Lab3-4: Single Photon Source

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ABSTRACT

In this lab, we studied the quantum dot excitation method of single photon source. First, the necessity and principle of making single photon emitter are introduced. Then we prepared quantum dot samples in index matching oil and liquid crystal, and observed blinking fluorescence emission from quantum dots with a CCD camera. In a confocal microscope setup with two APDs, we recorded the image of quantum dots fluorescence. By a Hanbury Brown and Twiss setup, we demonstrated antibunching from a histogram of photon separation times and observed no photons arrive when the delay time is zero.

Keywords: single photon source, Hanbury Brown and Twiss setup, antibunching

1. INTRODUCTION

1.1 Single photon source

Heisenberg uncertainty fundamental laws show that every quantum measurement significantly influences the observed system. This feature verifies the security communication of quantum cryptography. One of the most important parts of the quantum communication is single photon source (SPS). Though the quantum communication has a large potential market, its development is held back because the difficulties to get SPS.

For quantum communication, we need quantum SPS, which is different from the attenuated classical photon source. Because when we attenuate the class photon source, we may get single photon at some times, but it is possible that at some other times, there are two or more photons at one time. While, the quantum SPS should be one photon one time. There are three characters of a photon source as a reliable quantum photon source. First, the single photon source emits only one photon one time; second, the source is synchronized to an external clock; third, the polarization of the single photons can be well-defined. Currently, there are three approaches for room temperature single photon source: single molecules, colloidal semiconductor quantum dots (nanocrystals) and color centers in diamond. The drawback of using the color centers in diamond is that the polarizations of photons are random. In our lab, we use the quantum dots as single photon source.

Quantum dots are often referred to as “artificial atoms”. There electron motion is quantized in all three spatial directions, resulting in a discrete energy-level spectrum as that of an atom. This kind of single photon source is different from the single photon source by attenuating a classic light source. To get quantum single photon source, the critical issue is low concentration of photon emitters. In this situation, only one emitter gets excited as shown in Figure 1 (Fig. 1), and emits only one photon at a time. The former is decided by the density of emitters and the diameter of focus area of laser beam, and the later is decided by character of emitter.

Figure (Fig.) 1 Excitation of a single emitter by a focused laser beam

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In our experiment, the concentration of the quantum dots is about 1 nano Mol/Liter. And we use an oil immerse lens to get smaller laser diameter and increase the resolution. Oil immersion is a technique used to increase the resolution of a microscope. Another critical factor for single photon source is fluorescence lifetime. The triple energy level of a single atom is showed in Fig. 2. S₀ is the ground state, S₁, S₂ are excited energy levels. Within each state, there are vibration energy states. After the electrons are excited from ground state to the excited states, the electrons will return to the ground state. The typical emission rates are on the order of \(10^8 - 10^9\) s\(^{-1}\). Reciprocal of the emission is named as fluorescence lifetime, which is in the order of 10ns. Within each lifetime, a single photon emitter will only emit one photon. So it is possible to get only one photon one time.

![Fig. 2 Sketch of energy diagram of a single atom](image)

### 1.2 Antibunching

According to Grangier, Roger and Aspect, “a single photon can only be detected once”. They performed an experiment to examine correlations between photodetections at the transmission and reflection outputs of a 50/50 beam splitter [1]. The first experiment to examine correlations of photons was carried out by Hanbury Brown and Twiss [2]. They found a positive correlation between the continuous currents from photomultiplier tubes. Even though, they development a new criterion for single photon source.

![Fig. 3 Coincidence measurement](image)

As shown in Fig. 3, the correlation between the intensities of the transmitted beams \(I_T\) and the reflected beams \(I_R\) are given by the degree of second-order (temporal) correlation,

\[
g^{(2)}_{T,R}(\tau) = \frac{\langle I_T(t + \tau)I_R(t) \rangle}{\langle I_T(t)I_R(t) \rangle} \tag{1}
\]

