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Third-Order Frequency-Resolved Photon Correlations from a Single Quantum Dot's Resonance Fluorescence

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Third-Order Frequency-Resolved Photon Correlations from a Single Quantum Dot’s
Resonance Fluorescence

by

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A dissertation submitted in partial fulfillment
of the requirements for the degree of
Doctor of Philosophy
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Dedication

Para mi padre y madre, Aníbal y Zoraida,
quienes no saben inglés y, por tanto, no comprenderán nada en esta disertación y aún así se enorgullecerán igual

y

para mi abuelo, Luis Camargo,
quién, de niño, me empujó a una carrera en medicina, pero mi rebelde ser optó por estudiar Física.

Ellos fueron quienes me criaron para alcanzar este nivel con todo su amor y apoyo.
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**List of Abbreviations**

FWHM. ................................................ Full Width at Half Maximum

HBT. .................................................. Hanbury-Brown and Twiss

QD. ................................................... Quantum Dot

SPCM. ............................................. Single-Photon Counting Module

SD. .................................................... Spectral Diffusion

TLS. ................................................ Two-Level System
Abstract

The ability to control the photon emission from light sources has sparked an increasing interest in the field of quantum optics due to potential applications in the development of quantum technologies. Motivated by such applications, photon statistics have been a powerful investigative tool used to characterize the properties of light sources capable of emitting a bundle of $N$ photons. As such, the first-order and second-order photon correlations continue to be researched extensively providing invaluable information about the light and the mechanism underlying its generation. Meanwhile, third-order photon correlations have remained majorly unexplored due to the high degree of experimental challenges involving such measurements. This work presents experimental progress towards constructing the complete spectrally filtered three-photon spectrum of the resonance fluorescence generated from a single semiconductor quantum dot under strong monochromatic light excitation, and explores the effects imposed by the limited resolution of the filter bandwidth and environmental influences due to spectral diffusion. We investigated third-order auto-correlation measurements, in which photons were identically filtered, resulting in correlation maps that are functions of two relative time-delays. These measurements revealed accentuated nonclassical characteristics amongst the distinguishing features of the three-photon spectra following faithful agreement with theory. Additionally, we investigated third-order correlations where photons originate from opposite sidebands of the power spectrum. These three-photon cross-correlation measurements resulted in correlation maps with “anti-bunching” features as a consequence of correlations arising from the same sideband, and “bunching ridges” due to opposite sideband correlated photons.
Chapter 1
Introduction

Information is all around us—on books, pictures, internet, DNA, etc.—and the way we process this information is constantly changing. From Pascal’s arithmetic machine to small computers that can fit in your pocket (a.k.a. smartphones) we seek smaller, faster and more powerful devices that are able to store and process this information. With the coalition of quantum physics and computing theory we are on the verge of a new technological revolution in quantum computation & information processing. By harnessing the laws of quantum mechanics, quantum computers are expected to enhance considerably the information processing power to the point that computation currently out of reach, even for classical super-computers, would become feasible. Before achieving such technology, a fine control over the quantum particle used as the information carrier is required. Quantum states of light are expected to play a crucial role in transmitting quantum information over distances. Therefore, a fascinating subject of research in quantum optics is the science of correlations between photons.

1.1 The Concept of the Photon

The study of the properties of light is one of the most fundamental aspects of the natural sciences that has puzzled scientific minds for centuries. The foundations of the field of optics (such as reflection, refraction, diffusion, and vision) were established by philosophers from ancient Greece during Antiquity (between 8th century B.C. and the 2nd A.D.) from Plato, to Aristotle, to Euclid and Ptolemy, followed by scholars of the “House of Wisdom” during the
Islamic Golden Age (between 8th – 14th century) from Al-Kindi, to Ibn-Sahl, all the way to “the father of modern optics” al-Haytham. These concepts on light rendered a large effect in the development of optics up until the appearance of Newton in the late 17th century.

Sir Isaac Newton is without a doubt one of the defining figures in the history of science. Newton is mostly known by his *Principia* (which states his universal law of gravitation and his laws of motion that formed the foundations of classical mechanics) and by co-discovering mathematical methods now included in the field of calculus. It is, however, important to note that one of Newton’s major contributions was in the field of optics. He was the first to show that color is an intrinsic property of light, and advocated for its corpuscular theory claiming that “light is comprised of colored particles”, despite evidence that supported the wave behavior (e.g., Grimaldi’s observation of the diffraction of light).

After a long debate among prestigious names that spanned for decades, it seemed that Huygens’s wave theory, Young’s double-slit experiments, Fresnel’s work on diffraction and polarization, and Maxwell’s electromagnetic field theory were able to explain most of the phenomena of light, electricity, and magnetism. The electromagnetic wave theory had triumphed and the particle interpretation was believed to be definitely buried. No one suspected that the dawn of the twentieth century would bring a revolution in our understanding of the nature of light yet again.

The beginning of the 20th century carried two uncertainties involving light that brought forth the birth of two revolutionary theories that cause major transformations in the field of physics. One was the unexpected result by the Michelson-Morley experiment [1] that showed the speed of light is the same regardless of the motion of the Earth. The other was the uncertainty surrounding the nature of blackbody radiation at high frequencies, commonly known as the Rayleigh-Jeans ultraviolet catastrophe. The former lead to Einstein’s famous Theory of Special Relativity [2], while the latter ultimately led Max Planck to postulate that radiation energy can be divided into a number of discrete “energy elements”, $\epsilon$, that must be proportional to the frequency $\nu$, that is $\epsilon = h\nu$, where $h$ is a constant that eventually
carried Planck’s name [3]. Planck’s seminal work is generally regarded today as the birth of Quantum Theory.

The arrival of quantum mechanics brought with it the revival of the particle versus wave nature of light controversy. In 1905, Einstein used Planck’s “energy elements” to explain the photoelectric effect (emission of electrons from a material after absorbing electromagnetic radiation). Einstein proposed that, not just the energy but the radiation itself is quantized in discrete energy packets which he called “light quantum” [4]. This theory marked the beginning of the modern concept of the photon.

A photon is generally created via a process known as *spontaneous emission*, which occurs when a quantum mechanical system—such as a molecule, an atom, or an artificial atom—decays from an excited state to a lower energy state by releasing a quantized amount of energy.

In 1956, Hanbury-Brown and Twiss (HBT) discovered another surprising property of light as they were trying to measure the angular diameter of the stars by studying the correlation between intensity fluctuations [5]. The HBT experiment consisted on splitting a low-intensity beam of light (simulating that of a star) into two components using a half-silvered mirror, each component detected separately through two photo-multipliers. In their experiment, HBT claimed that two photons had been positively correlated (i.e., detected at the same time) when it had been expected that only single photons were to be detected due to the low intensity of the source. This effect brought in a lot of controversy into the world of quantum theory [6].

At the time, this result seemed to go against the foundations of quantum theory since, assuming a photon as a small localized indivisible particle, there would be no way to detect
two photons at the same time. Although initially rejected, the HBT effect was soon understood to be the manifestation of the wave-particle duality of light [7]. By simply treating photons as a system of bosons (wave-like objects) which can be accumulated in the same energy state, the quantum effect is then a result of the “bunching” of monochromatic photons (photons with a single optical wavelength or frequency). Along with the HBT development, another famous optical effect of similar quantum nature was discovered which provided further insight on the behaviour between two identical photons.

In 1987, physicists Chung Ki Hong, Zhe Yu Ou and Leonard Mandel demonstrated a technique based on the interference of two identical single-photons as they enter through the two input modes of a 50:50 beamsplitter, where both photons have an equal chance (50%) of either being reflected or transmitted [8]. Due to destructive interference, the probability that both photons are reflected (or transmitted) decreases the more indistinguishable the photons are. Therefore, two photons with identical properties will always exit the beamsplitter together as a pair, this is commonly known as the Hung-Ou-Mandel (HOM) effect. The discovery of the HBT and HOM effects, along with the advent of the laser, led to the development of modern quantum optics as it was shown that all kinds of light sources can be differentiated in terms of their photon statistics [9].

1.2 Types of Electromagnetic Radiation

For a broad range of quantum processes, including those involving multiparticle interactions [10], photon statistics have been a powerful investigative tool used to characterize the coherence properties of an electromagnetic field. Instead of simply being linked to monochromatic wave sources and a constant phase difference, in its essence, the degree of coherence is the measure of the temporal correlations between photons.

The correlations between the detection time of subsequently emitted photons serve as the primary tool for characterizing the type of radiation emitted through light-matter interactions at the few or single quantum level. For example, for photons originating from
blackbody radiation (i.e., incoherent light sources such as the sun or an incandescence light bulb) coincidences are systematic since photons arrive together in groups of two or more at the detector, exhibiting strong positive correlations as seen in the bunching reported by the HBT experiment. Meanwhile, for stimulated radiation (i.e., coherent light sources such as a laser), no mutual correlations are shown since photons arrive at the detectors arbitrarily indicating no time relationship between them. In addition to the thermal and stimulated light sources, there is a third type of radiation that cannot be explained through classical methods, displaying negative correlations: the single-photon sources.

A single-photon source emits light as well spaced photons within its emission lifetime after being optically, or electrically, excited causing them to arrive at the detector one at a time and therefore producing the characteristic of photon antibunching. Ideally, a single-photon source: (1) emits deterministic (i.e., on-demand) single photons at any arbitrary time defined by the user, (2) have a 0\% probability of emitting multiple-photons, (3) emits identical (indistinguishable) photons, and (4) allows for a fast repetition rate [11–13]. These non-classical states of light serve a wide range of applications in the so called “quantum information processing” where the data processing is done using a quantum system as the information carrier.

1.3 Applications

The ability to control the photon emission from light sources is a key component in the development of quantum technologies [14–17]. These systems use the quantum-mechanical states of a two-level quantum system (a system with only excited and ground energy states) as quantum bits (also know as ‘qubits’). These qubits replace the traditional binary bits (0 or 1) used in classical computers in order to enhance considerably the processing power. While the traditional binary bits can only be in one state or another, qubits harness the laws of quantum mechanics where coherent superposition allows for both states to exists simultaneously.
Through massive parallel processing, quantum computers are expected to perform a wide variety of applications like the ability to: crack popular encryption schemes with quantum cryptography [18–21], extend the range of distribution of quantum resources through quantum repeaters [22–24], optical imaging using light that has never interacted physically with the object to be imaged through ghost imaging [25–27], complete transfer of information between particles through quantum teleportation [28–31], or making highly sensitive measurements with high-resolution through quantum metrology [32–35]. The pursuit of such quantum applications makes it increasingly important to expand the available quantum sources in order to be capable of emitting a bundle of $N$ photons.

### 1.4 $N$-photon Sources

One useful approach to produce $N$-photons is by generating a correlated bundle of photons, where one photon may be used to ‘herald’ the existence of the other photons. A common example of such approach is via a nonlinear process in bulk such as spontaneous parametric down-conversion in crystals [36–39] and waveguides [40], or four wave mixing in optical fibers [41–43] and cavities [44]. While heralded single-photon sources are very practical for many of the applications mentioned on Section 1.3, these methods suffer from significant drawbacks as the repetition rate must be significantly reduced in order to avoid producing more than the desired amount of photons. This has led researchers into developing truly deterministic single-photon sources.

A potential approach to produce deterministic single-photons (or entangled photons through cascade emission [45]) is from two-level quantum systems such as atoms [46–49], molecules [50–52], ions [53, 54], mesoscopic quantum wells [55], or a nitrogen-vacancy colored center in diamond [56–59]. Another promising $N$-photon emitter with excellent optical stability, compared to other solid state systems, is the semiconductor quantum dot [60].
Quantum Dots

Quantum dots (QD) are semiconductor crystals which size ranges within only a few nanometers depending on their production process. QD nanotechnology has successfully entered numerous electronic and biomedical industries due to their wide variety of applications such as single-electron transistors [61], photovoltaic solar cells [62], and medical imaging [63], just to name a few. Their small size causes the motion of conduction band electrons and valence band holes, or excitons (bound state of an electron and electron hole) to be confined in all three spatial dimensions [64]. This confinement results in discrete energy states which are characteristic of individual atoms (Figure 1.2), which is why QDs are commonly referred to as “artificial atoms”. These atom-like energy states contribute to the QD’s special optical properties, such as the emission of photoluminescence light.

