Ultrafast Sources of Entangled Photons for Quantum Information Processing

by

Oktay Onur Kuzucu

B.S. Electrical and Electronics Engineering, Middle East Technical University (2001)
S.M. Electrical Engineering and Computer Science, Massachusetts Institute of Technology (2003)

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Abstract

Recent advances in quantum information processing (QIP) have enabled practical applications of quantum mechanics in various fields such as cryptography, computation, and metrology. Most of these applications use photons as carriers of quantum information. Therefore, engineering the quantum state of photons is essential for the realization of novel QIP schemes. A practical and flexible technique to generate high-purity entangled photon pairs is spontaneous parametric downconversion (SPDC) which finds its use in many QIP applications such as quantum key distribution (QKD) and linear optics quantum computation. SPDC is often used with ultrafast lasers to generate photon pairs with precise timing and engineered spectral properties. In this thesis, we focused on two photonic QIP applications using ultrafast-pumped SPDC. We first pursued the design and implementation of a pulsed narrowband polarization-entangled photon pairs at 780 nm for free-space entanglement-based QKD. We built and characterized a compact narrowband ultraviolet pump source and a polarization-entangled photon source based on SPDC in a polarization Sagnac interferometer. We then studied the generation of coincident-frequency entangled photons for Heisenberg-limited quantum metrology. Using extended phase-matching conditions in a periodically-poled KTP crystal, generation of coincident-frequency entanglement was verified and frequency indistinguishability was achieved for broadband signal and idler photons at \( \sim 1.58 \mu m \). We also developed a novel time domain characterization technique based on time-resolved single-photon upconversion. Using this technique, we measured the joint temporal density of a two-photon state for the first time and observed temporal anti-correlation for the coincident-frequency entangled state as predicted by Fourier duality. This new technique complements existing frequency domain methods for a more complete characterization of two-photon states.

Thesis Supervisor: Franco N.C. Wong
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Chapter 1

Introduction

Quantum mechanics and its implications have produced a paradigm shift in many well-established fields such as communication theory, coding and error correction, cryptography, metrology and computational complexity of algorithms. The “quantum” counterparts of classical research areas benefited from novel resources, such as superposition and entanglement, that can be utilized to cast a fresh look at well-known problems with new tools. Using this “quantum toolbox” many schemes have been proposed for providing secure communication between two parties [4, 5], realizing polynomial-time computation for classically intractable problems [6, 7], building fault tolerant computational schemes against decoherence and errors [8, 9], and beating the standard quantum limit in precision metrology [10, 11]. One of these schemes, quantum key distribution (QKD), has been developed into a practical tool that is now moving from the academic laboratory into a business. There are high hopes that more novelties of the quantum toolbox will find commercial applications in the years to come.

Among the physical systems that currently serve as testbeds for quantum information processing (QIP), the photon is an excellent candidate for encoding and long-distance transmission of quantum information. Because of its limited interaction with the transparent environment, a photon suffers minimal decoherence. As the carrier of quantum information, a photon can be interfaced with a multitude of physical systems for QIP. For instance, extremely narrowband photons can mediate the transfer
of quantum information coherently between remote atomic quantum memories, where information can be encoded on the polarization states of single photons [12]. We can also consider the other limit, where a multi-photon broadband coincident-frequency entanglement can achieve the ultimate physical limits for timing and precision measurements [11, 13]. Alternatively, photonic entanglement can be in more than one degree of freedom for testing the validity of quantum mechanics or superdense encoding of classical information [14, 15]. We also observe that two-photon entanglement in any given degree of freedom (polarization, momentum, time-bin, etc.) can find its use in either building or testing the security limits of today’s QKD technology [5, 16, 17]. A further survey of other photonic QIP applications would reveal that each application requires a specific photonic quantum state for an effective interface with the underlying physical system. Therefore, a special emphasis can be placed on application-specific engineering of the quantum state of single photons or entangled photons. Only with the capability of fully controlling and manipulating the single- or multi-photon quantum states can we expect the future prospects of QIP, such as large-scale linear optics quantum computation (LOQC) [18], to materialize. With this motivation, we undertake the task of engineering entangled photon sources for various QIP applications.

In our work, we exclusively use spontaneous parametric downconversion (SPDC) for generation of the entangled photon pairs. SPDC is a nonlinear process in which a single energetic pump photon is converted into a pair of photons that obey energy and momentum conservation. Over the years, SPDC has been used as an efficient technique that can be characterized in various degrees of freedom such as polarization, frequency, momentum and orbital angular momentum. As compared to other single- or few-photon emitters, such as quantum dots [19] or four-wave mixing in fibers [20, 21, 22], SPDC presents itself with flexible control over the relevant degrees of freedom which is desirable for photonic quantum-state engineering. Other notable features of SPDC include high-efficiency and high-flux generation of heralded or entangled photon pairs with low background and its potential for compact packaging to be deployed in practical applications. SPDC has been extensively used in
many QIP applications such as QKD [23, 24, 25], quantum teleportation [26] and cluster-state quantum computation [27, 28, 29]. SPDC has also been utilized for generating two-photon states that are entangled in multiple degrees of freedom. Using this hyper-entanglement it was possible to demonstrate the complete Bell-state measurements [14], perform entanglement distillation [30, 31, 32, 33], purification [34], and superdense coding [15].

We note that the high degree-of-control in SPDC enabled the tailoring of the two-photon output state for many important QIP applications. In practice, the quantum-state engineering for a specific application can be accomplished through controlling the internal and external dynamics. For instance, the SPDC source output can be externally controlled or filtered to achieve the desired properties. We can exemplify this by noting that the emission modes of a downconverter can be made to coincide with an atomic resonance using an external cavity [35, 36]. Alternatively, one can pursue controlling the internal dynamics of SPDC to engineer the quantum state directly from the source. For example, SPDC can be configured as a heralded source of single-photons for LOQC and QKD [37]. Here, the heralding process relies on the successful detection of one of the conjugate photons and ideally these photons are generated in a well-defined spatio-temporal mode. This requirement puts a special emphasis on spectral engineering for the joint output state, where coherent spectral control can be provided by manipulating the phase-matching function and the pump envelope for ultrafast SPDC [38, 39, 40, 41]. In other applications, such as free-space entanglement-based QKD, providing both internal and external controls may be necessary.

Let us briefly touch upon the properties of entanglement. Entanglement is a quantum mechanical resource that has no classical analog. A multi-partite pure quantum state is said to be entangled when it is not possible to express the joint state as a product state. The simplest example of this class of states is a Bell state of two photons $A$ and $B$, $|\Phi^+\rangle = (|00\rangle_{AB} + |11\rangle_{AB})/\sqrt{2}$, where $|0\rangle$ and $|1\rangle$ constitute the two possible states of the photons such as horizontal (H) and vertical (V) polarizations. This maximally-entangled bipartite state can exhibit stronger-than-classical
and nonlocal correlations when its joint statistics are measured by two parties. This argument extends to multi-partite case \((n > 2)\), where quantum correlations can be observed over multiple systems.

SPDC has been frequently utilized for the generation of two-photon or multi-photon entangled states. One can consider polarization entanglement as an example. A maximally-entangled two-photon state such as a singlet-state \(|\psi^-\rangle = (|HV\rangle_{SI} - |VH\rangle_{SI})/\sqrt{2}\), can be achieved through the coherent superposition of two distinct polarization output states generated by the SPDC process. The subscripts S and I refer to signal and idler modes, respectively. We consider the combination of the outputs of two degenerate type-II downconverters at a polarizing beam-splitter, sketched in Fig. 1-1 [1].

![Figure 1-1: Combination of two type-II SPDC outputs generating orthogonally polarized signal and idler photons at a polarizing beam-splitter (PBS), resulting in the polarization-entangled signal and idler photons being deterministically separated at the output, see Ref. [1].](image)

In this configuration, the first type-II downconverter 1 is arranged to produce an \(|HV\rangle_{SI}\) output state, whereas the second downconverter generates a \(|VH\rangle_{SI}\) output. Provided that we drive both downconverters coherently to maintain the phase coherence between the two output terms, we can generate any desired Bell state by adjusting the relative phase of the two outputs. It is essential to erase any distin-
guishing information between the two output terms which can potentially degrade the output state purity and the entanglement quality. This calls for arranging both downconversion processes identically, i.e., with the same spectral bandwidth and the same spatio-temporal mode. In the literature one can find other methods of generating the coherent superposition of the two SPDC outputs, which may have their own practical advantages. For instance one can use either a single nonlinear crystal [42, 43, 44] or two identical nonlinear crystals [45, 46]. The interferometric combination as illustrated in Fig. 1-1 may require a free-space arrangement of overlapping beam paths such as a Mach-Zehnder [44, 47] or Sagnac-type interferometer [48, 42]. Alternatively, directly overlapping emission cones from two perpendicularly aligned nonlinear crystals can also be used [49, 45]. In all of these arrangements, the common theme is to provide quantum interference through phase-coherent superposition of two possible downconverter outputs.

Since the first demonstration of high efficiency polarization entanglement by Kwiat et al. in 1995 [45], the performance of SPDC-based entanglement generation has shown tremendous improvement. The progress is reflected by the quality of entanglement generation in recent bulk crystal SPDC experiments. Source performance is typically quantified by the quantum-interference visibility and the source brightness, which is given by the number of detected coincidences per unit time per unit pump power per detection bandwidth. The state-of-the-art polarization-entangled photon source comes from our group exhibiting $>99.5\%$ quantum interference visibility and $10^4$ detected pairs/s/mW/nm [42, 50]. Comparing this with the original 1995 Kwiat’s result of $\sim98\%$ interference visibility with 0.07 detected pairs/s/mW/nm, we observe that current SPDC sources have become much more efficient through better crystals and improved control over the quantum superposition process. For example, a coherent combination of two collinear downconverter outputs with a Mach-Zehnder [44] or a Sagnac [42, 50] interferometer can achieve the required superposition over the entire transverse extent of the optical beam. Similarly, the superposition realized by perpendicularly oriented type-I phase-matched BBO crystals with overlapping emission cones comes with an enhanced source brightness [51] as compared to intersecting
emission cones of ordinary and extraordinary photons from a single type-II BBO crystal with a limited output flux [45].

In this dissertation, we focus on two distinct problems in the QIP framework. First, we investigate a source suitable for Ekert’s entanglement-based QKD (eQKD) protocol for a line-of-sight free-space communication channel [5]. Specifically, we design an ultraviolet pulsed pump and an efficient downconverter to implement a high-purity, high-flux source of narrowband polarization-entangled photons at near-infrared wavelengths. The second problem we address concerns ultrafast-pumped SPDC for the generation of frequency-entangled photons. By manipulating the phase-matching properties of the downconversion crystal, we construct and characterize a broadband coincident-frequency entangled two-photon source. This implementation preserves the frequency indistinguishability of the output photons which can be used for clock synchronization, Heisenberg-limited precision metrology or LOQC.

QKD has been extensively studied with weak coherent state inputs and tested for free-space and fiber communication links [52, 53, 24, 25]. The eQKD system based on polarization entanglement can offer some advantages over attenuated laser based QKD schemes. One can eliminate the active modulation components in laser-based QKD for the selection of generation and measurement bases. The joint measurement of the two-photon state can be performed with passive components such as polarizing beam-splitters, half wave-plates, high extinction-ratio polarizers and single-photon detectors. Furthermore, eQKD can be immune to certain types of attacks, such as photon number splitting attack, whereas the attenuated laser based design is not [54]. We note that as QKD moves deeper into the commercial phase, the practical requirements for the transceiver design will be more demanding. In particular, one should consider self-contained, compact and robust implementations that can be potentially used on mobile free-space platforms. For such a source design, it is necessary to establish the essential properties of the free-space channel in the presence of ambient solar radiation. The channel properties alone dictate most of the specifications for the design of the eQKD transceiver that is based on polarization entanglement generation in SPDC. Furthermore, one needs to address the practical limitations for
such a transceiver design on the security of the communication link. Note that a cost-effective and field-deployable design calls for a compact pump source for driving the SPDC process efficiently. Therefore, one should budget for an inexpensive and high-performance pump source design that can be seamlessly integrated with the downconverter for mobile eQKD applications.

Let us briefly review the requirements of free-space atmospheric communication for high signal-to-noise transmission of entangled photons in the presence of ambient solar radiation. As previous studies of QKD show, the near-infrared range of the spectrum is ideal for generating secret keys efficiently [3]. Given that the solar radiation is generally broadband, one can obtain the best signal-to-noise performance with appropriate temporal and spectral filtering. These filtering requirements call for narrowband, pulsed operation in the near-infrared regime for the downconversion source. We first introduce the design of the high-power, narrowband pump source at \( \sim 390 \text{ nm} \). The source design relies on a master oscillator – power amplifier (MOPA) configuration with a passively modelocked fiber ring laser at \( \sim 1560 \text{ nm} \). The amplified output of the fiber laser is then frequency quadrupled by two consecutive stages of efficient second harmonic generation. Later, we focus on the design of high-flux and high-purity generation of polarization-entangled photons from a type-II phase-matched degenerate SPDC process. We utilize a polarization Sagnac interferometer based design that uses a single bulk periodically-poled KTP crystal to generate inherently phase-stable polarization-entangled two-photon state [42, 50]. We then look at the entanglement quality through measurements of the quantum-interference visibility, Clauser-Horne-Shimony-Horne (CHSH) type Bell’s inequality violation and quantum state tomography. It is also important to quantify the entanglement quality in the low-power and high-power regimes with the latter case displaying degradation in the quantum-interference visibility due to multiple pair emissions. We analyze the impact of multi-pair events to deduce the practical limitation of the eQKD scheme in the pulsed regime as compared to the cw regime. For comparison, we introduce a theoretical model for the quantum-interference visibility degradation as a function of the mean photon pair probability, which gives additional insight into the effects of loss.
Finally, we suggest design alternatives for future free-space eQKD implementations.

We later concentrate our efforts on the generation and characterization of broadband frequency-entangled two-photon states. This class of continuous-variable entangled states are potentially useful for some important QIP applications such as quantum clock synchronization and precision metrology beyond the shot-noise limit [13, 55, 11]. The formalism of entanglement generation using discrete degrees of freedom also extends to continuous variables such as momentum (position) and frequency (time) spaces. Once again, providing the phase coherence for all terms in the superposition is an essential requirement towards the generation of high-purity multi-photon entanglement. As an example, consider the joint output state of an SPDC process with a continuous-wave (cw) pump and a spectral amplitude, $\phi(\omega)$:

$$
|TB\rangle = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \phi(\omega) |\Omega_C/2 - \omega\rangle_S |\Omega_C/2 + \omega\rangle_I 
$$

(1.1)

where $\Omega_C$ is the center frequency of the pump and $|\omega_j\rangle_K$ represents a single-photon Fock state at frequency $\omega_j$ for $K=S, I$. This so-called “twin-beam” state corresponds to a frequency anti-correlated two-photon state [56, 57]. Because of Fourier duality, the generation of the two twin-beam photons are positively-correlated or “coincident” in the time domain. As we introduce a broadband, femtosecond pump source, the spectral and temporal correlations of the two-photon state become more complex. In general, we observe that the interplay between the pump spectral amplitude and phase-matching function determines the joint spectral characteristics for the two-photon state [58, 59, 41]. Without any control over the dispersive properties of the nonlinear crystal or the broadband pump spectrum, the joint state yields a distinguishable output state where individual spectral distributions of the signal and idler can be substantially different [60, 58, 59, 38, 41]. This distinguishability can be measured with a Hong-Ou-Mandel (HOM) interference experiment as depicted in Fig. 1-2 [2]. The signal and idler photons interfering at a 50–50 beam-splitter generate a dip in the coincidence profile, as registered by two single-photon detectors. As the two photons become spectrally discernable, the interference visibility degrades. The
underlying two-photon distinguishability constitutes one of the practical barriers in broadband femtosecond laser pumped SPDC.

Figure 1-2: Hong-Ou-Mandel interferometer, which measures fourth order correlation for two incoming photons interfering at a 50–50 beam-splitter [2]. For indistinguishable photons arriving at the beam-splitter simultaneously, we observe destructive interference between the probability amplitudes for the case in which one photon is detected at each output port, resulting in a dip in the coincidence profile, as depicted in the inset.

The conventional method for erasing the frequency distinguishability relies on using either a thin nonlinear crystal [61] or narrowband spectral filtering of the two-photon output [62]. The former approach can improve the spectral indistinguishability by limiting the effects of temporal and spatial walk-off for signal and idler in the birefringent nonlinear crystal. However the output photon flux from the thin crystal is quite limited. The latter approach for restoring the two-photon visibility is restricting the signal and idler to a narrowband spectral regime with a filter at the expense of a prohibitively low output flux. Since both methods have undesirable features, a direct technique to create spectrally indistinguishable photon pairs is needed. For this, it is necessary to assess the dispersive and phase-matching properties of the nonlinear crystal at hand and observe how the spectral densities of the signal and idler can be made identical. A few groups have suggested and implemented novel methods of restoring frequency indistinguishability for SPDC under ultrafast
pumping without resorting to tight filtering schemes. Composite systems of nonlinear and anisotropic crystals [63] or controlled spatial dispersion and focusing geometries [64, 65] for managing group velocities in SPDC have provided means of generating frequency-indistinguishable photons. In these implementations, we note that coherent manipulation of joint spectral properties of the two-photon state can modify the underlying frequency correlations between signal and idler. In particular, we look at the generation of the positive frequency-correlated two-photon state. This type of frequency entanglement carries interesting prospects for quantum state metrology. It has been known that the standard quantum limit (or shot noise limit) on time-of-flight measurements with photons is not a fundamental limit. We can consider the timing uncertainty resulting from the averaging measurement within $M$ channels, each containing $N$ independent photons. If we pick the spectral width of all the photons to be the same, $\Delta \omega$, the timing uncertainty for this measurement defines the standard quantum limit [66],

$$\Delta t_{SQL} = \frac{1}{\Delta \omega \sqrt{MN}}.$$  \hfill (1.2)

However, entanglement can give us an edge and many theoretical studies focused on using novel entangled states for interferometric sensitivities beyond the standard quantum limit [10, 67, 68, 69, 70, 13]. For example, Giovannetti et al. proposed a method to enhance the time-of-flight measurement sensitivity by using a coincident-frequency entangled state over $M$ distinct modes, each containing exactly $N$ photons [13, 11]. Given that spectral width for all photons is $\Delta \omega$, this quantum state can achieve Heisenberg-limited timing sensitivity:

$$\Delta t_H = \frac{1}{\Delta \omega MN}.$$  \hfill (1.3)

The factor of $\sqrt{MN}$ enhancement comes from the fact that while the arrival times of photons can be random, their average is highly localized in time and the detection is performed over a number-squeezed state. Therefore, it is of great interest to experiment with the approximate physical systems that can exhibit the prescribed
frequency correlations and to investigate whether the timing sensitivity can be improved beyond the standard quantum limit. We focus on the simplest case, where the output state comprises two coincident-frequency entangled modes, each containing a single photon \((M = 2\) and \(N = 1\)). Giovannetti et al. proposed to generate the necessary frequency correlations from ultrafast-pumped SPDC by using extended phase-matching conditions \([56, 57]\). We also note that similar schemes have been proposed in the context of SPDC spectral engineering and restoration of two-photon frequency indistinguishability \([58, 71, 59, 41]\). In these schemes, the manipulation of two-photon frequency correlations is achieved by controlling the joint spectral amplitude, \(\tilde{A}(\omega_S, \omega_I)\), in terms of which the joint state is expressed:

\[
|\psi\rangle = \int \int \frac{d\omega_S}{2\pi} \frac{d\omega_I}{2\pi} \tilde{A}(\omega_S, \omega_I) |\omega_S\rangle_S |\omega_I\rangle_I \tag{1.4}
\]

where, \(\tilde{A}(\omega_S, \omega_I)\) is proportional to the product of the phase-matching function and the pump spectral amplitude. If one can engineer the joint spectral amplitude appropriately, the frequencies of the two photons can be tailored to exhibit positive correlation and in the time domain one would observe anti-correlation. Extended phase-matching corresponds to inverse group-velocity matching in ultrafast optics and it enables full utilization of the broadband pump spectrum. Application of extended phase-matching conditions results in ultrabroad phase-matching bandwidth which can be seen in second harmonic generation \([72]\). When the same phase-matching configuration is used for ultrafast-pumped SPDC, we can observe that the output state consists of frequency indistinguishable photon pairs without any external spectral filtering and the HOM dip is deep without spectral filtering \([73]\). We then proceed with verifying the coincident-frequency entanglement by contrasting single-photon autocorrelation times against two-photon correlation times. This comparison is instrumental to checking whether the joint state can be expressed as a product state. We further investigate the variations of the spectral correlations by filtering the broadband pump spectrum and comparing the resulting single-photon and two-photon correlation times.
The quantification of coincident-frequency entanglement is of practical importance for many QIP applications. We know that unless the joint spectral amplitude is engineered in a special way, the resulting two-photon state will not be factorizable. An important consequence of this fact relates to the output state purity of a heralded SPDC photon. For a typical two-photon state, the heralding detection of a conjugate photon projects the other photon to a random mixture of pure states [39, 74]. However, most of the aforementioned QIP tasks require pure single-photon states with well-defined spectral modes. Therefore, one of the challenges in frequency-entangled states is to eliminate the spectral correlations between the two photons altogether to generate two independent photons [38]. This requires one to measure the joint spectral amplitude of the two-photon state.

The conventional method to observe the frequency correlations relies on tunable narrowband filtering of the signal and idler photons and subsequent coincidence detection [38, 39, 37]. As both filters are tuned around the relevant spectral region, the resulting coincidence profile effectively measures the modulus square of the joint spectral amplitude, i.e., the joint spectral density. The shape of the joint spectral density gives important information about the factorizability of the output state [38, 39, 37]. However, we note that there are some practical difficulties with the joint spectral density measurement scheme. Measuring the joint spectral density, which can be alternatively viewed as the two-dimensional probability distribution for the output state, does not reveal the complex phase information of the joint spectral amplitude. Therefore, we cannot tell apart transform-limited single photons from chirped single photons. Another technical difficulty with this measurement concerns the detection of the filtered single-photons. The induced spectral slicing on signal and idler can reduce the detected singles and coincidences rates drastically. This loss factor can be tolerated in near-infrared range of the spectrum, where efficient silicon avalanche photodetectors (Si APDs) are available. However, the detection at infrared wavelengths is traditionally performed with low quantum-efficiency InGaAs APDs, which are operated in gated mode with long dead times. The overall reduction in the count rate due to spectral filtering of both photons can slow the joint spectral density measure-
ment to inconveniently low count rates. Therefore, we seek to develop an alternative characterization technique for monitoring joint statistics of the two-photon state.

The two-photon joint statistics in the frequency domain can be mapped onto the time domain via two-dimensional Fourier transformation. Expressing the joint state in time-variables and observing the two-photon correlations in the time domain is as powerful as the joint spectral density measurement. Instead of the narrow spectral filtering, we need to implement a narrow temporal filtering scheme followed by coincidence detection to monitor temporal correlations. This can be achieved with an ultrafast single-photon detection scheme with timing resolution on the order of 100 fs. However, the current single-photon detector technology can only deliver a timing resolution of several tens of picoseconds. Thus, we investigate another well-established photodetection scheme that was previously used to detect infrared photons at telecom wavelengths efficiently. Namely, we look at single-photon upconversion through sum-frequency generation [75, 76, 77, 78]. We note that the timing resolution feature for the single-photon upconversion can be added by using an ultrafast laser as the classical pump source. Effectively we use an ultrafast laser pulse as a sampling probe to pinpoint the single-photon arrival times and simultaneously monitor the coincidences to map the joint temporal statistics of the two-photon state.

