ON THE SQUEEZING AND OVER-SQUEEZING OF PHOTONS

by

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Graduate Department of Physics
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Abstract

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Quantum mechanics allows us to use nonclassical states of light to make measurements with a greater precision than comparable classical states. Here an experiment is presented that squeezes the polarization state of three photons. We demonstrate the deep connection that exists between squeezing and entanglement, unifying the squeezed state and multi-photon entangled state approaches to quantum metrology. For the first time we observe the phenomenon of over-squeezing where a system is squeezed to the point that further squeezing leads to a counter-intuitive increase in measurement uncertainty. Quasi-probability distributions on the surface of a Poincaré sphere are the most natural way to represent the topology of our polarization states. Using this representation it is easy to observe the squeezing and over-squeezing behaviour of our photon states.

Work is also presented on two different technologies for generating nonclassical states of light. The first is based on the nonlinear process of spontaneous parametric downconversion to produce pairs of photons. With this source up to 200,000 pairs of photons/s have been collected into single-mode fibre, and over 100 double pairs/s have been detected. This downconversion source is suitable for use in a wide variety of multi-qubit quantum information applications. The second source presented is a single-photon source based on semiconductor quantum dots. The single-photon character of the source is verified using a Hanbury Brown-Twiss interferometer.
Dedication

To my family.
Acknowledgements

To everyone who has spent time with me in the lab: I apologize for constantly dancing in the same room as your sensitive interferometers.

On a personal level I would like to thank my girlfriend, Jaime Almond, for her support, love, and patience the past five years. I would also like to thank my parents for always being there when I needed them. They may not grasp the contents of this thesis, but they have a deep understanding of the truly important things in life. And of course there is my (not so) little sister, Lindsey Davis. Thank you for being the best sister a brother could ask for.

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my thesis, and a special thanks to Mandi Gould for being my dance mentor. *Veni, Vidi,*
*Lindi.*
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Publication List

The following papers were published during the completion of this Ph.D.


The following papers have been submitted or are in preparation


Photon! Photon!
With apologies to William Blake

Photon! Photon! Forming Flight,
In the lab as dark as night.
What nonlinear order Chi
Could spring entangled Symmetry?

In what distant laser dyes
Sparked thy subtle wave-like guise?
What on resonance transpires?
On what level lasing fires?

And what calcite, and what part?
Could squeeze thy spin with such art?
And when thy mode began to lock,
What short pulse, and what short clock

What the Amplitude? What the Phase?
Of this single-photon craze?
What the particle? What dread thought
In what paradox are we caught?

When the stars threw down their spears,
That travelled for a million years,
Did the correlations bunch?
Just like Twiss and the other’s hunch?

Photon! Photon! Forming Flight,
In the lab as dark as night.
What nonlinear order Chi
Could spring entangled Symmetry?

-Krister Shalm, 2009
Chapter 1

Background

1.1 Precision Measurements

measure:
ascertain the size, amount, or degree of (something) by using an instrument
or device marked in standard units: the amount of water collected is measured

Measurement lies at the heart of science. Whenever a leap forward in our ability
to make measurements occurs, new revolutions and advances take place. For example,
the invention of the microscope and telescope greatly expanded the range of human
experience from what we could see to encompass that which was previously hidden. The
microscopic world with its strange single-celled organisms was for the first time accessible,
and distant galaxies and stars began to reveal their mysteries.

Today our most accurate measuring devices are based on the interference of light. The
interferometer, first invented in 1881 by Albert Michelson [Michelson 81, Michelson 87],
is capable of measuring displacements a tiny fraction of the wavelength of light. The
“best” interferometer currently in operation is at the Laser Interferometry Gravitational
Observatory (LIGO). This interferometer, with arms over three kilometres in length, is
capable of measuring displacements as small as $10^{-18}$ m—less than one-thousandth the
width of a proton [Dowling 09]. This is nine orders of magnitude more sensitive than
Michelson's original interferometer 130 years ago. Using this precision LIGO hopes to soon detect the presence of gravitational waves.

Despite their impressive performance, current interferometers are restricted in their precision by the shot-noise limit\(^1\). In the early 1980s it was discovered that quantum states of light in an interferometer could beat the shot-noise limit using a technique known as “squeezing” [Caves 81, Caves 80]. Squeezed states that achieve more than a 10 db reduction in measurement uncertainty below the shot-noise limit have been demonstrated experimentally [Vahlbruch 08], and squeezed light is being considered for use in the next generation of LIGO [Goda 08]. Another approach to quantum interferometry is based on multi-photon entanglement [Scully 93, Dowling 98]. Using entangled states it is possible to reach the Heisenberg limit in measurement uncertainty—the ultimate bound dictated by quantum mechanics. Experiments exploring these multi-photon entangled states have been carried out by our group [Mitchell 04] and others [Walther 04, Afek 10, Nagata 07, Khoury 06].

The heart of this thesis is an experiment, presented in chapter 4, that highlights the deep relationship that exists between squeezed states and multi-photon entanglement. We create a variety of polarization entangled three photon states and study their properties using quasi-probability distributions on the surface of a Poincaré sphere. These distributions reveal the connection between squeezing and entanglement as well as previously unseen features. For the first time we observe “over-squeezing”, the counter-intuitive \(\text{increase}\) in measurement uncertainty when a system has been squeezed beyond a certain point. We show that the effects of over-squeezing can be overcome using a detection system sensitive to the quantum correlations present in the squeezed state.

To generate our entangled states, sources of single and multiple photons are required. A significant portion of my doctoral work involved investigating and developing single

\(^1\)The shot-noise limit is also known as the standard quantum limit. Both terms are used interchangeably throughout this thesis.
and multiple photon sources appropriate for use in quantum metrology and quantum information applications. In chapter 2 I discuss the multiple-photon sources I built based on the process of spontaneous parametric downconversion. In chapter 3 I outline the details of a single-photon source using semiconductor quantum dots in microcavities that I set up. The remainder of this chapter introduces the fundamental concepts used in the rest of this thesis. Topics covered include the estimation of an unknown phase, the operation of an interferometer, the origin of the shot-noise limit and the Heisenberg limit in measurement uncertainty, the connection between angular momentum and interferometry, and the introduction of spin-squeezed states and their representation as Wigner quasi-probability distributions on the surface of the Poincaré sphere.

1.2 The Harmonic Oscillator

The quantization of the electromagnetic field treats each optical mode as a linearly independent harmonic oscillator. Discrete excitations of these modes are called photons. It is the properties of the harmonic oscillator that set the limits on the precision and sensitivity achievable through interferometry. Here I will briefly recount some of the important results from the quantum harmonic oscillator that will be used later in this chapter. I will not cover the quantization of the electromagnetic field; instead I refer to the interested reader to Gerry and Knight’s excellent textbook *Introduction to Quantum Optics* for a more in depth discussion on the topic [Gerry 04].

1.2.1 The Quantization of the Harmonic Oscillator

In the classical version of the one-dimensional harmonic oscillator, a particle of mass $m$ is subject to a potential $V(x) = \frac{1}{2} m w^2 x^2$. Any system with a potential that is quadratic in $x$ can be described by a harmonic oscillator. A common example of such a system is a spring that oscillates with a frequency of $w^2 = \frac{k}{m}$ where $k$ is the stiffness of the spring.
Chapter 1. Background

The Hamiltonian for the 1-D harmonic oscillator is

\[ H = \frac{p^2}{2m} + \frac{1}{2}mw^2x^2, \quad (1.1) \]

where \( p \) is the momentum of the system. For this Hamiltonian, one can find the equations of motion and solve for the position of the system at any given time [Shankar 80]

\[ x(t) = 2A \cos(\phi - wt), \quad (1.2) \]

where \( \phi \) is the phase of the oscillator and \( A \) is the amplitude of the oscillations.

In the quantum version of the 1-D harmonic oscillator, the classical canonical variables \( x \) and \( p \) are replaced by the Hermitian operators \( \hat{x} \) and \( \hat{p} \) using the correspondence principle. Here \( \hat{x} \) and \( \hat{p} \) must satisfy the commutation relationship \([\hat{x}, \hat{p}] = i\hbar\). This yields the Hamiltonian

\[ \hat{H} = \frac{\hat{p}^2}{2m} + \frac{1}{2}mw^2\hat{x}^2. \quad (1.3) \]

Because the operators \( \hat{x} \) and \( \hat{p} \) do not commute, they must satisfy the following uncertainty relation:

\[ \Delta X \Delta P \geq \frac{\hbar}{2}. \quad (1.4) \]

It is convenient to introduce the non-Hermitian annihilation operator \( \hat{a} \) and creation operator \( \hat{a}^\dagger \):

\[ \hat{a} = \sqrt{\frac{mw}{2\hbar}} \left( \hat{x} + \frac{i}{mw}\hat{p} \right), \]
\[ \hat{a}^\dagger = \sqrt{\frac{mw}{2\hbar}} \left( \hat{x} - \frac{i}{mw}\hat{p} \right). \quad (1.5) \]

The annihilation and creation operators do not commute \([\hat{a}^\dagger, \hat{a}] = 1\). After rearranging we find:

\[ \hat{x} = \sqrt{\frac{\hbar}{2mw}} (\hat{a}^\dagger + \hat{a}), \]
\[ \hat{p} = i\sqrt{\frac{\hbar mw}{2}} (\hat{a}^\dagger - \hat{a}). \quad (1.6) \]
Now the Hamiltonian in equation 1.3 can be expressed in terms of the operators \( \hat{a} \) and \( \hat{a}^\dagger \)

\[
\hat{H} = \hbar \omega \left( \hat{a}^\dagger \hat{a} + \frac{1}{2} \right).
\] (1.7)

The eigenvalue equation for the Hamiltonian is

\[
\hat{H} |n\rangle = E_n |n\rangle
\] (1.8)

where \( |n\rangle \) is an eigenstate with eigenvalue \( E_n \). The eigenstate \( |n\rangle \) represents a mode that has been excited \( n \) times; for this reason \( |n\rangle \) is referred to as a number state\(^2\). The number states are orthogonal to one another

\[
\langle n' | n \rangle = \delta_{n',n},
\]

and form a complete basis

\[
\sum_n |n\rangle \langle n| = \hat{I}.
\] (1.9)

The action on the creation (annihilation) operator on \( |n\rangle \) adds (subtracts) a quanta of energy \( \hbar \omega \) to (from) the system. The energy of the harmonic oscillator is constrained to be positive, and the annihilation operator \( \hat{a} \) must be bounded from below

\[
\hat{a} |0\rangle = 0,
\] (1.10)

where \( |0\rangle \) is the quantum vacuum. When the Hamiltonian in equation 1.7 acts on the vacuum state, we obtain

\[
\hat{H} |0\rangle = \hbar \omega \left( \hat{a}^\dagger \hat{a} + \frac{1}{2} \right) |0\rangle = \frac{1}{2} \hbar \omega |0\rangle.
\] (1.11)

The vacuum is not empty; it contains energy that is subject to fluctuations.

It is convenient to introduce the number operator \( \hat{n} = \hat{a}^\dagger \hat{a} \) which has the following eigenequation:

\[
\hat{n} |n\rangle = n |n\rangle.
\] (1.12)

The number states are also eigenvectors of the number operator with eigenvalues that correspond to the number of excitations in the harmonic oscillator.

\(^2\)Often in quantum optics, number states are also called Fock states.
The action of the creation and annihilation operators on the number states yields:

\[
\hat{a} |n\rangle = \sqrt{n} |n-1\rangle,
\]

(1.13)

\[
\hat{a}^\dagger |n\rangle = \sqrt{n+1} |n+1\rangle.
\]

(1.14)

By repeatedly applying the creation operator to the ground state $|0\rangle$ of the system, it is possible to generate the number state

\[
|n\rangle = \frac{\hat{a}^n}{\sqrt{n!}} |0\rangle.
\]

(1.15)

Successive number states are separated by the same amount $\hbar \omega$ from one another much like rungs on a ladder. Repeatedly applying the creation or annihilation operators to the system moves one up and down this energy “ladder”. Consequently, the creation and annihilation operators are sometimes referred to as ladder operators.

### 1.2.2 Coherent States

The solution to the classical harmonic oscillator given in equation 1.2 represents a system that oscillates back and forth around a stable equilibrium. The quantum state that comes closest to the classical behaviour is the coherent state $|\alpha\rangle$. The coherent state is a Poissonian distribution of the number states, and is an eigenstate of the annihilation operator

\[
\hat{a} |\alpha\rangle = \alpha |\alpha\rangle,
\]

(1.16)

\[
|\alpha\rangle = e^{-\frac{1}{2} |\alpha|^2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle
\]

(1.17)

where $\alpha$ is a complex number. In 1963, in his seminal work on the quantum theory of light [Glauber 63], Roy Glauber introduced coherent states in to quantum optics. Glauber’s coherent states represent an oscillating electric field and describe the properties of light emitted from a laser.
When working with coherent states it is helpful to define a displacement operator

\[
\hat{D}(\alpha) = e^{(\alpha \hat{a}^\dagger - \alpha^* \hat{a})},
\]

\[
|\alpha\rangle = \hat{D}(\alpha) |0\rangle.
\]

The displacement operator is both Hermitian and unitary, \(\hat{D}^\dagger \hat{D} = \hat{D} \hat{D}^\dagger = 1\). In phase space, the action of the displacement operator is to “displace” the vacuum state away from the origin. A coherent state has the following properties:

\[
\langle \hat{n} \rangle = e^{-|\alpha|^2} \sum_n \frac{(\alpha^* \alpha)^n}{n!} n = |\alpha|^2 = \bar{n},
\]

\[
\langle \hat{n}^2 \rangle = e^{-|\alpha|^2} \sum_n \frac{(\alpha^* \alpha)^n}{n!} n^2 = |\alpha|^4 + |\alpha|^2;
\]

\[
\Delta \hat{n} = \sqrt{\langle \hat{n}^2 \rangle - \langle \hat{n} \rangle^2} = |\alpha| = \sqrt{\bar{n}},
\]

\[
\langle \alpha | \alpha \rangle = 1.
\]

The average number of photons, \(\bar{n}\), in a coherent state is \(|\alpha|^2\) and the fluctuations in the mean photon number is \(\sqrt{\bar{n}}\). The signal-noise ratio of a coherent state scales as \(\frac{\langle \hat{n} \rangle}{\Delta \hat{n}} = \sqrt{\bar{n}}\).

### 1.2.3 The Phase Operator

Let us take a closer look at the structure of the creation and annihilation operators. Consider the solution to the classical simple harmonic oscillator given in equation 1.24

\[
x(t) = 2A \cos(\phi - wt) = A(e^{iwt}e^{-i\phi} + e^{-iwt}e^{i\phi}).
\]

The time-dependent version of \(\hat{x}\) defined in equation 1.6 is:

\[
\hat{x}(t) = \sqrt{\frac{\hbar}{2mw}} (\hat{a}(t)^\dagger + \hat{a}(t)),
\]

\[
= x_0 (\hat{a}(0)^\dagger e^{iwt} + \hat{a}(0)e^{-iwt}),
\]

with \(x_0 = \sqrt{\frac{\hbar}{2mw}}\). Comparing the form of classical and quantum solutions for position, it is tempting to write the annihilation and creation operators in polar form. Dirac
introduced such a polar decomposition of the form [Dirac 81]:

\[
\hat{a} = e^{i\hat{\phi}}\sqrt{n}, \quad (1.26)
\]
\[
\hat{a}^\dagger = \sqrt{n}e^{-i\hat{\phi}}. \quad (1.27)
\]

Here $\phi$ is to be interpreted as the Hermitian phase operator. From the commutation relation $[\hat{a}, \hat{a}^\dagger] = 1$ we have

\[
\hat{a}\hat{a}^\dagger - \hat{a}^\dagger\hat{a} = 1,
\]
\[
e^{i\hat{\phi}}\sqrt{n}\sqrt{n}e^{-i\hat{\phi}} - \sqrt{n}e^{-i\hat{\phi}}e^{i\hat{\phi}}\sqrt{n} = 1,
\]
\[
e^{i\hat{\phi}}\hat{n} - \hat{n}e^{i\hat{\phi}} = e^{i\hat{\phi}}. \quad (1.28)
\]

By expanding the exponentials, it can be shown that this relation is satisfied when $[\hat{n}, \hat{\phi}] = i$. This implies that an uncertainty relationship should exist between number and phase

\[
\Delta\hat{n}\Delta\hat{\phi} \geq \frac{1}{2}. \quad (1.29)
\]

The problem is that $\hat{\phi}$ is not Hermitian and therefore not an observable. A true phase operator does not exist in quantum mechanics. It is not possible to directly measure a phase; it is only possible to indirectly estimate a phase. A second problem with the uncertainty relationship in equation 1.29 is that $\Delta\hat{\phi}$ can take on values larger than $2\pi$. The operator $\phi$ as defined is continuous over the range $-\infty ... \infty$, but a phase shift is periodic modulo $2\pi$. An uncertainty in phase larger than $2\pi$ is ill defined.

A great deal of effort has been made to derive alternate forms of the phase operator. For a more detailed discussion of the various approaches to constructing phase operators I refer the curious reader to [Ban 95, Hradil 92, Loudon 73, Gerry 04]. The difficulties developing a phase operator were first discussed at length by Susskind and Glogower who developed a kind of one-sided “unitary” phase operator from which the phase distribution for any state over the range $-\pi ... \pi$ can be calculated [Susskind 64, Carruthers 68].
Later work by Pegg and Barnett developed a Hermitian phase operator over a truncated Hilbert space [Barnett 86]. While these approaches do not fully overcome all of the problems plaguing the phase operator, they do support the implications of the uncertainty relationship defined in equation 1.29. Thus, despite the non-existence of a true phase operator in quantum mechanics, for the rest of this thesis the uncertainty relationship between number and phase will be used as a convenient “fiction”.

1.3 The Mach-Zehnder Interferometer

The Mach-Zehnder (MZ) interferometer is one of the most common types of interferometers in use [Mach 03]. While it differs in configuration from a Michelson interferometer, it has identical performance characteristics. In the MZ interferometer light impinges on a beamsplitter which splits it between two paths as shown in figure 1.1. The length difference between the two paths can be adjusted, changing the relative phase $\phi$ between them. The two paths are then interfered on a second beamsplitter and the outputs measured, allowing $\phi$ to be estimated. It is worth spending more time introducing the beamsplitter as it plays a central role in the operation of an interferometer.

1.3.1 The Beamsplitter

The beamsplitter is a fundamental tool in the quantum optician’s toolbox. In its simplest configuration a beamsplitter takes two input modes and couples them to two output modes. Light entering an input port is either reflected or transmitted, and the amplitudes for reflectance $r$ and transmittance $t$ are in general complex. The reflectance and transmittance coefficients must satisfy the following relations:

$$|r|^2 + |t|^2 = 1,$$  
$$r^*t + rt^* = 0.$$  

(1.30)  

(1.31)
Figure 1.1: A Mach-Zehnder interferometer. Light enters a Mach-Zehnder (MZ) interferometer through either port 1 or 2 of a 50:50 beamsplitter (BS) and is “split” between the two output modes 3 and 4. By adjusting the path length in mode 4, a relative phase shift $\phi$ between the two arms is introduced. Both modes are interfered on a second BS and the light exits via the two output ports 5 and 6 where photodetectors measure the intensity.
For a typical 50:50 beamsplitter, like those used in the Michelson and MZ interferometers, \(|r| = |t| = 1/\sqrt{2}\). In this instance light is equally likely to be transmitted or reflected. In general this does not need to be the case; beamsplitters with different ratios of transmittance to reflectance can be constructed.

Now that the relationship between the reflectance and transmittance is known, the action of the beamsplitter on the two input modes is:

\[
\hat{a}_3 = t\hat{a}_1 + r\hat{a}_2, \\
\hat{a}_4 = r\hat{a}_1 + t\hat{a}_2,
\]

(1.32)

where \(\hat{a}_1(2)\) are the annihilation operators of the input modes and \(\hat{a}_3(4)\) are the annihilation operators of the output modes. The form of the beamsplitter relations depends on the details of the physical system that implements them. A typical 50:50 beamsplitter is made up of a dielectric coating. Light that is reflected off of this coating picks up a phase shift of \(e^{i\pi/2} = i\) relative to the transmitted beam. In this case the beamsplitter relations are:

\[
\hat{a}_3 = \frac{1}{\sqrt{2}}(\hat{a}_1 + i\hat{a}_2), \\
\hat{a}_4 = \frac{1}{\sqrt{2}}(i\hat{a}_1 + \hat{a}_2).
\]

(1.33)

Rearranging these relations, we find

\[
\hat{a}_1 = \frac{1}{\sqrt{2}}(\hat{a}_3 - i\hat{a}_4), \\
\hat{a}_2 = \frac{1}{\sqrt{2}}(-i\hat{a}_3 + \hat{a}_4),
\]

(1.34)

and

\[
\hat{a}^\dagger_1 = \frac{1}{\sqrt{2}}(\hat{a}^\dagger_3 + i\hat{a}^\dagger_4), \\
\hat{a}^\dagger_2 = \frac{1}{\sqrt{2}}(i\hat{a}^\dagger_3 + \hat{a}^\dagger_4).
\]

(1.35)
Consider the case of a single photon entering port 1 of the beamsplitter and nothing (vacuum) in the second port. Here the beamsplitter relations give the following transformation of the state

\[ |1\rangle_1 |0\rangle_2 = \frac{1}{\sqrt{2}} (|1\rangle_3 |0\rangle_4 + i |1\rangle_4 |0\rangle_3). \] (1.36)

It may be tempting to disregard the second input port as “nothing” enters it. Such a device would not be unitary: it implies that two new modes can be created from a pre-existing single mode. In linear optics one is restricted to transforming, coupling, and manipulating pre-existing modes. The vacuum mode in equation 1.36 is an integral part of the relationship.

1.3.2 A Single-Photon in a MZ Interferometer

We now consider a MZ interferometer where photons are sent in one at a time. Here each photon enters port 1 of the first beam splitter and is “split” between paths 3 and 4 with equal probability. A mirror in the upper arm of the interferometer can move, altering the length of path 4 with respect to path 3. For photons at a wavelength of \( \lambda \), the displacement of the mirror by an amount \( x \) leads to a phase shift of \( \phi = 2\pi x/\lambda \).

\[ |1\rangle_1 |0\rangle_2 = \frac{1}{\sqrt{2}} (|1\rangle_3 |0\rangle_4 + i |1\rangle_4 |0\rangle_3) \]
\[ \rightarrow \frac{1}{\sqrt{2}} (|1\rangle_3 |0\rangle_4 + i e^{i\phi} |1\rangle_4 |0\rangle_3). \] (1.37)

After picking up a relative phase shift, the two paths are interfered on a second beamsplitter. Here paths 3 and 4 are the input modes of the second beamsplitter and modes 5 and 6 are the outputs

\[ \frac{1}{\sqrt{2}} (|1\rangle_3 |0\rangle_4 + i e^{i\phi} |1\rangle_4 |0\rangle_3) = \frac{1}{2} (|1\rangle_6 |0\rangle_5 (1 - e^{i\phi}) + i |0\rangle_6 |1\rangle_5 (1 + e^{i\phi})). \] (1.38)
At each of the output modes, a photodetector is placed that measures the intensity of light in each mode

\[
\langle \hat{n}_5 \rangle = \langle \hat{a}_5^\dagger \hat{a}_5 \rangle = \frac{1}{2} |i(1 + e^{i\phi})|^2 = \cos^2(\phi/2), \tag{1.39}
\]

\[
\langle \hat{n}_6 \rangle = \langle \hat{a}_6^\dagger \hat{a}_6 \rangle = \frac{1}{2} |(1 - e^{i\phi})|^2 = \sin^2(\phi/2). \tag{1.40}
\]

When the paths are balanced and \( \phi = 0 \), the photon always emerges in path 5. When the two arms are out of phase with one another (\( \phi = \pi \)), then the photon will always leave via port 6.

With only a single photon it is impossible to gain any information about the relative phase shift between the two arms. Instead, the experiment must be repeated in order for enough statistics to be collected so that the phase can be estimated. If the experiment is repeated \( N \) times the total signal measured by the detectors is given as:

\[
\langle \hat{n}_5 \rangle \otimes N = N \cos^2(\phi/2), \tag{1.41}
\]

\[
\langle \hat{n}_6 \rangle \otimes N = N \sin^2(\phi/2). \tag{1.42}
\]

For the purposes of estimating the phase, it is useful to look at the difference in the signal between the two output arms of the interferometer. The operator associated with this measurement can be written as:

\[
\hat{M} = \hat{n}_5 - \hat{n}_6
= \hat{a}_6^\dagger \hat{a}_6 - \hat{a}_5^\dagger \hat{a}_5. \tag{1.43}
\]

For a single photon the expectation value of \( \hat{M} \) is

\[
\langle \hat{M} \rangle = \langle \hat{n}_5 \rangle - \langle \hat{n}_6 \rangle
= \cos^2(\phi/2) - \sin^2(\phi/2) = \cos(\phi). \tag{1.44}
\]

Using the beamsplitter relations in equation 1.35 it is possible to back out the action
of the second beamsplitter on the measurement operator $\hat{M}$

$$\hat{M} = \hat{a}_3^\dagger \hat{a}_4 + \hat{a}_4^\dagger \hat{a}_3$$

$$\hat{M} = |1\rangle_3 \langle 0|_3 |0\rangle_4 \langle 1|_4 + |1\rangle_4 \langle 0|_4 |0\rangle_3 \langle 1|_3$$

$$\hat{M} = |1,0\rangle \langle 0,1|_{3,4} + |0,1\rangle \langle 1,0|_{3,4}, \quad (1.45)$$

where, for notational convenience, $|a\rangle_3 |b\rangle_4$ is rewritten as $|a,b\rangle_{3,4}$. The action of this operator is to measure the coherence between the two modes. In the density matrix formalism of quantum mechanics, where a state is represented as a density matrix instead of a state vector, this operator measures the off-diagonal elements which contain the coherence information.

