Imaging of Single Photon Emitter Fluorescence and Photon Antibunching

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Abstract
In this experiment we demonstrate the Confocal Microscope fluorescence imaging of various single photon emitters like carbon nanotube, quantum dots and color centre in nano-diamonds. We also measure their spectrum to characterize them. Finally we use the Hanbury Brown and Twiss Setup to test if the photons emitted from our source has antibunching properties.

Keywords: Confocal Microscope, Single Photon Emitter, Antibunching, Hanbury Brown and Twiss Setup.

1. Introduction:

The advent of quantum information and computation science has placed stringent demands on optical sources, namely that produce single photons. Single photon sources based on single-emitter fluorescence are simply sources of photon emission that emit a single photon at a time. This property is known as antibunching [1] where each photon is separated in time. In our experiment, we probe different single photon emitters by imaging their fluorescence with a confocal microscope and test for antibunching characteristics using the famous Hanbury Brown and Twiss Steup [2].
2. Theory:

Antibunching is a purely quantum effect and cannot be realized, in anyway, from the classical theory of light. A simple interpretation of antibunching may be realized from the understanding that, light is a manifestation of discrete quantized packets of energy (photons). From this model, it is evident that if only one photon is incident on the beam splitter, then it can not be simultaneously detected at both the detectors. In that case the joint probability of equal time photo detection at the two detectors will be zero. To prove this mathematically, let us consider, a typical Hanbury Brown-Twiss setup with two detectors placed at two different points with positions \( r_1 \) and \( r_2 \), the joint photo-detection probability at the two detectors, at two different times \( t \) and \( t + \tau \) may be given by the formula

\[
g^{(2)}(r_1, t; r_2, t + \tau) = \frac{\langle I(r_1, t)I(r_2, t + \tau) \rangle}{\langle I(r_1, t) \rangle \langle I(r_2, t + \tau) \rangle}.
\]  

(1)

In their experiment Hanbury Brown and the Twiss found the equal time photo-detection (coincidence photon counts) to be more probable than the unequal time photo-detection, i.e,

\[
g^{(2)}(r_1, t; r_2, t + \tau) \geq g^{(2)}(r_1, t; r_2, t).
\]

Their result perfectly matched with the prediction made by classical theory of light.

In the quantum mechanical formulation, the formula for the joint photo-detection

*Fig 1: The correlation function can become greater than or equal to one in classical light field (eg thermal or coherent light) and can be lower than one in quantum light field (sub-poisson state, number state, etc)*[3]
The probability, given in Eq. (1), is replaced by the formula
\[ g^{(2)}(r_1, t; r_2, t + \tau) = \frac{\langle I(r_1, t)I(r_2, t + \tau) \rangle}{\langle I(r_1, t) \rangle \langle I(r_2, t + \tau) \rangle}, \]  
where the measurable quantities have been replaced by corresponding operators in the usual sense of quantum theory and the colons denote the normal ordering. In the quantum mechanical treatment, it turns out that the joint photo-detection probability at equal time may be smaller than unequal time photo-detection probabilities, unlike the classical treatment. Particularly, if a Hanbury Brown-Twiss experiment is performed with single photons, then,
\[ g^{(2)}(r_1, t; r_2, t) = 0 \leq g^{(2)}(r_1, t; r_2, t + \tau). \]  
This phenomenon is known as antibunching. The correlation function for bunched, coherent and antibunched state of photon is shown in figure 1.

In this experiment we use single emitter quantum dots, CNT, NV color centre in Nano-Diamonds and other fluorescent molecules to produce single photons and observe antibunching using a Hanbury Brown-Twiss setup. The reason for using single emitter quantum dots (or, fluorescent molecules) is the fact that, single photons cannot be produced by attenuating normal laser beams. Attenuation never negates the chance of producing photons in pair or triplet or sometimes zero photon per pulse which causes pollution from other photons in any quantum information processing applications. So far, the only known way of producing single photons is by electronic transitions in certain kinds of atoms, molecules or quantum dots. We will illustrate the theory by considering a
model of single atom whose energy level structure is given by Fig. 2. When this atom is excited by a trigger pulse, the electron moves from the ground state to the exited state. This electron might return to the ground state in two steps emitting photons of two different wavelengths. The unwanted wavelength is removed by using a proper filter. In this experiment, we use optical pulses to excite the prospective single photon emitters. We also measure the spectra of these nanostructures with a built-in spectrometer in our experimental setup.

