Quantum Optics in Coupled Quantum Dots

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Mauricio Garrido
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This dissertation titled
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ABSTRACT

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Coupled quantum dots present an active field of study, both at the fundamental and applied level, due to their atomic and molecular-like energy structure and the ability to design and tune their parameters. Being single-photon emitters, they are systems that behave fully according to the laws of quantum mechanics. The work presented here involved the experimental study of the electro-optical properties of Indium Arsenide, coupled quantum dots. Initial experiments involved the use of spectroscopic methods such as photoluminescence and photoluminescence excitation (PLE). Through such techniques, the top dot’s hole energy level structure was mapped and different types of resonant absorption were identified. The characterization of these excited states and the knowledge of how to resonantly excite into them is an integral part of the development of certain controlled spin gates in quantum computation. Additionally, a shift of the spectra in the electric field was observed with varying excitation wavelength through and above the wetting layer, which allowed for direct measurement of the optically-created electric field within the device. This extends the quantum dots’ capabilities to using them as electric-field nano-probes and opens up the possibility of an all-optical, fast switching mechanism. In the course of these studies, a novel data visualization method for PLE in this type of system was developed. Finally, to study correlated photon effects, a Hanbury Brown - Twiss experiment was built which revealed bunching and antibunching signals
typical of quantum statistics in biexciton cascade emissions. This is an important step towards the experimental investigation of entangled states in coupled quantum dots.

Approved: ________________________________

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I humbly place this dissertation at the lotus feet of my beloved spiritual master, His Holiness Radhanatha Swami. It is for his pleasure that it has come to be.

To Mother Kamagiri Devi Dasi, Yajna Purusa Dasa, Balarama Candra Dasa, Jaya Jagannatha Dasa, Sri Kisore Dasa, Gauranga Kisore Dasa, Caitanya Dasa, Anandavidya Dasa, Vamsi Dasa, Emily Tessle (Tess), Jason Friggens, Narendran Siddan, Samantha Carey, Cameron Goodyear, Marian Thomas, Kevin Vaught and all the Columbus devotees. They brought me to my spiritual master. My eternal gratitude to them.

Hare Krishna Hare Krishna Krishna Krishna Hare Hare
Hare Rama Hare Rama Rama Rama Hare Hare
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I was born in the darkest ignorance, and my spiritual master opened my eyes with the torch of knowledge. I offer my respectful obeisances unto him.

*Bhagavad Gita as It Is, Introduction*

Within the last two decades, a great deal of attention has grown in semiconductor nanostructures for the wide range of possible technological applications and for the simplicity with which basic principles of quantum mechanics can be studied experimentally. Among these semiconductor nanostructures are quantum dots (QDs), which are sought after for their optoelectronic properties. QDs are semiconductor heterostructures whose size is on the order of nanometers, where charge carriers are confined in all three spatial directions to lengths which are smaller than their de Broglie wavelengths. Mean field theories like Hartree, Hartree-Fock and density-functional theories, being the simplest approach, are used to describe QDs by assuming the charge carriers to be moving independently within an average field created by particle interactions and external confinement. Empirically, the mean field is approximated by an effective potential. Due to quantum mechanics, the three-dimensional confinement leads to discrete energy levels much like the ones found in atoms, thus giving them the common-known name of “artificial atoms”.

Not only are the energy levels of the charge carriers in the mean field potential of the QD atomic-like in their being discrete; but they also present a “shell” structure analogous to atoms, where degenerate or nearly-degenerate levels bunch up together and
are separated from other bunches by energy gaps\textsuperscript{5} (Figure 1). This level bunching is a result of dimensionality and symmetry of the mean field potential, and is not exclusive to QD systems, but can be found in the properties of other finite quantum, many-fermion systems, such as conductance, ionization energies, etc. A high degree of symmetry, hence high degeneracy, results in a pronounced level bunching.\textsuperscript{9}

\textbf{Figure 1.} Level bunching. Qualitative illustration of the level bunching (shell structure) at the Fermi energy for different binding energies. The binding energy is lower, i.e. the particle is more bound, if the single-particle level density at the Fermi energy has a minimum. After Brack et al.\textsuperscript{9}.

Having the density of single-particle states at a minimum at the Fermi surface (as shown on the left of Figure 1) implies that the particles occupy states with a smaller energy on average, i.e. the system is more bound. Therefore, shell filling leads to particularly stable states, as in the atomic case.
One advantage of these ‘artificial atoms’ is that, by embedding them in a device structure, they can be controllably tuned through a wide range of charge states similar to ionic states of an atom.\textsuperscript{10}

There are differences, though, between atoms and QDs. New electronic and optical properties become present, because of phenomena arising from the semiconductor nature of the QDs. Unlike electrons in an atomic potential, electrons in a QD are within the conduction band of the semiconductor, which means that there will be interactions with the underlying lattice and with valence and core electrons, as well as modified Coulomb interactions with other electrons in the conduction band. Given the low electron density (~10 nm mean distance between electrons), the effective mass approximation can be used to account for these effects.\textsuperscript{5} This effective mass approximation entails viewing the conduction electrons in the QD as a separate interacting system with an effective mass $m^*$, and a screened Coulomb interaction between the conduction electrons with the static dielectric constant $\varepsilon$ of the semiconductor. The many-body Hamiltonian $H$ of the quantum dot decoupled from its environment is thus.\textsuperscript{5}

$$H = \sum_{i=1}^{N} \left( \frac{\vec{p}_i^2}{2m^*} + V(\vec{r}_i) \right) + \sum_{i<j}^{N} \frac{e^2}{4\pi\varepsilon\varepsilon_0|\vec{r}_i - \vec{r}_j|}$$

Where the first sum is that of the single-particle kinetic and potential contributions, and the second sum is that of the Coulomb interaction between the electrons.
Even though the effective mass approximation is a good initial approximation, there are additional contributions and approximations that must be added into the theory (such as hole-hole interaction, electron-hole exchange interaction, slowly-varying envelope approximation, dipole approximation, effective nuclear magnetic field approximation, phonon modes, hyperfine interaction, etc.) in order to describe observed effects like fine structure splitting, phonon-broadening of the spectral lines, spin interactions between the charge carriers and the lattice nuclei, etc.

It is clear that with more refined models and better experimental understanding many exciting, high-yield applications such as new generations of lasers, biological markers, memory devices, quantum computing, photovoltaic cells, and light-emitting diodes may be possible. This has provided a strong motivation for both academia and industry to endeavor toward a better understanding of QDs.

Of special interest for our research are Stranski-Krastanow self-assembled QDs. During epitaxial (layer-by-layer) growth in Molecular Beam Epitaxy (MBE) or Metalorganic Vapor Phase Deposition (MVPD), lattice spacing mismatch between the wetting layer semiconductor and the buffering layer semiconductor induces islands to spontaneously nucleate on the wetting layer, see Figure 2. The wetting layer can then be defined as the thin 2-dimensional semiconductor layer from which the QDs form. Current technology allows control of the thickness of the layers and the heights of the quantum dots with atomic precision. The exquisite control makes it possible to engineer the confinement, and thus, the energy levels. Having this ability to tune the discrete energy levels of the quantum dots is one reason they are so popular. To probe these energy levels, we study the exciton states through optical spectroscopy. Excitons, or electron-hole pairs, are formed when an electron is excited by optical means from the
valence band of the QD into the conduction band. The relaxation of the pair into the lowest available states and subsequent recombination of the electron and the hole into a photon, allow the mapping of the discrete energy levels present in the QDs from the detection of these emitted photons, or photoluminescence (PL). Furthermore, information on the spin and charge dynamics of the electron and hole can be inferred from these studies through energy shifts and polarization.

One commonly studied type of S-K self-assembled QD is that formed with InAs/GaAs. A wetting layer of InAs, with its 0.42 eV direct band gap in bulk, can form quantum dots on a GaAs buffering layer, with its 1.52 eV direct band gap. Using the “indium flush” growth method, the height of the quantum dots can be controlled; leaving the QDs looking pancake-like as seen in Figure 3 right. In this method, the quantum dots are capped to the desired height after growing a thin layer of GaAs on top of them and annealing them for determined periods of time. Heights range typically
around 2-5 nm, while the lateral dimensions of the QDs (disk diameter) ranges around 10-20 nm, see Figure 4.

**Figure 3.** XSTM and AFM images of QDs. After Z. R. Wasilewski, et. al., J. Cryst. Growth 201/202, 1131 (1999).

**Figure 4.** QD structure. Right: Physical appearance of QD. Left: Band edge diagram of QD embedded in a Schottky diode. Applying a voltage changes the height of the energy levels within the QD with respect to the Fermi level, thereby allowing charges to tunnel from the n+ contact to the QD.
Charging the QDs can be done by embedding them in an electric-field-tunable Schottky diode structure. By applying specific biases, we can control the height of the gap at the Schottky contact relative to the n+ contact, thereby controlling the height of the Fermi level and hence, the number and type of charges inside the QDs \(^{10}\) (see Figure 4 left).

Upon growing a second layer of QDs over the first layer of QDs with a barrier layer of GaAs in between them, a top QD will tend to form right above where a QD has formed in the bottom layer due to the strain field created by the bottom dot (see Figure 5 upper right). Being able to easily tune the energy levels of the top and bottom QDs into and out of resonance by means of an applied bias in the Schottky diode structure (see Figure 5 upper left and lower), spectroscopic studies have revealed a coupling between these two QDs, which give them a “molecule-like” behavior.\(^{23}\) Hence, they are often referred to as Coupled Quantum Dots (CQDs) or Quantum Dot Molecules (QDMs).

Significant advances have been made in the study of single QDs (SQDs). Optical quantum gates \(^{24}\) and entangled-photon-pair sources \(^{25}\) through the use of SQDs, by means of multiexciton spin and charge states, have already been implemented and are being further developed. CQDs with their tunable resonance energy levels between their top and bottom QDs offer new alternatives in terms of control mechanisms which are now being studied.\(^{26}\) This opens up a world of possibilities in regards to tunable quantum mechanical interactions between physically separated entities, which may eventually translate into the development of scalable building blocks for quantum information technology.
Figure 5. CQD structure. Upper Right: Physical appearance of a set of CQDs with a 4nm GaAs barrier between them. Upper Left: Band edge diagram of CQDs. Lower Right and Left: Hole energy levels in bottom and top dot can be brought into and out of resonance by application of an external electric field that moves the Schottky contact energy level with respect to the one of the n-doped GaAs.

