solid helium, quantum optics, cavity quantum electrodynamics, quantum information and quantum theory of measurement, quantum chaos, high-precision measurements. The themes lead not only to a better understanding of fundamental phenomena, but also to important applications, like more precise atomic clocks, improvement of interferometric gravitational wave detectors and new methods for biomedical imaging[1].

Quantum Optics

The group research area is centered on the quantum properties of light that is produced by many different optical systems. It consists of experimental and theoretical studies concerning the shaping of quantum fluctuations of light, the generation of quantum correlations and entangled states, the interaction between quantum light and matter either dilute or condensed, and the use of non-classical light to improve the sensitivity of optical measurements. The groups is mostly interested in the so-called "continuous variable regime", in which one considers the quantum properties of the electric field of the optical wave, and where single photons are not distinguishable.
The rapidly growing field of quantum information requires in particular specific resources to operate its protocols. The group puts lots of efforts in the realization of quantum devices such as non-classical light sources, quantum memories and quantum repeaters. In this very competitive area, the group has proposed and developed many original contributions.
Recently, the group has extended the study of quantum effects in optical systems to the more general thematics of multimode quantum optics where the system under study has many degrees of freedom and thus possibly conveys a huge amount of information. This is for example the case of optical images, which span over many transverse modes, and of optical frequency combs which span over many frequency modes. The group explores this subject both theoretically and experimentally, from the introduction of new concepts to the improvement of highly sensitive measurement and applications to quantum information processing.
The group is also interested in interaction between quantum optics and condensed matter: polariton quantum gas in semi-conductor quantum well microcavities, generation of single and entangled photons by semiconductor nanocrystals.[2]

1 Basic Concept

1.1 Exciton

An exciton is a bound state of an electron and an electron hole which are attracted to each other by the electrostatic coulomb force. The exciton, an electrically neutral quasiparticle, exists in insulators, semiconductors and in some liquids. It can transport energy without transporting net electric charge, so it is regarded as an elementary excitation in the condensed matter[3].

1.2 Biexciton

A biexciton is created from two free excitons in the condensed matter. In quantum information and computation, it is essential to construct coherent combinations of quantum states. We can use the basic quantum operations to perform the physically distinguishable quantum bits. Therefore, biexcitons can be illustrated by a simple four-level system.

In an optically driven system which is shown on Figure 1, the $|01\rangle$ and $|10\rangle$ states can be directly excited by $|00\rangle$ state. However, direct excitation of the upper $|11\rangle$ state from the ground state is usually forbidden. In order to get the biexciton state, the most efficient alternative is coherent nondegenerate two-photon excitation. In other words, it is used $|01\rangle$ or $|10\rangle$ state as an intermediate state.

$|01\rangle$ and $|10\rangle$ states are the cases of the linearly polarized (i.e. the symmetric and antisymmetric combinations of spin up and down) exciton states. Their energy is different due to the different combination of spin. Moreover, due to coulomb coupling, the biexciton state $|11\rangle$ state is combined with the two antiparallel-spin excitons[4].

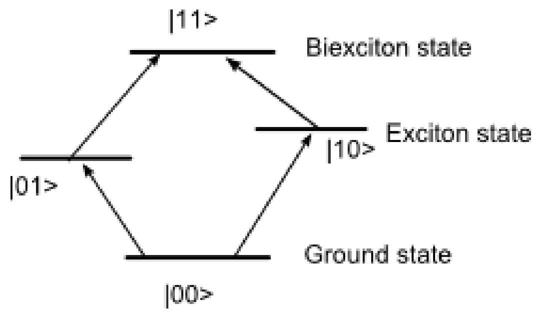

Figure 1: Model of single Quantum Dot. There are 4 states: ground state:$|00\rangle$, direct excited state: $|01\rangle$ and $|10\rangle$, upper excited state: $|11\rangle$. The selection rule is shown by the arrow.[5]

1.3 Quantum dot

Quantum dots are artificial atoms that can be custom designed for a variety of applications. Specifically, it is made of semiconductor materials which are small enough to exist quantum mechanical properties. However, quantum dots have some man-made nanoscale structures in which electrons can be confined in all 3 dimensions. Due to its structure, its electronic properties are intermediate between those of bulk semiconductors and of discrete molecules[6].

1.4 Photo-luminescence

Photoluminescence is the phenomenon of luminescence from direct photoexcitation the emitting species. Specially, its light emits from any form of matter after the absorption of photons (electromagnetic radiation). According to the quantum theory, this process can be treated as the transition from the high energy state to the low energy state. In the experiment measure, the absorption of photons is provided by the laser[7].

1.5 Colloidal core/shell nanocrystal

Colloidal core/shell nanocrystals(NCs) contain at least two semiconductor materials in an onionlike structure. The core nanocrystals have lot of basic optical properties, for example, their fluorescence wavelength, quantum yield and lifetime[8]. Particularly, wet-chemically synthesized colloidal core/shell nanocrystals will emit non classical light at room temperature, which makes them suitable for the application of the quantum optics[9].

2 Experimental measurement

M.Manceau etc [9] has already done the experiment of the CdSe/CdS core-shell dot-in-rods(DRs) synthesis and photoluminescence measurement in the lab. In their paper, they have measured the flickering between the bright state and grey state of the nanocrystal and showed that the autocorrelation function can be identified the exciton and biexciton state.

2.1 Exciton Experiment

In the experiment, a fast switching(flicking) between two states of emission(a bright state with a high emission efficiency and a grey state with a lower emission efficiency is shown by the Figure 2.

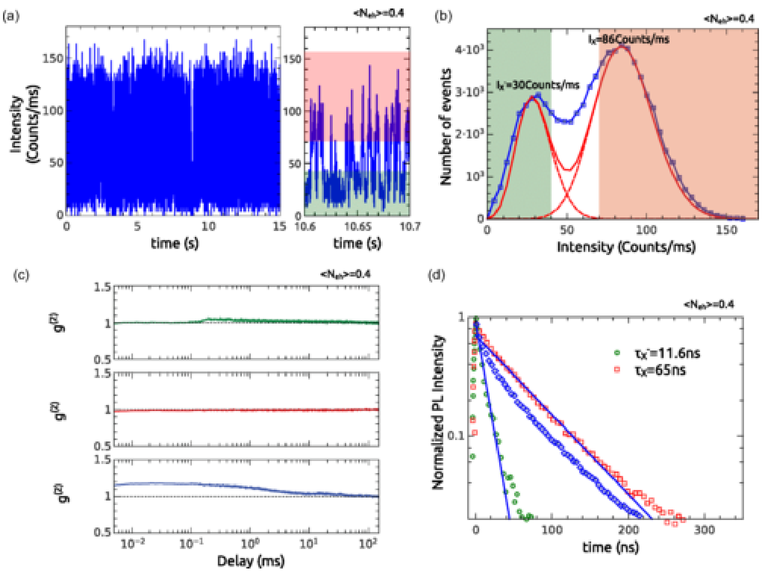

Figure 2: (a)PL intensity, (b)histogram of PL intensity, (c)normalized ACF and (d)PL decay in the dot-in-rod sample 1(DR1)[9].