Photoluminescence occurs when the light is absorbed by the quantum system exciting the electron to a higher unstable energy state and, after some time, the electron decays via spontaneous emission as shown in Figure 1.3. When this process involves a change in the electron’s spin multiplicity, it results in a longer lifetime of the excited state (seconds to minutes) producing phosphorescent light. However, when no such spin change is involved, the absorption/emission process occurs almost instantaneously (<10^{-5} s) emitting fluorescent light [65]. Typically, the energy of the emitted photon is lower than that of the absorbed...
photon, however, this study focuses on one particular type of fluorescence emission where the frequency of the absorbed radiation corresponds almost exactly to the natural frequency of the QD, known as resonance fluorescence. Essentially, this makes the QD a system consisting of only two energy levels: ground and excited states.

As a consequence of these fascinating optical properties, QDs have been exploited for a wide range of applications including single-electron transistors [66], quantum computing [67], lasers [68], second-harmonic generation [69], LEDs [70], photovoltaic solar cells [62], medical imaging [71], and single-photon sources [72]. In an attempt to improve feasibility for large scale production and reduce fabrication defects, scientist continue to search for different processes to manufacture QDs such as colloidal synthesis [73], and chemical vapour deposi-
This work focuses on light generated from InAs QDs grown by molecular-beam epitaxy.

QDs grown epitaxially in the Stranski-Krastanov growth mode (or “self-assembly” effect) consist on the deposition of a so-called “wetting layer” (successive atomic layers) of a material with a small band gap, such as indium arsenide (0.36 eV at 300 K), on substrate with a larger band gap such as gallium arsenide (1.43 eV at 300 K). The difference in lattice structures between the InAs layers and the GaAs substrate begins a phase transition once the critical thickness of the wetting layer is achieved. Due to the compressive strain, it becomes energetically favorable for the InAs layer to form nano-scale islands, aka QDs. Finally, to preserve the QDs from the external environment, a capping layer of the substrate is deposited over the wetting layer.

Figure 1.4: Schematic illustration of the epitaxial growth of self-assembled QDs: (a) Deposition of a thin layer of InAs on top of a GaAs substrate leading to an accumulation of biaxial strain due to the lattice mismatch. (b) Once the critical thickness of the wetting layer is reached, QDs islands are formed to minimize the total energy of the system. (c) Deposition of additional GaAs serving as a capping layer to avoid interaction with the external environment. (d) Band structure scheme illustration for self-organized quantum dots.

1.6 Overview

In this work, we aim to understand the spectral and time-correlation properties of a light scattered by a strongly driven semiconductor quantum dot. Chapter 2 is devoted to a review on the theoretical study of the interaction between a two-level system and a monochromatic
resonant (or near-resonant) laser field. In Section 2.1, treat the laser field as a single-mode quantum field to clearly identify the elementary processes of the spontaneous emission for fluorescence photons. We start by neglecting the interaction between the two-level system (TLS) and the laser photon system, such that the two systems are treated separately as a superposition of one another. This way the processes of absorption and stimulated emission of laser photons appear simply in the basis of uncoupled states which describes the two-level system driven under low power laser illumination. Then, we treat them as a coupled-system such that the two-level system is “dressed” by the laser photons and the process of spontaneous emission appear now in the basis of dressed states which describes the two-level system driven under high power laser illumination. Finally, we derive the optical Bloch equations which compose the master equation of the driven two-level model.

In Chapter 3, we explore the theory behind first-order correlations and the power spectrum of a strongly-driven two-level system, and provide the dressed-state picture to illustrate the origin of the features present in the correlation spectra. In Section 3.2 we discuss the experimental aspect of such measurements and include the introduction of non-idealities due to filter bandwidth and spectral diffusion into theoretical simulations.

Chapter 4 is devoted to the second-order correlations for the arrival times of the photons emitted by the coupled-system. First, we discuss the temporal aspect of the two-photon correlations and how it is used to characterize the type of emission process from a light source. Then, in Sections 4.1 and 4.2, we make use of the sensing method to explore the frequency-resolved two-photon spectrum and its underlying emission process down the dressed-states ladder.

In Chapter 5, we have performed an experiment to further our knowledge in the non-classical aspect of the scattered light emitted by a resonantly-driven quantum dot. In Section 5.1, we show the experimental results of the third-order auto-correlations measurements, i.e., correlations between the arrival times of three photons detected at the same frequency. The results of this investigation were published in a peer-reviewed journal: “Third-order
frequency-resolved photon correlations in resonance fluorescence” [Nieves et al., Phys. Rev. B 98, 165432 (2018), 77]. In Section 5.2, we show the experimental results of the third-order cross-correlations measurements, i.e., correlations between the arrival times of three photons filtered at different frequencies. The results of this investigation were published in a peer-reviewed journal: “Third-order photon cross-correlations in resonance fluorescence” [Nieves et al., Phys. Rev. B 102, 155418 (2020), 78].
Quantum dots are often referred to as “artificial atoms” due to their well-defined spectrum of energy levels [79]. The pumping of photons from a monochromatic laser excites the electron in the quantum dot which later goes through a relaxation process from a higher energy level back to the lower energy state. This results in the excited quantum dot to emit light via spontaneous emission, known as fluorescence. When the driving laser closely matches the natural frequency of the quantum dot, this fluorescence light is said to be in resonance with the quantum system. This so called resonance fluorescence is a fundamental source of light that has been particularly fit for the study of correlations with combined time and frequency information. This process of fluorescent emission from the quantum dot results in a simple model known as a two-level system which resembles the electronic shells in atoms featuring exceptional optical properties at cryogenic temperatures [80].

2.1 The Driven Two-Level Model

A two-level system can be described by a 2-dimensional complex Hilbert space consisting of two independent quantum states: a ground state, $|g\rangle = (\frac{1}{1})$, with energy $\hbar \omega_g$, and an excited state, $|e\rangle = (\frac{0}{1})$, with energy $\hbar \omega_e$. The atomic operator of the system can be defined
by a linear combination of the four basis operators:

Creation operator: \( a^\dagger = S_{eg} = |e\rangle\langle g| = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \),

Annihilation operator: \( a = S_{ge} = |g\rangle\langle e| = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \),

Ground steady-state operator: \( aa^\dagger = S_{gg} = |g\rangle\langle g| = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \),

Excited steady-state operator: \( a^\dagger a = S_{ee} = |e\rangle\langle e| = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \).

2.1.1 Non-Interacting Hamiltonian

We begin by neglecting the interaction between the two-level system and the laser photons, the Hamiltonian for this system is then in the form of \( H = H_0 + H_L \) where \( H_0 = \hbar \omega_g S_{gg} + \hbar \omega_e S_{ee} \) describes the Hamiltonian of the unperturbed two-level system, and \( H_L = \hbar \omega_L (S_{ee} + 1/2) \) represents the Hamiltonian of the laser mode with frequency \( \omega_L \). Therefore the global Hamiltonian of the system is given by:

\[
H = \hbar \omega_g S_{gg} + \hbar \omega_e S_{ee} + \hbar \omega_L \left( S_{ee} + \frac{1}{2} \right). \tag{2.2}
\]

Since \( S_{gg} + S_{ee} = 1 \), where 1 is the identity matrix \( \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \), the Hamiltonian above can be expressed in terms of the system’s resonance frequency, \( \omega_0 = \omega_e - \omega_g \), by subtracting the term \( \hbar \omega_g \mathbb{1} \) effectively moving the reference point. The Hamiltonian of the system then becomes:

\[
H = \hbar \omega_0 S_{ee} + \hbar \omega_L \left( S_{ee} + \frac{1}{2} \right), \tag{2.3}
\]
The eigenstates of $H$ are labeled by two quantum numbers: (1) the two-level system quantum numbers, $g$ or $e$, and (2) the number of laser photons, $N$. The states $|g, N\rangle$ and $|e, N - 1\rangle$ are close to each other when the detuning of the laser central frequency, $\Delta$, is very small compared to the resonance frequency of the two-level system, $|\Delta| \ll \omega_0$, where the detuning is given by:

$$\Delta = \omega_L - \omega_0.$$  \hfill (2.4)

This superposition between the two-level system and monochromatic electric field is illustrated in Figure 2.1.

Figure 2.1: Schematic representation of the superposition of a two-level system, with a resonance frequency $\omega_0$, driven at near-resonant condition by a monochromatic light field with frequency $\omega_L$, resulting in a manifold of two uncoupled states (e.g., $\mathcal{E}(N) = \{ |g, N\rangle , |e, N - 1\rangle \}$). The energy difference between the two states of each doublet depends on the detuning of resonance frequencies between the laser and the two-level system, $\Delta = \omega_L - \omega_0$. The order between the doublet states change depending on the sign of $\Delta$ (e.g., if $\omega_0 > \omega_L$, $|e, N\rangle$ is placed above $|g, N + 1\rangle$).
2.1.2 Coupled Hamiltonian and the Dressed State Picture

The Hamiltonian for the coupling between the two-level system and monochromatic electric field is in the form $H = H_0 + H_I$, where the interaction Hamiltonian $H_I$ in the electric dipole approximation is given by:

$$H_I = -\mu E(S_{eg} + S_{ge})$$

(2.5)

where $\mu$ is the transition dipole moment of the system, and $E$ is the applied electric field assumed to oscillate harmonically around the central frequency, $\omega_L$, that is:

$$E(t) = \frac{E_0}{2} (e^{i\omega_L t} + e^{-i\omega_L t})$$

(2.6)

The next stage is to find a modified effective Hamiltonian in the rotating frame, so that it satisfies the Schrödinger equation in all frames. The unitary matrix required for the transformation to the interaction frame is $U = \left( \begin{array}{cc} 1 & 0 \\ 0 & e^{-i\omega_L t} \end{array} \right)$. If we define a modified effective Hamiltonian in the rotating frame as $H_{rot} = UHU^\dagger - iU^\dagger \frac{dt}{dt}U$, it can be easily shown that this results in the same form for the Schrödinger equation in both the stationary and rotating frames. Thus, the corresponding matrix for the effective Hamiltonian in the rotating frame has the form:

$$H_{rot} = \begin{pmatrix} 0 & -\frac{\Omega_R}{2} - \frac{\Omega_R}{2} e^{-2i\omega_L t} \\ -\frac{\Omega_R}{2} - \frac{\Omega_R}{2} e^{2i\omega_L t} & -\Delta \end{pmatrix}$$

(2.7)

where $\Omega_R = \frac{\mu E_0}{\hbar}$ is the induced Rabi frequency of the laser mode which plays a big part in the separation between the uncoupled states shown in Figure 2.1.

Finally, we can get rid of the time dependency since, in a typical atomic system, the fast oscillating terms (i.e., those that oscillate at twice the optical frequency in the effective Hamiltonian) lead to weak excitations that can be neglected. This is known as the rotating-
wave approximation which yields a new time-independent Hamiltonian in the form:

\[
H = -\hbar \begin{pmatrix} 0 & \frac{\Omega R}{\Delta} \\ \frac{\Omega R}{\Delta} & \Delta \end{pmatrix}.
\] (2.8)

This Hamiltonian couples the states of each doublet in Figure 2.1 to each other such that, when this coupling is taken into account, the doublets become perturbed. These states are commonly referred to as “dressed” by the laser [81]. When the driven laser is at near-resonant condition, the separation between dressed states is dictated by the generalized Rabi frequency:

\[
\Omega = \sqrt{\Delta^2 + \Omega^2_R}.
\] (2.9)

For simplicity, we use integers ...0, 1, 2... to refer to the coupled states, and the quantum numbers + and − to label the upper and lower state of each doublet, as shown in Figure 2.2.