The aim of the work presented here is to show the advancements to the experimental study of the electro-optical properties of CQDs done by our group at Ohio University. The section on Photoluminescence Excitation includes results and text from two publications. The first followed from the observation and characterization of excited states (Garrido, M., Wijesundara, K. C., Ramanathan, S., Stinaff, E. A., Scheibner, M., Bracker, A. S. & Gammon, D. Characterization of the Shell Structure in Coupled Quantum Dots through Resonant Optical Probing. Mater. Res. Soc. Symp. Proc. 1117E, 1117-J05-03 (2009)). The second involved the study of photovoltaic effects within the devices (Garrido, M., Wijesundara, K. C., Ramanathan, S., Bracker, A. S. & Gammon, D. Electric Field Control of a Quantum Dot Molecule through Optical Excitation. Appl. Phys. Lett. Accepted). The final experimental section discusses Time-Domain experiments and the investigation of correlation effects in the emission from CQDs.
The splendor of the sun, which dissipates the darkness of this whole world, comes from Me. And the splendor of the moon and the splendor of fire are also from Me.

_Bhagavad-Gita as It Is, 15.12_

Beginning with an empty room our research group quickly built up a full-working laboratory capable of a multitude of spectroscopic experiments on single CQDs. Throughout this period, we have been constructing experiments to extend the knowledge of the physics of these and other systems as well as the techniques used to probe their properties. Important milestones have been achieved in the development of the laboratory including: (Polarized) Photoluminescence, Photoluminescence Excitation, and Time-Domain Experiments.

_A. Photoluminescence_

_Thus the original creation is directly from the Supreme Personality of Godhead, or Parambrahman, and the secondary creation, as a reactionary result of the original ingredients, is made by Brahmā. Thus the activities of the whole universe are started._

_Srimad Bhagavatam, 2.10.3, Purport_

The first step was to set up a system that would recreate the basic experiments that would bring our capabilities to the level of doing what other groups were presently doing. This meant obtaining photoluminescence from individual CQDs.

As mentioned in the introduction, the light emitted from the sample coming from the recombination of the optically excited electron-hole pair is called photoluminescence
Depending on what energy levels the charge and spin states of the exciton were in before the recombination, the resulting photons will have different energies corresponding to the different states, thereby creating a spectrum. By spectroscopically studying this emission, information related to the charge and spin states created in the QDs can be inferred.\textsuperscript{10, 27}

**Experimental setup**

CQDs samples have been provided by collaborators at the Naval Research Laboratory (NRL) in Washington, DC. These are InAs/GaAs S-K self-assembled CQDs, embedded in the insulating region of an n-I Schottky diode. Using MBE techniques, they were grown to heights of \(~2.5\) nm, with barrier thicknesses between the dots of 2, 4, and 6 nm. The top electrical contact is made up of a semi-transparent layer of titanium to provide a uniform electric field throughout the sample, and an 80 nm aluminum shadow mask with 0.2-1.0 µm apertures to be able to focus on single CQDs, so that relevant shifts in PL lines due to charging and multiexciton effects from single dots (on the order of a few meV) are not obscured by inhomogeneous line broadening arising from observing multiple CQDs at the same time (around 50-100 meV).\textsuperscript{28}

PL is detected using a Trivista triple grating spectrometer manufactured by Princeton Instruments, with a maximal resolution of \(~0.006\)nm (at 600nm). The detector is a liquid-nitrogen-cooled, charge-coupled device (CCD) with a detection wavelength range of 200 to 1050 nm.

In order to create excitons in the CQDs, a continuously tunable (700-1000 nm), continuous-wave/pulsed (100 fs - 4 ps) Spectra-Physics Ti:Sapphire laser is used.
In addition to broadening the PL lines, phonon scattering introduces unwanted dephasing and relaxation mechanisms into the system. For this reason, it is desirable to cool the QDs to temperatures ~20 K with the help of an Advanced-Research-Systems closed-cycle cryostat. Because vibrations can severely hinder the detection of single nanostructures, this cryostat also allows us to keep vibrations ~ 1μm.
Figure 6. Lab set up and experimental schematic.


Bias maps and CQDs

Because of the discrete nature of the energy levels in the QDs, the spectrum will be made up of peaks representing the different states of the QDs. Since there exist other peaks coming from other sources, like nearby QDs or stray light, most spectroscopic studies made on QDs have been devoted to accurately recognizing those peaks coming from a single QD and what information they convey from the quantum state of the excitons in the QD. One very useful tool developed for this recognition is the bias map (See Figure 7).

Figure 7. Bias maps of SQDs and CQDs. Left: Neutral excitons X0, positively charged excitons X+ and negatively charged excitons X- are identified in SQDs as straight lines that are quasi-independent of the electric field (the small dependence comes from the quantum confined Stark effect). Right: Anti-crossings and lines with strong electric field dependence are optical signatures of interactions between the top and bottom dots of the CQD (tunneling and indirect recombination, respectively). After Stinaff et al.23
By taking spectra at different voltage biases (i.e. at different electric field strengths), the energy levels in the QDs shift, giving rise to changes in the quantum state of the excitons and thus in the peaks’ height and position. Bias maps are contour plots of the bias versus PL spectra energy in the X and Y coordinates, while the PL spectra intensity in the Z coordinate is represented by a color gradient. Changes in peak height and position are thus much clearer to see. Instead of comparing minute differences in peak heights and positions in two PL spectra (intensity vs. energy plots) at a time for a series of biases by superposing the plots or going back and forth between them; bias maps allow us to do the same simply by pattern recognition. Thus, the appearance, disappearance and shapes of lines in the bias map will easily indicate different states and their dynamics.

In SQDs, the PL energy as a function of applied field show up mostly straight with a slight shift. This shift is an indication of the quantum confined Stark effect. In essence, when the electron and hole in the QD are subjected to an external electric field, the center of mass of each particle is shifted to opposite barrier walls that define the QD. This polarization creates a small, electric-field-dependent dipole, in addition to the built-in dipole, that will shift the energy levels of the exciton, and thus, also of the recombining photon seen in the bias map. Due to different Coulomb interactions, we can see different charge states appear as different lines. For example, when the dot becomes charged with an additional electron, the PL emission shifts to lower energy by approximately 6 meV. The appearance of this negatively charged exciton line (sometimes referred to as a negative trion) coincides with the disappearance of the neutral exciton line.

In CQDs, interesting features are added to the bias maps. Inter-dot (indirect) transitions (i.e. transitions in which the electron is in either the top or the bottom dot, and
the hole is in the other dot) are recognized by strongly electric field dependent (diagonal) lines, compared to the weakly electric field dependent (straight) lines produced by intra-dot (direct) transitions (i.e. transitions within the same dot (top or bottom)) which show a weaker electric field dependence. An interesting feature in these CQD spectra is the x-form pattern, which arise from charged states. The x-pattern was shown to be due to the molecule-like behavior of the charged CQDs when their energy levels come into resonance and the carriers are given the possibility to tunnel between the top and bottom dots. The x-pattern is thus evidence for the transition from the state in one dot to another.

Figure 8. CQD dynamics. Upper Left: Energy diagram of a CQD. Right: Corresponding bias map where an x-pattern can be seen. Lower Right: Hole level resonance. After Stinaff et al.²³
As seen in Figure 8 upper left and right, the x-pattern comes from the direct (same dot) and indirect (different dot) recombinations of electron-hole pairs. The upper left figure shows the energy diagram of the initial (charged-exciton/black lines) states, the final (hole/red lines) states, and the type of recombination between them (blue lines), which give rise to the x-pattern seen on the right figure. An important point to note is that the element that gives rise to the x-pattern (i.e. anticrossings at two different electric fields), as opposed to a single anticrossing at a single electric field, is the difference ($\Gamma^-$) in Coulomb interaction between the holes in the charged exciton. The Coulomb interaction between the two holes is larger when they’re close together in the bottom dot than the Coulomb interaction between them when they’re in separate dots. This makes the horizontal charged exciton line with the two holes in the bottom dot shift up in energy with respect to the diagonal charged exciton line with the holes in separate dots by an amount $\Gamma^-$. By doing this, the crossing point between the two lines is shifted to the left, giving rise to a second anticrossing point that creates the x-pattern we see.

The lower right figure is a schematic of the point at which the hole levels come into resonance, which corresponds to the crossing of the red lines in the upper left figure. Analysis of the fine structure in the bias maps’ patterns has also given insight into the spin fine structure of the QDs.\textsuperscript{31}

Another useful tool for studying the spin states and their dynamics is polarization. By circularly polarizing the exciting laser beam, its photons are selected to have either (angular momentum in units of $\hbar$) spin $+1$ or $-1$. The creation of the electron-hole pair must obey spin (angular momentum) conservation. Therefore, the electron will obtain a spin of $-1/2$ and the hole a spin of $+3/2$ for a spin $+1$ photon, while the electron will obtain a spin of $+1/2$ and the hole a spin of $-3/2$ for a spin $-1$ photon. The reason why this
is so, is clear from the fact that the electron, being an s-wave Bloch wavefunction, has a total angular momentum of 1/2 and can only be in a state of +1/2 or -1/2. The hole, on the other hand, being a p-wave Bloch wavefunction, has a total angular momentum of 3/2 and would be expected to be in any one of 4 states corresponding to spin states +3/2, +1/2, -1/2, and -3/2; but because of tensile stress between the GaAs and the InAs (due to the difference in their lattice constants), the valence band on which these states reside is split into two: a heavy-hole valence band and a light-hole valence band. The heavy-hole valence band allows only ±3/2 states, while the light-hole valence band allows only ±1/2 states. Since the heavy hole valence band is lower in energy (and thus defines the bottom edge of the band gap) and given that the energy splitting between discrete states derived from the heavy hole and light-hole valence bands is much larger than the orbital quantization energy, we can effectively neglect the ±1/2 hole states, while regarding the recombining heavy holes as particles with only ±3/2 states. Hence, the only possibilities for polarized photons to create an exciton are the above-mentioned states, as shown in Figure 9.

Inversely, the polarization of the light emitted from the QD will reveal what the spin state of the exciton was before the electron and hole recombined. Example: a photon with spin +1 will reveal that the recombining exciton was made up of a -1/2 electron and a +3/2 hole. Thus, it is possible to selectively excite the QDs into specific spin states by correctly polarizing the laser beam.
Utilizing the capability to obtain spectra from individual nanostructures, one of our accomplishments was a collaboration with a group at the University of Arkansas on a publication on the topic of quantum rings. Furthermore, being able to map out the ground charge and spin states of CQDs, one of the members of the group, Swati Ramanathan, presented work dealing with polarization memory in charged states for her Master’s Thesis, which also resulted in the group’s first publication in an international meeting’s proceedings. Another member of our group, Kushal Wijesundara, used this facility to study the exchange interaction in CQDs, resulting in another publication. Thus, the work done to set up the experiments and obtain polarized PL of ground state emissions, in addition to being very fruitful, provides the background for the next step, and one of the key results of this dissertation, excited states in coupled quantum dots. Being able to individually recognize and address the excited states in the CQDs is of great interest, and may provide a means to manipulate spin states necessary for many quantum computation schemes.
B. Photoluminescence Excitation

Inert matter is undoubtedly energy with potential to interact, but it has no initiative of its own.

