ABSTRACT

We experimentally studied and demonstrated the non-classical photon interpretation, called antibunched light, which is incredibly important in quantum optics. In contrast to the first observation made by Kimble, Dagenais and Mandel, we prepared and used colloidal quantum dots as emitters; the emission was improved by introducing them in a photonic band-gap material, made by cholesteric liquid crystals. The use of Hanbury Brown and Twiss setup allowed us to achieve an interesting histogram, from which we concluded that we have antibunching and therefore the quantum dots act as single photon sources.

Keywords: Antibunching, Single Photon Sources, Quantum Dots, Photonic band-gap material, Hanbury Brown and Twiss setup.

1. Introduction

Recently we have been witness of all the new concepts and terminology related with quantum communications and how fundamental phenomena, which arise from quantum theory, have allowed developing technology and new knowledge in this field.

Basically the light can be classified in coherent, bunched and antibunched. The last one exhibits a purely non-classical behavior, this means that the emitted field cannot be described by continuous electromagnetic field, it is quantized which is of incredible interest in quantum theory of light, and the main aim to do this experiment.

As we will discuss it in the next section the nature of light is due to the source. We can have different light sources; each of them is described by certain statistic. Experimentally those can be distinguished by correlating the same light at different types. In fact this is the idea behind the Hanbury Brown and Twiss experiment, which is one important part in our current experiment.

The purpose of this experiment is to study antibunching, such proof requires of many different issues. First we need to have a single emitter, in fact scientists have used a lot of sources such as single atoms, single molecules, semiconductor crystals, color center nanodiamonds, and photonic crystals. In fact Kimble, Dagenais and Mandel [1] using single atoms made the first antibunching observation. We used quantum dots as single emitters, and we enhanced their emission by introducing them in a photonic bandgap material cavity of cholesteric liquid crystal, then we excited the quantum dots using a laser with a wavelength of 632.8 nm and we measured its fluorescence and its statistical behavior using the Hanbury Brown and Twiss setup [2], which in fact obeys to a completely different statistic to the one obtained from an attenuated beam.

Other important point to mention is that quantum dots as most of the confined structures have resonance in optical frequencies; in fact the resonant frequency depends of the size of the structure, this can be clearly understood if we consider the model of a particle in a box [3], where the particles are the carriers, this will emulate the model of hydrogen atom therein the reason to call quantum dots as artificial atoms, where we have quantized levels.

Regarding to fluorescence of electrons this process consists on excite the confined carriers from ground state into excited states, as the time evolves this electron will go back into
the ground state, this happens in a short range of time that goes from some nanoseconds to hundreds nanoseconds, this is illustrated in the sketch shown in figure 1, where $S_0$ is the ground state and the singlet states $S_1$ and $S_2$ are the successive energy levels. After one electron is excited, this may acquires an energy state higher than $S_1$ or $S_2$, in spite of that in the relaxation process it will achieve lower states before the emission, this transition is called internal conversion. It is important to mention that the transition from $S_1$ to the ground state occurs on a very small period of time, in the order of 10 ns, this value is named lifetime of fluorophore, this means that we will have only one photon separated in time, or antibunched photon, this part of the transition is called fluorescence.

![Energy diagram which describe the transitions in quantum dot.](image)

2. Antibunching

Antibunching is fundamental and quite important to the quantum information field, telecom and in general in quantum optics. Antibunching is the separation of photons in time [4]. In spite of many technologies and applications based in antibunched light have been developed, it is amazing the physics behind this behavior, which is purely quantum optical, and it has not a classical counter part, this in contrast to bunched and coherent light.

The light is classified according to the statistic that it follows; such nature can be also distinguished by the second order correlation function $g^{(2)}(\tau)$, which is a comparison of two functions at different positions or time, this expression can be calculated as follow:

$$g^{(2)}(\tau) = \frac{\langle n_1(t)n_2(t+\tau)\rangle}{\langle n_1(t)\rangle\langle n_2(t+\tau)\rangle}$$

(1)