In our experiment, the light source is stationary and the time delay between \(I_R\) and \(I_T\) is 0 (simultaneous), so equation (1) can be written as:

\[
g^{(2)}_{T,R}(0) = \frac{\langle I_T(t) \rangle^2}{\langle I_T(t) \rangle^2} = g^{(2)}(0) \tag{2}
\]
Because $[\langle I_j(t) \rangle]^2 \leq \langle |I_j(t)|^2 \rangle$, 

$$g^{(2)}_{T,R} = g^{(2)}(0) \geq 1$$  \hspace{1cm} (3)

This is the case in classic field, which is assigned as “bunched”. Because the photons tend to come in bunches. Once they strike at a beam splitter, some will be reflected and others will be transmitted. In the fully quantum theory, the correlations between the output fields from the beam splitter are described by the degree of second-order coherence in the form of quantum mechanical operators as:

$$g^{(2)}_{T,R}(0) = \frac{\langle \hat{n}_T \hat{n}_R \rangle}{\langle \hat{n}_T \rangle \langle \hat{n}_R \rangle}$$  \hspace{1cm} (4)

Here $\hat{n}$ is the intensity operator, and $\hat{n} = \hat{a}^* \hat{a}$, so

$$g^{(2)}_{T,R}(0) = \frac{\langle \hat{a}_T^* \hat{a}_R \hat{a}_R^* \hat{a}_T \rangle}{\langle \hat{a}_T^* \hat{a}_T \rangle \langle \hat{a}_R^* \hat{a}_R \rangle}$$  \hspace{1cm} (5)

$\hat{a}^*$ is the creation operator, which lowers the number of particles in a given state by one. And $\hat{a}$ is the annihilation operator, which lowers the number of particles in a given state by one. For a 50/50 beam splitter, and one particular choice of phase of the beam splitter, (4) becomes to

$$g^{(2)}_{T,R}(0) = \frac{\langle \hat{n}_i (\hat{n}_i - 1) \rangle}{\langle \hat{n}_i \rangle^2} = g^{(2)}_{I,I}(0) = g^{(2)}(0)$$  \hspace{1cm} (6)

In a single photon source, $\hat{n} |1 \rangle = 1$, so $g^{(2)}(0) = 0$. The first experiments demonstrated the existence of nonclassical fields was performed by Kimble, Dagenais and Mandel in 1977 [3]. They got $g^{(2)}(0) = 0.4$, which suggested an antibunched field. In these antibunching fields, the individual photons strike the beam splitter, either transmit or reflect.

### 1.3 Cholesteric liquid crystal 1-D- photonic band-gap materials

Cholesteric liquid crystal is a type of liquid crystal with a helical structure. The axis direction of layers in cholesteric liquid crystals is periodical arranged. The period of the variation is denoted as pitch, over which a full rotation of 360° is completed, as shown in Fig. 4.

![Fig. 4 Sketch of cholesteric liquid periodical structure and transmission and reflection within bandgap](image)

Cholesteric liquid crystal has selective 100% reflection within a band centered at $\lambda_o = n_{av} P_o$ towards circularly polarized light with opposite electric-field vector-rotation to the rotation of molecules in the helical structure. Here, $n_{av} = (n_e + n_o)/2$, and the band width is $\Delta \lambda = \lambda_o (n_e - n_o) / n_{av}$. Cholesteric liquid crystal can also be assumed as 1-D photonic crystal. When emitters are put in the liquid crystal, it feels the periodical structures. If the emission of emit-
ters is within the band gap of the liquid crystal, it will be suppressed. While at the band gap edge, the emission will be enhanced.

2. EXPERIMENTAL SETUP, PROCEDURE AND RESULTS

2.1 Experimental setup

The experimental setup is showed in Fig. 5, and can be divided into two parts: the optical part and the electrical part. The optical part is made up of laser source, confocal fluorescence microscopy and detector. A diode-pumped solid-state laser is used in this laboratory, with 532-nm wavelength, 6 ps pulse duration, 76 MHz pulse repetition rate and 40 mW average output power. A spatial filter is used to clean and widen the beam, and make the beam circular polarized. Another three mirrors are used for calibration. Two more mirrors are used to raise and redirect the beam. Then the beam will pass a filter holder which is used to contain neutral density filters. These filters are used to adjust the laser intensity during the experiment.