Srimad Bhagavatam, 2.10.45, Purport

Having a map of the ground spin and charge states, it becomes possible to study the excited states in CQDs. A motivation for this is the fact that many logic gate and teleportation schemes have been devised in QDs through the use of excited charge and spin states.\textsuperscript{24, 38} Thus, information regarding the excited spin and charge states, and the ability to excite the QD to a desired state are required. For this we use photoluminescence excitation (PLE).

Usually, when mapping the ground states of the QDs, non-resonant excitation is performed; i.e. the laser wavelength is set such that the exciting photon will have enough energy to create an exciton above the QD energy levels (in the wetting layer or higher); see Figure 10. The reason for doing this is that exciting into such high energies ensures that the majority of photons will be absorbed, and thus, the QD structure may be seen after relaxation. By exciting into the wetting layer or higher, the electron-hole pair relaxes down to the ground and lowest occupied states of the QD, where it eventually recombines exposing the level it settled into. On the other hand, if the wavelength of the exciting photon is lower than the wetting layer and within the QD energy levels, given that the QDs energy levels are quantized, there may or may not exist an energy level at that specific energy on which to create the exciton and the photon may not be absorbed.

There are circumstances for which exciting into the QD is desired. If the ground states of the QD are known through the previous non-resonant PL, it is now possible to set the laser wavelength such that it will directly excite into a specific level above that
ground state, hence the name resonant excitation. The result of this action is then the creation of an excited state. (See Figure 10)

PLE studies consist of stepping the laser wavelength in small increments and detecting the PL emitted by the QDs. Because the laser will not be hitting an energy level at most wavelengths, due to the quantized nature of the levels, most of the PL will not be showing anything. When the laser becomes resonant with an optically allowed energy level, an exciton may be created there. The exciton will then either relax into lower levels, or directly recombine and emit. This will show up as a peak in the spectrum at the energy of the excited state and at the energy of the lower energy states. PLE thus has the ability to map out excited states of the CQDs.

From the above discussion, it can be seen that the intensity of the PLE spectra depends on the three steps to PLE:

1) Absorption
2) Relaxation
3) Emission

Thus,

\[ I_{PLE} = P_{\text{Emission}} P_{\text{Relax}} P_{\text{Abs}} I_{\text{Exc}} \]

Where \( I_{\text{Exc}} \) is the excitation intensity and \( I_{PLE} \) is the intensity of the detected PLE, \( P_{\text{Abs}} \) is the probability of absorbing a photon at the specific energy it is sent (this depends strongly on the wavelength of the laser photon), \( P_{\text{Relax}} \) is the probability an electron-hole pair relaxes down to a lower energy level (this depends less strongly on the wavelength
of the laser photon), and $P_{\text{Emission}}$ is the probability that the electron-hole will recombine and emit a photon.

![Diagram](image)

*Figure 10.* Resonant and Non-resonant absorption in a QD.

PLE is, therefore, a powerful tool for studying the structure of the excited states of the QDs, their fine structure, carrier relaxation processes, and spin relaxation processes (essential for decoherence). To analyze the fine structure of the excited states and the spin relaxation processes, however, PLE must be combined with polarization dependence studies. Interesting results on SQDs have come from these types of experiments, such as the role of the symmetric and asymmetric parts of the electron-hole and electron-electron exchange interactions in splitting and mixing triplet states of an excited trion, and the importance of phonon-related processes for charge carrier relaxation.

The most common way to present results from PLE studies in SQDs is to plot a non-resonant PL spectrum of the QD at a specific voltage and attach a plot of intensity vs. exciting laser wavelength of a specific peak monitored in the PL, see Figure 11. The
first plot allows the identification of the states present at the given voltage. The second plot shows the dependence of the chosen state on the exciting laser wavelength, i.e. what the excited states of the chosen state are. As it was seen in the last section, bias maps played a major role in the clear identification of charge states in SQDs and CQDs, and in the dynamics of CQDs. With the use of the bias maps, results from PLE studies are more easily interpreted.27 Thus the way the data is visualized can play a big role in the progress of a study. In this spirit, a new way to visualize PLE spectra was sought. Different ideas from 3D plots to bias maps sequences (see Figure 12) were investigated.

![Typical PLE](image)

*Figure 11.* Typical PLE. A single peak at a specific wavelength from the PL spectra is chosen to be monitored. The variation of this peak’s intensity as the laser changes in wavelength is then plotted.

Besides analyzing how the entire PL changes as a function of the excitation wavelength, interesting physics can also be obtained by focusing in on specific transitions unique to the CQDs. From PL obtained with non-resonant excitation, inter-dot (indirect) transitions can be recognized. Thus, by setting the laser wavelength at such points to
create that specific state, bias maps can reveal information like tunneling between
different shell levels, e.g. between a top-dot s-shell and a bottom-dot p-shell (See Figure
13), from the detected PL of the intra-dot (direct) transition.

![Figure 12. Sequence of bias maps excited by different laser wavelengths to visualize differences in absorption.](image)

With the tools that were developed for creating bias maps for PL, a common data
plotting tool was implemented in the lab using Labview for plotting wavelength maps.
Wavelength maps are contour maps, similar to bias maps, where the bias axis has been
replaced by a laser wavelength axis. With this type of plot, several peaks from the PL spectra can be viewed at the same time. This cuts down the time in taking data (the dependence of several peaks can be taken in one run), enhances the perception of changes in peak heights (small color changes in the foreground are easier to perceive than small height changes in peaks), and allows easy comparison of different excited states of different charge and spin states for correlations.

Figure 13. Tunneling between shells. Left: (1) The QD is first excited into an indirect transition. (2) The hole tunnels from the top-dot s-shell into the resonant bottom-dot p-shell. (3) Relaxation to the bottom-dot s-shell. (4) E-h recombination and detection of PL. Right: Example of usage of bias maps to define laser wavelength and expected PL detection energy.

The following is an example of data taken using this imaging tool. (See Figure 14). The PLE wavelength map on the right of Figure 14 taken at -1.5V (as shown on the bias map on the left) easily identifies resonances for not just one single line, but for a few
at the same time, including the neutral, negatively, and positively charged excitons. Simultaneous and non-simultaneous resonances for different lines can be seen. Being able to monitor PL at such long wavelengths was a challenge.

Figure 14. Example of usage of bias maps (left) to define bias and expected PL detection energy in the PLE wavelength maps (right).

Fluorescence coming from the Ti:sapphire laser would present a background noise that could potentially drown the feeble PL signal coming from the CQD. High-pass filters in the laser’s path and additional low-pass filters in the detection path helped us reduce the laser background. Also, the use of the three stages of the spectrometer in
subtractive mode further eliminated laser background, allowing us to observe excited states that were in the order of a few meV from the emission (See Figure 15).

This was just one of the many obstacles in this specific type of experiment. Additional difficulties included very low number of photon counts, vibrations and virtual leaks from cryostat, drops in laser power as wavelength is swept, only partial automation of wavelength meter, and non-clear identification of apertures in sample. How these different challenges were addressed is explained in the appendix.

![Figure 15. Schematic of PLE setup using the triple stage spectrometer in subtractive mode.](image)

The resulting bias maps had good signal over background within reasonable periods of data-taking time. With this configuration we could not only take PL spectra for different wavelengths at a single bias, but also a whole bias map for different wavelengths. To be able to see the differences between these bias maps, jpeg “movies”
were created made up of bias map frames. This opened up a whole new way of analyzing PLE data from our experiments and allowed for significant advances in the lab.

By learning to interpret the appearance and disappearance of dots and lines in the PLE movies, two interesting physical phenomena were determined: a) the type of absorption that took place upon excitation and b) the shell structure of the top dot’s hole. The mapping of the shell structure made use and corroborated the findings published in Nature by Scheibner et al. through a different method (level anticrossing spectroscopy, or LACS). Additionally, by using PLE, unlike the purely PL based method, this research opens up the possibility of using various laser spectroscopy techniques including polarization, time-resolved measurements, and pump-probe experiments to investigate and ultimately control the excited states of these CQDs. This initial work resulted in a publication in the 2009 MRS Fall Meeting Proceedings.

In CQDs, not only are there excited states within each individual dot but also the possibility of interdot excitations and recombinations, therefore being able to individually recognize and address the excited states in the CQDs is of great interest, and may provide a means to manipulate spin states necessary for many quantum computation schemes. As introduced earlier, individual bias maps were taken at different laser energies and then stacked together and viewed one after another. This results in a sequence or movie which presents this multi-dimensional information in a clear and simple way, allowing for straightforward recognition of important and interesting effects. To compare the patterns observed in these PLE movies with measured PL emission, a static bias map with a wider range of PL energies is included in the PLE movie along with a moving marker (red line in Figure 16) indicating where the laser energy is for each PLE frame.
This technique for visualizing PLE has allowed for the relatively easy identification of various PLE signatures in CQD that would otherwise have been difficult to interpret. For example, determining whether a given absorption (or emission) was due to a direct or indirect exciton would be extremely difficult (Figure 17), unless additional tests are performed.

Figure 16. Selected frames from a PLE movie. Left bias maps show some of the observed PL signatures from the ground state of the neutral exciton as laser energy is tuned. The right bias map is static and taken at a wider range energy range at lower resolution. The blue square indicates the energy range of left bias maps. The red line is a the marker to show where the laser energy is for the indicated bias map on the left.
To understand the signature of a PLE image, we must consider the absorption, relaxation and emission. The emission is given by the form (straight for direct emission, or diagonal for indirect emission) of the PL of the monitored ground state. The absorption is given by the way the laser line intersects the PL of the excited state in the above-mentioned static bias map (see Figure 18). Relaxation to the final states can involve both tunneling and phonon mediated relaxation.

**Figure 17.** Limitations of typical PLE for CQDs. Left: Schematic bias map depicting PL from a neutral exciton. Right: Representation of a PL spectra (black spectra) at a single bias (electric field), a specific PL energy is selected (in this case, that of the neutral exciton). The intensity of this line is then monitored as a function of laser energy (red spectra). Blue diagrams on top are schematics of the state of the CQD system. Blue points represent electrons, red points represent holes. Dashes on the left side of these diagrams represent the bottom dot’s energy levels, while dashes on the right side represent the top dot’s. In such an experiment it would be difficult to discern whether the peaks came from the recombination of direct or indirect states. After Garrido et al.43

Direct exciton states, when a photon is absorbed within a single dot only, will shift only slightly due to the quantum confined Stark effect,44 therefore, direct absorption into a single dot results in a mostly bias-independent state, which in the bias map appears
as a nearly straight (single PL energy) bias-independent PL line. Therefore, a single straight laser line will intersect most of the PL of the direct states.