2.1.1 PL intensity

In their experiment, the photoluminescence(PL) was collected by a confocal microscope with a high numerical aperture oil immersion objective. In the Figure 2a, the PL intensity is counted in second(x-axis) and the intensity counting is done with the bin time $250\mu s$. Specially, the zoom of the PL intensity Figure 2a can show that there will be two different types of light: red one belongs to the bright state and green one belongs to the grey state. However, the blank zone between two states are disorder, which implies that the mechanism between the switch of two states is not clear.

2.1.2 Histogram of PL intensity

In order to know the transition between the bright and grey states, histogram of the PL intensity will be used when fitting the data. In the Figure 2b, a two poissonian distribution fit is used to know the mean emission intensity of the grey/bright state. Two peaks appear in the histogram and the data histogram curve doesn't coincide the poissonian fitting curve.

2.1.3 PL decay

In the Figure 2d, the whole PL intensity will be separated into two curves(Bright state and grey state) depending on the intensity count number in Figure 2a.

2.1.4 Autocorrelation function(ACF)

As a fast switching between a bright and a grey mode with biexcitons, histogram of the PL intensity is not enough to describe the detail of the transition between two states. In order to characterize PL photons emission from the DRs, Time-Correlated Single Photon Counting(TCSPC) is used in the experiment, which is corresponding to the Figure 2c. In the experiment, due to the too fast flicking e.g. switching between two states is less than 100 ms, TCSPC can't detect two photons in the same time, which implies that the PL beam have to be separated into two beams.

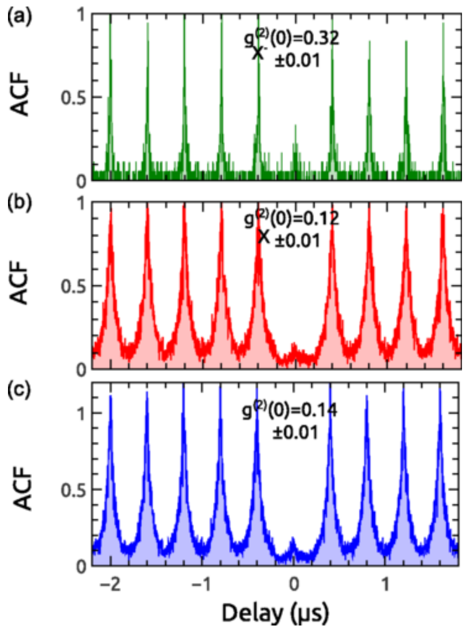

Figure 3: Normalized ACF for DR at short timescales. a. Grey state Normalized ACF. b. Bright state Normalized ACF. c. Whole PL normalized ACF.[9]

Autocorrelation function is used to know the detail of the photons inforamtion with two different time instants $t_1$and t_2 . It can be measured by the second order autocorrelation function of the electric field intensity which is related to the beam. It is defined as

$$g^{(2)}(t_1,t_2) = \frac{\langle E^*(t_1)E^*(t_2)E(t_1)E(t_2)\rangle}{\langle |E(t_1)|^2\rangle\langle |E(t_2)|^2\rangle} = \frac{\langle I_i(t_1)I_i(t_2)\rangle}{\langle I_i(t_1)\rangle\langle I_i(t_2)\rangle} \tag{1}$$

where<> represents the statistic average value and E is the electrostatic field and I is the emission intensity of each state.

In the experiment, autocorrelation function in the classical case is written as:

$$g^{(2)}(\tau) = \frac{\langle I_1(t)I_2(t-\tau)\rangle}{\langle I_1(t)\rangle\langle I_2(t-\tau)\rangle} \tag{2}$$

where τ is the delay time between two channels of TCSPC and $I_i(t)$ (i=1, 2)is the emission intensity of bright and grey state in the experiment.

In the quantum autocorrelation function, it will be:

$$g^{(2)}(\tau) = \frac{\langle S_1(t)S_2(t-\tau)\rangle}{\langle S_1(t)\rangle\langle S_2(t-\tau)\rangle} \tag{3}$$

where $S_i(t)$ is the time of each photon and τ is the delay time.

When we consider the delay between the photon with its neighbor photons, quantum autocorrelation will be used, for example, the normalized autocorrelation function with short time delay in Figure 3. When we consider the classical autocorrelation between the intensity event of the histogram, its classical autocorrelation function can be written function 2. Its figure is Figure 2c in several delays between the photons.

3 Simulation Model of photons with Excitons

In order to describe the mechanism of the transition between bright and grey state, the new model of photoluminescence(PL) will be simulated in this internship report. From the Figure 2, it is proved that there will be two states in the PL emission. In this model, there will be two different kinds of photons with two kinds of emission microtime, two kinds of time distributions of the photon, two probabilities of detection by different APD(avalanche photodiode) with two macrotimes.
Note: 1) Microtime is the time of certain PL photon emission after the corresponding laser photon exciting. Its unit is ns. 2) Macrotime is the time of certain PL photon emission after the first laser photon exciting the whole system. Its unit is μs.

3.1 Exponential statistics coefficient τ_A and τ_B

The time of PL photons emitted from the nanocrystal will follow the exponential statistics, which is shown by F.Pisanello[10]. In the semiconductor CdSe/CdS DRs, there exists two states(bright state and grey state), which implies that two different exponential statistics coefficient will fit the photon emission time.

The equations of the emission time for the bright state with exponential statistics coefficient τ_A and the grey state with another exponential statistics coefficient τ_B are shown below:

$$P(\delta t_A) = e^{-\frac{\delta t_A}{\tau_A}} \tag{4}$$

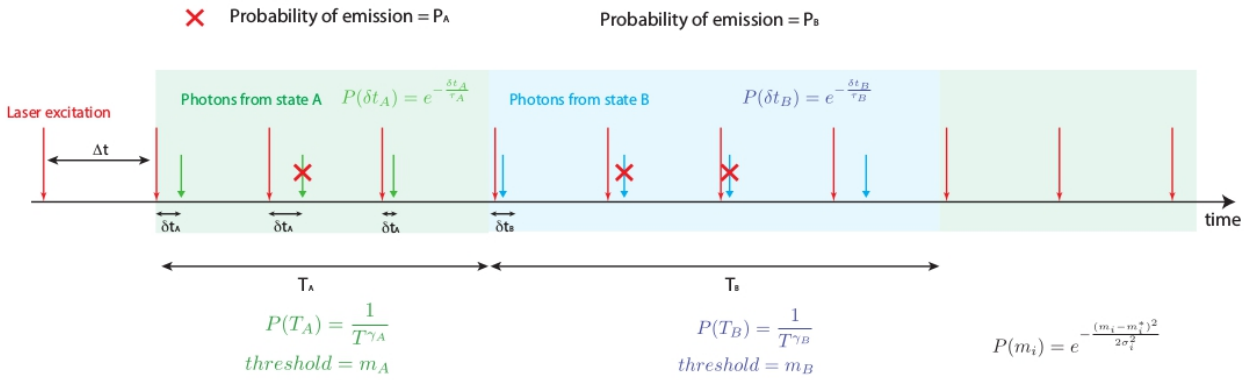

Figure 4: Exciton photon detection in microtime

$$P(\delta t_B) = e^{-\frac{\delta t_B}{\tau B}} \tag{5}$$

where $\delta t_{A/B}$ is the microtime of the PL photon and P is the PL emission time of each state.