Rather different from previous single-photon upconversion experiments, we use a non-collinear phase-matching scheme in a $\chi^{(2)}$ nonlinear crystal. The non-collinear phase-matching geometry enables us to implement two independent upconverters in a single nonlinear crystal. This configuration is advantageous because the detection of the infrared photons by frequency conversion is performed with efficient Si APDs. Furthermore, we can achieve high temporal resolution without being limited by the speed of the single-photon detectors. We discuss the design and implementation of this time-resolved photodetection scheme, which is synchronously driven by the same pump source that is used for the coincident-frequency entangled two-photon state experiment. We characterize the two-photon temporal correlations with the time-resolved photodetection scheme and also monitor the changes in the joint temporal density when the SPDC pump spectrum is modified. We observe that the amount of
continuous-variable entanglement can be varied for different values of the input pump bandwidth. Furthermore, we present a photodetection model based on single-photon upconversion with a finite pump bandwidth and compare the theoretically predicted coincidence profiles with the experimental results. We also discuss how the time-resolved upconversion scheme can be utilized for other practical purposes such as the evaluation of the grating homogeneity for quasi-phasematched nonlinear crystals.

This thesis is organized as follows. We present a brief theoretical background on SPDC in Chapter 2 that gives the preliminaries for the generation of non-classical photon pairs from a $\chi^{(2)}$ nonlinear crystal and single-photon detection via sum-frequency generation. Chapter 3 details the design and characterization of the pump and down-conversion sources for a free-space entanglement based QKD system. We focus on the source characterization in low-flux and high-flux limits and investigate the performance limitations due to the generation of multiple photon pairs in the same pump pulse. We then shift our attention to broadband SPDC in Chapter 4, where we address the generation of the coincident-frequency entangled two-photon state. We elaborate on the extended phase-matching scheme in a PPKTP crystal with a broadband femtosecond pump input [57]. We detail our measurements with HOM and Mach-Zehnder interferometers to verify the frequency indistinguishability of downconverted photons and the coincident-frequency entanglement therein. The characterization and quantification of frequency entanglement is investigated in Chapter 5, where we introduce a novel time domain characterization technique for two-photon temporal correlations. Finally, Chapter 6 concludes the thesis with the summary of contributions and future prospects of the presented work for quantum information processing.
Chapter 2

Preliminaries

2.1 Introduction

The process that involves the destruction of a single energetic photon and generation of two daughter photons is a nonlinear interaction known as spontaneous parametric downconversion (SPDC). It is the fundamental process that we utilize in this thesis, along with other three-wave mixing interactions such as second harmonic generation (SHG) and single-photon upconversion via sum-frequency generation (SFG). Therefore, it would be appropriate to give a brief background for these processes. The theoretical underpinnings of SPDC and single-photon upconversion would later come in handy in understanding and controlling the quantum states generated from these interactions. In this chapter, we summarize the theoretical aspects of SPDC and single-photon upconversion and at appropriate points we try to connect them to some of the experimental configurations utilized in this thesis.

2.2 Spontaneous Parametric Downconversion

We first study spontaneous parametric generation of photon pairs in a nonlinear crystal. The theory of SPDC has already been extensively developed in previous works [79, 80, 81, 58, 38, 61]. A common theme in these works is to understand the physical properties of SPDC by adopting the quantum theory of electromagnetic fields and
studying the weak nonlinear interaction between input and output fields. Expressing the joint state in this framework is instrumental to understanding the constraints on energy and momentum of these spontaneously generated photons. Experimentally, these photon pairs are produced by passing intense classical laser light, monochromatic or pulsed, through a nonlinear medium without inversion symmetry. Figure 2-1 depicts such an experimental arrangement, where a classical pump beam is incident on a nonlinear crystal and occasionally downconverts into two lower energy photons.

Figure 2-1: Spontaneous parametric downconversion in a $\chi^{(2)}$ nonlinear crystal. A strong horizontally polarized pump beam at $\omega_P$ is incident on the crystal input facet. A pump photon is occasionally converted to orthogonally polarized signal ($\omega_S$) and idler ($\omega_I$) photons for a type-II phase-matched interaction. We depict the collinear propagation of the output photons which exemplifies most of our experimental configurations.

Three-wave mixing between a classical pump field and two vacuum input modes can be studied in the interaction picture [81, 58, 38]. In this approach, the joint state of the downconverted photons is perturbatively calculated by introducing an interaction Hamiltonian. We can consider the case where a broadband pump pulse is incident on a $\chi^{(2)}$ nonlinear crystal and all other input modes are unexcited. We adopt a simple interaction Hamiltonian, $\hat{H}_{int}(t)$, defined over the effective interaction volume, $V$ [81, 38]:

$$\hat{H}_{int}(t) = \int_V d^3r \ \chi^{(2)} \hat{E}_P^{(+)}(r,t) \hat{E}_S^{(-)}(r,t) \hat{E}_I^{(-)}(r,t) + H.c. \quad (2.1)$$

where, $H.c.$ denotes the Hermitian conjugate of the previous term. The positive frequency operator $\hat{E}_j^{(+)}(r,t)$ is defined for pump, signal and idler modes ($j = P, S, I$) as $^1$:

$^1$We express the vectorial quantities in bold face, i.e. $\mathbf{k} = \hat{x}k_x + \hat{y}k_y + \hat{z}k_z$. 

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\[ \hat{E}_j^{(+)}(r, t) = i \int_0^\infty \frac{d\omega_j}{2\pi} \xi_j \hat{a}_j(\omega_j) \exp \left[ i(\mathbf{k}_j(\omega_j) \cdot \mathbf{r} - \omega_j t) \right] \quad j = P, S, I. \tag{2.2} \]

where the continuum-field single-mode annihilation operator \( \hat{a}_j(\omega_j) \) satisfies the canonical commutation relations: \( \left[ \hat{a}_j(\omega_j), \hat{a}_k^\dagger(\omega'_k) \right] = 2\pi \delta_{jk} \delta(\omega_j - \omega'_k) \). We have the normalization factor \( \xi_j = \sqrt{\frac{\hbar \omega_j}{(2\epsilon_0 n^2 V_Q)}} \) with \( V_Q \) being the quantization volume. We ignore the frequency dependence of \( \xi_j \) as it is a slowly-varying function of \( \omega_j \) [38].

Note that, we have also suppressed the polarization vectors associated with the electric fields. These polarization vectors are absorbed into the nonlinear susceptibility, \( \chi^{(2)} \), as it is reduced to a scalar form from the original tensorial expression \( \chi^{(2)}_{ijk} \). Such a simplification is valid because we configure the polarizations of the fields for an efficient phase-matched interaction that utilizes a particular tensor element, e.g., \( \chi^{(2)}_{333} \) for type-0 phase matching in periodically-poled lithium niobate (PPLN) where all fields are polarized along the crystallographic \( z \)-axis. Since the nonlinear interaction is usually weak, the pump pulse can be treated as an undepleted classical plane wave given by the Fourier transformation:

\[ E_P^{(+)}(r, t) = \int_0^\infty \frac{d\omega_P}{2\pi} \xi_P(\omega_P) \exp \left[ i(\mathbf{k}_P \cdot \mathbf{r} - \omega_P t) \right] \tag{2.3} \]

Without any loss of generality, we can assume that the pump beam propagates in the \(+\hat{z}\)-direction. We also introduce a physical interaction volume bounded by the crystal dimensions. Let us focus on the integration over the spatial variables. In practice, the transverse extent of the crystal is much larger than the pump diameter. Therefore, we can ignore the diffraction effects of the pump and safely extend the transverse plane spatial integral limits to infinity. The integrals over the transverse coordinates can be evaluated to yield,

\[ \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dx dy \exp[-i(k_{S,x} + k_{I,x})x] \exp[-i(k_{S,y} + k_{I,y})y] = (2\pi)^2 \delta(k_{S,x} + k_{I,x})\delta(k_{S,y} + k_{I,y}), \tag{2.4} \]
which defines the momentum conservation for the signal and idler photons in the \( xy \)-plane. We can observe the anti-correlation for the signal and idler momenta for a plane-wave pump input, which is a key mechanism to obtain momentum entanglement. We further simplify the transverse momentum picture for the signal and idler by constraining our treatment to collinearly propagating output modes. This restriction applies to our experimental arrangements where the detected modes have a limited transverse wavevector range limited either by fiber coupling or an aperture. Therefore, it would be legitimate to reduce the transverse wavevectors for signal and idler to plane-wave modes propagating along the \( z \)-axis as well, i.e. \( (k_{j,x} = k_{j,y} = 0, \text{ for } j = S, I) \). Thus, we can substitute the pump envelope and field operator expressions in the interaction Hamiltonian to obtain

\[
\hat{H}_{\text{int}}(t) = \kappa \int_{-L/2}^{L/2} dz \iint d\omega_P d\omega_S d\omega_I \mathcal{E}_P(\omega_P) \hat{a}_S^\dagger(\omega_S) \hat{a}_I^\dagger(\omega_I) \times \exp \left[ -i(k_S(\omega_S) + k_I(\omega_I) - k_P(\omega_P))z \right] \exp \left[ i(\omega_S + \omega_I - \omega_P)t \right] + H.c.
\]

where \( \kappa \) contains the proportionality terms which exhibit little variation over the bandwidth that is much smaller than the center wavelength. Without loss of generality, we have also positioned the nonlinear crystal at the \( z \)-axis origin for simplifying the evaluation of the integral.

Note that, before the incidence of the pump pulse, the joint state of the signal and idler is the vacuum state. The coupling introduced by the interaction Hamiltonian drives the time evolution of the vacuum-mode signal and idler. Through time-dependent perturbation theory, this evolution can be expressed as [82]:

\[
|\Psi(t)\rangle \approx \exp \left[ -\frac{i}{\hbar} \int_{-\infty}^t dt' \hat{H}_{\text{int}}(t') \right] |0\rangle \tag{2.6}
\]

\[
|\Psi(t)\rangle \approx |0\rangle - \frac{i}{\hbar} \int_{-\infty}^t dt' \hat{H}_{\text{int}}(t') |0\rangle \tag{2.7}
\]

where we use the first-order approximation since the nonlinear interaction is assumed
to be weak. Also, we only consider the second term in the above expression, because in practice we post-select the non-vacuum events by single-photon detection. Since this process is mediated with a finite duration pump pulse, the limits of the time integral in Eq. (2.7) can be extended to infinity. Thus, we evaluate the time-dependent integral in Eq. (2.7) as

$$\int dt' \exp \left[ i(\omega_S + \omega_I - \omega_P)t' \right] = 2\pi \delta(\omega_S + \omega_I - \omega_P).$$

With this delta function, we can eliminate one of the frequency integrals to express the joint state as:

$$|\psi\rangle = \frac{2\pi \kappa}{i\hbar} \int d\omega_S d\omega_I \mathcal{E}_P(\omega_S + \omega_I) \hat{a}_S^\dagger(\omega_S) \hat{a}_I^\dagger(\omega_I) \times \int_{-L/2}^{L/2} dz \exp \left[ -i(k_S(\omega_S) + k_I(\omega_I) - k_P(\omega_P))z \right] |0\rangle \quad (2.8)$$

The last integral in Eq. (2.8) can be evaluated to yield the well-known phase-matching function, $\Phi(\omega_S, \omega_I)$:

$$\Phi(\omega_S, \omega_I) = \int_{-L/2}^{L/2} dz \exp \left[ -i(k_S(\omega_S) + k_I(\omega_I) - k_P(\omega_P))z \right] = -i \frac{L}{2} \text{sinc} \left( \frac{\Delta k(\omega_S, \omega_I)L}{2} \right) \quad (2.9)$$

where, $\Delta k(\omega_S, \omega_I) = k_S(\omega_S) + k_I(\omega_I) - k_P(\omega_P)$ is the wavevector mismatch. An additional contribution to this expression is necessary for the quasi-phase matching (QPM) scheme [83]. In QPM, the ferroelectric domain orientations are periodically modulated by electric field poling. This modulation with period $\Lambda$ induces an additional wavevector, $2\pi/\Lambda$, which can be viewed as a crystal momentum. We can consider the periodic modulation for an infinitely long crystal, where the sign of the nonlinear coefficient $\chi^{(2)}$ alternates in every half period. The longitudinal-position dependent nonlinear susceptibility can then be expressed as a Fourier series:

$$\tilde{\chi}^{(2)}(z) = \chi^{(2)} \sum_{l=-\infty}^{\infty} c_l \exp \left( -i \frac{2\pi l}{\Lambda} \right) \quad (2.10)$$

The square modulation in the sign of the nonlinear susceptibility introduces crystal
wavevectors with quasi-momenta of $\pm (2\pi l)/\Lambda$ with $l = 1, 3, 5, \ldots$. We typically use first-order quasi-phase matching where only $l = \pm 1 (c = 2/\pi)$ terms produce phase matching such that

$$\Delta k(\omega_S, \omega_I) = k_S(\omega_S) + k_I(\omega_I) - k_P(\omega_P) \pm \frac{2\pi}{\Lambda} \approx 0 \quad (2.11)$$

In that case, the effective crystal nonlinearity becomes $\chi^{(2)}_{\text{eff}} = \frac{2}{\pi} \chi^{(2)}$. We can also define an alternative effective nonlinear coefficient that is introduced by another convention of units, where $d_{\text{eff}} = \frac{4}{\pi} \chi^{(2)}$. In the later stages of the thesis, the nonlinearity of the QPM crystals will be quantified by $d_{\text{eff}}$.

Finally, we can write the output state for downconversion in a compact form:

$$\left| \psi \right> = \frac{2\pi \kappa}{\hbar} \int\int d\omega_S d\omega_I \tilde{A}(\omega_S, \omega_I) \hat{a}_S^\dagger(\omega_S) \hat{a}_I^\dagger(\omega_I) |0\rangle, \quad (2.12)$$

$$\tilde{A}(\omega_S, \omega_I) = E_P(\omega_S + \omega_I) \Phi(\omega_S, \omega_I), \quad (2.13)$$

where $\tilde{A}(\omega_S, \omega_I)$ is the joint spectral amplitude (JSA) for the two-photon state. We will also define $\left| \tilde{A}(\omega_S, \omega_I) \right|^2$ as the joint spectral density, which can be viewed as a two-dimensional probability distribution for the frequency content of the joint state. Thus, the fluorescence spectra for signal and idler are given by

$$S_S(\omega_S) = \int d\omega_I \left| \tilde{A}(\omega_S, \omega_I) \right|^2, \quad (2.14)$$

$$S_I(\omega_I) = \int d\omega_S \left| \tilde{A}(\omega_S, \omega_I) \right|^2. \quad (2.15)$$

In general, the spectra for signal and idler are different. Consequently, even if other degrees of freedom for signal and idler are kept identical, a simple spectrometer measurement will be sufficient to distinguish these photons. Such distinguishability is reflected in a Hong-Ou-Mandel interference experiment [2], where one would observe degraded quantum-interference visibility. We emphasize that JSA is central to the distinguishability aspect and the spectral entanglement of the two-photon state. Studying JSA and engineering two-photon spectral properties will be deferred to
In Fig. 2-2, we plot a sample JSA distribution for a realistic scenario where we consider pulsed pumped SPDC. In this configuration, we take an ultrafast pump input with a Gaussian spectrum centered at 792 nm. The pump spectral bandwidth is set to 6 nm, full width at half-maximum. The nonlinear crystal is a 5-mm long periodically-poled KTP (PPKTP) with a grating period of 46.15 μm. We consider degenerate type-II phase matching for SPDC where signal and idler photons emerge at ~1584 nm with orthogonal polarizations. The left panel in Fig. 2-2 shows the pump envelope, $\mathcal{E}_P(\omega_S + \omega_I)$, sketched in a contour plot with signal and idler wavelengths as variables. Similarly, we plot the phase-matching function, $\Phi(\omega_S, \omega_I)$, for PPKTP in the right panel.

![Figure 2-2: Pump envelope (left panel) and phase-matching function (right panel) for an ultrafast type-II SPDC with a 5-mm long PPKTP crystal. The pump is a pulsed laser with a Gaussian spectrum centered at 792 nm and 6 nm bandwidth (FWHM). The contour plots for both functions are plotted with signal and idler wavelengths as two input variables.](image)

The product of the two distributions gives the joint spectral amplitude for the given experimental arrangement, as shown in Fig. 2-3. Note that for this particular shape and distribution of the JSA, the signal and idler fluorescence spectra are identical. Also the diagonal orientation of the JSA results in positively correlated signal and idler wavelengths, $\omega_S = \omega_I$, which cannot be achieved with cw-pumped SPDC.
Figure 2-3: Joint spectral amplitude for ultrafast type-II SPDC with a 5-mm long PPKTP crystal.

2.3 Single-Photon Upconversion by Sum-Frequency Generation

We now consider sum-frequency generation (SFG), another three-wave mixing process and a well-known tool in nonlinear optics. SFG is typically used for frequency conversion of weak optical fields which is useful for studies in molecular spectroscopy, ultrafast phenomena and photodetection. SFG also find applications in quantum optics, since it was successfully utilized as a single-photon detection technique [75, 76, 84, 78, 77]. The main motivation behind this scheme lies with the quantum efficiency and speed of single-photon detectors at infrared wavelengths which are usually inferior to their visible counterparts. As an example, we can consider the low quantum efficiencies, high dark currents and afterpulsing rates of InGaAs avalanche photodetectors (APDs). Because of these limitations, InGaAs APDs are usually operated in Geiger mode with low duty-cycle gating. However, silicon APDs at visible wavelengths are
more efficient and faster. Thus, using SFG to upconvert infrared photons to visible wavelengths has been considered as a promising technique to perform efficient photon counting of infrared photons.

Let us examine the classical SFG for frequency upconversion in a highly nonlinear crystal, such as PPLN, with length \( L \). The three-wave mixing process for SFG has two classical input fields as sketched in Fig. 2-4. We neglect the propagation losses in the nonlinear crystal and also limit the geometry to longitudinal propagation along \(+\hat{z}\) direction. We consider a high-power, pulsed pump field at \( \omega_P \), polarized along the appropriate crystallographic axis for phase-matched interaction,

\[
\mathcal{E}_P(z, t) = \text{Re} \left[ \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} E_P(z, \omega_1) e^{i(k_P(\omega_P+\omega_1)z-(\omega_P+\omega_1)t)} \right]. \tag{2.16}
\]

![Figure 2-4: Single photon detection with sum-frequency generation in a \( \chi^{(2)} \) nonlinear crystal. The two input fields comprise a strong classical pump, \( \mathcal{E}_P(z, t) \), and a weak probe signal, \( \mathcal{E}_S(z, t) \), collinearly combined at a dichroic mirror (DM1). The upconverted output field, \( \mathcal{E}_U(z, t) \), can be separated with another dichroic mirror (DM2) and efficiently detected with visible wavelength photodetectors.](image)

As a usual approximation, the strong pump can be taken as undepleted, hence \( \frac{\partial}{\partial z} E_P = 0 \). \( \mathcal{E}_P(z, t) \) is collinearly combined with the weak signal field, \( \mathcal{E}_S(z, t) \), on a dichroic beam-splitter. We assume the signal field to be narrowband and centered around \( \omega_S \), with the correct polarization orientation for phase-matched interaction,

\[
\mathcal{E}_S(z, t) = \text{Re} \left[ \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} E_S(z, \omega_2) e^{i(k_S(\omega_S+\omega_2)z-(\omega_S+\omega_2)t)} \right]. \tag{2.17}
\]

The output field centered at \( \omega_U = \omega_S + \omega_P \) is separated from the strong pump beam by another dichroic beam-splitter. For classical SFG, the upconverter input-output relations are obtained through the well-known coupled mode equations, under
the slowly varying envelope approximation [83, 85]:

\[ \frac{\partial}{\partial z} E_S(z, \omega_2) = -i \frac{(\omega_S + \omega_2) \chi^{(2)}}{2n_S(\omega_S + \omega_2)c} E_P^*(\omega_1) E_U(\omega_1 + \omega_2) e^{-i \Delta k(\omega_1, \omega_2)z} \]  
\[ \frac{\partial}{\partial z} E_U(z, \omega_1 + \omega_2) = -i \frac{(\omega_U + \omega_2 + \omega_1) \chi^{(2)}}{2n_U(\omega_U + \omega_2 + \omega_1)c} E_P(\omega_1) E_S(\omega_2) e^{i \Delta k(\omega_1, \omega_2)z} \]

where the phase mismatch is

\[ \Delta k(\omega_1, \omega_2) = [k_P(\omega_P + \omega_1) + k_S(\omega_S + \omega_2) - k_U(\omega_U + \omega_2 + \omega_1)] z. \]  
\[ (2.20) \]

In the case of quasi-phase matching we also include the crystal wavevector, \( 2\pi/\Lambda \), in the phase mismatch expression. Note that, for a given frequency detuning in signal or pump, all terms other than the exponential factor in Eqs. (2.18) and (2.19) are slowly varying. This allows simplification of the coupled mode equations to the following form:

\[ \frac{\partial}{\partial z} E_S(z, \omega_2) = -i \frac{\omega_S \chi^{(2)}}{2n_S(\omega_S)c} E_P^*(\omega_1) E_U(\omega_1 + \omega_2) e^{-i (\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)z} \]  
\[ \frac{\partial}{\partial z} E_U(z, \omega_1 + \omega_2) = -i \frac{\omega_U \chi^{(2)}}{2n_U(\omega_U)c} E_P(\omega_1) E_S(\omega_2) e^{i (\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)z} \]

(2.21)

(2.22)

where we implicitly assumed that \( \Delta k(\omega_1 = 0, \omega_2 = 0) = 0 \) and used a first-order Taylor series expansion to obtain the frequency derivatives of the wavevector mismatch for narrowband detunings:

\[ \Delta k_1(\omega_1) = \left[ \frac{\partial k_P(\omega_P + \omega_1)}{\partial \omega_1} - \frac{\partial k_U(\omega_U + \omega_1)}{\partial \omega_1} \right] \]  
\[ \Delta k_2(\omega_2) = \left[ \frac{\partial k_S(\omega_S + \omega_2)}{\partial \omega_2} - \frac{\partial k_U(\omega_U + \omega_2)}{\partial \omega_2} \right] \]

(2.23)

(2.24)

For convenience, the field amplitudes can be expressed in terms of photon number.
units. Doing so facilitates the derivation of coupled-mode equations for quantized fields. This requires the following normalization factors to be applied [86]:

\[ E_j(\omega_j) = \sqrt{\frac{2\hbar\omega_j}{cn_j(\omega_j)\epsilon_0 A}} A_j, \quad j = P, S, U. \]  

(2.25)

where, \( A \) is the cross-sectional area of the nonlinear medium. Hence, the simplified form for the coupled mode equations can be expressed in terms of photon units as,

\[
\frac{\partial}{\partial z} A_S(z, \omega_2) = -i\kappa^*(\omega_1) A_U(\omega_1 + \omega_2) e^{-i(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)z} \tag{2.26}
\]

\[
\frac{\partial}{\partial z} A_U(z, \omega_2 + \omega_1) = -i\kappa(\omega_1) A_S(\omega_2) e^{i(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)z} \tag{2.27}
\]

for \( 0 \leq z \leq L \) and

\[
\kappa(\omega_1) \equiv \sqrt{\frac{\hbar\omega_P\omega_S\omega_U}{2\epsilon_0^2 n_P n_S n_U \chi^2 A P(\omega_1)}}. \tag{2.28}
\]

In Ref. [86], Eqs. (2.26) and (2.27) are solved by using Laplace transformation. We follow the same approach to obtain the input-output relations for arbitrary pump and signal detunings:

\[
A_S(L, \omega_2) = i\kappa^*(\omega_1)L \sin(qL) qL e^{iL(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)/2} A_U(0, \omega_1 + \omega_2)
\]

\[
+ \left( \cos(qL) - iL \frac{\Delta k_1\omega_1 + \Delta k_2\omega_2 \sin(qL)}{qL} \right) e^{iL(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)/2} A_S(0, \omega_2) \tag{2.29}
\]

\[
A_U(L, \omega_1 + \omega_2) = i\kappa(\omega_1)L \sin(qL) qL e^{-iL(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)/2} A_S(0, \omega_2)
\]

\[
+ \left( \cos(qL) + iL \frac{\Delta k_1\omega_1 + \Delta k_2\omega_2 \sin(qL)}{qL} \right) e^{-iL(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)/2} A_U(0, \omega_1 + \omega_2) \tag{2.30}
\]

where \( q = \sqrt{|\kappa(\omega_1)|^2 + ((\Delta k_1\omega_1 + \Delta k_2\omega_2)L/2)^2} \). We can define a frequency response for the upconverter that determines the bandwidth limitations. Using the classical input-output relations, we get,
\[ A_U(L, \omega_1 + \omega_2) = H(\omega_1, \omega_2) A_S(0, \omega_2), \tag{2.31} \]

where

\[ H(\omega_1, \omega_2) = i\kappa(\omega_1)L \frac{\sin(qL)}{qL} e^{-iL(\Delta k_1(\omega_1) \omega_1 + \Delta k_2(\omega_2) \omega_2)/2}. \tag{2.32} \]

The two main contributing factors for bandwidth limitations of the upconverter are the length of the nonlinear crystal and the material dispersion. The pump power also affects the operational bandwidth, but it is of secondary importance. The bivariate profile of this frequency response is essential for the upconverter design. As a practical example, we consider a broadband SFG process in 1-mm long periodically-poled stoichiometric lithium tantalate (PPSLT) crystal. The two broadly tunable input fields consist of a probe signal centered around 1580 nm and a strong pump around 790 nm. The type-0 collinear sum-frequency interaction can be phase matched for a grating period of 8.5 \( \mu \)m at \( \sim 55^\circ \)C. Using the Sellmeier equations for PPSLT [87], we plot \( |H(\omega_1, \omega_2)|^2 \) as a function of signal and pump wavelengths in Fig. 2-5.