![Figure 2.2: Dressed state picture as a consequence the interaction between the two-level system and monochromatic laser field where the two states of each doublet in Figure 2.1 become “coupled” by the Rabi frequency of the electric field.](image-url)
2.1.3 Optical Bloch Equations

When dealing with mixed states it is specially helpful to describe the statistical state of a quantum system using a density matrix. The mixed state, $|\psi\rangle$, of a quantum system is represented to be in a statistical ensemble of different pure state vectors, $|\psi_n\rangle$. For a finite-dimensional system, the most general density operator is in the form:

$$\rho = \sum_n P_n |\psi_n\rangle\langle\psi_n|,$$  \hspace{1cm} (2.10)

where $P_n$ is the probability that the system is in the pure state $|\psi_n\rangle$. Let $S$ be an observable of the system, the expectation value of the measurement can be found as:

$$\langle S \rangle = \text{tr}\{S\rho\}.$$  \hspace{1cm} (2.11)

The density operator in Equation (2.10) can be written as a linear combination of the atomic operators in Equation (2.1) as:

$$\rho(t) = x_1(t)S_{eg} + x_2(t)S_{ge} + x_3(t)S_{gg} + x_4(t)S_{ee} = \begin{pmatrix} x_3(t) & x_2(t) \\ x_1(t) & x_4(t) \end{pmatrix}.$$  \hspace{1cm} (2.12)

Using Equation (2.11), one can find the expectation values of the atomic operators:

$$\langle S_{eg} \rangle = \text{tr}\{S_{eg}\rho(t)\} = x_2(t),$$

$$\langle S_{ge} \rangle = \text{tr}\{S_{ge}\rho(t)\} = x_1(t),$$

$$\langle S_{gg} \rangle = \text{tr}\{S_{gg}\rho(t)\} = x_3(t),$$

$$\langle S_{ee} \rangle = \text{tr}\{S_{ee}\rho(t)\} = x_4(t).$$  \hspace{1cm} (2.13)
From these we obtain the density operator:

\[
\rho(t) = \langle S_{ge} \rangle S_{eg} + \langle S_{eg} \rangle S_{ge} + \langle S_{gg} \rangle S_{gg} + \langle S_{ee} \rangle S_{ee} = \begin{pmatrix}
\langle S_{gg} \rangle & \langle S_{ge} \rangle \\
\langle S_{ge} \rangle & \langle S_{ee} \rangle
\end{pmatrix}.
\] (2.14)

The master equation describing the evolution of the density matrix can be described by a generalization of the quantum Louisville equation, known as the Lindbladian equation [82]:

\[
\frac{d\rho(t)}{dt} = -\frac{1}{\hbar}[H, \rho(t)] + \kappa \sum_{m,n} S_m \rho(t) S_n^\dagger - \frac{\kappa}{2} \sum_{m,n} \left( S_m^\dagger S_n \rho(t) + \rho(t) S_m^\dagger S_n \right),
\] (2.15)

where the first term is the usual Schrödinger term that generates the unitary evolution for a closed system, the second term describes the possible quantum jumps due to a radiative decay at a rate \(\kappa\) that the system may undergo due to interactions with the environment, and the third term is needed to normalize properly the case in which no jumps occur. By substituting Equations (2.8) and (2.14) into Equation (2.15), and making use of the definitions in Equation (2.1), we obtain a set of four equations known as the optical Bloch equations:

\[
\begin{align*}
\frac{d}{dt} \langle S_{ee} \rangle &= -\kappa \langle S_{ee} \rangle - \frac{i\Omega_R}{2} \langle S_{eg} \rangle + \frac{i\Omega_R}{2} \langle S_{ge} \rangle, \\
\frac{d}{dt} \langle S_{eg} \rangle &= -\frac{i\Omega_R}{2} \langle S_{ee} \rangle + \left( i\Delta - \frac{\kappa}{2} \right) \langle S_{eg} \rangle + \frac{i\Omega_R}{2} \langle S_{gg} \rangle, \\
\frac{d}{dt} \langle S_{ge} \rangle &= \frac{i\Omega_R}{2} \langle S_{ee} \rangle + \left( -i\Delta - \frac{\kappa}{2} \right) \langle S_{ge} \rangle - \frac{i\Omega_R}{2} \langle S_{gg} \rangle, \\
\frac{d}{dt} \langle S_{gg} \rangle &= \kappa \langle S_{ee} \rangle + \frac{i\Omega_R}{2} \langle S_{eg} \rangle - \frac{i\Omega_R}{2} \langle S_{ge} \rangle.
\end{align*}
\] (2.16)

The optical Bloch equation in the rotating-wave approximation given in Equation (2.16) may be rewritten in matrix form as:

\[
\frac{d}{dt} \mathcal{R}(t) = \mathcal{M} \cdot \mathcal{R}(t),
\] (2.17)
where $\mathbb{R}$ contains the expectation values of the observables and $\mathbb{M}$ is a single matrix capturing the parameters of the coupled system which may be used to find its dynamical evolution:

$$
\mathbb{R}(t) = \begin{pmatrix}
\langle S_{ee} \rangle \\
\langle S_{eg} \rangle \\
\langle S_{ge} \rangle \\
\langle S_{gg} \rangle 
\end{pmatrix}, \quad \text{and} \quad \mathbb{M} = \begin{pmatrix}
-\kappa & -i\frac{\Omega_R}{2} & i\frac{\Omega_R}{2} & 0 \\
-i\frac{\Omega_R}{2} & i\Delta - \frac{\kappa}{2} & 0 & i\frac{\Omega_R}{2} \\
i\frac{\Omega_R}{2} & 0 & -i\Delta - \frac{\kappa}{2} & -i\frac{\Omega_R}{2} \\
\kappa & i\frac{\Omega_R}{2} & -i\frac{\Omega_R}{2} & 0 
\end{pmatrix}, \quad (2.18)
$$

The solution of Equation (2.17) is important as it leads to the correlation functions required in the study of light-matter interactions in a two-level system.
Chapter 3

First-Order Photon Correlations

In the pursuit of the applications described in Section 1.3, the science of correlations between subsequently emitted photons from a light source has become a powerful investigative tool in the field of quantum optics. The concept of the $N$-photon spectrum, $g^{(N)}(t_1, \omega_1; \ldots; t_N, \omega_N)$, denotes the joint probability density of detecting photons at times $t_i$ with frequencies $\omega_i$, for $i = 1, 2, ..., N$ [83–85]. Starting at the simplest case where $N = 1$, the seminal work from Benjamin Mollow provided the first complete theoretical description of the spectrum of light from resonance fluorescence.

3.1 The Mollow Triplet

The first-order correlation function is proportional to the correlation of atomic operators when the two-level system is in equilibrium with the driving field, $g^{(1)}(t_1) \equiv \langle a^\dagger(t)a(t + t_1) \rangle$. In 1969, Mollow showed that the power spectrum (the distribution of the signal’s power in frequency bins) of resonance fluorescence can be obtained by performing a Fourier transform of the first-order correlation function, that is:

$$\tilde{g}^{(1)}(\omega_1) \equiv \int_{-\infty}^{\infty} g^{(1)}(t_1)e^{i\omega_1 t_1} dt_1 .$$ (3.1)

The expression for the “one-photon spectrum” is then given explicitly by Mollow [86]:

$$g^{(1)}(\omega_1) = 2\pi |\alpha_\infty|^2 \delta(\omega_1 - \omega_L) + n_\infty \kappa \omega_1^2 \left( \frac{(\omega_1 - \omega_L)^2 + (\omega_1^2/2 + \kappa^2)}{|f(i(\omega_1 - \omega_L))|^2} \right)$$ (3.2)
where the steady-state probability of finding the two-level system in its excited state $n_\infty$, the quantum mechanical expectation value of the two-level coherence $\alpha_\infty$, and the third-order polynomial $f(s)$ are given by:

$$n_\infty = \omega_1^2 \left( \frac{1}{4\Delta^2 + \kappa^2 + 2\omega_1^2} \right), \quad \alpha_\infty = i\omega_1 \left( \frac{\kappa + 2i\Delta}{4\Delta^2 + \kappa^2 + 2\omega_1^2} \right),$$

$$f(s) = s^3 + (\omega_1^2 + \Delta^2 + 5\kappa^2/4)s + \kappa(\omega_1^2/2 + \Delta^2 + 2\kappa + \kappa^2/4).$$

For a two-level system under low incident field power compared to the natural decay rate (that is, $\Omega \leq \kappa$), the spectral lineshape of the power spectrum consists of a single peak situated at the frequency of the driving laser $\omega_L$. Meanwhile, for a two-level system under an intense driving field (i.e., $\Omega \gg \kappa$), the fluorescence profile consists of a peak centered at the laser frequency, $\omega_L$, and peaks at the displaced frequencies $\omega_L \pm \Omega$, with widths proportional to the decay rate $\kappa$, as shown in Figure 3.1 for a system with decay rate $\kappa = 0.2$ GHz.

![Figure 3.1: Power spectrum for a two-level system driven exactly on resonance ($\Delta = 0$), with decay rate $\kappa = 0.2$ GHz, for different Rabi frequencies as given by Benjamin Mollow [86] through Equation (3.2).](image)

These peaks, commonly known as the “Mollow triplets”, are a consequence of the Autler-Townes effect (or AC Stark effect). For a system under low power illumination ($\Omega \leq \kappa$) driven
exactly at resonance ($\Delta = 0$), each doublet in the resulting manifold shown in Figure 2.1 becomes a superposition of the uncoupled states, allowing the system only one possible transition at the laser frequency. Meanwhile in the high power regime, that is, when the Rabi frequency far exceeds the natural relaxation rate of the system ($\Omega \gg \kappa$), the light emission process may then be viewed as a cascade down the ladder of the dressed states where the Mollow triplet emerges out of the only four possible spontaneous transitions between successive doublets.

![Dressed state picture of a resonantly driven two-level system generating resonance fluorescence with a decay rate $\kappa$.](image)

Figure 3.2: Dressed state picture of a resonantly driven two-level system generating resonance fluorescence with a decay rate $\kappa$. In the low power regime ($\Omega \leq \kappa$), when $\Delta = 0$, only one possible transition is allowed at the laser frequency. At high field intensities, $\Omega \gg \kappa$, the four possible transitions between successive doublets result in the Mollow triplet lineshape of the power spectrum.

As shown in Figure 3.2, the resonance fluorescence process originating from the lower energy (lower frequency) peak of the Mollow triplet, $\omega_L - \Omega$, occur due to the transition from the lower state of a top doublet to the upper state of a bottom doublet of the manifold, $|1, -\rangle \rightarrow |0, +\rangle$. Meanwhile, the photons originating from the higher energy (higher frequency) peak can be viewed as a transition from the upper state of a top doublet to the lower state of a bottom doublet, $|1, +\rangle \rightarrow |0, -\rangle$. Finally, the central peak is a consequence of the doubly degenerate transitions from the upper state of the top doublet to the upper state of the bottom doublet, $|1, +\rangle \rightarrow |0, +\rangle$, and transitions from the lower state of the top doublet to the lower state of the bottom doublet, $|1, -\rangle \rightarrow |0, -\rangle$. 
3.2 Experimental One-photon Spectrum

Experimentally, the frequency-resolved first-order photon correlation function, $g_{\Gamma_f}^{(1)}(t_1, \omega_f)$, of a light source is readily obtained interferometrically by using a spectral filter (such as a Fabry-Perot interferometer) with bandwidth $\Gamma_f$. The first-order photon correlation function is then proportional to the count rate of one photo detector as a function of the filtered frequency, $\omega_f$.

![Diagram of experimental setup](image)

Figure 3.3: Schematic representation for the experimental setup for the measurement of the power spectrum from a resonantly driven two-level system.