3. Experimental Setup:

The principle of the main experimental setup is explained in Fig. 3. Light from the 532 nm laser source is focused on the sample by the objective of a confocal microscope. The light emitted by the single emitter is also collected by the same objective. This radiated light may be directed to a EM-CCD camera or to the Hanbury Brown-Twiss interferometer, by choosing a proper out going port of the confocal microscope. The

![Fig 3: Schematics of our Experimental Setup.](image)
Hanbury Brown-Twiss interferometer is used to test whether the emitted photons are antibunched. Here we direct the radiated light to the APD where we test only one single emitter at a time. However, to image or to capture the video of many fluorescing emitters throughtout the area of the sample, the EMCCD can be used. The microscope should be used in the wide field mode for EMCCD capture as it images many emitters at a time.

4. Procedure:

A. Sample Preparation:

Three types of sample were used during this experiment. Quantum dots, CNT and nano diamonds.

- Preparation of quantum dots: A quantum dot is a portion of matter whose excitons are confined in all three spatial dimensions. Consequently, such materials have electronic properties intermediate between those of bulk semiconductors and those of discrete molecules. A cartoon of the CdSe quantum dot showing the orientation of Cd and Se atoms are shown in figure 4. We took 10nM concentration of 800 nm wavelength of colloidal CdSe quantum dots solution in a micropipette and dispensed in on a glass cover slip and spin-coated at around 3000 rpm for few seconds to get a uniform layer of quantum dots on the cover slip.

Fig 4. Cartoon of CdSe quantum dot showing the orientation of a group of Cd (Blue spheres) and Se (Red Spheres) into a confined spherical structure.[5]
• Preparation of Single Walled CNT: Single-walled carbon nanotubes (SWNTs) are tubular graphitic molecules (figure 5) that can be microns long and less than one nanometer wide. The combination of the unique properties of the carbon–carbon bond and their interesting molecular structure have given SWNTs unique physical properties and provided the potential for optical and electrical applications. The CNT samples were obtained from Prof. Krauss’ Group. The SW-CNT were dispersed in surfactant micelle solution to avoid agglomeration. A drop of each of them were taken and put on a glass cover slip and loaded on the microscope in the same way as the quantum dots.

![Fig 5: Cartoon of Single walled carbon nanotube [6]](image)

• Preparation of Nano-diamonds: The Nano-diamonds were commercially bought and prepared in the same way for our experiment as the SW-CNT.

B. Loading the sample:

We loaded the cover slip with the sample on sample space of the microscope objective with the help of two small magnets after adding a drop of oil before the objective. The oil drop helps us to work with high numerical aperture objective. In order to assure proper mounting, only a very small drop of oil is used. Larger drops increase the likelihood that the sample will not be level which affects the focus of the system due to the very small depth of field of the confocal microscope. The sample was held on a piezo translation stage, which allowed translation of the sample in x, y and z direction. The
apparatus allowed for the excited sample to be studied using an eyepiece, a EMCCD camera, or by monitoring via the avalanche photo diodes (APDs) in the Hanbury Brown and Twiss setup. In our experiment we use only eyepiece and APD ports.

C. Focussing and Imaging the Single Emitter Fluorescence using Confocal Microscope Imaging:

Our setup sent the laser light through a spatial filter that changed the elliptic beam cross-section into a round Gaussian cross-section, and broadened the beam diameter. The beam then passed through a series of removable neutral density filters used to control the incident intensity on the quantum dot sample in the objective of our confocal fluorescence microscope. We used a confocal microscope in order to selectively detect only emitted light from single molecule CdSe fluorescence originating at the focal point of the microscope. Confocal microscopes decrease side-lobe noise in detection signals. This microscope thus allowed us to scan a sample of quantum dots and focus on a single-emitter to obtain images with high resolution (using special LabVIEW software by Prof. Lukas Novotny). This is achieved within the confocal microscope by using a focusing lens (can also use two lenses) and a pinhole, as shown in Fig. 6. In our setup, the single photon counting avalanche photo-diode (APD) diameter is small enough, at 150µm,

Fig 6. A schematic of confocal microscope with standard elements [4]
that we used the APD detector surface area as the pinhole. We used a special high numerical aperture oil-immersion objective to achieve high resolution and intensity on the sample. A dichroic mirror was implemented to reflect the laser light but transmit the fluorescent light. The output fluorescent light is finally passed through a filter to block the laser mode and sent to the APDs in the Hanbury Brown and Twiss setup component of our experiment.