The classical input-output relations also enable us to derive the upconversion efficiency. While it is possible to obtain an efficiency figure for the plane-wave case, a more relevant derivation would involve Gaussian beams with well-defined beam parameters. Such an efficiency estimate would be a useful guideline for comparing the experimental data with theoretically expected values. For SFG, the effective frequency conversion efficiency and optimal focusing conditions for Gaussian beams were derived by Boyd and Kleinman [88]. In this derivation, the beam focusing factor \( h_m(B, L/b_p) \) was parametrized by the refraction parameter \( B = \rho(Lk_P)^{1/2}/2 \) and the pump confocal parameter \( b_p = 2\pi w_n^2 n_P/\lambda_P \) with \( w_n \) being the pump beam waist. In our experiments, we mostly used quasi-phase-matched nonlinear crystals such as PPLN. Thus, if we consider SFG in PPLN or a similar QPM crystal, the double-refraction parameter, \( \rho \), can be neglected (\( B \approx 0 \)). We can further assume that the focusing for signal and pump beams are arranged to provide a mode-matched interaction, i.e. \( (b_p = b_s) \). The upconversion efficiency for a QPM crystal under
Figure 2-5: Two-dimensional bandwidth profile, $|H(\omega_1, \omega_2)|^2$, for sum-frequency generation plotted as a function of signal and pump wavelengths. We considered type-0 phase matching in a 1-mm long PPSLT. The tunable probe (pump) input was centered around 1580 nm (790 nm).

perfect phase-matching and mode-matching conditions with a nonlinear coefficient of $d_{\text{eff}}$ has been derived elsewhere [89, 90]:

$$
\eta = \frac{P_U}{P_S} = \frac{16\pi d_{\text{eff}}^2 P_P L}{n_P n_S n_U c \epsilon_0 \lambda_U^2} \frac{h_m(0, L/b_p)}{k_p^{-1} + k_s^{-1}}.
$$

The set of classical input-output relations in Eqs. (2.29) and (2.30) can be considered as a precursor to the quantum version. For efficient single-photon detection by frequency upconversion, it is necessary to show that the quantum state of the upconverted photon is preserved. The state variables in classical input-output relations have been given in terms of the photon-number units. This is convenient for the reason that the same input-output relations can be used for quantum-state frequency upconversion by introducing the quantized field operators [86]. In this procedure the classical field amplitudes are replaced with the positive-frequency field operators $\{\hat{A}_j(L, \omega_j), \hat{A}_j^\dagger(L, \omega_j)\}$. These operators satisfy the bosonic commutation relations:
\[
\begin{align*}
\left[\hat{A}_j(z,\omega_j), \hat{A}_k(z,\omega_k)\right] &= 0 \\
\left[\hat{A}_j(z,\omega_j), \hat{A}_k^\dagger(z,\omega_k)\right] &= 2\pi\delta_{jk}\delta(\omega_j - \omega_k) \quad \text{for } j, k = S, U. \quad (2.34)
\end{align*}
\]

Once we substitute the field operators for signal and upconverted modes, the resulting input-output relations are:

\[
\begin{align*}
\hat{A}_S(L, \omega_2) &= i\kappa(\omega_1)L\frac{\sin(qL)}{qL} e^{iL(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)/2} \hat{A}_U(0, \omega_1 + \omega_2) \\
&\quad + \left(\cos(qL) - iL\frac{\Delta k_1\omega_1 + \Delta k_2\omega_2\sin(qL)}{2}ight) \frac{qL}{e^{iL(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)/2}} \hat{A}_S(0, \omega_2), \quad (2.35)
\end{align*}
\]

\[
\begin{align*}
\hat{A}_U(L, \omega_1 + \omega_2) &= i\kappa(\omega_1)L\frac{\sin(qL)}{qL} e^{-iL(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)/2} \hat{A}_S(0, \omega_2) \\
&\quad + \left(\cos(qL) + iL\frac{\Delta k_1\omega_1 + \Delta k_2\omega_2\sin(qL)}{2}\right) \frac{qL}{e^{-iL(\Delta k_1(\omega_1)\omega_1 + \Delta k_2(\omega_2)\omega_2)/2}} \hat{A}_U(0, \omega_1 + \omega_2). \quad (2.36)
\end{align*}
\]

Note that this substitution is acceptable, since the output field operators, \(\{\hat{A}_j(L, \omega_j), \hat{A}_j^\dagger(L, \omega_j)\}\), satisfy the commutation brackets of Eq. (2.34). Without explicit substitution, we can verify this by observing that the input-output relations given by Eqs. (2.35) and (2.36) define a unitary transformation. It can be shown that the output field operators expressed by a unitary transformation can preserve the commutator brackets. We can immediately observe the isomorphism between these transformations and lossless beam-splitter equations, which couple input modes \(\{\hat{a}_{i,1}, \hat{a}_{i,2}\}\) to output modes \(\{\hat{a}_{o,1}, \hat{a}_{o,2}\}\):

\[
\begin{bmatrix}
\hat{a}_{o,1} \\
\hat{a}_{o,2}
\end{bmatrix}
= e^{i\theta}
\begin{bmatrix}
T & iR \\
iR & T
\end{bmatrix}
\begin{bmatrix}
\hat{a}_{i,1} \\
\hat{a}_{i,2}
\end{bmatrix}
\begin{cases}
|R|^2 + |T|^2 = 1 \\
RT^* - TR^* = 0
\end{cases}
\]

In this intuitive picture, the frequency of a mode can be associated with an input or output port of the beam-splitter, the reflectivity of which is given by \(|R|^2 = \sin^2(\kappa(0)L)\) for perfect phase-matching conditions. An analogous 4-port beam-splitter
device based on frequency upconversion is sketched in Fig. 2-6. The input modes are colored with respect to their frequency, in this case black represents the infrared photon, whereas green stands for the upconverted photon. The coupling between the two frequencies (colors) is provided by the pump (beam-splitter), which is colored in red.

\[
\begin{align*}
\hat{A}_U(0, \omega_1 + \omega_2) & \quad \text{to} \quad \hat{A}_U(L, \omega_1 + \omega_2) \\
\hat{A}_S(0, \omega_2) & \quad \text{to} \quad \hat{A}_S(L, \omega_2) \\
\theta(\omega_1, \omega_2), R(\omega_1, \omega_2), T(\omega_1, \omega_2) & \quad \text{in the beam-splitter}
\end{align*}
\]

Figure 2-6: Analogous beam-splitter representation for single-photon frequency upconversion. The input (output) modes are defined at the \( z = 0 \) (\( z = L \)) plane. The frequency dependent beam-splitter parameters \( \theta(\omega_1, \omega_2), R(\omega_1, \omega_2), \) and \( T(\omega_1, \omega_2) \) can be extracted from input-output relations.

The brief theoretical treatment here is presented for quantum-state frequency upconversion for single-photon detection in the visible range. This technique constitutes the backbone of the time-resolved upconversion method we present in Chapter 5 for mapping the temporal correlations of single photons. There, we also develop a continuous-time photodetection model that describes our measurement with broadband single-photon upconversion.
Chapter 3

Pulsed Sagnac Source of Narrowband Polarization Entangled Photons

3.1 Introduction

The motivation for secure storage and transmission of information has initiated the discipline of cryptography. The classical cryptographic protocols relied on information theoretical analysis to prove that the particular scheme is secure under every conceivable situation. As an example to a provably secure protocol, we can consider the “Vernam cipher” or one-time pad scheme [91]. In this scheme, two parties, conventionally named Alice and Bob, can communicate securely if they encode their data with a random ciphertext which was determined prior to encoding and used only once. A simple demonstration of this protocol is sketched in Fig. 3-1 where Alice can encode her secret binary message by adding a random cipher or key on an XOR gate. Bob, having the knowledge of the identical key, can recover the secret message by performing the same operation. Provided that the key lengths are sufficiently long and each key is used only once, the mutual information that an eavesdropper (Eve) is able to extract from the secret message can be made arbitrarily small [92].
Therefore, we can reduce the problem of secure communication to generating and distributing long and random bitstreams securely between Alice and Bob. With the exclusive knowledge of the key, they can follow the one-time pad protocol over any public or private channel for secure communication. However, the classical key distribution protocols have several drawbacks due to either insecure methods or impractical requirements, such as face-to-face meeting for key exchange.

### 3.1.1 BB84 Quantum Key Distribution Protocol

In 1984, Bennett and Brassard introduced a novel key distribution protocol that relied on the fundamental principles of quantum mechanics and measurement [4]. In this protocol, which is commonly abbreviated as BB84 after its creators, Alice and Bob use a true single-photon source. Alice encodes random binary data on the polarization of her photon in either one of two incompatible bases, horizontal (H)/vertical (V) or ±45° anti-diagonal (A)/diagonal (D). The random selection for the polarization basis can be performed by wave plates or electro-optic modulators. Bob measures the polarization of the received photon by randomly choosing from the same pair of incompatible measurement bases. For this purpose, he can use a combination of a half-wave plate and a polarizing beam-splitter as sketched in Fig. 3-2. Bob uses single-photon detectors to register his measurement result. After this process is repeated many times, Alice and Bob compare their measurement bases over a public channel, in which they discard the events belonging to opposite basis selections. Considering all possible polarizations, \( |H\rangle, |V\rangle, |A\rangle, |D\rangle \), an eavesdropper cannot...
discriminate the state of each photon perfectly. As a consequence, any intercept-and-resend attempt will result in a polarization state that is occasionally different from the original photon. A more fundamental argument for this situation is given by the no-cloning theorem, which dictates that an arbitrary quantum state cannot be copied [93, 94]. Therefore, the intervention of an eavesdropper would induce errors for which Alice and Bob’s results differ for the same basis selection. Both parties can identify these errors and follow classical error correction [95] and privacy amplification [96] protocols to distill a secret key, where the mutual information of an eavesdropper can be shown to decay exponentially. This key can then be used in the one-time pad protocol to establish provably secure communication without having full control over the communication medium [97, 98]. We have to point out that in practice one has to work with quasi-single-photon sources, noisy channels, propagation losses and detection inefficiencies. These non-ideal conditions require extensive studies to establish the security limits of a practical QKD system [99, 100, 101, 102].

Figure 3-2: BB84 quantum key distribution system. Here, Alice encodes a random bitstream into the polarization of single photons in either (H/V) basis or (A/D) basis. This encoding is achieved by changing the half-wave plate (HWP) orientation angle, $\theta_A$. Bob also selects his measurement basis randomly, by varying his HWP angle, $\theta_B$. After passing through a polarizing beam-splitter (PBS), the photon is detected by single-photon detectors.

3.1.2 Entanglement-Based Quantum Key Distribution

The BB84 scheme seeded many other variants in quantum cryptosystems. In 1991, Artur Ekert introduced another quantum key distribution scheme that relied on the quantum correlations between two entangled particles [5]. In this scheme, Alice and Bob share a maximally entangled pair of particles, where the correlation between these
particles cannot be described by classical physics \[103, 104, 105\]. Alice and Bob can perform a series of joint measurements on their shared pair of entangled particles. As in BB84, they reveal their measurement bases over a public channel, but not the measurement results. From matching measurement bases, they can generate a secret key, whereas for other basis combinations they calculate a classical parameter that can violate a local correlation model. Such a parameter calculation for a joint measurement scheme would be similar to the $S$-parameter in Clauser-Horne-Shimony-Holt-type Bell’s inequality violation \[104, 106\]. Note that, any measurement-induced disturbance of an eavesdropper is imprinted on these correlation results by virtue of the no-cloning theorem \[93, 94\]. Namely, for the undisturbed channel, Alice and Bob’s joint measurement results violate the classical correlation limits arising from the entanglement. Upon measuring a transmitted photon, the eavesdropper cannot re-establish the quantum correlation between Alice’s and Bob’s photon pair, since entanglement cannot be created by a local operation. Hence the joint measurement data for the disturbed scenario yields a different outcome that signifies an error to Alice and Bob.

Entanglement-based QKD (eQKD) can have several advantages over BB84-type systems. For instance, a polarization-entanglement based QKD system will not require any active modulation component for random measurement basis selection for Alice and Bob. This is due to the fact that the polarization of a single photon from a maximally-entangled pair is totally random before being measured. Hence, in principle, an eQKD system would not necessitate the use of a random number generator for data encoding or an electro-optic modulator. Secondly, we note that the eQKD scheme can be more resilient against photon number splitting (PNS) attacks \[54\]. For non-ideal single-photon sources, such as an attenuated laser, each pulse may contain more than one photon and the polarization state of these photons is identical. This gives Eve an opportunity to selectively filter one photon for her measurement and gain partial knowledge of the secret key \[99\]. Multiple pair generation is also an issue for eQKD schemes. However, these pairs are independent from each other, consequently Eve does not gain any information about the photon she does not measure.
Another advantage relates to the properties of entanglement. When a photon pair is characterized as highly entangled, it is certain that no distinguishing information exists in other degrees of freedom [102]. However, the encoding and basis selection procedure for a single-photon source in BB84 scheme can imprint distinguishing information to unused degrees of freedom such as momentum or frequency. This information could in principle be measured by Eve [54]. Furthermore, using an entangled photon source enables monitoring the degree of entanglement continuously by means of CHSH-type Bell’s inequality violation or other measurements to account for systematic drifts in the output state quality.

### 3.1.3 Experimental Realizations

We note that both BB84 and Ekert’s QKD protocols and their variants have been implemented by many groups. The first implementation of a proof-of-principle BB84-type QKD dates back to 1992 where the photons were transmitted over 30 cm free-space for successful key generation [107]. The length-scale for free-space BB84 or its variants had first been enhanced to kilometer range [108, 109, 53, 52] and then to tens of kilometers [24, 25]. The fiber-based BB84 protocols have also been demonstrated by several groups where the communication links improved from hundreds of meters [110] to beyond one hundred kilometers [111]. The main improvement over these length scales came from more efficient detectors, reduced propagation losses, effective beam control and compensation of time-varying effects such as temperature-dependent birefringence drifts or turbulence.

The entanglement-based QKD protocols have been demonstrated more recently. The first proof-of-principle experiment based on polarization entanglement focused on free-space propagation [112]. Progress in the entanglement quality, single-photon detectors and active polarization control enabled free-space link spans of almost 150 km [113]. We can also see similar achievements in fiber-based eQKD systems using either time-bin entanglement [16, 114] or polarization entanglement [115]. A detailed review of QKD implementations with various protocols can be found in Ref. [54].

As we motivated earlier, QKD is an active field with ongoing work related to both
theoretical and experimental aspects. The context of QKD is quite broad with many possible implementations for the protocol to be adopted, the type of photon source to be used and the specific communication channel to be utilized. We were specifically interested in the source design and practical limitations for entanglement-based QKD over free-space communication channels. Our motivation came from the remarkable improvement of entanglement quality and output flux from spontaneous parametric downconversion experiments [42, 50]. This progress was highlighted by the fact that entanglement generation from SPDC can be stable without active control and the experimental setup can be extremely compact. This paves the way for maintenance-free and field-deployable SPDC modules which can be employed for mobile free-space line-of-sight QKD links. We were also motivated by a cost-effective implementation, which would contain a high-performance yet inexpensive and compact pump source that can efficiently drive the downconversion process. In the following section we investigate the possibilities of interfacing state-of-the-art high-flux sources of polarization-entangled photons from SPDC with entanglement-based QKD for line-of-sight free-space communication links. A bottom-up approach will be pursued to understand the restrictions imposed by the free-space eQKD scheme. We will then focus on the design, implementation and characterization of the polarization-entangled photon source from a polarization Sagnac interferometer. We will also elaborate on the practical limitations of our design and suggest improvements for future implementations.

3.2 Design Considerations for a Free-Space eQKD Source

The design of a free-space QKD system is primarily dictated by the properties of the line-of-sight communication channel and the receiver subsystems. In order to maximize the key generation rate, the entangled photon pairs should be generated, transmitted and detected at wavelengths where the interplay between the atmospheric
channel loss and the quantum efficiency of the single-photon detectors is optimal. The research group headed by Richard Hughes of the Los Alamos National Laboratory investigated the achievable key generation rate for a free-space QKD system with a line-of-sight communication channel using Si avalanche photodiodes (APDs) in Geiger-mode operation [3]. Figure 3-3 shows that the maximum secret key rate is obtained at \( \sim 780 \) nm wavelength. At shorter wavelengths, the Si single-photon counters are more efficient but atmospheric scattering losses are worse, and at longer wavelengths, the efficiency of Si APDs drops significantly to more than offset by the lower atmospheric losses. Therefore, it is ideal to configure and drive a degenerate SPDC source that can produce both signal and idler in this optimal wavelength range.

![Secret Bit Yield vs Wavelength](image)

**Figure 3-3:** The secret key yield for a free-space BB84 protocol that uses highly attenuated laser pulses and Si APDs, after Ref. [3]. The line-of-sight channel losses and Si APD quantum efficiency determine the optimal transmission window at \( \sim 780 \) nm.

Since the free-space communication channel incurs a significant amount of noise due to ambient solar radiation, additional care has to be taken in adequate filtering to maintain a high signal-to-noise ratio (SNR). The free-space ambient background is spectrally broadband, therefore a narrowband operation for SPDC is preferred to provide spectral discrimination. To permit temporal discrimination, pulsed rather than
continuous-wave (cw) operation is preferred. In pulsed mode, one can synchronize the detection electronics with the system clock provided by the pulsed source. The timing shutter for the detection of single photons is expected to enhance the SNR over temporally constant background light. These considerations for the spectral and temporal filtering dictate a pulsed or quasi-cw pumped degenerate SPDC source with a narrowband collinear output. Narrowband operation calls for a long nonlinear crystal with a narrow phase-matching bandwidth, which also provides a higher photon flux. In order to take advantage of a long nonlinear crystal, we operate the downconverter in a collinearly propagating geometry to maximize the overlap in the crystal and to optimize the light collection at the output. Before discussing further details on the downconversion, however, we will focus on the design of the pump source for pulsed SPDC as the first component of the free-space eQKD scheme.

3.3 Pump Source Design and Characterization

For degenerate downconversion at $\sim 780\, \text{nm}$ with a narrowband output spectrum, it is necessary to use a pulsed pump source centered at $\sim 390\, \text{nm}$ that is capable of reaching high power levels. We estimate the necessary pump power level by assuming values of the downconversion efficiency and system losses typical of our setup. For instance, a cw ultraviolet (UV) pumped 1-cm long PPKTP crystal can generate approximately $10^5$ pairs per second for 1 mW of input power and within 1 nm output bandwidth [42]. In general, most QKD protocols using SPDC sources require low pair generation probabilities for a given generation/detection event to minimize multi-pair events that the eavesdropper can exploit. Single-pass SPDC generation for multi-spatiotemporal mode output follows Poisson statistics. Thus, occasionally more than one pair will be generated for a pump pulse. An excess rate of multiple pairs can increase the bit error rates for the QKD scheme [116]. In order to avoid multi-pair events for the same pump pulse, it is desirable to keep pair generation probability low. Since we aim for a high key generation rate without generating excess number of pairs, we prefer dense packing of the pump pulses in time to increase the generation duty cycle.
In other words, the pulsed source should be able to sustain a high repetition rate. Thus, one can take \( \sim 1\% \) pair generation probability per pulse and \( \sim 100 \text{MHz} \) pulse repetition rate as realistic numbers for a practical system. For spectral discrimination against the ambient background, we prefer the detection bandwidth to be very narrow. Given the current interference filter capabilities, it is reasonable to assume a detection bandwidth of \( \sim 0.1 \text{nm} \). Combining these modest estimates on the system parameters, we require that the UV pump source should have an average power of \( \sim 100 \text{mW} \) in order to provide \( \sim 1\% \) pair generation probability per pulse. The pump bandwidth should also be comparable to or less than the SPDC working bandwidth. The main reason for this requirement concerns the efficient utilization of the pump spectrum. A broadband UV pump would produce degenerate broadband photon pairs that will spill out of the detection bandwidth and reduce the system efficiency. In order to generate the majority of the pairs within the detection bandwidth, the pump source should have a spectrum that appears as a quasi-cw source with \(< 0.1 \text{nm} \) spectral width. For UV wavelengths, this corresponds to pulses on the order of \( \sim 25 \text{ps} \), which is still shorter than the typical Si APD risetime of 300 ps.

It is rather challenging to find a commercial pulsed UV source that can satisfy the above specifications. Due to the large band gap requirement, the solid-state gain material selection is rather limited for 390 nm. For GaN and InGaN based pulsed lasers, the average power hardly exceeds 20 mW [117, 118]. As an alternative scheme, one can consider one- or two-stage nonlinear frequency conversion with high peak power pulsed sources at long wavelengths. For instance, an actively modelocked picosecond Ti:sapphire laser at 780 nm was initially considered as a suitable alternative. However, such a system would require a bulky and expensive diode pumped solid-state laser at 532 nm. For a field-deployable pump source, a Ti:sapphire based implementation is complicated, alignment-sensitive, costly and very difficult to be configured in a compact package.

An alternative scheme is to use fiber lasers in the 1.55 \( \mu \text{m} \) range. The advantages of using a fiber based design are apparent from many perspectives. A fiber laser can be easily designed to occupy a small footprint. Since light is guided in the fiber
medium, it is practically free of alignment problems. Furthermore, one can use a master-oscillator power amplifier (MOPA) configuration with a pulsed fiber laser as a seed source for an erbium-doped fiber amplifier (EDFA). State-of-the-art EDFAs can deliver up to 100 W of average power with a linearly polarized output. Thus, for a narrowband pulsed input at 1.55 μm, one can easily achieve kilowatt-level peak power at the output. The amplified fiber laser output with kilowatt-level peak powers can drive two cascaded second harmonic generation (SHG) stages to first reach the near-IR and then the UV range of the spectrum. In order to achieve high conversion efficiency for both SHG stages one can use the quasi-phase matching (QPM) scheme to provide custom non-critical phase matching in a collinearly propagating configuration for the desired input and output wavelengths [119, 120]. QPM is enabled by the periodical poling of nonlinear ferroelectric crystals such as lithium niobate or potassium titanyl phosphate.

Comparing the aforementioned alternatives for the high power pulsed UV source design, we chose the more compact and more stable fiber-based MOPA configuration with two SHG stages [121]. The block diagram of the UV source is shown in Fig. 3-4. Our design approach envisions the master oscillator as a passively mode-locked fiber laser with 0.07 nm bandwidth at 1561.4 nm and 31 MHz pulse repetition rate. For the power amplifier, a 5-W polarization-maintaining (PM) EDFA from a commercial vendor was chosen. Our selection for the QPM crystals were magnesium-oxide-doped periodically-poled lithium niobate (MgO:PPLN) for the first SHG stage and periodically-poled potassium titanyl phosphate (PPKTP) for the second SHG stage. Since both SHG stages are in a collinear, single-pass configuration the overall source design is highly modular. This enables us to test individual QPM crystals without disturbing the beam alignment. With 5-W EDFA average power and a modest 5% frequency quadrupling efficiency estimate, we expect that our design can produce 250 mW of narrowband, pulsed output at 390.35 nm. The following four sections detail the construction and characterization of this source.
Figure 3-4: Block diagram of the UV source design. The modular design with a fiber-based MOPA and two-stage SHG with MgO:PPLN and PPKTP is expected to generate $\sim$250 mW average power to drive the SPDC setup.