Therefore, the way the laser intersects the PL from the absorption state determines whether the PLE will be a continuous line, when exciting into a straight direct line (bottom Figure 18), or a series of points, when exciting into an indirect line (top Figure 18). The type of emission determines whether the continuous line or series of points of the PLE will be monitored along a straight line (direct recombination) or a diagonal line (indirect recombination). To show this, Figure 18 depicts $\lambda_{\text{laser1}}$ and $\lambda_{\text{laser2}}$ exciting into small sections of indirect states that then tunnel, relax and recombine in the direct ground state, creating a series of points that move as the laser energy is swept from $\lambda_{\text{laser1}}$ and $\lambda_{\text{laser2}}$. On the other hand, $\lambda_{\text{laser3}}$ excites almost entirely into a direct state that then relaxes and recombines in the direct ground state, creating a mostly continuous line, with the possible exception of a few gaps where the anticrossings are located in the PL from the absorbing state. Examples of direct and indirect types of PLE data are shown in the inset. Arrows draw attention to series of points in indirect absorption and gaps in a line in direct absorption.

In certain circumstances it is possible to observe corresponding emission in a non-resonant, broad-range bias map which may therefore be used to differentiate PL emission resulting from specific dots. Thus, we find that the PLE sequence displays emission along the entire ground state (independent of bias for direct emission, and dependent of bias for indirect emission) for a small range of laser energies. Since the state is discrete,
Figure 18. Schematic representation of the processes leading to some of the PLE features. Top: Indirect absorption. Bottom: Direct absorption.
excitation only happens when the laser energy matches the state’s energy, resulting in a single flashing, ground-state PL line in the movie, when the laser line crosses over the excited state’s PL line. We interpret the PL observed in the inset of Figure 19 at $E_{\text{Laser}} = 1316.01$ meV as such an intradot excitation.

Indirect transitions result in highly bias-dependent states since the energy levels of the top dot shift with respect to the bottom dot energy levels as the bias is changed. In a single PL bias map with non resonant excitation, this appears as a diagonal bias-dependent emission line, as represented in Figure 18. In a PLE experiment, the laser can excite into this level only when its energy matches that of the indirect state at a specific bias. Therefore, after tunneling and relaxation, this is seen as a single PL point in the bias map at the ground state emission energy, coming from the intersection of the diagonal bias-dependent PL line and the straight bias-independent laser line.

As the laser energy changes, the bias at which it can excite into the indirect state also changes, shifting the electric field at which the PL point is observed. Thus, a series of points is built up frame-by-frame, making it appear as a moving point along the bias-independent line of the direct, ground state emission (Figure 19). The PLE movie can now inform what type of absorption or recombination took place, simply by watching for flashing lines or moving dots in the PL.

Once these patterns are recognized, two benefits immediately arise: the dynamics of the states are better followed, and resonantly creating a specific direct or indirect state becomes just a matter of going back to the laser energy and bias at which the line or the point was observed.
Figure 19. PLE and LACS used to probe the shell structure of the top dot’s hole. Inset is a collection of PLE frames (all centered at 1291 meV) that show a series of points along the neutral exciton’s ground state energy (encircled by the magenta box) moving in tandem as the laser energy is changed. Arrow markers have been placed to better show the succession of points. When these points are plotted at the laser energy that created them, a series of lines is built up, which match the diagonal PL lines from the indirect excited states seen using LACS. It is interesting to note that the PL bias map taken at $E_{\text{laser}} = 1316.01$ meV is indicative of a direct recombination. After Garrido et al.43

**Excited State Identification through PLE**

We applied this knowledge to characterizing the shell structure of the top dot by creating a PLE movie of the energy range near the ground state of the neutral exciton in the bottom dot. Similar to the signature discussed in the previous section, a couple of
points appear along the neutral exciton’s straight bias-independent line, all moving in tandem as the laser energy is changed (See inset Figure 19). Therefore, it is clear that these come from an indirect absorption and a direct recombination. Also, from this last statement, it can be deduced that there must have been tunneling at some point in the dynamics of the created indirect neutral exciton to have ended up as a direct neutral exciton.

The points are inferred to have come from the indirect absorption into excited states of the indirect neutral exciton (i.e. shell structure of the top dot). This inference is based on two observations: the laser being so close to the ground state, and the separation between the points being roughly what would be expected between the different excited states of the top dot. To verify this assumption, LACS was performed on the CQD, where the electric fields at which the top dots excited states become resonant with the bottom dot are revealed as a series of anticrossings in the exciton PL as the electric field is increases. This not only shows the separation between the different excited states, but also their position in the bias map. Graphing the points where the laser created the PLE points, produced a series of lines that were then interpolated and seen to match the anti-crossings associated with the excited states of the top dot. This not only confirmed the assumption of the PLE signal resulting from the excited states of the hole in the top dot (Figure 19), but also helped identify each of the points with a specific excited state. One advantage of the PLE experiment is that, although the ground state PL where the level anti-crossings are observed becomes much weaker as the electric field increases making their recognition more difficult, the resonant excitation into the excited states results in relatively high signal intensities even for high lying excited states.
The study was extended by comparing the top dot’s hole’s shell structure for different barriers. The experiment was done for a sample with a 2nm barrier separation between the top and bottom dot (see Figure 20 and Table 1).

![Figure 20. PLE on CQDs with a 2nm barrier separation.](image)

Table 1: Energy separation between excited states in CQD samples with 4nm and 2nm barriers.

<table>
<thead>
<tr>
<th>(meV)</th>
<th>4nm</th>
<th>2nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>ΔE0-1</td>
<td>6.947</td>
<td>6.14</td>
</tr>
<tr>
<td>ΔE1-2</td>
<td>5.069</td>
<td>4.09</td>
</tr>
<tr>
<td>ΔE2-3</td>
<td>2.253</td>
<td></td>
</tr>
<tr>
<td>ΔE3-4</td>
<td>2.441</td>
<td></td>
</tr>
<tr>
<td>ΔE4-5</td>
<td>2.816</td>
<td></td>
</tr>
<tr>
<td>ΔE5-6</td>
<td>3.004</td>
<td></td>
</tr>
<tr>
<td>ΔE6-7</td>
<td>3.004</td>
<td></td>
</tr>
</tbody>
</table>
It can be seen that the energy separations for a sample with a 4nm barrier and a 6nm barrier are similar. This should be expected, since the CQDs of both samples have similar sizes, and it is their sizes which define the quantization (hence the energy separation between the levels) and not the barrier. As discussed previously, in addition to allowing for the clear identification of excited states that may not be observable in PL, this research opens up the possibility of using various laser spectroscopy techniques to investigate and ultimately control the excited states of these CQDs.

*Photovoltaic Effects Analyzed Through Indirect Emission*

The effects just described were seen at the end of the movies, where the laser energy was close to the ground state; but upon looking at the beginning of the movie, where the laser energy was close to the wetting layer (i.e. the 2-dimensional InAs layer where the CQDs form from and which acts like a quantum well), we noticed the entire bias map shifted in bias. This was an indication of the appearance of an electric field connected to the laser energy.

Even though the shifting effect was clear to see, accurately measuring it was a challenge. The first attempt was aimed at trying to determine the exact location of the anti-crossing points, so that definite initial and final points to measure the displacement in bias could be set. However, due to the different spin configurations joining at the anti-crossing and the indirect lines’ weak signal, broad peaks and their small non-linear dependence on the effective electric field; the location of the anti-crossing points were highly sensitive to error, even after fitting the peaks to Gaussian curves to account for inhomogeneous broadening.

In the end, the chosen method consisted in fixing an energy through which an indirect line would cut, fitting the indirect line to a straight line and determining the bias
at which it would cut it. This eliminated the problem with the multiple spin configurations joining at the crossing point. To minimize the error from fitting the indirect line with its small non-linear dependence on the effective electric field to a straight line, the bias map viewing program was added a function to draw a straight line between two points picked on the bias map. This allowed us to easily see how far along the indirect line we could take points to fit for which the indirect line remained linear. During this period we also experienced significant down time due to problems with the cryostat and having to resend it to the manufacturing company for testing and repairing.

After a year of hard work, the results were submitted and have now been accepted for publication in Applied Physics Letters.\(^45\) Of particular importance is the fact that even though this effect has been known to be produced in quantum wells,\(^46\) a quantitative approach to directly measuring the optically-created electric field \textit{in-situ} had not been available until now. This was an innovative way of using QDs as electric field probes inside nanostructures. By monitoring the interdot recombination associated with an electron and hole in different dots we were able to precisely monitor the internal electric field generated.

Due to the high linear Stark shift associated with the interdot exciton states, we can directly observe the effect of the optically created electric field as shown in Figure 21. In contrast, for an intradot exciton, we observe only a very small shift due to the quantum confined Stark effect. The average interdot “Stark” shift of 0.74 meV/kV·cm\(^{-1}\) (i.e. 20 meV/V) is up to 100 times larger than that of the intradot exciton. The large enhancement comes from the displacement of the electron and hole and is proportional to the distance between the two dots. Also, comparing the linewidth of the interdot PL lines in the CQDs to that of the WL (Figure 22), we can see the advantage of using the interdot
PL to measure small changes. As seen in Figure 21, any change in the electric field will result in a measurable shift in the emission energy of the interdot lines. Conversely, we can observe this as a shift in the electric field value at which the interdot line is equal to a specific PL energy (Figure 21). With this method we can measure changes in the electric field down to a few tenths of a kilovolt per cm.

As the laser is tuned in energy from below the WL to above, a shift of the electric field dependent spectra is observed (Figure 21), which varies with the energy of the excitation. This is observed as a shift in the spectra measured by the field dependent interdot recombination. Figure 24 shows the dependence on different laser wavelengths.