1.3.3 Sensitivity of a Phase Measurement

It is important to quantify how accurately our apparatus can estimate a phase shift. If we assume our detection system is perfect, meaning the detectors have negligible noise, then we can study the intrinsic performance of the interferometer by itself. For small phase shifts, from the standard propagation of errors we obtain

$$\frac{\Delta \hat{M}}{\Delta \phi} = \left| \frac{\partial \langle \hat{M} \rangle}{\partial \phi} \right|, \quad \Delta \phi = \left| \frac{\Delta \hat{M}}{\partial \langle \hat{M} \rangle}{\partial \phi} \right|, \quad (1.46)$$

where $\Delta \hat{M}$ is the measurement uncertainty in $\hat{M}$ and $\Delta \phi$ is the uncertainty in $\phi$ [Lee 02b]. The uncertainty in the phase shift depends both on the uncertainty in detection as well as the slope of the detection signal with respect to the phase. The uncertainty in phase measurements can be improved by reducing $\Delta \hat{M}$ or by increasing the sensitivity of $\phi$ to changes in $\hat{M}$ (make the slope bigger).
1.3.4 The Sensitivity of Single Photons in a MZ Interferometer

We can now determine how accurately a train of single photons is able to estimate a phase shift in an interferometer. From equation 1.45 it is simple to verify that $\hat{M}^2 = \hat{I}$ for the single photon case. Using this fact and the results from equation 1.44 we find

$$\Delta\hat{M}^2 = \langle \hat{M}^2 \rangle - \langle \hat{M} \rangle^2$$

$$= 1 - \cos^2(\phi)$$

$$= \sin^2(\phi), \quad (1.47)$$

and from equation 1.44 we obtain

$$\left| \frac{\partial \langle \hat{M} \rangle}{\partial \phi} \right| = \left| \frac{\partial \cos(\phi)}{\partial \phi} \right| = |\sin(\phi)|. \quad (1.48)$$

Repeating the single-photon experiment $N$ times is analogous to flipping a weighted coin $N$ times (where the probability of obtaining “heads” is $\cos(\theta)$). The photons in successive experiments are uncorrelated, and the clicks between the two detectors at the output of the interferometer will be binomially distributed. For the $N$ samples the uncertainty in $\hat{M}$ will be proportional to $\sqrt{N}$. Repeating the experiment $N$ times yields:

$$\left( \Delta\hat{M}^{\otimes N} \right)^2 = N \sin^2(\phi), \quad (1.49)$$

$$\left| \frac{\partial \langle \hat{M}^{\otimes N} \rangle}{\partial \phi} \right| = N |\sin(\phi)|. \quad (1.50)$$

From this we find that the sensitivity of an interferometer with $N$ single-photons sent through it to be

$$\Delta \phi = \frac{\sqrt{N} |\sin \phi|}{N |\sin \phi|} = \frac{1}{\sqrt{N}}. \quad (1.51)$$

The uncertainty in the phase scales as the inverse square of the number of photons present. This bound is often referred to as the standard quantum limit (SQL) or the shot-noise limit (SNL).
1.3.5 The Sensitivity of a Coherent State in a MZ Interferometer

Instead of single photons, consider what happens when a coherent state is sent through a MZ interferometer. Before the coherent state enters the MZ interferometer it is in the state $|\alpha\rangle_1 |0\rangle_2$. It is convenient to use the displacement operator from equation 1.18 when working with coherent states. After hitting the first beamsplitter the coherent state is transformed to

$$
|\alpha\rangle_1 |0\rangle_2 = \exp\left(\frac{\alpha}{\sqrt{2}} (\hat{a}_3^\dagger + i\hat{a}_4^\dagger) - \frac{\alpha^*}{\sqrt{2}} (\hat{a}_3 - i\hat{a}_4)\right) |0\rangle_3 |0\rangle_4
$$

$$
= \exp\left(\frac{1}{\sqrt{2}} \left[ \alpha \hat{a}_3^\dagger \alpha^* \hat{a}_3 \right] \right) \exp\left(\frac{i}{\sqrt{2}} \left[ \alpha \hat{a}_4 - (-\alpha^*) \hat{a}_4 \right] \right) |0\rangle_3 |0\rangle_4
$$

$$
= \left| \frac{\alpha}{\sqrt{2}} \right\rangle_3 \left| \frac{i\alpha}{\sqrt{2}} \right\rangle_4.
$$

(1.52)

The action of the beamsplitter takes the input coherent state and splits it into two equal coherent states. The states in modes 3 and 4 are two independent (separable) coherent states. This is in contrast to the case with the single photon where the state of the system after the beamsplitter cannot be factored; the single-photon is left in a superposition of both output modes of the beamsplitter.

The coherent state in arm 4 picks up an extra phase shift of $e^{i\phi}$ as it propagates through the interferometer:

$$
\left| \frac{\alpha}{\sqrt{2}} \right\rangle_3 \left| \frac{i\alpha}{\sqrt{2}} \right\rangle_4 \rightarrow \left| \frac{\alpha}{\sqrt{2}} \right\rangle_3 \left| \frac{i e^{i\phi} \alpha}{\sqrt{2}} \right\rangle_4.
$$

(1.53)

After hitting the second beamsplitter, the state is transformed in the following way

$$
\left| \frac{\alpha}{\sqrt{2}} \right\rangle_3 \left| \frac{i e^{i\phi} \alpha}{\sqrt{2}} \right\rangle_4 = \exp\left(\frac{1}{2} \left[ \alpha \hat{a}_6^\dagger - \alpha^* \hat{a}_6 \right] \right) \exp\left(\frac{i}{2} \left[ \alpha \hat{a}_5^\dagger - (-\alpha^*) \hat{a}_5 \right] \right) |0\rangle_5 |0\rangle_6,
$$

$$
\left| \frac{i e^{i\phi} \alpha}{\sqrt{2}} \right\rangle_4 = \exp\left(-\frac{e^{i\phi}}{2} \left[ \alpha \hat{a}_6^\dagger - \alpha^* \hat{a}_6 \right] \right) \exp\left(\frac{e^{i\phi}}{2} \left[ \alpha \hat{a}_5^\dagger - (-\alpha^*) \hat{a}_5 \right] \right) |0\rangle_5 |0\rangle_6,
$$

$$
\left| \frac{\alpha}{\sqrt{2}} \right\rangle_3 \left| \frac{i\alpha}{\sqrt{2}} \right\rangle_4 = \left| \frac{i(1 + e^{i\phi})\alpha}{2} \right\rangle_5 \left| \frac{(1 - e^{i\phi})\alpha}{2} \right\rangle_6
$$

$$
= |\alpha_5\rangle_5 |\alpha_6\rangle_6.
$$

(1.54)
To estimate the phase shift the normalized difference in the signals between the detectors at port 5 and 6 is measured:

\[
M = \frac{\langle \hat{n}_5 \rangle - \langle \hat{n}_6 \rangle}{\langle \hat{n}_5 \rangle + \langle \hat{n}_6 \rangle}.
\]  

(1.55)

Using the relations defined in equations 1.20-1.23 we find

\[
\langle \hat{n}_5 \rangle = |\alpha_5|^2 = \left| \frac{i(1+e^{i\phi})\alpha}{2} \right|^2 = \bar{n} \cos^2(\phi/2),
\]  

(1.56)

\[
\langle \hat{n}_6 \rangle = |\alpha_6|^2 = \left| \frac{1-e^{i\phi}\alpha}{2} \right|^2 = \bar{n} \sin^2(\phi/2),
\]  

(1.57)

\[
\Delta n_5^2 = \langle \hat{n}_5 \rangle^2 = \bar{n} \cos^2(\phi/2),
\]  

(1.58)

\[
\Delta n_6^2 = \langle \hat{n}_6 \rangle^2 = \bar{n} \sin^2(\phi/2),
\]  

(1.59)

\[
M = \frac{\langle \hat{n}_5 \rangle - \langle \hat{n}_6 \rangle}{\langle \hat{n}_5 \rangle + \langle \hat{n}_6 \rangle} = \frac{\bar{n}}{\bar{n}} \cos(\phi) = \cos(\phi),
\]  

(1.60)

where again $|\alpha|^2 = \bar{n}$ is the average number of photons. Just as in the single photon case, the detected signal oscillates with a $\cos(\phi)$ dependence.

Using the propagation of errors we can find the variance in $M$:

\[
\Delta M^2 = \left( \frac{\partial M}{\partial \langle \hat{n}_5 \rangle} \right)^2 \Delta \hat{n}_5^2 + \left( \frac{\partial M}{\partial \langle \hat{n}_6 \rangle} \right)^2 \Delta \hat{n}_6^2
\]

\[
= \left( \frac{2\langle \hat{n}_6 \rangle}{(\langle \hat{n}_5 \rangle + \langle \hat{n}_6 \rangle)^2} \right)^2 \Delta \hat{n}_5^2 + \left( \frac{2\langle \hat{n}_5 \rangle}{(\langle \hat{n}_5 \rangle + \langle \hat{n}_6 \rangle)^2} \right)^2 \Delta \hat{n}_6^2
\]

\[
= \frac{4}{\bar{n}} \left( \sin^4(\phi/2) \cos^2(\phi/2) + \sin^2(\phi/2) \cos^4(\phi/2) \right)
\]

\[
= \frac{\sin^2(\phi)}{\bar{n}}.
\]  

(1.61)

The uncertainty in a phase measurement is then just

\[
\Delta \phi = \frac{\Delta M}{\frac{\partial M}{\partial \phi}} = \frac{|\sin \phi|}{\sqrt{\bar{n}} |\sin \phi|} = \frac{1}{\sqrt{\bar{n}}}.
\]  

(1.62)

Both single photons and coherent states are capable of reaching the standard quantum limit in phase sensitivity interferometrically [Lee 02b].
1.3.6 The N00N State

The coherent state is a minimum uncertainty state (MUS): it saturates the uncertainty relation in equation 1.29. For a coherent state, the uncertainty is evenly distributed between its conjugate variables. The origin of shot-noise is from the vacuum fluctuations that enter the “empty” port of the first beamsplitter in an interferometer. Carl Caves suggested that by “plugging” this empty port with another quantum state it would be possible to reduce the fluctuations, leading to an improvement in measurement uncertainty [Caves 81]. This approach leads to squeezed states: states of light where the uncertainty relation in equation 1.29 is manipulated so that the uncertainty in one variable is decreased at the expense of an increase in uncertainty in the conjugate variable. For a MUS, increasing the uncertainty in the photon number distribution between the two modes inside the interferometer leads to a corresponding decrease in the phase uncertainty. For a state with \( N \) photons and a total energy of \( E = N\hbar \omega \), the maximum uncertainty in the energy \( \Delta E \) should not exceed the total energy of the system. This means that at most \( \Delta E \sim O(E) \) and \( \Delta \hat{N} \sim O(N) \) which puts a lower bound on the Heisenberg limit of phase uncertainty [Caves 81, Ou 96, Ou 97, Dowling 09]:

\[
\Delta \hat{\phi} \gtrsim \frac{1}{N}.
\]  

(1.63)

There have been several schemes proposed for states that are able to reach this Heisenberg limit [Yurke 86, Caves 81]. In this section I will focus on a particular state that is capable of reaching the Heisenberg limit in phase uncertainty [Holland 93, Scully 93, Ou 97, Dowling 98, Gilbert 10]. An ordinary beamsplitter leads to a binomial distribution of the photons between the output modes: each photon has a 50:50 chance of ending up in either path much like a flipped coin has even odds of landing heads or tails. Replacing this 50:50 BS with a “magic” beamsplitter that takes all \( N \) input photons and puts them in a superposition of either all \( N \) photons being in one arm or the other of
the interferometer produces the state
\[ |N :: 0\rangle = \frac{1}{\sqrt{2}} \left( |N, 0\rangle_{3,4} + |0, N\rangle_{3,4} \right). \] (1.64)

This state is commonly referred to as the “N00N” state.\(^3\)

As the N00N state evolves in the interferometer it picks up a phase shift of \(e^{iN\phi}\).
\[ |N :: 0\rangle = \frac{1}{\sqrt{2}} \left( |N, 0\rangle_{3,4} + e^{iN\phi} |0, N\rangle_{3,4} \right). \] (1.65)

To estimate the phase shift, an operator is needed that is sensitive to the coherence between the two arms. Such an operator is
\[ \hat{M}_N = |N, 0\rangle \langle 0, N|_{3,4} + |0, N\rangle \langle N, 0|_{3,4}, \] (1.66)
which is sensitive to \(N\)th order correlations in the state. The expectation value of this operator is
\[ \langle N :: 0| \hat{M}_N |N :: 0\rangle = \cos(N\phi), \] (1.67)
and \(\langle \hat{M}_N^2 \rangle = \hat{I}\) for a N00N state. In an interferometer, \(\langle \hat{M}_N \rangle\) for a N00N state oscillates \(N\) times as fast as a classical state. The N00N state behaves like a classical state with a wavelength of \(\lambda/N\). This property is known as phase super resolution. It should be noted that phase super resolution is not a purely quantum effect; it has been demonstrated using classical light sources [Resch 07]. Classical super resolution can be achieved by multiplying \(N\) copies of a sinusoidal signal together, with each copy phase shifted from the previous one by \(2\pi/N\). The resulting multiplied signal will oscillate \(N\) times faster than the original sinusoid. The trade-off with such a scheme is that it is unable to beat the shot-noise limit in phase sensitivity [Resch 07, Plick 09, Nagata 07].

The variance in \(\hat{M}_N\) for a N00N state is
\[ \Delta \hat{M}_N^2 = 1 - \cos^2(N\phi) = \sin^2(N\phi). \] (1.68)

\(^3\)Removing the bra-ket notation from the N00N equation gives \(N0 + 0N\) which is then shortened to “N00N”. This has led to an entire series of Gary Cooper themed conference jokes involving posters for the movie “High Noon”
From equation 1.46, we find the phase uncertainty associated with this measurement to be

\[ \Delta \phi = \frac{\Delta \hat{M}_N}{\left| \frac{\partial \langle \hat{M}_N \rangle}{\partial \phi} \right|} = \frac{|\sin(N\phi)|}{|N\sin(N\phi)|} = \frac{1}{N}, \]  

which is precisely the Heisenberg limit in phase uncertainty. This represents a \( \sqrt{N} \) improvement in the phase sensitivity over the standard quantum limit, and is a property referred to as **phase super sensitivity**.

### 1.3.7 Measuring Nth Order Correlations

To implement the measurement operator \( \hat{M}_N \) described in equation 1.66 in the lab, a second “non-magical” beamsplitter is used to interfere paths 3 and 4. At the output ports the number of photons are counted (instead of a simple intensity measurement). The operator for counting \( N \) photons is \( \hat{N} = \hat{a}^\dagger N \hat{a}^N \) [Loudon 73]. Using the beamsplitter relations and some algebra, it is possible to show that

\[ \langle \hat{N}_5 \rangle \propto \cos^2(\frac{N\phi}{2}), \quad \langle \hat{N}_6 \rangle \propto \sin^2(\frac{N\phi}{2}). \] 

For the N00N state, the difference between the photon number counting at the two output ports recovers the desired measurement operator \( \hat{M}_N \):

\[ \hat{M}_N = \hat{a}_5^\dagger N \hat{a}_5^N - \hat{a}_6^\dagger N \hat{a}_6^N \]

\[ \propto |N, 0\rangle \langle 0, N|_{3,4} + |0, N\rangle \langle N, 0|_{3,4}. \] 

By counting the number of photons using number-resolving detectors, it is possible to measure the \( N \)th order correlations that exist. This detection scheme is inherently quantum mechanical as true number-resolving detectors are quantum in nature [Lundeen 08].
1.4 Schwinger Representation

There is an interesting connection between the algebra of two uncoupled harmonic oscillators and the algebra of angular momentum. Schwinger pointed out that a system of two uncoupled harmonic oscillators, sharing a total of $N$ excitations, is isomorphic to a spin $j = N/2$ particle [Sakurai 94]. Schwinger’s representation can be applied to the MZ interferometer, bringing to bear the full power of the angular momentum formalism to the problem.

In the MZ the two input modes are transformed sequentially by a beamsplitter, a phase shifter, and a final beamsplitter before a measurement is made. $N$ particles distributed between the two input modes, now labelled $a$ and $b$, can be described as a collection of $N$ spin-$1/2$ particles. Here the number of particles $n_a$ in mode $a$ corresponds to the number of spins pointing up while the number of particle $n_b$ in mode $b$ corresponds to the number of spins pointing down. We define the following angular momentum operators in terms of the optical modes (setting $\hbar = 1$)

\[
\hat{J}_x = \frac{1}{2} \left( \hat{a}^\dagger \hat{b}^\dagger \hat{a} - \hat{b}^\dagger \hat{a}^\dagger \hat{b} \right),
\]

\[
\hat{J}_y = \frac{-i}{2} \left( \hat{a}^\dagger \hat{b}^\dagger - \hat{b}^\dagger \hat{a}^\dagger \hat{a} \right),
\]

\[
\hat{J}_z = \frac{1}{2} \left( \hat{a}^\dagger \hat{a} - \hat{b}^\dagger \hat{b} \right) = \frac{1}{2} (\hat{n}_a - \hat{n}_b),
\]

\[
\hat{J}^2 = \frac{1}{2} \hat{n} \left( \frac{\hat{n}}{2} + 1 \right),
\]

where $\hat{n} = \hat{n}_a + \hat{n}_b = \hat{a}^\dagger \hat{a} + \hat{b}^\dagger \hat{b}$ is the total number of photons. The operators obey the commutation relations $[\hat{J}_\alpha, \hat{J}_\beta] = i \in_{\alpha,\beta,\gamma} \hat{J}_\gamma$. Both $\hat{J}^2$ and $\hat{J}_z$ share a common eigenstate $|j, m\rangle_z$

\[
\hat{J}^2 |j, m\rangle_z = j(j + 1) |j, m\rangle_z,
\]

\[
\hat{J}_z |j, m\rangle_z = m |j, m\rangle_z,
\]

with eigenvalues $j = (n_a + n_b)/2$ and $m = (n_a - n_b)/2$. Since $\hat{J}^2$ also commutes with $\hat{J}_x$ and $\hat{J}_y$, both $\hat{J}_x$ and $\hat{J}_y$ each share an eigenstate with $\hat{J}^2$ as well. Here we choose to work
with $\hat{J}^2$ and $\hat{J}_z$ to follow convention and for convenience. From now on we will simply write $|j, m\rangle_z$ as $|j, m\rangle$.

In terms of the creation operators for modes $a$ and $b$, the eigenstate $|j, m\rangle$ can be written as

$$|j, m\rangle = \frac{(a^\dagger)^{j+m} (b^\dagger)^{j-m}}{\sqrt{(j+m)!(j-m)!}} |0\rangle,$$

with $n_a = j + m$ and $n_b = j - b$. For a Fock state containing $N$ photons in mode $a$ the spin representation is

$$|N, 0\rangle_{a,b} = \frac{(a^\dagger)^{2j}}{\sqrt{(2j)!}} |0\rangle = |j, j\rangle,$$

which corresponds to having a system where all $N$ particles are spin up. Similarly a Fock state with all $N$ particles in mode $b$ can be written as $|j, -j\rangle$, corresponding to a system with all $N$ particles aligned spin down. In this language a $N00N$ state is a superposition of having all spins pointing up with all spins pointing down

$$|N :: 0\rangle_{a,b} = \frac{1}{\sqrt{2}} (|j, j\rangle + |j, -j\rangle).$$

### 1.4.1 The Spin Coherent State

For a single harmonic oscillator, the closest quantum state to the classical solution is the coherent state. For the two-mode spin system, the analogue to the coherent state is the spin coherent state [Arecchi 72]. A spin coherent state is one where all of the spins are aligned in the same direction. The total state of the system can be written as a product state of the individual spins: there is no entanglement between each of the spins. In this sense the spin coherent state is the closest approximation to a classical state. A spin-coherent state can be described by a vector that starts at the origin and terminates at a point on the surface of a sphere of radius $\sqrt{j(j+1)}$. This sphere can be thought of as a generalized Bloch sphere; when $j = 1/2$, the sphere is equivalent to the ordinary Bloch sphere used extensively in quantum information. The direction the vector points,
corresponding to the direction in which all of the spins are aligned, is given by a polar angle $\theta$ and azimuthal angle $\gamma$. This allows the spin-coherent state to be written as $|\theta, \gamma\rangle$.

For a state with all of the spins pointing up, we have $|j, j\rangle = |\theta = 0, \gamma = 0\rangle$. Likewise for the spin-coherent state with all spins aligned down $|j, -j\rangle = |\theta = -\pi, \gamma = 0\rangle$. In general [Agarwal 81],

$$|\theta, \gamma\rangle = \sum_{m=-j}^{j} \left( \frac{2j}{j+m} \right)^{\frac{1}{4}} \left( \cos \frac{\theta}{2} \right)^{j-m} \left( \sin \frac{\theta}{2} \right)^{j+m} e^{i(j+m)|j, m\rangle}. \quad (1.82)$$

For a spin coherent state $|\theta = \pi/2, \gamma = 0\rangle$ pointing along the $x$ axis, the expansion in terms of $|j, m\rangle$ is

$$|\theta = \pi/2, \gamma = 0\rangle = \left( \frac{1}{\sqrt{2}} \right)^{j} \sum_{m=-j}^{j} \left( \frac{2j}{j+m} \right) |j, m\rangle, \quad (1.83)$$

which is a coherent superposition of the $N$ photons binomially distributed between the two modes.

The spin coherent state is also a minimum uncertainty state in the sense that the angular momentum Heisenberg uncertainty relationship,

$$\Delta \hat{J}_x' \Delta \hat{J}_y' \leq \frac{1}{2} \left| \langle \hat{J}_z' \rangle \right|, \quad (1.84)$$

is satisfied with equality. Here $\hat{J}_x'$, $\hat{J}_y'$, and $\hat{J}_z'$ are the angular momentum operators in a rotated basis such that $\hat{J}_z'$ lies along the axis $(\theta, \gamma)$ passing through the centre of the coherent state. The uncertainty between the two non commuting angular momentum operators is equally distributed $\Delta \hat{J}_x' = \Delta \hat{J}_y' = \frac{1}{\sqrt{2}} \left| \langle \hat{J}_z' \rangle \right|^{1/2} = \frac{\sqrt{N}}{2}$. This leads to a circularly symmetric uncertainty distribution on the surface of the sphere centred about $\hat{J}_z'$ as shown in figure 1.2.

### 1.4.2 Spin Squeezing

A spin-squeezed state is one where the uncertainty relations in the angular momentum operators are manipulated to decrease the variance in one operator at the expense of
Chapter 1. Background

Figure 1.2: A spin-coherent state and a spin-squeezed state on a generalized Bloch sphere.

a) A spin-coherent state $|\theta, \gamma\rangle$ can be described by a vector that terminates on the surface of a sphere of radius $\sqrt{j(j + 1)}$. The expectation value of angular momentum operators orthogonal to the state vector will have an uncertainty of $\sqrt{N/2}$. The uncertainty in the state can be represented as the intersection of a cone with of radius $\sqrt{N/2}$ and the surface of the sphere. b) Applying the squeezing operator $U_{sq} = e^{\chi J_z^2}$ to $|\theta, \gamma\rangle$ stretches the uncertainty towards the poles of the Bloch sphere, leading to a decrease in the uncertainty in one of the angular momentum operators orthogonal to state vector. Strictly speaking, for a squeezed state a simple vector is no longer sufficient to completely describe the state on the Bloch sphere. The squeezing breaks the circular symmetry in the uncertainty distribution. A full description of the state must include the shape of the uncertainty distribution as well.
an increase in variance in the conjugate operator. A standard way to accomplish this is to take a spin-coherent state and “squeeze” it, reducing the variance in one of the orthogonal angular momentum operators below the shot-noise limit of $\sqrt{N}$ as shown in figure 1.2. The squeezing operator is not unique; here we introduce a squeezing operator commonly discussed in the literature [Agarwal 81]:

$$U_{sq} = e^{\chi \hat{J}_z^2},$$

with squeezing parameter $\chi$, that takes a spin-coherent state and moves it towards the poles of $\hat{J}_z$. This form of squeezing operator can be constructed for $\hat{J}_x$ and $\hat{J}_y$ as well. For a spin-coherent state already aligned along $\hat{J}_z$ the squeezing operator has no effect. For spin-coherent states aligned in other directions, this squeezing operator leads to a “stretching” of the uncertainty along $\hat{J}_z$ as the state moves towards the poles, leading to a decrease in uncertainty in an orthogonal angular momentum operator. For a spin-coherent state initially aligned along either the $x$ or $y$ axis, the maximum amount of squeezing leads to the formation of a N00N state along the $z$ axis. In this case, half the population along the equator coherently “moves” to the north pole while the other half of the population “moves” to the south pole.

### 1.4.3 The Angular Momentum Picture of a MZ Interferometer

A MZ interferometer takes two input modes and then unitarily transforms them into two output modes. We have seen that the Schwinger representation allows the two-mode input state (and output state) to be expressed in terms of a spin-$j$ particle. In [Yurke 86] Yurke points out that the action of the MZ interferometer is to unitarily rotate the input state to the output state. If we consider each of the components of the MZ, we can determine what this rotation is. The action of the 50:50 beamsplitters is equivalent to $\pm \frac{\pi}{2}$ rotation about the $x$ axis, i.e. $exp(\pm i \frac{\pi}{2} \hat{J}_x)$. The relative phase shift $\phi$ applied in one of the arms is equivalent to a rotation of $\phi$ about the $z$ axis, i.e $exp(-i\phi \hat{J}_z)$. The total
action of the MZ interferometer in the angular momentum picture is then

\[
|\psi_{out}\rangle = e^{i\frac{\pi}{2}\hat{J}_x} e^{-i\phi \hat{J}_z} e^{i\frac{\pi}{2}\hat{J}_x} |j, m\rangle = e^{i\phi \hat{J}_y} |j, m\rangle.
\] (1.86)

(1.87)

The entire action of the interferometer is to perform a rotation by \(\phi\) about the \(y\) axis. This can be seen pictorially in figure 1.3. To estimate the phase shift, the difference in intensity at the two output ports is measured. This corresponds to measuring \(\langle \hat{J}_z \rangle = \frac{1}{2} (\langle \hat{n}_a \rangle - \langle \hat{n}_b \rangle)\), the difference in the number of spin up and spin down particles.

Using this angular momentum picture, the connection between the MZ interferometer and atomic interferometers, like the Ramsey interferometer, becomes clear. The first beamsplitter is equivalent to applying a \(\pi/2\) pulse. The relative phase shift is equivalent to the atomic state rotating about the \(z\) axis (perhaps due to a magnetic field) after the \(\pi/2\) pulse. The second beamsplitter is equivalent to then applying a second \(\pi/2\) pulse to the system after some specified time. Detection is performed by measuring \(\langle \hat{J}_z \rangle\). Despite radically different physical implementations, atomic interferometers used in atomic clocks and the optical interferometers used by LIGO share the same basic description. The power of the angular momentum formalism is the consistent framework it provides for studying interferometers independent of their physical implementation. This enables us to examine the properties of quantum states using one physical implementation and draw general conclusions about their behaviour in other systems.