3.2 Power law statistics coefficients γ_A, γ_B and their threshold values m_A, m_B

The switching of the emission photons between the grey mode and the bright mode will follow the power law statistics[11]. In the bright mode, γ_A is the power law coefficient and m_A is its threshold value. In the grey mode, γ_B represents the coefficient of the power law in the grey state and m_B is its corresponding threshold value.
Note: Threshold value means the minimum number of photons series in the power law statistics.

When considering the bright state with the power law coefficient γ_A and the grey state with the power law coefficient γ_B, their corresponding switch emission series are shown below:

$$P(T_A) = \frac{1}{T^{\gamma_A}} \tag{6}$$

$$P(T_B) = \frac{1}{T^{\gamma_B}} \tag{7}$$

where γ is the coefficient of the power law statistics and P is the PL emission time.

3.3 Normal distribution coefficients σ_A or σ_B in each state

When the power law statistics is considered as the transition between the bright and grey mode, its threshold value will have changed. In order to control the change of the threshold value, normal distribution will be considered. In this model, 2 kinds of normal distribution are designed to control the change of the threshold values in the corresponding power law statistics:

$$P(x) = e^{-\left(m_i - m_i^*\right)^2 / 2\sigma^2} \tag{8}$$

where m_i^*(i=A,B) is the mean value of the threshold in the power law distribution and the σ_i (i=A,B) is the standard deviation.

When the bright state is considered, its threshold value has mean value m_A^* and the normal distribution coefficient σ_A. The grey state has the similar normal distribution coefficient σ_B and mean value of threshold m_B^*.

3.4 Probabilities coefficients P_A and P_B

As the limitation of the avalanche photodiode, especially for double photons emitted by the biexciton excitation, some photons can't be detected. Therefore, the probabilities of the detection P_A and P_B will be added into the exciton simulation part. Specifically, P_A is the probability of the bright state photons which can be detected by (channel 1 of APD)APD1 and P_B is the probability of the grey state photons which can be detected by APD2(channel 2 of APD).

3.5 Precision of the detection

The model of photons with excitons has to be considered the measurement accuracy by the instruments. The photon emission time has smallest value, which is treated as the precision in this model.

3.6 Pulse time of the laser excitation $\triangle t$

In the real experiment, the laser with pulses will be used. The interval of each pulse of the laser will be treat as $\triangle t$.

3.7 Number of Photon

In this model, it will simulate from one to ten million photons system.

4 Model of Biexciton

As the structure of biexciton, sometimes the nanocrystal will emit two photons after a pulse laser excitation. Model of biexciton is designed to describe this case. In this model, four extra coefficients will be added into the simulation. Cconsidering the biexciton state of the bright state, an exponential statistics coefficient τ_1 and its corresponding probability P_1 will be utilized. They has the similar equation as the exciton part 3.1 and 3.4. An biexciton of the grey state also has its exponential statistics coefficient τ_2 and probability P_2.
Note: In the model of biexciton, the probabilities P_2, P_1 of the biexcition and the probability of exciton P_A, P_B are independent.

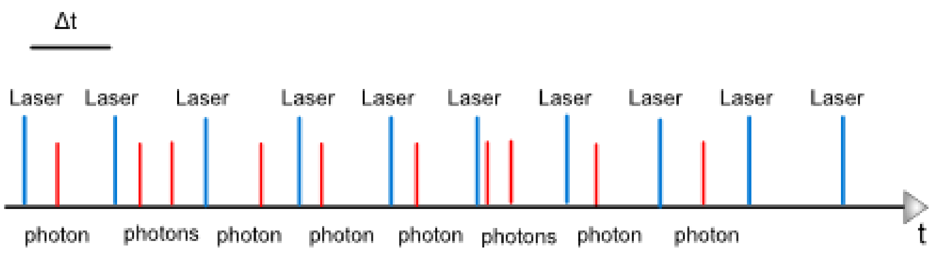

Figure 5: Biexciton photon detection in microtime

4.1 Exponential statistics coefficient τ_1 and τ_2 of biexciton

Biexciton sometimes will emit 2 photons when laser pumping the nanocrystal. The photon emission time of biexciton also follows the exponential statistics. Accurately, the photon emitted by the bright state biexciton has an exponential statistics coefficient τ_1 and the photon emitted by the grey state biexciton has an exponential statistics coefficient τ_2
In detail,

$$P(\delta t_1) = e^{-\frac{\delta t_1}{\tau 1}} \tag{9}$$

$$P(\delta t_2) = e^{-\frac{\delta t_2}{\tau 2}} \tag{10}$$

where $\delta t_{1/2}$ is the microtime of the PL photon and P is the PL emission time of each state.

4.2 Probabilities of photon emitted by biexciton in the detector

In the real experiment, not all photons will be detected by the APD. In order to improve the precision of the Biexciton Model, probabilities P_1 is considered the bright state and P_2 is for the grey state. Moreover, the bright state emission has only P_1 probability data from the APD1, the similar probability P_2 for the grey by APD2.

5 Simulation

In the simulation part, Matlab will be used as the programming Language. All the Matlab codes are in the appendix.

5.1 Simulation of exponential statistics

When simulating the PL time, the inverse function method is used to build the exponential statistics.
Original function:

$$f(x) = e^{-\frac{t}{\tau}} \tag{11}$$

Inverse function:

$$t = -\tau ln(f(x)) \tag{12}$$

where f(x) is the uniform distribution between 0 and 1.

5.2 Simulation of power law statistics

The power law statistics is also built through the inverse function.
Mathematical Method:
Original function:

$$P(T) = \frac{1}{T^{\gamma}} \tag{13}$$

where the equation has the minimum value 'm' of the PL emission time P.

$$x = m(1-r)^{\frac{-1}{\gamma}} \tag{14}$$

where m is the threshold value of the power law distribution, γ is the coefficient of the power law distribution and the r is the uniform distribution between 0 and 1[12].

5.3 Simulation of PL intensity figure

In the Matlab, Step to build the figure: 1) Use the histogram bin count order "histc" to count the number of values in each bin time that are within each specified bin range. The bin time can be different as it is set by the function. 2) Build the histogram through the order "plot".

5.4 Simulation of Quantum correlation function(ACF)

Step to build the figure of quantum correlation function: 1) Separate the photon emission time into two APDs with the same probability 50%. 2) Build two same size of emission time vector of the bright and the grey state. 3) Use the element translation and subtraction of the vector to get the delay of the each photon. 4) Calculate the ACF and plot the figure.

5.5 Simulation of Classical correlation function(ACF)

Step to build the figure of classical correlation function: 1) Calculate the PL intensity value as the section 5.3. 2) Build two same size of time vector of the intensity. 3) Use the element translation and subtraction to build the ACF function. 4) Calculate the ACF values with different delay time.