It is seen that for laser energies below the WL no shift occurs, that the shift increases with increasing laser photon energy, and that once the laser energy is above the GaAs band edge no further shift is observed. The points in Figure 24 were obtained by fitting the interdot PL peaks in the spectra for different applied fields at each laser energy. These peaks were then fitted to a line which was used to calculate the shift in electric field for an arbitrary PL energy. Power dependence curves show that, at a given laser energy, as the photon flux (laser power) is increased the electric field shift also increases and ultimately saturates (Figure 25). In this case, the saturation was measured to be around 1.2 kV/cm at an excitation wavelength just above the WL ($\lambda_e = 860.1$ nm). The same trend was observed in 8 different CQDs within the same sample, with an average saturation of 1.12 kV/cm ($\lambda_e = 860.1$ nm).
Figure 21. Bias shift in the PL of CQDs. Due to this optically generated electric field, a shift of $\Delta E$ to higher energy (or equivalently, in field $\Delta F$) for the interdot emission at a fixed applied field $F_A$ (fixed PL energy) is observed. After Garrido et al.\textsuperscript{45}

Figure 22. PL spectra of device. The narrowness of the interdot PL lines in the CQDs (lower right red circle), compared to the broadness of the WL (upper left red circle), make them ideal to detect small changes of 1 meV. After Garrido et al.\textsuperscript{45}
Figure 23. CQDs as probes for photovoltaic effect. Band-edge diagram (not to scale) of the sample structure showing intradot (solid blue arrow) and interdot (dashed blue arrow) recombination. The laser excitation generates e-h pairs in the top and bottom wetting layers (WLs) (red arrows). Because of the applied electric field, the charges tunnel and accumulate, generating an electric field. After Garrido et al.\textsuperscript{45}

We also observe in Figure 25 a dependence of the optically created shift on the applied field itself. At a single PL energy the positively charged exciton ($X^+$) is observed at a lower applied field than the neutral exciton ($X^0$), and it is seen to saturate faster than the $X^0$. With a given laser power and energy, say 50 mW and 860.1 nm, for an applied electric field of $F_A = 31.7$ kV/cm we find an optically created electric field of $F_{\text{optical}} = 1.4$ kV/cm; while for a higher applied field of $F_A = 48.0$ kV/cm we find a lower optically created field of $F_{\text{optical}} = 1.0$ kV/cm. The exact saturation point at these two biases
appears to be different, but the error at high powers is large enough that it is difficult to ascertain this with confidence.

The observed effects are consistent with photovoltaic band flattening,\textsuperscript{46, 47} which is produced by the ionization of the photogenerated e-h pairs within the Schottky diode. Basically, the shift of the PL in electric field indicates a local electric field which is the result of creating electron-hole pairs by means of optical absorption into the WLs and consequent tunneling of charges to opposite sides of the device (Figure 23). Due to the applied electric field, electrons will tunnel to one side of the device (bottom WL, doped GaAs, etc.), while the holes will tunnel to the other side (AlGaAs barrier, top contact of the diode, etc.). This will create separate charge accumulations of extra holes on one side and extra electrons on the other. As opposite sides of the device become charged, they begin to generate a local electric field opposing the applied electric field. To induce an opposing field, however, the ionized electrons and holes must not only separate, but remain within the device region. In our device, this is most likely done through trapping at impurity sites and materials boundaries (e.g. the AlGaAs/GaAs interface, the WLs, or the doped/intrinsic GaAs interface), while those not trapped will contribute to the measured photocurrent. Steady state equilibrium will be reached when the Coulomb interaction prohibits additional charges from tunneling. The optically generated field ($F_{\text{optical}}$) opposes the applied electric field; therefore it is now necessary to increase the applied field to achieve the same effective local electric field.

The optically generated field ($F_{\text{optical}}$) dependence on laser power, laser energy and the applied field is a complicated function of the various tunneling rates, recombination rates, generation rates, and density of traps within the device. However, since the photoluminescence excitation (PLE) intensity and the photocurrent are proportional to the
absorption of photons and tunneling of carriers out of the device we can deduce some qualitative trends. In Figure 24 we see that at \( \lambda_e = 880 \) nm there is a PLE signal but nearly no photocurrent, suggesting that a fraction of the absorbed photons, those which do not relax and recombine, are ionized and trapped, thereby contributing to \( F_{\text{optical}} \). As the laser energy is increased we observe an increase in PLE, photocurrent, and the optically generated field. Above the GaAs both the PLE and photocurrent increase dramatically indicating efficient absorption, ionization, and tunneling, however \( F_{\text{optical}} \) still saturates indicating a saturation of the trapping rate.

![Figure 24](image.png)

**Figure 24.** Left: Wavelength dependence of the optically generated electric field (left axis), device photocurrent (right axis) and PLE intensity (arbitrary units). The optically generated electric field increases as the laser is tuned above the WL, saturating after the GaAs. The photocurrent and PLE intensity show a significant increase only above the GaAs. Right: Photocurrent was taken at an electric field in between the neutral exciton and positive trion (where the shift was mainly observed), denoted by a vertical green dotted line in I-V curves. Brown vertical dotted lines denote electric fields where neutral exciton and positive trion were observed. After Garrido et al.45
The saturation in the power dependence, as seen in Figure 25, demonstrates a limit to the optically generated electric field most likely due also to the saturation of the trapping rate. When analyzing saturation limits for different excitation energies, Figure 25 shows that at a laser energy of $\lambda_e = 860.1$ nm (above the WL), the field shift appears to saturate at a higher limit of 1.4 kV/cm than at $\lambda_e = 892.0$ nm (below the WL), where saturation is observed at a reduced magnitude of 0.35 kV/cm. This is consistent with a large decrease in absorption (expected from the analysis in Figure 24, whereby exciting into the low energy tail of the WL the absorption is reduced, reducing $F_{\text{Optical}}$) and possibly a lower trapping rate.

The dependence of the optically created shift on the applied field, we attribute to the applied field dependence of the tunneling rates of the electron ($t_{e-s}$) and hole ($t_{h-c}$). At higher applied electric field, charges are less easily trapped and tunnel more easily out of the device, resulting in reduced charge build up at a given laser power, hence reducing the optically created field. Therefore, even though the entire electric field dependent spectra are seen to be shifting, the higher-field spectra are doing so at a slower pace. This is seen in contour plots such as Figure 21 as a shift and compression along the electric field axis of the PL lines. However, regardless of applied field we would expect the saturation point to be nearly the same which is consistent with the results in Figure 25.

Following up on this observation, lifetime measurements of the optically-created electric field were taken to quantify how long it takes for the field to create upon excitation and decay after it. As expected, the decay took much longer (~125$\mu$s) than the creation (< 2-3 $\mu$s—our system’s resolution), because of the long waiting times for the trapped charges to recombine or tunnel out of the device. This meant that possible
applications in electric field modulation could be performed in the kHz range. These additional results have been published in the 2010 MRS Fall Meeting Proceedings.48

Figure 25. Power dependence curves showing the shift in bias as a function of laser power for a wavelength above and below the WL. For excitation above the WL a clear dependence on the applied field is observed. The saturation occurs much more rapidly at lower applied field (F_A = 31.7 kV/cm) whereas, at a higher field (F_A = 48.0 kV/cm) the optically generated charges more easily escape to the device. Exponential curves were fitted to the data to better show the trends. Inset: Bias map showing where the 2 curves were measured at 860.1 nm. After Garrido et al.45

The need to measure the creation and decay times for the optically-generated electric field pushed us to advance the lab’s capabilities a step further to measure events in the time domain. This is a powerful tool and the topic of the last chapter of this dissertation.
C. Time-Domain Experiments

\[
\begin{align*}
evam kālo 'py anumitaḥ \\
saukṣmye sthaulye ca sattama \\
saṁsthāna-bhuktyā bhagavān \\
avyakto vyakta-bhug vibhuḥ
\end{align*}
\]

One can estimate time by measuring the movement of the atomic combination of bodies.

Time is the potency of the almighty Personality of Godhead, Hari, who controls all physical movement although He is not visible in the physical world.

*Śrimad Bhagavatam, 3.11.3*

Time is destruction, and all manifestations are to be vanquished by the desire of the Supreme Lord. That is the law of nature.

*Bhagavad-Gita as It Is, 11.32, Purport*

The time domain is a remarkable subject within quantum optics. For the systems at hand, it entails being able to measure and manipulate events on the scale of pico to microseconds. It allows us to understand and develop many interesting physical phenomena, such as lifetimes, temporal-correlations, tunneling times, entanglement, etc. Among these, entanglement is an interesting, strange and challenging phenomenon that is appealing both for fundamental and applied research.

Given that QDs are single-photon emitters, they may offer the opportunity to create on-demand, entangled photon pairs. In fact, a few groups have already demonstrated the ability to create these, albeit not on demand, on SQDs.25, 49 They have
used the radiative biexciton decay cascade, which has two possible decay channels. Each channel emits a photon pair and both photons in the pair will have the same polarization depending on the channel (see Figure 26).

![Figure 26. Biexciton cascade. There are two decay channels, each with its own specific polarization. The intermediate exciton states are separated by the anisotropic electron-hole exchange interaction. After Akopian et al.](image)

This scheme of entanglement between the two polarization states relies on the non-distinguishability between the two decay channels. The important factor here is to have the two intermediate exciton states be degenerate, so that there is no “which path” information that destroys the entanglement. The two exciton states are, however, naturally separated by the anisotropic electron-hole exchange interaction, which is a consequence of the non-perfect cylindrical symmetry in the grown QDs and is on the order of tens of μeV. The above mentioned groups have tackled this issue by applying an
external magnetic field to reduce this splitting and by spectrally filtering the incoming photons to detect only those which spectrally lie in the overlap between the two polarization states. There are inconveniences to these approaches, though. The first group requires the use of superconducting magnets capable of producing a few Teslas and is also suspected by others 49, 50 not to have truly demonstrated entanglement. The second group requires spectral filtering, which introduces randomness, thereby yielding low rates and non-event ready entangled photon pairs.

Because of their ability to tune the energy levels of the top and bottom dots into and out of resonance by means of an applied voltage, CQDs may provide a way to more easily erase the “which path” information and thus also not require spectral filtering. It is true that tuning the exciton energy levels into resonance does not get rid of the anisotropic electron-hole exchange interaction, for it is this that separates the observed, molecular-like bonding and antibonding states, 23 but Scheibner et al. 51 and studies in our group 35 have shown that the anisotropic electron-hole exchange interaction can be modified with applied electric field and recently Ramirez and Cheng 52 have theoretically shown that the tunneling effects in strongly coupled dots lead to a substantial reduction of optical fine structure splitting without any drop in the optical oscillator strength.

Looking in Figure 27 at the level diagram of the neutral biexciton cascade, we searched for the specific energies that satisfied the requirements of the biexciton cascade in Figure 26. In our CQD implementation, the two possible intermediate exciton states are the direct and indirect exciton, which according to the diagram, are almost degenerate near the anticrossing. The enlargement of this area shows that the anisotropic exchange interaction which splits the direct exciton into two parallel lines decreases as the direct exciton line continuously turns into the indirect exciton line. This means that most
probable sites to look for entanglement are those very near the anticrossing along the indirect exciton line. Since the biexciton is already a few times weaker than the exciton and especially at the electric field where the anticrossings can be seen, the initial state of the biexciton cascade was chosen to be along the direct biexciton line.