In practice, however, complications such as the stabilization and uniformity of the properties of the spectral filter may introduce some non-idealities into the measurements. For instance, the filter’s resonance frequency will inevitably fluctuate during the measurement as will the filter bandwidth. In addition, the precise width of each spectral filter may be difficult to set to an arbitrary independent value, and to maintain over time. Small variations in filter bandwidths are particularly consequential when the filter bandwidth is near the natural linewidth of the emitter. In fact it was recently shown that, even in the weak excitation regime, the photon statistics of a filtered single QD’s resonance fluorescence depend dramatically on the filter bandwidth [87]. Therefore, the spectral bandwidth of the filter must be taken into account in the theoretical simulations.

Given the solution of Equation (2.17) can be written as:

$$R(t + t_1) = e^{eta t_1} \cdot R(t),$$

(3.3)
and making use of the quantum regression theorem [88], which states that the time evolution of an operator $O$ whose expectation value is known is given by:

$$\langle O(t + t_1) \rangle = \sum_i a_j(t_1) \langle O_j(t) \rangle ,$$  \hspace{1cm} (3.4)

we can calculate the first-order correlation function as:

$$g^{(1)}(t_1) = \langle e^{M_{t_1}}_{\mathcal{L}} \mathcal{H}, (e^{M_{t_1}}_{\mathcal{L}} \mathcal{H}, e^{-i\omega_L t_1}}. $$  \hspace{1cm} (3.5)

The resolution of the Fabry-Perot interferometer, $\Gamma_f$, is introduced into Equation (3.5) by multiplying the correlation function by $e^{-t_1\Gamma_f/2}$ and finding its Laplace transform. The expression for the spectrally filtered “one-photon spectrum” is given by Konthasinghe et al. [89]:

$$g^{(1)}(\omega_1) = 2 \text{Re} \left\{ \left[ \left( i(\omega_L - \omega_1) + \frac{\Gamma_f}{2} \right) \mathbb{1} - M \right]^{-1}_{(2,2)} n_\infty^e + \left[ \left( i(\omega_L - \omega_1) + \frac{\Gamma_f}{2} \right) \mathbb{1} - M \right]^{-1}_{(2,4)} \alpha_\infty^e \right\} ,$$  \hspace{1cm} (3.6)

where $\mathbb{1}$ is the $4 \times 4$ identity matrix, $M$ is the matrix capturing the inputs of the equation of motion for the system’s observables given in Equation (2.18), and $\left[ \cdots \right]_{(i,j)}$ denotes the element $(i,j)$ of the resulting matrix.

Additionally, the transition frequency of the two-level system being probed will fluctuate to some extent. This “spectral diffusion” results in inhomogeneous broadening, i.e., an averaging over different resonance frequencies. To account for the fluctuations of the resonance frequency, $\omega_0$, due to spectral diffusion [90, 91], as in Ref. [89], we assume a Gaussian distribution of the detuning between the QD resonance frequency and the laser frequency, $\Delta$ (centered at $\Delta_\mu = 0$ with a standard deviation $\Delta_\sigma = 2$ GHz), and integrating Equation (3.6)
over all possible random detunings:

\[
g^{(1)}_{\Gamma_f}(\omega_f) \propto \int g^{(1)}_{\Gamma_f}(\omega_f, \Delta)e^{-\frac{(\Delta - \Delta_\mu)^2}{2\Delta^2}} d\Delta. \tag{3.7}
\]

This results in a theoretical simulation of the Mollow triplet in the one-photon spectrum that demonstrates a closer interpretation to experimental results as shown in fig. 3.4.

Figure 3.4: Theoretical and experimental one-photon spectrum of the resonance fluorescence from a single QD. The allowed spontaneous transitions in the dressed-state picture are shown at their corresponding emitted frequency.

Although the first-order correlations provide invaluable information about the light and the mechanism underlying its generation, this measurement alone reveals little about the source itself. For example, a light source consisting of a single radiating atom, and a light source consisting of an ensemble of non-interacting radiating atoms could have identical \( g^{(1)}_{\Gamma_f}(\omega_f) \). To distinguish the two, a second-order photon correlation measurement is required.
Chapter 4

Second-Order Photon Correlations

The second-order correlation function, \( g^{(2)}(t_1, t_2) \), measures the time correlation between two photons emitted by a continuous source arriving at the detectors at times \( t_1 \) and \( t_2 \) respectively. The normalized \( g^{(2)}(t_1, t_2) \) can be expressed on the form of the photon creation and annihilation operators as:

\[
g^{(2)}(t_1, t_2) = \frac{\langle a^\dagger(t_1)a^\dagger(t_2)a(t_1)a(t_2) \rangle}{\langle a^\dagger(t_1)a(t_1) \rangle \langle a^\dagger(t_2)a(t_2) \rangle}
\] (4.1)

From an experimental point of view, photon arrival times can be measured using the Hanbury-Brown and Twiss (HBT) setup [5], wherein a beamsplitter is used to divide the direction of the emitted photons into two pathways, and each path is equipped with a detector connected to electronics that records the arrival time of each photon as shown in Figure 4.1. The second-order correlations between two photons has attracted significant attention in the

![Figure 4.1: Schematic representation of a Hanbury Brown & Twiss setup for the measurement of the time-delay between photons, \( \tau = t_2 - t_1 \). An incoming light is split in two pathways using a 50:50 non-polarizing beamsplitter, then photon arrival times are recorded by a detector at each path. A histogram is build from the time difference between detected photons to obtain the measurement of the second-order photon correlations, \( g^{(2)}(\tau) \).](image)
field of quantum optics as it serves as the primary tool to distinguish between coherent, incoherent, and quantum light sources.

To distinguish the type of light source one most deal with the time-delay between subsequently emitted photons. The second-order photon correlation, $g^{(2)}(\tau)$, is measured by creating a histogram corresponding to the frequency of the time difference between photon arrival times, $\tau = t_2 - t_1$. The lineshape of $g^{(2)}(\tau)$ contains unique features that are directly dependent on the type of photon emission process. For example, photons originating from incoherent light sources (blackbody radiation sources such as stars or incandescent light bulbs) have the tendency of arriving together at the detectors, exhibiting a peak within a short time-delay interval due to the “bunching” of photons, $g^{(2)}(0) > 1$. Meanwhile, photons emitted from a coherent light source (such as a laser) tend to arrive at the detectors randomly, exhibiting a flat line in the second-order correlations, $g^{(2)}(\tau) \approx 1$. On the other hand, photons emitted from a typical quantum source (a two-level system such as an atom, molecule, or quantum dot) tend to be well spaced, arriving at the detectors one at a time producing a characteristic "anti-bunching" dip in the second-order correlation function, $g^{(2)}(0) < 1$. This pattern makes clear the importance of the behaviour of the second-order photon corre-

![Figure 4.2: Second-order photon correlations for incoherent (square), coherent (triangle), and single-photon (circle) sources.](image-url)
lations at $\tau = 0$, however, the temporal aspect alone cannot determine whether photons are indistinguishable.

It is important for quantum applications that photons remain indistinguishable from one another in order to avoid nullifying the quantum effects and reducing the emitted photons to a mere classical stream of bits. This entails that the frequency is to be resolved in the second-order correlation measurements. Recent theoretical work by Del Valle et al. has laid out a framework for studying these seemingly hidden correlations in a way that can be compared in a straightforward manner with experiments [92].

Following the supplemental material in Ref. 92, to compute the correlation between atomic operators $\langle a^\dagger(t)a(t + t_1)\rangle$, a vector $\mathbf{v}_{X,Y}(t_1)$ is defined for any two operators $X$ and $Y$ acting on the Hilbert space:

$$
\mathbf{v}_{X,Y}(t) = \begin{pmatrix}
\langle X(0)Y(0) \rangle \\
\langle X(0)a(t_1)Y(0) \rangle \\
\langle X(0)a^\dagger(t_1)Y(0) \rangle \\
\langle X(0)(a^\dagger a)(t_1)Y(0) \rangle
\end{pmatrix}.
$$

The dynamical evolution of $\mathbf{v}_{X,Y}(t_1)$ is ruled by the matrix $\mathbb{M}$:

$$
\partial_t \mathbf{v}_{X,Y}(t_1) = \mathbb{M} \mathbf{v}_{X,Y}(t_1); \quad \text{where} \quad \mathbb{M} = \begin{pmatrix}
0 & 0 & 0 & 0 \\
-\frac{\Omega}{2} & -\frac{\kappa}{2} - i\Delta & 0 & \Omega \\
-\frac{\Omega}{2} & 0 & -\frac{\kappa}{2} + i\Delta & \Omega \\
0 & -\frac{\Omega}{2} & -\frac{\Omega}{2} & -\kappa
\end{pmatrix},
$$

so that the steady-state observables for the two-level system are given by:

$$
\mathbf{v}^{\text{ss}} = \lim_{t \to \infty} \mathbf{v}_{1,1}(t_1) = \lim_{t \to \infty} e^{\mathbb{M}t_1} \begin{pmatrix}
1 \\
0 \\
0 \\
0
\end{pmatrix}.
$$
Two matrices $T_{\pm}$ are then defined which introduce an extra $a^\dagger$ for $T_+$ and an extra $a$ for $T_-$ between $X$ and $Y$ when acting on $v_{X,Y}(t_1)$:

$$
T_+ v_{X,Y}(t_1) = \begin{pmatrix}
\langle X(0)a^\dagger(t_1)Y(0) \rangle \\
\langle X(0)(a^\dagger a)(t_1)Y(0) \rangle \\
\langle X(0)a^{t_2}(t_1)Y(0) \rangle \\
\langle X(0)(a^{t_2}a)(t_1)Y(0) \rangle
\end{pmatrix},
$$

and

$$
T_- v_{X,Y}(t_1) = \begin{pmatrix}
\langle X(0)a(t_1)Y(0) \rangle \\
\langle X(0)a^2(t_1)Y(0) \rangle \\
\langle X(0)(a^\dagger a)(t_1)Y(0) \rangle \\
\langle X(0)(a^\dagger a^2)(t_1)Y(0) \rangle
\end{pmatrix}.
$$

Since $a^\mu a^\nu = 0$ for any $\mu$ or $\nu > 1$, these matrices read:

$$
T_+ = \begin{pmatrix}
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix}
\quad \text{and} \quad
T_- = \begin{pmatrix}
0 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix}.
$$

With Equations (2.18), (4.4) and (4.7), the one-photon spectrum may be obtained through the Fourier transform of $\langle a^\dagger(t)a(t+t_1) \rangle$. As given by Del Valle et al., the power spectrum in its integral form is given by:

$$
g_{1}^{(1)}(\omega_1) = \frac{1}{\pi} \Re \left[ T_+ \frac{1}{\mathbb{M} + (-i\omega_1 - \Gamma_1^2/i^2)} T_- v_{ss} \right]_1, \quad (4.8)
$$

where $[\cdot]_1$ denotes the first element of a vector. However, while the one-photon spectrum can be readily obtained by Equation (4.8), the computation of the correlation function becomes challenging even at the second order [93]. The pioneering theoretical work of Del
Valle et al. has introduced a method that allows to compute this quantity without the approximations performed before, making calculations manageable even at high orders.

4.1 Sensing Method

The correlations between $N$ photons can be computed exactly using the “sensors method” introduced by Del Valle et al. [92]. This method consists in the introduction of a set of sensors weakly coupled to the dynamics of the open quantum system through the resonance fluorescence as shown in Figure 4.3.

![Diagram of sensors theory](image)

Figure 4.3: Schematic representation of the sensors theory of frequency-resolved photon correlations. This theory consists on the introduction of $N$ sensors, each with bandwidths $\Gamma_i$, and transition frequencies $\omega_i$, weakly coupled to the two-level system (TLS) its resonance fluorescence with strength $\varepsilon_i$ assumed small enough such that the sensors leave the TLS undisturbed.