### 1.5 The Poincaré Sphere

To study the properties of classical and quantum states used in interferometry, the experiments in this thesis are implemented using the polarization of light. In 1852 Stokes developed a method for characterizing the polarization of classical light using a vector [Stokes 52]. The Stokes parameters \(S_1, S_2\) and \(S_3\) constitute a Cartesian coordinate system for the polarization state, which is represented by a Stokes vector that terminates
Figure 1.3: *Schwinger Representation of a Mach-Zehnder interferometer.* a) A beamsplitter acting on the state $|j, m\rangle$ rotates it by $\pm \frac{\pi}{2}$ about $x$. In a MZ interferometer, the first beamsplitter takes an input state (1) and rotates it by $\frac{\pi}{2}$ (2). The relative phase shift acquired between the two paths rotates the state about $z$ by $-\phi$ (3). The second beamsplitter applies a final $-\frac{\pi}{2}$ rotation about $x$ to the state (4). Measuring the difference in intensities between at the two outputs to the interferometer, $\langle n_a \rangle - \langle n_b \rangle$, is equivalent to measuring $2 \langle \hat{J}_z \rangle$. b) The overall action of the Mach-Zehnder interferometer is to apply the rotations $e^{-i\frac{\pi}{2} \hat{J}_x} e^{i\phi \hat{J}_z} e^{i\frac{\pi}{2} \hat{J}_x}$ to an input state. This is equivalent to applying a single rotation about $y$ by $\phi$, i.e. $e^{i\phi \hat{J}_y}$. c) The action of the MZ-interferometer on an initial state $|j, j\rangle$ in (1), corresponding to $2j$ photons entering the first beamsplitter via the first input port, is shown as a sequence of rotations about the generalized Bloch sphere. The three rotations the interferometer performs in (2)-(4) are equivalent to a single rotation about $y$ by $\phi$. 
on the surface of a sphere. This sphere is known as the Poincaré sphere. The Stokes parameters $S_1$, $S_2$, and $S_3$ describe the degree of linear, diagonal, and circular polarization respectively, and the intensity of the beam is characterized by the Stokes parameter $S_0 = S_1^2 + S_2^2 + S_3^2$. A quantum description of polarization can be obtained by replacing the Stokes parameters with the corresponding Stokes operators:

\[
\begin{align*}
\hat{S}_0 &= 2\hat{J}^2 = a_H^\dagger a_H + a_V^\dagger a_V = \hat{n}_H + \hat{n}_V = N, \\
\hat{S}_1 &= 2\hat{J}_z = a_H^\dagger a_H - a_V^\dagger a_V = \hat{n}_H - \hat{n}_V, \\
\hat{S}_2 &= 2\hat{J}_x = a_H^\dagger a_V + a_V^\dagger a_H = \hat{n}_D - \hat{n}_A, \\
\hat{S}_3 &= 2\hat{J}_y = i(a_V^\dagger a_H - a_H^\dagger a_V) = \hat{n}_R - \hat{n}_L
\end{align*}
\]

where $N$ is the total photon number and $a_i^\dagger, a_i$, and $\hat{n}_i$ are the creation, annihilation and photon number operators for polarization mode $i$, and where $i$ takes the values H,V,D,A,R and L corresponding to horizontal, vertical, diagonal, anti-diagonal, right-hand circular and left-hand circular polarizations respectively [Korolkova 01]. The Stokes operators do not commute with one another, leading to the uncertainty relations $V_i V_m \geq \left| \left\langle \hat{S}_n \right\rangle \right|^2 \in_{l,m,n}$ where $V_i$ represents the variance in the Stokes operator $\hat{S}_i$. Now the quantum analogue to the Poincaré sphere is simply a scaled version (by a factor of two) of the spin-$N/2$ generalized Bloch sphere discussed in section 1.4.1. In the next section we will explore how quantum states can be represented on this Poincaré sphere.

### 1.5.1 Quasi-Probability Distributions on the Poincaré Sphere

If a classical polarization state is represented by a Stokes vector, one might think that the most “classical” quantum polarization state should also be determined by a vector. Indeed this is the case. A spin-coherent state, composed of a collection of photons with the same polarization, can be specified by a vector terminating on the surface of the quantum Poincaré sphere. As seen in section 1.4.2, the spin-coherent state has an uncertainty in the orthogonal Stokes parameters that scales as $\sqrt{N}$. For squeezed states the uncertainty
between the non-commuting Stokes operators implies that rather than assuming definite values, the Stokes operators must be described by a joint quasi-probability distribution.

**Density Matrix Formalism**

Until this point, we have only considered pure quantum states described as a vector $|\psi\rangle$. A more general approach for treating quantum states is to use the density matrix formalism. A quantum state can be described by a density matrix which is a linear, Hermitian operator. The density matrix can be written as a probability distribution over projectors onto different basis states spanning the Hilbert space, i.e.

$$\rho = \sum_i p_i |\psi_i\rangle \langle \psi_i|,$$

where $p_i$ are probabilities and $\sum p_i = 1$ [Nielsen 00]. The diagonal elements of the density matrix are referred to as *populations* while the off-diagonal elements are called *coherences*. The expectation value of a Hermitian operator $\hat{O}$ acting on a quantum state is given as

$$\langle \hat{O} \rangle = Tr[\rho \hat{O}].$$

A pure state can be written as $\rho = |\psi\rangle \langle \psi|$. States that cannot be written in this form are said to be mixed. The maximally mixed state has a density operator that is an unbiased statistical *mixture* over all of the basis states $|\psi_i\rangle$. In terms of polarization, the purity of a state is related to the degree of polarization. An unpolarized (maximally mixed) state has a degree of polarization of 0 while a pure polarization state has a degree of polarization of 1. Using this density matrix formalism we can tackle the problem of defining quasi-probability distributions on the surface of the Poincaré sphere.

**Wigner Quasi-Probability Distributions**

The state of a classical system can be represented by a probability distribution in phase space. For quantum systems it is impossible to represent it using a phase-space prob-
ability distribution: doing so would require simultaneously knowing the value of two non-commuting observables. Instead a quasi-probability distribution can be formulated that retains many of the useful features of a classical probability distribution. The penalty for doing this is that the quasi-probability distribution can contain negative values. For the states of angular momentum under consideration, the phase space covers the surface of the Poincaré sphere. The choice of quasi-probability distributions is not unique: there exists a continuum of possibilities. Here we introduce Wigner quasi-probability distributions following the approach of Agarwal [Agarwal 81] to provide an informationally complete description of the polarization state space and to calculate any of the desired properties of the state [Dowling 94, Stratonovich 56, Luis 05, Luis 06].

The Wigner function that describes a spin-$j$ system relies on a multipole expansion of the density matrix $\rho$ using the multipole operators $\hat{T}_{kq}$ [Agarwal 81]:

$$W(\theta, \gamma) = \sum_{k=0}^{2j} \sum_{q=-k}^{k} Y_{kq}(\theta, \gamma) \text{Tr}[\rho \hat{T}_{kq}],$$  

(1.91)

where $Y_{kq}(\theta, \gamma)$ are the spherical harmonic functions. The multipole operators allow the density matrix to be expanded in terms of the spherical harmonics, and contain all of the necessary angular momentum coupling terms

$$\hat{T}_{kq} = \sum_{m=-j}^{j} \sum_{m'=-j}^{j} (-1)^{j+m} \sqrt{2k+1} \begin{bmatrix} j & k & j \\ -m & q & m' \end{bmatrix} |j, m\rangle \langle j, m'|,$$  

(1.92)

where $\begin{bmatrix} j & k & j \\ -m & q & m' \end{bmatrix}$ is the Wigner 3$j$ symbol. The resulting quasi-probability distribution is spread out over the surface of a Poincaré sphere.

**Spin-Coherent States on the Poincaré Sphere**

The Wigner distribution for a spin-coherent state is a circularly symmetric distribution of width $\sqrt{N}$ centred about the point where the Stokes vector terminates on the surface of the Poincaré sphere. As the number of photons in the spin-coherent state is increased,
the relative uncertainty area on the surface of the sphere decreases as shown in Figure 1.4. In the limit of large $N$ the distribution shrinks to a point, recovering the classical Stokes description.

**N00N States on the Poincaré Sphere**

The N00N state is a superposition of all the photons being spin up and spin down. On the Poincaré sphere this corresponds to a superposition of all the photons being either at the “North” or “South” poles. The resulting probability distribution is more complex than that of the spin-coherent state, containing “fringes” that cycle around the equator. The Wigner distributions for several different N00N states are plotted in figure 1.4. As $N$ grows larger, the Wigner functions move further and further away from the classical result. It is interesting to note that the N00N states contain an $N$ fold symmetry around the equator that corresponds to their phase super resolution properties.

**Spin-Squeezed States on the Poincaré Sphere**

Starting with a spin-coherent state composed of horizontally polarized photons, the action of the operator $U_{sq} = e^{\chi \hat{S}_3}$ squeezes the distribution towards the poles away from the equator. This increases the uncertainty in $\hat{S}_3$ while decreasing the uncertainty in $\hat{S}_2$. The maximally squeezed state is the N00N state, corresponding to a superposition of all of the photons being right or left handed circularly polarized. The Wigner distributions for a variety of squeezed states are shown in figure 1.5.

**Mixed States on the Poincaré Sphere**

Classically a partially-polarized beam of light is described by a Stokes vector that lies inside the surface of the Poincaré sphere. In contrast, the Wigner quasi-probability distribution for a partially mixed state is a “broad” distribution across the surface of the sphere. For the maximally-mixed state the Wigner distribution is constant across the
Figure 1.4: *Wigner distributions on the Poincaré sphere for spin-coherent and N00N states of 3, 10, and 20 photons.* The radius of the spheres are normalized for ease of comparison. The spin-coherent states are made up of horizontally polarized photons, hence the distribution is centred along $\hat{S}_1$ with a width of $\sqrt{N}$. As $N$ grows larger, the uncertainty distribution shrinks relative to the radius of the sphere. In the limit of large $N$ the uncertainty distribution collapses to a single point, mimicking the classical Poincaré sphere. The N00N states pictured are in a superposition of all the photons being right-hand or left-hand circularly polarized. At either pole along $\hat{S}_3$ sits a circular “blob” of probability corresponding to this superposition. Around the equator there are $N$ fringes corresponding to the coherence that exists in the N00N state. These fringes are related to the *phase super resolution* and *phase super sensitivity* of the N00N state.
Figure 1.5: Wigner distributions on the Poincaré sphere for different 10-photon spin squeezed states. A horizontally polarized spin-coherent state is squeezed by the operator $U_{sq} = e^{\chi \hat{S}_3^2}$. The squeezing parameter $\chi$ determines the amount of squeezing with $\chi = 0$ corresponding to a spin-coherent state and $\chi = 1$ to the maximally squeezed N00N state. As squeezing parameter is increased, the distribution “stretches” towards the two poles of $\hat{S}_3$, leading to a reduced uncertainty in $\hat{S}_2$. 
surface of the sphere: a projection along any direction is equally likely.

1.5.2 Marginals

An important property of traditional planar Wigner functions is that the marginal probability distribution obtained by integrating the quasi-probability distribution over one conjugate variable yields the correct probability distributions of the other variable. For spin systems where the Wigner functions are based on the angular momentum variables, the marginals do not correspond to physically measurable observables, i.e. a phase operator. However, the marginals can still be used to predict the performance of a quantum state in something like an interferometer. A phase shift in a MZ interferometer corresponds to a rotation about the z axis. By integrating over the polar angle $\theta$ of the Wigner quasi-probability distribution $W(\theta, \gamma)$, we obtain the marginal distribution $M(\gamma)$ for the azimuthal angle $\gamma$,

$$M_W(\gamma) = \int W(\theta, \gamma) \sin(\theta) d\theta,$$

which qualitatively predicts the fringe pattern that would be obtained in a MZ interferometer. The reason the marginals do not correspond to probability distributions as in the planar case is related to the fact that the SU(2) Wigner quasi-probability distribution is not designed with this marginal property in mind: rather they are based on the more abstract properties of angular momentum operators [Luis 02, Luis 06, Luis 05]. For example, the Wigner distribution contains negative values for every state. This can be seen by the fact that the spin-coherent state is always orthogonal to an antipodal spin-coherent state. To satisfy this orthogonality condition and remain normalized, the Wigner quasi-probability distribution for each state must take on negative values at some point. This presence of negative values can cause sharper oscillations that lead to deviations from the expected marginal probability distribution.

An alternative approach is to use the Q quasi-probability distribution (also known as
the Husimi distribution) [Luis 05] to study properties of spin states. For spin systems, the Q distribution is positive everywhere and is given in terms of the overlap of the the density matrix with the spin-coherent states [Agarwal 98]:

\[
Q(\theta, \gamma) = \frac{2j + 1}{4\pi} \langle \theta, \gamma | \rho | \theta, \gamma \rangle,
\]

with the normalization requirement

\[
\int Q(\theta, \gamma) \sin(\theta) d\theta d\gamma = 1.
\]

Using the Q distribution, the expected fringe pattern in an interferometer is exactly equal to a normalized slice of the distribution around the equator \(Q(0, \gamma)\). Thus, slices of the Q distribution are useful for visualizing and studying the properties of spin states.

### 1.6 Prospects for Quantum Interferometry

The \(\sqrt{N}\) improvement in measurement sensitivity that N00N states promise is enormous. Consider LIGO which has on average \(10^{24}\) photons circulating in the interferometer. With a N00N state a factor of \(10^{12}\) improvement in the phase sensitivity is possible! This would enable displacements on the order of \(10^{-30}\) m to be detected, putting LIGO within reach of the Planck length. Despite this tremendous promise, we are still a long ways away from implementing Heisenberg-limited interferometry in a practical manner. The large nonlinearities required to create and detect the states used in quantum metrology are not yet available. What is needed to make this technology feasible is a large-scale material science revolution. Of course this revolution may never fully happen, but it is still instructive to study what fundamental limits quantum mechanics places on our ability to interact, manipulate, and transform the world around us. At the most basic level, the new way of thinking that quantum information has brought about has deepened our understanding of quantum mechanics and quantum phenomena. The road to reaching Heisenberg limited interferometry may be long and difficult, but the payoff in our ability
to measure the world around us is huge. Right now we are at the start of an exciting journey of discovery.
Chapter 2

Spontaneous Parametric
Downconversion

2.1 Motivation

Producing the nonclassical states needed in quantum metrology requires novel photon sources. In addition, these sources are useful for other applications in quantum information that require multiple entangled qubits. A qubit can be thought of as a spin-1/2 particle that encodes information in the direction of the spin. Unlike a regular bit of information that can take the values 1 or 0, a qubit is capable of storing superpositions of 1 and 0. Qubits can be entangled with one another and then processed in a variety of ways. A quantum computer capable of processing entangled qubits using quantum gates can implement quantum algorithms that outperform their classical counterparts. A practical quantum computer requires hundreds (and possibly thousands) of entangled qubits to function. Unfortunately, the entanglement between qubits is fragile. Theoretical work-arounds have been developed to deal with the inevitable errors and decoherence that will occur in quantum logic gates [Nielsen 00, Shor 95], and clever proposals like cluster state computing simplify some of the complexity of quantum gates.
[Briegel 01, Walther 05a, Walther 05b, Kai 07]. Still, generating the robust entangled states needed for quantum computing remains a daunting task.

It is known that linear optics alone is not sufficient for implementing scalable quantum computation [Knill 01, Lee 02a]. The number of optical elements like beamsplitters, wave plates, crystals, mirrors, and lasers scales exponentially with the number of qubits. The reason for this terrible scaling is that photons weakly interact with each other; quantum gates require strong interactions between the photons. While photons may not be the final architecture used to implement a quantum processor, they are useful for testing many of the building blocks that will be used in the final architecture.

A qubit can be encoded within the many degrees of freedom of a photon. Entangled states using polarization [Ou 88, Shih 88], [Rubin 96, Grice 97, Keller 97], momentum [Rarity 90], and energy-time [Franson 89] degrees of freedom have all been implemented. A photon can also be made to multitask: multiple qubits can be simultaneously encoded within different degrees of freedom of the same photon [Barreiro 05, Wei 07, Barreiro 08]. A disadvantage is that it can be difficult to entangle, manipulate, and measure the different qubits entangled on the same photon. The most “straight-forward” approach is to entangle multiple photons.

For the past twenty-five years, the most reliable and robust manner of generating single photons and entangled photons has been through the use of spontaneous parametric down conversion (SPDC). In SPDC, a single photon from a pump beam is converted to two less energetic photons through the mediation of a nonlinear medium. SPDC has been used to study everything from fundamental tests of quantum mechanics to implementations of various quantum technologies.

In the rest of this chapter I will outline the work I carried out designing and building different SPDC multi-photon sources. The experiment described in chapter 4 relies on SPDC. Much of my initial work was spent on optimizing this source. These lessons were then applied to build a next generation source that is one of the brightest sources of four
photons in the world.

2.2 Introduction to Spontaneous Parametric Down Conversion

In a typical nonlinear medium an electric field drives a dipole moment nonlinearly [Boyd 03]. The polarizability of the material, the dipole moment per unit volume, can be expressed in terms of the following power series

\[
P(t) \equiv \chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + \ldots ,
\]

where \( \chi \) is the nonlinear susceptibility tensor that determines the strength of the response of the medium to a driving field \( E(t) \). SPDC is a second order nonlinear effect (\( \chi^{(2)} \)), and can be thought of as the time-revered process of sum-frequency generation. In sum-frequency generation two input driving fields \( E_1(t) \) and \( E_2(t) \), at frequencies \( w_1 \) and \( w_2 \) respectively, produce a third field \( E_3(t) \) with a frequency \( w_3 = w_1 + w_2 \) as shown in figure 2.1 a). The effect of the two driving fields in this nonlinear regime is multiplicative instead of additive \(^1\). Second-harmonic generation (SHG) is a special case of sum-frequency generation where the two input frequencies oscillate at the same frequency \( w \) producing an output field at \( 2w \). This is analogous to a musician plucking a guitar string; not only will a note at the fundamental frequency be created, but also an overtone at twice the frequency.

Quantum mechanics is symmetric with respect to time; the equations work equally well for time moving forward as well as backwards. Why we perceive there to be a definite “arrow of time” is still an open question [Leggett 08, Maccone 09]. If we take sum-frequency generation and run it in reverse we should return back to the initial state as shown in figure 2.1 b). Now the inputs to the crystal are the fields \( E_1(t) \), \( E_2(t) \)

\(^1\)This can be seen in the trigonometric identity \( \sin w_1 \cos w_2 = 2 \sin(w_1 + w_2) \).
Figure 2.1: a) *Sum-Frequency Generation* Two beams with frequencies $w_1$ and $w_2$ interact in a nonlinear $\chi^{(2)}$ crystal to generate a third beam with a frequency $w_3 = w_1 + w_2$. b) *Time reverse of sum-frequency generation.* Two input seed beams at $w_1$ and $w_2$ are stimulated by a pump at $w_3$. c) *Spontaneous Parametric Downconversion.* A pump beam, seeded by vacuum fluctuations, downconverts into a pair of daughter photons.

The output is $E_1(t)$ and $E_2(t)$; all the photons in $E_3(t)$ are *downconverted* into the lower frequency outputs. The presence of $E_1(t)$ and $E_2(t)$ at the input of this downconversion process is necessary; they act as seed beams that stimulate the emission. Blocking either $E_1(t)$ or $E_2(t)$ stops the stimulated emission from taking place. This effect can be used to build a switch that operates at single-photon intensities [Resch 01]. All that is required is that the seed beams share a definite phase relationship with $E_3(t)$. This switch was successfully used by Jeffrey Lundeen to implement an experimental test of Hardy’s paradox [Lundeen 09].

Blocking both seed fields at the input should prevent any stimulated emission from occurring. But this is not the case! Vacuum fluctuations generate virtual photons that can act as the seed fields. The result is that a photon from the pump $E_3(t)$ is spontaneously converted into two daughter photons as illustrated in figure 2.1 c). The only constraint on the frequency of the daughter photons is that they must sum to the frequency of the pump. Just as photons from spontaneous emission in an atom can be emitted in any
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direction, the daughter photons from SPDC are not constrained to a single plane; their emission pattern forms a cone. Historically, one daughter photon is referred to as the signal while the other is referred to as the idler.

In the limit of a strong pump field (treated classically), the Hamiltonian describing SPDC is

\[ \hat{H} = i\kappa (\hat{a}_s^\dagger \hat{a}_s^\dagger - \hat{a}_s \hat{a}_i), \]  

(2.2)

where \( \kappa \) is related to the strength of the nonlinear interaction and is assumed to be weak. This Hamiltonian does not take into account the effects of dispersion, mode mismatch, pump bandwidth and a host of other effects; nevertheless, it serves as a useful approximation. The result of this nonlinear interaction is to produce the state

\[ |\epsilon\rangle = \frac{1}{\cosh \tau} \sum n |n, n\rangle_{s,i}, \]  

(2.3)

where \( |n, n\rangle_{s,i} \) represents \( n \) photons emitted into the signal and idler modes, \( \tau = \kappa t \) is the scaled time of the nonlinear interaction, and \( \epsilon = \tanh \tau \) is the amplitude for obtaining a SPDC pair per pulse [Kok 00]. As \( \kappa \ll 1 \) most of the time nothing happens, occasionally a pair of photons is produced, and very rarely two or more pairs of photons are created. Detecting the presence of a single photon in the idler arm indicates with a high probability that a single photon is in the signal arm. Such a single-photon source is said to be heralded.

2.3 Phase Matching

In order for SPDC to take place energy must be conserved. For photons with energy \( \hbar \omega \), this leads to the condition

\[ \hbar \omega_p = \hbar \omega_s + \hbar \omega_i, \]  

(2.4)

where \( p, s, \) and \( i \) refer to the pump, signal and idler respectively. The total momentum of the process must also be conserved. SPDC is a parametric process that returns the
nonlinear medium to its initial state. Consequently, the momentum of the daughter photons must match the momentum of the pump. For a nonlinear material of infinite dimension this leads to the condition

\[ \mathbf{k}_p = \mathbf{k}_s + \mathbf{k}_i, \]  

(2.5)

where \( \hbar \mathbf{k} \) is the momentum of a single photon. When this condition is met the process is said to be phase matched. Most nonlinear mediums are not infinitely long, tall, or wide\(^2\), but are finite. For a nonlinear crystal with a finite thickness the phase matching need not be perfect. In this case, it is convenient to write the momentum mismatch \( \Delta \mathbf{k} \) in terms of its vector components

\[ \Delta k^x = k^x_s + k^x_i - k^x_p, \]  
\[ \Delta k^y = k^y_s + k^y_i - k^y_p, \]  
\[ \Delta k^z = k^z_s + k^z_i - k^z_p, \]  

(2.6)  
(2.7)  
(2.8)

where \( \mathbf{x} \) and \( \mathbf{y} \) are transverse to and \( \mathbf{z} \) is along the direction of the pump’s propagation. Along the \( \mathbf{z} \) direction, the condition of infinite length is only satisfied when the crystal thickness is \( \gg \) than the Rayleigh range of the pump. In the lab nonlinear crystals used for SPDC have thickness that range from 0.1 mm up to 10 cm – shorter than than the Rayleigh length of the pump beams typically used. In this case it is possible for imperfect phase matching to occur, but the efficiency of the process rapidly falls off as the mismatch becomes large. It is therefore desirable to engineer a system for perfect phase matching. For a focused pump beam, imperfect phase matching is also possible along the transverse \( \mathbf{x} \) and \( \mathbf{y} \) directions.

\(^2\)Unless one considers space-time itself as a nonlinear medium! An intense enough field (\( \sim 10^{30} \text{W/cm}^2 \)) will undergo nonlinear effects mediated by the quantum vacuum [Gerstner 07].


2.3.1 Birefringence

For SPDC to occur, materials are needed that satisfy the phase matching conditions and have a large $\chi^{(2)}$. Most materials experience normal dispersion – the index of refraction increases with frequency. In such mediums it is impossible to phase match the pump with the daughter photons. One method to overcome this is through the use of birefringent materials where the index of refraction depends on the polarization of the light passing through it. A birefringent crystal is not isotropic; its response to an electric field depends on both the electric field’s direction and polarization. Within the crystal the axes along which two orthogonal polarization components move with the same phase velocity is called the optic axis. An uniaxial crystal contains one optic axis while a biaxial crystal has two optic axes. Beta-Barium Borate (BBO) is an uniaxial crystal commonly used to generate SPDC. It has a relatively large $\chi^{(2)}$, is resistant to moisture, and has a high damage threshold. Recently a biaxial crystal $\text{BiB}_3\text{O}_6$ (BIBO) with an even larger $\chi^{(2)}$ has been considered for both SHG and SPDC.

A uniaxial crystal contains two indices of refraction. The index along the optic axis is called the ordinary index $n_o$ while the index perpendicular to this is called the extraordinary index $n_e$. A crystal is positive when $n_o < n_e$ and negative when $n_e < n_o$. Due to dispersion $n_0$ and $n_e$ are wavelength dependent. For a specific wavelength $\lambda$ the Sellmeier coefficients can be used to calculate appropriate values of $n_0(\lambda)$ and $n_e(\lambda)$. For a beam of light that makes an angle $\theta$ with the optic axis, the component of polarization perpendicular (parallel) to the plane formed by the optic axis and direction of propagation is said to be ordinarily (extraordinarily) polarized. The ordinary component of polarization experiences an index of refraction $n_o$ while the index of refraction of the extraordinary polarization depends on the angle $\theta$ with the optic axis

$$n'_e(\lambda, \theta) = \frac{1}{\sqrt{\frac{\cos^2 \theta}{n_o^2(\lambda)} + \frac{\sin^2 \theta}{n_e^2(\lambda)}}} \quad (2.9)$$

An interesting effect of birefringence is that the direction of energy propagation of ex-
traordinarily polarized light is not necessarily perpendicular to the planes of constant phase; the Poynting vector and \( \mathbf{k} \) do not point in the same direction. As a result, the extraordinary polarization component walks off from the ordinary polarization. This effect is used in polarizers to separate out two polarization components. Calcite polarizers made in this fashion offer extinction ratios of 100,000:1.

### 2.3.2 Type-I and II Phase Matching

Phase matching can occur two different ways in a birefringent material like BBO. In type-I phase matching, the pump beam is extraordinarily polarized while the two daughter photons are ordinarily polarized \(^3 e \rightarrow o + o\). The two daughter photons are polarized the same way leading to a symmetric behaviour in the crystal. In degenerate SPDC where both daughter photons are the same wavelength, each photon appears on opposite sides of a cone centred about the pump. Tuning \( \theta \) changes the phase matching conditions causing the radius of the cone increase or decrease. The cone can be shrunk until both daughter photons travel collinear with the pump. Often, a small opening angle for the cone is preferable if individual access to either daughter photon is required.

The polarizations of the signal and idler photons from degenerate type-I SPDC are not entangled. In 1999 Paul Kwiat developed a technique to use type-I phase matching to generate entangled photons by sandwiching together two identical type-I crystals rotated by 90° with respect to one another [Kwiat 99, Dehlinger 02b, Dehlinger 02a]. The daughter photons generated in the two crystals will have orthogonal polarizations.

Consider a pair of crystals oriented so that the extraordinary polarization is horizontal \((H)\) in the first crystal and vertical \((V)\) in the second crystal. A \(H(V)\) polarized pump beam will generate a pair of \(V(H)\) photons in the first (second) crystal. If instead the pump is diagonally polarized \( \frac{1}{\sqrt{2}}(|H\rangle + |V\rangle)\), there is an equal probability of obtaining

---

\(^3\)This is true for BBO and other negative crystals. For crystals like BIBO that are positive, the phase matching conditions are met when \(o \rightarrow e + e\).
downconversion from either crystal. These two processes interfere and we must add their amplitudes. This leads to the entangled state

$$|\Phi^+\rangle = \frac{1}{\sqrt{2}} (|H, H\rangle + |V, V\rangle).$$  \hspace{1cm} (2.10)$$

The astute reader will notice that $|\Phi^+\rangle$ is one of the Bell states. Using a half wave-plate to set the polarization of the pump beam to $\frac{1}{\sqrt{2}} (|H\rangle - |V\rangle)$ creates the Bell state $|\Phi^+\rangle = \frac{1}{\sqrt{2}} (|H, H\rangle - |V, V\rangle)$. The remaining Bell states $|\psi^+\rangle = \frac{1}{\sqrt{2}} (|H, V\rangle + |V, H\rangle)$ and $|\psi^-\rangle = \frac{1}{\sqrt{2}} (|H, V\rangle - |V, H\rangle)$ can be generated using an additional half waveplate to flip the polarization of one of the daughter photons. The degree of entanglement can also be varied by changing the pump half waveplate angle while the purity of the state can be tuned by imposing a polarization dependent time delay on the pump.