D. Studying the spectrum of the emitters:

To observe the spectrum of our samples, the laser light was first calibrated to 532 nm. Next spectrum from CNT and NV colour centres were taken with 5 sec exposure with our spectrometer (fig). Additionally we also obtained the spectrum from gold nanoparticles which are plasmonic samples. We have observed the characteristic peaks from our sample spectrum as determined in literature.

F. Setup for observing Antibunching

i) Alignment of APD with laser: Before starting the antibunching experiment, we need to align the APD such that the laser spot passes through the centre of the square spot of APD.

ii) Measuring of Zero Point: Zero point is the point on the histogram (figure 14) of the antibunching where the time delay between the one and the next incident photon is zero [7]. Zero point was measured by splitting the signal from a single APD with the help of BNC Cable and feeding it to the start and stop channel.

iii) Proving the Antibunching: The beam-splitter of the Hanbury Brown-Twiss setup directed about half of the incident photons to APD 1 and the other half to APD 2 (see Fig. 7). These two APD's were used to compensate a deadtime of each detector in measuring the time intervals of two consecutive photons. One of the APD's provided a “start” signal and the other provided a “stop” signal. By measuring the time difference between the “start” and “stop” signal, one can find the delay between two photons (inter-photon time). The number of “coincidence counts” were plotted against the “interphoton time” in the form of histograms by the Time Harp software in the computer.
5. Results and Analysis:

A. Confocal Microscopy Imaging of Single Emitter Fluorescence:
We focused our spot through the eyepiece to obtain four leaved clover. To start with, we raster scanned a 25µm by 25 µm of our CNT sample around the laser spot with the help of the Nanodrive. We used a 532 nm laser with pulse duration of 6 picoseconds and repetition rate of 76 MHz. The experiment was controlled by a LabView code created by Prof. Novotny’s group where the input parameters like the scan area and number of pixels were fed. A snapshot of the user interface with images from CNT sample (fig 9) is shown. The images from the vertical and horizontal APD were captured during the raster scan. A linegraph of the intensity along a single line of raster scan could also be noted.

![Fig 8: Confocal microscope fluorescence image from single colour centres in nanodiamond (left) and 10nM of800nm wavelength CdSe quantum dot (right).]
during our experiment. The fluorescence images were obtained which showed optimum point spread function giving us the hope that these emitters could emit antibunched photons. We also imaged the fluorescence of color centre in nano-diamonds (fig 8 left) and 10nM of 800nm Wavelength of CdSe Quantum Dots (fig 8 right).

![Labview scan Display](image)

**Fig 8: Labview scan Display.** Distinct bright spots from fluorescence of individual Carbon nanotube sample can be seen here. The two panels on the right give the results of the scan through a color-coded display of photon count per pixel from each of the two APDs. The panel on the bottom displays the photon count against position for a given line scan. The bottom panel could also be used to observed photon count against time, where we were observing the photon count from a single pixel (indicated with a number and cross-hairs in the top right panel of fig). Scan area 25µm²

**B. Spectrum of the samples:**

A spectrometer was placed at one port where the radiated light from the sample could be fed into a spectrometer. The laser light was calibrated to 532 nm (fig 10). Next,
Figure 10. Calibration of the diode pumped solid state laser beam at around 532 nm.

the spectrum of our samples were noted. In addition to our CNT, and nanodiamond sample, we had also measured the spectra of gold nanoparticles. Fluorescence spectra were detected for different sample position and the intensity of emission were plotted against the excitation wavelength (fig 11-13) Distinct maxima for the characteristic fluorescence spectra were obtained for our samples which matched the values in literature [8].