![Schematic diagram of experiment setup.](image)

The second part of the optical part of the experiment setup is a confocal fluorescence microscopy. Cofocal fluorescent microscopy is applied in many research areas. The specialty of the confocal detection is that the light not originating from the focal plane will not pass through the pinhole before the detector and hence cannot be detected, as showed in Fig. 6. In fact, the confocal microscopy is not necessary for single photon source. Here, we used it for convenience of imaging and scanning. In our setup, the diameter of the APD’s is very small (150um²), and can be used as pinhole. Usually, between the objective lens and the object, index match oil is used to increase the numerical apertures of objective lens. The purpose of using of confocal fluorescent microscopy is for the convenience of real time imaging by a CCD camera, for increasing resolution and brightness. After passing through the filters, the laser beam get into the confocal fluorescence microscopy system. A dichroic mirror is used to reflect the 532nm and transmit the fluorescence beam, which are not included in this diagram. An orange glass and interference filter are used to eliminate remaining 532nm light. Then the fluorescence light can be directed towards the detection part.

The detection part is made up of electro multiplying (EM) cooled CCD camera and two single photon counting avalanche photodiode modules (APD’s) for different purposes. Due to the sensitivity of APD and EM-CCD, the whole experiments were conducted in the dark. The EM-CCD is used to align the system and take real time images of fluorescence from emitters. The 50/50 beam splitter and the two APD’s is the optical part of a Hanbury Brown and Twiss setup for antibunching measurements. The beam splitter directs half of the incident photons to APD1 and the other half to APD2. The signal from APD1 is assigned as “start”, and signal from APD2, which is on a delay, is used to provide “stop” signal. Measuring the time delay between “start” and “stop” signals, a histogram of time delay between two pho-
tons and the coincidence count can be formed. Here the coincidence count is defined as number of second photons which appeared at a definite time interval (resolution) after the first photons.

Fig. 6 Schematic diagram of confocal microscope.

The electrical part of the experiment has two parts. For single-emitter fluorescent imaging, the signals from APD’s are connected to a computer with two cards (counter/timer and controller board) and Labview software. The software also controls a nano driver to run a piezo-translation stage which holds the sample, to create images of emitters and analyze the data. For antibunching, the APD’s are also connected to a Timeharp 200 time–correlated single photon counting PCI card. As shown in Fig. 5, the coincidence count is recorded by this card. Using the so-called “start”- “stop” electronics, a histogram of how many second photons will appear at the definite time interval after the first photons in pairs will be built in the computer screen, which is used to judge whether there is antibunching or not.

2.2 Experiment Procedure and Results

A. Preparation of Samples

Two types of samples were used in our experiments. They are colloidal and CdSeTe quantum dots. Preparation of the quantum dots are listed below:

1) Colloidal quantum dots in a 1D photonic bandgap cholesteric liquid crystal: use a capillary pipette to drop a small drop of quantum dots on a Corning No.1 microscope cover glass slip, then applied another capillary pipette to drop some liquid crystal, and mixed the quantum dots and liquid crystal for at least 5 minutes. Cover another glass slip on the former glass slip, and shift slightly in one direction to make the liquid crystal self-assemble to form 1D photonic bandgap structure. Within the bandgap of the structure, propagation of light is forbidden. So if the spontaneous emission is in the bandgap, it will be suppressed. However, the spontaneous emission will be enhanced near the band edge. This is similar to the cavity in laser: the bandgap acts as 100% reflecting mirror, and the band edge acts as partly transmitted mirror. The concentration of the quantum dots used is very important for obtaining single photon source. And the dosage of the liquid crystal also should be carefully looked out. If adopt too much liquid crystal, liquid will flow between the slips, which is not good for observation. We made two of such samples as Sample 1 and Sample 2.

2) CdSeTe quantum dots by spin coating: a Corning No.1 microscopic cover glass slip was mounted in a vacuum chuck, and the rotation speed was turned to about 3000 rpm. The solution with proper concentration was dropped on the glass slip when it was spinning and lasted about 40s until it was dry. This sample was assigned as Sample 3.
Fig. 7 Images of photon emitting quantum dots from (a) Sample 1 (b) Sample 2 and (c) Sample 3.

