Laboratory 3&4: Confocal Microscopy Imaging of Single-Emitter Fluorescence and Hanbury Brown and Twiss setup for Photon Antibunching

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Abstract
This experiment purpose was the study of single photon sources and confirm whether or not they exhibit photon antibunching in their behavior. We imaged fluorescence using a confocal microscope. We also used the Hanbury Brown and Twiss interferometer with Avalanche Photodiode Detectors to obtain antibunching histograms. The photon antibunching histogram was not achieved.

Background
In order to understand the experiment, one needs to understand the concept of single photon sources. Single photon sources are in essence a light source that emits a photons separated by time. Also they release, as the name implies, one photon at the time. This is highly valuable in cryptography because photons are elementary particles that cannot be separated and thus they cannot be separated into smaller particles. Examples of sources that we can use as single photon sources are dye molecules, carbon nanotubes, quantum dots and color center nanodiamonds.

A single emitter excited to a higher energy state will release a single photon to decay to a more stable state. The average amount of time it takes to this emitter to decay and emit a photon is called the fluorescence lifetime. The other concept to understand in this lab is antibunching. Antibunching is basically photons spaced evenly through time. Single photon sources are then sources that create these antibunched photons. It might appear then than an attenuated laser might be able to create photons to the single photon level but we have to think that the laser produces this single photon level in average. This means that at a single instance in time there might be a time where there might be a pair of photons, a triplet or not any photons at all. Then we can see our second order correlation function to understand antibunching in a mathematical form:

\[ g^{(2)}(\tau) = \frac{\langle N_1(t)N_2(t + \tau) \rangle}{\langle N_1(t) \rangle \langle N_2(t + \tau) \rangle} \]

where \( g^{(2)} \) is the second order correlation function at two different times \( t \) and \( t + \tau \). Coherent light has \( g^{(2)}(\tau) \) of 1, also at \( \tau = 0 \). Classical light has a \( g^{(2)}(0) \) > 1 and \( g^{(2)}(0) > g^{(2)}(\tau) \). Antibunched light has a \( g^{(2)}(0) < 1 \) and a \( g^{(2)}_{\text{max}}(\tau) = 1 \) [1].
In the observation of single photon sources, a microcavity is used to enhance the fluorescence. In this lab the microcavity was developed with photonic bandgap materials. What these materials do is that they create selective reflectivity through periodicity. In the lab, cholesteric liquid crystals were used which suppress emissions in the blocked frequencies and enhance the ones at the band edge. These materials can be used in tandem with different materials such as color center nanodiamonds, carbon nanotubes, quantum dots or dye molecules. In this lab we used nanodiamonds, carbon nanotubes and quantum dots.

Quantum dots are semiconductor structures that confine excitons in all directions. Since electrons are confined in this matter, they will have discrete quantum energy states just like a single atom does and this is mainly the reason why quantum dots act like a single atom when they get excited. Color center nanodiamonds are nano-sized diamonds that have a “defect” on their structure which act as single emitter. Carbon nanotubes present a carbon configuration that allows them to have observable single photon fluorescence.

An instrument that we use in this experiment is the confocal microscope. This microscope images an object inside a pinhole. What this pinhole does is that it effectively allows the microscope to image the object in the pinhole while blocking out other light sources not in the pinhole and thus increasing the contrast and the details in our image. To increase our resolution we used a high numerical aperture oil immersion objective.
The instruments used in this lab consisted of a confocal microscope and a Hanbury Brown and Twiss interferometer setup. Single emitters were made on glass microscopes slides, where they could be pumped by a laser. The sample was held on a piezo translation stage, which allowed for a translation of the sample. The apparatus allowed for the excited sample to be observed using an eyepiece, an EM-CCD camera, or by using the avalanche photo diodes (APDs) in the Hanbury, Brown and Twiss interferometer setup. The full setup is shown in figure 1. Two lasers were used in this lab to excite the sample. One laser was a 5 mW HeNe laser, and the other was a diode-pumped solid-state pulsed laser that operated at a wavelength of 532 nm. The apparatus uses a dichroic mirror to direct laser beams to the sample, but passes the light emitted by the sample to the interferometer setup. The Hanbury, Brown and Twiss interferometer setup consisted of a beam splitter and two single photon counting avalanche photo diodes (APD). The beam splitter will split any incoming photon beams into the two APDs, which allows observation of photons spaced in time in spite of the “dead” time of detectors immediately after photon detection events. This setup is used for testing for photon antibunching.
APDs emit TTL pulses in response to incident photons. TTL signals were monitored with a Lab View program that allowed for confocal microscopy. The program controlled the piezo translation stage and could thus be used to scan the sample through the pump beam while the number of incident photons for a given pixel and time size were recorded by the program. This allowed us to image our sample based on these regions of varying intensity. Testing for antibunching was accomplished with a Time Harp card in conjunction with the Hanbury Brown & Twiss interferometer. One APD would operate a start channel for a Timeharp 200 board while the other channel would have a variable delay and operate a stop channel on the same board.