In type-II phase matching the two daughter photons are orthogonally polarized, $e \to e + o$, and experience different indices of refraction traveling through the medium. This breaks the symmetry present in type-I SPDC. The signal and idler are emitted into cones than do not perfectly overlap; one cone will be centred above and the other below the pump beam. Changing $\theta$ changes the phase matching conditions and alters the emission angles and radiuses of the cones. The cones can be expanded so they intersect or can be collapsed down to a point. The collapsed cones case is also referred to as “beam-like” emission [Takeuchi 01] and is used in the experiment discussed in chapter 4.

It is possible to generate entanglement using a single type-II crystal by adjusting the phase matching so the signal and idler cones intersect. A photon at the point of intersection is equally likely to have come from either the signal or the idler. Again theses two processes interfere leading to the entangled state

$$|\Psi^+\rangle = \frac{1}{\sqrt{2}} \left(|o, e\rangle_{1,2} + |e, o\rangle_{1,2}\right),$$  \hspace{1cm} (2.11)$$

where 1 and 2 are the point of intersections. As in type-I it is possible to generate all of the Bell states using additional waveplates. Unlike type-I it is not easy to directly vary
the degree of entanglement or purity; to do so requires a realignment of the collection system so that distinguishing information between the two photons can be introduced.

2.4 Hong-Ou-Mandel Interferometer

To entangle photons from different pairs, two-photon interactions are required. The problem is that a photon-photon interaction is incredibly weak. Paul Dirac remarked that a photon interferes only with itself, not with other photons [Dirac 81]. Nonlinear mediums like SPDC can mediate these two photon interactions but are inefficient. In 1986 Chung Hong, Zhe-Yu Ou, and Leonard Mandel developed a two-photon interference effect that succeeds 100% of the time [Hong 87]. This Hong-Ou-Mandel (HOM) interferometer is now found at the heart of nearly every quantum computing experiment involving photons, and is one of the basic tools in a quantum optician’s toolbox.

Consider two distinguishable photons entering opposite ports of a 50:50 beamsplitter. By distinguishable we mean that the photons are different; they may be different colours, be in different spatial modes, have different transverse profiles, arrive at different times, and so on. To account for this distinguishing information each of the photons is described by a different creation operator. Using the beamsplitter relations from equation 1.35 the two-photon state at the output of the beamsplitter is

$$\hat{a}_1^\dagger \hat{b}_2^\dagger |0\rangle = \frac{1}{2} \left( \hat{a}_3^\dagger + i \hat{a}_4^\dagger \right) \left( i \hat{b}_3^\dagger + \hat{b}_4^\dagger \right) |0\rangle = \frac{1}{2} \left( i \hat{a}_3^\dagger \hat{b}_3^\dagger + \hat{a}_3^\dagger \hat{b}_4^\dagger - \hat{a}_4^\dagger \hat{b}_3^\dagger + i \hat{a}_4^\dagger \hat{b}_4^\dagger \right) |0\rangle.$$  

Counting the number of photons at each of the beamsplitter output ports we find that half the time the photons exit in different modes, and half the time they exit in the same mode.

If the distinguishing information is erased so that the photons now are the same colour, have the same polarization, and arrive at the same time, there is no longer any
way to tell the photons apart at the output of the beamsplitter. This leads to interference between the two photons

\[
\hat{a}_1^\dagger \hat{a}_2^\dagger |0\rangle = \frac{1}{2} \left( \hat{a}_3^\dagger + i\hat{a}_4^\dagger \right) \left( \hat{a}_a^\dagger + \hat{b}_1^\dagger \right) |0\rangle \\
= \frac{1}{2} \left( i\hat{a}_3^\dagger \hat{a}_3^\dagger + \hat{a}_4^\dagger \hat{a}_4^\dagger - \hat{a}_4^\dagger \hat{a}_3^\dagger + i\hat{a}_4^\dagger \hat{a}_4^\dagger \right) |0\rangle \\
= \frac{i}{2} \left( \hat{a}_3^\dagger \hat{a}_3^\dagger + \hat{a}_4^\dagger \hat{a}_4^\dagger \right) |0\rangle.
\]

Now the photons always exit through the same port of the beamsplitter; the output is a two-photon N00N state. For two photons the 50:50 beamsplitter is the magic beamsplitter (discussed in 1.3.6) needed to make a N00N state!

The HOM interferometer is an excellent tool for determining how indistinguishable two photons are. Placing a single-photon detector at each of the output ports we can measure when the detectors fire in coincidence. When the photons are completely distinguishable a steady stream of coincidence counts is registered. As the distinguishing information is erased, the number of coincidence counts decreases until it vanishes when the photons are indistinguishable. This provides a useful method for determining the arrival time of two photons. Fast single-photon detectors based on silicon have a jitter of tens of picoseconds; this is orders of magnitude slower than the coherence length of photons produced in SPDC. Using the HOM interferometer, the path length difference between two photons is varied and the coincidence counts monitored. When the dip in coincidence is observed the path length are balanced to within the coherence length of the photons. The minimum value of the dip is a measure of the distinguishing information that remains between the two photons.

The HOM interferometer measures second-order interference effects (fourth-order in amplitude). Zhe-Yu Ou showed that two classical fields can achieve a maximum visibility of 50% using linear optics and an appropriate detector scheme; any dip larger than this is quantum [Ou 91]. Recently Kevin Resch demonstrated a novel technique to generate a classical version of the HOM interferometer with 100% visibility [Kaltenbaek 08]. The
main difference in Resch’s version of the HOM interferometer is that a nonlinear crystal is used to create the necessary two-photon interactions. In the experiment described in chapter 4, the standard version of the HOM interferometer is used to set the arrival time of two different “classical” beams.

2.5 Fibre Coupling SPDC into Single-Mode Fibre

There are many situations where it is desirable to couple the emission from SPDC into single-mode fibre. A single-mode fibre acts as a spatial filter, erasing any distinguishing transverse information between the photons. Eliminating this mode mismatch can increase the quality of the output states in an experiment [Altepeter 05, Langford 05, Pan 03, Gasparoni 04]. Many experiments require the precise alignment of many optical paths; any instabilities in the pump laser or SPDC source can lead to significant misalignment. A fibre coupled SPDC source can be decoupled from the rest of an experiment. The frustration of spending weeks realigning an experiment due to a bumped mirror\(^4\) is a good reason to fibre couple. Once photon pairs are coupled into fibre it is “easy” to move the photons around, making it possible to share a single SPDC source between multiple different experiments.

There are also situations where it is not advisable to fibre couple. If an experiment depends critically on the efficiency with which photon pairs are detected, the use of fibre couplers should be minimized. Coupling in and out of fibre multiple times will lead to losses and decrease the overall collection efficiency. If the experiment is relatively simple and the count rates are low, then fibre coupling can be avoided. If long term polarization stability is required, then fibres may not be advisable. Fibre optic cables are slightly birefringent and cause random polarization rotations to occur. Loops in a cable will also alter the polarization of the photons inside. Temperature fluctuations, movement

\(^4\)Which has happened many times too myself and most other experimentalist I have talked to.
in the cable, or applied strain will also lead to variations in the polarization. Periodic corrections to the polarization of the photons in a fibre need to be made to fix the drift that occurs. By taping a fibre down and keeping it isolated the polarization can remain stable up to several days [Altepeter].

There is a extensive body of literature covering strategies for fibre coupling SPDC into a single-mode fibre [Migdall 97, Takeuchi 01, Migdall 02, Castelletto 03, URen 04, Castelletto 04, Torres 05, Ljunggren 05, Kurtsiefer 01, Dragan 04, Ling 08a], but the optimal scheme is still unknown. The reason for this is that there is not a closed form general solution to the problem. Numerous approximations are made in each case that affect the range over which a solution is valid. A common approach is to first use one of theoretical models to calculate the rough parameters that are needed to “get close”, and then experimentally test different lenses and fibre-coupling geometries until an optimum is reached.

The strategy I have found most useful for fibre coupling is due to Christian Kurtsiefer [Ling 08a]. The problem Kurtsiefer considers is a gaussian pump with a waist radius $W_p$ that downconverts into gaussian signal and idler modes with $1/e^2$ waist radiuses of $W_s$ and $W_i$ respectively. The signal (idler) mode makes an angle $\theta_s$ ($\theta_i$) with the pump as shown in figure 2.2. In this case, only SPDC within the $yz$ plane is considered and the phase mismatch along $x$ is set to zero ($\Delta k_x = 0$). For a crystal of length $l$, the amplitude of a pair being emitted from the pump into the signal and idler modes is related to the
overlap integral $\Phi(\Delta k_z)$

$$
\Phi(\Delta k) = \frac{\pi}{\sqrt{AC}} e^{-\frac{\Delta k_z^2}{4C}} \int dz e^{-4Z^2+iKZ},
$$

$$
A = \frac{1}{W_p^2} + \frac{1}{W_s^2} + \frac{1}{W_i^2},
$$

$$
C = \frac{1}{W_p^2} + \frac{\cos^2 \theta_s}{W_s^2} + \frac{\cos^2 \theta_s}{W_i^2},
$$

$$
D = \frac{\sin 2\theta_s}{W_s^2} - \frac{\sin 2\theta_s}{W_i^2},
$$

$$
F = \frac{\sin 2\theta_s}{W_s^2} + \frac{\sin 2\theta_s}{W_i^2},
$$

$$
H = F - \frac{D^2}{4C},
$$

$$
K = \Delta k_y \frac{D}{2C} + \Delta k_z. \quad (2.15)
$$

In a typical experimental setup the collected SPDC photons have a filtered bandwidth of only a few nanometers. In this case, we will assume perfect transverse phase matching $\Delta k_y = 0$. To fully account for transverse phase matching requires numerical methods [Boeuf 00]. The overlap integral now includes only the longitudinal phase mismatch $\Delta k_z$ and can be solved analytically:

$$
\Phi(\Delta k_z) \propto \int_{l/2}^{l/2} dz e^{-4Z^2+iKZ}
$$

$$
= l \int_0^1 du e^{-\zeta^2 u + \cos \left( \frac{\Delta k_z l}{2} u \right)} . \quad (2.16)
$$

The parameter $\zeta = \sqrt{Hl/2}$ is related to the walk off by the beams in the crystal. For thin crystals, where $l$ is much smaller than the Rayleigh length of the pump, signal, and idler modes, the walk off term is small and can be neglected. In this case the overlap integral takes the well known form of a sinc function. When the Rayleigh lengths of the beams are small relative to the crystal length, then the walk off term $\zeta$ dominates and the overlap integral takes on a Gaussian profile. In the limit where the Rayleigh ranges are $\ll l$, the length of the crystal ceases to play a role. This is because the interaction region where the SPDC takes place becomes so tiny that only a small amount of walk off is needed to halt the process.
Figure 2.2: *Model for optimizing the emission of SPDC into single-mode fibre.* A gaussian pump beam of waist $W_p$ is focused into a $\chi^{(2)}$ crystal of thickness $l$. The signal and idler photons are emitted at an angle $\theta_s$ and $\theta_i$ into gaussian modes with waists of $W_s$ and $W_i$ centred on the crystal.
When the waist of the beams is relatively constant over the length of the crystal, the emission rate of the SPDC into the daughter modes is [Ling 08b]:

\[
R_T \propto \frac{1}{(W_p W_s W_i)^2 \left( \frac{1}{W_p^2} + \frac{1}{W_s^2} + \frac{1}{W_i^2} \right)^2}.
\] (2.17)

In most experimental situations the signal and idler waists are the same \((W_s = W_i = W)\); usually matched fibre couplers with identical lensing systems are used. In this case we can determine the optimal ratio of signal and idler waist to the pump waist \(W_p = \gamma W\) from

\[
R_T \propto \frac{1}{W^2 \left( \frac{1}{\gamma^2} + 2\gamma \right)^2}.
\] (2.18)

The maximum downconversion rate occurs when \(\gamma = 1/\sqrt{2}\). To obtain the optimum emission the pump waist should be smaller than the signal and idler waists by a factor of \(1/\sqrt{2}\).

The rate equation implies that the tighter the focus, the brighter the emission. Taken to the extreme, focusing infinitely tightly should lead to infinitely bright emission. Of course this is incorrect; the derivation is only valid when the length of the crystal is small compared to the Rayleigh range of the beams. Focusing too tightly leads to large walk off effects that will decrease the brightness.

A way to estimate an appropriate beam radius is to estimate the range of angles that SPDC of a specific bandwidth emits into. SPDC is a very broadband process; it is not uncommon for the photon wave packets to have a bandwidth in excess of 100 nm. Narrow spectral filters are often employed to select out a smaller frequency range. In a typical degenerate SPDC experiment, the angles of emission of the daughter photons are \(\theta_s = \theta_i = \theta\). If both the signal and idler are passed through a filter with a centre wavelength of \(\lambda_s\) and a \(1/e^2\) bandwidth of \(\Delta\lambda\), the angular spread of the emission is

\[
\Delta\theta \approx \left| \frac{d\theta}{d\lambda} \right| \Delta\lambda.
\] (2.19)
Figure 2.3: Angular emission spectrum for the signal photon from type-I SPDC in BBO. The solid line represents perfect phase matching. The angular spectrum is calculated using Alan Migdall’s numerical phase matching program [Boeuf 00]. The angle for the BBO crystal is cut at 29.3° and the pump is at 404 nm.

The slope of the angular emission as a function of the wavelength \( \frac{d\theta}{d\lambda} \) can be calculated from the phase matching relations. Figure 2.3 shows a typical angular spectrum for the signal photon in BBO cut for type-I SPDC.

The goal is to collect all of the light that is emitted within this angular spread into a single-mode fibre. The angular spread sets the waist of the signal and idler beams

\[
W = \frac{\lambda_s \Delta \theta}{\pi}.
\]  

This beam radius must be matched to the mode-field radius of a single-mode fibre in
order to maximize the collection efficiency. For a typical single-mode fibre designed for \( \lambda = 800 \ \text{nm} \), the mode-field radius is 2.8 \( \mu \text{m} \). Mode matching can be accomplished using a lens system that images the mode-field radius of the fibre onto a spot of radius \( W \) at the surface of the crystal.

This spot size should only be taken as a starting point: the results from this method may not be the optimal solution. The ratio of the spot sizes between the daughter beams and the pump is arrived at using the thin crystal approximation. Also, \( \frac{d\theta}{d\lambda} \) depends on how tightly focused the pump is and its bandwidth. A numerical approach is required to fully optimize the problem. Alan Migdall’s group has developed a free FORTRAN program [Boeuf 00] to calculate many of the relevant parameters from SPDC, and the results from this program provide a useful check.

Experimentally I have found the best approach is to begin with the parameters specified from Kurtsiefer’s calculations, and then to test different combinations of focusing lenses for the pump and the collection system as well as various fibre coupling geometries. Fibre coupling can be a dark art at times: the number of elements and the quality of the optics are critical to the success of any scheme. A high end fibre coupling solution for a single beam can cost many of thousands of dollars, and it is often the case that the optimal predicted solution is not the best solution for the imaging optics available.

### 2.6 Experiments with Type-II SPDC

When I began my doctoral work, I inherited a pulsed type-II SPDC source designed to generate orthogonally polarized unentangled photon pairs. This source is used in the experiment discussed in chapter 4. The crystal is a 0.5 mm thick piece of BBO cut so that the external emission angle for the signal and idler photons is \( \sim 3^\circ \). The pump is produced by frequency doubling a mode-locked Ti:sapphire crystal. The mode-locked laser is a homebuilt kit purchased from KM Labs that operates at 80 MHz and outputs
400 mW of light at 810 nm in 100 fs pulses. Another 0.5 mm BBO crystal is used in the frequency doubling, leading to 40 mW of pump power at 405 nm. This pump is then focused onto the SPDC crystal to produce daughter photons at 810 nm. The crystal was tilted to collapse the downconversion cones and obtain beam-like emission [Takeuchi 01] with the daughter photons emerging noncollinearly at an angle of $\sim 3^\circ$. The two photons were then combined into the same spatial mode using a polarizing beamsplitter and collimated using a two lens system. This collimated beam was then sent to a single-mode fibre coupler.

One might wonder why we do not use the much higher powered Ti:sapphire laser as the pump to generate many more pairs at telecom wavelengths. Excellent low-cost fibre equipment exists in the telecom band that can not be found (easily) in the near infrared region. The reason is that the Si single-photon detectors needed in these experiments have a peak efficiency of $\sim$50-65% around 800 nm. For wavelengths above 900 nm the detection efficiency rapidly falls off. Recent developments have led to the development of high efficiency number-resolving detectors capable of operating over a wide wavelength range [Hadfield 05, Lita 08, Rosenberg 05, Divochiy 08, Hadfield 09, Kardynal 08]. Currently these detectors are not commercially available, but may become prevalent in the near future. If this happens, it may be desirable to move to telecom wavelengths.

The source was constructed in a small corner of the optical table; there was not much room available to make adjustments to the system. It was impossible to accurately measure the spot sizes on the crystals, but the pump beam and the two daughter beams were estimated to have a waist diameter of $\sim$100 $\mu$m in the crystal. I was also unable to verify whether the collapsed cone geometry provided better results over a collinear setup, but my predecessor claims to have seen about a 30% improvement in brightness [Mitchell].

In the first experiments conducted with the setup we were able to collect a maximum of several hundred photon pairs/second into single-mode fibre [Adamson 07]. Future
experiments required a brighter source of photons, so I set out to try and optimize the setup. At the time, the Ti:sapphire laser had suffered from some damaged optics. While fixing it, I took the opportunity to insert a 1:1 telescope into the cavity that doubled the cavity length. This lowered the repetition rate of the laser to 40 MHz, doubling the intensity of each pulse. This improvement doubled the SHG power. Each pulse generates four times the SHG intensity, but there are only half as many pulses. After the upgrade, the SHG power was measured to be 90 mW. Over time, damage to the doubling crystal as well as a failing pump laser reduced the SHG power back to 50mW.

The second change was to replace the spectral filter used. The original spectral filter had a bandwidth of 10 nm and transmitted 50% of the light. This was replaced by a new filter from the Japanese company Asahi Spectra which had a peak transmission of 70 % and a bandwidth of 12 nm at a central wavelength of 810 nm. The final adjustment was to replace the fibre coupler used. The original fibre coupler was a cheap fixed lens coupler in a mirror mount from Thorlabs mounted on a home-built \textit{xyz} translation stage. A high quality fibre coupler from New Focus (part #9131) with a better aspheric lens and more precise control over the relevant degrees of freedom was used. With these changes, the maximum fibre coupling efficiency observed was $\sim 6000$ pairs/s. In an experimental situation with lossy optics present, this decreased to a maximum of $\sim 2000$ detected pairs/s.

Due to the space constraints, further optimizations could not be made. Both daughter photons were combined into the same spatial mode using a polarizing beamsplitter and subsequently coupled into the \textit{same} fibre coupler. This prevented us from independently optimizing both beams simultaneously. There was always an asymmetry in the collection efficiencies of the signal and idler photons. This asymmetry may be a result of the collapsed cone geometry used; the signal and idler are orthogonally polarized which can lead to walk off effects [Lee 05], but a definitive reason was never discovered.
2.7 An Improved SPDC Source

After the completion of the experiment described in chapter 4, I began work on building a brighter SPDC source suitable for multi-pair experiments. Lee Rozema has assisted in the building and aligning of this new system. Type-I downconversion was chosen for its higher nonlinearity. A 2 mm long BBO crystal, with its optic axis cut at $29.3^\circ$, was used to downconvert pump photons at 404 nm to daughter photons at 808 nm with an emission angle of $\theta_s = \theta_i = 3.2^\circ$. The reason 808 nm was chosen is that high quality 3 nm bandwidth spectral filters with a transmission of greater than 90% are available from CVI Lasers. These filters were originally designed for Raman spectroscopy, but are suitable for use in SPDC experiments.

2.7.1 A New SHG System

The most important step in the development of a new SPDC source was the purchase of a commercial mode-locked Ti:sapphire laser from Coherent (Mira-HP). This laser has an output power of 4 W of pulsed light at 810 nm with a repetition rate of 76 MHz. The length of the pulses are $\sim 160$ fs in length. This large increase in laser intensity enables stronger nonlinear effects to be observed. A new doubling system was developed using this laser to pump the downconversion crystal. At first BIBO was tested as it has a higher nonlinearity than BBO. Two anti-reflection (AR) coated BIBO crystals, one with a length of 1.0 mm and the other with a length of 2.0 mm, were purchased from NewLight Photonics. In testing, both crystals suffered damage when the pump power exceeded 1 W. It was suspected that the coatings were burning in the presence of the high pump intensities. An uncoated 2 mm BIBO crystal was tested and did not suffer damage; however, reflection losses hurt the conversion efficiency. The unconverted beam was also elliptical and had a poor mode quality. A 2mm BBO crystal was instead chosen for the SHG. The AR coatings on BBO did not suffer from damage under the high pump
Figure 2.4: Second-harmonic power from a 2 mm AR coated BBO crystal. The 808 nm pulsed pump is focused into the crystal with a 50 mm focal length lens. A maximum conversion efficiency of \(\sim 50\%\) is achieved; pump depletion prevents higher conversion efficiencies from being obtained.

The output power and conversion efficiency for different pump powers is shown in figure 2.4. The conversion efficiency saturates at \(\sim 50\%\) as the pump is depleted. With BBO it is possible to generate over 1 W of doubled power, a factor of twenty improvement over the old laser system.

The tight focusing required in SHG leads to an elliptical beam; at the focus the walk off experienced by the pump is of the same order as the beam waist. To correct for this ellipticity, two different cylindrical lenses are used to collimate the frequency doubled output. A circularly symmetric pump should improve the mode matching between the pump and the SPDC photons.

---

When running at high power, occasionally the coating will be damaged. In this case the crystal is moved to an unburnt spot. With BBO this happens every couple of weeks under frequent operation.
2.7.2 Collecting the SPDC

A number of different collection geometries and imaging systems were tested. I will describe the setup that is the simplest to use and align, and consistently gave the highest count rates. I refer to this geometry, shown in figure 2.5, as the “bow tie” due to the criss-crossing paths the SPDC photons take to the fibre coupler. This double pass setup is similar to other SPDC geometries used in other experiments [Jennewein 02, Pan 03].

The pump is initially focused using a single lens into the crystal. On the other side of the crystal an achromatic lens placed one focal length away is used to collimate the SPDC and the pump beam. Two pairs of mirrors are then used to pick off the SPDC and send them to different single-mode fibre couplers. The pump is either absorbed by a beam block or retro-reflected to make a second pass through the crystal as shown in figure 2.5.

In the double pass, an identical collection setup is constructed on the other side of the crystal in order to pick off a second pair of downconverted photons.

Several combinations of focusing and collimating lenses for the pump and SPDC were tested. For the lenses available in the lab, the optimal focal length for the pump was found to be 15 cm. The New Focus fibre couplers have several aspheric lens with different focal lengths available. The 8 mm, 11 mm, and 15 mm focal length aspheres were tested with the 8 mm lenses performing the best. This was true when the couplers were placed anywhere from 30 cm to 1.2 m away from the pick off mirrors.

Once the photons are collected into fibre, they are sent to fibre-coupled single-photon detectors from PerkinElmer (SPCM-AQRH series). Coincidences between the detectors are recorded using a circuit designed by our electronics technician Alan Stummer [Stummera]. The circuit has eleven channels and is capable of detecting up to eleven-fold coincidences. The coincidence window is adjustable from 2 ns to 256 ns. For the measurements recorded, the coincidence window was set to 4 ns – less than the 13.1 ns separation between pump pulses. For a pump power of \(~700\, mW\), 200,000 pairs of photons per second were detected with a raw efficiency of \(~10\%\) (not including detector and other
Figure 2.5: A bow tie double pass setup to collect two pairs of SPDC photons into single-mode fibre. A strong pump (blue beam) is focused with a lens (L1) on a translation stage into a $\chi^{(2)}$ crystal, generating a pair of daughter photons each at half the wavelength of the pump (red beams). A second lens (L2) collimates both the pump and the SPDC. A pair of mirrors is used to pick the daughter photons off, sending them to different fibre couplers (D1 and D2) connected to single-photon detectors. A narrow-band interference filter (IF) is used to spectrally filter the photons. To generate a second pair, the pump can be retro-reflected back through the crystal and a similar setup used to pick off the resulting daughter photons. Coincidence detection events between all four detectors can be monitored with a coincidence circuit.
losses). Retro-reflecting the pump, we were able to collect 40,000 pairs of photons from
the second pass. The reason for the lower coincidence rate on the second pass is that the
lens used to focus the pump and collimate the SPDC is not achromatic; the focal length
depends on wavelength. Replacing this lens should lead to a significant improvement in
the collection rates. A total of 100 four-fold coincidences per second were observed. This
is nearly ten times better than other four-fold rates reported [Langford 05].

2.8 Alignment Tips and Tricks

The following is a guide for aligning a downconversion setup and coupling it into fibre.
These are the strategies, tricks, and techniques I have found most useful. I do not claim
they are optimal or the best way of doing things, but they have been helpful to myself.
While I will be addressing the bow tie geometry, most of the tips are general and can be
used for systems in different configurations.

2.8.1 Determining the correct orientation of the SPDC crystal

Phase matching will only occur when a crystal is correctly oriented with respect to the
pump beam’s polarization. To determine the correct orientation, we make use of the fact
that SPDC is the time-reverse of SHG. Place the crystal in a rotation mount and use the
light from the Ti:sapphire laser to look for SHG. The Ti:sapphire should be calibrated
with the polarization reference used to define the direction of linear polarization on the
table. It may be necessary to focus the Ti:sapphire depending on the strength of the
laser, but the SHG signal is usually a strong blue colour that is easy to see. Using a
power meter, it is easy to find the rotation angle that maximizes the SHG. Once this
angle is found, the crystal polarization has been calibrated to the table.
2.8.2 Aligning the pump beam

First make sure that the pump is well collimated and parallel to the table. A useful way to make sure the beam height is constant is to take two pieces of anodized aluminum (or any material that is not reflective and not combustible) and drill a hole in each of them at the desired beam height. With the addition of a mounting screw, these can serve as useful alignment irises in a variety of scenarios. I will assume that two irises are available, but having more is often beneficial. Whenever possible align the beam path to run along the holes in the table.

Inserting the SPDC Crystal. Make sure there are at least two mirrors in place so that the beam can be walked if needed. Send the beam down the path where the SPDC crystal will be placed, and then insert the SPDC crystal in the desired location. Set one alignment iris between the steering mirror and the crystal, and set the other iris after the crystal as far down the table as possible. Make sure that the pump is centred on both of these irises.