3.3.1 Master Oscillator

The design choice for the seed source of the MOPA scheme is a passively modelocked fiber laser. Over the last couple of decades, ultrafast fiber laser technology has progressed to approach the performance of their bulk solid-state counterparts [122]. A typical fiber laser resonator construction can be very compact to build the device in a letter-size area, including the fiber-pigtailed 980 nm pump diode. There are three critical aspects of the fiber laser design: pulsing mechanism, narrowband intracavity filtering and cavity length scaling for a desired repetition rate.

Erbium-doped and erbium-ytterbium co-doped fibers are two possible choices as a gain medium for a fiber laser. Both types of fibers can be pumped with either a 980 nm or a 1480 nm diode laser through wavelength division multiplexers (WDM) and both can provide broadband gain in the 1550 nm regime. Typical small signal gain profile of the erbium-doped fiber peaks around 1535 nm when excited with a short wavelength pump laser (either 980 nm or 1480 nm). The gain at $\sim$1560 nm is about half of the peak gain with the exact ratio depending on the ion concentration and fiber manufacturing process. We obtained an erbium-doped fiber sample from a commercial vendor to measure the gain profile as a function of the signal wavelength. The experimental setup was similar to the one sketched in Fig. 3-5. The gain fiber was pumped with a 250 mW diode laser at 980 nm and the gain was measured using a low-power tunable infrared diode laser (1530 nm-1570 nm) that was injected together with the pump laser in a WDM fiber coupler. The unsaturated gain profile of the tested fiber is plotted in Fig. 3-6.

Erbium-ytterbium co-doped fibers offer the advantage of higher pump absorption due to the presence of ytterbium ions. The non-radiative energy exchange between ytterbium and erbium ions enables us to reach significant gain levels at 1550 nm.
Figure 3-5: Gain fiber testing experiment with a 980 nm pump laser and a 1559 nm diode laser combined in a WDM. The output of the WDM was spliced to the tested fiber and the optimal fiber length for 250 mW pump power was determined by the standard cut-back method.

with relatively short fiber lengths. Thus, the co-doping scheme is useful if there is a limitation on the length of the gain fiber. Along with other intracavity fiber components, minimizing the gain fiber length is important if the repetition rate is to be increased beyond 100 MHz [123].

Figure 3-6: Gain profile of a typical erbium-doped fiber. The injected pump (signal) power level was ∼250 mW (0.35 mW). The erbium-doped fiber length was 62 cm. The achievable gain at 1560 nm is ∼7 dB lower than the peak gain.

Since the exact gain and loss profile of rare-earth doped fibers from different vendors exhibit significant variation, it was necessary to individually obtain and test
sample fibers. Both erbium and erbium-ytterbium type gain fibers were obtained from various commercial vendors. The standard testing apparatus for gain measurement is sketched in Fig. 3-5. A 300-mW, 980 nm diode laser from JDS Uniphase was combined with a 20-mW, 1559 nm Alcatel fiber-pigtailed diode laser in a WDM combiner. The output port of the WDM combiner was spliced to the tested gain fiber. The opposite end of the gain fiber was cleaved and the output was filtered by an infrared (IR) bandpass filter, and we measured the amplified signal with a power meter. In order to assess the optimal fiber length for a given pump power level, the gain fiber was initially kept long and gradually cut back and cleaved into shorter pieces to find the balance between longitudinal pump absorption and propagation loss of the gain fiber. For four different gain fibers and 250-mW input pump power, the optimal lengths and corresponding gain values were determined and tabulated in Table 3.1.

<table>
<thead>
<tr>
<th>Gain Fiber</th>
<th>Input Signal Power [mW]</th>
<th>Optimal Length [cm]</th>
<th>Gain [dB]</th>
</tr>
</thead>
<tbody>
<tr>
<td>INO EY103 (Er)</td>
<td>4.8</td>
<td>21</td>
<td>0.03</td>
</tr>
<tr>
<td>INO EY105 (Er)</td>
<td>4.8</td>
<td>107</td>
<td>4.1</td>
</tr>
<tr>
<td>Nufern SM-EYDF (Er/Yb)</td>
<td>1.57</td>
<td>14.6</td>
<td>1.82</td>
</tr>
<tr>
<td>Liekki Er110</td>
<td>4</td>
<td>92</td>
<td>9.6</td>
</tr>
</tbody>
</table>

Table 3.1: Gain fiber measurement results

As Table 3.1 indicates, the optimal gain at 1559 nm was achieved by a very highly doped Liekki fiber with a reasonable length of 92 cm. In order to maintain a high repetition rate, it was necessary to keep all other intracavity fiber components as short as possible. Besides the gain fiber, the cavity design included a narrowband intracavity filter for narrowband output and a saturable absorbing Bragg mirror to provide pulsed operation. As a practical choice we preferred a ring type cavity configuration, because a standing-wave type cavity would inherently yield a lower repetition rate (by a factor of 2) with the same component lengths. The layout of the fiber ring cavity design is sketched in Fig. 3-7.

In order to achieve a narrowband fiber laser output (<0.1 nm) at a desired center wavelength, it was necessary to use a low insertion-loss intracavity filter. For this
purpose, a fiber Bragg grating (FBG) was an ideal choice. Within the filter bandwidth, many commercial FBG products can provide a very high reflectivity which reduces the intracavity loss. One can also control the pulse spreading due to the grating structure to minimize the group delay. This can be achieved by varying the grating apodization of the FBG filter. As a disadvantage, most FBG filters operate in reflection. For a ring cavity design, this necessitates integrating the FBG filter with a fiber circulator. A custom-designed FBG filter was obtained from Teraxion with a 25 GHz bandwidth (flat-top filter profile) and less than 50 ps/nm group delay. The test FBG filter had more than 90% reflectivity.

One can operate the laser in the pulsed regime with various gain-switching or modelocking techniques. The gain-switching scheme requires complex electronic control circuitry and hinders the compact cavity design. Passive modelocking scheme, on the other hand, is more advantageous. Using an intracavity saturable absorbing mirror (SAM), an ideal turn-key operation can be achieved with minimal space requirements. For a fiber laser, given the magnitude of intracavity gain and loss, the pulse energy can fluctuate significantly within one round-trip. This requires using a saturable absorber with high loss modulation to achieve self-starting modelocking. A commercial SAM from BATOP Optoelectronics was obtained with 25% saturable and 5% nonsaturable loss and 50 \( \mu \)J saturation fluence. The SAM was mounted on a one-inch aluminum plate and inserted in a cage mount for confocal configuration. In this configuration the intracavity light was directed to a fiber collimator via another fiber circulator and focused by an aspheric lens (4-mm focal length) onto the SAM. The retro-reflected light is coupled back into the cavity, following the reverse path, and exiting the output port of fiber circulator.

The highly-doped gain fiber, narrowband FBG and high-modulation SAM constitute the most critical components of the fiber laser design. Apart from these, the output coupling element was a fiber 90/10 coupler. The pump beam was coupled through a WDM in a counter-propagating configuration. Since the gain profile along the Er-doped fiber is not homogeneous, co-propagating and counter-propagation pulse gains can be different. Experimentally, better amplification was observed for
the counter-propagating case. The only free-space section in the cavity was the SAM holder stage, which was needed to accurately position the SAM for focusing.

Figure 3-7: Schematic of passively modelocked fiber laser in a ring configuration. Pulsed operation was provided by the SAM. We used an intracavity FBG filter to fix the center wavelength and output bandwidth of the fiber laser. Both SAM and FBG filter operated in reflection, and two fiber circulators were used to preserve the ring structure. SAM, saturable absorber mirror; FBG, fiber Bragg grating.

For testing laser alignment and modelocking, we monitored the laser output with a power meter and with an amplified InGaAs photodiode whose rf output was connected to a spectrum analyzer. After the proper alignment and confocal positioning of the SAM was achieved, cw lasing was observed for input pump power levels between 60 and 100 mW. As we increased the pump power beyond 100 mW the laser transitioned to Q-switched mode-locking rather than cw modelocking, because the pulse energy was not sufficient to saturate the gain. Above 275 mW of pump power, self-starting cw modelocking was observed. The output power level of the fiber laser is plotted in Fig. 3-8(a) as a function of the input pump power, where we indicate the observed modelocking regimes. Fig. 3-8(b) shows the rf spectra measured around 31.2 MHz for the Q-switched and cw modelocking regimes.
Figure 3-8: (a) Fiber laser output power as a function of pump power. The boundary between Q-switched modelocking (QSML) and cw modelocking (CWML) occurs at \( \sim 275 \text{ mW} \) pump power. (b) Rf spectrum of fast photodiode signal indicating the two pulsed regimes of the fiber laser. QSML operation (red-dashed trace) with the typical low-frequency modulation and CWML operation with a single frequency comb line (blue-solid trace) are shown.

The average output power of the laser at \( \sim 290 \text{ mW} \) input pump power was 1.8 mW. At this power level, the observed output spectrum of the fiber laser is plotted in Fig. 3-9. The repetition rate was adjusted to be around 31.125 MHz, a sub-multiple of a 1.25 GHz clock from a QKD test bed at the National Institute of Standards and Technology (NIST). This was achieved by shortening some of the passive fiber components and re-splicing them into the cavity. With fine length adjustments, we obtained a repetition rate of 31.2 MHz.

It is important to emphasize that the fiber laser was made of non-polarizing fiber components. Thus, when any fiber component’s position or bending was varied, the output beam polarization could change. Preserving the polarization state was crucial for the rest of the pump source. A more robust cavity design would result if all intracavity components were made of PM fibers. However, we observed that as long as the fiber laser components were kept motionless, the output polarization would stay constant. Thus, even if the output polarization state was elliptical, it would be corrected into a linear polarization state and coupled into a PM fiber. In order to obtain a well-defined output polarization we used the arrangement sketched in Fig. 3-10. With an inline polarization controller and fiber polarization splitter, we sent the output of the fiber laser into two PM fibers for guiding the two orthogonal polarizations. By adjusting the polarization controller, we directed most of the output
power to the PM EDFA and a small amount to the tap fiber from which its optical/rf spectrum could be monitored. After these adjustments were made, the laser power for seeding the EDFA was observed to be almost constant for many months. Even though the laser design inherently lacked polarization stability, it provided turn-key operation in the absence of mechanical perturbations. Recently, we constructed a second generation of the fiber laser with all PM components and we utilized this source to directly seed the PM EDFA without additional polarization controllers. Finally, the timing jitter of the free-running laser was characterized. We measured the laser output with a high-speed photodetector showing a stable rf-comb, where the single-sideband phase noise measurement (integrated from 100 Hz to 2.9 MHz) showed a residual timing jitter of 2.7 ps.

### 3.3.2 Polarization-Maintaining EDFA

To achieve the high peak power levels necessary for two-stage SHG the output of the passively modelocked fiber laser should be amplified with a high power EDFA
Figure 3-10: Polarization controlling stage for coupling the non-PM output of the fiber laser into two PM fibers. An inline polarization controller was used with a polarization beam splitter. The orthogonal polarization outputs of the splitter were coupled into PM fibers, one of which was connected to the EDFA and the tap output was used for monitoring purposes.

with minimal pulse distortion. In the experiment, a 5-W polarization-maintaining EDFA manufactured by IPG was used as the power amplifier. The EDFA had a PM fiber input and a collimated free-space output with a 4-mm Gaussian beam diameter. Initial tests with the EDFA showed excessive self-phase modulation (SPM). In Fig.3-11(a) we plot the EDFA output spectrum as a function of the EDFA pump current. At high power levels, the EDFA spectrum exhibited clear splitting that was undesirable for the system performance.

Silica-based optical fibers are known to have non-zero $\chi^{(3)}$ susceptibilities. When high peak power levels are attained in the fiber core, nonlinear effects start distorting the pulse spectrum through SPM [124]. We monitored the amplified fiber laser spectrum to keep track of the spectral broadening effect of SPM because of the need to maintain a narrow bandwidth for our QKD application. To gain insight into the high-intensity pulse propagation in an optical fiber one can calculate the SPM-induced peak nonlinear phase shift [124]. For instance, for an average power of 5W in the EDFA, the corresponding peak power for a 50-ps Gaussian pulse is $\sim3$ kW. If the amplified pulse propagates $L = 1$ meter in a standard SMF-28 silica core fiber, the peak nonlinear phase shift can be calculated as:

$$\phi = \gamma P_{\text{peak}} L = \frac{n_2 \omega_0}{c A_{\text{eff}}} P_{\text{peak}} L \approx \pi,$$

(3.1)

where, $n_2$ is the nonlinear refractive index ($2.2 \times 10^{-20}$ m$^2$W$^{-1}$), $\omega_0$ is the angular center frequency, $c$ is the speed of light and $A_{\text{eff}}$ is the effective mode area in the
fiber. A nonlinear phase shift of $\pi$ is large enough to indicate spectral splitting, although the final shape of the output spectrum also depends on the initial pulse chirp. Yet, this is a useful calculation that illustrates the importance of minimizing the length of passive fiber components to avoid spectral broadening. A full numerical simulation, if necessary, can also be made by including the effects of gain and the actual fiber core structure. One can then use the split-step Fourier method to predict the pulse evolution in time and frequency [124]. The EDFA amplification details and fiber parameters for such a simulation were not readily available. Instead we pursued a trial and error approach with the vendor to modify the EDFA and to minimize the detrimental effects of SPM.

For improved SPM performance, we made several critical modifications on the EDFA. We removed an intermediate amplifier stage, an isolator stage and some lengths of passive fiber including the output collimator cord, which was shortened as much as possible. The length of the large-mode active fiber with a $15 \mu m$ effective mode diameter was cut by half to 1.5 m. A shorter gain fiber would give rise to a more pronounced amplified spontaneous emission (ASE) noise centered mainly at 1535 nm. Nevertheless, the broadband ASE power never exceeded 1% of the output power, even at the highest gain regime. After these measures were taken, the EDFA spectra exhibited less broadening, as shown in Fig. 3-11(b).

![Figure 3-11: EDFA output spectrum measurements (a) before and (b) after modifications. Each trace on the plot was recorded at the indicated EDFA current.](image)

Note that, these measurements were taken when a 1560-nm FBG was installed in the fiber laser. At a later stage of the experiment, we changed the center wavelength
to 1561.4 nm. This change was imposed by the phase-matching temperature range of the type-0 PPKTP crystal. Switching to a longer wavelength enabled us to work with the PPKTP crystal near room temperature. This wavelength shift required a new FBG filter to be ordered and installed in the fiber laser. The new FBG filter ended up being 25% broader in bandwidth, which corresponded to shorter pulses and higher peak power levels. The output EDFA spectra for the modified fiber laser input are plotted in Fig. 3-12, which shows more pronounced spectral splitting. However, the overall performance of the EDFA was still acceptable for driving the subsequent SHG stages.

### 3.3.3 First SHG Stage with MgO:PPLN

The collimated EDFA output was aligned such that the output polarization was vertical, which corresponded to the z-axis of the first SHG stage using a 10-mm long MgO:PPLN crystal. From the MgO:PPLN Sellmeier equations, type-0 phase matching for SHG at 780.7 nm requires a crystal poling period of 19.47 $\mu$m at around 90$^\circ$C crystal temperature. The 10-mm long MgO:PPLN crystal from HC Photonics was housed in a crystal oven that was temperature controlled with 0.1$^\circ$C accuracy. We used a tunable IR diode laser for cw SHG characterization of the crystal, indicating that for the 10-mm long MgO:PPLN crystal, the wavelength and temperature phase-matching bandwidths were 1.3 nm and 9.8$^\circ$C, respectively. The effective crystal non-linear coefficient, $d_{eff} = \frac{2}{\pi} d_{33}$ was measured to be 13 pm/V. The crystal temperature for peak SHG efficiency at 1561.4 nm was 93.8$^\circ$C. After cw characterization, to confirm its phase matching and efficiency parameters the crystal was dual-wavelength anti-reflection (AR) coated for 780 nm and 1560 nm.

We focused the pulsed output of the EDFA with a 10-cm focal length plano-convex lens into the dual-AR coated MgO:PPLN crystal and the beam diameter at the focal plane was measured to be $\sim$56 $\mu$m. The generated SHG output was collimated with a 20-cm focal length plano-convex lens (AR-coated for near-IR wavelengths) and separated from the fundamental by dichroic mirrors. The output at 780.7 nm had a spatial mode that was very close to an ideal TEM$_{00}$-mode with negligible ellipticity.
Figure 3-13 shows the measured SHG output power and SHG conversion efficiency as a function of input EDFA power. The conversion efficiency exceeded 50% for pump inputs above 2 W with a maximum efficiency of 68%. At a later stage in the experiment, the EDFA focusing lens was replaced with a 25-cm plano-convex lens that focused the pump to a beam diameter of \( \sim 140 \mu m \). The resulting conversion efficiency was comparable with the 10-cm focusing lens results. The longer focal length lens improved the thermal loading of the crystal and output power stability of the source with \( \sim 1\% \) power fluctuations.

![Figure 3-12: Measured spectra of laser output centered at \( \sim 1561.4 \text{ nm} \) and its SHG output at different EDFA power levels of (a) 1.3 W, (b) 3.4 W, and (c) 4.5 W. Dotted curve is the fiber laser output before EDFA.](image)

The spectra of the amplified fiber laser and the first SHG output are plotted in Fig. 3-12. Note that, due to saturation effects, the splitting in the SHG spectra becomes less pronounced. We also constructed an autocorrelation setup for the temporal characterization of the pulses from the fiber laser and the first SHG output. Since the expected pulse durations were on the order of \( \sim 100 \text{ ps} \), a standard background-free intensity autocorrelation measurement was sufficient. This measurement scheme employed noncollinear SHG in a broadband nonlinear crystal with time-delayed copies.
of the fundamental [125]. For the fiber laser output, a type-II phase matched PPKTP crystal with \( \sim 70 \) nm phase-matching bandwidth (\( \Lambda=46.1 \mu m \)) was used. The intensity autocorrelation measurement for the fiber laser using a Gaussian pulse shape revealed a pulse duration of 75 ps and a time-bandwidth product of 0.55. A similar measurement for the first SHG output was performed with a 1-mm long BBO crystal (\( \theta=30 ^{\circ} \) cut for normal incidence). At 2 W average output power, a pulse width of 49 ps was measured yielding a time-bandwidth product of 2.6. Both autocorrelation traces are sketched in Fig. 3-14.

![Figure 3-13: SHG output power at 780.7 nm with 10 mm long PPLN crystal at 93.8°C and 56 \( \mu m \) beam diameter. Corresponding SHG Efficiency is plotted against the right axis.](image)

The noticeable increase in the time-bandwidth product for the EDFA output was due to spectral broadening inside the amplifier. Both autocorrelation measurements necessitated long translation stages for tracing the complete autocorrelation peak. This situation is especially evident for the fiber laser autocorrelation measurement, where the one-inch travel range of the translation stage could only capture the upper half of the autocorrelation peak.
Figure 3-14: Measured intensity autocorrelation and Gaussian fits for laser output at 1560 nm and first SHG stage output at 780 nm. Fiber laser output gives 75 ps pulses, whereas first SHG stage output gives 49 ps pulses at FWHM, assuming a Gaussian pulse shape. The data points are collected only near the center for the fiber laser output due to the limited range of the translation stage.
3.3.4 Second SHG Stage with PPKTP

For the second SHG stage we initially planned to use a 10-mm long PPKTP crystal, designed for type-0 phase matching for a fundamental wavelength of 780 nm. It is worth noting that most of the transparent nonlinear crystals in the visible range start absorbing significantly at the ultraviolet end of the spectrum. Due to Kramers-Kröning relations, an absorption resonance increases the refractive index gradients. Due to this index run-off and wavelength scaling, the first-order quasi-phase matching condition is usually satisfied with a much shorter poling period. For instance with 780.7 nm fundamental input, first-order type-0 phase matching for PPKTP requires a poling period of 2.95 μm at room temperature. Such short poling periods can be difficult to fabricate and SHG efficiency may suffer as a result of poor poling quality. However, a more critical aspect of KTP turns out to be the UV-induced absorption and related photochromic damage, as explained below.

KTP is a nonlinear crystal with two well-established growth techniques, the flux method and the hydrothermal method. These growth techniques are known to affect certain absorption properties of KTP, most famous of which is gray-tracking problem usually observed in SHG and optical parametric oscillator (OPO) operation with high power Nd:YVO$_4$ lasers [126, 127, 128, 129, 130]. Due to the presence of impurities, defects and color centers in KTP, the intensity-induced absorption is spectrally inhomogeneous and strongly wavelength dependent. For instance, absorption lines at 420 nm and 510 nm can be associated with the formation of Ti$^{3+}$ and Fe$^{2+}$ color centers [131]. To understand the possible effects of high intensity UV generation at 390.35 nm, periodically-poled samples from both growth techniques were obtained and tested under cw and pulsed pumping.

We first obtained a flux-grown PPKTP sample from Raicol. Raicol was able to pole KTP with short periods and a first-order type-0 grating was ordered with a 2.95 μm poling period. We first tested the Raicol crystal with a cw, tunable Ti:sapphire laser. Around 780.7 nm fundamental input, ~0.06 nm phase-matching bandwidth was measured at 22°C crystal temperature, and the measured tempera-
ture bandwidth was 0.9°C. The crystal was poled very homogeneously and yielded a nonlinear \(d_{\text{eff}}\) coefficient of 7.6 pm/V, compared with the expected \(d_{\text{eff}}\) of 9.8 pm/V. The crystal was then tested under pulsed pumping with the 780.7 nm first SHG output. We observed that the flux-grown PPKTP exhibited catastrophic damage at high intensities. With 1 W pulsed input focused into a \(\sim 100 \mu m\) spot size, the UV power at the output only went up to 45 mW and decayed to 10 mW within minutes. During this period, significant spatial mode distortions developed and eventually a permanent photochromic damage along the beam path was observed. A looser focusing with a \(\sim 250 \mu m\) spot size did not cause observable damage at such a short time scale, yet the output power barely reached 50 mW with 3 W input power. Thus, given the long term spatial mode and output power degradation, this particular flux-grown sample was not useful for generating high power 390-nm light.

Additional PPKTP samples were obtained from our collaboration with the Air Force Research Laboratory at Wright-Patterson Air Force Base and AdvR, Inc. These samples were either hydrothermally-grown or high gray-tracking resistant flux-grown (HGTR) for type-II phase-matched SHG. The type-II scheme is inferior to type-0 since it uses the nonlinear coefficient \(d_{24}\) as compared to \(d_{33}\) for type-0 phase matching. For a typical KTP crystal, \(d_{33}\) value is \(\sim 3-4\) times larger than \(d_{24}\). Previous testing with flux-grown PPKTP samples poled for type-0 and type-II phase matching indicated a 9-to-1 output power factor. However, as compared to a first-order type-0 grating, a type-II grating is easier to fabricate with typical poling periods on the order of 7.6 \(\mu m\). A total of three HGTR and one hydrothermal type-II PPKTP crystals were characterized with the tunable cw Ti:sapphire source. We measured all relevant crystal parameters such as the effective nonlinearity, the phase-matching bandwidth and the phase-matching temperature. These measurement results are summarized in Table 3.2:

We also tested these uncoated samples under pulsed pumping. The focusing and the collimating lenses were both 75-mm focal length plano-convex type, broadband AR-coated for near-infrared and ultraviolet ranges, respectively. For all samples, the long-term UV output power variation for \(\sim 2.8\) W input pump power was recorded.
and they are plotted as a function of time in Fig. 3-15. All the flux-grown PPKTP samples exhibited fast degradation in their output powers in a short time scale and a slower decay in the long time scale, whereas the hydrothermally-grown PPKTP sample generated more steady output. Ripples in SHG power were caused by thermal fluctuations and their amplitudes can be reduced with better temperature control. Although the effective nonlinearity and the poling quality of the hydrothermally-grown PPKTP was not optimal, the output power and long term stability performance readily surpassed the flux-grown samples. A similar hydrothermally-grown PPKTP crystal with 22-mm length and improved grating structure yielded a much better $d_{\text{eff}}$ of 2.4 pm/V. However, due to the length of the crystal, the phase-matching bandwidth was prohibitively low (0.042 nm). Thus, this crystal was cut and polished into two sections (10-mm and 12-mm long) and dual-AR coated for 390.35 nm and 780.7 nm. For 2.15 W pulsed input power at 780.7 nm (3.3 W EDFA power), we measured a SHG power of 410 mW. This corresponded to a $\sim$5.5% frequency quadrupling efficiency. The spatial mode was very close to an ideal TEM$_{00}$, and the long term output power was stable. We observed that the achieved power level and output stability of the UV source was satisfactory for driving our narrowband pulsed SPDC.