We need to adjust a zero time in our lab first. If photons are antibunched there should be no delay in our signals. The variable delay is used here to adjust our zero time for the histogram. To obtain it we shoot a signal from one APD to be split between both ports of our Timeharp card. This will create two pulses for a simultaneous arrival. The truth is that there is a delay between the ports which will be represented as a single line in our histogram. This will be our zero time.

![Figure 2: The signals from our APD that should have arrived at the same time will show approx. between 55~63 ns.](image)

As photons impinge on the APDs, the counter timer board is started and stopped to give a measure for the time difference between photons incident on the two different APDs. These time different values are recorded and used to build up an antibunching
histogram by the Time Harp program. When an antibunched histogram is formed there should be a dip at the zero time. If two photons are bunched together, then the beam splitter could send them to separate APDs, thus being binned at the zero time. This is not desirable with antibunched light.

**Procedure**

1. Prepare concentration of the desired sample. Use a precise pipette to take a drop of the sample on a microscope cover slip. Use a glass rod to distribute the sample uniformly on the cover.
2. Prepare microscope objective by cleaning with acetone or methane. Place a drop of the immersion oil on the objective lens. Place the sample over the objective and secure the piezoelectric stage with the magnets provided.
3. To attenuate laser you might have to remove filter, if you do then make sure to remove filters one at the time.
4. Using the eyepiece make sure you obtain the “four leaved clover” shape of the light.
5. Obtain spectra of the emitters to be used in the experiment.
6. Start the raster scan. Adjust the Hanbury Brown & Twiss interferometer to make sure the laser light goes correctly through the pinholes and to improve the raster scan.
7. Select a bright spot in the scan. Try to select emitter fluorescence on the sample rather than clusters.
8. Determine the zero time in histogram.
9. Start the histogram of data from the Timeharp card. The antibunching histogram will have the signature “dip” at the zero time.

**Results and Analysis**

Before starting our raster scans, we obtained the spectra of our selected emitters which were quantum dots, carbon nanotubes, gold nanorods and nanodiamonds. Note that the peaks are what the emitters are since that is their emission spectrum.
Figure 3: Spectrum of Gold Nano rods sample with 1 sec exposure.
Figure 3: Spectrum of carbon nanotube sample with 1.30641 sec of exposure.
Figure 3: Spectrum of nanodiamond sample with 5 sec of exposure.
Figure 3: Spectrum of CdSe Quantum Dot sample with 1.30641 sec of exposure.

For our samples of confocal microscopy we used nanodiamonds and quantum dots but since we used quantum dots for our anitbunching analysis only. We will see how the color center nanodiamonds scan looks like.
In this lab, we used 3 different kinds of samples but only one in which we created a histogram. We focused on Quantum Dots in order to create an antibunching histogram so we are going to present the results from the Quantum Dots sample. Unfortunately we did not obtain the result that we would have desired which is a dip at the zero time in the histograms to prove antibunching.
Figure 5: Histogram of the quantum dot. If a dip is missing at the zero time, then we can expect the sample to not contain antibunching like this one. This one is not antibunched.

We tried to modify the sample by selecting a different bright spot in our scan.

Figure 6: Same quantum dot sample but with different selected bright spot.
Figure 7: Histogram of the new selected spot. No antibunching in this histogram.

After obtaining again an undesirable result in our lab, we decided to execute a different scan by moving the sample and thus obtaining new bright spots.
Figure 8: Same quantum dot but different scanning location. In other words we moved the sample to obtain a new scan.

Figure 9: Histogram of the new moved sample. No antibunching recorded.

Conclusion

As seen in our results, antibunching could not be obtained. There are multiple reasons why. It could have been a lack of proper alignment in the interferometer; it could have also been bleaching of samples. This experiment is difficult in that one must take care of all these variables that can influence our results and also that even if one takes the proper precaution one must also account for the behavior of the samples such as blinking of the quantum dots.

References