In the simplest case, each sensor $n_i$ (for $i = 1, 2, \ldots, N$) is modeled as a two-level system with resonance frequency $\omega_i$, bandwidth $\Gamma_i$, and creation and annihilation operators $\zeta_i^\dagger$ and $\zeta_i$ such that $\langle n_i(t_i) \rangle = \langle \zeta_i^\dagger \zeta_i(t_i) \rangle$. Sensors correlations are then computed in the limit of their
vanishing coupling \( \varepsilon_i \), by introducing a sensing vector \( \mathbf{w} \) of steady-state correlators:

\[
\mathbf{w}_{[\mu_1\nu_1, \ldots, \mu_N\nu_N]} = \begin{pmatrix}
\langle \varsigma_{\mu_1}^\dagger \varsigma_{\nu_1} (t_1) \ldots \varsigma_{\mu_N}^\dagger \varsigma_{\nu_N} (t_N) \rangle \\
\langle \varsigma_{\mu_1}^\dagger \varsigma_{\nu_1} (t_1) \ldots \varsigma_{\mu_N}^\dagger \varsigma_{\nu_N} (t_N) a \rangle \\
\langle \varsigma_{\mu_1}^\dagger \varsigma_{\nu_1} (t_1) \ldots \varsigma_{\mu_N}^\dagger \varsigma_{\nu_N} (t_N) a^\dagger \rangle \\
\langle \varsigma_{\mu_1}^\dagger \varsigma_{\nu_1} (t_1) \ldots \varsigma_{\mu_N}^\dagger \varsigma_{\nu_N} (t_N) a^\dagger a \rangle
\end{pmatrix},
\]

(4.9)

where the indices \( \mu_i \) and \( \nu_i \) take values of 0 or 1. In the supplemental material to Ref. 92, Del Valle \textit{et al.} provided the system’s equation of motion in the couplings \( \varepsilon_i \) derived from a master equation, which reads:

\[
\partial_t \mathbf{w}_{[\mu_1\nu_1, \ldots, \mu_N\nu_N]} = \left[ \mathcal{M} + \mathbb{1} \sum_{i=1}^{N} \left( (\mu_i - \nu_i) i\omega_i - (\mu_i + \nu_i) \frac{\Gamma_i}{2} \right) \right] \mathbf{w}_{[\mu_1\nu_1, \ldots, \mu_N\nu_N]} \\
+ \sum_{i=1}^{N} \left( i\varepsilon_i \mu_i T_+ \mathbf{w}_{[\nu_i, \ldots, \mu_N\nu_N]} - i\varepsilon_i \nu_i T_- \mathbf{w}_{[\mu_i, \ldots, \mu_N\nu_N]} \right),
\]

(4.10)

Using this equation of motion, any relevant correlation function, \( g^{(N)}(t_1, \omega_1; \ldots; t_N, \omega_N) \), can be obtained by applying the quantum regression theorem. For example, recursively computing the steady-state solutions (solutions at zero-time delay, that is \( t_1 = t_2 = \ldots = t_N = 0 \), Equation (4.10) becomes:

\[
\mathbf{w}_{[\mu_1\nu_1, \ldots, \mu_N\nu_N]}^{ss} = \frac{1}{-\mathcal{M} - \mathbb{1} \sum_{i=1}^{N} \left[ (\mu_i - \nu_i) i\omega_i - (\mu_i + \nu_i) \frac{\Gamma_i}{2} \right]} \\
\times \sum_{i=1}^{N} \left( i\varepsilon_i \mu_i T_+ \mathbf{w}^{ss}_{[\nu_i, \ldots, \mu_N\nu_N]} - i\varepsilon_i \nu_i T_- \mathbf{w}^{ss}_{[\mu_i, \ldots, \mu_N\nu_N]} \right),
\]

(4.11)

down to the vector \( \mathbf{w}^{ss}_{[00, \ldots, 00]} = \mathbf{v}^{ss} \). The one-photon spectrum through the sensors method can then be calculated by:

\[
\tilde{g}_{\Gamma_1}^{(1)}(\omega_1) = \Re \left[ \mathbf{w}^{ss}_{[11]} \right]_1.
\]

(4.12)
Meanwhile, the steady-state two-photon spectrum may be easily computed by:

\[
g^{(2)}_{\Gamma_1, \Gamma_2}(\omega_1, \omega_2) = \Re \left[ \frac{\omega_{ss}^{11}}{g^{(1)}_{\Gamma_1}(\omega_1)g^{(1)}_{\Gamma_2}(\omega_2)} \right]. \tag{4.13}
\]

Furthermore, the steady-state \(N^{th}\)-photon spectrum for \(N \geq 2\) can then be found straightforwardly as:

\[
g^{(N)}_{\Gamma_1 \ldots \Gamma_N}(\omega_1, \ldots, \omega_N) = \Re \left[ \frac{\omega_{ss}^{11 \ldots 11}}{\prod_{n=1}^{N} g^{(1)}_{\Gamma_n}(\omega_n)} \right]. \tag{4.14}
\]

The importance of being able to observe photon correlations in both time and frequency, led us to the discovery of a new type of photon emission which had remained unnoticed.

### 4.2 The Two-photon Spectrum

Experimentally, the frequency-resolved second-order photon correlations, \(g^{(2)}_{\Gamma_1, \Gamma_2}(\tau, \omega_1, \omega_2)\), associated with the resonance fluorescence scattered light can be measured by altering the HBT setup in Figure 4.1. Two filters, with bandwidths \(\Gamma_1\) and \(\Gamma_2\) respectively, are placed at each pathway such that the detected photons will have known frequencies, \(\omega_1\) and \(\omega_2\), as shown in Figure 4.4. The “two-photon spectrum” can then be obtained through a histogram of the photon arrival times for a matrix of the filtered photons at frequencies \(\omega_1\) and \(\omega_2\) [94].

![Figure 4.4: Schematic representation of the modified Hanbury Brown & Twiss setup for the measurement of correlations between two filtered photons.](image-url)
The two-photon spectrum measures the correlations between photons detected at $\tau = 0$ for all possible combinations of frequencies $\omega_1$ and $\omega_2$. This results in a rich two-dimensional landscape as shown in Figure 4.5, wherein the color code indicates the correlation statistics for bunched photons $g^{(2)} > 1$ (red), anti-bunched photons $g^{(2)} < 1$ (blue), and no correlations between photons $g^{(2)} = 1$ (white). This spectrum revealed that most of the interesting quantum emission processes arise away from the Mollow triplet peaks in the one-photon spectrum.

Figure 4.5: Two-photon resonance fluorescence spectrum, for photons filtered at frequencies $\omega_1$ and $\omega_2$ respectively, arriving simultaneously ($\tau = t_2 - t_1 = 0$) at the detectors. Theoretical simulation was performed using Equation (4.13) with filter bandwidths $\Gamma_1 = \Gamma_2 = 0.33$ GHz for a system with decay rate $\kappa = 0.2$ GHz driven at a Rabi frequency of $\Omega = 1.88$ GHz and assuming no detuning $\Delta = 0$.

Before further scrutinizing the origin of the features in the two-photon spectrum, it is helpful to compare it with its predecessor. A direct comparison between $g^{(1)}_{\Gamma_1}(\omega_1)$ and $g^{(2)}_{\Gamma_1,\Gamma_2}(\omega_1, \omega_2)$ can be observed by taking the auto-correlations of the two-photon spectrum, that is, correlations between photons emitted at the same frequency corresponding to the diagonal of Figure 4.5 at $\omega_1 = \omega_2$. 

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Figure 4.6: Theoretical and experimental measurements between (a) the one-photon spectrum and (b) the two-photon auto-correlation spectrum. The two-photon auto-correlations (i.e., correlations between photons emitted at the same frequency $\omega_1 = \omega_2$) corresponds to the diagonal of the two-photon spectrum in Figure 4.5. The allowed spontaneous dressed-state transitions are shown, each at their corresponding emitted frequency.

Figure 4.6 shows a direct comparison between (a) the first-order and (b) second-order auto-correlations measurements. In this juxtaposition, we can clearly observe the features in the two-photon auto-correlations, $g_{11}^{(2)}(\tau = 0, \omega_1 = \omega_2)$, occurring at the frequency of the Mollow triplet peaks. The origin of these features can be illustrated in the dressed state picture for the two-photon resonance fluorescence (Figure 4.7).

In this picture, disconnected pathways in the transitions between doublets result in photon anti-bunching since the system requires a change of its internal state to shift levels before a second transition can occur. This anti-bunching behaviour is seen with emissions
from the Mollow triplet sidebands at frequencies $\pm \Omega$. On the other hand, photon bunching from the central peak occurs due to successive transitions illustrated as connected pathways between states. In the full picture, however, Figure 4.6 reveals this bunching sitting at a local maximum.

The two-photon spectrum revealed some interesting features in the quantum emission process occurring away from the frequencies of the Mollow triplet peaks. Although the central peak is indeed bunched, this comes in a region of suppressed bunching, $g^{(2)} \approx 1$, meaning simultaneous transitions are likely, but at a low repetition rate. This occurs because the first transition simply reached a state in which is favorable for the second transition to take place. While photon bunching and anti-bunching features are observed at the Mollow triplet central peak and sidebands respectively, the most notable feature in the two-photon auto-correlation spectrum are the high resonances observed at the region in-between these peaks.

The high resonance between photons emitted in the region in-between peaks occurs due to another class of transition in the same ladder commonly known as “leapfrog transitions” [95].

Figure 4.7: Dressed-state picture formalism for the quantization of energy states by two-photons.
Rather than simply transitioning to a neighbouring doublet, leapfrog transitions in the ladder of states jump over one of these doublets. This happens because the second photon lifts the quantization of the spectrum to another state $\mathcal{E}(N + 2)$ with frequency $\frac{E}{h} = \omega_1 + \omega_2$. This allows for new transitions in the dressed-states picture to occur through an intermediate “virtual” (highly unstable) state as shown in Figure 4.7. The high instability of the virtual state then allows for subsequent photons to be emitted instantaneously exhibiting photon “superbunching” in the two-photon spectrum, $g^{(2)}(\omega_f) \gg 1$.

Similar results are generalized when turning to higher orders. While the correlations of the peaks retain the same qualitative behaviours, new features associated to the leapfrog transitions thus appear away from the peaks [95]. This have led scientists to explore correlations at higher orders.
Single-photon sources are at the core of the highly sought after quantum technology, utilizing photons as the information carriers. One of the cornerstones of quantum theory is the principle that any property of an object cannot be measured without affecting the object itself. In quantum optics, measuring the light field means destroying the photon leaving only vacuum behind. This principle is an important feature in quantum cryptography as it removes the possibility of an eavesdropper. However, for other applications such as ghost imaging and quantum teleportation, it is useful to employ a bundle of $N$-photons. With a bundle of $N$-photons, one photon from the bundle may be measured to ‘herald’ the existence of the others. For this purpose, there has been increasing interest in correlation measurements which go beyond the second order.

In Chapter 4, we have seen how a single-photon source is characterized by determining the value of the second-order photon correlations, $g^{(2)}(\tau)$, at $\tau = t_2 - t_1 = 0$ to evaluate the suppression of multi-photon states. We also discussed how the frequency-resolved two-photon spectrum, $g^{(2)}_{\Gamma_1\Gamma_2}(\tau, \omega_1, \omega_2)$, reveals a new type of photon emission where the generation of a bundle of photons is also possible through the filtered resonance fluorescence from a two-level system. However, just like the two-photon correlations are necessary to measure the quality of detection of single-photons, three-photon correlations measurements are crucial to produce pure quantum states of light associated with two-photon generators.
The third-order photon correlation function, $g^{(3)}(t_1, t_2, t_3)$, measures the correlation between the arrival times involving three photons emitted by a continuous source. Early work using a streak camera showed strong photon bunching statistics in third-order correlation measurements of microcavity laser light \([96, 97]\). The third-order correlation function was shown to provide more refined information about non-classical light sources such as multiphoton differentiation \([98]\) and the ability to analyze components \([99]\). The aim of this chapter is to provide the initial steps towards constructing the full three-photon spectrum, $g^{(3)}_{\Gamma_1 \Gamma_2 \Gamma_3}(\tau_1, \tau_2; \omega_1, \omega_2, \omega_3)$, which constitute a coincidence “cube” of photons detected simultaneously (i.e., $\tau_1 = \tau_2 = 0$ where $\tau_1 = t_2 - t_1$ and $\tau_2 = t_3 - t_1$) as shown in Figure 5.1. Here we explore three-photon correlations which make up the three-photon spectrum from single quantum dot resonance fluorescence under strong monochromatic laser illumination. It is worth mentioning these measurements are not specific to quantum dots, but rather are a characteristic of the resonance fluorescence from a two-level system.