![Figure 27. Biexciton level diagram.](image)

(a) The calculated energy level diagram of the neutral biexciton-exciton cascade. The areas in the boxes $X$ and $XX$ have been enlarged to show the fine structure caused by spin exchange interactions in the vicinity of level resonances. (b) Extended electric field dependent optical spectrum of the QDM with neutral exciton ($X$) and neutral biexciton ($XX$) transitions. After Scheibner et al.51

To prove the entanglement between the two photons, one must know their quantum state to determine the probability that a photon pair is detected, for only then can tests for entanglement be applied. The quantum state of a system is fully described
by the density matrix, therefore, the above-mentioned groups both constructed it by means of quantum state tomography analysis, but chose different tests on it to make their claim. Quantum state tomography is a construction of the density matrix by measurement of the polarization state of the system. The polarization state of this two-photon system can be determined using 16 measurements of selected polarization combinations on two-photon temporal correlations. The reason for taking the polarization measurements on the temporal correlations is to make sure that the polarization being analyzed is actually coming from an exciton and a biexciton from the same cascade, i.e. the exciton we’re looking at actually came from the biexciton we’re also looking at. With these 16 polarized correlation measurements, the density matrix is built on the basis of polarization combinations as seen on Figure 28.

For the entanglement tests, the first group chose that of the largest eigenvalue, where a value >0.5 signifies the presence of entanglement, since the probability that a photon pairs exists in any given polarization state in an unpolarized classical source cannot exceed 0.5. The second group chose to apply the Peres criterion to the density matrix, which is a form of violation of Bell’s inequality where one of the off-diagonal matrix elements must be non-zero for entanglement to exist. Given that this parameter is bounded by the interval [0,0.5], with 0 signifying unentangled and 0.5 maximally entangled, we’ll use this second method to assess the degree of entanglement and also avoid the controversial questionings that the first group has had.

The question now arises as to what the experimental setup must be in order to measure polarization-dependent temporal correlations. Typically, temporal correlation measurements are made using the Hanbury Brown-Twiss (HBT) arrangement. This arrangement is characterized by a beam splitter (BS) that separates the signal light into
two beams that go to two different monochromators (MCs), i.e. one wavelength spectrometers (see Figure 29).

Figure 28. Example of graphical representation of the density matrix of a state as estimated by linear tomography. Upper plot is the real part of the density matrix, the lower plot is the imaginary part. After D.F.V. James et al.59

Avalanche photodiodes (APDs) are put at the end of each monochromator to detect single photons. Their signals are fed to a time to amplitude converter (TAC), which outputs the time difference between the detection of one APD and the other. Finally, this signal is fed to a multichannel analyzer (MCA), which creates a histogram of
number of coincidence counts versus the time difference between the detection of the two photons, i.e. whenever a signal from the TAC comes (meaning the two APDs have been triggered with a certain time difference between the two), the MCA adds one coincidence count to the bin in the histogram to which it has assigned the time difference indicated by the TAC. This histogram is proportional to the correlation function, which represents the probability that one photon arrives a certain time after the other one. This setup was first implemented to analyze the correlation function in SQDs by Regelman, Mizrahi, Gershoni and Ehrenfreund and showed that photon statistics in SQDs can be varied with excitation rate.

Figure 29. Hanbury Brown Twiss arrangement.

To measure polarization simultaneously, retarders and polarizers are placed between the beam splitter and the MCs, to allow only photons of a specific polarization to go through.

We began by setting up the HBT arrangement and trying to detect first the characteristic antibunching that is expected when two photons cannot be detected at the
same time. In our system, this translates to detecting in the two APDs two excitons that are not in a same cascade, since QDs are true quantum emitters and can only consistently emit one photon from one state at a time. Because the very low number of coincidence counts ranging from 5 to 20 counts per second depending on the TAC settings, the alignment of the optics, the exciton type and the excitation power and wavelength, one set of data typically took at least 4 hours to record. Additionally, the random drifting of the sample can sometimes make it quite difficult to maintain the number of counts steady and can thus make the runs take longer. The following plots are thus our preliminary data. Antibunching in CQDs was observed when detecting two neutral excitons (i.e. autocorrelation) and a biexciton and exciton not belonging to a same cascade, as seen in Figure 30

The next step was to obtain the antibunching-bunching signature of photons that are emitted sequentially, such as the biexciton-exciton cascade. The coincidence counts for this were even lower because of the weaker biexciton emission, but after 4 hours one could start to see the antibunching-bunching signal begin to appear in the data. To be able to have a discernable signal, though, the run would require 12-24 hours. This was done for the positively charged biexciton-exciton cascade, since it had higher counts than the neutral biexciton-exciton cascade (see Figure 31).
Figure 30. Antibunchings and multiple peaks. Antibunches were observed where they were expected and in the proper type of correlation. Multiple extra peaks were constantly observed in the beginning for different types of correlation, which indicated unreal signals. These were later removed after troubleshooting the electronics. Solid green curves are Gaussian fits to the data.

From the plots, we can see an offset, which is the result of a background noise of ~2 counts per second. By smoothing the plots with a 5-point average and fitting it to a Gaussian, the widths of the peaks and troughs are determined to be 1.68 ns for the
autocorrelation of the neutral exciton, and 1.21 ns for the bunching and 1.70 ns for the antibunching of the positively charged biexciton correlation. These values are similar to those obtained by the exciton lifetime studies done by another member of our group.

The autocorrelation also presents some strange peaks and troughs, which were a consequence of our earlier setup. In it, a photon counter that we were using to determine the rate of photon counts on the APDs along with a different delay generator were introducing anomalous systematic pulses that were producing these strange peaks and troughs in all our data, as seen by their constant appearance under very different types of experiments. Making sure that the data makes sense in these preliminary stages is therefore critical. Using a different delay generator and electronics configuration eliminated these signals.

To test and calibrate our new setup, two different sources of uncorrelated light were placed in front of the MCs. A constant background noise without any noticeable kinks in it convinced us that our electronics were working as they should. To determine whether the signal was centered in the right histogram time bin, the signal from just one APD was sent to the TAC through the two different channels that trigger the start and stop in the TAC. This created a single-bin peak on the data that showed where the “zero-time” would be roughly. The antibunching and bunching-antibunching were both around that single-bin peak. To test whether what we were looking at was really coming from the sample, one APD’s signal was delayed a specific amount of time with respect to the other APD’s signal with a delay generator. The signals showed a shift in time as expected from the delay.
Figure 31. Bunching-antibunching of a positively charged biexciton-exciton cascade in a CQD. Red and blue circles in the schematic and bias map show the electric field and energies of the cascade around where entanglement could be expected to be observed. The red and blue solid dots show the electric field where the bunching-antibunching graph on the left was actually taken to better observe this type of signal, because of its higher number of counts.

A final test to prove the origin of the signals was performed by switching which APD would provide the start pulse and which would provide the stop pulse in the TAC. The antibunching showed no difference, as expected. The bunching-antibunching, however, was expected to be flipped with respect to the zero-time (i.e. to be an antibunching-bunching graph). What we saw was that when the biexciton was used as the start pulse and the exciton as the stop pulse, no signal would be present in the data, whereas having the exciton as the start pulse and biexciton as the stop pulse would yield
the figures shown. This difference, we attribute to the fact that there are many more excitons than biexcitons, therefore there are many more possible uncorrelated stop pulses that drown the signal when the biexciton is the start pulse.

As mentioned above, these runs are just preliminary data that served as proof of principle to us that we were able to perform correlation spectroscopy on CQDs and that there doesn’t seem to be anything blatantly wrong with it. The fact that our counts are so low is at least consistent with, although still lower than, the counts per hour that Gershoni et al. were getting.49

The last step in our experimental setup was to introduce polarizers and retarders to perform polarized correlation measurements. To be able to analyze the horizontal-vertical, diagonal(+45°)-diagonal(-45°), and right-left circular polarizations described by James et al.,53 3 variable retarders and a polarizer were introduced in each of the two paths going from the non-polarizing beam splitter to the MCs. For this we were required to first characterize our system to see if the beam splitter or other optical elements were introducing any attenuation or retardance that could skew our polarization results.

The polarization state is characterized by the Stokes parameters, which are a set of numbers in a vector normalized to the total light intensity, and measured in the horizontal-vertical, the diagonal(+45°)-diagonal(-45°), and the right-left circular polarizations basis.

\[
S = \begin{pmatrix}
I \\
Q \\
U \\
V
\end{pmatrix} = \begin{pmatrix}
\text{Total light intensity} \\
\text{Intensity diff. between H & V linearly polarized components} \\
\text{Intensity diff. between linearly polarized components oriented at ± 45°} \\
\text{Intensity diff. between L & R circular components}
\end{pmatrix}
\]
Sending photons with known polarization, detecting them, and calculating the Stokes parameters, the following results were obtained for the transmitted and reflected beams coming from the beam splitter (the ideal vector of the perfectly polarized case is included for comparison):

**Horizontal Polarized Excitation**

\[
S_{\text{ideal}} = \begin{pmatrix} 1 \\ 1 \\ 0 \\ 0 \end{pmatrix}, \quad S_{\text{trans}} = \begin{pmatrix} 1 \\ 0.922045 \\ -0.01247 \\ -0.16605 \end{pmatrix}, \quad S_{\text{refl}} = \begin{pmatrix} 1 \\ 0.930995 \\ -0.2005 \\ -0.0936 \end{pmatrix}
\]

**Right Circular Polarized Excitation**

\[
S_{\text{ideal}} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ -1 \end{pmatrix}, \quad S_{\text{trans}} = \begin{pmatrix} 1 \\ 0.134494 \\ -0.13173 \\ -0.90807 \end{pmatrix}, \quad S_{\text{refl}} = \begin{pmatrix} 1 \\ 0.277525 \\ -0.4975 \\ 0.781633 \end{pmatrix}
\]

**Left Circular Polarized Excitation**

\[
S_{\text{ideal}} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 1 \end{pmatrix}, \quad S_{\text{trans}} = \begin{pmatrix} 1 \\ 0.339888 \\ -0.34541 \\ 0.820276 \end{pmatrix}, \quad S_{\text{refl}} = \begin{pmatrix} 1 \\ 0.013452 \\ 0.893283 \\ -0.56066 \end{pmatrix}
\]
Comparing these results of the transmitted and reflected beam polarization vectors with the ideal polarization vector, it is seen that the horizontal polarization is well detected within a 20% margin. Right and left circular polarizations are within a 30% margin for the transmitted beam. However, the right and left circular polarizations for the reflected beam show a strong retardance that makes the diagonal and circular polarizations undistinguishable. It is believed from previous experiences, that the retardance from the beam splitter is sensitive to its position angle. Trying to position the beam splitter to minimize this retardance can be quite time-involved and sensitive to small perturbations, because of the amount of measurements that must be taken to see the retardance after every change in position. Also, it is not straightforward how and what changes in position will affect the different parameters in the different vectors. Therefore, the easiest method is just to create a Mueller matrix for each beam path that describes the system’s behavior and use them to correct for the retardance.