Inserting the focusing lenses. Place the lens that will focus the pump into the beam path. The lens is usually mounted on a translation stage that moves parallel to the beam path. Position the lens so that the beam passes through its centre. The reflected spot from the front surface of the lens can be used to help ensure the face of the lens is perpendicular to the incident beam. When the lens is aligned, the beam will be centred on the second iris (although it may be expanded from the focusing). Check to make sure that the focus of the pump is at the crystal. Once the first lens is well aligned, insert the second lens (again mounted on a translation stage) designed to collimate the pump. Again, make sure to position the lens so that the beam passes through its centre. Adjust the distance of the lens from the crystal until the beam is collimated. If the setup has been performed correctly, the two lenses should act as a 1:1 telescope. The pump should behave as if the crystal and two lenses are not present and follow the original path. If these lenses are achromatic, they should also collimate the SPDC emission.
2.8.3 Setting up the Fibre Couplers

*Some observations on the different fibre couplers available.* There are five degrees of freedom that must be controlled when fibre coupling: these are the two tilt angles perpendicular to the coupler's face, the transverse $xy$ positions of the coupler, and the distance of the focusing element from the tip of the fibre. The best (most expensive) couplers allow all five degrees of freedom to be controlled independently. In the New Focus couplers we use, the tilt controls are coupled to the focusing degree of freedom, and the $xy$ degrees of freedom were controlled using external translation stages. Recently we have been experimenting with the Thorlabs MBT61X series of couplers. The lens remains fixed while the fibre is moved by and $xyz$ translation stage. There is some coupling between the tilt and focus degrees of freedom, but not as much as in the New Focus Couplers. It is also much easier to set the focusing up with the Thorlabs stages. To control the beam position, two steering mirrors can be placed in front of the coupler. With both the Thorlabs and the New Focus couplers, removing the fibre requires a realignment of the coupler. This can be time consuming if there is a system where fibres need to be swapped frequently. Optics For Research (OFR) is a company (now owned by Thorlabs) that specializes in miniature fibre couplers that are of a high quality. These fibre couplers are about one inch in diameter and contain all five degrees of freedom built in. Because of their small size, all five degrees of freedom are strongly coupled to one another. These couplers are very difficult to align, but once aligned are very stable. It is possible to swap fibres without any discernible coupling loss. There are places where we have used these couplers for many months, swapping the fibres frequently, and not had to realign things. If high coupling efficiency is not required, these fibre couplers are serviceable. They are also useful as a stable collimator to out couple light into other parts of an experiment.

*Collimating the coupler.* The lens in most fibre couplers can be used to image the fibre core onto the surface of the crystal, but this can be difficult to setup and align. It
is often simpler to use the fibre coupler to couple a collimated beam. Trying to couple a collimated beam into a fibre is the same problem as trying to collimate light emitted by a fibre. Using a fibre coupled alignment laser (preferably in the visible), send light through the fibre so it is emitted by the coupler. Using the controls on the coupler, collimate the beam. The irises should be used to make sure the collimated beam is parallel to the table and the same height as the SPDC. The alignment laser may have a different wavelength than the SPDC, but it should still be close to being collimated when switching to the downconversion. A notable exception are the couplers from OFR where going from an alignment laser at 630 nm to SPDC at 808 nm can cause an 80-90% drop in coupling efficiency. Even with this large wavelength dependence there will still be some signal that can be collected. Once this signal is found the collimation parameters can be tweaked to increase the collection efficiency.

2.8.4 Picking off the Downconverted Photons

Setting up the mirrors. The SPDC beams are close to the pump beam, so care needs to be taken when separating them from the pump. The easiest geometry I have found to accomplish this is to use two pick off mirrors in the orientation shown in figure 2.5. This allows the mirror mounts to be oriented away from the pump beam. It may be necessary to glue or tape the mirror to the mount so that the mirror extends outside the edge of the mount. Place the pick off mirrors as close to the pump as possible after the SPDC collimating lens. Set up two steering mirrors to send the SPDC to the fibre couplers.

Aligning the beam paths. Place the collimated fibre couplers on the table as shown in figure 2.5. Use an alignment laser to send light backwards from the couplers to the crystal. To set the correct angle of the beams to match the emission angle of the SPDC, it is helpful to use a translation stage and a tall thin object like a small Allen key. Place the stage so that it moves perpendicular to the pump beam. Insert the Allen key so that it is between the pick off mirrors and centred on the pump. Using trigonometry,
calculate how far the two SPDC beam should be from the pump. Move the allan key so that it is this distance away from the beam. Adjust the pick off mirror so that it is nearly touching the pin (don’t actually touch the pin though lest mirror be scratched). This is where the SPDC will be hitting the mirror. Using the steering mirror, direct the SPDC onto the centre of the pin. Move the pin out of the way, and then use the pick off mirror to centre the alignment beam on the pump beam within the crystal. Make sure to use a low pump power and laser safety goggles while doing this. Repeat this procedure for the other SPDC beam.

2.8.5 Collecting photon pairs.

Hook the two collection fibres up to the single-photon detectors and insert the spectral filters. Turn up the pump power and look to see if there are coincidences. If you are fortunate, there will be some pairs observed. Most likely there will be none. The first thing to do is to pick one of the SPDC beams and use the two steering mirrors to walk the beam around on the crystal. This may take some time. Be careful not to walk too far away – if this happens, start over with the alignment laser. Once SPDC pairs are seen, it is relatively straight forward to optimize the collection rate. Iterate between adjusting the mirror positions and the Fibre couplers. Also tilt the crystal to ensure that the phase matching angle is optimal.

2.8.6 Setting up the second pass

If a double pass geometry is required, insert a mirror after the first set of pick off mirrors to retro-reflect the pump back through the crystal. Repeat the setup procedures of the pick off mirrors and couplers on the other side of the crystal. There are fewer degrees of freedom as the collimating lens for the SPDC on one side is the pump focus for the other. It may be necessary to iterate optimizing the collection from one side of the crystal and then the other.
2.8.7 Other Tips

*Pick mirrors carefully.* Many dielectric mirrors are not designed for short femtosecond pulses. This can cause strange dispersive effects. Metal mirrors avoid many of these problems, but are subject to higher losses. For the blue light losses can be be much higher, so make sure to check the mirror specifications carefully before ordering.

*Minimize stray light* With an intense blue pump, most things fluoresce. Make sure that there are no misbehaving coatings or pieces of glass in the beam path that are giving off fluorescence. The SHG process involves intense beams and can lead to a lot of scatter. To minimize this scatter, beam blocks or even containment boxes can be constructed. Many fibre claddings allow light to leak in. Make sure that fibre cables are hidden away from any stray light sources.

*Clean the fibres regularly* Inserting and removing fibres can cover their cores in dirt and oils. Use a fibre inspection scope to check to see if there is dirt, oil, or scratches on the fibre core. After cleaning the fibre, be sure to check that the dirt has been removed. Cleaning a fibre can lead to dramatic enhancements in collection efficiencies. A dirty fibre is often the culprit behind drops in efficiency.

*Install Climate Controls.* The alignment and stability of a Ti:sapphire laser is strongly dependent on the humidity and temperature of the lab. If it is very humid or there is a large temperature swing, the laser may drift out of alignment or cease to mode-lock. The best thing to do in these cases is not try and undertake a major realignment of the laser. Doing so will almost certainly necessitate a realignment of everything downstream. With the use of irises this is made simpler, but will still most likely take several days. What will most likely happen is that within a day or two the humidity/temperature will return to normal. If you have just finished realigning the laser, you will now have to start realigning things again. This is especially true in the summer when it rains. On days like these, find a nice theory problem to work on.

*Leave enough room.* A common mistake made in nearly every experiment is that not
enough room is left. Invariably, experiment designs change as more optics need to be added. Unless there are severe constraints, a useful rule of thumb is to leave double the amount of room you think you need\textsuperscript{6}.

2.9 Conclusions

When I first started in the lab our brightest source was able to collect several hundred coincidences/second into single-mode fibre. Through the optimizations and upgrades I have undertaken, our brightest source is now more than two orders of magnitude brighter. We are currently setting up an Type-I sandwich source to generate entangled photons pairs, and investigating new collection geometries to allow six and eight photon experiments to be carried out. With the improvements made to our collection and pump systems, our lab will be able to continue carrying out cut edge experiments in quantum information and metrology for many years to come.

\textsuperscript{6}This rule of double also applies to memory in computers, to hard drive space, and to toilet paper. Buy twice as much as you think you need.
Chapter 3

Semiconductor Quantum Dot

Single-Photon Sources

One way to engineer quantum states of light is to use single-photon sources. A heralded downconversion setup can be used as a single-photon source [Hong 86, Pittman 05, Fasel 04], but the probability of emitting multiple pairs can hurt the single-photon character of the source, and a trigger is required. Ions [Kimble 77, Diedrich 87] and molecules [Basche 92, Fleury 00] can also be used as single-photon sources, but it can be difficult to efficiently isolate and collect the emission. Colour centres, such as nitrogen vacancies in diamond, can be used as room temperature single-photon sources [Kurtsiefer 00, Broui 9], but they are highly multi-modal and unsuitable for many quantum information applications. A relatively new single-photon technology is based on self-assembled semiconductor quantum dots in cavities [Benson 00, Santori 02b, Santori 02c, Waks 02, Yoshie 04, Flagg 09]. These devices can be integrated into all optical semiconductor circuits and are promising sources of single photons. Such a source can be thought of as a type of photon “turnstile”, allowing only one photon to be emitted at a time.

A portion of my doctoral work was spent setting up a single-photon source using semiconductor quantum dots in microcavities manufactured at NIST by our collaborators
Richard Mirin and Martin Stevens [Mirin 04]. Our original interest in quantum dots was to see if they would be a viable method for generating N00N states. While the quality of the dots is not yet high enough to generate entangled multi-photon states, the setup has been a useful source of single-photons in other experiments.

### 3.1 Quantum Dot Properties

Quantum dots are often called artificial atoms as they exhibit many of the same properties and behaviours as real atoms. This atom-like phenomena arises from the fact that quantum dots are confined in three-dimensions, leading to a quantization of the energy levels present. In semiconductors, this occurs when the physical size of a quantum dot is comparable to the DeBroglie wavelength of an exciton (electron-hole pair) that occupies the dot [Seidl 06, Gerardot 07]. For a quantum dot of radius $r$, the confinement leads to discrete energy levels that are evenly spaced by $\sim 1/r^2$. The close proximity of charges in a quantum dot can give rise to Coulomb effects that scale as $\sim 1/r$. In small dots the confinement dominates while in larger dots Coulomb effects shift the energy levels.

The self-assembled semiconductor quantum dots we use are based on InGaAs, and are grown on top of a GaAs substrate using molecular beam epitaxy (MBE). MBE allows a single monolayer at a time to be deposited. There is a lattice mismatch between InGaAs and GaAs that induces a strain in the system. For the first couple of monolayers it is energetically favourable for the InGaAs to follow the lattice structure of the GaAs substrate. This base layer of InGaAs is called the “wetting layer”. When additional material is deposited onto the wetting layer, the InGaAs clumps together around strain locations to form quantum dots. The situation is similar to the behaviour of water on the hood of a freshly waxed car; it is energetically favourable for the water to “bead” up into tiny droplets. By controlling the temperature and rate of growth, it is possible to control the size and density of the dots, but the process is inherently random. The
quantum dots we use are typically 5-10 nm high and 20-40 nm in diameter. Once the
dots have formed, another layer of GaAs is grown on top to passivate the surface. A
document page
of picoseconds) to the ground state of the quantum dot before recombining to emit a photon [Santori 02a]. This relaxation process is nearly two orders of magnitude faster than the lifetime of the upper states, hence the emission from these upper states is suppressed. In a perfectly symmetric quantum dot the emission of the single exciton state X will either be right-hand circularly polarized or left-hand circularly polarized. In practice, remnant strain fields and asymmetric shapes break the symmetry of the dot. This lifts the polarization degeneracy of a single-exciton recombination and leads to an energy splitting. In this case, the emitted photon will either be horizontally or vertically polarized with different wavelengths.

When two excitons are captured by a quantum dot, the Coulomb interaction between them shifts the energy levels as shown in figure 3.2. The energy level of this biexciton state XX can be either red or blue shifted depending on the specific dot in question. The first exciton to recombine will be emitted as a photon with a wavelength $\lambda_{XX}$. Once the first exciton recombines, the Coulomb interaction disappears and the remaining exciton recombines to emit a photon at a different wavelength $\lambda_X$. As more excitons are captured by the quantum dot, other excited states can be populated. The dynamics between these different excitons may be complex, but the last exciton remaining in the dot will not be subject to any Coulomb interactions; it will recombine to emit one photon with a wavelength $\lambda_X$. This guarantees that the emission from X contains only one photon at a time. The separation in wavelength between the emission from X, XX, and the other excited states can be up to a nanometer—allowing the X line to be spectrally filtered to create a single-photon source.

In principle, the biexciton decay in a symmetric quantum dot should lead to a polarization entangled state. In practice, the polarization splitting of the X state is often larger than the natural line width of the exciton emission – leading to distinguishing information that destroys the entanglement. To generate entangled photon pairs using a quantum dot, the degeneracy in the X state needs to be restored. This has so far been accom-
Figure 3.2:  

**a) Schematic of above-band excitation process.** An optical pump creates electrons and holes inside the wetting layer. These carriers can be captured by a quantum dot which has quantized energy levels. The electron and hole rapidly relax to the ground state (a process that lasts only tens of picoseconds) before recombining and emitting a photon at the wavelength $\lambda_X$. 

**b) Energy level diagram for a quantum dot.** A quantum dot with one exciton present is in the state $X$. The presence of asymmetry in the dot structure breaks the polarization degeneracy in the $X$ state, leading to an energy splitting between the polarizations of $\Delta E_s$. When two excitons are present, the quantum dot is in the state $XX$. The Coulomb interaction between the two excitons leads to a shift in the energy level of the $XX$ state by $\Delta E_c$. From here the dot can return to an empty state 0 by emitting either two $H$ or $V$ polarized photons.
plished through strong spectral filtering of the emission from the X state [Akopian 06], and by the application of an in-plane magnetic field [Stevenson 06, Young 06, Shields 07]. While these techniques show promise, the degree of entanglement present is still low and the pair production rates are many orders of magnitude lower than that of SPDC.

3.2 Micropillar Cavities

The emission from a quantum dot can occur in any direction, making the efficient collection of photons difficult. To help increase the collection efficiency, a cavity can be constructed around a quantum dot. When a quantum dot is in resonance with the cavity, it will preferentially emit photons into the cavity mode. The emission pattern of the cavity is narrow, allowing a larger fraction of the photons to be collected. When a quantum dot is weakly coupled to a cavity the spontaneous lifetime of the emission is also reduced, an effect first noticed by Edward Purcell [Purcell 46, Yablonovitch 87]. For a semiconductor cavity with quality $Q$ and the mode volume $V$, the enhancement in the spontaneous emission lifetime is referred to as the Purcell factor

$$F \propto \frac{3Q\lambda_0^3}{4\pi^2Vn^3},$$

(3.1)

where $n$ is the index of refraction of the material and $\lambda_0$ is the wavelength of the emission in free space. By increasing the quality of the cavity or reducing its mode volume (making the cavity smaller), it is possible to obtain large enhancements in the spontaneous emission lifetime. This result is only valid in the weak-coupling regime; in the presence of strong coupling a quantum dot can emit a photon into the cavity mode and resonantly reabsorb it many times before the photon leaks out of the cavity.

The cavities used in our quantum dot samples are micropillar structures; a picture from a typical micropillar taken with a scanning electron micrograph is shown in figure 3.1 b). The first step in constructing a micropillar cavity is to sandwich the layer of quantum dots between two distributed Bragg reflectors (DBR), creating a 2D planar cavity. DBRs
are mirrors that are formed by growing alternating, quarter-wavelength thick, layers of different semiconductor materials. In our dots, the alternating layers are AlAs and GaAs. The bottom DBR consists of 26.5 layers to form a high reflector while the top DBR is made up of 14 layers and serves as an out-coupler; the photons are more likely to escape out of the top of the cavity rather than the bottom. To provide transverse confinement, the sample is etched to form pillar structures. Before the etching takes place, a mask is applied to the sample that sets the diameters and locations of the pillars. All the material not protected by the mask is removed by the etching. The large index gap between air and the semiconductor material provides strong transverse confinement through total internal reflection leading to a 3D cavity. The cavities surrounding the dots we study typically have a $Q \sim 1000 - 2000$. To help find the cavities, a numbered grid system is also etched into the sample.

As the formation of quantum dots occurs randomly on a sample, not every cavity is guaranteed to have a quantum dot. Many of the cavities will be empty while others may have more than one dot. Because of the fluctuations in the dot size, different dots will emit at different wavelengths; this makes it possible to filter out the emission of a single dot of interest. Figure 3.3 shows a typical spectrum from a quantum dot in a $1.5 \mu m$ diameter micopillar structure. The exciton and biexciton lines are clearly visible. The cavity mode has a line width ($>1$ nm) that is much broader than emission from the dots. The other peak in the diagram is most likely from another dot that is not as strongly coupled to the cavity.

For a dot to optimally couple to a cavity it must be placed precisely at the centre of the structure, but the randomness in the dot distribution means this rarely occurs. A quantum dot can be brought into resonance with a cavity through temperature tuning. As the temperature is varied, the quantum dot’s centre frequency changes rapidly as compared to the cavity frequency as shown in figure 3.4. For this particular dot, resonance with the cavity and the maximum emission rate is achieved at a temperature of $\sim 25$ K.
Figure 3.3: Spectrum from a quantum dot in a cavity. The peak labeled $X$ is from single-exciton emission while the $XX$ peak is from the biexciton emission. The broad background is from the cavity mode. There is a clear shift in the energy between the $X$ and $XX$ that is easily resolvable by the spectrometer with a resolution of $\sim 0.2$ nm. The third peak is most likely from a second dot that is not on resonance with the cavity.
Although each cavity may have more than one quantum dot in it, it is extremely unlikely that both quantum dots will be co-resonant with the cavity at the same time.

3.3 Experimental Setup

3.3.1 Cooling the Quantum Dot

At the heart of the quantum dot experimental setup is a liquid Helium cryostat from Cryo Industries (model CFM-1738-102). This cryostat can operate at higher temperatures using liquid Nitrogen instead of Helium, and comes with a “no-move” option. With the no-move option, the sample holder is mechanically decoupled from the transfer line and is specified by the manufacturer to drift < 150 nm after thermal equilibrium is reached. Compared to other cryostats this is very stable. A typical experimental data run can last for several hours and involve coupling the dot emission into a single-mode fibre; with this cryostat the dot rarely drifted out of alignment with the collection system—even when the transfer line was accidentally bumped.

The cryostat comes with two AR coated quartz windows allowing optical access to the sample from two sides if needed. To mount the quantum dots, one of the quartz windows is removed granting access to the sample holder. Low-temperature vacuum-friendly Apezion N thermal grease is used to affix the quantum dots to the sample holder. The grease brings the dots into thermal contact with the cooled sample holder and also affixes them in place. As the cryostat is operated vertically, the chip containing the quantum dots may sometimes lose contact and fall off the sample holder. Some micropillars may be damaged by this, but the structures are surprisingly robust and most will remain undamaged. The most common reason for a sample to fall off is because either too much or too little thermal grease has been applied to the back of the chip. When it comes to thermal grease it is better to err on the side of too little; when too much is applied it can coat the surface of the chip and ruin the optical coupling from the
Figure 3.4: Temperature tuning a quantum dot. By varying the temperature, it is possible to shift the emission line of a quantum dot. Temperature tuning is used to bring a dot in resonance with a cavity. The emission from the dot is red-shifted as the temperature increases.
Built into the cryostat sample holder is a small heater that can be used to control the temperature of the sample. A Lakeshore Model 331S temperature controller is used to set and maintain the temperature of the sample. The temperature controller includes a feedback loop that allows it to keep the temperature stable to an accuracy of 0.01 K. The temperature controller is useful for tuning the quantum dot into resonance with the cavity.

The cryostat is mounted vertically on the table using two long 1.5” diameter stainless steel posts from Thorlabs. When unlocked, the cryostat can move up and down the post to roughly adjust the vertical position. Once locked, the cryostat remains in position. Before cooling down, it is important to use a pump to create a vacuum in the sample chamber and thermally insulate the dot. A sufficient vacuum ($\sim 10^{-6}$ torr) can be achieved using a standard mechanical pump. The transfer tube can also be pumped down using the mechanical pump to thermally isolate it and prevent excess heating of the liquid He.

Two people are required to insert the transfer tube into the cryostat and liquid He tank; trying to do this by one’s self can lead to bodily harm or, worse, damage to the transfer tube. One person is responsible for inserting the transfer tube into the liquid He tank while the other is responsible for inserting the transfer tube into the cryostat. To start, one end of the transfer tube is lowered into the liquid He tank causing the liquid to boil off and raising the pressure inside the tank. Typically the pressure will rise to $\sim 5$ psi but may hit $\sim 10$ psi at which point the safety release valves will vent the excess pressure. While the pressure is building inside the tank, the other side of the transfer tube should be partially lowered into the cryostat. Opening the valve on the transfer line allows the liquid He to start flowing from the tank to the cryostat. Once there is a steady flow, He gas will begin to escape from the vent valve on the cryostat. Once this happens, finish lowering the transfer line into the cryostat and *loosely* tighten the valve.
that seals the transfer tube in place. If any of the fixtures are too tight, it may be very difficult to undo them once they are frozen. Once the He starts to come into contact with the cold finger attached to the sample holder, the temperature will rapidly drop. It takes \( \sim 15 \) minutes to cool down to 4 K from room temperature.

To reduce the amount of He consumed, slowly tighten the valve on the transfer line that regulates the flow. When the transfer line is closed too tightly, the temperature will begin to rise quickly. Open up the valve a bit and wait for the temperature to come down. Then try to close the valve some more. This may take \( \sim 15-20 \) minutes before the valve is closed as far as it can be. Once it is closed, the cryostat consumes about 0.5-0.7 liters of He/hour and requires 1-1.5 litres to cool down. If the pressure in the He tank drops too low the transfer may stop; in this case connect a tank of He gas to the liquid He tank to boost the pressure. If during the cool down process condensation forms on the cryostat window, remove the transfer line and warm the sample up. Condensation occurs when the sample chamber is not under vacuum. Check to make sure the O rings form a correct seal and try to pump down the sample chamber again. If the transfer suddenly stops while there is still plenty of liquid He left in the tank and sufficient pressure built up, the transfer line may be blocked inside by ice. In this case, warm the system up and wait for the ice to melt. To speed this process up, a heat gun can be used to rapidly warm the transfer line. Make sure that the transfer line is not in the liquid He tank when you do this! Heating up the transfer line while it is directly connected to the liquid He tank can cause the He to rapidly boil off and rapidly raise the pressure inside the tank to dangerous levels.

To bring the sample back to room temperature again requires two people. Make sure that there is no exposed skin on the arms and that thick insulating gloves are worn. The person who is removing the transfer line from the cryostat should wear safety goggles. First close the valve on the transfer line to stop the transfer of liquid He. Both sides of the transfer tube should then be raised simultaneously. The end on the cryostat will be
removed first as it is shorter. Take care while removing the transfer line from the liquid He tank—the tube will be very cold and a large amount of pressure may have built up. When the tube is removed, a geyser of cold He gas will erupt from the top of the tank. If the cold gas comes in contact with skin it can cause burns. Once the transfer line is removed, it takes approximately one hour for the sample chamber to return to room temperature.

### 3.3.2 Collecting Light from the Quantum Dot

In our experiment a Ti:Sapphire laser was used to pump the quantum dots. This is the same laser that is used in the SPDC experiments. A small portion of the laser light is picked off and coupled into a single-mode fibre that is then sent through 15 m of fibre to another room containing the quantum dot. The Ti:Sapphire laser has a centre frequency of 810 nm and is above the band gap of InGaAs. As a result, we excite carriers in the wetting layer which are then captured by the quantum dot. Most of the dots we study emit photons in a wavelength range of 925-945 nm. To resonantly excite the dots using our laser would require the purchase and installation of a picosecond optics kit for the Ti:Sapphire laser. It is possible to tune our laser in CW mode out to the dot wavelengths, but the laser’s centre frequency is not very stable and jumps around (this is because the system is optimized for mode-locked operation). It is also difficult to filter out the dot emission from a resonant pump laser. For our purposes, above-band excitation is sufficient.

If the Ti:Sapphire is mode locked, the short 100 fs pulses are significantly broadened by the passage through the fibre. A 10 nm narrowband filter centred at 810 nm is used to ensure that the light from the excitation laser is spectrally distinct from the quantum dot emission. The light from the Ti:Sapphire is reflected by a dichroic mirror designed to reflect light below and transmit light above 890 nm. The Ti:Sapphire light is focused onto the quantum dot using an aspheric lens with a numerical aperture (NA) of 0.5
as shown in figure 3.5. The same lens used to focus the excitation laser is also used to
collect the dot emission. The advantage of using an asphere is that it has a relatively long
working distance of 5.9 mm allowing it to focus on the quantum dots through the cryostat
window. An asphere is able to focus to a small spot size and works well to collimate the
dot emission. The disadvantage of an asphere is that it is wavelength dependent; the
optimum focus for the dot emission is different than the excitation laser. When the
collection from the quantum dot is optimized, the spot size of the pump will be much
larger and more pump power is required to excite the dots. It is possible to use a
microscope objective with a long working distance, but these are rarely designed for the
wavelength range used in the quantum dots. To have one custom build is prohibitively
expensive and the multiple elements used will reduce the collection efficiency. Another
possibility is to bring the pump in from a steep angle and focus it separately onto the
quantum dot. For our purposes, the confocal geometry proved sufficient, but for more
complex resonant pumping schemes a side pumping geometry might be necessary. In this
case, the polarized pump can be brought in at Brewster’s angle to minimize unwanted
reflections into the collection system.

The lens is mounted on a specially constructed three-axis stage designed to keep the
beam in alignment independent of the stage motion. This allows the cryostat to remain
fixed while the collection system is moved around the dot surface. The quantum dot
emission is collimated by the asphere and transmitted through the dichroic mirror. An
AR coated piece of GaAs provides extra filtering by blocking all remaining light below
890 nm. The quantum dot light is then focused into a spectrometer using a 5 mm focal
length lens. The spectrometer is an Andor SR303I which comes equipped with three
switchable gratings. For the measurements in this thesis the 1800 lines/mm holographic
grating was used giving a spectral resolution of \(\sim 0.2\) nm.