Figure 5.1: Three-photon resonance fluorescence spectrum for photons filtered at frequencies $\omega_1$, $\omega_2$ and $\omega_3$ respectively, arriving simultaneously ($\tau_1 = t_2 - t_1 = 0$ and $\tau_2 = t_3 - t_1 = 0$) at the detectors. Theoretical simulation was performed using Equation (4.14) with filter bandwidths $\Gamma_1 = \Gamma_2 = \Gamma_3 = 0.33$ GHz for a system with decay rate $\kappa = 0.2$ GHz driven at a Rabi frequency of $\Omega = 1.88$ GHz and assuming no detuning $\Delta = 0$. 

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5.1 Three-photon Auto-correlations Measurements

From an experimental point of view, photon arrival times between three photons can be measured from a setup based on a straightforward extension of the Hanbury-Brown and Twiss type measurement wherein light from a source is split into 3-channels, each channel equipped with a spectral filter and detector. This work focuses on InAs Quantum Dots grown by molecular-beam epitaxy held in a cryostat at a base temperature of 4 K and interacting with a resonant wave-guided monochromatic laser beam.

The simplest measurements to obtain from the three-photon spectrum are the three-photon auto-correlations, that is, measurements involving a single filter. In this section, we focus exclusively on third-order auto-correlation measurements which constitute a small subset of the data required to reconstruct the complete three-photon spectrum.

After briefly describing our experimental setup in Section 5.1.1, we report our measurements in a juxtaposition of the extracted N-Photon spectra of auto-correlations for \( N = 1, 2 \) and \( 3 \), and present the raw time third-order correlation density maps for the filtering frequencies \( \omega_f - \omega_L \approx \Omega/2 \) and \( \omega_f - \omega_L \approx \Omega \) in Section 5.1.2. Furthermore, in Section 5.1.3, theoretical simulations are presented for the ideal case of a two-level system under a more intense driving field, and provided a simple dressed-states interpretation to further understand the origin of its spectral features.

5.1.1 Experimental Setup

Here we probed epitaxially grown InAs QDs held in a cryostat (at a base temperature of 4 K) and strongly interacting with a resonant wave-guided monochromatic laser beam. Our setup, depicted in Figure 5.4, is optimized to efficiently collect the light scattered by a single QD while minimizing unwanted background laser scattering. This optimization was performed by using an orthogonal excitation/detection geometry where QDs were grown between two distributed Bragg reflectors of moderate reflectivity as shown in Figure 5.2 [100].
The resonance fluorescence from the QD is then collected by lens and a single spectral filter was placed in the path of the QD scattered light.

![Figure 5.2: Photograph of the inside of the cryostation illustrating the orthogonal excitation/detection geometry of the InAs quantum dots samples in a planar microcavity containing alternating layers of GaAs/AlAs Bragg mirrors.](image)

The home-made optical filter consists of a thick solid ethalon, a monolithic transparent cuboid made of fused silica with parallel surfaces reflectively coated [101]. When inserted into the path of the electromagnetic radiation, the etalon acts as an optical cavity with the transmission periodically varying with the optical frequency. In resonance, the reflections from the two surfaces cancel each other via destructive interference. The resonance frequency of the filter, \( \omega_f \), can be adjusted by controlling its temperature to change the index of refraction of the ethalon. For optimal heat distribution, the ethalon is placed between two

![Figure 5.3: Fabry-Perot etalon with adjustable resonance frequency between two thermoelectric coolers (TEC) for optimal heat distribution.](image)
thermoelectric coolers (as shown in Figure 5.3). The bandwidth of the filter, $\Gamma_f$, was fixed to approximately match the radiative decay rate of InAs QDs by adjusting the incidence angle of the laser in the cuboid. The filtered QD scattered light is then split in three pathways using two non-polarizing beam-splitters.

The filtered resonance fluorescence is then detected by single photon detector modules (Excelitas SPCM-AQRH-14). Tagging of photon arrival times was performed for each detection channel independently with the help of a router (PicoQuant PHR 800 router with 100 ns cable delay to avoid the router dead-time window) and time-tagging electronics (PicoQuant PicoHarp 300) at an overall detector-limited resolution of about 400 ps. Note that the router is used here for no purpose other than as a replacement for a more expensive multichannel time-tagging system.

Figure 5.4: Schematic representation of the experimental setup for the measurements of third-order photons auto-correlations. The light near-resonantly scattered by a strongly-driven QD excitonic two-level system was collected by a lens, spectrally filtered using a Fabry-Perot etalon, split in three ways by two non-polarizing beam splitters, and detected at an overall detector-limited resolution of about 400 ps. Time-tagging of photon arrival times was performed on each channel independently.
5.1.2 Auto-correlation Measurements

Similarly to its one-photon counterpart, a two-photon source can be characterized by the third-order photon auto-correlations, \( g_{(3)}^{\Gamma_f}(\omega_f, \tau_1, \tau_2) \). The third-order photon auto-correlations measures the time difference between the first and second detected photons, \( \tau_1 = t_2 - t_1 \), and the first and third detected photons, \( \tau_2 = t_3 - t_1 \), for all three photons filtered at the same frequency, \( \omega_f \), with a filter resolution \( \Gamma_f \). Due to the long exposure times needed to collect sufficiently third-order coincidence events (on the order of days), we focus here on two specific measurements, each with a different filter frequency setting.

From the two-photon auto-correlation spectrum, it is evident the areas of interest are located at the bunching, and antibunching, regions of the Mollow triplet (i.e., the regions in-between peaks, and at the sidebands, respectively). For this reason, we focused specifically on measurements of the third-order auto-correlations at the filter frequencies \( \omega_f - \omega_L = \Omega/2 \) and \( \omega_f - \omega_L = \Omega \). Figure 5.5, shows a juxtaposition of the theoretical and experimental correlations between one, two, and three photons detected at the same frequency window using a fixed Rabi frequency \( \Omega/2\pi = 1.88 \text{GHz} \) and a filter bandwidth \( \Gamma_f/2\pi = 0.33 \text{GHz} \). The radiative decay rate of the QD and the laser-QD detuning are assumed to be \( \kappa/2\pi = 0.2 \text{GHz} \) and \( \Delta = 0 \) respectively.

Figure 5.5 then reveals the three-photon spectrum exhibits significantly more pronounced photon anti-bunching at \( \omega_f - \omega_L = \Omega \) compared to its two-photon counterpart. Meaning the likelihood of the system emitting three photons simultaneously is far lower than the probability of emitting two photons concurrently at this frequency configuration. In addition, three-photon pathways through virtual states are found to lead to more strongly correlated emission than at second order at \( \omega_f - \omega_L = \Omega/2 \). To better appreciate the two experimental data points in Figure 5.5c, we turn to the raw three-photon time correlations illustrated by creating a histogram of the time-delays \( \tau_1 \) and \( \tau_2 \). These histograms are best visualized as a three-dimensional density plot as shown in Figure 5.6.
Figure 5.5: First (a), second (b), and third (c) order filtered resonance fluorescence spectrum at time-delays $\tau_1 = \tau_2 = 0$. Dressed-state diagrams illustrate the allowed transitions at each corresponding filter frequency.
In Figure 5.6(a-b), the experimental and theoretical three-photon time correlation maps (with a 1 ns bin width) are shown for a filter frequency $\omega_f - \omega_L \approx \Omega/2$. These maps reveal a strong coincidence peak at $\tau_1 = \tau_2 = 0$ demonstrating that, for this filter configuration, photons are more likely to be detected in bunches than spread over time. In addition, the map also reveals diagonal ridges which indicate events in which two photons are detected simultaneously and one photon is accidental are more frequent than events in which all three detectors “click” accidentally at different times. In a second configuration, namely for a filter frequency $\omega_f - \omega_L \approx \Omega$, dispersing correlations are detected producing anti-bunching troughs shown in Figure 5.6(c-d).

Figure 5.6: Third-order correlation maps are shown for experimental (a) and theoretical (b) data for a filtering frequency $\omega_f - \omega_L \approx \Omega/2$, and for experimental (c) and theoretical (d) data for a filtering frequency $\omega_f - \omega_L \approx \Omega$. Measurements were taken using a fixed Rabi frequency $\Omega/2\pi = 1.88$ GHz, a quantum dot radiating fluorescence at a decay rate of $\kappa/2\pi = 0.2$ GHz, and a filter bandwidth $\Gamma_f/2\pi = 0.33$ GHz. The detuning between laser and two-level system of is assumed to be $\Delta = 0$. 
The addition of a third photon into the system increases the quantization of the spectrum to yet another state $E(N+3)$ with frequency $\frac{E}{\hbar} = \omega_1 + \omega_2 + \omega_3$. Similar to its two-photon counterpart, a new peak was expected to arise in the three-photon spectrum at the filter frequency $\omega_f - \omega_L \approx \pm \Omega/3$ [95]. The absence of this feature in our experiment can be attributed to the low resolution of the used filter relative to the Rabi frequency. More precisely, a ratio between filter bandwidth and Rabi frequency in our experiment of $\Gamma_f/\Omega = 0.18$. This can be remedied by either decreasing the filter bandwidth or increasing the Rabi frequency in our experiment, such that the ratio between the filter resolution and the frequency of the laser Rabi oscillations is at least $\Gamma_f/\Omega \leq 0.1$ when the feature at $\pm \Omega/3$ initiates to become evident according to theory.

### 5.1.3 Discussion

To further scrutinize the three-photon auto-correlation measurements in Figures 5.5 and 5.6, we turn to simulated correlations where the only difference is an increased Rabi frequency, specifically $\Omega/2\pi = 15 \text{ GHz}$. With the increased laser power, the ratio between the filter resolution and the Rabi frequency of the driving field is now $\Gamma_f/\Omega = 0.02$. Under this condition, $g_{\Gamma_f}^{(3)}(\omega_f)$ then reveals that, what seemed to be single correlation peaks in Figure 5.5c, are actually two pairs of correlation peaks as shown in Figure 5.7b.

One set of peaks, at $\omega_f - \omega_L = \pm \Omega/2$, corresponds to emissions in which only two of the three photons are entangled by a transition through a virtual intermediate state while a third photon originates from a simultaneous but independent transition. This occurs due to the latter being left in a highly unstable state. Meanwhile, the relevant emission process are those occurring between states separated by two rungs were all three transitions are entangled. The new set of peaks on the three-photon auto-correlation spectra originating at the frequencies $\omega_f - \omega_L = \pm \Omega/3$ are associated with transitions in which all three photons are entangled. At these frequency windows, leap-frog transitions occur over three doublets in the dressed-states ladder via two virtual intermediate states, as shown in Figure 5.8.
Figure 5.7: Simulation for the normalized one-photon spectrum (a) the auto-correlated three-photon spectrum at zero time-delay (b) for which all parameters are identical to those in Figure 5.5 except for and increased Rabi frequency, $\Omega/2\pi = 15$ GHz.