In our case such functions are $n_i(t)$ which represent the number of counts registered by the detector $i$ at time $t$ or $t+\tau$, those detectors are part of the Hanbury Brown and Twiss experiment shown in the following figure 2. This experiment consist on a 50:50 beam splitter which divide the initial beam in two equal portions that impinge two single photon detectors, the output signals of the detectors are send to a timer-correlated single-photon counting PC board module which records the time elapsed between the pulse of each detector and count the number of pulses at each input.
Using such experiment we can discuss three different scenarios. First we can discuss the case where \( g^{(2)}(0) = 1 \), this implies that we have perfectly coherent light, therefore the probability of obtaining a stop pulse is the same for any \( \tau \). Moreover we have the scenario when we will measure that \( g^{(2)}(0) > 1 \), corresponding this to bunched light, this is the case for thermal sources, naturally the probability to detect one photon is higher if we measure in short scales of time than long intervals. Finally, we can have the case where photons are separated in time, this means that we have antibunched light; in such case it is physically impossible to detect two photons at the same time, therefore \( g^{(2)}(0) < 1 \). As we already mentioned, Kimble, Dagenais and Mandel demonstrated such non-classical behavior of light, they obtained \( g^{2}(0) = 0.4 \). After this demonstration many scientist have been working with antibunching experiments and investigating different methods to produce single photon sources.

3.- Photonic band-gap materials

It is easy to figure it out all the variables and factors that one can have in a real experiment; unfortunately our case is not different, therein the reason to use different tools to improve the conditions and the effects under study. In our case we used photonic band gap materials to enhance the emission of the quantum dots. We will review the fundamental ideas of photonic crystals in this section and we will explain how we experimentally used them in the next section.

Photonic crystals are materials that inhibit the propagation of certain light frequencies by the use of a periodic structure [5], which alternates the refraction index with the periodicity of the order of light wavelength a well-defined thickness, see figure 3 a, such structure is for light what an atomic lattice for electrons, where the interaction of light with the structure results in bands called photonic bandgap, see figure 3b, just similar to the energy bandgaps in a semiconductor crystal, but such forbidden bands are bands where it does not exist a solution for the electric field, then the light is reflected for such light with certain frequency see figure 3c.
Figure 3-a) Periodic structure, 3-b) Dispersion relation, 3-c) Transmission behavior of photonic crystals.

Now that we have described the idea behind photonic crystal and photonic bandgap we can move forward in our explanation of bandgap materials. It is important to mention that we are using cholesteric liquid crystals (LCs) to fabricate band gap materials. In general liquid crystals are materials with properties between liquid and solid crystals, made of large organic anisotropic molecules which can acquired a specific orientation forming different phases. Cholesteric crystals, for visual demonstration these structures can be described by the layers with different orientations from 0° to 360°, such a period of structures are called pitch; this is presented in figure 4.

Figure 4 Photonic bandgap materials with the reflected and transmitted circular polarized light.

Liquid crystals form periodic structures with periodically changing refractive index for circularly polarized light with one handedness, which behaves quite similar to photonic crystals, therein the reason to call them photonic bandgap materials, in this case we have reflection of one circular polarization for a wavelength given by \( \lambda_0 = n_{av} P_0 \) where \( n_{av} \) is the average of refraction index given by \( \frac{(n_e+n_o)}{2} \) with a band width given by \( \lambda_0 \frac{(n_e-n_o)}{n_{av}} \).
4.- Experimental Setup

In this section we will describe the experimental setup used to study antibunching, in fact results very interesting all the different disciplines and elements that this experiment implies.

We will split the experiment in three parts; we divided it in such manner because is the natural procedure that we used every time that we studied different samples. First of all we have our excitation source, in this experiment we used two types of lasers with different wavelengths. The first is a CW laser with a wavelength of 632.8 nm and the second is a mode locked laser with a wavelength of 532 nm with pulse duration of 6 ps and a repetition rate of 76 MHz, the average power was approximately 40 mW. We spatially cleaned the laser beam and we modified the initial polarization to circular polarization, then we raised the height of the beam using two mirrors, then we attenuated the beam before this goes into the confocal fluorescence microscope, key part of this experimental setup, which starts with the dichroic mirror, see figure 5.

![Sketch of the experimental setup.](image)

This experiment makes use of expensive equipment, which is very sensible to certain levels of intensity, therein, the need to work in a dark room, and always being careful of the laser intensity or other sources of light, such as quantum dots fluorescence, this in order to not denied EM-CCD or the single photon APDs. Before continuing describing this setup is important to describe the idea behind the confocal microscopy and why is important in our setup, we show a basic diagram in figure 6.

The need of confocal fluorescence microscopy regards in the possibility to imaging with increased resolution and contrast, this is possible because confocal microscope blocks the light which is not originated in the focal plane, this thanks to the pinhole positioned before the detector.

Continuing with the description of the first part, which has as a main function to image the quantum dots fluorescence, we needed to select the appropriated port, basically we used the port
number 2 and 4, if we wanted to use the EM-CCD camera we selected the port number 2, for the Hanbury Brown and Twiss setup we used port number 4.