\footnote{It was not possible to separately focus the pump beam before the dichroic as the distance from the
dichroic to the sample was \(\sim 1\) m}
Figure 3.5: *Diagram of quantum dot apparatus.* The quantum dots are placed in a liquid He cryostat. A fibre-coupled Ti:Sapphire laser, operated in either CW or pulsed mode, can be used to excite the dots. A polarizing beamsplitter (PBS) and a half waveplate (HWP) are used to control the pump intensity. The emission from the dots is collected using a confocal geometry. A dichroic mirror that reflects light below and transmits above 870 nm is used to separate the pump from the dot emission, and a second filter (IF2) is used to remove any excess pump contamination. The dot emission is then sent to a spectrometer where a cooled CCD camera can be used to study the spectrum of the light. Alternatively, the spectrometer can act as a monochromator and transmit a selected wavelength through an output slit. This spectrally filtered light is then coupled into a fibre after passing through a HWP and PBS to select out a single polarization. To image the surface of the dot sample, a white light source and CCD can be inserted into the system. *Inset: A Hanbury Brown-Twiss Interferometer to measure the second-order coherence properties of light. A fibre beamsplitter is used to split the emission from the quantum dot between two SPCM. The arrival times between these two SPCM is measured using a home-built timing circuit described in the text.*
A thermo-electrically cooled CCD (Andor DU401A-BR-DD) is used to study the spectrum from the quantum dot. The camera can be cooled to $-85^\circ$ C to suppress unwanted dark counts. At this temperature the quantum efficiency of the CCD in the dot wavelength range is $\sim 30\%$ —higher than other Si based detectors. The detector contains 1024x128 pixels and each pixel is a 26 $\mu$m x 26 $\mu$m square. To obtain the spectra, the vertical pixel bins are summed. A mercury lamp with well known spectral lines is used to calibrate the spectrometer and camera. The camera system is quite flexible and can be removed from the spectrometer to be used in other experiments.

A flip mirror inside the spectrometer can be activated to direct the quantum dot light towards an output slit. Closing down the output slit enables a single quantum dot line to be selectively filtered. As the slit is narrowed the spectral resolution increases, but the number of photons collected decreases. Different slit sizes must be tested to find the optimal trade off between resolution and count rates (which depends on the intended application). After the output slit, a 5 mm focal length lens collimates the light from the spectrometer. A half-wave plate and polarizing beamsplitter are used to filter out a single polarization from the X state. The light is then coupled into a fibre and is ready to be sent to other experiments. The light is no longer in a single Gaussian transverse mode due to the clipping from the spectrometer slits, making the coupling into single-mode fibre difficult. Typically a multi-mode fibre is used to perform the initial coupling before switching to a single-mode fibre.

To find the quantum dots of interest, a simple imaging system is used. A flip mount with a piece of glass is used to couple a bright white-light source into the beam path. The white light reflects off the dot sample and is then sent to a web cam located after the dichroic mirror and GaAs filter. The web cam is the Philips SPC900NC. This web cam has very good low light performance and is used by amateur astronomers to build inexpensive telescope imaging systems. The image collected from the quantum dot surface is magnified using a 30 cm focal length lens. The etched numbers on the quantum dot
grid are easy to make out on the CCD camera, allowing a desired dot to be easily found. When the white light source is turned off, the emission from the cooled dots should be readily visible on the CCD. It may take some searching to find a micropillar cavity that contains a quantum dot. The numbers that are etched into structure may also contain quantum dots. In this case the etched numbers act like 2D planar cavities and have a broader emission spectrum. Care must be taken to avoid accidentally collecting from a number instead of a cavity. The spectrum of the emission can help distinguish between these two cases.

### 3.4 Photon Statistics

To verify that our quantum dots are acting as single-photon sources, it is necessary to study their emission statistics. To characterize the statistical properties of light correlation functions must be measured. The first order coherence of a beam of light is defined as the correlation between two field amplitudes, and is typically measured using an interferometer. The second order coherence is a measure of the correlations of two optical intensities, and provides information on the photon statistics between the two beams. For a single mode stationary light source, the second order coherence between the intensity at a time \( t \) and \( t + \tau \) is defined as

\[
g^{(2)}(\tau) = \frac{\langle \hat{a}^{\dagger}(t)\hat{a}^{\dagger}(t + \tau)\hat{a}(t + \tau)\hat{a}(t) \rangle}{\langle \hat{a}^{\dagger}(t)\hat{a}(t) \rangle^2}.
\] (3.2)

In principle it is possible to directly measure \( g^{(2)}(\tau) \) using an ideal number-resolving detector; however, real detectors have a finite time resolution and suffer from dead time. After a photon is detected there is a recovery period before the detector can be triggered again. Another method to measure \( g^{(2)}(\tau) \) is to use a Hanbury Brown-Twiss (HBT) interferometer that consist of a single beamsplitter that sends the light to two different detectors [Loudon 73, Kim 99]. Using the beamsplitter relations from equation 1.35, we find that for a beam entering arm 1 of a beamsplitter (vacuum in arm 2) and detectors
at the output ports 3 and 4 [Loudon 73]:

\[ g^{(2)}(\tau) = \frac{\langle \hat{a}_1^\dagger(t)\hat{a}_1^\dagger(t+\tau)\hat{a}_1(t+\tau)\hat{a}_1(t) \rangle}{\langle \hat{a}_1^\dagger(t)\hat{a}_1(t) \rangle^2} = \frac{\langle \hat{a}_3^\dagger(t)\hat{a}_3(t) \rangle \langle \hat{a}_4^\dagger(t+\tau)\hat{a}_4(t+\tau) \rangle}{\langle \hat{a}_3^\dagger(t)\hat{a}_3(t) \rangle \langle \hat{a}_4^\dagger(t)\hat{a}_4(t) \rangle} = \frac{\langle \hat{n}_3(t)\hat{n}_4(t+\tau) \rangle}{\langle \hat{n}_3(t) \rangle \langle \hat{n}_4(t) \rangle}. \] (3.3)

The time difference \( \tau \) between detection events at the two output ports provides a measure of \( g^{(2)}(\tau) \). To measure the time between detection events, a coincidence timing circuit is used. When one of the detectors fires, an internal clock within the timing circuit is activated. The clock is stopped a time \( \tau \) later when the other detector fires. After many detection events have accumulated, the statistics for the source can be evaluated.

For a coherent state with Poissonian statistics \( g^{(2)}(\tau) = 1 \). This means that the photons are uncorrelated in their arrival times: each photon acts independently. For a single-mode thermal source, \( g^{(2)}(0) = 2 \). If one photon is detected it is twice as likely that a second one will be detected immediately after; the photons arrive in bunches. Hence a source with a \( g^{(2)}(0) > 1 \) is said to be bunched. For an ideal single-photon source, \( g^{(2)}(\tau) = 0 \). Immediately after a photon has been detected a second photon is never seen. When \( g^{(2)}(\tau) < 1 \) photons are less likely to arrive one after the other, and the source is said to be antibunched. Antibunching is a purely quantum effect as classically \( g^{(2)}(\tau) \geq 1 \) [Loudon 73].

### 3.4.1 The \( g^{(2)}(\tau) \) of a single atom

For the subsequent analysis, we treat our quantum dots as if they were ideal atoms. A single driven atom has a second order coherence function of

\[ g^{(2)}(\tau) = 1 - e^{-|\tau|/\tau_\text{TR}}, \] (3.4)
Chapter 3. Semiconductor Quantum Dot Single-Photon Sources

where $T_R$ is the radiative lifetime. The $g^{(2)}(\tau)$ function is a double-sided Lorentzian that dips to 0 when $\tau = 0$. All physical detectors have a finite time resolution $T$. As long as $T$ is comparable to $T_R$, the detector’s finite response must be taken into account. The ideal $g^{(2)}(\tau)$ for a single atom is integrated over the response time of each detector. Assuming that both of the detectors at output ports 3 and 4 have the same time resolution $T$,

$$g^{(2)}_{\text{det}}(\tau) = \frac{1}{T^2} \int_t^{T+t} \int_{t'}^{T+t'} dt \left( 1 - e^{-|\tau|/T_R} \right)$$

$$= \frac{1}{T^2} \int_t^{T+t} dt_3 \int_{t'}^{T+t'} dt_4 \left( 1 - e^{-|t_4-t_3|/T_R} \right)$$

$$= \begin{cases} 
1 - \frac{T^2}{T_R} \left( e^{-\frac{(T+|\tau|)}{T_R}} + e^{-\frac{(T-|\tau|)}{T_R}} - 2e^{-\frac{|\tau|}{T_R}} + 2\frac{T}{T_R} - 2e^{-\frac{|\tau|}{T_R}} \right) & \text{if } \tau \leq T \\
1 - \frac{T^2}{T_R} \left( e^{-\frac{(T+|\tau|)}{T_R}} + e^{-\frac{(T-|\tau|)}{T_R}} - 2e^{-\frac{|\tau|}{T_R}} \right) & \text{otherwise}
\end{cases}$$

(3.5)

For $\tau = 0$, this reduces to

$$g^{(2)}_{\text{det}}(0) = 1 - \frac{T^2}{T_R} \left( 2e^{-\frac{T}{T_R}} + 2\frac{T}{T_R} - 2 \right).$$

(3.6)

From this, we see that the dip no longer reaches 0, reducing the measured $g^{(2)}_{\text{det}}(0)$ for the system.

### 3.4.2 Measuring the $g^{(2)}(\tau)$ from a quantum dot

Using a HBT interferometer, as shown in figure 3.5, we can measure the $g^{(2)}(\tau)$ of the emission from a quantum dot. The spectrometer is used to select out only photons from the X transition at $\lambda_X$. The filtered photons from the dot are then coupled into multimode fibre. Multimode fibre is used to increase the count rates, decreasing the acquisition time.

As long as the spectrometer is able to adequately filter the dot emission, it was found that using single-mode fibre provided a negligible enhancement to the measured $g^{(2)}(\tau)$. A fibre beamsplitter is used to split the incident photons between two PerkinElmer single photon counting modules (SPCM). At the quantum dot emission wavelengths, these detectors have a quantum efficiency of $\sim 20\%$ and a specified jitter of $\sim 400$ ps.
a detector fires, it sends a TTL pulse to a coincidence circuit that then measures the
time between detector events. The circuit was built by our electronics technician Alan
Stummer; it has a time resolution of < 50 ps, and can record times delays over 200 ns in
length. This circuit costs approximately $500 CDN to build and the designs are freely
available at Alan Stummer’s website [Stummerb]. In comparison, similar commercial
circuits can cost 20-40 times more. An artificial delay of 26.2 ns is introduced into one
detector to shift the $\tau = 0$ events. This is because it is difficult for a circuit to accurately
measure two simultaneous events with good accuracy. A normalized plot of the number
of detection events as a function of $\tau$ provides a direct measure of the $g^{(2)}(\tau)$ of the
source.

The SPCMs have a significant probability of after-pulsing (0.5%). After-pulsing can
occur due to feedback within the detector. When this happens, some time after the de-
tector registers a detection event it subsequently registers a second false event [Zhao 03].
After-pulsing can also arise when the detector emits a photon after it fires. This photon
can be coupled back into the optical system and trigger the other SPCM. When using
multimode fibres, there is a significant probability that the after-pulse will register a co-
incidence. This leads to a large peak for a specific time delay (dependent on the optical
path length and the time it takes after a detection event for a detector to after-pulse).
Using single-mode fibre eliminates this problem as the probability that the emission from
the SPCM can couple into the fibre is negligible.

We found one very bright dot in a 2 $\mu$m pillar on the sample provided to us by
our collaborators at NIST with the spectrum shown in figure 3.3. The $g^{(2)}(\tau)$ for this
quantum dot under both CW and pulsed above band excitation was measured. It should
be noted that the data from the CW and pulsed pumping were taken during different
cooling cycles of the dot. The quantum dot was first brought into resonance with the
cavity through temperature tuning; the resonance temperature for this dot was found to
be $25.5 \pm 0.2$ K. When in resonance, the number of counts on the spectrometer increased
by a factor of three. The output slit of the spectrometer was set to 200 \( \mu \)m and the coupling into multi-mode fibre optimized.

For reference, figure 3.6 a) shows an unnormalized histogram of the detection events from our Ti:Sapphire laser operating in CW mode at 810 nm. The histogram is flat for all values of \( \tau \) as expected for a Poissonian source. The \( g^{(2)}(\tau) \) data from the quantum dot pumped with the CW laser is shown in figure 3.6 b). For this run, data was acquired for 45 minutes. In multimode fibre, both detectors recorded in excess of 65,000 counts/s—the maximum data rate our home-built timing circuit could handle. A clear dip is seen for time delay of 26.2 ns, corresponding to the zero time delay position. A fit to the data using the model for the single atom emitter gives a \( g^{(2)}(0) = 0.16 \pm 0.04 \). In comparison, an ideal single-photon source would have a \( g^{(2)}(0) = 0.07 \pm 0.01 \) in the same detection system (from equation 3.5 for detectors having a jitter of 400 ps).

The width of the dip is consistent with a \( T_R = 1.7 \pm 0.1 \)ns. While the decay rate of this dot was not measured, other dots in similar samples were found to have lifetimes of \( \sim 0.4-1 \) ns\(^2\). This broader dip width is consistent with other experimental observations [Santori 02a]. The cause for wider dip is unknown, but is most likely a combination of the above-band excitation of carriers, the pump power used, the presence of excess charges in the dot, and the emission from multiple levels within the dot. To thoroughly investigate the physics behind the wider dip more sophisticated spectroscopic tools are needed.

We are primarily interested in using the quantum dots as single-photon sources. The bulk of the spectroscopy work needed to study the dynamics of the dots is being carried out at the same time by our collaborators. Our collaborators have the tools necessary to conduct more in depth studies on the samples with the aim of improving their manufacturing processes while we are well positioned to use the dots to study foundational issues in quantum mechanics and quantum information. Consequently, we have not spent much time performing spectroscopic studies.

\(^2\)These measurements were made by our collaborators at NIST.
Figure 3.6: Results from a Hanbury Brown-Twiss Interferometer for a CW laser (a), a quantum dot under CW above-band excitation (b), a mode-locked Ti:Sapphire laser (c), and a quantum dot pumped with the mode-locked laser (d). A time delay of 26.2 ns corresponds to $\tau = 0$. For the CW data in a), the measured coincidences are independent of the time delay as expected for a Poissonian source. In b) a sharp dip is observed, indicating antibunching. The solid line corresponds to a fit to the data and gives a $g^{(2)}(0) = 0.16 \pm 0.04$. The dashed line represents the maximum achievable $g^{(2)}(\tau)$ for an ideal source in our detection system. In c) the pulsed nature of the Ti:Sapphire is seen. Pulses occur every 13.1 ns, corresponding the repetition rate of the laser. Each peak is fit by a Gaussian distribution. In d) the peaked character of the pulsed excitation is observed, with a peak noticeably absent at $\tau = 0$. There is an additional “background” component with a dip that most likely due to long-lived carriers in the wetting layer. A fit to this data gives a $g^{(2)}(0) = 0.16 \pm 0.04$. 
The $g^{(2)}(\tau)$ of the Ti:Sapphire in pulsed mode is shown in figure 3.6 c). For this run, data was taken for 5 min. The pulses from the Ti:Sapphire are approximately 100 fs in length, providing precise timing information. On the histogram peaks appear every 13.1 ns—corresponding to the repetition rate of the laser. The width of the peaks confirms the 400 ps jitter of the detectors. The height of the peaks are the same, as expected from a pulsed classical source. Figure 3.6 d) shows the $g^{(2)}(\tau)$ for the quantum dot pumped with this pulsed source. Data in this run was acquired for 25 minutes. Again peaks are seen every 13.1 ns, except for at a delay of 26.2 ns ($\tau = 0$) where a dip is observed. The histogram appears to be a combination of a CW and pulsed spectrum with a combination of peaks and a constant “background” level with a dip. Ideally there should just be peaks with zero background and a missing peak at $\tau = 0$. At first it was thought that the Ti:Sapphire was not modelocked properly and suffering from CW breakthrough. It was confirmed that this was not the case by looking at the spectrum of the Ti:Sapph and repeating the $g^{(2)}(\tau)$ measurements of the laser by itself. Repeated pulsed measurements of the $g^{(2)}(\tau)$ of the dot all showed the same behaviour. The most likely explanation for this behaviour is that the carriers generated in the wetting layer persist for a long time. The peaks indicate that a dot is most likely to emit within a few ns after being excited by a pulse, but the large “background” indicates a significant number of carriers remain available for capture at longer time scales. The broad bandwidth of the pump may be responsible for this longer relaxation time of the carriers. In most other quantum dot experiments picosecond pulsed sources are used, and this behaviour is not observed [Santori 04, Mirin 04, Santori 02a]. One way to test this would be to purchase a picosecond kit for our laser or use a bandpass spectral filter to generate narrowband pulses.

Ordinarily, the $g^{(2)}(0)$ for a pulsed quantum dot is determined by comparing the area of the peak at $\tau = 0$ with the area of the adjacent peaks. A better value for $g^{(2)}(0)$ is often measured under pulsed excitation compared to CW excitation as the finite detector
response is no longer a factor. In our case this area method is not feasible due to the dot emission at time scales much longer than the duration of the excitation pulse. Using a Lorentzian fit for the peaks and the dips, a $g^{(2)}(0) = 0.16 \pm 0.04$ is found. This value is determined by comparing the average height of the peaks to the minimum value of the dip.

In the experiments we carried out that used the quantum dot as a source of single photons the dot was pumped by a CW laser. This leads to a higher count rate as the dot is excited more frequently. For these experiments it is the CW pumped $g^{(2)}(\tau)$ data, like that shown in Figure 3.6 b), that is used to characterize the single-photon nature of the source.

For both the CW and the pulsed cases, the measured $g^{(2)}(0)$ provides strong evidence that our quantum dot is a good single-photon emitter. The remaining small probability of detecting more than one photon at $\tau = 0$ can be attributed to contamination from nearby lines. As seen in the spectrum in figure 3.3, the cavity has a broad emission spectrum. Narrowing the output slits to provide better spectral filtering from the spectrometer and reducing the pump power should lead to better $g^{(2)}(0)$ values, but at the expense of significantly reducing our count rates.

### 3.5 Indistinguishability

The ability to fabricate high quality waveguides and cavities with dots inside has already enabled many cavity-QED experiments to be performed on a chip [Yoshie 04, Flagg 09, Englund 8, Santori 02c, Englund 07, Faraon 08]. An important requirement for further exploiting these sources for quantum information applications is that the photons emitted from the quantum dots be indistinguishable. For example, one could imagine a device that uses single photons emitted simultaneously from different quantum dots to engineer quantum states or implement quantum circuits [Lindner 09]. Currently, there are too
many inconsistencies in the manufacturing techniques to practically allow different dots to emit indistinguishable photons. The next best alternative is to interfere successive photons emitted by a single-dot with one another. This can be done probabilistically using beamsplitters and delay lines, but requires that successive photons be indistinguishable from one another.

For the photons to be indistinguishable, the emission from the quantum dot should be transform limited with $\Delta w \Delta t = 1$. For this requirement to be met $\frac{2T_{sp}}{T_c} = 1$ where $T_{sp}$ is the spontaneous emission lifetime and $T_c$ is the coherence time of the dot emission\(^3\). If $T_{sp} > T_c/2$, it implies that the emission is multi-mode and there is distinguishing information present.

There are several factors that can hurt the indistinguishability of the photons in a quantum dot. The first is the relaxation time of carriers within the dot. It can take $\sim 100$ ps for carriers to be captured and then relax into the ground state using above band excitation. This can be reduced to 10 ps for resonant excitation schemes [Santori 02c]. Typically pulsed excitation is used in schemes where successive photons from a single quantum dot are interfered due to the accurate timing information provided by the pump. A possible point of concern in our dots is the strong “CW” like behaviour in the $g^{(2)}(\tau)$ data as well as the width of the peaks. If the dots rapidly capture an electron-hole pair and then emit this should not be a problem. If, however, the dots take much longer to capture an electron-hole pair this could introduce a significant amount of jitter. Based on the lifetimes for dots in our sample, it is most likely that the capture process is rapid and that the dots are able to be excited multiple times by a single excitation pulse. A second mechanism that could lead to inhomogeneous broadening is the simultaneous capture of multiple excitons. A quantum dot cannot emit on the X line until all but one exciton remain. If many carriers are captured, it may take some

\(^3\)For a Lorentzian line shape, the bandwidth of the photons is related to the coherence time $\Delta w = \frac{2}{T_c}$ [Santori 04, Loudon 73]. When there is no broadening mechanisms or dephasing, $\frac{2}{T_c} = \frac{1}{T_{sp}}$. 
time before all of the required recombination events have taken place. Both of these sources of jitter can be reduced through the use of resonant excitation. Another source of inhomogeneous broadening stems from spectral diffusion. Excess charges surrounding the dot can lead to decoherence. It has been speculated that this process occurs on a longer scale [Santori 02a], so photons emitted in short time intervals may not be as adversely affected by this. A benefit to using a cavity is that it is possible to decrease the spontaneous emission lifetime of the quantum dots using the Purcell effect to better match the coherence time.

While we have not performed a direct measurement of the indistinguishability of the photons emitted by our quantum dot, our collaborators have performed some preliminary experiments using a HOM interferometer. So far they have not found any evidence that the photons are indistinguishable. The coherence time of the photons emitted from the dots is typically $\sim 100 - 200$ ps and the spontaneous emission lifetime ranges from $400$ ps to $1$ ns. In the literature, the best report HOM dip visibility for photons emitted from a single quantum dot is 0.81 [Santori 02c]. There is still work to be done on the manufacturing side to reduce the amount of distinguishing information present in the dots.

### 3.6 Conclusions

While quantum dots are not ready to replace SPDC as a multi-photon source, they are good single-photon sources. Unlike SPDC, the single photons from a quantum dot do not need to be heralded and, if the collection technology improves, can be generated on-demand. Already our quantum dot source has proved useful in experiments that require single-photons, and with improvements in dot technology, may allow multi-photon experiments to be carried out. The setup is also easily adaptable to other novel photon sources such as colloidal quantum dots, and diamond NV centres, or any other sample
that requires a cryostat. This flexibility, along with the spectrometer and TEC cooled
CCD open a range of experimental avenues to be explored in the future.
Chapter 4

Squeezing and Over-Squeezing Triphotons

4.1 Introduction

Physical measurements have a fundamental limit placed on their accuracy by quantum mechanics and the Heisenberg uncertainty principle. Using squeezing techniques, it is possible to reduce the uncertainty of a desired property below the standard quantum limit at the expense of increasing that of the complementary one. As pointed out in section 1.4.2 spin squeezing plays a fundamental role in enhancing the precision of interferometric measurements, and has gained interest as it is simple to generate the required nonclassical states [Geremia 04, Hald 99, Bowen 02, Heersink 03, Marquardt 07, Klose 01, Smith 06, Chaudhury 07, Ghose 08]. Squeezing is already being applied to enhance the precision of gravity-wave detectors [Goda 08], and may play a critical role in other high precision applications such as atomic clocks [Ye 08] and optical communications [Furusawa 98]. While impressive gains in squeezing have been made recently, spin-squeezed systems are still many orders of magnitude away from reaching the Heisenberg limit in uncertainty.

In this chapter I present an experiment where we produce and study a family of states
that bridge the gap between Heisenberg-limited interferometry and spin-squeezing. These spin-squeezed states are constructed by overlapping three indistinguishable photons in an optical fibre and manipulating their polarization (spin), resulting in the formation of a “triphoton” particle. The symmetry properties of polarization imply that the triphoton states we measure are most naturally represented by quasi-probability distributions on the surface of a sphere as shown in section 1.5. We observe for the first time the effect of this spherical topology of polarization, which imposes a limit to how much squeezing can occur and can lead to the quasi-probability distributions wrapping around the sphere, a phenomenon we call “over-squeezing”.

The experiment was carried out with Robert Adamson, another graduate student in our lab. I designed, built, and carried out the experimental measurements while Rob contributed to some of the data analysis and theory. The resulting work was published in Nature in 2009 [Shalm 09].

4.2 The Triphoton Family

The three photons we study are indistinguishable in every degree of freedom (colour, spatial mode, temporal overlap, etc...) save for their polarization. The state of the three photons can be written as

\[ |\Psi\rangle = c_3 |3,0\rangle_{H,V} + c_2 |2,1\rangle_{H,V} + c_1 |1,2\rangle_{H,V} + c_0 |0,3\rangle_{H,V}, \]  

(4.1)

where \(|m,n\rangle_{H,V}\) represents \(m\) and \(n\) photons in the orthogonal horizontal \((H)\) and vertical \((V)\) polarization modes respectively. The photons no longer have an individual identity of their own - there is no way to tell the photons apart from one another. All that can be said about the state is whether there is a superposition of three, two, one, or zero horizontally polarized photons. Using the Schwinger representation, discussed in section 1.4, the polarization of each photon can be treated as spin-1/2 particle. A collection of three indistinguishable spin-1/2 particles form a composite spin-3/2 particle. This
composite particle is what we term a triphoton. The most famous triphoton is the N00N state

\[ |\Psi\rangle = \frac{1}{\sqrt{2}}(|3,0\rangle_{H,V} + |0,3\rangle_{H,V}) \]  

(4.2)

which is able to reach the Heisenberg limit in phase sensitivity as well as display super-resolution.

We can rewrite the state of the triphotons in equation 4.1 as

\[ |\Psi\rangle = \left( \frac{c_3}{\sqrt{3!}} (\hat{a}_H^{\dagger})^3 + \frac{c_2}{\sqrt{2!}} (\hat{a}_H^{\dagger})^2 \hat{a}_V^{\dagger} + \frac{c_1}{\sqrt{2!}} \hat{a}_H^{\dagger} (\hat{a}_V^{\dagger})^2 + \frac{c_0}{\sqrt{3!}} (\hat{a}_V^{\dagger})^3 \right) |0\rangle, \]  

(4.3)

where \( \hat{a}_{H,V}^{\dagger} \) represent the creation operator for a single \( H \) or \( V \) photon. As first discussed in [Lee 02a, Fiurasek 02], it is possible to factor this state into a product of creation operators:

\[ |\psi\rangle = (A \hat{a}_H^{\dagger} + B \hat{a}_V^{\dagger})(C \hat{a}_H^{\dagger} + D \hat{a}_V^{\dagger})(E \hat{a}_H^{\dagger} + F \hat{a}_V^{\dagger}) |0\rangle \]

\[ = \hat{a}_1^{\dagger} \hat{a}_2^{\dagger} \hat{a}_3^{\dagger} |0\rangle, \]  

(4.4)

where \( A, B, C, D, E, \) and \( F \) are complex coefficients and \( \hat{a}_1^{\dagger}, \hat{a}_2^{\dagger}, \) and \( \hat{a}_3^{\dagger} \) are the creation operators defined as \( (A \hat{a}_H^{\dagger} + B \hat{a}_V^{\dagger}), \) \( (C \hat{a}_H^{\dagger} + D \hat{a}_V^{\dagger}), \) \( (E \hat{a}_H^{\dagger} + F \hat{a}_V^{\dagger}) \) respectively. By taking three photons, each in a polarization state \( \hat{a}_1^{\dagger}, \hat{a}_2^{\dagger}, \) and \( \hat{a}_3^{\dagger} \), and placing them into the same mode it is possible to create any desired triphoton state.