A. Single emitters

(1) Mount the sample. Before the experiment, we need to mount a sample on the sample holder of the confocal microscope. First, use methanol to clean the lens and the sample holder, then drop a small oil drop on the objective aperture, and then put the sample on the sample holder, two magnets are used to hold the sample tight.

(2) Put the output light to EM-CCD port and focus the light on the sample. As shown in Fig. 7, when the CCD is well focused, in the screen we can see quantum dots emitting light. Due to blinking of quantum dots, some emitters are blinking. Blinking is a drawback for stable single photon source. Now, the non-blinking quantum dots are available [4], which is better single photon source.

(3) Find the single emitters. After well adjust focusing, we should turn off the laser, and then turn on the APD’s put many filters in the filter hold, to avoid too much light get into the APD’s and potential damage, then change the output port to APD’s. Before scanning, make sure the electrical part is working. The Labview software is used to record the image of raster scanning. The panel of the software is showed in Fig. 8. On the right, two images come from two APD’s. The vertical and horizontal axes are pixels. The left bottom image is the photon counting of the current scanning line. The upper left is panel used to select scan area and control the nano drive. At first, the scan area was set to 25umx25um, and then we reduced the scan area by selecting specific area to eliminate the obvious affection from cluster emitters. As shown in Fig. 8, we did a 3.5 μm × 3.5 μm scanning.

Fig. 8 Labview panel for scanning
Fig. 9 Colloidal Quantum Dots in cholesteric photonic bandgap liquid crystal host (a) 3.5x3.5 um scanning, (b) Photon counting at spot indicated in (a) by a cross.

From this image of Fig. 9 (a), we do not only get the information in space, but also in time. Look at the spots indicated by arrows, we can see some strips. Due to raster scanning, the sample is scanned line by line. The velocity of the scanner is \( v \) nm/s, which means every line takes \( \ell / v \) second, \( \ell \) is the length in x direction. At one time when the quantum dots release photons, it is recorded by detectors. At another time, when the detector comes back, maybe the quantum dots don’t release photons due to blinking. If there are many emitters, it is not easy to observe this strips and blinking because of average effects. So observing the strips area is a convenient way to find single emitters.

(4) Antibunching. The software allows selecting a particular emitter and records the photon counting according to time. As shown in Fig. 9 (b), it can be seen that the photon counting is changing with time because of blinking. But observation of blinking can’t guarantee that the emitter is single photon source. In fact, in Sample 1 and Sample 2, we observed blinking but no antibunching, mainly because that in the process of fabrication, the quantum dots may not be mixed well enough with the liquid crystal and maybe too many quantum dots has been added to the sample.

Sample 3 was prepared by spin coating, which uses centrifugal force to apply uniform thin films on flat substrates. We did scanning in 12.5 \( \mu \text{m} \times 12.5 \mu \text{m} \) area, as shown in Fig. 10. In Fig. 10, the confocal microscopy was a little out of focus. In Fig. 11, we refocused, and the image is much clearer than Fig 10. Focusing is very important. It helps to locate the positions of emitters easier.

We focused on five spots on the sample to do single photon counting, and the spots are shown in Fig. 11 as indicated by the circle and number. The photon counting correlation to time is shown in Fig. 12 (a) and (b), the green and red curves are signals from two APD’s. As shown in Fig. 12 (a), the blinking of single photon emitter is very obvious. From time 90000ms to 110000ms, it stopped, and no photon at all, later it emitted again. The possibility of spot 2 is single photon emitter is very large. And in Fig. 12 (b), we can see blinking, but there is no totally stop of emission, so there are maybe two or more emitters. At the same time, the Time-Harp software is recording the temporal relationship of the APD’s, and the relationship of photon counting at specific spots and time interval are recorded in the Time-Harp soft-
ware as shown in Fig. 13.