Figure 11: Characteristic spectrum of single walled CNT with 5 ns exposure.
Figure 12: Characteristic spectrum of NV color centre in nanodiamond with 5 ns exposure.

Fig 13: Characteristic spectrum of gold nanoparticles.
C. Antibunching Measurements:

i) Measuring Zero Point: The Hanbury Brown and Twiss setup uses a Timeharp card to create an antibunching histogram. Once a photon is detected in one of the APDs, a TTL pulse signals a capacitor in the Timeharp card to start charging. When a photon is detected in the other APD, a signal is sent to stop the charging of the capacitor. The charge on the capacitor corresponds to the time that has elapsed between the arrivals of photons. If the photons are truly antibunched, then one would expect that there should never be zero time between the start and stop signals. An intentional delay is used to adjust where the zero time is on the histogram. To determine the zero time of the histogram setup, a signal from one APD was split and sent to both ports of the Timeharp card. This creates two pulses that represent a simultaneous arrival. In reality there is a delay between the arrival at the two ports, and this will be represented as a line (single bin) on one particular time (fig 14). This particular time is the zero time. When an antibunched histogram is collected there should be a dip at the zero time. If two photons

![Graph showing antibunching measurements](image)

**Fig 14** The signals that should have arrived at the same time all show up in the same histogram bin at around 63 ns.
are bunched together, then the beam splitter could send them to separate APDs, thus being binned at the zero time. This should not happen with antibunched light.

**ii) Measuring Antibunching:** As can be seen in figure 15, scans show clear regions of higher intensity photon emission, indicative of fluorescing quantum dots. Stripping in this region shows that the quantum dot is blinking. Thus, we were able to image single emitters in the form of colloidal quantum dots using confocal microscopy. Based on these scans, we focused on pixels in this scanned region that looked to be locations of single emitters and tested for antibunching. Results of testing observed quantum dots for antibunching were varied. A frequently encountered problem was quantum dots bleaching before adequate data was collected to determine whether antibunching was occurring. A sample histogram made using the Time Harp program are shown in figure 17. Unfortunately we were not able to find evidence of photon antibunching, as shown in figure 17 (left). In the ideal case (figure 17 left), we see that there is a notch like structure at the delay line, but in our case (fig 17 right) the structure is not sufficient to give proof.

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*Fig 15 : Sample scan displaying fluorescence intensity of different parts of the sample. The scan was taken using a sample of CdSe colloidal quantum dots. Regions of high photon counts are indicative of the presence of quantum dots in that part of the sample. The streaky nature of the circled region is indicative of a blinking quantum dot, because different line scans see different fluorescence intensities.*
Figure 16: This figure shows a time trace of quantum dot fluorescence. The counts as a function of cumulative data intervals captured (time) shows the blinking properties of quantum dots. Lowering of count could be a possible indication of bleaching of the quantum dot.

Fig 17: Sample image of histogram constructed via the Time Harp program. While dip in the graph, as can be seen in the image obtained from Luke Bissel’s sample in (left), are indicative of antibunching, we saw scans like the sample image in (right) that gave no evidence of antibunching. Although it has a very small dip at the delay point, yet it is not sufficient to prove antibunching. X axis shows the interphoton time while the y axis gives the coincidence count.

of antibunching even after multiple scans. In further measurements we are planning to enhance the optical properties of these quantum dots in photonic microcavities using cholesteric liquid crystal and carry out antibunching measurements.
6. Conclusion:

Thus in this experiment we have prepared samples that are known to emit single photons and obtained their fluorescence imaging with a help of confocal microscopy. We have also obtained spectrum from our nanostructured samples with the help of a built-in spectrometer in our experimental setup. Apart from that we have observed blinking from quantum dots. While carrying out antibunching measurement, it was seen that the histograms showing photon count with interphoton time, had no dips at the delay points. Hence antibunching was not observed with our samples. This could be possibly because our sample had large bunches of quantum dot and the fluorescence we obtained was not from a single quantum dot or any other emitter used. The sample concentration should therefore be as less as possible. However, we plan to further investigate the antibunching phenomenon by enhancing the optical properties of the quantum dots by embedding them in photonic chiral microcavities using cholesteric liquid crystal host. This might show promising result.

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List of References:


