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Kristen Gram April 10, 2024

Even Dispersion of Single Photon Emitters for the Purpose of High Intensity Single Photon Generation

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Abstract

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By Kristen Gram

Reliable, deterministic single photon emission is a necessary criterion for many quantum information and sensing protocols. However, as of now, near-ideal single photon sources have not been realized. One of the most promising platforms for single photon generation currently is the semiconductor quantum dot, which can reliably emit identical single photons after excitation with a quantum yield >50%. By preparing thin, evenly dispersed films of semiconductor quantum dots far enough apart that collective effects are negligible, we can effectively convert coherent laser light into single photons within a large region (~100 μm^2- 1 mm^2). We can then design a setup capable of measuring the emission (and correlations) across this region to determine whether or not we have single photon sources. Although we were unable to conclusively show that we had single photon emission as we did not measure correlations between photons, adjustments to the setup to measure this should be relatively easy to implement in the future.

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“The probability of one receptor (rod) cell receiving more than one photon is very small. The experiments on human behavior therefore indicate that individual photoreceptor cells generate reliable responses to single photons.”

-William Bialek, on the ability of the human eye to sense single photons

Chapter 1

Introduction

1.1 Single Photon Sources: why

The discovery of the dual wave-particle nature of light was an important catalyst for the quantum revolution of the 20th century. Likewise, the concept of bosons (including photons) as discrete excitations of the electromagnetic field was deeply important in the formulation of quantum field theory. Photons can mediate information transfer between matter particles, and matter can mediate information between photons. Single photons are particularly interesting physical objects. They are frequently used as qubits in quantum computing both because of their long coherence times and (being massless particle) because they move at the speed of light. To reliably perform quantum algorithms, it is necessary for a photon source to be as reliable as possible. This begs the question: what makes a good single photon source?

1.2 Single Photon Sources: theory

The criteria for an ideal single photon source are relatively straightforward.[1] Photon emission must be deterministically controllable. In other words, the user of a single photon source must be able to instruct the source to emit or not emit – it should not emit otherwise, and it should always emit when instructed. Only single photons should be emitted at a particular time, not multiple. Emitted photons should be indistinguishable – all emitted photons should have the same intrinsic properties- frequency, polarization, and wave packet spread in space/time.[2,3] Additionally, the user should always be able to generate photons at a rate that is limited only by the pulse duration. In reality, no single photon source will be 100% ideal- it will always have some nondeterministic properties.

Crucially, only one photon should be emitted at a particular time, not multiple. This is referred to as "antibunching". This criterion requires a high level of time resolution – it can be tested, to a reasonable level, by calculating the second-order correlation function between photons. Experimentally, this is done using a Hanbury-Brown and Twiss interferometer (Figure 1).[4]

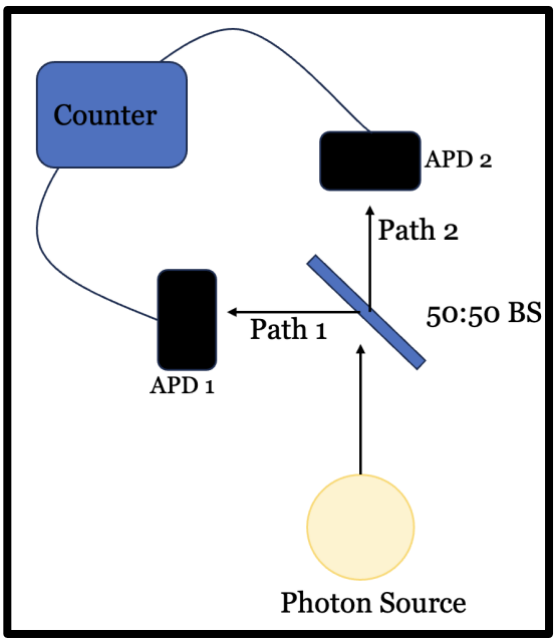

Figure 1: Diagram of a Hanbury-Brown and Twiss Interferometer. Photons travel from the photon source to a 50:50 beam splitter, which then forces them to travel along either path 1 (length r_1) or path 2 (length r_2). They are then detected with a single photon detector, typically an avalanche photodiode (APD)

The measurement and subsequent analysis are commonly referred to as a "$g^{(2)}$" measurement, as the correlation function (when using a Hanbury-Brown Twiss interferometer setup) is

$$g^{(2)}(\vec{r_1}, \vec{r_2}, \tau = t_2 - t_1) = \frac{\langle \hat{n}(\vec{r_1}, t_1)\hat{n}(\vec{r_2}, t_2)\rangle}{\langle \hat{n}(\vec{r_1}, t_1)\rangle\langle \hat{n}(\vec{r_2}, t_2)\rangle} \quad (1)$$

where $\hat{n}$ is the photon number operator; $\vec{r_1}, t_1$ and $\vec{r_2}, t_2$ are the distances and times measured between the photon source and the detectors for the first and second light paths respectively; and τ is the measured time difference between detections at the first and second detectors. If a source is emitting single photons, $g^{(2)}(\tau)$ must be nonzero everywhere other than $\tau = 0$. This makes intuitive sense, as a single photon cannot travel along both light paths at once, so there must be some time delay between subsequent photon detection.

1.1.1 Photon Number Operator

The Hamiltonian of the quantum harmonic oscillator is defined as

$$\hat{\mathrm{H}} = \hbar\omega_0 \left(\hat{a}^\dagger \hat{a} + \frac{1}{2} \right) \tag{2}$$

Where ω_0 is the base frequency of the harmonic oscillator, $\hat{a}^\dagger$ is the creation operator, and $\hat{a}$ is the annihilation operator. As their names suggest, these operators correspond to the creation and annihilation of bosonic particles or quanta of energy. $\hat{a}^\dagger \hat{a} = \hat{n}$ defines the number operator, meaning that the Hamiltonian can also be written as

$$\hat{\mathrm{H}} = \hbar\omega_0 \left(\hat{n} + \frac{1}{2} \right) \tag{3}$$

The eigenstates of this Hamiltonian are number states, labeled by an integer n. The number of photons present in some region of an electromagnetic field is given by this integer. Then, for photons to be anti-bunched, the expectation of the number operator $\langle \hat{n} \rangle$ must be zero for either detector 1 or detector 2 when $\tau = 0$.

1.3 Quantum Dots

There have been many proposed ways to produce near-ideal single photons sources. In principle, single photons can be reliably produced by any radiative source; however, it is generally easier to produce single photons if the source is effectively a two-level system, with a ground state and an excited state separated by a definite energy. (Interestingly, this is also the criteria for a qubit!). While single atoms themselves fit into this category (due to their discrete energy levels), they are not easy to work with due to their small size, relative instability at room temperatures, and complex setups required

for control.[5] Nanoscale condensed matter emitters that behave like atoms, on the other hand, avoid these disadvantages.

Quantum dots (QDs), referred to as "artificial atoms", have been created in a number of different media. QDs are several orders of magnitude larger than atoms, and they can be extremely stable and easy to work with at room temperature. There are several types of QD, including the semiconductor nanocrystals whose creators were recently the recipients of the Nobel prize in chemistry.[6] The basic principle behind the atom-like properties of QDs is quantum confinement. As the size of an object made from bulk material (i.e., containing many atoms) decreases, it approaches the Bohr radius of a bound electron-hole pair, also known as an exciton. The exciton can then be "trapped" in the potential well defined by the crystallite.

1.4 Excitons

Excitons form in semiconducting materials when an electron is promoted from the valence band to the conduction band, leaving behind a "hole" or excess positive charge in the valence band. Excitons are usually created in semiconductor QDs after the QD absorbs a photon with energy equal to or larger than the spacing between the permitted energy levels of the semiconductor material. Excitons are described by the Hamiltonian

$$\hat{H} = -\frac{\hbar^2}{2m_h}\nabla^2{}_h - \frac{\hbar^2}{2m_e}\nabla^2{}_e - \frac{e^2}{\epsilon|r_e - r_h|}, \tag{4}$$

Where m_h is the effective mass of a hole, m_e is the effective mass of an electron, ϵ is the dielectric constant of the semiconductor, and $|r_e - r_h|$ is the relative position of the hole and the electron.[7] The third term in this Hamiltonian (the potential energy term) can be neglected for small particle sizes, due to screening.

In the case of semiconductor nanocrystals, the exciton Bohr radius is on the order of 10 nm; any isolated crystallites with diameters of ~2 nm-10 nm will begin to show the effects of quantum confinement[8]. This is because the semiconductor material acts as a potential well, and the exciton is preferentially located within the crystal. As the crystallite radius shrinks, uncertainty in the position of the exciton decreases, and higher momentum states become accessible. This means that the kinetic energy of the both the electron and the hole increases substantially.[9] Then, any excited states of a quantum dot are shifted to higher energy. In practice, this means that decreasing the radius of a quantum dot causes the emission spectrum to undergo a blueshift.

1.4.1 Recombination

Excitons recombine (i.e., the electron returns to the valence band/ground state) after a characteristic lifetime due to the presence of external/vacuum fields; typically, this recombination process is radiative, and the exciton energy is released in the form of a photon. This makes QDs, which typically host a single exciton at a time, good candidates for being ideal single photon sources. However, defects in the crystalline structure of a QD can open non-radiative pathways for the exciton to recombine and dissipate its energy. Non-radiative relaxation lowers the photoluminescence quantum yield – the ratio of emitted photons to absorbed photons - of a QD.[10] Recalling that one of the criteria for an ideal single photon source is that it can be deterministically controlled, if a QD "blinks" (does not release a photon after excitation) due to non-radiative relaxation, its efficacy as a single photon source is reduced.

1.4.2 Quantum Dot Shells

One solution that successfully reduces the rate of non-radiative relaxation is the addition of a "shell" on the outside of a QD, made from another semiconducting material

with a similar lattice structure and (for emission in the visible spectral region) a larger bandgap. QDs with this type of structure are typically referred to as “core-shell” type, with the core acting as the confining region (Figure 2). Because the core is protected from chemical changes and subsequent defects by the shell, core-shell type QDs typically have much higher quantum yields than core-only QDs, as well as lower rates of blinking.[11]

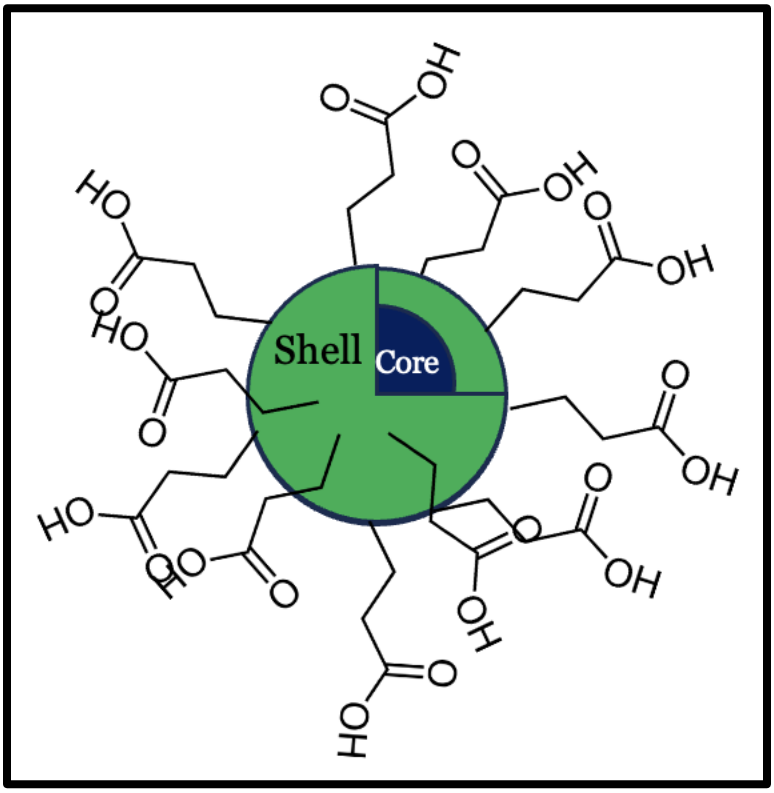

Figure 2: Schematic of a core-shell type quantum dot with surface functionalization.

1.5 Motivation

Although quantum dots themselves are not ideal single photon sources, they are good enough to be useful when designing setups for detecting single photon emission from

discrete points on a sample. This is because most semiconductor QDs are prepared as colloids, which can then be turned into thin films on top of a substrate. The eventual goal of this project is to design a platform for high-intensity single photon generation using hybrid exciton-polariton systems. We intend to couple quantum-confined (in one or more dimensions) materials that can support excitons to gold nanoparticles, which support plasmon-polariton modes. Polaritons are hybrid light-matter eigenstates of a quantum system – in this case, an exciton coupled to a cavity mode of light, with the gold nanoparticles acting as the cavity.[12,13] When a polariton is formed, the energy levels of the excitons are shifted. If the shift in energy is larger than the polariton linewidth, a phenomenon called "polariton blockade" occurs such that the system cannot be doubly excited. We hope to eventually examine whether plasmonic cavities can support polariton blockade without overwhelming loss of energy. If this is the case, we will be much closer to creating deterministic single photon sources and photonic transistors.

Chapter 2

Methods

2.1 Specifications

To achieve high-intensity single photon emission, it was necessary to use high quality quantum dots. We compared colloidal monodisperse quantum dots from two different sources, with approximately the same emission spectra and composition (Table 1).

	Sigma-Aldrich	Emory Department of Chemistry (Lian Lab)
Emission Peak	615 nm	600 nm
Core/Shell	CdSe/CdS	CdSe/CdS
Functionalization	Carboxylic Acid (4 nm)	n/a
Quantum Yield	>50%	Unknown
Solution	H_2O	Hexane (C_6H_{14})
Size	~20-30 nm	~15-20 nm
Concentration	1 mg/mL: ~ 10^{-4} M	Unknown

Table 1: Quantum Dot Specifications

2.2 Sample Preparation

To measure single photon emission, it was necessary to dispense the QDs evenly on a material that was transparent in the visible spectrum. We used standard glass coverslips for microscope imaging, cleaned using the following procedure:

a. Sonicate for 20 minutes in de-ionized water with industrial grade soap.

b. Sonicate for 20 minutes in de-ionized water.

c. Sonicate for 20 minutes in acetone.

d. Sonicate for 20 minutes in isopropanol.

e. Store in isopropanol, with a cover to prevent evaporation.

We compared three different methods to distribute single QDs on glass coverslips: drop-casting, spin-coating, and dip-coating.

2.2.1 Drop Casting

Drop casting is a standard technique used to disperse nanoparticles onto a substrate (Figure 3).[14]

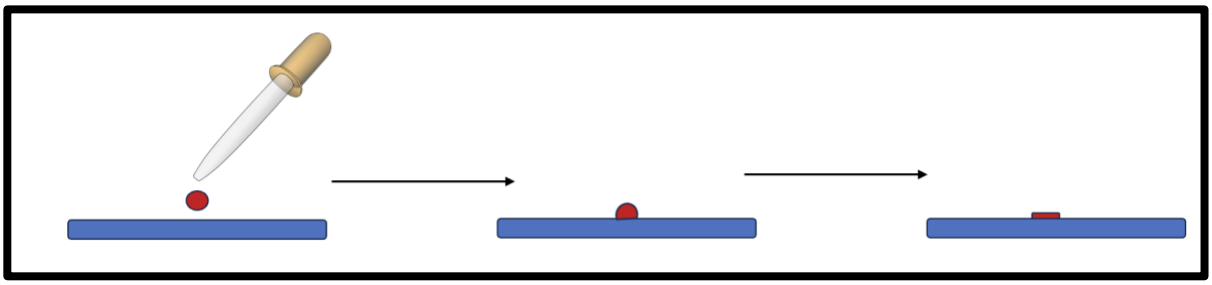

Figure 3: Drop Casting Procedure. A small amount (~10-100 µL) of liquid is dispensed on the substrate using a micropipette. It is then allowed to dry, typically under nitrogen flow, until all that is left is a thin film of nanoparticles.

However, it is notorious for leaving a macroscopic "coffee" ring, such that the concentration of nanoparticles is much higher at the edges than the center of the ring (Figure 4).

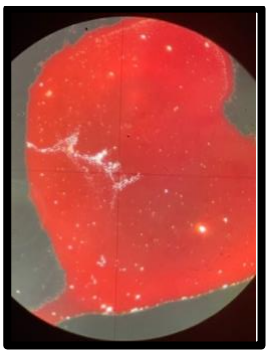

Figure 4: Dark-field scattering image taken of a region of the coffee ring of a drop-casted 0.1 mg/mL solution of quantum dots.

The strength of this effect is dependent on the surface tension of the solvent in which a colloid is dissolved. When the drop initially falls on the substrate, its edges will be "pinned", preventing the solvent from spreading out. Radial outward capillary flow driven by evaporation causes nanoparticles to concentrate at the edges of the droplet. This effect can be suppressed in many ways, including a reduction of the surface tension and an increase of the evaporation rate of the solvent.

Because the surface tension of water is quite high (~73 N/mm) compared to nonpolar substances like hexane, this preparation technique was inadequate for creating even distributions of the Sigma Aldrich QDs.[15] However, it provided a reference measurement for the emission spectra, as well as a demonstration of the effect of decreasing QD concentrations. The Sigma Aldrich QDs were labeled as having an emission peak at 620 nm, but the real emission peak was consistently closer to 610 nm.

2.2.2 Spin Coating

Spin coating is another method frequently used to prepare thin films on a substrate. The basic technique involves dispensing a small volume of liquid onto a substrate and spinning it at a high rate for a short period of time (Figure 5).

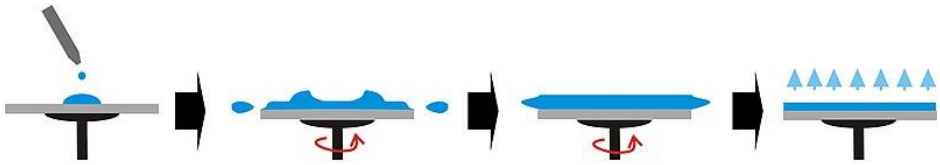

Figure 5: A schematic of the spin-coating process. Taken from Wikimedia Commons.

Generally, the goal is to produce a thin film of nanoparticles or polymers. Spin-coating is more effective when the colloid/polymer is suspended in a volatile solvent with the same polarity as the substrate, so the liquid will disperse quickly and evenly when spun.[16] One of the downsides of this technique is that much of the solution is wasted during the process; additionally, it is ineffective for aqueous solutions (i.e., it was not very useful for producing even films of the Sigma Aldrich QDs). There are several parameters that must be considered while performing spin-coating, beyond the material parameters: acceleration time, final RPM, total spin time, and method of dispersal (static vs. dynamic). Previous studies used solutions diluted to around 10^{-9} M: nanomolar concentrations.[17,18]

2.2.3 Dip Coating

The final technique that we used to produce films of QDs was the dip-coating method. The dip-coating method is one of the oldest commercial methods used to coat substrates/objects with thin films. However, there are several factors that make dip-coating an attractive method for preparing thin films of QDs from a solution. They both involve one of the most basic properties of liquid solutions: polarity. When a small volume of some non-polar substance is added to a larger volume of a polar substance (or vice versa), it tends to spread evenly over the surface. Additionally, Colloidal particles tend to spontaneously attach themselves to the

interface of two opposite-polarity solvents.[19] Taking these two ideas into account, we designed the following procedure to prepare even films of quantum dots using dip-casting:

a. Prepare ~50 mL of DI water or hexane in a 100 mL beaker.
b. Add 20 μL of a QD colloidal suspension of opposite polarity (dilute) with a micropipette.
c. Submerge one side of a cleaned glass slide in the liquid, taking care to keep the other side dry.

When the thickness of the layer of the solvent on top is close to the order of the diameter of the colloidal particles, it can be assumed that all colloidal particles will rest directly on the interface. Additionally, we can assume that colloidal particles will be spread evenly by the initial motion of the solvent. Then, the QDs that are deposited on the glass slide should be evenly distributed.

2.3 Light path Design

To characterize our QD samples, we needed to have (1) a method of excitation and (2) a way to detect the photoluminescence.

2.3.1 Excitation Path

We used a monochromatic 523 nm laser as a high-intensity photon source to excite both the Sigma Aldrich and Emory QDs. The laser driver used to power the picosecond laser diode head has both pulsed and continuous wave (CW) modes, with base frequencies of 80 MHz and 1 MHz in pulsed mode. A schematic of the excitation light path is shown in Figure 6.

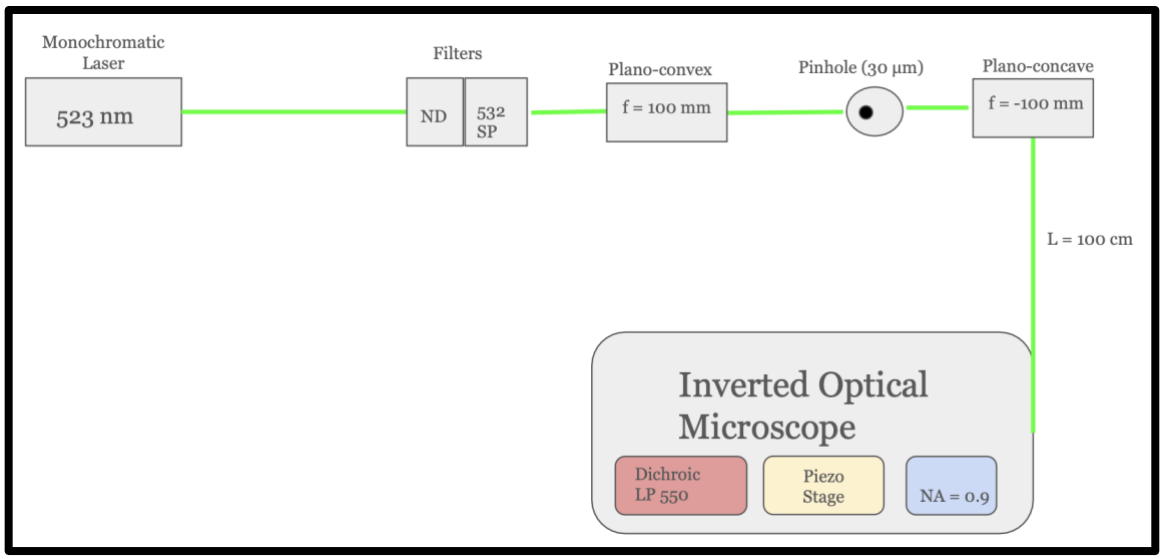

Figure 6: Excitation Light path

Notable features of the excitation light path are the neutral density (ND) filter, used to reduce emission intensity; the short pass filter, used to remove wavelengths longer than 532 nm from the excitation source (otherwise they may overlap with the emission spectrum); and the pinhole, used to filter out non-Gaussian modes of the electromagnetic field produced by the laser. The beam is focused and directed into an inverted optical microscope. The microscope objective used has a numerical aperture (NA) of 0.9- this means that the diffraction limited spot size of the Gaussian beam is approximately

$$d = \frac{\lambda}{2\,NA} = \frac{523\ nm}{2 \cdot 0.9} = 290nm. \tag{5}$$

This is much larger than a single quantum dot, so the emission spot should be spherical, and no features of the quantum dot should be discernible. To prevent the laser light from being detected as part of the emission, there is also a 550 nm long pass dichroic mirror (DCM) in the microscope. This can be easily swapped with a 50:50 beam splitter (BS).

2.3.2 Emission light path

A diagram of the emission light path is shown in Figure 7.

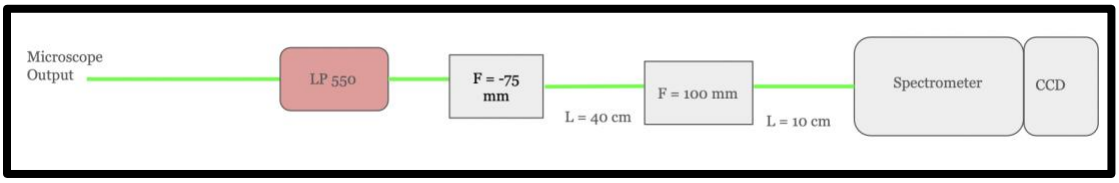

Figure 7: Emission light path.

The emission light path has an additional long pass filter to prevent any laser light from being detected. The emission is then focused and fed into a spectrograph with variable slit size and center wavelength. The emitted and wavelength-separated photons are directed to a sensor and converted to electric signals by a charge-coupled device (CCD). The CCD is capable of recording images or spectra in full vertical binning (FVB) mode.

Chapter 3

Results and Discussion

3.1 Drop Casting

Although the drop-casting method was ineffective as a method to prepare single emitters (see 2.1.1), the presence of a visible coffee ring allowed us to definitively measure the emission spectrum of the Sigma Aldrich QDs (figure 8).

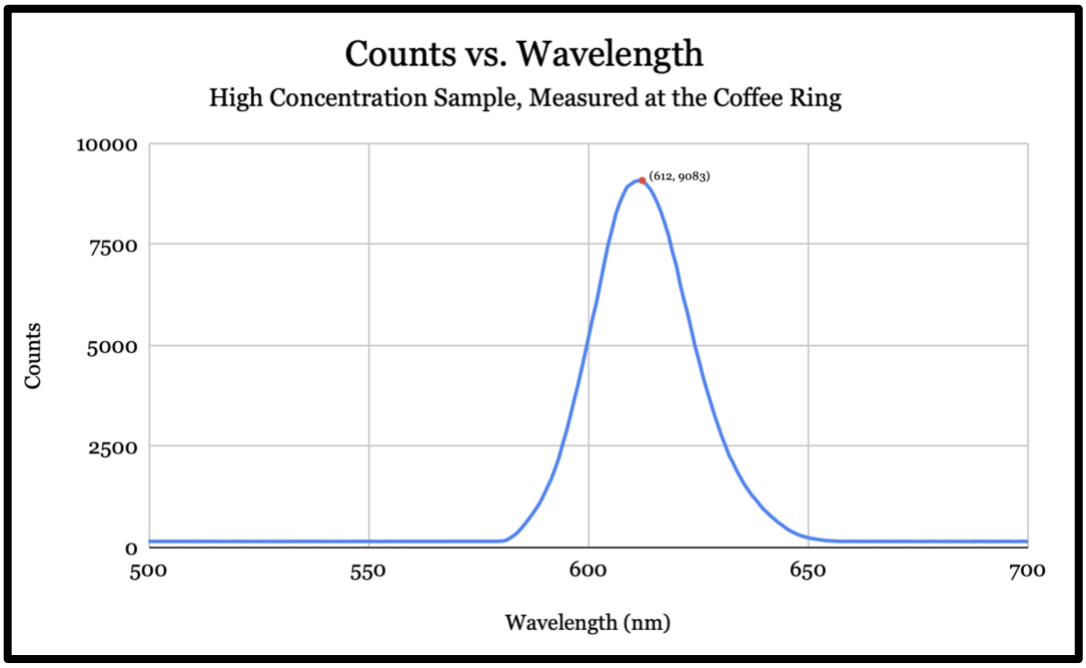

Figure 8: QD Spectrum for a drop-cast sample. The FWHM of the distribution is approximately 27 nm.

Additionally, we captured an image of the emission in the region inside of the coffee ring (Figure 9).

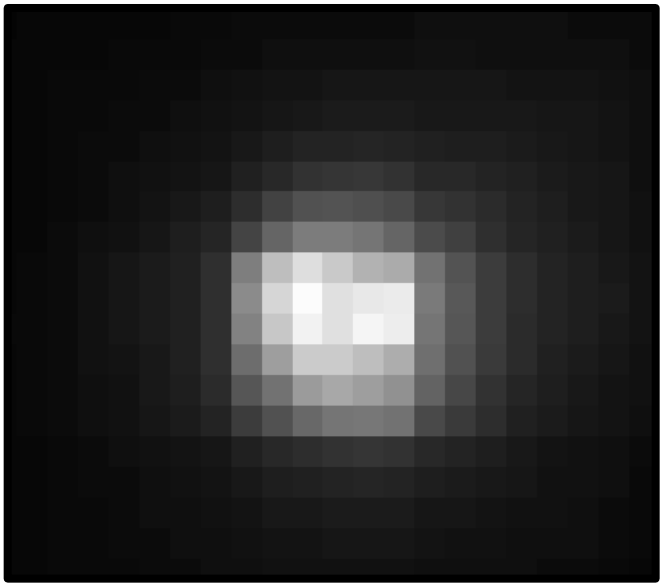

Figure 9: Image of drop-casted QD/QDs under continuous wave laser excitation. The diffraction limited spot size is 290 nm, and this corresponds to about Each pixel of the CCD camera corresponds to about 50 nm on the sample.

The near-uniform spherical shape of the emission suggests that the light is diffraction limited – the imaged PL source is likely a small cluster or homogenous film of quantum dots.

3.2 Spin Coating

The spin coating method allowed us to achieve isolated clusters of QDs. After diluting the Sigma Aldrich QDs 100 times, the concentration of nanoparticles in the solution prior to spin coating was on the order of 10^{-6} M. Spectra taken of the 1:100 dilution sample can be seen in Figure 10.

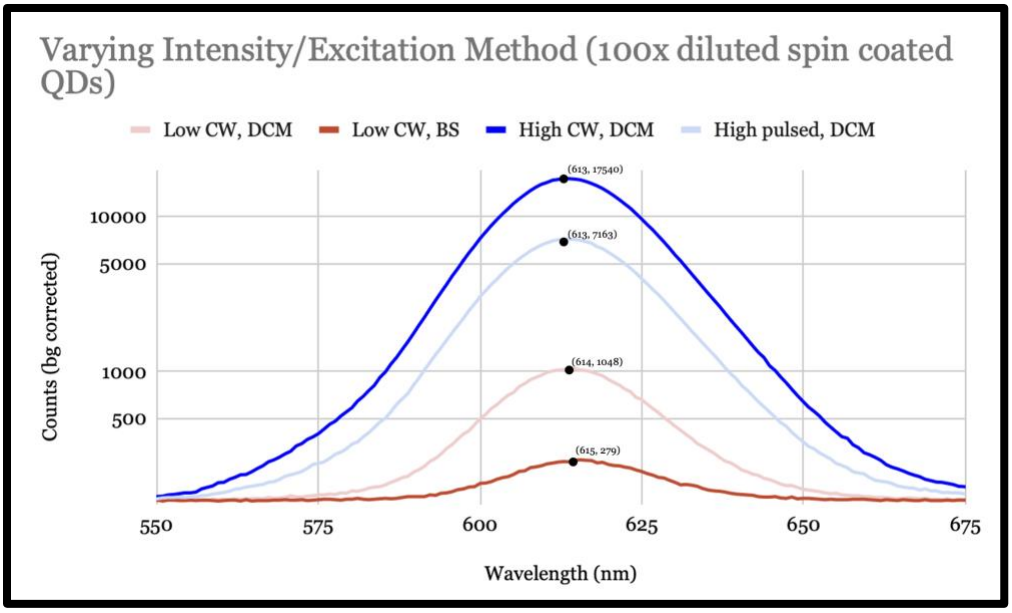

Figure 10: Log-Scale spectra taken for a spin-coated sample prepared with a 10-6 M aqeuous suspension of QDs. The emission peaks are consistently around 614 nm. Counts vary depending on both the intensity of the laser and the mirror used.

To replicate the results of prior studies and isolate single emitters (albeit without having an even distribution across the sample), we diluted the Sigma Aldrich QD solution to a molarity on the order of 10^{-9} M (Figure 11).

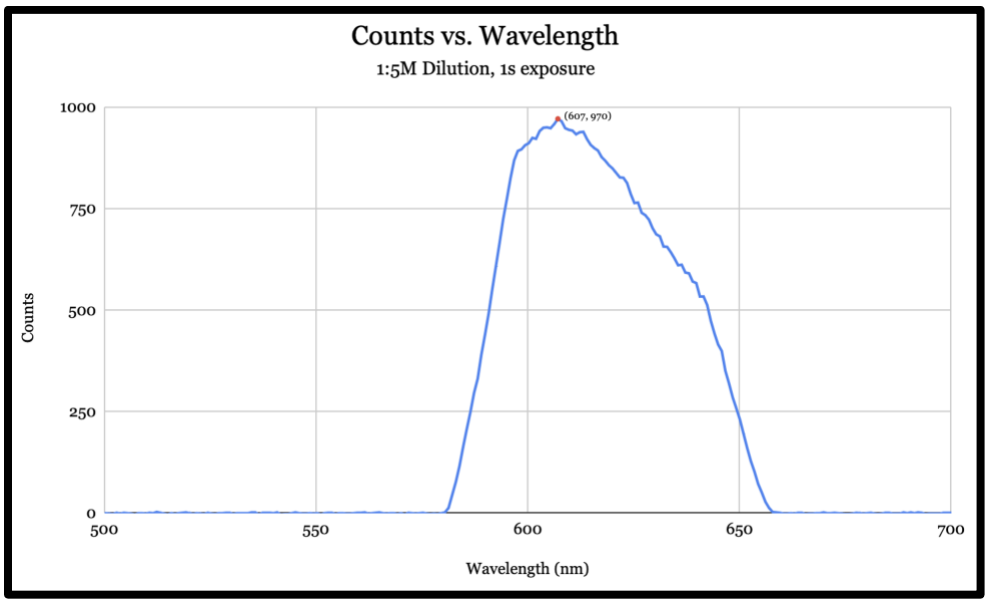

Figure 11: Emission spectrum taken for a 1:5M dilution sample. The emission peak is at around 607, slightly blueshifted compared to higher concentration samples.

Once again, though, it is unclear whether the source of the detected light is a cluster of emitters or a single emitter.

Another way to test whether single photon emitters are present, prior to taking a g-2 measurement, is to perform a scan using an avalanche photodiode (APD) . To do this, we moved the sample using a piezo stage and measured the changes in emission intensity using a single APD. This produces a 2D map of the emission coming from the sample in a given region. Preliminary scans were done on high concentration samples (figures 12, 13). We were unable to discern single emitters, although a strange wavelike pattern was present in a 25 μm^2 region of the sample.

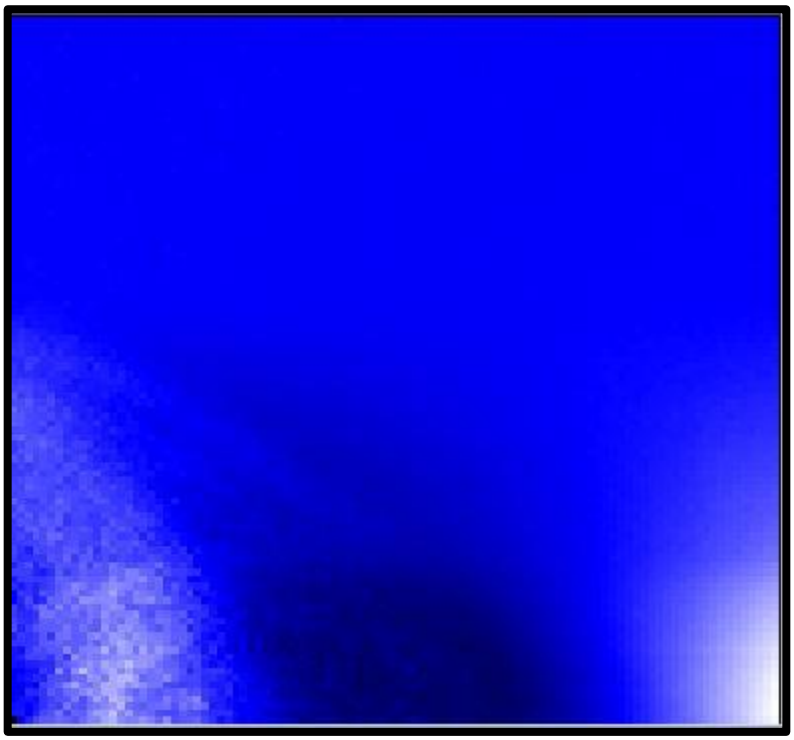

Figure 12: APD Scan of a high-density region of a spin coated sample. The scan area was 625 μm^2, and the scan speed was 5 ms/pixel. The maximum intensity detected was 60k counts/pixel.

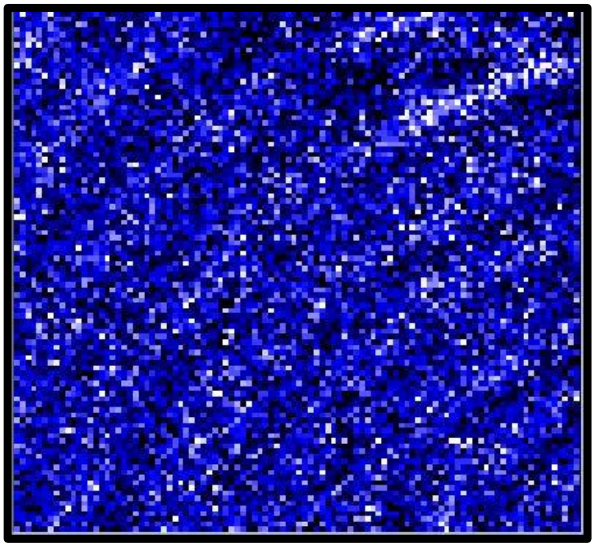

Figure 13: APD scan of a high density region of a spin coated sample. The scan area was 25 μm² and the scan rate was 10 ms/pixel. The maximum intensity was 8000 counts/pixel. The origin of the wave pattern is unclear.

3.3 Dip Coating

The dip coating method was the most promising for yielding an even distribution of emitters. We collected spectra and images for a 1:100 dilution dip-coated sample (Figures 14, 15, 16).

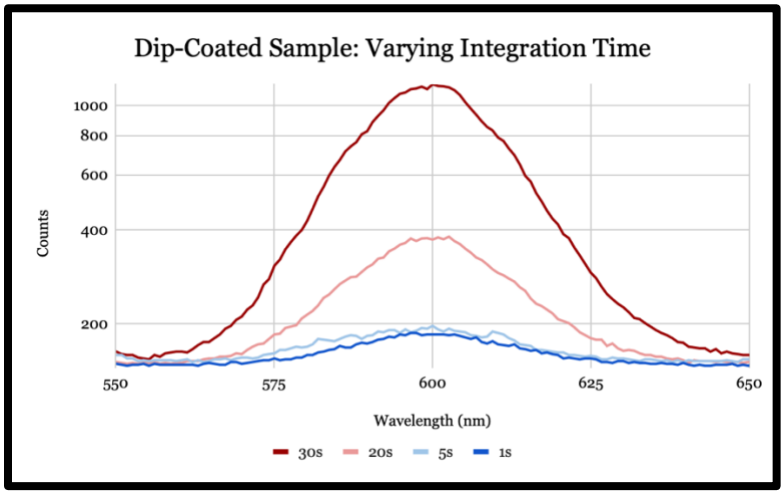

Figure 14:Log-scale emission spectra of a dip-coated sample.

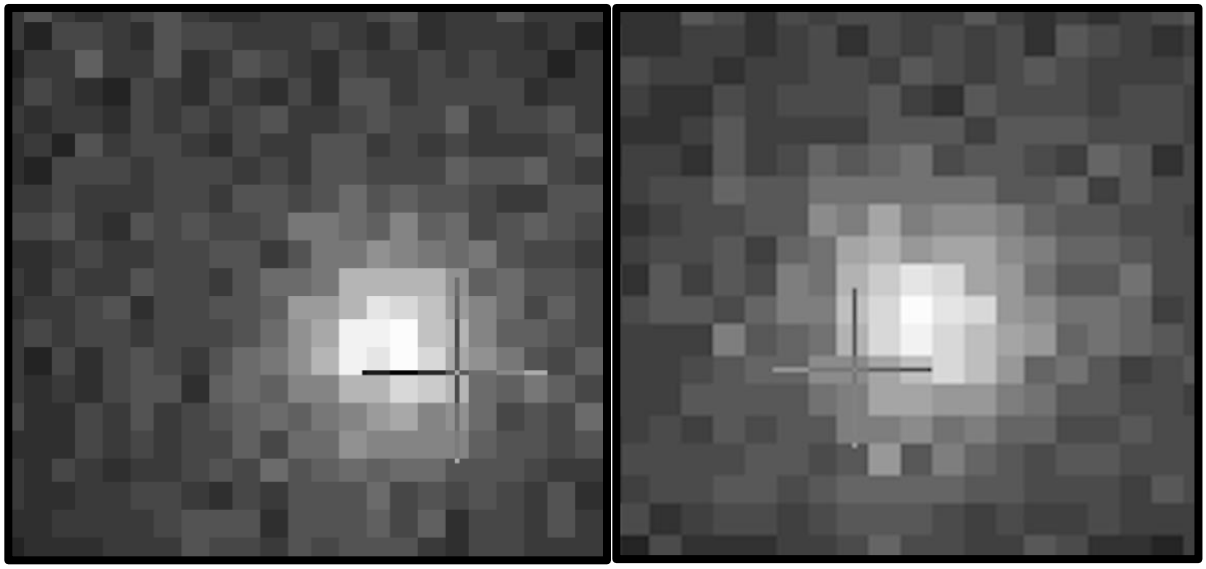

Figure 15: CCD image of an emitter, 1s exposure, 2 consecutive measurements

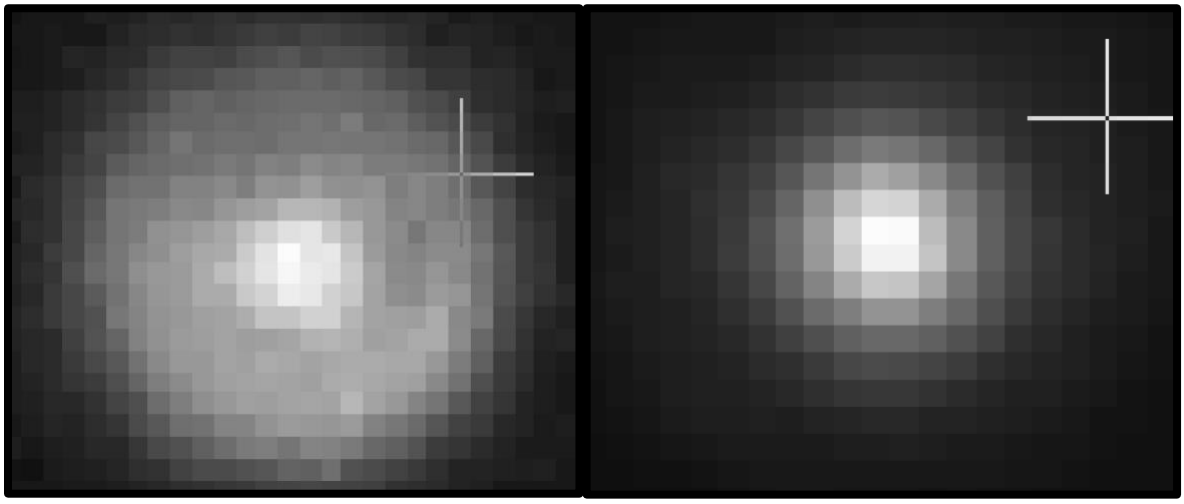

Figure 16: CCD Image of an emitter, 5 second exposure (left), 30 second exposure (right)

The images indicate that the emission is extremely spherical, so the source of photoluminescence is at least diffraction limited – either a single emitter or two emitters within a very small (<1/2 of the diffraction-limited spot size) radius.

3.4 Future Work

The only way to determine that a source is a single photon emitter and not a cluster is to measure second-order correlation. In the future, we intend to change the emission light path to a Hanbury-Brown and Twiss interferometer. Additionally, we intend to test the drop-casting and spin-coating methods on the Emory QDs in hexane, as the volatility of hexane makes these methods more effective. Finally, once we have demonstrated that we are able to reliably find and measure single photon emitters at the nanoscale, we will be able to begin examining exciton-polariton systems.

Conclusion

To create a proof-of-concept setup that can reliably measure single photon emission over a large (~μm^2-mm^2) region, we prepared thin films of quantum dots on glass. We used three different techniques to do this, including spin-coating, drop-casting, and dip-coating. Our results demonstrated that we were able to measure diffraction-limited photoluminescence, but it was not clear whether this was from single emitters or clusters. In the future, we should be able to determine this clearly by measuring second order correlations between photons traveling on different light paths.

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