Fig. 11 Refocus of objective lens and scan in area 12.5 \( \mu m \times 12.5 \mu m 

Fig. 12 (a) Blinking of quantum dots of spot 2 (b) Blinking of quantum dots of spot 3
Fig. 13 Antibunching of CdSeTe QDs (The white curve is fitting curve, and the black curve is experimental data.)
To decide how photons do we have for each emitter, first, we normalized the photon counting. Then, fit the antibunching curves according to equation

\[ \text{Normalized photon counting} = 1 - \frac{1}{N} \exp\left(\frac{-\Delta t}{\tau}\right) \]  

(1)

we can decide how many emitters in each spot. Here \( N \) is the number of emitters, and \( \tau \) is the fluorescence lifetime, and \( \Delta t \) is the time interval of photons from APD1 and APD2.

Table 1. Parameters of fitting of antibunching curves.

<table>
<thead>
<tr>
<th>Point</th>
<th>( N )</th>
<th>Err. ( N )</th>
<th>( \tau ) (ns)</th>
<th>Err. ( \tau ) (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.500083 ± 0.01092</td>
<td></td>
<td>70.90803</td>
<td>± 1.61769</td>
</tr>
<tr>
<td>2</td>
<td>1.068239 ± 0.00495</td>
<td></td>
<td>56.21986</td>
<td>± 0.61649</td>
</tr>
<tr>
<td>3</td>
<td>1.637841 ± 0.01081</td>
<td></td>
<td>117.7176</td>
<td>± 3.28286</td>
</tr>
<tr>
<td>4</td>
<td>2.146844 ± 0.01702</td>
<td></td>
<td>167.89746</td>
<td>± 7.721</td>
</tr>
<tr>
<td>5</td>
<td>2.131742 ± 0.01664</td>
<td></td>
<td>108.68102</td>
<td>± 3.30924</td>
</tr>
</tbody>
</table>

From Table 1, it can be seen that spot 2 is a single photon emitter. Its antibunching curve in Fig. 13 (2) also proves this. The minimum of the second order correlation reaches zero. Compared with Fig. 11, we found that there is no direct relationship with brightness of the emitter and single photon source. Though spot 3 is much brighter than spot 1, they have almost the same number of emitters. It can also see that the fluorescence lifetime of quantum dots is very sensitive to the environment. In spot 2, there is only one emitter, and it has the shortest fluorescence lifetime. As the increasing of emitters, the fluorescence lifetime increase.

B. Fluorescence Lifetime

The fluorescence lifetime refers to the average time the molecule stays in its excited state before emitting a photon. Due to this delay of radiation and excitation, we can make sure that the single photon emits one photon one time. If the concentration of excited state at time \( t \) is \([S_2]_t\), and the initial concentration is \([S_2]_0\), then the relationship of \([S_2]_t\) and \([S_2]_0\) will be:

\[ [S_2]_t = [S_2]_0 e^{-\frac{t}{\tau}} \]  

(2)

Here \( \tau \) is the fluorescence lifetime. Here we measured the fluorescence life time of DiI dye solution in toluene. The sample is DiI dye solution in toluene with a concentration of \( 10^6 \) Mol/Liter, and it was prepared by spin coating. Due to the large concentration of the solution, we can see lots of clusters in the scan image as shown in Fig. 14.
Fig. 15 Fluorescence lifetime of DiI dye molecules with fitted curve (a) spot 1 (b) spot 2 (c) spot 3 (d) spot 4

To measure the fluorescence lifetime, we should disconnect one of the APD, and connect the electrical output of the laser to the variable delay card. Change the delay between the electrical pulse and fluorescence signal from another APD, and select interested area from the Labview software and control the nano drive go the position, run the Time-Harp software, we can get the curves show the decay of fluorescence as in Fig. 15.

The curves are fitted according to

\[ \text{Photoncounting} = A \exp(-\Delta t / \tau) \]  \hspace{1cm} (3)

Here \( A \) is affected by laser intensity, \( \Delta t \) is the time interval between laser pulse and fluorescence, and \( \tau \) is the fluorescence lifetime.

Table 2 Fluorescence lifetime of DiI dye molecules of different environments.