6 Result

Exciton

In this case, I simulate the photoluminescence(PL) emission excited by exciton from the DRs and compare the result with the experiment data in Figure 2.

Table 1: Simulation data of photon emitted by the exciton of dot-in-rod nanocrystal

τ_A	τ_B	P_A	P_B	γ_A	γ_B	m_A	m_B	σ_A	σ_B	step(ns)	$\triangle t(\mu s)$	num
60	10	0.4	0.13	2	2	5	3	10	16	0.4	512	10^6

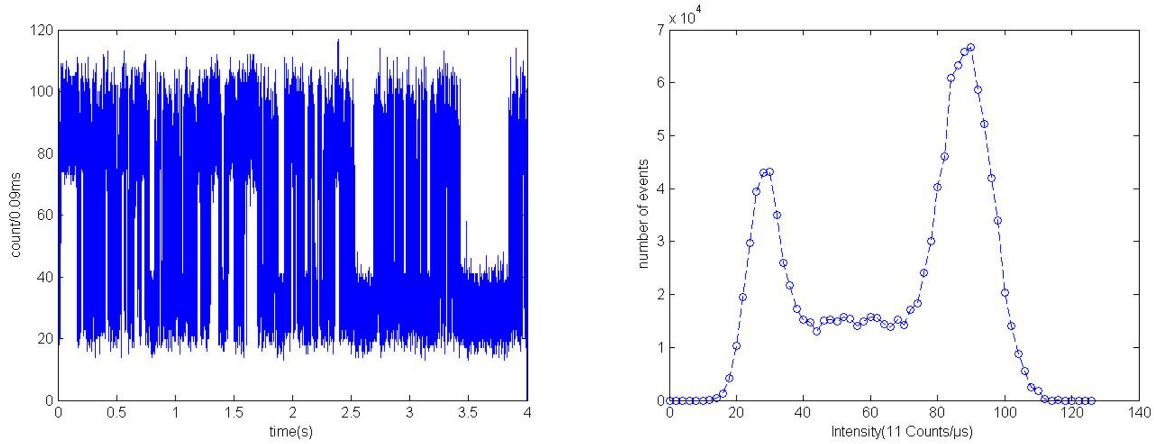

Figure 6: PL intensity of the DR (dot-in-rod) emission (left) and Histogram of PL intensity of the DR (right)

In these two figures(Figure 2a, 2b and Figure 6), there all exist two types of emission time photons. Compared with the position of peaks, both cases have two peaks at the almost same intensity(x-axis). However, the switch between the bright and grey state is different in the simulation part. Specifically, the number of event drops a little in the experiment figure, but the switch zone of two state has a concave plane in the simulation. The reason of the difference is that double photons (biexciton) sometimes will emit in the experiment. To some extent, the model of exciton can describe the mechanism of the transition.

In the Figure 2c, the autocorrelation function in the bottom increases a little at a short time scale and then decreases until the value of 1. In the simulation Figure 7, $g^{(2)}$ factor has decreased from about 1.3 to 1.1 at the large delay time. The most difference between 2 cases is in the range of 10^{-2}. When delay time is large, both cases $g^{(2)}$ factor will decrease to the value of near 1 finally.

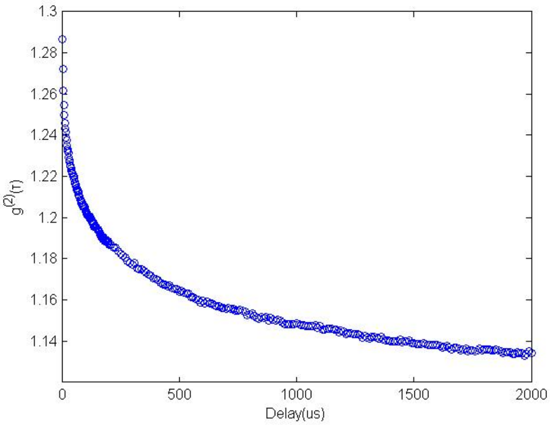

Figure 7: $g^{(2)}$ of the PL intensity emission from the excitons on the certain delay between photons

Biexciton

In Table 2, four extra terms will be added into simulation (τ_1 , τ_2 , P_1 and P_2). In this simulation, two photons will be emitted at the same time with their probabilities P_1 and P_2 due to the biexciton.

Table 2: Simulation data in the Biexciton model

τ_A	τ_B	P_A	P_B	τ_1	τ_2	P_1	P_2	γ_A	γ_B	m_A	m_B	σ_A	σ_B	step(ns)	$\triangle t(\mu s)$	num
60	10	0.08	0.5	2	2	0.02	0.02	2	2	25	1	5	2	512	0.4	10^7

Compared with the Figure 8a, The PL intensity Histogram in the simulation has the similar intensity distribution. The ratio between the bright state and the grey state is about 7.5. In the Figure 8a, the ratio between the bright and grey state is about 7.3. In the switching zone of two states, the simulating curve has better fit with the experiment curve in Figure 8a compared with the Model of exciton.

When we consider the classical correlation in the Figure 10(Right), we can observe the $g^{(2)}$ is about 1.4 at the short delay times and it will turn to 1.05 at a much larger delay time. In the quantum autocorrelation function, the $g^{(2)}(0)$ has ratio about 0.16 compared with the highest value of ACF in Figure 10(Left). On the whole, $g^{(2)}(0)$ in the simulation part fits well compared with the experiment measurement the Figure 8d.

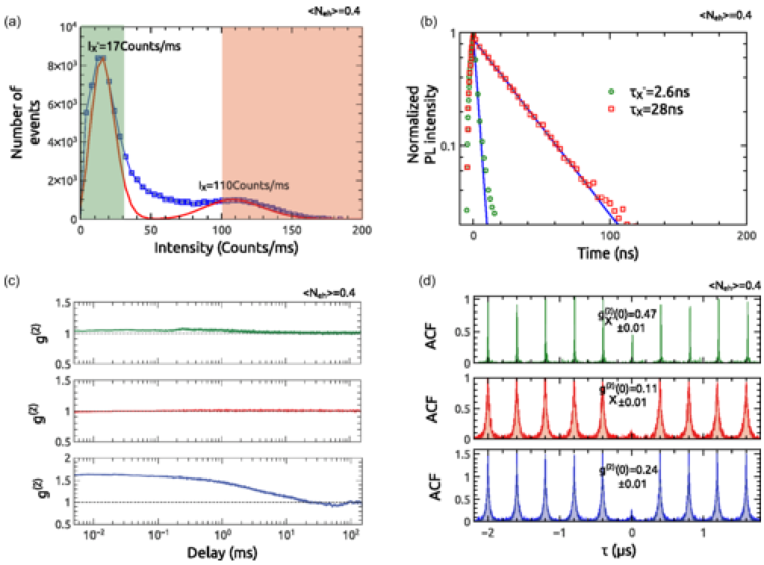

Figure 8: (a) Histogram of the PL intensity of DR2 whose Count rates below 30 counts/ms in green are associated with the grey state, while the part of the histogram above 100 counts/ms is attributed to the bright state. (b) PL decay curves for grey state photons (green circles) and bright state photons (red squares). (c) g(2) for DR2 on several decades of delays between the photons. From top to bottom: g(2) for the grey state photons, bright state photons together with the g(2) of the whole PL intensity. (d) ACF for DR2 at short time scales. From top to bottom: ACF for the grey state photons, bright state photons together with the ACF of the whole PL intensity[9].
Note: DR2 is the second type of dot-in-rod nanocrystal in the experiment