As a final note, the second pulsed SHG stage was also tested with a 10-mm long 1%-mol MgO-doped periodically-poled stoichiometric lithium tantalate (MgO:PPSLT). MgO:PPSLT is a promising candidate for UV applications due to its extended UV transparency range [119]. For third-order type-0 phase matching ($\Lambda=8.5 \mu m$, $T=91.5^\circ C$) a 10-mm MgO:PPSLT yielded a 0.05 nm phase-matching bandwidth. Under pulsed

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Length [mm]</th>
<th>$d_{\text{eff}}$ [pm/V]</th>
<th>Phasematching Bandwidth [nm]</th>
<th>Temperature at 780.7 nm [°C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>HGTR-1</td>
<td>10</td>
<td>1.55</td>
<td>0.085</td>
<td>58.6</td>
</tr>
<tr>
<td>HGTR-2</td>
<td>10</td>
<td>1.2</td>
<td>0.179</td>
<td>57.1</td>
</tr>
<tr>
<td>HGTR-3</td>
<td>10</td>
<td>1.5</td>
<td>0.090</td>
<td>63.2</td>
</tr>
<tr>
<td>Hydrothermal</td>
<td>14</td>
<td>1</td>
<td>0.112</td>
<td>94.6</td>
</tr>
</tbody>
</table>

Table 3.2: Type-II PPKTP Sample Testing.
Figure 3-15: Long term high power pulsed SHG outputs for various crystals. FG: Flux-grown, HT: Hydrothermally-grown. For all FG-PPKTP samples, the input pump power was 2.5 W, for HG-PPKTP the pump power was 2.8 W.
pumping, the same crystal reached 140 mW with 2.5 W input power. However, its long term output power test showed signs of photorefractive damage, which may have resulted from the low MgO concentration and a nonideal (50/50) stoichiometry ratio.

### 3.3.5 Future Design Considerations

Learning from our experience in laser design, nonlinear crystal testing and performance evaluation, it is instructive to emphasize several key points for future implementations of the UV source. First of all, the current passively modelocked fiber laser design does not offer the flexibility of varying the pulse period easily. However, it may be quite advantageous to be able to vary the pulse repetition rate to study the variation of multi-pair generation probability. There are several possible methods to implement a UV source with variable repetition rate. A hybrid (active-passive) modelocked fiber laser that employs an intracavity electro-optic modulator is one alternative [132, 133]. This scheme has the disadvantage of reduced pulse energies for subsequent SHG stages for a given average EDFA power. With a more efficient SHG process, the conversion efficiency loss due to the reduced pulse energy of the hybrid modelocking scheme can be alleviated. Another approach involves repetition rate scaling after the UV generation by passive beam splitting and combining with the appropriate delays. However, this alternative also involves significant power level reduction and alignment sensitivity due to the spatial length of the pulse period (∼9.7 m).

It is worth noting that given the resilience of the hydrothermally-grown PPKTP against UV-induced absorption, a type-0 first order grating of hydrothermally-grown PPKTP could potentially exceed 1 W output power level. Scaling up the power of UV source is especially attractive for driving multiple downconverters, which may be useful for on-demand single photon sources [134].
3.4 Polarization Sagnac Interferometer for Pulsed Entangled Photon Source

The second half of the pulsed polarization entangled photon source design focuses on the generation of high-quality entanglement from an SPDC process. With abundant UV power in a well-defined spatial mode, one can adjust the necessary parameters for the SPDC system for high-purity and high-flux generation and detection of polarization entangled photon pairs. The generation of polarization entanglement requires interferometric combination of two downconverter outputs. To date, polarization entanglement has been realized in a number of interferometric configurations and by using different crystals [45, 44, 46, 43, 47, 135]. Performance comparison between different implementations can be achieved by examining several metrics such as the quantum-interference visibility for a given beam divergence angle, source brightness per unit detection bandwidth per unit pump power, etc. However, another important and rather qualitative attribute of the source concerns interferometric stability of the output quantum state against environmental perturbations. For a source that is targeted towards field-deployable free-space applications, it is desirable that the output state phase stability should be an inherent property of the system rather than an actively-controlled one.

The Sagnac configuration is a single-crystal implementation of interferometrically combined outputs of two identical, coherently-driven downconverters [136, 1]. A bidirectionally pumped polarization Sagnac interferometric (PSI) SPDC source offers the required output state stability. The PSI configuration eliminates the need for path-length stabilization through its common path arrangement and it is immune to path-length perturbations [42]. Moreover, a particular phase of the output state can be chosen by simply adjusting the relative phase between the horizontally (H) and vertically (V) polarized components of the pump. This can be seen by tracking the individual phase terms for clockwise (CW) and counter-clockwise (CCW) beam paths. As Fig. 3-16 indicates, the horizontal (vertical) component of the pump travels clockwise (counter-clockwise) in the PSI interferometer. Following the analysis in [42],
one can write the unnormalized output quantum state \( |\Psi\rangle = |\Psi_{CW}\rangle + |\Psi_{CCW}\rangle \) in terms of the superposed amplitudes of the two downconverter outputs, by following the beam paths.

For an initial relative phase, \( \phi_P \), between the \( H \) and \( V \) components of the input pump, one can transfer this phase difference to the downconverter outputs. The pump phase is imprinted on the generated signal and idler photon pair [137]: \( \phi_P = \phi_S + \phi_I \). After labeling the output ports of the interferometer as A and B, one can express the unnormalized output states of the two downconverters as follows:

\[
|\Psi_{CW}\rangle = e^{i(\pi/2 + \phi_P + k_P l_1 + (k_S + k_I) l_2 + \phi_{D,S} + \phi_{D,I})} \eta_{CW} E_H |H_{S,A}\rangle |V_{I,B}\rangle 
\]

(3.2)

\[
|\Psi_{CCW}\rangle = e^{i(\pi + \phi_P + k_P l_2 + (k_S + k_I) l_1 + \phi_{D,P})} \eta_{CCW} E_V |V_{S,A}\rangle |H_{I,B}\rangle 
\]

(3.3)

where \( \phi_{D,X} (X = S, I, P) \) accounts for the phase acquired by \( X \) when the beam passes through the dual-wavelength half-wave plate. \( E_H \) and \( E_V \) stand for pump field amplitudes for the \( H \) and \( V \) components, respectively. For the CCW term in Eq. (3.3), the fixed phase factor of \( \pi \) is produced by the reflection of the pump beam and the signal photon at the polarizing beam-splitter. Similarly, for the CW term, a \( \pi/2 \) phase factor is incurred by the reflection of the idler photon. The generation efficiencies for both paths and the beam propagation losses are encapsulated by \( \eta_{CW} \) and \( \eta_{CCW} \).

Note that, the phase accumulated for propagation through PPKTP is common for both terms and left out as part of the inconsequential global phase. An important simplification for calculation of the relative phase between \( |\Psi_{CW}\rangle \) and \( |\Psi_{CCW}\rangle \) is obtained by noting that in free space \( k_P = k_S + k_I \). This enables us to eliminate some of the terms in the relative phase to obtain \( \phi_{PSI} = \phi_{D,P} - \phi_{D,S} - \phi_{D,I} + \pi + \phi_P \). All terms in this expression are fixed except \( \phi_P \). By factoring out the global phase terms, we can write the unnormalized output state as:

\[
|\Psi\rangle = |H_{S,A}\rangle |V_{I,B}\rangle + e^{i\phi_{PSI}} \frac{\eta_{CCW} E_V}{\eta_{CW} E_H} |V_{S,A}\rangle |H_{I,B}\rangle
\]

(3.4)

Since the interferometer’s length-dependent terms drop out from the relative phase
expression, it is obvious that mechanical perturbations do not change the output state. In order to generate a desired Bell state from the PSI configuration, for instance a singlet state, the required conditions are $\phi_{\text{PSI}} = \pi$ and $\eta_{\text{CCW}} = \eta_{\text{CW}}$. By controlling the orientation of the half-wave plate and quarter-wave plate at the input of the PSI source, these conditions can be satisfied.

Figure 3-16: Phase-stable polarization Sagnac interferometer pumped with a UV source. For phase stability the clockwise and the counter-clockwise SPDC outputs have a fixed relative phase difference which can be externally controlled by adjusting the input pump polarization. The relative phase between orthogonally polarized pump components is controlled with a half-wave plate (HWP) and quarter-wave plate (QWP) combination. PBS, polarizing beam splitter; DM, dichroic mirror; DHWP, dual-wavelength half-wave plate.

The PSI design eliminates the need for spatial (aperture), spectral (interference filter), and temporal (timing compensator) filtering because the two downconverter outputs are completely indistinguishable [44] due to the common-path arrangement. In principle, all the output photons are strongly polarization entangled without the need for filtering, thus leading to a much higher generation efficiency than that of other approaches.

A previously developed cw SPDC source of polarization-entangled photons using the PSI configuration was efficient and yielded high visibility two-photon quantum
interference [42, 50]. The work of Kim et al. was the first exploration of PSI configuration in pursuit of generating high-flux polarization entangled photons with minimal filtering requirements and with the capability of performing the complete Bell-state measurements through hyper-entanglement [42]. Pulsed operation of a Sagnac interferometer was realized by Shi and Tomita in a scheme where the generation of entanglement was limited by the use of a non-polarizing beam splitter [48]. The non-polarizing Sagnac scheme produces polarization-entangled photons at a 50% success rate, which the PSI design overcomes. The remaining sections in this chapter detail the integration of a PSI SPDC configuration with the developed UV source and summarizes the testing and characterization results in the low and high photon flux regimes and their implications on the output quantum state purity.

3.4.1 Experimental Setup

Fig. 3-17 shows the experimental setup for the pulsed Sagnac source together with the measurement apparatus. For the pulsed PSI source of polarization entangled photons, we used a 10-mm long flux-grown PPKTP from Raicol as the nonlinear crystal. This crystal was poled with a grating period of 7.85 μm for type-II phase matching and degenerate wavelength output at 780.7 nm. The crystal phase-matching bandwidth and nonlinearity was characterized by type-II SHG measurements with a tunable cw Ti:sapphire source. With 305 mW input pump power at 790.54 nm and a 24°C crystal temperature, 95 μW of SHG power was obtained, which yielded a crystal nonlinear coefficient $d_{\text{eff}}$ of 2.4 pm/V. We show the wavelength tuning curve for type-II SHG characterization of a 10-mm PPKTP crystal in Fig. 3-18. The measured phase-matching bandwidth for SHG at a constant temperature was 0.084 nm. The required phase-matching temperature for degenerate emission with a 390.35 nm pulsed pump was 28.6°C, which was controlled with a thermoelectric heater with a temperature stability of better than 0.1°C.

Given the observation of crystal damage in flux-grown crystals in our UV source development, the 10-mm crystal was first tested for any observable damage with the UV source at high power. When the UV input was focused into a ~50 μm spot size
Figure 3-17: Experimental setup showing the bidirectionally pumped polarization Sagnac interferometer (PSI) in the dashed box. The generated polarization-entangled signal and idler outputs are analyzed in coincidence measurements under different operating conditions. IF, interference filter; PBS, polarizing beam splitter; DM, dichroic mirror; HWP, half-wave plate; QWP quarter-wave plate; DHWP, dual-wavelength half-wave plate.
within the crystal for various power levels, no degradation in transmission or spatial mode quality was observed. Therefore, it appeared that the flux-grown PPKTP worked well for downconversion for pulsed UV pumping up to a maximum average power of $\sim 100 \text{ mW}$.

For driving the PSI source with the UV pump, it was necessary to filter the UV pump source to remove the residual 780.7 nm light coming from the first SHG stage, which would contaminate the SPDC output at the same wavelength. This filtering was accomplished by a multi-bounce arrangement of two dichroic mirrors (high-reflecting at 390.35 nm, high-transmission at 780.7 nm). After a total of 7 bounces on these mirrors, the estimated suppression for the residual 780.7 nm beam was 140 dB. This multi-bounce two-mirror filtering was sufficient to reduce the residual 780.7-nm light in the UV pump to a level low enough that no noticeable counts were measured when the downconversion crystal was removed.

The PSI, as shown in the dashed box of Fig. 3-17, was composed of two flat mirrors which were high-reflecting both at the pump and downconversion wavelengths and a PBS that served as the input and output optical element. The Sagnac configuration calls for an input/output PBS that works at both the pump and downconversion wavelengths. Instead, a commercially available standard PBS cube was used, which was originally designed and AR-coated for 780 nm but not for 390 nm. The extinction ratio (ER) and reflection losses that could be achieved with the PBS at the pump wavelength was highly dependent on the incidence angle. Note that deviating from normal incidence changes the right-angle triangular shape for the Sagnac interferometer. However, a useful trade-off can be achieved for optimizing the extinction ratios at both wavelengths. At approximately $2.5^\circ$ incidence angle, the PBS provided $\sim 20:1$ ER for the pump and $\sim 770:1$ ER for 780.7 nm. The absorption and reflection losses of the PBS for the pump in this configuration was 35%. The low PBS ER at 390.35 nm resulted in a small amount of $V$-polarized pump at the crystal but it was not phasematched to generate any downconverted photon pairs.

For type-II phase-matched SPDC in PPKTP, the pump beam polarization should be aligned along the crystal’s $y$-axis. This was achieved by using a dual-wavelength
half-wave plate (DHWP) oriented at 45°, which converted the vertical pump polarization in CCW propagation into horizontal polarization. The DHWP accomplished another essential task, namely, it rotated the clockwise propagating signal and idler pair by 90° so that for bidirectional pumping, the signal and idler photons from CW or CCW directions always exited from the same PBS port. A 1-mm thick multiple-order quartz DHWP was obtained from OptiSource LLC, with both surfaces dual-AR coated at 390.35 nm and 780.7 nm. The measured ER of the DHWP at both wavelengths were better than 100:1.

![Type-II PPKTP cw characterization](image)

Figure 3-18: Wavelength tuning curve for 10-mm long type-II phasematched Raicol PPKTP crystal, showing a 0.084 nm phase-matching bandwidth and a nonlinear coefficient $d_{\text{eff}}$ of $\sim 2.4$ pm/V.

The linearly polarized, collimated UV beam after the dichroic filter was passed through a half-wave plate (HWP) and a quarter-wave plate (QWP) to adjust the relative phase and amplitude between the $H$- and $V$-polarized components as outlined in Sec. 3.4. The pump was focused into the PPKTP crystal positioned at the center of the PSI using a plano-convex lens with a 15-cm focal length and with broadband AR-coating that covered both UV and near-IR wavelengths. To ensure that the counter-
propagating pump components reach the crystal at the same time, thus eliminating any temporal distinguishability between the two outputs, the crystal was positioned at the center of the interferometer to within $\sim 1\, \text{mm}$. This was much less than the coherence length of the 50-ps pump pulse. The pump focal spot at the PSI center plane was measured to have a beam diameter of $\sim 90\, \mu\text{m}$.

The Sagnac interferometer alignment for the two downconverter outputs necessitated erasing any spatial or temporal “which-path” information at the PBS output. In addition to the time-symmetric positioning of the PPKTP crystal, the alignment procedure must ensure that signal and idler beam paths were perfectly overlapped for bidirectional pumping. In order to achieve the coarse interferometer alignment, a 780.7 nm classical source was constructed by tapping and fiber coupling the output of the first SHG stage in the UV-source. The classical 780.7 nm beam path was overlapped with the UV-pump beam by using a dichroic mirror. As we varied the polarization of the classical input, a roughly 50:50 power split at the PBS was achieved. The DHWP orientation was changed to $0^\circ$ (hence no rotation) to allow the classical interference to be observed at the other PBS output port. A $45^\circ$ oriented high extinction ratio Glan-Thompson polarizer separated the diagonally and anti-diagonally polarized outputs from the PSI. For classical 780.7 nm input oriented at $45^\circ$, an interference visibility of 99.4% was obtained after PSI alignment. The same procedure for the classical UV input resulted in 97.5% peak visibility for an uneven power split, resulting from the low ER of the PBS at the UV wavelengths.

For the polarization correlation measurements, we collimated the signal and idler outputs with a 15-cm focal length plano-convex lens and each beam passed through a matching circular aperture for spatial-mode filtering. A set of circular apertures with diameters varying from 1 mm to 4 mm were used to define a full divergence angle for the measured spatial mode. Corresponding full divergence angles for the fixed apertures ranged from 6.7 mrad to 26.7 mrad. Spatially-filtered output beams were directed to polarization analyzers each comprising a half-wave plate and a high-extinction polarizer to measure the polarization states. A quarter-wave plate was also optionally added for quantum state tomography measurements. Both beams
were filtered spectrally using interference-filters (IFs) with a full-width half-maximum (FWHM) bandwidth of \( \sim 0.15 \text{ nm} \) centered at 780.7 nm and with peak transmission of 60%. The double-Lorentzian filter shape caused additional filtering losses, which reduced the aggregate filter transmission to \( \sim 36\% \). Additional suppression of the pump light was provided by inserting two dichroic mirrors on both signal and idler beam paths, where all mirrors were AR-coated for near-infrared wavelengths and highly-reflecting for UV wavelengths. The outputs were subsequently focused by AR-coated 5-cm focal length doublet lenses onto two PerkinElmer Si APDs with \( \sim 50\% \) detection efficiency. Upon a detection event, the APDs generated sharp \( \sim 30 \text{ ns} \) pulses with a \( \sim 300 \text{ ps} \) rise time. Individual APD counts as well as coincidence counts were recorded by a computer controlled counter-timer board. A home-built fast electronic circuit provided 1.8-ns coincidence window for monitoring coincidence counts [138].

We balanced the pair generation rates from both paths and made the necessary input pump polarization adjustments to produce a singlet-state output \( |\psi^-\rangle = (|H_SV_I\rangle - |V_SH_I\rangle)/\sqrt{2} \). In the next two sections, we show the results of our source characterization in the low and high flux regimes.

### 3.5 Low-Flux Characterization of the PSI Source

Unlike most cw SPDC sources, the combination of a high-power pulsed UV pump and the efficient PSI source can lead to a substantial pair generation probability per pulse. In order to assess the entanglement quality of the pulsed Sagnac source with minimal degradation due to multiple-pair events, the PSI output was characterized at low pump power (\( \leq 1 \text{ mW} \)). For the study of low-flux polarization entangled photons various measurement schemes can be used to characterize the output state. In this study three distinct and complementary measurements were performed: Two-photon quantum interference, CHSH form of Bell’s inequality, and quantum state tomography.
3.5.1 Quantum-Interference Visibility

The signal-idler quantum-interference measurement in two incompatible polarization bases, $H-V$ and $\pm 45^\circ$ antidiagonal-diagonal ($A-D$), is a common and relatively simple method to assess the entanglement quality of the polarization-entangled output state. For each of the polarization measurement bases, one can set the signal polarization analyzer angle at one of the basis polarization axes and monitor the coincidence counts as a function of the idler analyzer angle. The quantum-interference visibility is given by

$$V = \frac{C_{\text{max}} - C_{\text{min}}}{C_{\text{max}} + C_{\text{min}}},$$

(3.5)

where $C_{\text{max}}$ and $C_{\text{min}}$ are the maximum and minimum coincidence counts, respectively, and an ideal entanglement source yields $V = 1$.

![Graph showing quantum interference measurements for input pump of 0.1 mW and sinusoidal fits in the $H-V$ (squares) and $A-D$ (circles) bases. $H-V$ ($A-D$) visibility is 99.79% (98.11%).]

A distinct feature of the singlet (equivalently triplet) state as compared to a classical mixture of two opposite polarization photons is the shape of the quantum interference fringe. When both polarization analysis angles are set to $45^\circ$, one gets a
dip in the coincidence counts for the entangled state. On the other hand, the classical mixture of opposite polarizations would only yield a constant coincidence profile. The fringe pattern in the ±45° basis is a direct signature of the quantum correlation of two photons that does not have a classical counterpart. Experimentally, the coincidence dip resulting from the $|AA⟩⟨AA|$ projection operation is quite useful to perform fine adjustments for interferometer alignment and to find the correct pump phase with HWP and QWP. After these adjustments were completed and interference quality was optimized, we observed the fringes at different pump power levels and different aperture sizes. Fig. 3-19 shows the coincidence counts in the $H-V$ and $A-D$ bases and the corresponding sinusoidal fits to data at a pump power of 0.1 mW, as measured at the entrance to the PSI. Each data point represents an average of 30 1-s measurements, and background count levels were not subtracted from the coincidence count data in Fig. 3-19. Two main contributions to the background counts are the Si APD dark-counts ($\sim50$/s) and broadband non-parametric fluorescence, which approximately represents 5% of the total singles counts. These background count levels did not contribute to significant accidental coincidences. Using the measured maximum and minimum coincidence counts and substituting them into Eq. (3.5), an interference visibility of $99.79\pm0.38\%$ was obtained in the $H-V$ basis and $98.11\pm1.16\%$ in the $A-D$ basis. The data in Fig. 3-19 were taken using a circular aperture with a full collection divergence angle of $\sim13$ mrad (2-mm aperture diameter). From the singles count rate of $\sim1,600$/s, a conditional detection probability of $\sim9.5\%$ was inferred, which was limited in large part by the Si APD detector efficiency of $\sim50\%$, the effective IF transmission of $\sim36\%$ and the measured 95% transmission efficiency through the other optical components. The remaining reduction in the conditional detection probability can be attributed to spatial filtering that was provided by the 2-mm diameter aperture, which is a typical problem due to the multimode nature of SPDC with an unfocused or weakly focused pump [43].

After this very low-power measurement, we measured the quantum interference at an increased pump power of 1.1 mW, as shown in Fig. 3-20 and with all other parameters being the same as in Fig. 3-19. The singles rate increased to $16,000\,\text{s}^{-1}$ at this
power level, and the quantum-interference visibilities were found to be 98.04 ± 0.35% in the $H-V$ basis and 96.64 ± 0.46% in the $A-D$ basis. From the singles count rate and the 9.5% conditional detection probability, one can estimate the pair generation rate to be 1.1% per pulse. This high pump power caused the slight reduction in visibilities due to multiple-pair emission events, which are analyzed in detail in the next section.

In practice, the quantum interference visibility depends strongly on the beam divergence angle associated with spatial filtering. For the PSI source, this variation in the quantum-interference visibility was measured at different divergence angles, as set by the aperture diameters, and at a constant input power of 1.1 mW. For each data point in Fig. 3-21, an average of 30 1-s measurements of $C_{\text{max}}$ and $C_{\text{min}}$ were recorded and the visibility was obtained from Eq. (3.5). At a given pump power a larger aperture size allowed more light to be collected which increased the effective pair generation rate. As a result, multiple-pair events increased (which are analyzed in more detail in Sec. 3.6) and caused the visibility to degrade slightly, as shown in
Fig. 3-21 for the measurements in the $H-V$ (squares) basis. The $A-D$ data shows a more pronounced deterioration in the visibility at larger collection angles. The $H-V$ basis was the natural basis aligned with the PPKTP’s crystal principal axes. Therefore, measurements in the $A-D$ basis required the coherent superposition of the two counter-propagating downconverter outputs, and the entanglement quality was sensitive to their spatial mode distinguishability and to the phase variation of the output state across the spatial extent. Crystal grating inhomogeneity can give rise to spatial mode distinguishability if the confocal parameter associated with the collection beam divergence is smaller than the crystal length (see measurements in Chapter 5 for clear evidence of crystal inhomogeneity). For multimode SPDC, the deviation from one type of Bell state to the other type can be caused by a relative phase shift between $|HV\rangle$ and $|VH\rangle$ terms for different transverse wavevectors [139]. In this case, the $A-D$ data in Fig. 3-21 clearly show that the singlet-state entangled output of the PSI degraded at larger divergence angles. In designing the Sagnac source for a specific application, one must therefore consider the trade-off between generation efficiencies and entanglement quality as a function of the collection angles.