The larger Rabi frequency relative to the filter bandwidth is the reason that the peaks can be separably viewed in the three-photon spectrum of Figure 5.7b but not in that of Figure 5.5c. While it is straightforward to increase the Rabi frequency in our experiments by increasing the power of the applied laser, the emission rate then becomes low enough that the overall recording time becomes impractical. This problem can likely be remedied by improving the collection and propagation efficiency, as well as the detector quantum efficiency in our experiments (combined these amount to $\approx 1\%$ here). In addition, other more complex approaches have been proposed to overcome the reduced emission rate with increased ...
Rabi frequency, such as the use of photonic nanowires [102], microlenses [103], broadband enhancement solution using bullseye structures [104], and using the Purcell effect of cavity quantum electrodynamics [105] to enhance the rate of emission. Then, according to the simulations of Figure 5.7b, correlations larger by many orders of magnitude than in the current experiments may be obtained.

For the presently considered case of three photons with the same frequency, the distinguishing pathways are the ones from the upper (lower) state of the top rung to the lower (upper) state of the bottom doublet which necessarily must proceed via two virtual intermediate states. These cascades require the photons to have frequencies of $\omega_L \pm \Omega/3$ and $\omega_L \pm \Omega/2$ as shown in Figure 5.8. To better understand the emission process in these cascades, we turn to simulated third-order time correlation maps.

Figure 5.9 shows the time correlation maps for the frequencies associated with the three-photon cascades involving virtual states corresponding to the set of peaks at $\Omega/2$ and $\Omega/3$. Accordingly, the transitions involving virtual states at $\Omega/2$ are associated with correlation maps exhibiting bunching ridges at times in which only two out of three photons are detected simultaneously while the third photon is accidental (i.e., at $\tau_1 = 0$, $\tau_2 = 0$, and at $\tau_1 = \tau_2$) as shown in Figure 5.9a. Meanwhile, the transitions involving virtual states at $\Omega/3$ are associated with correlations maps in which the ridges are nearly vanishing relative to the coincidence peak at $\tau_1 = \tau_2 = 0$ as shown in Figure 5.9b. The vanishing of the horizontal, vertical and diagonal ridges are consequence of the three-photon time-energy entanglement in which two-photon correlations are restricted.

An interesting prospect emerging from the $N$-photon spectrum measurement and analysis is the possibility of generating heralded emission of groups of photons of any desirable number. For example, when considering the pathways analyzed in Figure 5.9b which consist of three photons emitted subsequently, it is clear that no second such sequence is allowed immediately following the first since the system is left in a lower dressed state. Therefore, there is a tendency to emit in bundles in between other emission events. This feature has
been analyzed in detail theoretically and was explicitly verified using Monte Carlo simulations [92]. With the use of independently tunable filters, a large parameter space exists to select pathways for the generation of heralded $N$-photon sequences [95].

5.1.4 Conclusion

In conclusion, spectrally-filtered third-order photon auto-correlations were recorded for resonance fluorescence from an individual semiconductor QD. Only a single filter was used, thus yielding the auto-correlation cross-section of the three-photon spectrum as introduced by Del Valle et al. Our measurements establish a firmer foothold of this concept and show its utility for characterizing non-classical light sources.
Comparing correlations measured for second and third order it can nevertheless already be seen the advances of higher-order correlations. A three-photon spectrum provides more pronounced photon anti-bunching compared to the two-photon spectrum, when the filter frequency matches that of the Mollow triplet sidebands. In addition, three-photon transitions through virtual states corresponding to photons filtered at frequencies between the sidebands and the central peak are found to lead to more strongly correlated emission than at second order adding a new dimension to potential communication schemes in quantum information science applications [95]. Additionally, we discussed how three-photon frequency entanglement could be obtained under high resolution spectroscopy of the resonance fluorescence from a strongly driven two-level system.

This experimental investigation have resulted in a peer-reviewed journal: “Third-order frequency-resolved photon correlations in resonance fluorescence” [Nieves et al., Phys. Rev. B 98, 165432 (2018), 77]. In the next section we build up on this investigation through the measurements of photons cross-correlations at third-order, i.e. the use of independently tunable filters for each channel which brings about new experimental challenges.
5.2 Three-photon Cross-correlations Measurements

In Section 5.1, we focused on auto-correlation measurements of the three-photon spectrum of resonance fluorescence from a quantum dot, \( g^{(3)}_{\Gamma_f}(\tau_1, \tau_2; \omega_f) \). In other words, three-photon correlations made using only one filter, such that all three emitted photons are detected at the same frequency, \( \omega_1 = \omega_2 = \omega_3 = \omega_f \). However, the dressed-state picture in the simulation of Figure 5.9(a) shows that a three-photon coincidence can be measured while only two of the three photons are entangled. Therefore, although the photons reaching the detectors in this experiment are filtered at identical frequencies, it is possible to observe interference even if the photons are distinguishable.

To measure correlations between distinct photons, one must be able to distinguish the resonance frequencies of each photon and build a histogram of photon arrival times in order to obtain the third-order cross-correlations maps, \( g^{(3)}_{\Gamma_1 \Gamma_2 \Gamma_3}(\tau_1, \tau_2; \omega_1, \omega_2, \omega_3) \). That is, correlation measurements using three independently tunable filters, each with bandwidth \( \Gamma_i \), such that the detected photons have independently known frequencies \( \omega_i \), where \( i = 1, 2, 3 \) indicates each of the independent filters.

In this section, we focus exclusively on third-order cross-correlation measurements which constitute a small subset of the data required to reconstruct the complete three-photon spectrum shown in Figure 5.1.

5.2.1 Experimental Setup

The three-photon cross-correlation maps can be measured by altering the setup depicted in Figure 5.4 to include three independent tunable filters (instead of just one) in order to distinguish between detected photons. This extension is shown in Figure 5.10, both schematically (top) and the actual setup (bottom). A polarized beam-splitter was used to adjust the power of the excitation laser, and therefore manipulate the Rabi frequency to increased the separation between the Mollow triplet peaks.
Figure 5.10: Schematic (top) and actual (bottom) experimental setup for the measurement of three-photon cross-correlations.

Here we use three home-made filters, similar to the one used in Figure 5.4, consisting of Fabry-Perot etalons inserted between two thermo-electric coolers (as shown in the inset in Figure 5.10) in order to control the filter’s resonance frequencies $\omega_1$, $\omega_2$, and $\omega_3$. The bandwidth of each filter, $\Gamma_1/2\pi = 0.5$ GHz, $\Gamma_2/2\pi = 0.75$ GHz and $\Gamma_3/2\pi = 0.67$ GHz, were fixed to approximately match the linewidths of the peaks in the Mollow triplet by adjusting the incidence angle of the laser in the etalon.

Similar to the setup described in Section 5.1.1, the filtered resonance fluorescence is then detected by single photon detector modules while tagging of photon arrival times is performed for each detection channel independently at an overall detector-limited resolution of about 400 ps.
5.2.2 Cross-correlation Measurements

As with the auto-correlation measurements in Figure 5.6, cross-correlation maps were recorded by creating a histogram of photon arrival times from each channel, only this time for a set of configurations from filter frequencies ($\omega_1, \omega_2, \omega_3$). The three-photon cross-correlation maps are shown at the bottom of Figure 5.11 where the color code is chosen so that the anti-bunching statistics ($g^{(3)} < 1$) are shown in blue, the classical correlations ($g^{(3)} \approx 1$) in white, and the bunching statistics ($g^{(3)} > 1$) are indicated in red.

![Cross-correlation maps](image)

Figure 5.11: Experimental three-photon correlation maps as a function of time delays $\tau_1 = t_2 - t_1$ and $\tau_2 = t_3 - t_1$ with fixed Rabi frequency of $\Omega/2\pi \approx 1.88$ GHz for different filtering frequency configurations with filter bandwidths of $\Gamma_1/2\pi = 0.5$ GHz, $\Gamma_2/2\pi = 0.75$ GHz, and $\Gamma_3/2\pi = 0.67$ GHz. Numerals in (a) show areas with different permutations of a fixed order of photon detection events (e.g., quadrant I includes detection events for which $t_2 < t_1 < t_3$).

Experimentally, we obtained $g^{(3)}_{\Gamma_1 \Gamma_2 \Gamma_3}(\tau_1, \tau_2; \omega_1, \omega_2, \omega_3)$ by measuring the total coincidences at $\tau_1$ and $\tau_2$ (i.e., count of simultaneous detections) and normalizing by the total of accidentals in the same time bin (i.e., the coincidences between a time-delay interval in which detections
are uncorrelated, for example, the average of the total coincidences in the anti-diagonal region where $\tau_1 = -\tau_2$, away from $\tau_1 = \tau_2 = 0$):

$$g^{(3)}_{\Gamma_1,\Gamma_2,\Gamma_3}(\tau_1, \tau_2; \omega_1, \omega_2, \omega_3) = \frac{\text{Total Coincidences at } \tau_1, \tau_2}{\text{Average of Total Accidentals at } \tau_1 = -\tau_2 \neq 0}.$$  \hspace{1cm} (5.1)

The spectral lineshapes above the correlation maps in Figure 5.11 show the filter frequency configuration used for each correlation map relative to the power spectrum, wherein the resonance frequency of one filter is set to match one of the sidebands of the Mollow triplet while the other two filters are set to match the opposite sideband.

In contrast to the auto-correlation maps (Figure 5.6), the cross-correlation maps in Figure 5.11 contain a mixture of both bunching and anti-bunching statistics. It can be seen that correlations from photons originating from the same sideband remain as anti-bunching troughs, that is, the diagonal $\tau_1 = \tau_2$ in Figure 5.11a, the horizontal at $\tau_2 = 0$ in Figure 5.11b, and the vertical at $\tau_1 = 0$ in Figure 5.11c. Meanwhile, the remaining features correspond to coincidence ridges due to two-photon bunching from cross-correlated photons.

Furthermore, it can be seen that the nonclassical character at $\tau_1 = \tau_2 = 0$ in the auto-correlation maps in Figure 5.6, have turned into Poissonian statistics due to the superposition of the second-order auto-correlated and cross-correlated photon statistics. Table 5.1 provides a quantitative comparison of the two and three-photon correlations corresponding to the central feature from each correlation map.

**Table 5.1: Raw data for the indicated correlation maps showing the normalized 3rd and 2nd-order correlations at $\tau_1 = \tau_2 = 0$.**

<table>
<thead>
<tr>
<th>Figure</th>
<th>$g^{(3)}_{\Gamma_1,\Gamma_2,\Gamma_3}(0,0;\omega_1,\omega_2,\omega_3)$</th>
<th>$g^{(2)}_{\Gamma_1,\Gamma_2}(0;\omega_1,\omega_2)$</th>
<th>$g^{(2)}_{\Gamma_1,\Gamma_2}(0;\omega_1,\omega_3)$</th>
<th>$g^{(2)}_{\Gamma_1,\Gamma_2}(0;\omega_2,\omega_3)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.6a</td>
<td>$10.53 \pm 0.25$</td>
<td>$2.97$</td>
<td>$3.11$</td>
<td>$3.14$</td>
</tr>
<tr>
<td>5.6c</td>
<td>$0.13 \pm 0.04$</td>
<td>$0.50$</td>
<td>$0.50$</td>
<td>$0.49$</td>
</tr>
<tr>
<td>5.11a</td>
<td>$1.14 \pm 0.08$</td>
<td>$1.46$</td>
<td>$1.51$</td>
<td>$0.51$</td>
</tr>
<tr>
<td>5.11b</td>
<td>$1.14 \pm 0.14$</td>
<td>$1.51$</td>
<td>$0.48$</td>
<td>$1.48$</td>
</tr>
<tr>
<td>5.11c</td>
<td>$1.22 \pm 0.13$</td>
<td>$0.48$</td>
<td>$1.47$</td>
<td>$1.60$</td>
</tr>
</tbody>
</table>
5.2.3 Discussion

To better understand the experimental maps presented here, we may divide them into six regions corresponding to a fixed order of photon detection events as illustrated in Figure 5.11a. For example, the quadrant I (at the upper-left corner) corresponds to a photon detection order $t_2 < t_1 < t_3$, meaning that a photon from channel 2 is detected first, followed by a photon detected in channel 1, and the last photon detected will result from channel 3. Meanwhile, the quadrant II (at the lower-right corner) corresponds to a channel detection order for which $t_3 < t_1 < t_2$, meaning a photon in channel 3 is detected first, followed by a photon in channel 1, and then by a photon from channel 2. The remaining octants at the top-right and bottom-left corners correspond to the remaining orders of channel detection events $t_1 < t_2 < t_3$ for octant III, $t_1 < t_3 < t_2$ for octant IV, $t_3 < t_2 < t_1$ for octant V, and $t_2 < t_3 < t_1$ for octant VI. This order is the same for all correlation maps shown here.