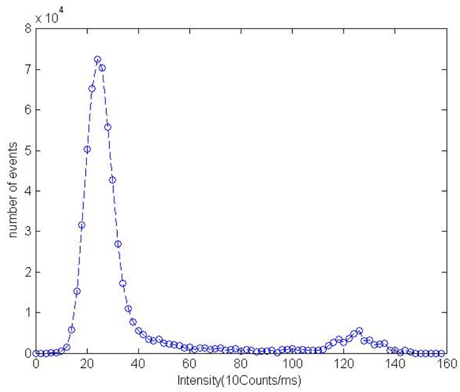

Figure 9: Histogram of the PL intensity in the biexciton simulation

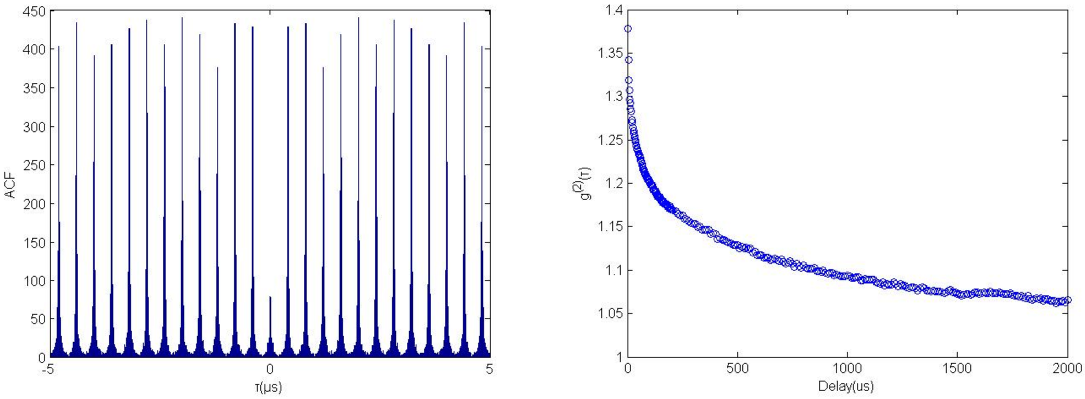

Figure 10: PL quantum autocorrelation for the DRs at short time scales(Left) and the classical correlation $g^{(2)}$ factor for the biexciton(Right)

6.1 Examples of the coefficient effect in the Model

Effect of the bintime in the histogram

Table 3: exciton in different bintime

τ_A	τ_B	P_A	P_B	γ_A	γ_B	m_A	m_B	σ_A	σ_B	step(ns)	$\triangle t(\mu s)$	num	bintime(μs)	label
60	10	0.4	0.13	2	2	1	3	10	6	0.4	512	10^6	100	1
60	10	0.4	0.13	2	2	1	3	10	6	0.4	512	10^6	300	2
60	10	0.4	0.13	2	2	1	3	10	6	0.4	512	10^6	500	3
60	10	0.4	0.13	2	2	1	3	10	6	0.4	512	10^6	700	4
60	10	0.4	0.13	2	2	1	3	10	6	0.4	512	10^6	1000	5

From Figure 11, it is easily to find that when the bin time increases, the peak will left move with the lower intensity peak. The transition area thebetween the grey and bright mode will appear a small peak when the bin time becomes larger. In order to know accurately the effect of the bin time, 3-D curve is plotted in Figure 12.

In Figure 12(Left), the tendency of the grey mode peak will increase with the increasing bin time, and the opposite intensity tendency for the bright mode. At the top view of the 3D curve(Figure 12(Right)), it is easy to find that two peaks will be separated increasingly with the growing bin time. The distribution of each peak will be broadened when the bin time increases.

Effect of the different probabilities in the biexciton emission time

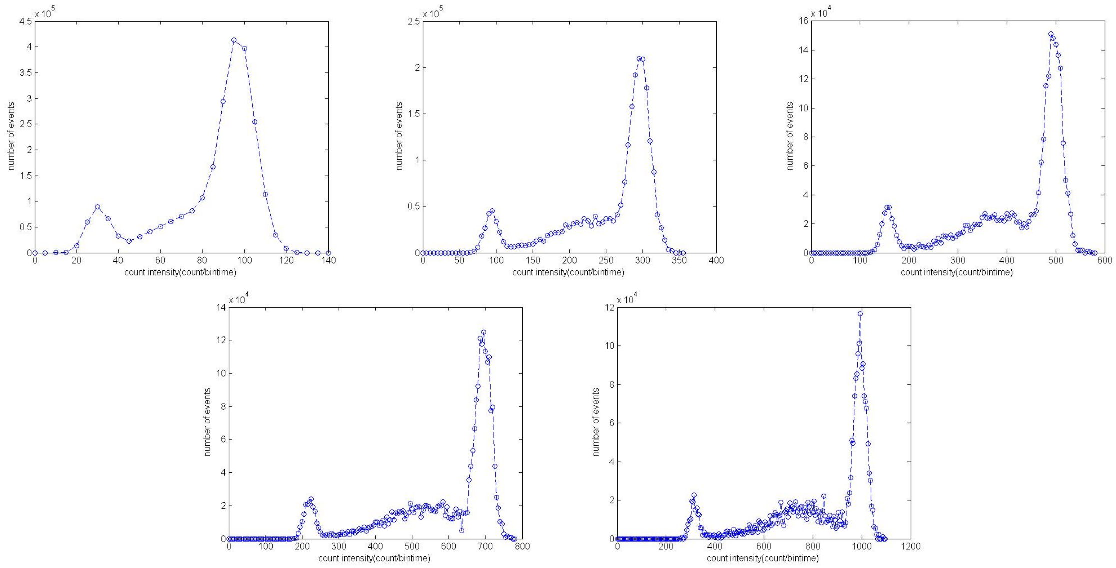

Figure 11: Biexciton PL intensity with different bin time.They are upper left $100\mu s$, upper right $500\mu s$, middle $300\mu s$, lower left 700 μs and lower right $1000\mu s$