Then using the port number 2, we can take an image of the quantum dots using the EM-CCD, then the image is obtained in computer 1. In this case we changed the position of focus, so with EM-CCD camera we worked not in confocal mode, but in a wide field microscopy mode.

![Figure 6 Sketch of confocal microscopy](image)

The next part of the experiment has as a main purpose to scan the sample using a nanodriver to control the piezo translation stage, this stage moves in xy direction, meanwhile the two APDs with area (150 um²) detect each photon and sent a TTL signal to photon counting card, such information is sent to a computer program that allows to form an image of the sample and analyze the number of counts, see figure 7:
Finally the third part of our experiment consists in a version of the Hanbury Brown and Twiss experiment that we already explained, but in our experiment it will allow us to obtain an antibunching histogram. It is important to mention that this part make use of the Time harp which is a programmable delay card connected to computer number 3. This part requires of electronics, here we will illustrate how we calibrated it. First we used only one APD, then we divided the pulse with a T-junction, it is important to mention that the length traveled by the electrical pulses must be equal, this in order to avoid delays, then we programmed the delay station to have a delay of 60 ns, finally we compared the delay between both signals, as it is shown in the following figure, the same logic was used to obtain the zero delay in our antibunching histograms.

The following figure illustrates the number of counts obtained after the delay station was programmed at 60 ns.
Once we discussed the previous topics we will be able to understand the experimental results and how we obtained them.

5.- Experimental Procedure and Results

We began preparing different samples of quantum dots. They are colloidal and CdSe quantum dots fluorescing at 579 nm and 800 nm respectively, at concentration of 10 nmol and 1 nmol. We prepared them by dropping quantum dots substance in a glass cover slip, then we mixed up with liquid crystals for 3 minutes, we repeated the same procedure in other slip, then we putted both slips together face to face forming a sandwich structure, finally we shifted in one direction, this in order to form the photonic bandgap structure which will become in the cavity of the quantum dot. The reason to have the quantum dots in the photonic bandgap material can be seen in the plots shown in figure 10, figure 10 a) shows the photonics gaps for our photonic bandgap material and figure 10 b shows how the spontaneous emission of the quantum dots (this is the case for CdSe QD) will be enhanced near the band edge.

For CdSe quantum dots we followed a similar process but this time we used the spin apparatus with a rotation rate 3000 rpm for a 40 seconds until it was dry.

Figure 10-a) Transmission bands for our cholesteric photonic bandgap material. b) Fluorescence spectrum of CdSe quantum dot and transmission band of cholesteric liquid crystal.
After we observed the quantum dots using the computer one, shown in figure 5, we obtained the images shown in figure 11, for the quantum dots, it is important to mention that we are showing the fluorescence of the quantum dot and not the structure, in fact we observed how the quantum dots blink, then suddenly they stopped blinking and after a period of inactivity they just start to blink again.

![Quantum dots blinking due to fluorescence.](image1)

Then we decided to scan the sample in specific area of 25 $\mu$m x 25 $\mu$m, the obtained image is shown in figure 12, in fact, such figure shows stripes because the nanodriver scan line per line, and as we already stated, suddenly one quantum dot can stop to emit, then the next line shows a dark pixel instead of a bright one as one can expect, this behavior generates such striped figure.

![Image of fluorescence of colloidal quantum dots.](image2)

Then we decided to select certain position or certain point spread function of any quantum dot, with the idea to obtain a histogram of such emitter, and see if it shows antibunching, for the sample with a concentration of 1nmol we did not observed antibunching, in fact we obtained the following behavior.
Figure 13.- Histogram of colloidal quantum dots fluorescence of ten nmols concentration (no antibunching).

We decided to study other sample, prepared by Luke Bissell to do that we repeated the same experiment and the same process, first we observed the quantum dots blinking, then we scanned the sample and finally we selected certain quantum dots and obtained their histograms. Surprisingly we observed antibunching, this can be seen in the following plot.

Figure 14.- Antibunching histogram obtained with Luke Bissell sample, the antibunching is clear.