3.5.2 CHSH $S$-Parameter Measurements

Violation of the Clauser-Horne-Shimony-Holt (CHSH) form of Bell’s inequality [106] is another common method of entanglement characterization by measuring its $S$-parameter. For an ideal entangled state a maximum value of $S = 2\sqrt{2}$ is predicted by quantum mechanics, whereas $S$ cannot be greater than 2 classically. Therefore, the state is considered nonclassical if $2 < S \leq 2\sqrt{2}$ and the closer $S$ is to $2\sqrt{2}$, the higher is the entanglement quality. $S$-parameter measurement is an appropriate joint measurement for the eQKD scheme. Alice and Bob can perform their measurements on randomly picked bases and later check for the violation of classical $S$-parameter bound over the public channel.

The cw PSI experiment offered a detailed account of the $S$-parameter measurement procedure [42] and the same methodology was pursued here for the pulsed PSI source. Calculation of the $S$-parameter depends on the results of a series of pro-
Figure 3-21: Quantum interference visibilities in the $H$-$V$ (squares) and $A$-$D$ (circles) bases for various aperture sizes at a constant input pump power of 1.1 mW.

Objective measurements on two-photon quantum state. Specifically, the value of the $S$-parameter is determined by four measurement expectation values [106]:

$$S = |E(\theta_S, \theta_I) + E(\theta_S, \theta_I') - E(\theta_S', \theta_I) + E(\theta_S', \theta_I')|,$$  \hspace{1cm} (3.6)

where $\theta_S$ and $\theta_I$ are the polarization analyzer angles for signal and idler, respectively. The value of $S$ can be maximized by a judicious choice of analyzer angles and for this measurement the following set was selected: $\theta_S = -\pi/4$, $\theta_S' = 0$, $\theta_I = 5\pi/8$, and $\theta_I' = 7\pi/8$. Each expectation value $E(\theta_S, \theta_I)$ is obtained from the coincidence measurements in 4 different polarization analyzer combinations: the chosen signal and idler set and their orthogonal sets, namely, $(\theta_S, \theta_I)$, $(\theta_S, \theta_I + \pi/2)$, $(\theta_S + \pi/2, \theta_I)$ and $(\theta_S + \pi/2, \theta_I + \pi/2)$. One calculates the expectation value according to

$$E(\theta_S, \theta_I) = \frac{C_{++} - C_{+-} - C_{-+} + C_{--}}{C_{++} + C_{+-} + C_{-+} + C_{--}},$$  \hspace{1cm} (3.7)

where the subscript $(++)$ represents the angle set $(\theta_S, \theta_I)$ and the $(--)$ subscript...
replaces it with the orthogonal angle.

As part of the low-flux characterization, the $S$-parameter measurements were performed at an input pump power of $\sim 70 \, \mu \text{W}$ at a full divergence angle of 13 mrad. We collected a total of 16 coincidence measurements, each consisting of an average of 30 1-s data sets. The observed expectation values and errors are tabulated in Table 3.3:

<table>
<thead>
<tr>
<th>$\theta_S, \theta_I$</th>
<th>$E(\theta_S, \theta_I)$</th>
<th>$\Delta E(\theta_S, \theta_I)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$-\pi/4, 5\pi/8$</td>
<td>-0.726</td>
<td>0.0547</td>
</tr>
<tr>
<td>$-\pi/4, 7\pi/8$</td>
<td>-0.647</td>
<td>0.0596</td>
</tr>
<tr>
<td>$0, 5\pi/8$</td>
<td>0.664</td>
<td>0.0621</td>
</tr>
<tr>
<td>$0, 7\pi/8$</td>
<td>-0.702</td>
<td>0.0619</td>
</tr>
</tbody>
</table>

Table 3.3: CHSH - Bell’s inequality measurement

From the measured values in Table 3.3 and Eq. (3.6), we obtain an $S$-parameter value of $S = 2.739 \pm 0.119$, indicating a violation of Bell’s inequality of more than 6 standard deviations. The entanglement quality given by $S$ is comparable to that indicated by the quantum-interference visibility measurements of Sect. 3.5.1. At a higher power level of 1.1 mW, the magnitude of $S$ drops slightly to $S = 2.675 \pm 0.033$. Once again, this degradation is caused by multi-pair generation events.

### 3.5.3 Quantum State Tomography

A more detailed characterization of the output state can be obtained by quantum state tomography. One can reconstruct the density matrix, $\hat{\rho}$, for a two-photon output state through a series of projective measurements. The coincidence counts recorded in these measurements can be used to obtain individual density matrix elements by a maximum likelihood algorithm. For the pulsed PSI source, the protocol outlined in [140, 141] was utilized to estimate $\hat{\rho}$ from 16 projective measurements for an input pump power of 0.1 mW at a full divergence angle of 13 mrad. Similar to the CHSH measurements, 30 1-s coincidence counting data points were averaged for each measurement point. Coincidence data from these measurements were used in a nonlinear least squares algorithm to estimate the bipartite density matrix. With the density
matrix we can calculate various fidelity measures to characterize the entangled state.

The real and imaginary parts of \( \hat{\rho} \) are plotted in Fig. 3-22. This density matrix estimation yielded 98.85% fidelity for the PSI singlet-state output, where the fidelity was calculated as \( \text{Tr}[\hat{\rho} \hat{\rho}_{\psi^-}] \), with \( \hat{\rho}_{\psi^-} \) being the singlet-state density operator. Another entanglement measure, tangle, could be calculated from the estimated density operator [142]. A factorizable state yields zero tangle and a maximally-entangled Bell state yields unity. The estimated density operator yielded a tangle value of 0.9589, indicating a highly entangled output state. We observed that all three types of measurements, two-photon quantum-interference visibility, CHSH-type Bell’s inequality violation, and quantum state tomography are consistent, equally effective and useful in characterizing the PSI output state as highly entangled.

### 3.6 High-Flux Characteristics

In the previous section the PSI source output was characterized in the low-flux limit. From the measurements of Fig. 3-20, the estimate for the pair generation rate was 1% per pulse per mW of average pump power for a full divergence angle of 13 mrad.
Availability of over 400 mW pump power gives the capability to drive the SPDC process at a significant generation rate at which multiple pairs could be produced in large numbers. This generation capability allows one to tailor the SPDC source for a specific application. One may, for instance, improve the entanglement source performance by increasing the pump power and reducing the divergence angle to achieve the desired generation rate with a higher entanglement quality. However, a higher flux output comes with its disadvantages. The main concern is an increase of accidental coincidences that reduces quantum-interference visibility, leading to potential errors in quantum information processing tasks. For instance, one can consider entanglement-based QKD as an example, where a higher secret key rate is expected for a mean pair generation number $\alpha \gg 1\%$. However, the maximum $\alpha$ that can be used without compromising its performance depends on a number of operational system parameters [116]. As a consequence, it is highly useful to have a simple and accurate estimate of the pair generation rate under all circumstances. The main focus of this section is the occurrence of accidental coincidences and how a pulsed source can lead to more errors than a cw source. Further attention will be given to the impact of multiple pair generation on entanglement quality and how that effect can be utilized to measure the pair generation probability accurately.

### 3.6.1 Accidental Coincidences

There are three main contributions to the observation of accidental coincidences in the typical measurement setup of Fig. 3-16. The first type of contribution is due to the generation of independent multiple entangled pairs from a multimode SPDC source under strong pumping. If the detected coincident photon pair originates from two different signal-idler pairs, there is no polarization correlation and therefore an error may occur. Consider the case of two-pair events where the production rate is proportional to the square of the pump power. For two independent photon pairs generated by the same pump pulse, all possible detection scenarios are depicted in Fig. 3-23. If the polarization analyzers for coincident photon detection are set to opposite polarizations, a maximum number of pairs are registered. In the presence
of a double pair, three out of four possible distributions will register a coincidence, where at least one detection event is required for two photon-number non-resolving detectors with unity detection efficiency. For coincidence minimum, the analyzers will be set to the same polarizations and two out of four distributions will yield a coincident count. In contrast, a single pair of entangled photons does not yield an accidental coincidence at the coincidence minimum point and half of the time it gives a coincidence click at the maximum point.

Figure 3-23: Four possible polarization distributions for two independent pairs of photons generated in the same pulse and detected by perfect polarization analyzers and single-photon counters. The polarization analyzers are set to opposite polarizations for maximum coincident counts where pairs yielding coincidences are enclosed in blue solid box. The minimum coincidence counts arising from double pairs are enclosed in red dotted box, where analyzer polarization settings are identical.

The second type of contribution is caused by the detection of one photon from a downconverted photon pair and a UV-induced fluorescence photon emitted by the PPKTP crystal which we measured to have a generation rate of less than 5% of the singles count rate. Since the fluorescence varies linearly with the pump power, the accidental coincidence rate is proportional to the square of the pump power. There is also the possibility of an accidental coincidence due to the detection of two independent fluorescence photons or detection of a fluorescence photon in one detector and a dark count in the other detector. But the probabilities of these events are much lower than the case when one of the detected photons belongs to a downconverted pair. The third contribution comes from background photons from stray light and from detector dark counts. Both events are independent of the pump power and
generally the background and dark counts are low enough that accidental coincidences caused by them are negligible.

The above discussion on accidental coincidences applies to both cw and pulsed SPDC. However, more accidental coincidences have been observed in the pulsed case than in the cw case for the same average pumping power. Given the same SPDC setup and the same average input power one would observe the same number of downconverted pairs per second for the cw and pulsed cases. In cw operation, the SPDC pair or fluorescence photon generation probability within a coincidence window of duration $T_c$ is proportional to $T_c$. The cw accidental coincidence rate resulting from two-pair events is then given by $f_{cw} = gT_c$, where $g$ is a proportionality constant. On the other hand, for pulsed SPDC with a repetition rate of $R_p$, the pair or fluorescence emission is localized within each pump pulse duration, whose width is typically smaller than $T_c$, as in the PSI source. Therefore, the SPDC pair (or fluorescence photon) generation probability per pulse (alternatively, per coincidence window) is proportional to $1/R_p$. The accidental coincidence probability per pulse is then proportional to $1/R_p^2$, and the accidental coincidence rate is $f_{pulsed} = g/R_p$. Note that the cw and pulsed cases have the same proportionality as long as the same type of accidental coincidences is considered. For the same input power, the ratio of their accidental rates is

$$\frac{f_{cw}}{f_{pulsed}} = R_pT_c. \quad (3.8)$$

Using a typical value for the coincidence window duration $T_c = 1$ ns, the repetition rate of $R_p = 31.1$ MHz for the pulsed pump implies that $f_{cw} \ll f_{pulsed}$. That is, the pulsed Sagnac SPDC source is much more susceptible to visibility degradation due to accidental coincidences as compared to its cw counterpart with the same entangled pair generation rate. Equivalently, a cw pumped source can tolerate a much higher average pump power than a pulsed source for the same amount of accidental coincidences. A quick comparison to the cw experiment in Ref.[42] with $T_c = 1.8$ ns and $R_p = 31.1$ MHz gives a scaling factor of $\sim 18$ for the input pump power that
will yield the same accidental coincidence rate. This problem can be minimized by increasing the repetition rate until it is comparable to \( 1/T_c \). Increasing the repetition rate, however, has its own problems. For instance, typical Si APDs are not able to handle detection rates much higher than a few MHz. Also, it is not desirable to have \( R_p T_c > 0.5 \) because of the need for temporal separation of the pulses. For a pulsed source designed for a specific application such as QKD, one must be aware of the trade-offs between the need for a high pair generation rate and the desire for high entanglement quality, and strike an application-specific balance for the appropriate combination of pump powers, repetition rates, detector speeds, and error budget.

### 3.6.2 Multiple-Pair Generation

The UV-induced fluorescence rate was measured by detuning the phase-matching temperature so that only fluorescence photons were detectable and their detection rate was only a small fraction (\( \sim 5\% \)) of the detected singles rate. Therefore multiple-pair generation is the main contributor to accidental coincidences in the PSI setup. The limitations due to multi-pair events have previously been observed [143, 144]. Eisenberg et al. measured the multi-pair visibility degradation in their study of stimulated parametric emission in a multi-pass downconversion configuration [145]. In this section, a simple Poisson model is developed to quantify the effect of multiple-pair generation on two-photon quantum interference measurements in spontaneous parametric emission in a single-pass downconversion setup. The theoretical predictions of this model are then compared to the experimental observations at various pump power levels.

Consider an ideal pulsed SPDC source that generates singlet-state polarization-entangled photon pairs, with no background or fluorescence photons. It is fair to assume that the free-space output is spatially multimode and that the output is also temporally multimode because the coincidence measurement time is large compared with the output pulse width. As a consequence, the number of entangled pairs in the
output pulse is Poisson distributed with a mean pair generation number $\alpha$:

$$p_n(\alpha) = \frac{e^{-\alpha} \alpha^n}{n!}, \quad \sum_{n=0}^{\infty} p_n(\alpha) = 1,$$

(3.9)

where $p_n(\alpha)$ is the probability weight for obtaining exactly $n$ pairs per pulse. One can further assume that the $n$ entangled pairs are independent because of the multimode nature of the output. That is, each mode has an occupation number of either 0 or 1. Note that the assumption of the Poisson distribution is not valid for single-mode outputs, in which case a Bose-Einstein distribution for the probability weights would be appropriate:

$$p_n(\alpha) = \frac{\alpha^n}{(1 + \alpha)^{n+1}}.$$

(3.10)

It will be insightful to compare both distributions for various values of $\alpha$. The effect of multiple pairs on the two-photon quantum-interference visibility can now be written explicitly. We first treat the multimode output case with Poisson statistics. In calculating the visibility of Eq. (3.5), the only necessary parameters are the maximum and minimum coincidence probabilities, $C_{\text{max}}$ and $C_{\text{min}}$, respectively. For singlet-state entangled output, one can consider a coincidence measurement along $H$ polarization for the signal photon and along $V$ ($H$) polarization for the idler photon to measure $C_{\text{max}}$ ($C_{\text{min}}$), assuming ideal polarization analyzers. Other polarization settings (such as $A-D$) would work just as well, as long as the polarizers are orthogonal for measuring $C_{\text{max}}$ and parallel for measuring $C_{\text{min}}$. To complete the model for the calculations, one can further assume ideal photon-number non-resolving detectors and both the signal and idler paths have a system detection efficiency $\eta$.

The minimum and maximum coincidence probabilities are given by

$$C_{\text{min}} = \sum_{n=1}^{\infty} p_n(\alpha)c_{\text{min}}(n),$$

(3.11)

$$C_{\text{max}} = \sum_{n=1}^{\infty} p_n(\alpha)c_{\text{max}}(n),$$

(3.12)

where $c_{\text{min}}(n)$ and $c_{\text{max}}(n)$ are the minimum and maximum coincidence probabi-
ties for an output with exactly \( n \) independent pairs of entangled photons in multiple spatio-temporal modes. Note that the summation starts from \( n = 1 \) because \( c_{\text{min}}(0) = c_{\text{max}}(0) = 0 \). For \( n \) independent photon pairs, there are \((n + 1)\) different ways to arrange their polarization orientations relative to the analyzer settings, and they follow the binomial distribution. For example, as shown in Fig. 3-24 among \( n \) signal photons \( k \) of them may be \( H \)-polarized (with corresponding \( V \)-polarized idler photons), and the remaining \( n - k \) photons will be \( V \)-polarized and this specific configuration appears with a probability of \((\frac{n}{k})/2^n \) with the binomial coefficient

\[
\binom{n}{k} = \frac{n!}{(n-k)!k!}
\]

(3.13)

Figure 3-24: A sample distribution of multiple photon pairs generated in a single pulse. The depicted distribution probability can be easily calculated from the binomial distribution \((\frac{n}{k})/2^n \).

for \( 0 \leq k \leq n \). For a given polarization arrangement, one requires at least one detected \( H \)-polarized event at the signal detector and at least one detected \( V \)-polarized event at the idler detector to yield a coincidence count for \( c_{\text{max}}(n) \). Similarly, to obtain \( c_{\text{min}}(n) \) it is required to detect an \( H \)-polarized event at each detector. Combining the above requirements, both coefficients can be expressed as:

\[
c_{\text{min}}(n) = \frac{1}{2^n} \sum_{k=1}^{n-1} \binom{n}{k} \left[ 1 - (1 - \eta)^k \right] \left[ 1 - (1 - \eta)^{n-k} \right],
\]

\[
c_{\text{max}}(n) = \frac{1}{2^n} \sum_{k=1}^{n} \binom{n}{k} \left[ 1 - (1 - \eta)^k \right]^2.
\]

(3.14)

Using the above coefficients, it is straightforward to compute the minimum and maximum coincidence probabilities from Eqs. (3.11-3.12) and obtain the visibility
of Eq. (3.5) as a function of the mean pair generation probability per pulse $\alpha$.

It is instructive to evaluate the multiple-pair effect for small $\alpha$ in which case the terms with more than two pairs of entangled photons can be ignored. The simplified Poisson distribution coefficients then read $p_1(\alpha) \approx \alpha(1 - \alpha)$ and $p_2(\alpha) \approx \alpha^2/2$. In this case, one obtains

$$C_{\min} = \frac{1}{4} \eta^2 \alpha^2,$$

$$C_{\max} = \frac{1}{2} \eta^2 \alpha \left[ 1 + \frac{\alpha}{4}(2 - 4\eta + \eta^2) \right],$$

and the corresponding visibility expression to the first order of $\alpha$ as

$$V = 1 - \alpha.$$

We can pursue the visibility calculation for the single spatio-temporal output with Bose-Einstein statistics. For a given mean pair generation probability, $\alpha$, and the probability of generating $n$ pairs of photons is given by Eq. (3.10). Due to the coherent superposition of the two downconverter outputs, we can obtain $n$ pairs at the output of the Sagnac interferometer in a number of possible ways, each with a distinct probability. In order to calculate this probability, we first note that while the frequency and spatial mode of the output photons are identical, the polarization states of these photons can be different depending on their propagation direction within the Sagnac interferometer. In that case, we can assign each polarization state an occupation number which enables us to express the output state for $n$ pairs as follows:

$$|\psi_{(n,m)}\rangle = |m\rangle_{H,S}|m\rangle_{V,I}|n-m\rangle_{V,S}|n-m\rangle_{H,I}$$

for $m \leq n$

where the subscript indicates the polarization and the output spatial mode of the photon. For the above output state, $m$ pairs with orthogonally polarized photons are generated by the clockwise propagating pump beam and the remaining $n - m$ pairs are generated by the counter-clockwise propagating pump beam. The probability
of generating this specific configuration for the \( n \)-pair event is determined by the Bose-Einstein statistics:

\[
p_{n,m}(\alpha) = \frac{\alpha^{m}}{(1 + \alpha)^{m+1}} \frac{\alpha^{n-m}}{(1 + \alpha)^{(n-m)+1}} = \frac{\alpha^{n}}{(1 + \alpha)^{n+2}} \tag{3.19}
\]

Therefore, similar to the multimode case, we can express the minimum and maximum coincidences for the quantum-interference visibility with a single spatio-temporal output and a mean pair generation probability \( \alpha \):

\[
C'_{\text{min}} = \sum_{n=1}^{\infty} \sum_{k=1}^{n} p_{n,k}(\alpha) \left[ 1 - (1 - \eta)^{k} \right] \left[ 1 - (1 - \eta)^{n-k} \right],
\]

\[
C'_{\text{max}} = \sum_{n=1}^{\infty} \sum_{k=1}^{n} p_{n,k}(\alpha) \left[ 1 - (1 - \eta)^{k} \right]^2. \tag{3.20}
\]

where the quantum interference visibility can be calculated by using Eq. (3.5). In the low-flux limit, the single spatio-temporal mode quantum-interference visibility can be approximated as \( V \approx 1 - 2\alpha \). In contrast to the visibility expression in Eq. (3.17), the single spatio-temporal output would exhibit faster visibility degradation for small \( \alpha \) values.

For the multimode case of Eq. (3.17) the linear dependence of the quantum-interference visibility on the mean pair generation probability shows clearly how the entanglement quality degrades with increasing pump powers. It is a little surprising to see in Eq. (3.17) that there is no dependence on the system detection efficiency \( \eta \). A physical explanation is that for a pair of photons that survive the system loss and are detected, one does not know if they belong to the same entangled pair or not and therefore it should only depend on the generation statistics and not on the system loss. For a system with high loss that results in a low detection rate, the entanglement quality may still be compromised by multi-pair events.

The above results can be directly compared with measurements in the setup shown in Fig. 3-16. The free-space output of the pulsed PSI source with a weakly focused pump had many spatial modes that can be described by a Poisson distribution. More-
over, the pumping rate was low enough that even at the highest measured $\alpha$ value in the experiment, stimulated emission can be neglected. The quantum-interference visibilities in both $H-V$ and $A-D$ bases were measured at different input pump powers and at a fixed full divergence angle of 13 mrad. The pair generation rate per pulse $\alpha$ was inferred from singles and coincidence measurements, which have yielded a conditional detection efficiency of $\eta = 9.5\%$ under these operating conditions. The maximum pump power was $\sim 70 \text{ mW}$ which corresponds to $\alpha = 0.7$. A pair detection rate of $\sim 110,000 / \text{s}$ was measured with $\sim 1 \text{ MHz}$ singles rate at this power level. Higher $\alpha$ values were not pursued to avoid any possible crystal damage at elevated pump power levels.

![Figure 3-25: Quantum-interference visibility in the $H-V$ (squares) and $A-D$ (circles) bases as a function of mean pair generation number $\alpha$. The solid blue curve is computed from a multimode Poisson model. The green dashed curve depicts the Bose-Einstein single mode theoretical predictions. Visibilities in the $A-D$ basis are lower than those in the $H-V$ basis due to other factors that are unrelated to multi-pair occurrences. Inset shows the linear behavior of the quantum interference visibility in the low-flux regime ($\alpha \leq 0.1$).](image)

Figure 3-25 plots the measured visibilities and compares them with the calculated
values from the Bose-Einstein (single mode) and Poisson (multimode) models. The visibility calculation with the single-mode assumption and associated pair generation statistics with the Bose-Einstein distribution yields a lower interference visibility profile and it does not match our experimental data. For the range of interest ($\alpha < 1$), the prediction of the Poisson model exhibits very good agreement with the experimental data, verifying our initial assumption about the multimode nature of the output state. The $H-V$ results match the theoretical values from the Poisson model very well in both the low and high $\alpha$ regimes. In particular, at low $\alpha$, the visibility degradation is linear with respect to the mean pair generation number. For high $\alpha$ values, we observe that the experimentally obtained visibility points slightly drift from the theoretical curve. We attribute this fact to the estimation errors for the mean pair generation rate at high flux values where the fluctuations in the singles and coincidence count rates are more pronounced. The $A-D$ results also show the expected linear dependence on $\alpha$ except there is a fixed amount of visibility loss, which is related to other factors. The discrepancy between the $H-V$ and $A-D$ visibilities can be attributed to some imperfect experimental components. The strongest candidate for this deterioration is the wavefront distortion in the optical components of the Sagnac interferometer that created partial spatial distinguishability. Certainly, if a single optical component along the pump or the output beam path induces spatial distortions, the later stages of the experiment will also suffer the same imperfection. It was verified that this problem did not originate from the input pump profile because no visibility improvement was observed after cleaning up the pump spatial mode by coupling it into a single mode fiber. The surface quality of the PBS might be responsible for the visibility loss, yet this was not verified. Further testing with a higher quality dual-wavelength PBS is needed to support this claim. Another possible cause is the crystal poling inhomogeneity that we discussed in Sect. 3.5.1. Since the $H-V$ measurements were made in the downconverters’ natural polarization basis, their results were not affected by these distinguishability issues. It is useful to note that one can take advantage of the $\alpha$ dependence of $H-V$ visibility measurements to deduce accurately the mean pair generation probability $\alpha$. 