Consider the special case of a triphoton N00N state consisting of a superposition of three right-handed circularly polarized (R) and three left-handed circularly polarized photons (L):

\[ |3 :: 0\rangle = \frac{1}{\sqrt{2}}(|3,0\rangle_{L,R} + |0,3\rangle_{L,R} \]

\[ = \frac{1}{2\sqrt{3}}((\hat{a}_L^{\dagger})^3 + (\hat{a}_R^{\dagger})^3) |0\rangle \]

\[ = \frac{1}{2\sqrt{3}}((\hat{a}_L^{\dagger} + \hat{a}_R^{\dagger})(\hat{a}_L^{\dagger} + e^{i\frac{2\pi}{3}} \hat{a}_R^{\dagger})(\hat{a}_L^{\dagger} + e^{i\frac{4\pi}{3}} \hat{a}_R^{\dagger}) |0\rangle \]

\[ = \hat{a}_0 \hat{a}_6 \hat{a}_{12} |0\rangle. \]  

(4.5)
Figure 4.1: A triphoton N00N state composed of an equal superposition of three right-handed and three left-handed circularly polarized photons is equivalent to three linearly polarized photons separated by 60°. It is impossible to combine three photons with non-orthogonal polarization states into the same mode with 100% efficiency.

The three terms in parenthesis create three different linear polarizations each separated by 60°. If three linearly polarized photons separated by 60° can be placed in the same mode, in the circular basis this will look like a N00N state as shown in Figure 4.1.

4.3 Experimentally Producing Triphotons

To create an arbitrary triphoton, equation 4.4 implies that three photons with the appropriate polarizations must be placed into the same spatial, temporal, and spectral mode. A straightforward way to do this is to first prepare three identical photons with each photon polarized in the desired manner. Using a series of 50:50 beamsplitters the three photons can be “mashed” together. A triphoton is produced whenever all three photons leave the network of beamsplitters in the same mode. In general, this “mode-mashing” cannot succeed with 100% efficiency if the polarizations of the photons are non-orthogonal; combining non-orthogonal states is a non-unitary operation. This loss means that the three photons will not always end up in the same mode. Instead we use single-photon detectors to study only the cases when a triphoton has successfully been created, a process known as post-selection. Post-selection can be thought of as a type of induced nonlinearity [Knill 01], and plays a pivotal role in most quantum information
experiments. A typical nonlinear process generates a weaker signal that must be filtered in some manner from the fundamental driving fields. In the same way, post-selection is used to filter out a desired signal from unwanted background. In our case the signal is three photons ending up in the same mode while the background is all other combinations of photons leaving the beamsplitters.

4.3.1 Efficient Mode-mashing

To create a triphoton in our experiment, two photons from SPDC are mode-mashed with a third photon from an attenuated laser. Unwanted contamination from higher order terms in the laser and the SPDC crystal can cause background three-photon events that reduce the quality of our triphoton states. A true single-photon source would minimize this background; unfortunately the photons from our quantum dots are not indistinguishable and cannot be used in this application. Because of the three-photon background terms it is important that the mode-mashing be as efficient as possible. For the triphoton states we are interested in, beamsplitters are not the most efficient method to combine the photons. Instead a variable partial polarizer is used to more efficiently mode-mash the photons. The family of states we study using this variable partial polarizer approach is

\[
|\Psi\rangle = c_3 |3, 0\rangle_{H,V} + c_1 |2, 1\rangle_{H,V} + c_1 |1, 2\rangle_{H,V} + c_3 |0, 3\rangle_{H,V}.
\] (4.6)

A polarizer is a device that transmits a specific polarization while blocking the orthogonal polarization. A partial polarizer is an imperfect polarizer: the orthogonal polarization state is only partially attenuated instead of being blocked. Using a partial polarizer (PP), it is possible to manipulate the polarization state of the photons passing through.

To create these triphoton, we start by combining an $H$ and $V$ photon together on a polarizing beamsplitter. As these two photons are orthogonally polarized, it is possible to place them in the same mode with 100% efficiency. Using a half waveplate the two
photons are rotated so that one photon is polarized at $45^\circ$ while the other is polarized at $-45^\circ$

$$\hat{a}^\dagger_H \hat{a}^\dagger_V \ket{0} \rightarrow \hat{a}^\dagger_{45^\circ} \hat{a}^\dagger_{-45^\circ} \ket{0}$$  

$$= \frac{1}{2} \left( \hat{a}^\dagger_H - \hat{a}^\dagger_V \right) \ket{0}$$  

These two photons are then sent through a variable partial polarizer (VPP) that is capable of attenuating either the horizontal or vertical components of the polarization state. The effect of the variable partial polarizer can be modelled using a beamsplitter with a variable transmittance that acts on the H and V modes independently. The beamsplitter takes a H (V) photon and transmits it into the target mode 2 with a transmission probability of $|T_H(V)|^2$ and reflects it into the loss mode 3 with a probability $|R_H(V)|^2$:

$$\hat{a}^\dagger_H \rightarrow T_H \hat{a}^\dagger_H + R_H \hat{a}^\dagger_H, \quad |T_H|^2 + |R_H|^2 = 1,$$

$$\hat{a}^\dagger_V \rightarrow T_V \hat{a}^\dagger_V + R_V \hat{a}^\dagger_V, \quad |T_V|^2 + |R_V|^2 = 1.$$  

After passing through the VPP, the two photons in equation 4.8 are transformed to

$$\hat{a}^\dagger_{45^\circ} \hat{a}^\dagger_{-45^\circ} \ket{0} \rightarrow \frac{1}{2} \left[ T_H^2 \hat{a}^\dagger_{H2} - T_V^2 \hat{a}^\dagger_{V2} \right] \ket{0}$$  

$$+ \frac{1}{2} \left( 2T_H R_H \hat{a}^\dagger_{H2} \hat{a}^\dagger_{H3} + R_H^2 \hat{a}^\dagger_{H3} \hat{a}^\dagger_{H2} - 2T_V R_V \hat{a}^\dagger_{V2} \hat{a}^\dagger_{V3} - R_V^2 \hat{a}^\dagger_{V3} \hat{a}^\dagger_{V2} \right) \ket{0}$$  

The probability of both photons being transmitted is given as $\frac{1}{2}(|T_H|^4 + |T_V|^4)$. Post-
selecting on the cases where both photons are transmitted and renormalizing yields

\[ \hat{a}_{45^\circ} \hat{a}_{-45^\circ} |0\rangle \rightarrow \frac{1}{\sqrt{2} \sqrt{T_H^4 + T_V^4}} \left[ T_H^2 \hat{a}_H^\dagger - T_V^2 \hat{a}_V^\dagger \right] |0\rangle \]

\[ = \frac{1}{\sqrt{2} \sqrt{T_H^4 + T_V^4}} (T_H \hat{a}_H^\dagger - T_V \hat{a}_V^\dagger)(T_H \hat{a}_H^\dagger + T_V \hat{a}_V^\dagger) |0\rangle \]

\[ = \frac{1}{\sqrt{2} \sqrt{1 + T^4}} (\hat{a}_H^\dagger - T \hat{a}_V^\dagger)(\hat{a}_H^\dagger + T \hat{a}_V^\dagger) |0\rangle \]

\[ = \frac{1}{\sqrt{2}(1 + T^2) \sqrt{1 + T^4}} (\cos \theta \hat{a}_H^\dagger - \sin \theta \hat{a}_V^\dagger)(\cos \theta \hat{a}_H^\dagger + \sin \theta \hat{a}_V^\dagger) |0\rangle \]

\[ = \frac{1}{\sqrt{2}(1 + T^2) \sqrt{1 + T^4}} \hat{a}_{-\theta}^\dagger \hat{a}_\theta^\dagger |0\rangle , \quad (4.11) \]

where \( T = T_V/T_H \) and \( \theta = \arccos(1/\sqrt{T^2 + 1}) \) is the angle that the linearly polarized photon makes with respect to the horizontally polarized photon. The effect of the partial polarizer is to rotate the polarization of the two photons into non-orthogonal polarization states. When \( T = 0 \) \((T_V = 0 \text{ and } T_H = 1)\), both photons emerge \( H \) polarized with a probability of 1/2. When \( T = \infty \) \((T_V = 1 \text{ and } T_H = 0)\), both photons emerge \( V \) polarized with a probability of 1/2. When \( T = 1 \) \((T_V = 1 \text{ and } T_H = 1)\), one photon emerges polarized \( 45^\circ \) while the other emerges polarized \(-45^\circ\). When \( T = \sqrt{3} \) \((T_H = 1/\sqrt{3} \text{ and } T_V = 1)\), one photon emerges at \( 60^\circ \) while the other emerges at \(-60^\circ\) with a probability of 5/9\(^\dagger\)

The final step is to add the third photon with the appropriate polarization in order to create a triphoton. There are a number of ways to accomplish this that depend on the type of VPP used and the characteristics of the photon source. If the efficiency of mode-mashing the third photon is \(|\alpha'|^2\), the probability of creating the desired triphoton state is \(|\alpha'|^2 \frac{1}{2}(|T_H|^4 + |T_V|^4)\). In our case, the third photon is the attenuated laser source. The efficiency \(|\alpha'|^2\) can be treated as part of the attenuation process; this allows us to only consider the efficiency of the VPP in the mode-mashing.

The action of the VPP is to squeeze the polarization state of the triphoton. It is convenient to define a squeezing operator \( A e^{\hat{S}_1 \theta} \) that represents the action of our VPP.

\(^\dagger\)Using beamsplitters to mode-mash would only succeed in producing this state 7/16 of the time.
on our input state $\hat{a}^\dagger_{+45} \hat{a}^\dagger_{-45} \hat{a}^\dagger_H |0\rangle$. The VPP can also be described by the operator

$$
\hat{V} = T^3_H \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & T & 0 & 0 \\
0 & 0 & T^2 & 0 \\
0 & 0 & 0 & T^3
\end{pmatrix}.
$$

Comparing $\hat{V}$ to $A e^{S_1 \theta}$ we find that $\theta = -\frac{\ln(T)}{2}$. After post-selection, the triphoton that is produced using the VPP is given by

$$
|\zeta\rangle = \eta e^{-S_1 \frac{\ln(T)}{2}} \hat{a}^\dagger_{+45} \hat{a}^\dagger_{-45} \hat{a}^\dagger_H |0\rangle,
$$

where $\eta$ is a normalization constant and $T = T_V/T_H$ is the squeezing factor. Here $e^{-S_1 \frac{\ln(T)}{2}}$ is the squeezing operator; it is not unitary as it models the action of the lossy VPP.

### 4.3.2 Experimental Setup

In the first step of the experiment, a pulsed 40 Mhz Ti:Sapphire laser operating at 810 nm is frequency doubled using a 0.5mm thick Beta-Barium Borate crystal (BBO) to produce 50 mW of pulsed light at 405 nm as shown in figure 4.2. These pulses are focused into a 0.5 mm BBO crystal phase-matched for Type-II spontaneous parametric downconversion with beam-like emission [Takeuchi 01], to generate a pair of photons at 810 nm. These photons are combined into the same mode using a polarizing-beamsplitter and then sent through a half waveplate at 22.5$^\circ$ to produce the state $\hat{a}^\dagger_{+45} \hat{a}^\dagger_{-45} |0\rangle = \frac{1}{2}(\hat{a}^\dagger_H + \hat{a}^\dagger_{V}) |0\rangle$.

Next, the downconverted pair of photons is combined with a horizontally polarized local oscillator (LO) photon, taken from the original Ti:Sapphire beam, on a highly transmissive beamsplitter to produce the unnormalized state $\hat{a}^\dagger_{+45} \hat{a}^\dagger_{-45} \hat{a}^\dagger_H |0\rangle = (\hat{a}^\dagger_H + \hat{a}^\dagger_{V}) \hat{a}^\dagger_H |0\rangle$.

This state is then sent to a VPP where either the horizontal or vertical polarization component can be attenuated and the polarization state of the three photons controlled.
Figure 4.2: Overview of the experiment. A pair of SPDC photons from Type II spontaneous parametric downconversion (SPDC) are placed into the same mode with a polarizing beamsplitter (PBS). A half waveplate (HWP) rotates the photons into the diagonal basis and a third photon from a local oscillator (LO) is combined into the same mode using a laser window. A variable partial polarizer (VPP) is used to control the polarization of the triphotons by attenuating either the horizontal $T_H$ or vertical $T_V$ polarization components. Varying $T_H$ and $T_V$ causes the angle $\theta$ that the two SPDC photons make with the $H$ polarized LO photon to change, leading to the creation of different triphoton states. The triphotons are coupled into a single mode fibre and spectrally filtered (IF). To characterize the states, quantum state tomography is carried out by using a polarization analyzer consisting of three waveplates and a PBS, and threefold coincidence counts are collected using coincidence circuitry and single-photon counting modules (SPCM).
The VPP is formed from a balanced polarization interferometer. For a polarization interferometer to successfully serve as a VPP, it must be stable for long periods of time. This can be achieved by minimizing the path lengths in the interferometer, combining as many elements as possible together in a monolithic design, and stably mounting the components. Calcite beam displacers offer a convenient way to build interferometers that satisfy all three of these requirements. A calcite beam displacer (BD) is a slab of birefringent material that uses walk-off to spatially separate two polarization components (see section 2.3.1). By placing two calcite beam displacers in a row with a half waveplate sandwiched between them it is possible to create a stable balanced polarization interferometer as shown in figure 4.3 b). In the first BD the ordinary component of polarization takes the bottom path while the extraordinary polarization walks off to the top path. The half waveplate “flips” the polarization of both beams. In the second BD, light in the top path is now transmitted straight through while light in the bottom path walks up. If the beam displacers are the same length, both components of polarization are coherently recombined and exit in the same mode. If need be a second half waveplate can be used to “flip” the polarization state back to compensate for the action of the sandwiched waveplate. For the VPP considered here, \( V \) and \( H \) constitute the ordinary and extraordinary polarization components respectively. To complete the VPP, a small beam block is placed between the two beam displacers. Moving the beam block alters the attenuation of the \( H \) or \( V \) paths, changing the squeezing parameter \( T \). Using this configuration it is possible to create the triphotons states in equation 4.13 by changing a single experimental parameter–the position of the beam block. A total of eleven different triphoton states are prepared and measured by varying the transmissivity \( T \) of the variable partial polarizer from \( T = 0 \) (a spin-coherent state) to \( T = 1.7 \) (a N00N state).

Following the partial polarizer, a quarter wave-plate is used to rotate the state from the circular to linear polarization basis. The three photons then pass through a 12 nm spectral filter centred at 810 nm and are coupled into a single-mode fibre. A polarization
Chapter 4. Squeezing and Over-Squeezing Triphotons

4.3.3 Removing Distinguishing Information

In order to form a triphoton all three photons must be indistinguishable from one another. Two of the photons come from SPDC while the third is from the original Ti:Sapphire beam. The SPDC photons and the LO photons will have similar properties as they are related: the photons from the Ti:Sapphire laser are the grandparents of the daughter photons created during SPDC. Despite these family ties, the photons from the laser and SPDC will contain differences. The nonlinear processes of SHG and SPDC are dispersive, altering the spectral characteristics of the SPDC relative to the LO. An interference filter is used to remove the spectral difference existing between the photons. Coupling the three photons into a single-mode fibre erases any transverse path information that could be used to label the photons.

It is important that all three photons are placed in the same temporal mode: they must exit the VPP at the same time. Since the photons have a coherence length of approximately 30 \( \mu m \), each path must be balanced to within this length. To ensure that the two photons from SPDC are in the same temporal mode, a translation stage in the \( V \) arm before the first beamsplitter is used to adjust the arrival time of the \( V \) polarized photon. If the photons are indistinguishable, after passing through the half waveplate they will ideally be in the state \((\hat{a}_H^{12} - \hat{a}_V^{12})\hat{a}_H^\dagger |0\rangle\). When this state is sent to a polarizing beamsplitter, either both photons will be horizontally polarized or vertically polarized. If there is any distinguishing information that can be used to tell the photons apart there will be some probability the photons will exit from opposite ports of the polarizing beamsplitter as discussed in section 2.4. Initially, the VPP and all other waveplates are
Figure 4.3: a) A polarization interferometer. Light hits a polarizing beamsplitter (PBS) that transmits $H$ polarized light and reflects $V$ polarized light. A second PBS coherently recombines the two polarizations. By varying the path length difference of the two arms, it is possible to rotate the state $|H\rangle + |V\rangle$ by an angle $\phi$ to $|H\rangle + e^{i\phi} |V\rangle$. b) A variable partial polarizer. Using two calcite beam displacers (BD), a stable and compact polarization interferometer can be created. The first BD walks $H$ polarized light up while transmitting $V$ polarized light. A half waveplate (HWP) flips the polarization state of each path. Now the top path is $V$ polarized and travels straight through the second BD while the bottom path is $H$ polarized and walks up. A thin beam block can be inserted into the two paths. As the beam block moves across the bottom (top) path it attenuates the $H$ ($V$) component of the polarization, changing the value of $T$ and allowing different triphoton states to be produced.
Table 4.1: The Hong-Ou-Mandel dip data between the two photons from SPDC. The two orthogonally polarized photons are combined together on a polarizing beamsplitter, sent through a half waveplate at $22.5^\circ$, coupled into a single-mode fibre, and then sent to a fibre polarizing beamsplitter. At each of the output ports $A$ and $B$ of the fibre polarizing beamsplitter a single-photon detector is placed and coincidence counts measured. Using a translation stage, the $V$ path from the SPDC is adjusted until the dip is found. Each datapoint is collected for 20 s. The data is then corrected for accidental coincidences. A HOM visibility of 97% is observed. As stated in the text, the repetition rate of the laser is 40 MHz.

removed from the setup and the photons coupled into the single-mode fibre and sent to the in-fibre polarizing beam splitter. A single-photon detector is placed at each output port of the fibre PBS. The translation stage is scanned by hand until a Hong-Ou-Mandel dip is observed. Table 4.3.3 shows the coincidence counts obtained when sitting at the dip minimum and when sitting outside the dip. After subtracting accidental coincidences, a HOM visibility of 97% is observed.

Once the HOM dip has been found the VPP is inserted into the setup. To ensure that the paths in polarization interferometer are balanced, the HOM is measured again. Now the half waveplate before the VPP is set to $0^\circ$ and a second half waveplate at $45^\circ$ is inserted after the VPP. The BD are mounted on gimbal mounts from NewPort allowing the calcite beam displacers to be precisely tilted. By tilting one BD small amounts relative to the other, it is possible to change the relative path length between the $H$ and $V$ arms. By adjusting the paths in this way the HOM dip is again found, and the polarization interferometer in the VPP balanced to within a coherence length.
To verify that the polarization interferometer is balanced, a half waveplate and quarter waveplate are inserted before the VPP enabling the preparation of the polarization states $H, V, H + V, H - V, H + iV$, and $H - iV$. These states are sent through the VPP and polarimetry is carried out. Minor adjustments to the path length of the interferometer are made until the VPP is balanced so as not to induce a polarization rotation. Once adjusted the interferometer is stable on the order of a week - more than enough time to collect the necessary measurements.

To place the LO photon in the same mode as the SPDC photons the classical HOM effect discussed in section 2.4 is used. The $V$ arm of the SPDC emission is blocked and the photons from the $H$ arm are combined with $V$ polarized photons from the LO. The photons then pass through the VPP and a half waveplate at $22.5^\circ$ and are sent to the fibre polarizing beamsplitter where coincidences are measured. Approximately 54,000 singles/s are collected from the $H$ channel of the SPDC while the LO is attenuated to approximately 70,000 singles/s. This is done to ensure a higher count rate. The statistics of the light from the single SPDC arm are Poisson-like, hence the maximum expected HOM visibility is the classical limit of 50% \cite{Ou91}. To find the HOM dip, an automated Melles-Griot Nanomover translation stage is used to scan the LO over approximately two inches of path length in 10 $\mu$m steps, counting the accidental coincidences between the two sources for 8 s at each step. Figure 4.4 shows the results of such a scan near the dip region. The Melles-Griot stage is then set to the centre of the dip, ensuring that the LO and SPDC photons end up in the same temporal mode. A clear dip can be seen with a visibility of $20 \pm 2\%$. This is lower than the maximum classical dip of 50$, indicating that distinguishing information still remains. Tighter spectral filtering would most likely remove most of the remaining distinguishability.
Figure 4.4: To balance the path lengths between the SPDC and the LO paths, a two-photon interference effect is looked for between the LO photons and photons from $H$ arm of the SPDC. Using an automated stage, the LO path is scanned in 10 $\mu$m increments and accidental coincidences between the two arms are counted for 8 s per step. When the path lengths are balanced, a clear Hong-Ou-Mandel dip is observed.
4.3.4 Experimental Background Contributions

The desired triphoton state is composed of a single SPDC pair and a single LO photon placed into the same mode. All other three-photon terms are background events that add incoherently to our desired state and reduce its fidelity, purity and visibility. To account for these background terms, the emission statistics of the SPDC and LO sources must be included in the analysis of the triphoton states. The LO emission is described by a coherent state $|\alpha\rangle$ defined in equation 1.17 with mean photon number $|\alpha|^2$. For the SPDC, the photon pair emission statistics are given in equation 2.3. In the low-intensity regime relevant to the present experiment, the probability per pulse of obtaining a single pair of SPDC photons is $|\epsilon|^2$ while the probability of obtaining a single LO photon is $|\alpha|^2$. To minimize background events, the pair generation rate and the local oscillator rate are carefully balanced to ensure that the majority of three-photon events detected originate from a single SPDC pair and a single LO photon.

Any source of loss in the system will lead to a decrease in the measured raw coupling efficiency. This includes mode-mismatch between the photons and the single-mode fibre, detector losses, losses due to the variable partial polarizer, and losses due to the spectral filters. The probability per pulse of collecting and detecting a SPDC pair into single-mode fibre is given as $|\epsilon|^2\eta_1\eta_2$, where $\eta_1$ and $\eta_2$ are the raw efficiencies for coupling and detecting either the first or second photons from the SPDC source. To accurately determine the effect of the background contributions it is necessary to measure the raw coupling efficiencies $\eta_1$ and $\eta_2$ for every triphoton state prepared. It is important to note that the coupling efficiency of the LO into the fibre is used to set the attenuation level, and therefore is already included in the $|\alpha|^2$ term. Once the values of $|\alpha|^2$, $|\epsilon|^2$, $\eta_1$, and $\eta_2$ have been experimentally measured, it is possible to calculate the contributions of each of the significant three-photon terms for the desired triphoton state as shown in Table 4.3.4.

The different combinations of photons from the LO and the SPDC form different
triphoton states. For example, the background term stemming from all three photon coming from the LO will always be $H$ polarized. Extending the results in equation 4.11 to include the emission statistics of the sources, it is possible to determine the polarization state of the triphoton background contributions. The polarization state of each of the major background contributions is shown in the second column of Table 4.3.4.

For the N00N state with $T = 1.7$, after the variable partial polarizer we collect $\sim 2000$ SPDC pairs/second into single-mode fibre, corresponding to collection efficiencies of $\eta_1 = 4\%$ and $\eta_2 = 6\%$. The LO is tuned so that $\sim 180,000$ photons/second are collected into the single-mode fibre. The repetition rate of both the SPDC and LO sources is 40 MHz as they are generated by the same pump laser. For these parameters, $|c|^2 = 0.0186$ and $|\alpha|^2 = 0.0045$. Using these values, it is possible to calculate the contributions of each of the major three-photon terms for the N00N state as shown in column 3 of table 1. For the N00N state, the desired combination of a single LO photon and a single pair of SPDC photons occurs $\sim 62\%$ of the time.

### 4.4 Quantum State Tomography

Quantum state tomography (QST) is a technique to fully characterize a quantum state of interest [James 01]. The idea originates from the tomography carried out in medical imaging where many different 2D pictures, or slices, are taken of the subject of interest. If enough slices are taken at differing angles, it is possible to reconstruct a full 3D image of the subject. In the same way, if enough linearly independent measurement are made on a quantum state it is possible to reconstruct its density matrix. Since a typical density matrix has $N^2$ elements, a minimum of $N^2 - 1$ linearly independent measurements are required to reconstruct the state after accounting for normalization. While $N^2 - 1$ measurements are the minimum number required to reconstruct a state, it is often experimentally more convenient to carry out an over-complete set of measurements.
Table 4.2: Significant three-photon contributions to the detected triphoton state. DC1 and DC2 are photons from Type-II SPDC while LO represents a photon from the local oscillator. The state of each three-photon contribution immediately after the variable partial polarizer is shown. For notational compactness the states are written in vector format with \([|3,0\rangle_{H,V},|2,1\rangle_{H,V},|1,2\rangle_{H,V},|0,3\rangle_{H,V}\]). The probability for generating each state in terms of the experimental parameters is given in the third column. For the specific case of the N00N state \((T = 1.7)\), the fourth column shows the calculated percentages of each of the major background contributions.
4.4.1 Hidden Differences

The creation of our triphoton states relies on the photons involved being indistinguishable. In general, an experimental apparatus that tries to combine indistinguishable particles from distinct sources will always leave some residual distinguishability. This distinguishing information may be inaccessible to the experimental measuring device and therefore will be “hidden”. For an ideal entangled triphoton composed of three indistinguishable photons it is impossible to label the polarization of each of the photons—instead the polarization state of the entire triphoton must be considered. In the presence of distinguishing information it becomes in principle (though not necessarily in practice) possible to label each of the individual photons. While the state of an ideal triphoton occupies a permutation symmetric four-dimensional Hilbert space, a triphoton with distinguishing information exists in an eight-dimensional Hilbert space where the ordering of the photons matters. For example, the ideal triphoton state $|2, 1\rangle_{H,V}$ consists of two $H$ and one $V$ photon. In the presence of distinguishing information it is now possible to label each of the photons; the state could really be in some unknown linear superposition of $|HHV\rangle$, $|HVH\rangle$, and $|VHH\rangle$.

The measurements that are accessible experimentally are those related to the angular momentum of the triphoton. An ideal triphoton forms a spin $j = \frac{3}{2}$ particle which corresponds to the four-dimensional Hilbert space of the permutation symmetric states. Any ordering information due to distinguishability can lead to the existence of two $j = \frac{1}{2}$ subspaces, each of which forms a two-dimensional Hilbert space composed of states of mixed symmetry. Each of the $j = \frac{1}{2}$ subspaces contain the same information, and their population places a lower bound on the amount of distinguishing information present [Adamson 07, Adamson 08b, Adamson 08a]. For an experimental apparatus that is not sensitive to permutations of these in-practice indistinguishable particles, coherences between states of different symmetry, which are ordering-dependent, have no influence on any measurement outcomes. This leads to a block diagonal form for the $8 \times 8$ density
matrix composed of a $4 \times 4$ sub matrix corresponding to the spin $j = \frac{3}{2}$ subspace and two $2 \times 2$ sub matrices corresponding to the spin $j = \frac{1}{2}$ subspaces. Due to the multiplicity of the $j = \frac{1}{2}$ subspaces resulting from the experimental distinguishability, there are a total of 20 linearly independent elements (instead of 24) that must be measured in order to specify the complete density matrix of the triphoton.