<table>
<thead>
<tr>
<th>Spot</th>
<th>A</th>
<th>Err. A</th>
<th>( \tau ) (ns)</th>
<th>( \Delta \tau ) (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3081.09266</td>
<td>27.01376</td>
<td>2.67953</td>
<td>0.01824</td>
</tr>
<tr>
<td>2</td>
<td>2625.69581</td>
<td>24.35865</td>
<td>2.7233</td>
<td>0.01981</td>
</tr>
<tr>
<td>3</td>
<td>2779.4804</td>
<td>21.36147</td>
<td>2.66492</td>
<td>0.01587</td>
</tr>
<tr>
<td>4</td>
<td>2824.2396</td>
<td>24.36072</td>
<td>2.67338</td>
<td>0.0179</td>
</tr>
</tbody>
</table>

From Fig. 14, it can be seen that spots 1-4 are in different environments. Spot 1 and spot 2 are clusters of dye molecules. Spot 3 is much darker than other spots, and spot 4 is at the edge of molecules cluster. Table 2 indicates that when the environments are different, the fluorescence lifetimes are slightly different, because fluorescence lifetime is an average effect, and affected by environment.

The fluorescence lifetime of CdSeTe quantum dots is very sensitive the environment. And it is in the order of 50-
150ns. This time is longer than the time interval of laser pulse. So the time interval is much longer than interval of laser excitation. The lifetime of DiI dye molecules is about 2.6ns, which is much shorter than the time interval of laser in our experiment. It will also possible to have single emitters using this kind of molecules.

We also observed the bleaching of dye molecules, which is also a problem for single photon source. Turn off the APDs and get rid of all filters, we expose an area of 1 \( \mu m^2 \) to the full energy of laser for 2min, 4min, and 8min, it can be seen from Fig. 16 that as the increase of time, the photon emission of dye molecules is greatly diminished. So in the experiment, we should avoid bleaching in the experiment by using filters.

![Fig. 16 Bleaching of DiI dye molecules for (a) 2 minutes (b) 4 minutes (c) 8 minutes](image)

**SUMMARY**

In this Lab, we successfully achieved antibunched single photon source with CdSeTe quantum dots, which was approved by antibunching measurement in the Hanbury and Twiss setup. The main difficulty for making single photon source is getting single emitters. In our experiment, the quantum dots tend to cluster together. So controlling the concentration of sample is very important. Spin coating is also helpful in distribute quantum dots. We also studied the mechanism of single photon emission. Due to fluorescence lifetime, electrons in quantum dots can’t absorb photons before the end of fluorescence, which guarantee single photon emission.

**APPENDIX**

1. Signal from APD. We used a digital oscillator to measure the photon current from one APD. When photons incident on the APD, it will trigger the avalanche of electrons, and generate a TTL pulse. The pulse is 35ns wide and 5 voltage height, as shown in Appendix Fig 1.

![Appendix Fig. 1 TTL pulse from APD](image)

![Appendix Fig. 2 Electrical output of laser](image)
2. Electrical output of laser. In our experiment, the repetition of laser is 76 MHz. The electrical output of laser is showed in Appendix Fig. 2. It has a high repetition rate and the width of the pulse is about 13ns.

3. Decision of zero time interval position. In the antibunching experiment, we used the second order correlation of output of two APDs. There are delays from the cable, from the computer, from the different distances of APDs and the beam splitters. So we do not know exactly the delay between the two signals. To determine the zero delay of the two outputs, a trick is needed. As shown in Appendix Fig. 3, use an inverter to guide put of the output from APD1 to the delay card, and then send to the Time-Harp card together with the other part of output (the route of this part is the same as that of APD2), to do a self second correlation of APD1 in the computer. The result is showed in Appendix Fig. 4. In this way, we can eliminate the time delay after the inverter of APD1 and APD2. Then in the experiment, slightly change the delay by delay card, it will be easy to get the zero delay time of the two detectors.

\[ \text{APD1} \quad \text{Delay card} \quad \text{Computer} \]
\[ \quad \text{Inverter} \quad \text{Time-Harp card} \]

Appendix Fig. 3 Circuit for zero time interval decision

\[ \text{Photon counting} \]
\[ \text{Time interval (ns)} \]

Appendix Fig. 4 Self second correlation of one APD

REFERENCES