Since the entanglement will be between the polarization states relative to the QD’s major axes, we want to align the polarizers along these axes. For that, the polarization measurements were made on the neutral exciton and positively charged exciton to determine Stokes vector. The results were:

\[
S_{x^+} = \begin{pmatrix}
1 \\
0.55052 \\
-0.55402 \\
-0.26235
\end{pmatrix},
S_{x^0} = \begin{pmatrix}
1 \\
-0.35333 \\
0.37406 \\
0.2483
\end{pmatrix}
\]
Before attempting to measure the complete set of polarized correlation combinations with which to construct the density matrix, which would require 3 retarders and 1 polarizer in each of the two paths, we decided to just try putting the polarizers to see if we still had enough signal strength and if this did anything interesting to the correlation data. The signal strength dropped down 20-30% and we needed about 24 hours of integration time to obtain some recognizable data. With this long integration time, being able to keep the already low number of counts from falling due to sample drifting becomes a big issue. Furthermore, putting in the other 3 retarders would make the counts go even lower (i.e. even longer integration times), and since it takes 16 of such runs to determine whether there is entanglement or not under certain experimental conditions, the experiment becomes too long to be feasible. Possible solutions we’re looking into are a) inserting very small solid immersion lenses inside the cryostat to increase the amount of collected photons, and b) diminishing the magnification of the sample’s image on the APDs to allow more room for drifting without dropping the number of counts.

Producing and clearly measuring entanglement is a very difficult experiment to perform, but patience and determination are key factors to achieve it. This chapter has shown the path, the difficulties and the progress made towards it, and certainly we’ve come very close. Future research will have to overcome the low number of counts and determine how far away from the neutral exciton anticrossing one can get to minimize the anisotropic exchange interaction, while being close enough to have almost degenerate energy levels.
CHAPTER 3: SUMMARY AND CONCLUSIONS

In this dissertation we have presented results of the experimental study of the electro-optical properties of CQDs done at Ohio University. These include the observation and characterization of excited states, the study of photovoltaic effects within the devices, and the investigation of correlation effects in the emission from CQDs. These results will provide a solid foundation for further investigations into the properties of these technologically relevant structures. The following major results were reported.

*Photoluminescence Excitation Study of the CQD Shell Structure*

Through this PLE study we have optically probed the shell structure of the top dot by resonantly creating an indirect neutral exciton and monitoring the direct neutral exciton. This was achieved using the understanding obtained from the new technique to visualize PLE.

The new technique entails creating .gif movies using bias maps taken at different excitation energies as frames for the movie. Absorption into indirect states was identified as points moving along the PL emission line of the state being monitored in the PLE movie. Absorption into direct states was identified as a mostly continuous line, in a single flashing frame, along the PL emission line of the state being monitored in the PLE movie.

The identification of the hole’s excited states in the top dot was given by relating the results obtained using PLE with those using LACS. This technique can be easily extended to further studies, and possible control, of the excited states in CQDs. Possible further studies can include polarized PLE to study spin dynamics in excited states of CQDs, and pump-probe experiments for initialization techniques into direct or indirect states that act as qubits (quantum bits). Relating information regarding the type of
absorption (moving dots or flashing lines) to the type of emission (indirect for bias-dependent lines or direct for bias-independent lines) may reveal some of the created exciton’s dynamics, such as relaxation and tunneling.

*Observation and Control of a Laser Induced Photovoltaic Effect*

We have used a CQD to study the optically created electric field due to photovoltaic effects. The field produces a shift in the applied electric field dependence of interdot exciton PL. Due to the physical separation of the electron and hole, the interdot exciton provides a sensitive probe of the strength of this field. Power and wavelength dependence curves show saturations due to a limit in the amount of charge the device can accumulate under steady state excitation. They also show a dependence of the optically created electric field on the applied electric field. A maximum optically generated field of ~3.25 kV/cm was observed which corresponds to 5.04% of the total field. The ability to optically generate localized electric fields may be useful as a simple method for creating a modulated signal for experiments such as differential transmission where a varying field on top of a constant applied field is required.

*Correlated Photon Emission from CQDs*

Initial studies of correlated photon emission from the biexciton cascade in CQDs were performed. These resulted in the observation of expected bunching and anti-bunching signatures associated with correlated photon emission from quantum mechanical, single photon emitters. We also measured the polarization state of the emission and characterized the effects of the experimental setup on the observed polarization states. These are necessary first steps toward the possibility of producing entangled photon pair emission from these samples. A possible experiment to follow the work done so far on the way to entanglement could be bias-dependence on the time-
correlated photon emission near and far from anticrossing regions. Finding better ways to diminish losses in light acquisition might allow for polarized time-correlation experiments.

CQDs have a great potential for studying basic principals of quantum mechanics in the lab and for high-yield applications, such as quantum computing. The results presented in this dissertation hopefully serve as a stepping stone in better understanding their dynamics and structure, and in providing experimental tools to work with them.
REFERENCES


APPENDIX A: MAKING PLE MOVIES

**Step 1: Taking data**

The PLE program is very similar to the bias map program in terms of setting it up to run (see Figure 32). Voltage Start is the lowest voltage and Voltage End is the highest, with Voltage Step defining the increments. Laser Wavelength Start is the lowest wavelength in nm, and Laser Wavelength End is the highest wavelength in nm, with Laser Wavelength step defining the wavelength increments in nm.

![Figure 32. PLE program. Front panel.](image)

Like the bias map program, it has a Monitor T and P button to choose to monitor the sample’s temperature and the laser power. There is also the Cosmic Ray Correction
button to choose to take 2 acquisitions at each bias, the Start button to start the run and the Stop button (in red color) to exit the PLE program at the end of the run.

When the Start button is pressed, a new screen will appear, which is nothing more than the usual bias map acquiring screen. After it is done with the first bias map for the first wavelength it will close, the acquired bias map will be displayed in the PLE program screen, change the wavelength and display it in the Wavelength box of the PLE program screen, open up the bias map acquiring screen again and start the new bias map at the new wavelength.

The Stop Run and Pause buttons will stop and pause the current run after the currently running bias map acquiring screen is done.

In addition, there is a feature which was incorporated in case one may desire to move the monitored energy (i.e. move the grating by changing the center wavelength) by keeping the center wavelength always a certain distance in nm from the laser wavelength. This is chosen with the Move Bias map with Laser button and the distance between the center wavelength and the laser wavelength is set in the Set Stage Within (nm) of Laser box.

Sequentially speaking, the program developed to run the PLE experiment does the following (see Figure 33):

1. If the moving Move Bias map with Laser button has been pressed, it sets the grating so that the center wavelength is within the given distance from the current laser wavelength.

2. Enters the Set & Meas Wavelength subVI:
a. Sets the laser at the given laser wavelength variable.

b. Waits 500ms to make sure everything has settled.

c. Reads the wavelength from the Bristol laser wavelength meter (see Figure 34).

   i. Evaluates whether the read laser wavelength is within a certain range of the desired wavelength or not (+-12nm). This is to ensure there was no mode hoping in the laser.

   ii. If the wavelength is within range it continues.

   iii. If it is not within range, because it cannot read the wavelength meter (due to the laser intensity being too low or too high for the Bristol wavelength meter to measure), or because the laser has mode hopped, it goes back to step (i) and will try reading again for up to 5 times.

      1. If by the end of the 5 tries the wavelength is still not within range, it will pause and show a warning message until the user decides to either skip the present reading of the laser wavelength (but still take the data) or the laser is checked for realignment and then goes back to step (i) to read again.

3. Sets the wavelength on the BEOC (laser power controller).

4. Sets the wavelength on the Meadowlark laser power meter.

5. Enters the Bias Map subVI
a. Runs a bias map.

b. Saves the bias map as a .odf data file with the wavelength it was taken at using the format given (FrameNumber_Wavelength”nm”.odf within a folder whose name contains all the information about the experiment).

6. Increases the laser wavelength variable by the user-specified amount and goes back to step 1 in a loop until the last wavelength is reached.

Figure 33. PLE program Block Diagram. Numbered red boxes indicate step outlined in text.
Once the PLE data has been gathered, a simple bias map must be taken which contains both the range of energies in which the data was taken and the range of energies across which the laser was swept (usually done with a 300 groves/mm grating in the spectrometer). This bias map will serve as our guiding map in the movies to see where the laser energy was exciting at the time the frame was taken.

**Step 2: Turning .odf data files into .png image files**

The bias map files for the PLE will have been saved in a single folder and labeled by a frame number, followed by an underscore “_” and the laser wavelength it was taken at. Example: 14_880.505386nm.odf. In order to have them all uploaded to the NRLData.exe viewing program at the same time in the correct order, all frame numbers must have the same amount of digits. This means that if there were 20 frames, which start with 1, 2, 3, …, 20; the file names of the frames must be changed so that they are
numbered 01, 02, 03, …, 19, 20. Similarly if there were 150 frames, the file names must be renamed so that they are numbered 001, 002, 003, …, 009, 010, 011, …, 099, 100, 101, …, 149, 150. This procedure will save a lot of time later. If it’s not done, the program will upload them in the following sequence: 1, 10, 100, 11, 110, 111, 112, etc. The alternative is to choose to either upload each frame individually in the correct order into the viewing program (very time consuming when there are many frames); or upload them all at the same time and then having to organize them by removing plots and re-adding them where they belong.

To open all the .odf files at the same time is very easy. First, the NRLData.exe program must be opened. The user must make sure there are no previous plots loaded by clicking the Remove Plot button on the lower right corner. then clicking on Open Folder option in the File tab. See Figure 35.
Figure 35. Open Folder option in File tab.

This will open a dialog box to choose a folder from. The user must find the folder where the PLE files are located and be within it, then click the Current Folder button on the bottom right corner of the dialog box. See Figure 36.
Figure 36. Current Folder button to open all files in the folder.

At this point, the frames can be manipulated with the program to bring out and highlight interesting features that the user may want to show. Once this is done, they can be saved individually as .png image files by clicking on the Save As Png option of the Export tab in the main menu bar. See Figure 37.
Figure 37. Save as Png option in the Export tab.

The same must be done with the reference bias map, but it should be saved on a different folder from the .png images of the PLE frames.