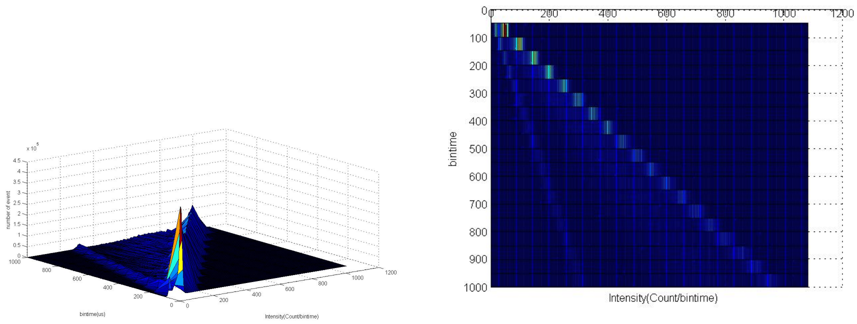

Figure 12: 3D plot of the exciton(Left) and Low view of the curve(Right)

Table 4: biexciton in different probabilities

τ_A	τ_B	P_A	P_B	τ_1	τ_2	P_1	P_2	γ_A	γ_B	m_A	m_B	σ_A	σ_B	step(ns)	$\triangle t(\mu s)$	num	label
60	10	0.13	0.4	1.9	2	0.05	0.05	2	2	5	2	15	2	512	0.4	10e7	1
60	10	0.13	0.4	1.9	2	0.2	0.2	2	2	5	2	15	2	512	0.4	10e7	2
60	10	0.13	0.4	1.9	2	0.3	0.3	2	2	5	2	15	2	512	0.4	10e7	3

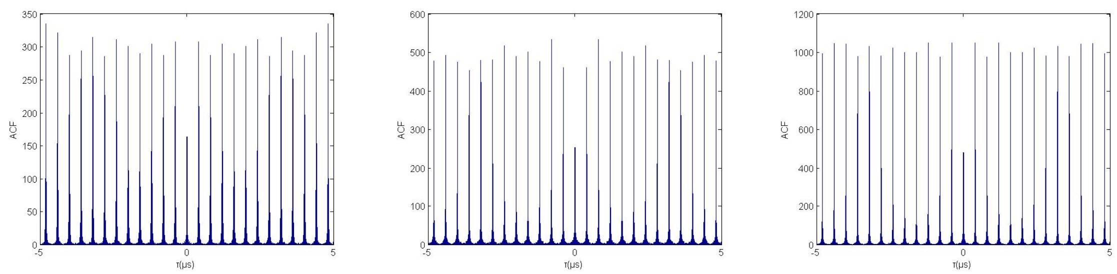

Figure 13: quantum correlation function with different probabilities in the biexciton emission.P_A and P_B for each figure: 0.05(Left), 0.2(Middle), 0.3(Right)

From the Figure 13, we can observe that $g^{(2)}(0)$ becomes larger with the increasing probability of the biexciton. When we increase the count density of the PL emission, the value of the $g^{(2)}(0)$ will change a little until the count intensity is very large. Moreover, $g^{(2)}(0)$ will trend a certain value at all cases. The probabilities of the biexciton will also change the trend of the $g^{(2)}$ in the non-zero delay.

Effect of the normal distribution factor in the biexciton emission time

In the switch of the two states, the normal distribution factor of the threshold value will have an influence on the distribution of the intensity for each state. From the Table 5, the distribution of the threshold value in state 1 is changed.

Table 5: biexciton in different normal distribution factor

τ_1	τ_2	P_1	P_2	τ_A	τ_B	P_A	P_B	γ_1	γ_2	m_1	m_2	σ_1	σ_2	step(ns)	$\triangle t(\mu s)$	num	label
60	10	0.13	0.4	1.9	2	0.05	0.05	2	2	5	2	15	2	512	0.4	10^7	1
60	10	0.13	0.4	1.9	2	0.05	0.05	2	2	5	2	10	2	512	0.4	10^7	2
60	10	0.13	0.4	1.9	2	0.05	0.05	2	2	5	2	5	2	512	0.4	10^7	3

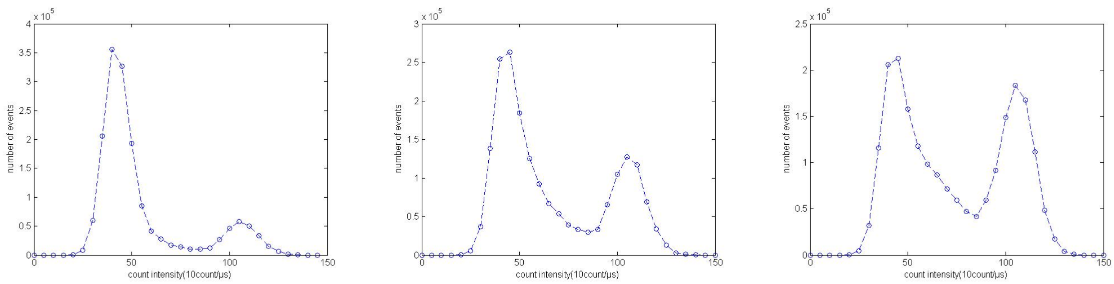

Figure 14: PL intensity with different threshold value distribution. Normal distribution factor σ_1 for each figure: 15(Left), 10(Middle), 5(Right)

From the Figure 14, we can find that the bright mode intensity increases when the normal distribution factor σ_1 decreases, which means that the normal distribution factor can affect the height of the intensity peak. And the position of two peaks can't be affected by the change of the normal distribution.

7 Conclusion

In this report, it is successfully to simulate the photoluminescence emission time of the bright and grey mode. The simulation models predict the different cases of the transition between the bright and grey mode. Through these cases, it can be found out the mechanism of the photoluminescence in the dot-in-rod nanocrystal.

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Appendices

Appendix A   Exciton Code

```
%input
%tau1 is the rate parameter of the exponential random statistics ...
    for the state A(bright mode). Its unit is ns.
%tau2 is the rate parameter of the exponential random statistics ...
    for the state B(grey mode). Its unit is ns.

%P1 is the probability of detection in state A
%P2 is the probability of detection in state B

%Gamma1 is the power law random number coefficient for state A
%Gamma2 is the power law random number coefficient for state B
%thre1 is the mean value of threshold in the state A power law ...
    statistics
%thre2 is the mean value of threshold in the state B power law ...
    statistics

%sigma11 is the normal distribution of the threshold for state A
%sigma12 is the normal distribution of the threshold for state B


%deltatime is the laser excitation time unit(us)
%step is the precision for each detector unit(ns)
%sizeOut is the total number of the photon

%output
%mac is the real photon emission time in us(macrotime)
%lmac is the label for each macrotime

%method: Inverse function of the distribution
% origional function:f(x)=exp(t/Tau)
% f(x) inverse function is t=-mu*ln(f(x)); and if f(x) distributes
% evenly,the t is the exponential random distribution .

% precision setting
% tau=step*number of step
% we can get the number of step first: mu.step=tau/step
% we get the intergal of step and then we multiply with the step ...
    and get
% the tau value whose minimum value is the step except

% state switch method: random vector for only 0 and 1 values
% we use power law statistics to bulid a random number of 0 and 1.


function [mac,lmac]= ...
    exciton(tau1,tau2,P1,P2,Gamma1,Gamma2,thre1,thre2,sigma1,sigma2,deltatime,step,si

% set the parameters use in the function
l1 = powerlaw(thre1,thre2,sigma1,sigma2,Gamma1,Gamma2,sizeOut);  % ...
    product random number between 0 and 1 for certain percentage of ...
```

```
    state 1
l2 = 1-l1;          % product random number between 0 and 1 for ...
    state 2

step=step/1000; %step in ps

mu1step=tau1/step; %get the number of step in tau 1
mu2step=tau2/step; %get the number of step in tau 2

stepval1=round(mu1step);
stepval2=round(mu2step);

b = rand(1,sizeOut);
b1 = round(stepval1.*(log(b)));
b2 = round(stepval2.*(log(b)));

r1 = -step.*b1;  %bulid the exponential random number for tau 1
r2 = -step.*b2;  %bulid the exponential random number for tau 2

%bulid the probability of emission
p1=rand(1,sizeOut);
p2=rand(1,sizeOut);

p1=p1<P1;
p2=p2<P2;

r1=r1.*p1;
r2=r2.*p2;


%bulid a mixing exponential random number
r1 = l1.* r1;  %select the exponential random number for tau 1
r2 = l2.* r2;  %select the exponential random number for tau 2

r = (r1+r2);  %combine all the selection random number in ns
l = l1+l2*2;  %find the corresponding states for each exponential ...
    random number

%macrotime random number in us
w=r>0;
mac=1:sizeOut;
mac=deltatime*mac+r/1000; %unit of mac is us

%delete the missing photon in the series
mac=w.*mac;
mac(mac==0)=[];
lmac=l.*w;
lmac(lmac==0)=[];

end
```