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3.7 Summary

This chapter focused on the development of a compact, pulsed, narrowband source of polarization-entangled photons that is of interest to entanglement-based free-space QKD. The design efforts were focused on a high-power field-deployable UV pump source and an efficient polarization entangled photon source with the phase-stable PSI configuration. The first half of this chapter explained the compact UV-source design, which relied on a passively modelocked fiber laser with a MOPA configuration and two stages of SHG. The UV-source performance was satisfactory for driving one or potentially more PSI sources efficiently. A significant part of the pump source development was devoted to understanding and remedying the UV-induced damage issues in PPKTP. This study offered a phenomenological perspective on some of the practical problems with using KTP for high-intensity UV generation.

The second half of the chapter elaborated on the construction and characterization of the pulsed, narrowband downconversion source of polarization-entangled photons. Pulsed operation allows a QKD system to easily manage the arrival times of the photons by synchronization of the transmitter and receiver clocks. It also provides temporal discrimination against background light that arrives outside of the clocked coincidence window. The narrowband SPDC output allows a matching narrowband spectral filter to be used to screen out ambient light especially during daylight QKD operation. Pumped by the developed UV source, the Sagnac geometry ensured a phase-stable generation of any desired Bell state at the degenerate wavelength of 780.7 nm. In principle, the bidirectionally pumped single-crystal Sagnac configuration eliminates spatial, spectral, and temporal distinguishability in the interferometric combination of the two counter-propagating downconverter outputs. Experimentally, a high quality entanglement was observed except for a slight quantum interference visibility loss caused most likely by the spatial distinguishability problem.

The PSI source was highly efficient and it was pumped to cover a range of pair generation probabilities, reaching almost unity probability at the high end. At the low end, a pair generation probability of 0.01 per pulse was achieved at an average pump
power of \( \sim 1 \text{mW} \) at a pump repetition rate of 31.1 MHz. However, achieving higher pair generation degrades the two-photon entanglement quality due to the presence of multiple pairs and this operational regime may not be suitable for some quantum information processing tasks. If the pair generation probability per pulse is limited by multi-pair events, then the only way to increase the flux without reducing the entanglement quality is to increase the repetition rate, up to the point where the detector speed may impose a practical limit.

It is common to evaluate the performance of a cw SPDC source by its quantum-interference visibility and its spectral brightness, which is the number of detected pairs per second per mW of pump in a 1-nm bandwidth. However, the spectral brightness is not an appropriate performance metric for a pulsed source. Instead of spectral brightness, it is more useful to specify the pair generation probability per pulse and the repetition rate of the source. The pulsed Sagnac source is capable of achieving a high enough pair generation rate (as high as 0.7/pulse) for optimal secret key generation in an entanglement-based QKD system in the presence of channel losses and non-ideal detection efficiencies [116]. High pair generation rates have been reported for fiber-based \( \chi^{(3)} \) downconversion sources [144, 146]. The PSI source performance compares well in terms of achievable generation efficiency with negligible amount of fluorescence and scattering effects.

In conclusion, a highly efficient SPDC source was developed utilizing the Sagnac configuration to generate pulsed, narrowband, polarization-entangled photons. With the PSI source, high entanglement quality and high pair generation rates were demonstrated. Under strong pumping, the pulsed source showed entanglement quality degradation due to multi-pair events, in accordance with the theoretical analysis. The source can be utilized in many quantum information processing applications including quantum key distribution. In addition, because of its high efficiency and compactness, the Sagnac source is also useful for more advanced quantum enabling technologies such as an on-demand source of single photons [147].
Chapter 4

Coincident-Frequency
Entanglement with Extended Phase-Matching

4.1 Introduction

Improvement in measurement accuracies beyond the classical limits is one of the most attractive prospects for quantum metrology. Quantum optics provides some of the essential tools for precision metrology, such as squeezed states or novel entangled states [10, 67, 69, 68]. Using non-classical light, interferometric measurements beyond the standard quantum limit have been demonstrated [148, 149]. A recent protocol for quantum clock synchronization and quantum metrology proposes the use of a positive frequency-correlated quantum state to achieve Heisenberg-limited precision [13, 55]. In this scheme it can be shown that the timing and positioning measurements with an $N$-photon coincident-frequency entangled state can be enhanced by a factor of $\sqrt{N}$ over the standard quantum limit, $\Delta t = 1/\sqrt{N}\Delta\omega$, where $\Delta t$ is the timing accuracy, and $\Delta\omega$ is the coherence bandwidth of each photon. The coincident-frequency entangled state with mean frequency $\bar{\omega}$ is given by $|\psi\rangle = \int d\omega \phi(\omega)|\bar{\omega}+\omega\rangle_1...|\bar{\omega}+\omega\rangle_N$, with the spectral amplitude $\phi(\omega)$. A proof-of-principle realization for this state with two
photons can be achieved by ultrafast-pumped spontaneous parametric downconversion (SPDC). As we will show in this chapter, the prescribed frequency correlations for coincident-frequency entanglement calls for precise engineering of the joint spectral distribution of the SPDC two-photon output state.

Besides quantum metrology, engineering the ultrafast SPDC two-photon correlations is useful for other quantum information processing applications as well. For instance, linear optics quantum computation requires ancilla photons in well-defined temporal modes for successful operation of LOQC quantum gates [18]. In addition to providing timing information, an ultrafast pump source also partially determines the spectral and temporal properties of photon pairs. Therefore, pure single-photon ancilla states for optimal LOQC gate fidelities can be generated by controlling and manipulating the joint state spectral correlations.

Because of the large pump bandwidth and frequency anti-correlation in typical downconverted light, ultrafast SPDC often generates photon pairs with signal and idler frequency spectra that are not identical [59, 41]. This spectral distinguishability under pulsed pumping causes Hong-Ou-Mandel (HOM) interferometric measurements [2] to have poor visibility [59, 41, 150, 151]. This was traditionally alleviated by narrowband filtering of the output photons or by using a thin nonlinear crystal, both causing the output flux to drop significantly [152, 61]. However, there are other techniques to impose or restore the spectral indistinguishability of the two-photon state. We especially note the pioneering work of Ian Walmsley’s group first at University of Rochester and later at Oxford University for characterizing the spectral information, distinguishability and entanglement in SPDC [59, 38, 41, 71]. They outlined methods to engineer the joint spectral amplitude of the two-photon state to achieve a desired amount of frequency entanglement depending on practical applications. By controlling the interplay between the phase-matching function and broadband pump spectrum, they demonstrated a highly frequency-entangled two-photon output state with indistinguishable single photon spectra [60, 153]. We also note that it was possible to eliminate the distinguishing timing information externally. For instance, by incorporating additional dispersive and birefringent crystals of precise
length within multiple SPDC nonlinear crystals, timing for signal and idler photons can be made indistinguishable which improves the visibility of HOM interference for ultrafast-pumped SPDC [63]. Even though this approach enables an effective long nonlinear medium with controllable dispersive properties and engineered two-photon timing, the requirement for multiple compensation and nonlinear crystals with precise thicknesses can constitute a practical limit on scalable design. Other groups have shown alternative methods of controlling two-photon spectral correlations by manipulating the phase-matching conditions through the spatial degrees of freedom. For instance, by controlling the spatial dispersion of the pump beam and using the angle dependence of the phase-matching function, the spectral correlations of signal and idler can be varied in a controllable manner [64, 65]. This method also offers the freedom of using arbitrary nonlinear crystals and choosing a desired wavelength range for signal and idler photons without being restricted by the dispersive and phase-matching properties of the nonlinear crystal. However, we note that the precise focusing and alignment requirements may be impractical for a robust system design. Our approach aims to generate the frequency-indistinguishable two-photon state directly from the downconversion process. We rely on the spectral engineering tools outlined by previous theoretical and experimental works [59, 38, 41, 57, 72] to implement an ultrafast-pumped SPDC source with a single efficient nonlinear crystal, simple focusing and alignment requirements and adjustable frequency-entanglement to be utilized in a specific QIP application.

Specifically, we study the generation of coincident-frequency entangled photons via extended phase-matching (EPM) conditions in PPKTP. The coincident-frequency generation of signal and idler directly from PPKTP remedies spectral distinguishability problems of broadband SPDC. We first give an overview of this scheme developed by Giovannetti et al. and explain how it provides identical signal and idler spectra [57, 56]. By applying the EPM conditions to a type-II phase-matched PPKTP crystal, we tested the frequency indistinguishability of the two-photon output with a fiber-based Hong-Ou-Mandel (HOM) interferometer. The two-photon autocorrelation times obtained from HOM interference were then compared with single-photon
autocorrelation times to verify the frequency entanglement. We further improved the frequency indistinguishability of the joint state by determining the degenerate operating conditions accurately for coincident-frequency generation of signal and idler. The improved conditions were obtained by performing classical difference-frequency generation with the same PPKTP crystal. Finally, we examine the need for characterizing the joint temporal statistics of the two-photon state.

4.2 Extended Phase-Matching Conditions for PPKTP

For the simplest nontrivial case of $N = 2$ the coincident-frequency state $|\psi\rangle_{CF} = \int d\omega \phi(\omega)[\omega + \omega]_1[\omega + \omega]_2$ consists of a pair of entangled photons with identical but uncertain frequencies; the two photons are positively correlated in frequency, and hence anti-correlated in time. Here, $\phi(\omega)$ is the joint spectral amplitude (JSA) that is a function of the pump and the phase-matching function. In contrast, conventional SPDC with cw pumping yields pairs of time-entangled photons that are correlated in time but anti-correlated in frequencies as described by the state $|\psi\rangle_{SPDC} = \int d\omega \phi(\omega)[\omega + \omega]_1[\omega - \omega]_2$.

Engineering the spectral and temporal correlations of the joint state relies on controlling the two independent components: The pump spectral amplitude and the phase-matching function. It is instructive to inspect the JSA with explicit frequency dependencies. We rewrite the JSA expression from Chapter 2 with the constant factor of $-iL/2$ omitted:

$$\tilde{A}(\omega_S, \omega_I) = \mathcal{E}_p(\omega_S + \omega_I) \frac{\sin(\Delta k(\omega_S, \omega_I)L/2)}{\Delta k(\omega_S, \omega_I)/2}$$

where $\omega_S$, $\omega_I$ are the frequencies for signal and idler, respectively, and their frequency dependent refractive indices are $n_S(\omega_S)$ and $n_I(\omega_I)$, $\mathcal{E}_p(\omega_S + \omega_I)$ is the pump spectral amplitude and $\sin(\Delta k(\omega_S, \omega_I)L/2)/(\Delta k(\omega_S, \omega_I)/2)$ is the phase-matching function.

Intuitively, the JSA for the positive frequency-correlated two-photon emission should
confine the frequencies of signal and idler to a narrow bandwidth for a given pump
frequency. But also it should enable a coherent superposition of these correlated pairs
for all possible pump frequencies. Let us first express the desired functional form for
the coincident-frequency two-photon state and then we can list the requirements for
the pump spectrum and phase-matching function to achieve the given form:

\[ \tilde{A}_{CF}(\omega_S, \omega_I) \propto P(\omega_S + \omega_I)Q(\omega_S - \omega_I) \] (4.2)

The above JSA for two-photon state has sum-frequency dependent, \( P(\omega_S + \omega_I) \),
and difference-frequency dependent, \( Q(\omega_S - \omega_I) \), components. The sum-frequency
term is assumed to be a broadband function with non-zero values along the \( \omega_S + \omega_I = \omega_P \) contour with the frequency spread determined by the pump bandwidth, \( \Omega_P \).
Here, \( \omega_P \) is the center frequency for the pump spectrum. For coincident-frequency
generation of the two-photon state, we expect the difference-frequency term to take
finite values only if signal and idler frequencies are the same and zero otherwise. In
the two-dimensional frequency-space, the squared magnitude of this function would
look similar to the distribution sketched in Fig. 2-3.

The pump dependent part of JSA already has the same functional form for the
summation of \( \omega_S \) and \( \omega_I \). Thus, the problem reduces to manipulating the phase-
matching function to have a parametrization with the difference of \( \omega_S \) and \( \omega_I \). One
may think that using a very long nonlinear crystal will achieve the required coincident-
frequency generation for signal and idler. In general, however, this is only true for
one specific pump frequency and does not hold for other pump frequencies, resulting
in poor utilization of the broad pump spectrum. Therefore, it is necessary to control
the variation of the phase-matching function for the entire spectral range where the
entire spectrum of the broadband pump can be utilized. First, assume a degenerate
phase-matching scheme with \( \omega_P = \omega_S + \omega_I \) and \( \omega_S = \omega_I \). As the pump frequency is
varied, the the signal and idler frequencies are detuned from the degenerate center of
half the pump frequency by different rates. These detuning rates for signal and idler
are determined by the phase-matching condition, \( \Delta k(\omega_S, \omega_I) = 0 \), and they can be
obtained from the first-order Taylor expansion for the wavevector mismatch:

\[
\Delta k(\omega_S + \Delta \omega_S, \omega_I + \Delta \omega_I) = \Delta k(\omega_S, \omega_I) + \frac{\partial k_p(\Delta \omega_S + \Delta \omega_I)}{\partial \omega_p} \bigg|_{(\omega_S, \omega_I)} (\Delta \omega_S + \Delta \omega_I) - \frac{\partial k_S(\Delta \omega_S)}{\partial \omega_S} \bigg|_{\omega_S} (\Delta \omega_S) - \frac{\partial k_I(\Delta \omega_I)}{\partial \omega_I} \bigg|_{\omega_I} (\Delta \omega_I)
\] (4.3)

where the signal and idler detunings from the center frequency, \(\omega_p/2\), are represented by \(\Delta \omega_S\) and \(\Delta \omega_I\), respectively. Conventional phase-matching dictates \(\Delta k(\omega_S, \omega_I)\) to be zero. Thus, the phase-matching condition for a different pump frequency \(\omega_p' = \omega_S + \omega_I + \Delta \omega_S + \Delta \omega_I\) determines the signal and idler detunings in terms of the inverse group velocities, \(k'_S(\omega_S), k'_I(\omega_I)\), and \(k_p'(\omega_S + \omega_I)\):

\[
\Delta \omega_S \gamma_S + \Delta \omega_I \gamma_I = 0; \\
\gamma_S = k'_p(\omega_S + \omega_I) - k'_S(\omega_S); \\
\gamma_I = k'_p(\omega_S + \omega_I) - k'_I(\omega_I).
\] (4.4, 4.5, 4.6)

We observe that in order to provide coincident signal and idler emission for a detuned pump frequency, it is required to match the "slope" terms \(\gamma_S\) and \(\gamma_I\). Thus, the parametrization of the phase-matching function for a broadband pump can be expressed to first-order as \(Q(\omega_S \gamma_S + \omega_I \gamma_I)\). The coincident-frequency emission for signal and idler is satisfied if \(\gamma_S = -\gamma_I\), or more explicitly

\[
k'_p(\omega_S + \omega_I) = \frac{k'_S(\omega_S) + k'_I(\omega_I)}{2}
\] (4.7)

which can be viewed as an inverse group velocity matching condition [58, 57]. In the pump-referenced time-frame, one can observe that the restriction imposed by Eq. (4.7) corresponds to signal and idler moving in opposite directions. This is an expected consequence of the positive frequency correlation by Fourier duality, which manifests
itself as anti-correlation in the time domain and is similar to frequency anti-correlation of conventional cw SPDC. A more detailed consideration of achieving this condition in a nonlinear crystal rules out a type-I phase-matching scheme. Since signal and idler are co-polarized in type-I phase-matching, it is not possible to obtain opposite signs for $\gamma_S$ and $\gamma_I$. Thus, only a type-II phase-matching scheme can potentially achieve the inverse group velocity matching condition for the coincident-frequency emission of signal and idler. Note that under the group velocity matching conditions, a second harmonic generation (SHG) process with a fundamental wavelength tuned around $\omega_P/2$ will reveal an extremely broad phase-matching bandwidth [72]. Frequency conversion by SHG ($\omega_1 + \omega_1 \rightarrow 2\omega_1$) coincides with the phase-matching function for SPDC with the restriction that $\omega_S = \omega_I$. Considering only the first order terms in the Taylor expansion naturally “extends” the phase-matching bandwidth until the higher order terms become significant for large detunings. Therefore, in the remainder of this chapter, the standard phase-matching condition $\Delta k = 0$ and Eq. (4.7) will be referred to as the extended phase-matching (EPM) conditions.

The survey of available nonlinear crystals for type-II phase-matching and EPM conditions has been done by various groups [38, 57]. Among these crystals, KTP seems to offer several practical advantages. Quasi-phase matching (QPM) with KTP allows us to use the grating period as a free parameter for phase matching at any set of operating wavelengths within the crystal’s transparency range; i.e., one can always choose a grating period to satisfy the conventional phase-matching condition $\Delta k = 0$. It is important to find the pump and degenerate downconversion wavelengths at which EPM conditions are satisfied. The EPM conditions and the Sellmeier equations for KTP dictate that the coincident-frequency emission for signal and idler can be achieved for degenerate type-II phase matching at a pump center wavelength of around 792 nm [57, 72]. The degenerate photon pairs centered at $\sim1584$ nm can then be detected with Geiger-mode InGaAs avalanche photodetectors (APDs). We note that many commercially available L-band fiber components can be used for this wavelength range. For the pump wavelength of $\sim792$ nm, the required poling period for type-II phase-matching in PPKTP is $\sim46.15$ μm. The aforementioned
ultra-broad SHG scheme had been tested previously with a 1-cm long PPKTP crystal 
($\Lambda=46.1 \mu m$), which displayed a tuning range in excess of $\sim 60$ nm for the fundamental 
centered around 1584 nm [72]. Thus, the prospects of using the same PPKTP crystal 
in downconversion to generate coincident-frequency entanglement was promising.

From the Taylor expansion of the wavevector mismatch, one can also define a 
phase-matching bandwidth for a monochromatic pump input [57]:

$$\Omega_f = \frac{4\pi}{L |\gamma_S - \gamma_I|} \quad (4.8)$$

Substituting the EPM condition ($\gamma_S = -\gamma_I$), one gets

$$\Omega_f = \frac{2\pi c}{L} \frac{|n_{g, P} - n_{g, S}|}{n_{g, P} n_{g, S}} \quad (4.9)$$

where, $n_{g,i}$ ($i = S, P, I$) is the group index with $n_{g,i}(\omega_i) = n_i(\omega_i) + \frac{\partial n_i(\omega)}{\partial \omega} \big|_{\omega_i} \omega_i$. Thus, the phase-matching bandwidth is directly related to the difference of the downconverted photon and pump group velocities. Note that the EPM scheme permits using long nonlinear crystals that effectively confine the emission wavelengths of signal and idler to a narrow bandwidth, as defined by the phase-matching bandwidth in Eq. (4.8). The only restriction to the crystal length arises from the higher order terms in the Taylor expansion. For PPKTP under EPM condition, this upper limit for the crystal length was estimated to be $\sim 20$ cm, which is longer than the crystal dimensions we normally use in practice [56].

In summary, EPM conditions enable a degenerate downconversion scheme where 
individual spectral lines of a broadband pump can be downconverted into distinct 
spectral bins defined by the crystal’s narrowband phase-matching bandwidth. The 
single-photon spectra of the signal and idler are kept identical in this scheme, which 
ensures spectral indistinguishability without any filtering. We can fully utilize the 
broadband spectrum of the pulsed pump without resorting to filtering schemes which 
limit the output flux of the source. The extent of the single-photon spectrum can be much larger than the phase-matching bandwidth. In the time domain, this cor-
responds to a coherent superposition of time anti-correlated photons, which we will explore in the next chapter. We now focus on the experimental characterization of the spectral indistinguishability of the broadband downconverted photons from EPM-conditioned PPKTP.

4.3 Experimental Setup and Initial Results

Figure 4-1 shows the experimental setup for the generation of coincident-frequency entanglement and the HOM interferometric analysis. HOM interference can be used as a tool to test the presence of distinguishable features of two interfering photons [2, 57]. The pump source was a Kerr-lens mode-locked Ti:sapphire laser (Spectra-Physics, Tsunami) that could be operated in either continuous-wave (cw) or pulsed mode without altering its output spatial mode characteristics. In pulsed operation, the mode-locked laser output had a 3-dB bandwidth of \( \sim 6 \) nm and an average power of 350 mW at an 80-MHz pulse repetition rate. For ultrafast pulse-pumped SPDC, we used a 1-cm long and 1-mm thick flux-grown PPKTP crystal (\( \Lambda = 46.1 \, \mu m \)). The crystal facets were dual-wavelength anti-reflection (AR) coated for 792 and 1584 nm. We focused the pump beam into the PPKTP crystal with a beam diameter of \( \sim 200 \, \mu m \).

The output was collimated and two dichroic mirrors (reflecting the 792 nm pump and transmitting the 1584 nm signal and idler) were used to block the pump beam. We did not apply any narrowband spectral filtering to restrict the SPDC output bandwidth. The output beam was then collimated with a 15-cm plano-convex lens and focused with an aspheric lens (\( f = 15.3 \, \text{mm} \)) into a single-mode polarization-maintaining (PM) optical fiber for subsequent HOM interferometric measurements.

The fiber-coupled light was sent to a fiber polarizing beam-splitter (FPBS) that separated the orthogonally polarized signal and idler photons into their respective fiber channels. A half-wave plate (HWP) at the entrance to the PM fiber was used to set up the signal and idler in their appropriate fiber polarization modes. For the construction of the HOM interferometer, the PM patch cords of FPBS were connected to single-mode fibers. In the signal arm of the HOM interferometer was a
Figure 4-1: Experimental setup showing the entanglement source and the fiber HOM interferometer used to test the coincidence-frequency entangled state. DM: dichroic mirror; HWP: half-wave plate; FPBS: fiber polarization beam splitter.

A fiber polarization rotator that was used to match the idler polarization in the second arm. The two arm lengths between the FPBS and the 50–50 fiber coupler were carefully matched by including an adjustable air gap in the idler arm. The air gap was nominally 50 mm-long and efficient coupling (>70%) between the two fibers was achieved by attaching a graded-refractive index collimator to each fiber.

The two outputs of the 50–50 coupler were sent to two fiber-coupled custom-made InGaAs APDs. The design and performance of similar devices have been described in more detail in Ref. [154]. The passively-quenched InGaAs APDs were operated in Geiger mode by simultaneously sending 20-ns gating pulses (3.9 V above the breakdown voltage) to each detector at a repetition rate of 50 kHz, yielding a detector duty cycle of $10^{-3}$. The counter D1 (D2) had a detection quantum efficiency of 16% (24%) at $\sim 1580$ nm, as measured with an attenuated fiber-coupled diode laser input. Under the above operating conditions, the measured dark count rates were 40/s and 20/s for D1 and D2, respectively, which correspond to dark count probabilities of $8 \times 10^{-4}$ (D1) and $4 \times 10^{-4}$ (D2) per gate. The photocount outputs were amplified and sent into a high-speed AND-gate logic circuit for coincidence detection within a 1.8-ns coincidence window [138].
The two-photon output was first analyzed in the case of cw pumping. For 350 mW input pump power, the average singles count probabilities were measured as $2.3 \times 10^{-3}$ (D1) and $2.5 \times 10^{-3}$ (D2) per 20-ns gate, corresponding to count rates of 125/s and 115/s respectively (not corrected for dark counts). The probability of detecting a pair per gate is $2 \times 10^{-5}$ corresponding to a count rate of $\sim 1$ coincidence/s. The low coincidence count rate was due to the interferometer losses ($\sim 65\%$ transmissivity, measured independently), low detector quantum efficiencies, and an estimated 30% coupling efficiency into the PM fiber. From the detection efficiencies and the measurement duty cycle, the estimated single spatial fiber-optic mode pair generation rate was $\sim 4 \times 10^6$/s at 350 mW of pump power, corresponding to $\sim 5\%$ pair generation probability per pulse.

For the HOM measurements, the coincidence counts and the two singles counts were measured as a function of the air gap distance. Accidental coincidence counts were individually measured by separating the gate pulses for both detectors by more than 20 ns so that the detection events are guaranteed to be registered from independent events. The induced optical delay is later compensated by using appropriate lengths of coaxial cables in order to simultaneously bring the time-separated counting events to the coincidence detector [154]. Typical accidental coincidence probabilities within the 1.8-ns coincidence window were $5 \times 10^{-7}$ per gate corresponding to 5 counts in a 200-s measurement time.