**Step 3: Creating a complete frame**

To create a complete frame that combines together the .png image files from the PLE frames and the reference bias map, the Movie Image Creator.vi will be used, which can be found in the following path: C:\Documents and Settings\Eric Stinaff\My Documents\Mauricio\Programs. Before beginning to run the program the fields on the right side must be filled. Highest Energy QD Bias map refers to the highest energy (in meV) that is displayed on the .png images of the PLE frames. Similarly for the lowest. Starting Laser Wavelength refers to the shortest wavelength (in nm) for which a PLE frame was taken, which can be found in the file name of the first of the PLE frames.
Note: it is assumed that the PLE frames were taken and are thus organized from shortest to longest laser wavelength, e.g. the first file name could be 00_862.6nm and the last file name could be 82_945.2nm.

The run button can now be clicked to start the program. This will prompt a dialog box asking for the Wetting Layer Bias map File. This is the path to the reference bias map. See Figure 38.

Figure 38. Reference bias map dialog box.

A second dialog box will then be prompted asking for the Image Sequence Folder. This is the path to the .png PLE frames. Note: the user should be within the folder containing the .png PLE frames and click the Current Folder button, not the Open button. See Figure 39.
The program will then begin to create a sequence of complete .png image frames with the PLE frame on top and a static reference bias map below with a red line defining where the laser energy was at the time the PLE frame on top was taken and two vertical blue lines defining the area within the reference bias map that corresponds to the PLE frame (the two diagonal lines extending from the bias map are to give the idea that the bias map on top is a blow up of the area defined by the two vertical blue lines on the reference bias map on the bottom). See Figure 40.
Step 4: Making a movie

The program that will create the movie is the Jasc Animation Shop 3, which is part of the Jasc Software. To open the files into a movie sequence, first the Animation Wizard button must be clicked. It is the icon below the File tab of the main menu. See Figure 41.
This will prompt the Animation Wizard dialog box. The first display will ask for the total dimensions of the complete .png files. The Movie Image Creator.vi will have made them of width: 699 and height: 968. See Figure 42.

The default canvas color for the animation is Opaque. See Figure 43.
In the next dialog box, it should be chosen that the images should be located in the upper left corner of the animation frame; and that when the image occupies only part of the frame, the rest should be filled with canvas color. See Figure 44.

In the next dialog box it should be chosen that Yes, the animation should be repeated indefinitely in a loop. As for the frame display time, 30 is a good number to
start. This time can later be changed when a simulation of the movie is run, before saving it. See Figure 45.

Figure 45. Loop and display time.

The following dialog box will ask to load the images. For this the Add Image… button is clicked. This will prompt a new dialog box to search for the image files. The user should then go to the CompletePng folder and select all the image files within the Open dialog box at the same time, starting with the last one. If the user begins selecting all of them starting with the first one, this will cause the last frame to be the first one listed in the Animation Wizard. This is not a big deal, since the user can then use the Move Up, Move Down or Remove Image buttons to sort the list, but this is just an easy trick to avoid it. See Figure 46.
When the Open button is clicked, the images will be uploaded into the Animation Wizard. See Figure 47.

In the last dialog box the Finish button is clicked. The frames will then be displayed within a single box. To run the animation, the Animation option in the View
tab of the main menu must be selected. This will display a small looping animation in a pop-up box. See Figure 48.

![Figure 48. Animation option in the View tab.](image)

If the speed is too fast or too slow, the user must first select all the frames by selecting the Select All option in the Edit tab of the main menu and then the Frame Properties… option of the Animation tab in the main menu. This will prompt a dialog box that allows one to change from the initial 30 frames per second that was selected in the Animation Wizard. Note: if all the frames are not selected, only the current frame will be affected by the change in the time.

To save the file the user can just select the Save As option in the File tab of the main menu. This will create the desired PLE movie.
APPENDIX B: EXPERIMENTAL CHALLENGES

The following are some of the experimental challenges we faced during the course of our experiments and the way they were addressed to push the research forward:

- Challenge: Given the fact that the system being studied is individual QDs, the number of photon counts is, of course, very low. Therefore, the experimental setup is crucial for not losing too many of those already few counts. After the initial loss of light from emission in every direction other than that of the collection optics; every mirror, polarizer, retarder, or beam splitter in the out coming PL introduces a decline in photon count.

  Solution: We had to align our system well from time to time and rearrange our set up so that the PL’s path to the spectrometer was as direct as possible.

- Challenge: Low number of counts implies long integration times for taking data are necessary. For polarization studies, both right and left polarized light coming from the sample must be detected, increasing the integration time by a factor of two.

  Solution: there were initial attempts to split the CCD camera into two separate regions and put a non-polarizing beam splitter before the retarders, so that we could simultaneously analyze the two polarizations in one run. Unfortunately, this became highly time consuming and technically very challenging, since precisely-placed collimators were needed after the beam splitter to send the signals parallel to each other and separated by a very small distance from each other.

- Challenge: The design of the cryostat is such that it suffers from mild vibrations from the pumping. Despite the company’s efforts to isolate the pumping from the rest of
the cryostat, the sample moves beyond the micrometer precision needed to do spectroscopy on QDs.

Solution: A movable infrastructure for the cryostat was designed at the lab to minimize the contact between the pumping shaft and the rest of the cryostat. Also, a temperature controller was placed so that small variations in temperature over time would not make the sample drift out of focus. The settings of such a PID temperature controller for most stable condition were found to be $P=90$, $I=999$, $D=0$ by process of trial and error.

-Challenge: Virtual leaks in the cryostat hinder the sample from cooling below 30K.

Solution: A complete system clean-up, periodic heating (baking) of the entire system for a few hours, long vacuum pumping times (a few days), and keeping a positive pressure of He gas (to prevent the cryostat bellows from collapsing and ruining the vacuum) seem to allow the system to reach nominal temperatures of 8.5K and maintain itself below 20K for a week or more sometimes, before it begins to slowly creep up in temperature again.

-Challenge: With the use of a BEOC laser power controller, the laser power can been kept relatively constant. However, due to the optics that split and steer the beam into the sample, the laser wavelength meter, and the laser itself; the power still oscillates as the wavelength changes.

Solution: Before running a PLE experiment, the laser wavelength would be swept along the range it would go through in the experiment and whenever there would be a decline in power, the laser would be re-tuned to peak up the power. At the end of the sweeping, a new quick sweep would be performed to make sure all the range was still
fairly constant in power. This procedure ensures a smoother taking of data in PLE experiments.

-Challenge: For accurate measurement of the wavelength being used to create excited states in the QD, a Bristol 821 Pulsed Laser Wavemeter was purchased. Even though this is the most accurate wavemeter in the market up to date, it can only be partially automated. Constant manual handling of the beam attenuator must be performed in order for the device to correctly detect the laser beam.

Solution: In order to make sure that the PLE experiments would record the wavelength every time it would take data, an alarm system was built into the program to stop it from taking any more data until the user either manually fixed the wavemeter to read or decided to skip the reading. This, however, meant that the system could potentially stop in the middle of a run for a long time if left unattended. The alignment procedure of the previous challenge was therefore crucial for long, over-night runs.

-Challenge: Within a QD sample there are hundreds to thousands of apertures (most of which contain a right amount of QDs to single out), but less than 100 of those actually contain a QD whose bias map exhibits an x-pattern to study. Thus a great deal of time is spent “hunting” for such apertures. Unfortunately, being able to clearly identify the place in the sample where an x-pattern is found, so that it can be returned to afterwards, proves extremely challenging.

Solution: Using e-beamed patterned samples with guiding markings completely solves this problem.

-Challenge: The sound of the pumping in the expander head of the cryostat goes from a “chugging” sound to a very hard metallic pounding sound. In addition to this, a strong vibration can be seen to build up in the expander head. This strong vibration can
be seen to translate to the sample. This is an indication of a strain in the system that could eventually turn out damaging it. The problem has presented itself at around four thousand hours of compressor running time, then at around eight thousand hours of compressor running time and the last time has been at around twelve thousand hours of compressor running time.

-Solution: On previous occasions, we sent the expander head of the cryostat back to Advanced Research Systems, the company that sold this system. After taking months of trying to contact them, shipping, testing, fixing, retesting and reshipping; the expander seemed to work fine again.

The last time this happened, in order to save months’ time in all this process, we decided to see if we could do something ourselves… even taking it apart if it was the case. We believed it was either impurities that were stuck in the expander head or something mechanical, like a misplacement of the rings that keep the different gas compartments in the expander separated from each other. Since a mechanical repair is a more involved procedure, the first option was to see if the banging noises were coming from impurities, by trying to clean them out.

To begin, the simplest procedure we tried was changing the adsorber. This was due anyway at twelve thousand hours of running, so we thought it might be what was causing the problem. After changing it and running for a few minutes, the hard metallic banging noise began again.

Next, we purged the expander head with ultra-high purity Helium a couple of times. Then we purged the whole system connected by introducing the ultra-high purity Helium into the compressor, letting it run for a few seconds, letting the Helium come out and repressurizing the compressor with new Helium. This we did it for a couple of times.
Accidentally, in one of the repressurizations, the compressor was over pressurized to 310 psi. The normal running pressure is 270 ± 20 psi. The expander head of the cryostat, however, sounded much quieter. In the following repressurization, we did not over pressurize and the sound was back up as loud as before. Therefore, the following repressurizations were done over pressurizing the compressor.

At the end of these purgings, we noted how much softer the expander head sounded. We also noted that in the last purging, we had not repressurized again to the right pressure, but had left the system pressurized at the high end of the normal running pressure at 290 psi.

The system was allowed to cool down and was monitored to check for any anomalies. The cooling down went fine the expander head kept sounding softly.

We concluded that it must have been impurities trapped inside the expander head that somehow were removed when the system was run over pressurized by being forcibly pushed out.

After a few days or so of running normally, we noticed the compressor gas pressure had dropped down a few tens of psi. This was a little worrisome, since it basically meant that there was a leak present.

In the past few days we have observed that the system loses about 10 psi per day and thus, repressurizing of the compressor must be done every week.

The gas lines and the compressor will be checked soon to determine where exactly the leak may be coming from.

-Challenge: All the electronics in the lab have had to be shut down due to the room temperature rising above 80 degrees Fahrenheit every spring, because the AC is not
turned on until a certain specified date, even though the outside temperature may be high. This is also happens to be the time when data is being taken for the APS Spring Meeting.

Solution: We have had to semi open the lab door and put fans to extract the hot air from the room. This helps a little, but depending on the outside temperature, sometimes we just have to shut down the electronics.

The design for the new building promises to handle these temperature issues much better with a new heating/cooling system. Also, the floors would be built in such a way to minimize building vibrations, which we can definitely see in our sample. Finally, flywheel back-up generators would prevent power failures from ruining equipment by kicking in much faster than diesel back-up generators would.