Appendix B Bieciton Code

```
%input
%tau1 is the rate parameter of the exponential random statistics ...
    for the state A exciton(bright mode). Its unit is ns.
%tau2 is the rate parameter of the exponential random statistics ...
    for the state B exciton(grey mode). Its unit is ns.

%P1 is the probability of detection in state A exciton
%P2 is the probability of detection in state B exciton

%tauA is the rate parameter of the exponential random statistics ...
    for the state A biexciton(bright mode). Its unit is ns.
%tauB is the rate parameter of the exponential random statistics ...
    for the state B biexciton(grey mode). Its unit is ns.

%PA is the probability of detection in state A biexciton
%PB is the probability of detection in state B biexciton

%Gamma1 is the power law random number coefficient for state A
%Gamma2 is the power law random number coefficient for state B
%thre1 is the mean value of threshold in the state A power law ...
    statistics
%thre2 is the mean value of threshold in the state B power law ...
    statistics

%sigmal1 is the normal distribution of the threshold for state A
%sigmal2 is the normal distribution of the threshold for state B


%deltatime is the laser excitation time unit(us)
%step is the precision for each detector unit(ns)
%sizeOut is the total number of the photon

%output
%mac is the real photon emission time in us(macrotime)
%lmac is the label for each macrotime
%mac1 is the real photon emission time series of state A
%mac2 is the real photon emission time series of state B

%method: Inverse function of the distribution
% origional function:f(x)=exp(t/Tau)
% f(x) inverse function is t=-mu*ln(f(x)); and if f(x) distributes
% evenly,the t is the exponential random distribution .

% precision setting
% tau=step*number of step
% we can get the number of step first: mu.step=tau/step
% we get the intergal of step and then we multiply with the step ...
    and get
% the tau value whose minimum value is the step except

% state switch method: random vector for only 0 and 1 values
% we use power law random number function to bulid a random number ...
    of 0 and 1.
```

```
% power law statistics is used following method:
% f(x)=threshold*(1-r)^(-1/(gamma-1)) where r is the random statistics
% between 0 and 1

function [mac,lmac,mac1,mac2]= ...
    biexciton(tau1,tau2,P1,P2,tauA,tauB,PA,PB,Gamma1,Gamma2,thre1,thre2,sigma1,sigma2

% set the parameters use in the function
l1 = powerlaw(thre1,thre2,sigma1,sigma2,Gamma1,Gamma2,sizeOut);  % ...
    product random number between 0 and 1 for certain percentage of ...
    state 1
l2 = 1-l1;           % product random number between 0 and 1 for ...
    state 2

step=step/1000; %step in ps

mu1step=tau1/step; %get the number of step in tau 1
mu2step=tau2/step; %get the number of step in tau 2
muAstep=tauA/step; %get the number of step in tau A
muBstep=tauB/step; %get the number of step in tau B


stepval1=round(mu1step);
stepval2=round(mu2step);
stepvalA=round(muAstep);
stepvalB=round(muBstep);

b = rand(1,sizeOut);
a = rand(1,sizeOut);
b1 = round(stepval1.*log(b));
b2 = round(stepval2.*log(b));
bA = round(stepvalA.*log(a));
bB = round(stepvalB.*log(a));


r1 = -step.*b1;  %bulid the exponential random number for tau 1
r2 = -step.*b2;  %bulid the exponential random number for tau 2
rA = -step.*bA;  %bulid the exponential random number for tau A
rB = -step.*bB;  %bulid the exponential random number for tau B

%bulid the probability of emission for biexciton time
p1=rand(1,sizeOut);
p2=rand(1,sizeOut);
p3=rand(1,sizeOut);
p4=rand(1,sizeOut);

% set the probability of detection
p1=p1<P1;
p2=p2<P2;
p3=p3<PA;
p4=p4<PB;

r1=r1.*p1;
r2=r2.*p2;
rA=rA.*p3;
rB=rB.*p4;
```

```
%bulid a mixing exponential random number

r1 = l1.* r1;  %select the exponential random number for tau 1
r2 = l2.* r2;  %select the exponential random number for tau 2

r = (r1+r2);  %combine all the selection random number in ns
l = l1+l2*2;  %find the corresponding states for each exponential ...
    random number

%bulid a excition mixing random number
rA = l1.* rA;  %select the exponential random number for tau A
rB = l2.* rB;  %select the exponential random number for tau B

rr = (rA+rB);  %combine all the selection random number in ns
ll = l1+l2*2;  %find the corresponding states for each exponential ...
    random number

%macrotime random number in us
w1=r>0;
macw=1:sizeOut;
mac=deltatime*macw+r/1000; %unit of mac is us

%bulid the biexcition
w2=rr>0;

mac1=deltatime*macw+rr/1000; %unit of mac1 is us
mac1=w2.*mac1;
lmac1=ll.*w2;

mac2=w1.*mac;
lmac2=l.*w1;

%delete the missing photon in the series
mac=[mac1;mac2];
mac=reshape(mac,1,[]);
mac(mac==0)=[];

lmac=[lmac1;lmac2];
lmac=reshape(lmac,1,[]);
lmac(lmac==0)=[];


end
```