Figure 15 shows the image of quantum dot emission spectrum (figure 16 is this image processing)

Figure 15.- Image of the quantum dots emission spectrum obtained with an EM-CCD camera.
We also studied the spectrum of emission of the quantum dots. The obtained spectrum was wide but surprisingly we observed an unexpected peak (which is an artifact), this can be seen in the following figures:

![Spectrum of emission of Quantum dots prepared by Luke Bissell, here it is clear the peak observed at 700 nm.](image1)

5.- Fluorescence Lifetime

In order to complete our research we decided to measure the fluorescence lifetime of the emitters. This means the rate of time that the molecule is in excited state before emits and acquired a smaller state of energy, the obtained behavior can be observed in the following plot:

![Fluorescence Lifetime, taken from different quantum dots.](image2)

In order to analyze such behaviors, we used OriginLab™. First of all we plot the experimental data, this can be seen in the following figure.
It is important to mention that the expected behavior was the one showed in fig 17 and 18, in fact the behavior should be described by the following equation;

\[
\psi_T = A e^{-\frac{t}{\tau_1}} + B e^{-\frac{t}{\tau_2}} + \psi_0
\]

(2)

Where \( \psi_i \) is the concentration in excited state at any time different of zero, \( A \) and \( B \) are arbitrary constants and \( \tau_1, \tau_2 \) are fluorescence lifetime constants, our main purpose of this section, in fact it is easy to realized that the obtained plots obeyed the behavior shown in equation 2.

In figure 18 we show each of the curves shown in figure 17 with its corresponding fitting, such fitting takes as a reference the time equal to zero when we have maximum counting rate, as we already stated we want to determine the fluorescence lifetime for each of the quantum dots.
Figure 19.- This figure shows the exponential fitting for each curve shown in figure 18-a, a) corresponds to red curve, b) to green, c) to blue and d) to yellow (CLC).

Then we determined the fluorescence lifetime and the coefficients shown in equation (2) for each of the plots shown in figure 19.

<table>
<thead>
<tr>
<th>Point</th>
<th>Ψ0</th>
<th>A</th>
<th>B</th>
<th>τ1(ns)</th>
<th>τ2(ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Red</td>
<td>-219.973</td>
<td>1027.2293</td>
<td>2127.3435</td>
<td>0.8844</td>
<td>33.6453</td>
</tr>
<tr>
<td>Green</td>
<td>99.1156</td>
<td>1026.8987</td>
<td>1777.6926</td>
<td>0.8254</td>
<td>32.1213</td>
</tr>
<tr>
<td>Blue</td>
<td>83.8989</td>
<td>959.5054</td>
<td>1603.2327</td>
<td>0.7933</td>
<td>29.0367</td>
</tr>
<tr>
<td>Yellow</td>
<td>-77.7901</td>
<td>2183.9514</td>
<td>858.2994</td>
<td>0.7459</td>
<td>30.2096</td>
</tr>
</tbody>
</table>

Table 1.- Fitting parameters of each curve shown in figure 18 and 19, according to equation (2)

In fact such time constants are quite similar, in spite of the selected emitter in the sample, the following figures taken in other quantum dots, show a similar behavior.
Figure 20.- This figure shows the exponential fitting for each curve shown in figure 18-b. a) corresponds to blue curve, b) to green, c) to yellow and d) to pink (CLC).

<table>
<thead>
<tr>
<th>Point</th>
<th>Ψ0</th>
<th>A</th>
<th>B</th>
<th>τ1(ns)</th>
<th>τ2(ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blue</td>
<td>27.9019</td>
<td>1304.6174</td>
<td>1646.876</td>
<td>0.8454</td>
<td>32.6274</td>
</tr>
<tr>
<td>Green</td>
<td>-200.6639</td>
<td>1547.040</td>
<td>1662.093</td>
<td>0.8224</td>
<td>32.1416</td>
</tr>
<tr>
<td>Yellow</td>
<td>-146.6667</td>
<td>1815.6742</td>
<td>1361.66</td>
<td>0.8133</td>
<td>30.6741</td>
</tr>
<tr>
<td>Pink</td>
<td>-290.7894</td>
<td>1868.8335</td>
<td>1492.728</td>
<td>0.7174</td>
<td>26.0798</td>
</tr>
</tbody>
</table>

Table 2.- Fitting parameters of each curve shown in figure 20, according to equation (2)

6.- Conclusions

We successfully observed photon antibunching; we used a real setup, which allowed us to perform cutting edge research in this field, therefore we experienced how difficult was to obtain such quantum mechanical effect. Besides the experimental difficulties, we realized that this experiment and in fact most of the real experiments make use of many other disciplines like, chemistry, material science and electrical as well as computational sciences.

Particularly in this experiment, we concluded that the knowledge of photonics play an important role, we observed how we enhanced the quantum dots emission by the use of photonic bandgap materials. We prepared quantum dots, which are the key element in this laboratory and also in many disciplines like quantum information and quantum cryptography.
Acknowledgments

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References