The locations of interest, however, are at the boundaries between each region, at which two out of three photons are detected simultaneously, or right at the center of the map at $\tau_1 = \tau_2 = 0$, at which all three photons are detected at the same time. At these boundaries, the presence of bunching or anti-bunching depends on whether it corresponds to emission from alternating sidebands, or if the emission process involves consecutive emission from the same sideband.

As done in previous chapters, we turn to the dressed-states formalism to further understand the origin of the features in the cross-correlation maps in Figure 5.11. In the dressed-states picture shown in Figure 5.12, the resonance fluorescence process originating from the “long” sideband of the Mollow triplet, $\omega_L + \Omega$, can be viewed as a transition from the upper state of a top doublet to the lower state of a bottom doublet (e.g., $|1, +\rangle \rightarrow |0, -\rangle$). Meanwhile, the photons originating from the “short” sideband, $\omega_L - \Omega$, occur due to the transitions from the lower state of a top manifold to the upper state of a bottom manifold (e.g., $|1, -\rangle \rightarrow |0, +\rangle$). The two-photon and three-photon emission can then be visualized as a cascade down this ladder as illustrated in Figure 5.12.
The vertical, horizontal, and diagonal features of each correlation map in Figure 5.11 are an aftereffect of the two-photon cascades (Figure 5.12b). The two-photon cascade from cross-correlated photons accounts for the bunching that occurs due to connected transitions between states. On the other hand, disconnected pathways, and thus photon anti-bunching, are associated with the two-photon cascade from auto-correlated transitions, i.e., second-order events involving photons from the same sideband.

Meanwhile, the three-photon correlations consist of three possible cascades which contain a combination of disconnected and connected pathways (Figure 5.12a). The cascades composed of disconnected transitions originate when either the first or third photon originate

Figure 5.12: Dressed-state picture formalism for (a) three-photon correlations corresponding to the origin of central feature of all correlation maps in Figure 5.11, and (b) two-photon correlations corresponding to the origin of the vertical, horizontal, and diagonal features of each correlation map.
from the long sideband of the Mollow triplet. This results in the need of simultaneous emission of photons originating from the short sideband, however, since the internal state of the system must change before a second emission from the same sideband can take place, these cascades result in photon anti-bunching. In turn, it is more favorable for the system that the secondly emitted photon originates from the long sideband allowing the bunching of three photons. As such, the three-photon correlations corresponding to the the central feature of all cross-correlation maps in Figure 5.11, that is $g^{(3)}_{\Gamma_1,\Gamma_2,\Gamma_3}(0, 0; \omega_1, \omega_2, \omega_3)$, is a consequence of the superposition of all three possible cascades. This results in the suppressed bunching, $g^{(3)} \gtrsim 1$, as seen in Table 5.1.

These central features at $\tau_1 = \tau_2 = 0$ of the experimental maps constitute a small subset of the data required to reconstruct the complete three-photon spectrum of resonance fluorescence. Specifically, by repeating such measurements for a matrix of frequency triplets $(\omega_1, \omega_2, \omega_3)$ and logging the normalized coincidence rate, an entire $g^{(3)}_{\Gamma_1,\Gamma_2,\Gamma_3}(0, 0; \omega_1, \omega_2, \omega_3)$ coincidence "cube" would be obtained as shown in Figure 5.1. It is worth mentioning these measurements are not specific to quantum dots, but rather are a characteristic of the resonance fluorescence from a two-level system. In practice, however, many hurdles remain before such a quantity can be measured.

An obvious obstacle in the measurement of three-photon correlations are the impractically long recording times that would be required. This limitation could at least in part be remedied by improving the collection and propagation efficiency, as well as the detector quantum efficiency in our experiments, or by using more complex approaches, for instance those which employ photonic nanowires [102], microlenses [103], broadband enhancement solutions using bullseye structures [104] or in general make use of the Purcell effect of cavity quantum electrodynamics [105].

Another complication that may introduce non-idealities is the stabilization and uniformity of the properties of the spectral filters. For instance, the filters resonance frequencies will inevitably fluctuate during the measurement as will the filter bandwidth. In addition,
the precise width of each spectral filter may be difficult to set to an arbitrary independent value, and to maintain over time. Small variations in filter bandwidths are particularly consequential when the filter bandwidth is near the natural linewidth of the emitter. In fact it was recently shown that, even in the weak excitation regime, the photon statistics of a filtered single QD’s resonance fluorescence depend dramatically on the filter bandwidth [87].

In the measurements presented here, the filters had slightly different bandwidths, which explains in part the variations between the three panels in Figure 5.11. Were it not for slightly different properties of individual filters the three configurations would be equivalent to those obtained by simply switching electric cables between detectors and time-tagging electronics. Lastly, the transition frequency of the QD being probed will fluctuate to some extent. This “spectral diffusion” results in inhomogeneous broadening, i.e., an averaging over different QD resonance frequencies. To examine the consequences of spectral diffusion under various filter bandwidths, we turn to theoretical simulations of the one-photon spectrum using Equation (3.7).

Figure 5.13 shows the normalized power spectrum obtained experimentally by scanning a filter with bandwidth $\Gamma_f/2\pi = 0.33$ GHz, and the theoretical simulations for $g^{(1)}_{\Gamma_f}(\omega_f)$ using the same bandwidth as the experimental trace, and for $\Gamma_f/2\pi = 0.75$ GHz which matches the largest bandwidth involved in the cross-correlation maps in Figure 5.11. The theoretical power spectrum in the absence of spectral diffusion, obtained directly using Equation (3.6), is also included in order to observed its impact in the experimental measurements.

From Figure 5.13 it is clear that spectral diffusion substantially impacts the measured correlations. In particular, it leads to a greater central peak magnitude relative to the sideband magnitude compared to the case of a purely radiatively broadened two-level system. In that sense spectral diffusion causes an effectively reduced interaction strength since, during part of the measurement, the laser is off-resonance with the QD.

In addition, Figure 5.13 reveals how a larger filter bandwidth increases the spectral overlap between the tails of the central peak and the sidebands, even more so in the presence of
Figure 5.13: Experimental and theoretical Mollow triplet lineshape with Rabi frequency $\Omega/2\pi = 1.88$ GHz for filter bandwidths $\Gamma_f/2\pi = 0.33$ GHz (black) and $\Gamma_f/2\pi = 0.75$ GHz (red). Theoretical traces with (solid) and without (dashed) spectral diffusion (SD) are based on Equations (3.6) and (3.7) respectively, with a radiative decay rate $\kappa/2\pi = 0.2$ GHz.

spectral diffusion. This source of “contamination” may be in principle avoided by increasing the Rabi frequency, which was not possible in our current experimental configuration. Altogether, variations over time in the QD resonance frequency and in the filter properties reduce the degree of bunching and anti-bunching in the maps of Figure 5.11.

5.2.4 Conclusion

In summary, spectrally-filtered third-order photon cross-correlations were recorded for resonance fluorescence from an individual semiconductor quantum dot under strong laser illumination. In this section, we presented considerably more challenging measurements involving three separate filter configurations. Namely, those for which the resonance frequency of two of the filters matches the lower frequency sideband of the Mollow triplet, while the remaining filter is set at the opposite sideband. The resulting correlation maps provide a
stepping-stone towards constructing a complete three-photon spectrum. Furthermore, the revelation of a rich landscape of bunching and antibunching features opens the way for potential new quantum devices capable of producing two-photon bunching or antibunching on demand.

Additionally, our measurements highlight how, in practice, these measurements are sensitive to filter bandwidths and spectral fluctuations of the QD resonance frequency over time. Future progress will likely hinge upon technical filter improvements and operation at higher Rabi frequency. A complete three photon spectrum may be mapped for quantum dot resonance fluorescence, but also any other light source, so long as photon emission rates are large enough.

This experimental investigation have resulted in a peer-reviewed journal: “Third-order photon cross-correlations in resonance fluorescence” [Nieves et al., Phys. Rev. B 102, 155418 (2020), 78]. Future directions include measurements of the three-photon spectrum at different frequency configurations. Additionally, one order up, measurements for the fourth-order photon correlations, $g_{\tau_1,\tau_2,\tau_3;\Gamma_1\Gamma_2\Gamma_3\Gamma_4}^{(4)}(\omega_1,\omega_2,\omega_3,\omega_4)$, are possible through a simple extension of the setup in Figure 5.10 by using another beamsplitter in order to divide the light scattered by the QD in four pathways. This will allow us to move towards the first step of detecting pure three-photon states and provide more clear indication of the nonclassical character of the light emitted due to resonance fluorescence of a two-level system.
The resonance fluorescence originating from a two-level system has been shown to provide a unique opportunity to study a host of fascinating phenomena. The ability to control the photon emission from resonance fluorescence has sparked an increasing interest in the field of quantum optics due to their potential applications in the development of highly sought after quantum technologies. As such, the first-order and second-order photon correlations continue to be researched extensively providing invaluable information about the light and the mechanism underlying its generation. The main goal of this study was to investigate the majorly unexplored frequency-resolved third-order photon correlations of the resonance fluorescence from two-level system. This work presented experimental progress towards constructing the complete spectrally filtered three-photon spectrum from a single semiconductor quantum dot under strong monochromatic light illumination.

First, we investigated third-order autocorrelation measurements in which photons were identically filtered at the two regions of the Mollow triplet where the two-photon autocorrelation spectrum showed fascinating photon statistics. A comparison with the correlation maps computed using the "sensors method" introduced by Del Valle et al. [92] reveals faithful agreement with theory. The three-photon autocorrelation measurements demonstrated significantly more pronounced photon antibunching compared to its two-photon counterpart. In addition, three-photon pathways through virtual states are found to lead to more strongly correlated emission than at second order. We characterized the correlations asso-
cient with these virtual transitions, and compared them with correlations at other Mollow triplet frequency windows.

Additionally, we investigated third-order cross-correlations, where two out of three photons originates from one of the Mollow triplet sidebands while the third photon originate from the opposite sideband of the power spectrum. The three-photon cross-correlation measurements resulted in correlation maps with antibunching features as a consequence of correlations arising from the same sideband, and bunching ridges due to opposite sideband correlated photons. The revelation of a rich landscape of bunching and antibunching features opens the way for potential new quantum devices capable of producing two-photon bunching or antibunching on demand. Furthermore, our measurements highlight how, in practice, these measurements are sensitive to filter bandwidths and spectral fluctuations of the QD resonance frequency over time. Future progress will likely hinge upon technical filter improvements and operation at higher Rabi frequency. A complete three-photon spectrum may be mapped for quantum dot resonance fluorescence, but also any other light source, so long as photon emission rates are large enough.


Appendix A

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Appendix B
Author’s Work

B.1 List of Publications


B.1.1 Conference Proceedings


Yamil A. Nieves González was born in Humacao, Puerto Rico. He attended the University of Puerto Rico-Humacao as an undergraduate where he majored in Physics Applied to Electronics. In 2014, he affiliated with the American Physical Society (APS) Bridge Program at the Physics department of the University of South Florida, where he joined the Digital Holography & Microscopy laboratory and received the Masters (M.S.) degree in Applied Physics in August 2016. The following Fall, he joined the Solid State Quantum Optics laboratory mainly studying resonant light-matter interactions in epitaxially-grown semiconductor quantum dots.