It is important to note that the population of the $j = \frac{1}{2}$ subspaces only places a lower bound on the amount of distinguishing information present. For example, the state $|HHH\rangle$ is fully symmetric and our measurement apparatus is not sensitive to any distinguishing information present. Similarly, the GHZ state $\frac{1}{\sqrt{2}} (|HHH\rangle + |VVV\rangle)$ with all of the photons in different “hidden” modes would appear identical to a N00N state $\frac{1}{\sqrt{2}} (|3, 0\rangle_{H,V} + |0, 3\rangle_{H,V})$ as shown in figure 4.5. For these states the distinguishing information present does not affect our measurement outcomes in any way—the GHZ state and the N00N state would yield identical fringe patterns in an interferometer. If there were hidden distinguishing information that could affect the outcomes of our measurements, then our technique for characterizing states would account for this in the density matrix. Ultimately it is the density matrix of the state that we are interested in as this will tell us how useful our state is for the purpose we intend to use it for.

While distinguishability does not affect ideal symmetric states, it does play an important role in the preparation of our triphotons. Consider the creation of a triphoton state using mode-mashing where each of the photons now occupies a distinct mode that is hidden from our apparatus:

$$
|\Psi\rangle \propto \left(\hat{a}_H^\dagger + T\hat{a}_V^\dagger\right) \left(\hat{b}_H^\dagger - T\hat{b}_V^\dagger\right) \hat{c}_H^\dagger |0\rangle
$$

$$
= \left(\hat{a}_H^\dagger \hat{b}_H^\dagger \hat{c}_H^\dagger + T \left[\hat{a}_V^\dagger \hat{b}_H^\dagger \hat{c}_H^\dagger - \hat{a}_H^\dagger \hat{b}_V^\dagger \hat{c}_H^\dagger\right] - T^2 \hat{a}_V^\dagger \hat{b}_V^\dagger \hat{c}_H^\dagger\right) |0\rangle .
$$

(4.14)

The overall wave function of the state must be symmetrized with respect to both the
polarization and hidden degrees of freedom. Each term can be written as:

\[
\hat{a}_H^\dagger \hat{b}_H^\dagger \hat{c}_H^\dagger |0\rangle = \frac{1}{\sqrt{6}} |HHH\rangle \left( |abc\rangle + |acb\rangle + |bca\rangle + |cab\rangle + |cba\rangle \right),
\]
\[
\hat{a}_V^\dagger \hat{b}_H^\dagger \hat{c}_H^\dagger |0\rangle = \frac{1}{\sqrt{6}} \left[ |VHH\rangle (|bca\rangle + |bc\rangle) + |HVH\rangle (|abc\rangle + |cba\rangle) + |HHV\rangle (|acb\rangle + |cab\rangle) \right],
\]
\[
\hat{a}_V^\dagger \hat{b}_V^\dagger \hat{c}_H^\dagger |0\rangle = \frac{1}{\sqrt{6}} \left[ |VVH\rangle (|abc\rangle + |bac\rangle) + |VHV\rangle (|acb\rangle + |bca\rangle) + |HVV\rangle (|cab\rangle + |cba\rangle) \right],
\]

(4.15)

where \(a, b,\) and \(c\) represent the hidden degrees of freedom for each photon.

Our measurement apparatus is not sensitive to the hidden degrees of freedom and traces over them; this yields the reduced density matrix for the polarization state of the triphoton

\[
\rho_{pol} = Tr_{abc} (|\Psi\rangle \langle \Psi|).
\]

(4.16)

The effect of distinguishing information on the preparation of a N00N state \((T = 1/\sqrt{3})\) is shown in figure 4.5 for the case where the two photons labelled \(a\) and \(b\) are indistinguishable \((a=b)\) while the third photon in mode \(c\) is distinguishable, and the case where all three photon are distinguishable. It is clear that “hidden” distinguishing information in our state preparation procedure can reduce the purity, fidelity, and performance of our states. Our density matrix formalism allows us to detect distinguishing information in our states that could affect the outcomes of measurements made by our apparatus. Distinguishing information in completely symmetric states, like the N00N state or the GHZ state, is not characterized by our tomographic technique, but this distinguishing information would never affect the outcome of any measurements our apparatus could make. The power of the technique is that we can quantify how much distinguishing information is present that can affect our measurements without having direct access to the degrees of freedom in which the distinguishing information resides.
Figure 4.5: The absolute value of the density matrix elements for a) an ideal GHZ state with hidden distinguishing information, b) an ideal N00N state, c) a N00N state prepared with two indistinguishable photons and one distinguishable photon, and d) a N00N state where all of the photons in the preparation stage are distinguishable. The ideal GHZ and N00N states have the same density matrix; however, the presence of distinguishing information in state preparation can affect the purity, fidelity and quality of the generated triphotons.
4.4.2 Tomographic Measurements

The necessary measurements to reconstruct the density matrix can be made using a polarization analyzer and counting the number of photons at the outputs. In our setup a quarter wave-plate, a half wave-plate, and a quarter wave-plate followed by a polarizing beam-splitter are used to implement the polarization analyzer. At the $H$ output port of the PBS is a single SPCM, while at the $V$ output port a network of beamsplitters divides the counts between three SPCMs. Coincidences from the four SPCMs are measured using a coincidence circuit.

When three of the detectors in the $V$ path fire the state is projected onto $|VVV\rangle\langle VVV|$ which measures $j = 3/2$, $m = -3/2$ (all spins are down). This projector is only able to project onto the completely symmetric subspace. Rotating the waveplates before the PBS is equivalent to performing a rotation on the analyzer; this allows us to “orbit” this projector around the symmetric subspace and fully characterize it. This fully symmetric subspace is most naturally represented by a quasi-probability distribution on the surface of a Poincaré sphere as discussed in section 1.5. Rotating the $m = -3/2$ projector is equivalent to measuring the polarization at different points on the surface of the sphere. This is analogous to classical polarimetry where the polarization is measured along different axes of the Poincaré sphere to determine the Stokes parameters.

Whenever a three-fold coincidence is registered between a photon in the $H$ path and two in the $V$ path a projection onto $|HVV\rangle\langle HVV| + |VHV\rangle\langle VHV| + |VVH\rangle\langle VVH|$, which is a convex sum over all of the $m = -1/2$ states, is performed. Rotating the waveplates allows us to fully characterize all of the subspaces containing $m = \pm 1/2$, which includes the $j = 1/2$ subspaces that contain information on how much distinguishing information is present. Therefore the projections onto $m = -3/2$ and $m = -1/2$ and their rotations are sufficient to completely characterize the density matrix of our triphoton states, even when “hidden” distinguishing information is present. This density matrix will correctly predict the outcomes of any measurements made on our states with
our apparatus. This polarization analyzer can be extended to arbitrary $N$ as long as
projections onto all of the $m$ states are performed [Adamson 08b, Adamson 08a].

To perform quantum state tomography, an over-complete set of 32 linearly independent
polarization measurements are carried out on our triphoton states using a polarization
analyzer composed of a quarter wave-plate, a half wave-plate, and a quarter wave-
plate followed by a polarizing beam-splitter as shown in figure 4.2. Using a constrained
convex-optimization routine, the density matrix for the triphoton states are reconstructed
from these polarization measurements. Figure 4.6 shows the magnitude of the density
matrix elements for the right-handed circularly polarized coherent state ($T = 0$), the
phase state ($T = 0.7$), the N00N state ($T = 1.7$), and the N00N state after the dom-
inant background contributions are tomographically characterized and subtracted off.
The Wigner quasi-probability distributions discussed in the paper are calculated based
on the symmetric portion of the density matrix that contains the relevant polarization
information about the triphoton state.

4.4.3 Measurement Setup

The polarization analyzer used in our experiment is shown in figure 4.2. A quarter wave-
plate and a half waveplate are used to rotate the measurement basis of the polarization
analyzer. A third quarter waveplate right before the fibre coupler is used to help calibrate
the system. The PBS is manufactured by Oz Optics and operates by first outcoupling the
light, passing it through a monolithic PBS, and then recoupling the light at the output
ports. The fibre from the $H$ port is sent to a SPCM (labelled $D$) while the fibre from
the other port is sent to a network of fibre beamsplitters. In the $V$ arm the first fibre
beamsplitter has a splitting ratio of 70:30. The output from the 30% output arm is sent
to a SPCM (labeled $A$) while the other arm is sent to a 50:50 beamsplitter which divides
the remaining light between two other SPCMs (labelled $B$ and $C$). This splitting scheme
allows the light to be split approximately equally between the three detectors. A coin-
cidence between detectors $ABC$ projects the state onto $m = -3/2$ while a coincidence between $D$ and any two of the other detectors projects the state onto $m = -1/2$.

The three single-photon detectors act as a single three-photon detector with a reduced efficiency. A three-fold coincidence will only be recorded when three photons end up at three different detectors (assuming perfect detection efficiency). With the beamsplitter network there is a chance that two or more photons may end up at the same detector, reducing the number of observed three-fold coincidences. This reduction in the observed counting rates needs to be properly accounted for when analyzing the tomography data.

In general, an $N$ photon detector constructed out of a beamsplitter network of $M$ detectors ($M \geq N$ and the photons equally likely to arrive at any one of the detectors) will have an efficiency of

$$E(M, N) = \frac{M!}{(M - N)!M^N}. \quad (4.17)$$

For the $m = 3/2$ three-photon detector composed of the three ideal SPCMs $ABC$, the probability of detecting a three-fold coincidence is $E(3, 3) = 2/9$. Accounting for the different intrinsic detector efficiencies and the fact that the fibre beamsplitters do not divide the counts evenly amongst the three detectors, the efficiency of the three-photon detector is:

$$E'(3, 3) = \frac{3!\eta_A\eta_B\eta_C}{0!} = 6\eta_A\eta_B\eta_C, \quad (4.18)$$

where $\eta_A$, $\eta_B$, and $\eta_C$ are the total efficiencies for a photon arriving at and being detected by detectors $A$, $B$, and $C$ respectively.

For a $m = -1/2$ detection composed of a three-fold coincidence between ideal SPCM $D(AB + AC + BC)$, the three-photon detector efficiency is $E_{1/2} = E(1, 1)E(3, 2) = \frac{2}{3}$. When a state with two $V$ photons and one $H$ photon enters the analyzer, the $H$ photon will always be detected at $D$ while there is a $2/3$ probability that the two $V$ photons will end up at different detectors and register a coincidence. Taking into account the detector inefficiencies, splitting imbalances, and path losses, the effective three-photon detector
efficiency is \(2\eta_D(\eta_A\eta_B + \eta_A\eta_C + \eta_B\eta_C)\). In our setup, the four efficiencies were measured to be \(\eta_D = 0.49\), \(\eta_A = 0.14\), \(\eta_B = 0.15\), and \(\eta_C = 0.14\).

A total of 16 waveplate settings are used to make 32 linearly independent measurements on our states. The measurement settings used and the observed count rates for a N00N state with \(T = 1.7\) are shown in Table 4.4.3. Data is collected for 150 seconds at each waveplate setting. It takes \(\sim 40\) minutes to characterize a single state in this way. After the detector inefficiencies are accounted for, the density matrix is reconstructed using maximum likelihood techniques [Bock 98]. The free Matlab toolkit Sedumi [Sturm 99] was used to carry out the maximum likelihood estimation. The solver was constrained to produce a positive-semidefinite density matrix with a block-diagonal structure that best matched our experimental measurements. To account for experimental errors, a Monte Carlo simulation was carried out to model the errors introduced by the waveplates and the counting statistics.

### 4.5 Results

A total of eleven different triphoton states were prepared by varying the transmissivity of the variable partial polarizer from \(T = 0\) to \(T = 1.7\) and then characterized using QST. The density matrices for a spin-coherent state \((T = 0)\), a phase state \((T = 0.7)\), and a N00N state \((T = 1.7)\) are shown in figure 4.6. While the density matrix description of a state is informationally complete, it has a complicated and non-intuitive basis dependence that makes visualizing the data difficult. A more natural way to visualize the properties of our triphotons is to plot their Wigner functions on the surface of the Poincaré sphere. The spherical symmetry of polarization allows features of the state, such as rotations and squeezing, to show up clearly on the surface of the Poincaré sphere in a basis independent manner.

It is the \(j = 3/2\) symmetric subspace that contains the polarization characteristics of
Chapter 4. Squeezing and Over-Squeezing Triphotons

Table 4.3: The 16 waveplate settings used to carry out quantum state tomography. The measured raw three-fold coincidence rates for the N00N state \((T = 1.7)\) at the \(m = -1/2\) and \(m = -3/2\) detectors, before the path efficiencies have been corrected for, are shown in columns three and four respectively. Data was collected for 150 seconds at each waveplate setting.

<table>
<thead>
<tr>
<th>HWP</th>
<th>QWP</th>
<th>(m = -1/2)</th>
<th>(m = -3/2)</th>
</tr>
</thead>
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<tr>
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<td>114</td>
</tr>
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<td>81</td>
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<tr>
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<td>0°</td>
<td>143</td>
<td>98</td>
</tr>
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<td>0°</td>
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<td>45°</td>
<td>152</td>
<td>103</td>
</tr>
</tbody>
</table>
the triphotons that we are interested in; we plot the quasi-probability distribution for this subspace on the surface of the Poincaré sphere. This representation is not informationally complete by itself—it is necessary to also specify the amount of distinguishing information present. This is found from the populations of the $j = 1/2$ subspaces in the density matrix. The Wigner functions for the eleven measured states are shown is Figure 4.7. There is clear evidence of squeezing in these pictures, something not easily seen in the density matrix representations of the states. It is useful to examine the properties of the spin-coherent, phase, and N00N states in more detail. Different projections of the spherical Wigner functions characterizing these three states are shown in figure 4.8.

### 4.5.1 Spin-Coherent State

When $T = 0$ the partial polarizer only transmits the horizontal polarization, producing a quasi-classical spin-coherent state composed of three horizontally polarized photons. A quarter wave-plate then rotates the coherent state into the circular basis. On the Poincaré sphere, this coherent state has a circularly symmetric quasi-probability distribution centred on the $\hat{S}_3$ axis as shown in Figure 4.8 a), and an uncertainty in $\hat{S}_1$ and $\hat{S}_2$ given by the shot-noise limit.

### 4.5.2 Phase State

Setting $T = 0.7$ produces the phase state $|\zeta\rangle = \frac{1}{2}(|3,0\rangle_{H,V} + |2,1\rangle_{H,V} + |1,2\rangle_{H,V} + |0,3\rangle_{H,V})$, which represents an equally weighted superposition of polarization states. On the Poincaré sphere the Wigner distribution is centred on the $\hat{S}_3$ axis as shown in Figure 4.8 b), but now takes a “banana” shape as the uncertainty along $\hat{S}_2$ decreases below the shot-noise limit while the uncertainty in $\hat{S}_1$ increases above it. The phase state has been called a “jack-of-all-trades” state [Durkin 07] for metrology as its symmetry and squeezing properties allow it to accurately acquire and then track an unknown phase.
Figure 4.6: *Reconstructed Triphoton Density Matrices.* The absolute value of the density matrix elements for a) the coherent state (T=0), b) the phase state (T=0.7), c) the N00N state without background subtraction (T=1.7), and d) the background subtracted N00N state (T=1.7). The binomial distribution of polarization states can be clearly seen for the coherent state in (a). For the N00N state (c), hidden distinguishing information and background terms reduce the fidelity of the state to \(0.69 \pm 0.01\) as compared to an ideal N00N state. After experimentally subtracting off the major background terms (d) and filtering the distinguishing information out, our N00N state has a fidelity of \(0.89 \pm 0.01\) with an ideal state. The density matrices take on a block diagonal form composed of a spin \(j = \frac{3}{2}\) and two spin \(j = \frac{1}{2}\) sub spaces.
Figure 4.7: Triphoton Wigner quasi-probability distributions on the Poincaré sphere for eleven measured states and a background subtracted N00N state. The states are ordered in the direction of increased squeezing. Also shown for each state is the population $D$ of the $j = 1/2$ subspace which puts a lower bound on the amount of distinguishing information present.
Figure 4.8: Triphoton Wigner quasi-probability distributions for a coherent state a), a phase state b), a N00N state c), and a background subtracted N00N state d). For each state the 2D projections and an equal-area cylindrical map projection of the quasi-probability distributions are shown. In a) the coherent state has a uniform distribution centered about the $\hat{S}_3$ axis. As the squeezing parameter is increased to produce a phase state, the width of the angular features decreases until the minimum feature size is reached in d) with the maximally-squeezed N00N state. In both the phase state and N00N state, large regions of quasi-probability distribution take on negative values.
4.5.3 N00N State

Tuning the partial polarizer to $T = 1.7 \approx \sqrt{3}$ creates the N00N state $|\zeta\rangle = \frac{1}{\sqrt{3}} (|3, 0\rangle_{H,V} + |0, 3\rangle_{H,V})$, which is capable of reaching the Heisenberg limit in phase-sensitive measurements and also of achieving super-resolution in lithographic applications [Dowling 98, Mitchell 04, Walther 04, Nagata 07, Ou 97]. Our N00N state has a fidelity of $0.68 \pm 0.01$ with an ideal N00N state. This low fidelity stems from background events in our state preparation and the fact that the three photons are not completely indistinguishable. To account for the background contributions, state tomography was independently carried out on each major background source. This was accomplished by first blocking the LO and measuring the three photon contributions arising from multiple SPDC pairs. Then the SPDC was blocked and the LO measured. The counts from these two processes were subtracted from the measured triphoton rates and then the density matrix was calculated. The resulting background subtracted density matrix is shown in figure 4.6 d), yielding a fidelity with an ideal N00N state of $0.80 \pm 0.03$.

In the metrology and lithography applications for which N00N states have been proposed, the detection schemes rely on N-photon absorbers which are sensitive only to symmetric states. Because the extra elements in the density matrix resulting from distinguishing information represent states of mixed symmetry, they will be automatically filtered out by the detectors in any practical implementation. After performing such filtering and accounting for the major background contributions, our N00N state has a fidelity of $0.89 \pm 0.03$ with an ideal state. As shown in Figure 4.8 d), the Wigner quasi-probability distribution is peaked at the poles of the $\hat{S}_1$ axis, corresponding to finding all three photons being either horizontally or vertically polarized, and displays a three-fold symmetry around the equator.
4.5.4 Marginals

In traditional planar Wigner functions the marginals can be used to obtain valid probability distributions. As discussed in section 1.5.2, the corresponding marginals in our SU(2) Wigner quasi-probability distributions do not yield valid probability distributions. Instead “slices” of the Q quasi-probability distribution can be used to predict the behaviour of our triphotons inside an interferometer. This is seen in Figure 4.9 where a slice about the equator of the Q distribution $Q(0, \gamma)$ for a coherent state, phase state, background-subtracted N00N state, and ideal N00N state exactly corresponds to the fringe pattern that would be obtained in an interferometer. For the N00N state the enhanced phase resolution and the three-fold symmetry are clearly visible.

4.5.5 Quantifying Entanglement

To quantify the degree of entanglement in our states we calculate the negativity $N(\rho) = \sum_j max(0, -\mu_j)$ [Miranowicz 04], a measure related to the Peres-Horodecki criterion [Peres 96, Horodecki 96] where the $\mu_j$s are the eigenvalues of partial transpose $\rho^T$ of the density matrix $\rho$. Any positive value of the negativity indicates entanglement with maximally-enangled states, such as the N00N state, having $N(\rho) = 1$. Using the negativity we can determine the bipartite entanglement between any single photon and the rest of the system. Starting with the coherent state at $T = 0$ with a negativity of $0.05 \pm 0.03$, the degree of entanglement steadily increases until the N00N state is reached which has a negativity of $0.33 \pm 0.03$. After correcting for the measured background contributions, the negativity of the N00N state is $0.52 \pm 0.03$.

4.6 Squeezing and Over-Squeezing

From the quasi-probability distributions in figure 4.7 squeezing is clearly seen. As $T$ increases, the feature sizes on the distributions become smaller indicating an improved
Figure 4.9: A slice around the equator of the $Q$ quasi-probability distribution $Q(0, \gamma)$ (black circles) and the expected interferometric fringe pattern (solid blue line) for a) the coherent state b) the phase state, c) the background subtracted N00N state, and d) the ideal N00N state. $Q(0, \gamma)$ exactly predicts the expected fringe pattern of a state in an interferometer. e) $Q(0, \gamma)$ data for the triphotons states ranging from $T = 0$ to $T = 1.7$ (no background subtraction). The data is offset along the y-axis to make it easier to compare the different expected fringe patterns.
sensitivity. As $T$ approaches 1 the distribution begins to wrap around itself and take on a more complex shape than the simpler “banana” distribution. The distribution for the N00N state is completely wrapped around the sphere, and contains the smallest features possible about the equator. Despite the small feature sizes present in these wrapped states, paradoxically the uncertainty in the Stokes operators begins to increase as further squeezing takes place; a phenomena termed over-squeezing.

The origin of over-squeezing lies in the nature of uncertainty. Uncertainty is the second moment of an intensity measurement, but higher order moments are required to fully take advantage of the increased sensitivity the wrapped states offer. An analogy to this situation is the behaviour of a water balloon as it is squeezed. Imagine that the balloon is perfectly spherical to start as shown in figure 4.10 a). Measuring the uncertainty is analogous to looking at the shadow cast by the balloon. For the spherical balloon the shadow is a circle, similar to the circularly symmetric distribution of a spin-coherent state. If the balloon is squeezed from the sides it will take on an oval shape as shown in figure 4.10 b). Now the shadow is also an oval. This is similar to the banana shaped distribution of a squeezed state. If the balloon is forcefully squished about its middle it will take on the shape of a dumb-bell; the centre will shrink while the areas above and below will expand as shown in figure 4.10 c). The projection of this dumb-bell balloon is a circle that has a larger radius than the unsqueezed balloon despite having a smaller radius at the centre. The balloon is “over-squeezed”: measuring just the shadow is insufficient to observe the fine structural details present.

The uncertainty in the Stokes operators is measured for each of the eleven states created as shown in Figure 4.11. Starting with the coherent state at $T = 0$ and continuing to the state with $T = 1$, the uncertainty in $\hat{S}_2$ decreases below the shot-noise limit while the degree of squeezing increases. After $T = 1$, however, the states begin to over-squeeze as further squeezing leads to an increase in the uncertainty of both $\hat{S}_1$ and $\hat{S}_2$. As over-squeezing occurs the three photons become more entangled, but they seem to depolarize
Figure 4.10: The essential features of over-squeezing can be demonstrated using water balloons. a) A perfectly spherical balloon casts a circular shadow on a flat plane. This is analogous to the uncertainty in the Stokes operators of a polarization spin-coherent state which forms a circle of radius $1/\sqrt{N}$. b) As the balloon is compressed it takes the shape of a spheroid and casts an elliptical shadow. In terms of a squeezed state this is consistent with the banana shaped distribution. c) When the balloon is aggressively squeezed the centre is compressed causing the ends of the balloon to mushroom over. The shadow cast by this highly squeezed balloon is a circle with a larger radius than the original balloon in a) despite the balloon having narrower features. In an over-squeezed quantum state, the uncertainty measurements measure the “shadow” cast by the distributions, and are not sensitive to the smaller features present. Higher order correlations are needed to take advantage of the enhanced precision offered by over-squeezed states.
with respect to the Stokes operators. The N00N state is maximally entangled and contains perfect correlations, yet appears completely unpolarized to intensity measurements. Unlike mixed states, which are represented as broad probability distribution on the surface of the Poincaré sphere (with the maximally mixed state represented by a uniform distribution), the N00N state distribution contains the sharpest features possible due to the quantum correlations that exist. This is analogous to the maximally-entangled GHZ state where any single member appears mixed, yet perfect correlations exist between all particles. For both the N00N and the GHZ states, these correlations only become evident when performing multi-particle measurements using, for example, number-resolving detectors. The three-fold symmetry of the N00N state is directly related to the three-fold enhancement in phase super-resolution of this N00N state [Mitchell 04], and the minimum feature width corresponds to the Heisenberg-limited phase super-resolution possible in interferometry using N00N states [Nagata 07, Resch 07]. Squeezed states are often studied in the context of improving phase measurements with sub shot-noise sensitivity, but for highly squeezed states in the over-squeezing limit (such as the N00N state) multi-photon detectors as opposed to intensity based measurements are required to take advantage of the enhanced precision offered.

4.7 Conclusions

Although spin-squeezing has been examined in numerous systems previously, it has always been in a limit where the sphere could be approximated by a tangent plane [Bowen 02, Heersink 03, Geremia 04, Marquardt 07]. In these experiments the uncertainty distributions are squeezed by $\sim 100 \, \mu$rad on the surface of the sphere. In contrast, the squeezing we measure in our experiment is profoundly affected by the topology of the Poincaré sphere. This represents the first time that over-squeezing has been observed. It is also the first time that spin-squeezing in the few photon limit has been
Figure 4.11: The uncertainty in the Stokes parameters $\hat{S}_1$ and $\hat{S}_2$ for eleven squeezed triphoton states after filtering for distinguishing information has been applied. As the squeezing parameter is increased to $T = 1$, the uncertainty in $\hat{S}_2$ steadily drops. After $T = 1$ the topology of the sphere causes the triphotons states to over-squeeze, and the uncertainty in $\hat{S}_2$ begins to increase again as the state becomes depolarized with respect to single-particle intensity measurements. The insets show the Wigner quasi-probability distributions for the spin-coherent state ($T = 0$), the phase state ($T = 0.7$), and the background-corrected N00N state ($T = 1.7$). The distributions demonstrate that the minimum feature size decreases as the degree of squeezing increases as expected. As the distributions begin to wrap around the Poincaré sphere the symmetry causes the measured uncertainties to increase as shown with the N00N state. The sharper features of the over-squeezed states are only recovered when higher order correlations are measured. Solid lines indicate theory, while crosses are experimental data points. The discrepancy between the measured states and theory is due to background contributions in our preparation method. The solid circles are the uncertainties in the N00N state after corrections for background events and distinguishing information have been made.
measured. Our work unifies the concepts of spin-squeezing of intense beams and multi-photon Heisenberg-limited metrology, highlighting the deep connection between squeezing and entanglement. This is indicated in the maximally squeezed N00N state where no single-particle information remains and all the information resides purely in non-classical correlations. With improvements in single-photon sources, this relationship between squeezing and entanglement can be further exploited to engineer custom nonclassical states required in a variety of quantum information technologies.
Chapter 5

Conclusions


In this thesis the quantum states of interest are built up photon-by-photon, giving us precise control over their properties. While our squeezed states contain only a modest number of photons compared to other squeezed systems, we are able to study the previously inaccessible over-squeezing regime. This bottom up approach is a powerful way to study the physics of more complex systems; over 50 years after Feynman’s famous lecture there is still plenty of room left at the bottom.

There are many open questions in quantum interferometry that need to be addressed. It is still unknown what combination of states and measurement strategies are best suited to the task of acquiring and tracking an unknown phase. There is also research to be done to investigate what class of squeezed states are most sensitive to arbitrary rotations and phase shifts—a problem known as quantum sensing. Deep questions remain concerning the meaning of entanglement and the appropriate methods to quantify it in highly squeezed systems. Our group is working on extending the techniques discussed in this thesis to begin tackling some of these issues.

Finally, the new photon sources that have been developed provide a strong platform for future experimental investigations into a wide variety of quantum information and
foundational issues. These sources should keep our group at the cutting-edge of Feynman’s bottom for many years to come.
Bibliography