Appendix C Power Law Statistics Code

```
%function pl is to bulid a random number of 0 and 1 which follows 2 ...
    powerlaw
%distribution series
%output l is the random number series of 0 and 1


function [series]=powerlaw(thre1,thre2,sigma1,sigma2,Gamma1,Gamma2,num)


%bulid 2 normal distribution for threshold values
thre11=normrnd(thre1,sigma1,1,num);
thre22=normrnd(thre2,sigma2,1,num);

%bulid 2 random number between 0 and 1
n1=rand(1,num);
n2=rand(1,num);
%bulid 2 power law random number series
rn1=thre11.*(1-n1).^(-1/(Gamma1-1));
rn2=thre22.*(1-n2).^(-1/(Gamma2-1));

%get the intergal and absolute series
rad1=abs(round(rn1));
rad2=abs(round(rn2));


%get a series to describe the number of subvector e.g bulid a ...
    series like[rad1(1),rad2(1),rad1(2),rad2(2),`````]
l = [rad1;rad2];
l = reshape(l,1,[]);

% delete the extra value in the vector to increase the running ...
    efficient
temp=cumsum(l);
n=sum(temp<num);
l(n+2:end)=[];

%limit the l vector values in order to

%%random number 0 and 1 series
%%e.g l=[1 2 3 4], then RR=[1 0 0 1 1 1 0 0 0 0]

R = arrayfun(@(x) mod(x,2)*ones(1,l(x)),1:length(l),'un',false);
series = cell2mat(R);
series(num+1:end)=[];

end
```

Appendix D Classical Correlation Code

```
%function classicalcorrelation is to bulid the classical ...
    correlation in
%exciton photon emission time series
%input
%mac is the emission time for exciton
%output
%g2 is the classical autocorrelation function

function [g2] = classicalcorrelation(mac)

%find the max value for mac
w=max(mac);

%bulid the time scale
l=0:2:w; %count in 2us for the hole photon time series

%count the intensity of every 2us
bin1=histc(mac,l);


g21=zeros(1,100);%bulid a 0 vector

%find the classical autocorrelation function with delay time in 2us.Its
%range is from 1 to 200us delay time.
for i=1:100

bin2=bin1;
bin3=bin1;
bin2(end-i+1:end)=[];%delete the area which is not calculated
bin3(1:i)=[];
g21(i)=mean(bin2.*bin3)/mean(bin2)/mean(bin3); %g2 ...
    =mean(I(t)*I(t+tau))/mean(I(t))/mean(I(t+tau))
end

%bulid another 0 vector
g22=zeros(1,81);
l=1;
%count the autocorrelation with 10us. Its range is from 200us to
%2000us(2ms)
for j=100:5:1000
bin2=bin1;
bin3=bin1;
bin2(end-j+1:end)=[];
bin3(1:j)=[];
g22(l)=mean(bin2.*bin3)/mean(bin2)/mean(bin3); %g2 ...
    =mean(I(t)*I(t+tau))/mean(I(t))/mean(I(t+tau))
l=l+1;
end


%combine 2 vectors to build a whole classical correlation time ...
    vector for
%the histogram y-axis
```

```
g2=[g21,g22];
%build the delay histogram x-axis
i=(1:100)*2;
j=(100:5:1000)*2;
k=[i,j];
%plot the figure of the classical autocorrelation  function
figure(1)
plot(k,g2,'o');
xlabel('Delay(us)')
ylabel('g(2)')

end
```

Appendix E   Quantum Correlation Code

```
%function quantumcorrelation is to bulid the histogram of quantum ...
    autocorrelation
%input
%mac1 is the photon emission time series of the state A
%mac2 is the photon emission time series of the state B
%output
%output is the autocorrelation value between APD1 and APD2 ...
    detection photon
%delay time

function [output]=quantumcorrelation(mac1,mac2)

%bulid 1 and 2 value random vector the probability of each state is 50%
len=rand(1,length(mac1));
len1=len>0.5;
len2=1-len1;

%bulid the detection data for each APD(2 cases)
w11=len1.*mac1; %detection for APD1 exciton of the bright mode
w12=len2.*mac2; %detection for APD2 biexciton of the grey mode

w1=[w11;w12]; %combine the detection
w1=reshape(w1,1,[]); %delete the undetection photon emission time
w1(w1==0)=[];

w21=len1.*mac2; %detection for APD1 exciton of the grey mode
w22=len2.*mac1; %detection for APD2 biexction of the bright mode

w2=[w21;w22];
w2=reshape(w2,1,[]);
w2(w2==0)=[];

%make 2 vector same shape in order to use the vector calculation
if length(w1)>length(w2)
    w1(length(w2)+1:end)=[];
else
    w2(length(w1)+1:end)=[];
end
%bulid a blanck vector
output=[];
%precions: 21 elements around delay time

for k=-10:1:10
%method: element shift for one vector and use another vector minus ...
    it to
%get the delay time

w3=w2(mod((1:end)-k-1,end)+1)-w1;
w4=w1(mod((1:end)-k-1,end)+1)-w2;
output=[output,w3,w4];
end
%make the matrix as 1*n vector
output=reshape(output,1,[]);
```

```
output(abs(output)>5)=[];
end
```

Appendix F   The Code Of PL Curve

```
%mac is the series of photon emission time in us
%l is the counting number per (bintime)us
%rangecount is the range of counting in s
%k1 is the intensity count(counts/bintime(us))
%numevent is the number of event


function [numevent,rangecount]=plotcurve(mac,bintime)


%find the max value for num
w=max(mac);
%bulid the time scale
%set the bin time
l=0:bintime:w; %count in us

%count the intensity
rangecount=histc(mac,l);

l1=l/1000000;%x axis in s
%plot the pl intensity on seconds
figure(1)
plot(l1,rangecount);
xlabel('time(s)')
ylabel('count/bintime(us)')


%intensity count in unit 5
k1=0:5:max(rangecount)+10;
%number of events
numevent=zeros(1,length(k1));

for w=1:1:length(k1)
    numevent(w)=sum((rangecount>5*(w-1)&rangecount<w*5).*rangecount);
end
%plot the intensity and the number of event figure
figure(2)
plot(k1,numevent,'--o');
xlabel('count intensity(count/bintime)');
ylabel('number of events')


end
```

Appendix G The Change Of Bin Time

```
%bulid the 3d curve of the intensity count with different bintime
%inensitycount is the intensity count vector
%bin is the bin time vector
%x is the number of the counting for the certain count intensity
%num is the input photon emission time
function [intensitycount,bin,x]= bintime(num)
%bulid a blank vector for the number of counting
x=[];


%bulid the matrix of the intensity and count number with different ... bin time
for bintime=100:100:1000

%find the max value for num
w=max(num);
%bulid the time scale
%set the bin time
l=0:bintime:w; %count in us

%count the intensity
k=histc(num,l);


%find the max value of the count intensity
l1=0:1000:w;
intensitycount=histc(num,l1);


%intensity count in unit 5
intensitycount=0:5:max(intensitycount)+10;
%number of events vector
su=zeros(1,length(intensitycount));

for w=1:1:length(intensitycount)
    su(w)=sum((k>5*(w-1)&k<w*5).*k);
end

%put each numbers of events into a matrix
x =[x;su];
end

%plot 3d curve
bin=100:100:1000;

surf(intensitycount,bin,x);
xlabel('Intensity(Count/bintime)');
ylabel('bintime')
zlabel('number of event')

end
```