Figure 4-2 shows the HOM measurements, corrected for the accidental counts, under cw pumping at the pump wavelength of 792 nm, and similar results were obtained when the pump wavelength was tuned to 787 nm and 797 nm. The two-photon coherence time is related to the phase-matching bandwidth as $\tau_c = \frac{\pi}{\Omega_c}$ [57]. Thus, it is useful to compare the experimentally measured two-photon coherence time to the phase-matching bandwidth of the PPKTP crystal that can be theoretically derived from its Sellmeier equations. From the experimental data, the two-photon coherence time can be related to the base-to-base displacement width of the HOM dip, $l_c$, as $\tau_c = l_c/2c$. Fitting the experimental data with the HOM triangular dip expected for type-II phase-matching yields a visibility of $95 \pm 5\%$ and a two-photon coherence
Figure 4-2: HOM interferometric measurements under cw pumping at 792 nm. The experimental data are fitted with a triangular-dip HOM function (solid line). Base-to-base width of the HOM dip is $l_c = 0.85 \pm 0.16$ mm corresponding to a two-photon coherence time $\tau_c = 1.42 \pm 0.26$ ps. HOM dip visibility is $95 \pm 5\%$.

The HOM visibility is defined by $V = 1 - C_{\min}/C_{\max}$, where $C_{\min}$ is the coincidence count rate at zero signal-idler time delay and $C_{\max}$ is the coincidence count rate at a long time delay (in the flat part of the HOM measurement). The fiber-to-fiber coupling efficiency of the air gap varied by $\sim 10\%$ over the scan, thus the coincidence data was normalized to the maximum singles count rate in Figs. 4-2 and 4-3; this normalization affects the visibility by $\sim 1\%$. The FPBS extinction ratio was measured to be $\sim 99\%$ for both ports. A 2% reduction in the visibility was caused by the 1% leakage of the signal field into the idler channel and vice versa. This 2% loss could be eliminated by using a clean-up polarizer in each of the two output ports of the FPBS. The cw measurements represent the first demonstration of tunable SPDC without any degradation of the two-photon entanglement, a result due to the large extended phase-matching bandwidth of $\sim 67$ nm. The tuning range of 10 nm centered at the
wavelength of 1584 nm is 3 times larger than the two-photon coherence bandwidth.

For the pulsed HOM measurement, the setup was identical to that used in the cw measurements, except the pump laser was set in the pulsed mode centered at 790 nm with a 3-dB bandwidth of 6 nm. Figure 4-3 shows the pulsed HOM measurement results with a visibility of $85 \pm 7\%$ and a two-photon coherence time $\tau_c = 1.3 \pm 0.3$ ps. This was the first observation of high HOM visibility, without spectral filtering, in pulsed SPDC in which the pump bandwidth $\Omega_P$ is much larger than the two-photon coherence bandwidth $\Omega_c$. The visibility in pulsed HOM was sensitive to deviations from the exact EPM operating conditions such as the crystal angle (changing the effective grating period and frequency degeneracy of the signal and idler) and the pump wavelength. The slightly lower visibility in pulsed HOM results compared with the cw measurements may be caused by such deviations. Another plausible explanation for the lower visibility in pulsed HOM interferometry was the presence of occasional independent multiple pairs that can give rise to accidental coincidences.

In order to show that the frequency correlations between the signal and idler are entangled, one can measure the autocorrelation of the signal and idler independently. Single-photon coherence times (equivalently bandwidths) give information about the spectral distribution of the signal and idler, which are not necessarily equal to two-photon coherence time. The autocorrelation can be easily measured by modifying the fiber interferometer. The PPKTP output light was sent through a polarizer to pass only the horizontally polarized signal or the vertically polarized idler photons, then rotated by 45° with a HWP before being coupled into the PM fiber. The fiber interferometer, as depicted in the setup of Fig. 4-1, effectively became a Mach-Zehnder interferometer because the FPBS now served as a 50–50 beam splitter for the input signal or idler (but not both) light. With this autocorrelation setup, we measured a signal coherence time of $\sim380$ fs, and an idler coherence time of $\sim350$ fs which are much shorter than the two-photon coherence time of $1.3 \pm 0.3$ ps.

The high visibility obtained in the pulsed HOM measurements was a consequence of operating under the EPM conditions which allowed for the generation of coincident-frequency entanglement. The signal and idler outputs are frequency-entangled if,
given the generic two-photon state \( |\psi\rangle_{DB} = \int \int \frac{d\omega_s}{2\pi} \frac{d\omega_i}{2\pi} A(\omega_s, \omega_i) |\omega_s\rangle |\omega_i\rangle \), the spectral amplitude \( A(\omega_s, \omega_s) \) cannot be factorized as \( f_S(\omega_S)f_I(\omega_I) \) [38]. If the two photons are not entangled, then \( S_S(\omega_S) = |f_S(\omega_S)|^2 \) and \( S_I(\omega_I) = |f_I(\omega_I)|^2 \) are the fluorescence spectra, which are directly related to their coherence times. If the two interfering photons at the 50–50 coupler of the HOM setup were not entangled, such as those from two single-photon sources, the HOM coherence time would be given by the overlap integral of the two pulses, as shown in Refs. [2, 155, 19]. In our case, the pulsed two-photon coherence time of 1.3 ps is much longer than the single photon coherence times which are less than 400 fs. Such a long two-photon coherence time would not be possible if the two photons were not entangled. The large difference between the measured two-photon coherence time and the single photon coherence times suggested a significant amount of frequency entanglement. However, there are still outstanding questions on the variation of HOM visibility and two-photon coherence time from cw input to pulsed input. In the following section, we present a detailed analysis in order to understand the effects of various pump and crystal parameters on the variation of HOM visibility.

4.4 Improved Two-Photon HOM Interference with Extended Phase-Matched PPKTP

In the previous section, we demonstrated the generation of a broadband frequency entangled two-photon state. By imposing the EPM conditions on a KTP crystal, the spectral distinguishability between signal and idler photons was reduced. Yet, the apparent degradation of quantum interference visibility points out to partial distinguishability between the signal and idler. Therefore, it would be insightful to revisit the EPM conditions in KTP. Specifically, we address the sensitivity of the HOM interference visibility with respect to deviations from the EPM conditions. We experimentally determined the ideal crystal settings that enabled perfectly degenerate emission for signal and idler photons by testing our PPKTP in a difference-frequency
For SPDC, a basic requirement for generating frequency-indistinguishable photons is to ensure that the signal and idler are degenerate. Otherwise, nondegenerate signal and idler interference would show a reduced visibility, because signal and idler fluorescence spectra do not overlap perfectly. This degradation would be more pronounced for a broadband pulsed pump input. Hence, for the particular crystal at hand, it was necessary to determine the exact operating conditions for degeneracy. We can revisit the phase-matching equation for SPDC to find the exact wavelength pair for signal and idler; however, this necessitates precise knowledge of frequency- and temperature-dependent refractive indices for KTP.

For imperfect EPM conditions, the group velocity matching condition is not fully satisfied. Therefore, we need to address the sensitivity of the quantum interference visibility to the deviations from the ideal group velocity matching. It is insightful to refer to the analysis in Ref. [57] which computes the HOM visibility variation as a function of group velocity mismatch. This mismatch is parametrized by the angle $\Theta$. 

Figure 4-3: HOM interferometric measurements for a pulsed pump centered at 790 nm with a 3-dB bandwidth of 6 nm. The experimental data are fitted with a triangular-dip HOM function (solid line). Base-to-base width of the HOM dip is $l_c = 0.8 \pm 0.2$ mm corresponding to a two-photon coherence time $\tau_c = 1.3 \pm 0.3$ ps. HOM dip visibility is $85 \pm 7\%$. 

generation setup.
where

$$\tan \Theta = \frac{k_P'(\omega_P) - k_I'(\omega_P/2)}{k_P'(\omega_P) - k_S'(\omega_P/2)}.$$  \hfill (4.10)

For a 2-dimensional plot of the joint spectral amplitude with EPM conditions, \(\tan \Theta\) gives the slope of the distribution. To understand the \(\Theta\) dependence of the HOM visibility, one can use the model outlined in Ref. [57], where a Gaussian pump spectrum \(\exp(- (\omega - \omega_P)^2 / \Omega_P^2)\) is assumed. We ignore the higher order dispersive terms in the Taylor expansion. The normalized coincidence counts for the HOM interferometer can then be expressed as [57]:

$$C(\tau) = \begin{cases} 
1 - \frac{\sqrt{\pi}}{2} \xi \text{erf} \left( \frac{1 - |\tau|/\tau_\Theta}{\xi} \right) \quad , \quad |\tau| < \tau_\Theta \\
1 \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \q
From the above analysis, one observes that even for a minute deviation from the ideal group velocity matching condition, the interference visibility can degrade very quickly for a broadband pulsed pump. Hence, it is critical to understand crystal dispersion around the operating wavelengths. Note that, the experimental setup in Ref. [72] did not explicitly mention crystal temperature as one of the key parameters and the experimental setup in Sec. 4.3 maintains the same crystal alignment as in Ref. [72], with crystal temperature kept around room temperature. In Ref. [72], a flux-grown PPKTP crystal that housed several gratings with substantially different poling periods was characterized by difference-frequency generation. After determining the EPM conditions by this technique, another flux-grown PPKTP crystal was used with 46.1 μm poling period, however it was not characterized completely. With this latter PPKTP crystal, which is also used in our demonstration of coincident-frequency entanglement, it is necessary to find the degenerate operation and ideal EPM conditions.
Figure 4-5: Sketch of the HOM interference dip as a function of the relative time delay between signal and idler. The coincidence profiles were calculated for the same group velocity mismatch parameters of Fig. 4-4, with the best visibility given by $\Theta = -\pi/4$.

4.4.1 Difference-Frequency Generation with PPKTP

KTP is a biaxial crystal with many practical uses such as efficient doubling of Nd:YAG lasers to 532 nm. At these wavelengths of practical interest, one can find detailed characterization of the material refractive indices. For the purposes of EPM, the wavelengths of interest are $\sim$790 nm and $\sim$1580 nm. Thus, the dispersive properties and refractive indices at these wavelengths are crucial in determining the exact EPM conditions. Namely, the precise knowledge of indices $n_y$ at 790 nm and $n_y$ and $n_z$ at 1580 nm range is necessary. First, we need to determine the variation of $n_z$ and its temperature gradients. Efficient three-wave mixing applications with KTP utilize type-0 interaction with high $d_{33}$ nonlinearity. Therefore, the wavelength dependence $n_z$ was accurately measured for KTP’s entire transparency range [156, 157]. For type-II interaction, the near-infrared refractive index variation for crystal $y$-axis ($n_y$) is also well-known [158], yet the variation at infrared wavelengths is not adequately characterized. König and Wong used a difference-frequency generation scheme with various grating periods for a flux-grown PPKTP to characterize $n_y$ with a modified set of Sellmeier equations [72]. However, the precise value of the refractive indices is
also dependent on the growth procedure and very likely to vary from a flux-grown KTP to a hydrothermally-grown KTP.

For SPDC with a cw pump, the dispersion of KTP and phase-matching condition together determine the output wavelengths. In order to ascertain that signal and idler wavelengths are degenerate, one can use difference-frequency generation (DFG). Through determination of the signal-idler wavelength pair for which the output DFG power is strongest, we can progressively move to the degenerate operating conditions by fine-tuning the crystal temperature and/or the incidence angle. In this scheme, a strong tunable cw Ti:sapphire laser (pump, 787 nm–798 nm) is combined with a tunable cw infrared diode laser (idler, 1569 nm–1595 nm), as seen in Fig. 4-6. The horizontally-polarized pump is modulated by a chopper and the modulation signal is used as the reference input for the lock-in amplifier. The average modulated pump power was 150 mW. The pump beam was focused using an AR-coated 175-mm plano-convex lens. The idler, which is vertically polarized was focused by an AR-coated 125-mm plano-convex lens and combined with the pump using a dichroic mirror. The PPKTP crystal was placed at the focal plane of both input beams. The crystal temperature was monitored with a temperature controller to 0.1°C precision. The pump beam was filtered from the output beam by passing through multiple dichroic mirrors. The DFG output (signal) was polarized horizontally and it was separated from the idler by multiple polarizing beam-splitters. The signal was then focused on an amplified InGaAs pin photodiode with a 25-mm plano-convex lens and the rf output was amplified with a filter bank and detected with the lock-in amplifier. We aligned the input beams and varied the crystal positions for optimal DFG output power.

Since the DFG output was weak and the idler input was in the same wavelength vicinity, it was necessary to use a modulated pump input, which maps the same modulation on the signal. In the experiment, the chopper modulation frequency was ∼660 Hz. Since signal and idler had opposite polarizations one could separate them with polarizing beam-splitters. However, due to imperfect polarization optics some portion of the idler leaked into the detector side and introducing a constant
Figure 4-6: Experimental setup for difference-frequency generation with tunable cw Ti:sapphire (pump) and tunable infrared diode laser (idler). The horizontally-polarized output signal beam was separated from pump and vertically-polarized idler by a combination of dichroic mirrors (DM) and polarizing beam-splitters (PBS).

background. The lock-in filtering scheme eliminated this background and detected the modulated signal beam.

We used the following measurement procedure: For a given pump wavelength, the idler wavelength was varied and the DFG signal power was monitored from the lock-in output. Due to the relatively broad phase-matching bandwidth (∼2.8 nm) of the 10-mm crystal, the tunable range of the IR diode laser was set at ∼7 nm around degeneracy. The DFG tuning curve was then fitted with a \((\sin(x)/x)^2\) function to determine the center wavelength for the idler. Since the pump wavelength was already known, the fitted center determined the corresponding peak wavelength for the signal output. The procedure was repeated for other pump wavelengths to generate a contour plot for the peak signal and idler wavelength pairs. A similar method was employed in Ref. [72] to determine the saddle of the contour plot to locate the EPM center wavelength. The DFG tuning curves for the 10-mm long PPKTP crystal \((\Lambda = 46.1 \mu\text{m}, T=58^\circ\text{C})\) recorded for various pump wavelengths are plotted in Fig. 4-7. The crystal was aligned for near-normal incidence. The recorded center wavelengths correspond to approximately degenerate conditions for signal and idler.

Using the same crystal, the DFG tuning curves and peak signal and idler wavelength pairs were recorded at various temperatures. From these curves, one can observe the sensitivity of the degenerate condition. In Fig. 4-8, the peak idler wavelengths obtained at different crystal temperatures are plotted as a function of the
pump wavelength. The temperature gradient for the idler variation was determined to be 0.041 nm/°C. The degeneracy line \( \omega_I = \omega_P/2 \) is also plotted to guide the eye. Near-degenerate conditions were obtained when the crystal temperature was set to 58°C. It is no surprise that the variation of signal and idler is very close to a unity slope line, a signature of EPM condition \( \Delta \omega_S/\Delta \omega_I \approx 1 \). Note that, one can also vary the incidence angle to obtain near-degenerate emission as in Ref. [72], where the specific incidence angle for near-degenerate case was \( \sim 4.8° \). However, this has various difficulties in practice. For instance, when a QPM grating has limited width, further increase in the incidence angle can cause clipping of the optical beam. Also a large incidence angle can introduce other problems such as spatial dispersive effects or reduced fiber coupling efficiency.

By using a different tunable infrared laser, the DFG measurement was extended to shorter wavelengths, covering 1540 nm to 1595 nm range. With the extended range data for optimal signal and idler wavelengths, we developed a modified Sellmeier
Figure 4-8: Center wavelengths obtained for the idler at various crystal temperatures. Degenerate conditions were obtained at 58°C crystal temperature. The dashed line represents the degenerate tuning line.

equation fit for $n_y$ using the phenomenological model described in Ref. [72]. Using the standard Sellmeier equation:

$$n_y^2 = A_1 + \frac{A_2}{1 - A_3/\lambda^2} - A_4 \lambda^2$$  \hspace{1cm} (4.14)

with $\lambda$ specified in microns, the fitted coefficients are: $A_1 = 2.001024$, $A_2 = 0.829828$, $A_3 = 0.146305$, $A_4 = 0.042750$. The DFG measurements were instrumental in setting up the PPKTP crystal for ideal degenerate emission. Under normal incidence and 58°C crystal temperature, the signal and idler spectra were expected to be very similar under broadband pulsed pumping.

Until now, the near-degenerate signal and idler emission was analyzed and tested for collinear propagation. Due to different phase-matching conditions, the non-collinear SPDC may not emit degenerate pairs. In particular, for the tightly focused pump in type-II SPDC, signal and idler with different polarizations can be generated in non-overlapping spatial modes [159]. Tight focusing for the pump beam is neces-
sary for efficient fiber coupling of signal and idler to achieve high conditional-coupling probability [160]. One has to be cautious about tight focusing for broadband pumping, because when the joint state spatial distribution is projected onto the fiber spatial mode, the fiber coupled signal and idler may end up having distinguishable spectra. Thus, instead of optimizing the fiber coupling efficiency with tight focusing, a loosely focused pump that approximated a plane wave was preferred. As a practical choice, loose focusing limits the single-mode fiber coupling efficiency; however, it preserved the spectral indistinguishability of signal and idler.

4.4.2 Optimized HOM Interference

Using the crystal parameters from our DFG characterization, the HOM interference was re-measured with a weakly focused pump beam. Unlike the first demonstration, we used an interference-filter bank with various bandwidths (1 nm, 2.5 nm, and 4 nm) placed before the PPKTP crystal to vary the input pump spectrum. This is especially useful for monitoring the single-photon autocorrelation times as a function of the pump bandwidth. A 30-cm plano-convex lens focused the pump beam into a $2\omega_0 \approx 200 \mu m$ spot size. As in the previous setup, the output was filtered by multiple dichroic mirrors to reflect the pump and then collimated with a 150 mm plano-convex lens. The polarization of the output was rotated by the half-wave plate to align the signal-idler polarization to match the PM fiber fundamental axes. Unlike the previous demonstration in Sect. 4.3, we inserted a broadband (25 nm FHWM) band-pass filter before the fiber-coupling stage. The band-pass filter was centered at 1585 nm to block the non-phase-matched fluorescence photons. The collimated output beam was then focused by an AR-coated 8 mm aspheric lens into the PM fiber.

The average output power of the pulsed pump was improved to $\sim 1 W$ because our replacement doubled Nd:YVO$_4$ laser had more pump power at 532 m. The pulsed pump spectrum was smoother and more stable at high power outputs. However, this much power was usually not necessary to drive the SPDC, because it was essential to keep multi-pair emissions to low values for HOM interference. The incident pump power level for the PPKTP crystal was controlled by a half-wave plate and a po-
larizing beam-splitter combination. The incident pump spectrum was manipulated by the interference-filter bank and fine adjustments with the pump intracavity-prism insertion. Under pulsed operation, this control gave us the capability to vary the input FWHM bandwidth from 1 nm to 7 nm.

In light of the analysis in Chapter 3, it was necessary to keep multi-pair emission rates as small as possible. Note that, even if the detection rate of single photons is low, the generation rate at the SPDC crystal can be high. Hence, the interference visibility may degrade due to the presence of independent pairs generated by the same pump pulse. It was also necessary to characterize the propagation losses to obtain the conditional-detection efficiency. In addition to the losses listed in Sec. 4.3, the broad-band interference-filter had a peak transmission of \( \sim 60\% \). The conditional-detection efficiency including the detector quantum efficiency for the HOM interferometer was re-measured to be \( \sim 1.5\% \). The main loss contribution comes from the fiber-coupling efficiency, which was measured for the new focusing conditions as \( \sim 20\% \).

If the mean pair generation number, \( \alpha \) for SPDC is kept around 1% per pulse, the observed detection rate due to interferometer loss and detector duty cycle will be only \( \sim 6 \text{ Hz} \). Note that, the Geiger-mode InGaAs APDs have an effective detection window of only 1 ms over one second interval (50 kHz gating frequency, 20 ns pulses). The slow detection inevitably results in low count rates, which necessitate long-term averaging. The targeted pair-generation probability for HOM measurements was \( \alpha \sim 0.5\% \). This required \( \sim 60 \text{ mW} \) input pump power. With the expected singles rate of \( \sim 3 \text{ Hz} \), one had to be careful with the contribution of detector dark counts. With improved detector isolation and bias circuitry, the detector dark count rates were measured as 30/s and 19/s for detector D1 and D2, respectively. The average accidental coincidences resulting from the dark counts were measured as \( 10^{-3} \) counts per second of measurement time for 50 kHz gating frequency, 1.8 ns coincidence window, 20 ns gate pulses.

The HOM interference measurements were performed under low-flux conditions with detected singles rates around \( \sim 35/s \) (\( \sim 25/s \)) for D1 (D2), which includes the detector dark counts. As the air gap delay is varied, each data point was averaged.
over 300 1-s measurements. We observed a peak averaged-coincidence rate at the
edge of the HOM dip of $\sim 0.1/s$. Unlike the first demonstration, we did not subtract
the accidental coincidences. We also used a more standard definition for interference
visibility, $V = (C_{\text{max}} - C_{\text{min}})/(C_{\text{max}} + C_{\text{min}})$. The input pump bandwidth was also
varied such that, four distinct HOM dip traces were obtained for cw, and 1 nm, 3 nm
and 7 nm pulsed pumping. The normalized HOM dip are shown in Fig. 4-9. Important
parameters such as quantum interference visibilities and two-photon coherence times
obtained from the functional-fits are listed in Table. 4.1:

<table>
<thead>
<tr>
<th>Bandwidth [nm]</th>
<th>HOM Visibility [%]</th>
<th>Two-photon coherence time [ps]</th>
</tr>
</thead>
<tbody>
<tr>
<td>cw</td>
<td>97.4 ± 0.25</td>
<td>1.368</td>
</tr>
<tr>
<td>1</td>
<td>85.97 ± 1.63</td>
<td>1.608 ± 0.238</td>
</tr>
<tr>
<td>3</td>
<td>84.19 ± 3.02</td>
<td>1.192 ± 0.328</td>
</tr>
<tr>
<td>7</td>
<td>94.44 ± 0.81</td>
<td>1.320 ± 0.2232</td>
</tr>
</tbody>
</table>

Table 4.1: HOM Interference for various pump spectra.

The tabulated interference visibilities were obtained from the standard visibility
expression, using raw count rates without accidental background subtraction. The
visibility figures for the intermediate pump bandwidths (1-nm and 3-nm) were slightly
reduced due to the use of the pump interference filters. The insertion of these filters
degraded the signal-to-noise ratio of our measurements which resulted in lower inter-
ference visibilities. The two-photon coherence times were extracted using a nonlinear
least-squares fitting algorithm. The functional form for the visibility is taken from
Ref. [57], with $V = \sqrt{\pi} \xi \text{erf}(1/\xi)/(4 - \sqrt{\pi} \xi \text{erf}(1/\xi))$. The same routine is also used
to estimate the 95% confidence intervals for visibility and coherence times. Since the
cw nonlinear fit displayed a rather large error margin, its confidence interval was not
included in the tabulated data set. Note that the two-photon coherence time does
not display a big change as we varied the input pump bandwidth. Again, this is
a consequence of the fact that the timing uncertainty between the signal and idler
is only a function of the phase-matching bandwidth, which in turn depends on the
bimodal group delays in the PPKTP crystal. In other words, the signal and idler pho-
tons experience different group delays due to the distinct group indices for PPKTP’s $y$-axis and $z$-axis, for both cw and pulsed pumping.

Figure 4-9: HOM interference dips for four different 3-dB input pump bandwidths (top-left: cw, top-right: 1 nm, bottom-left: 3 nm, bottom-right: 7 nm). The experimental data points are given by red circles whereas the fitted dip profiles are plotted with solid blue lines. Accidental coincidences were not subtracted.

It is also of interest to see the variation of the single-photon autocorrelation times for both signal and idler as a function of the input pump bandwidth. Once again, the fiber HOM interferometer was converted to a Mach-Zehnder interferometer by following the same procedure in Sec. 4.3. One of the downconverted photons was blocked by inserting a polarizing beam-splitter. The rotation of the half-wave plate in Fig. 4-1 effectively converted the fiber PBS into a 50–50 beam-splitter. The fringe variation resulting from the single-photon interference was observed by dithering the air gap with a piezoelectric transducer (PZT). Short-scale dithering range was 10 μm for recording the depth of local fringes, whereas the mean air gap separation was controlled with a manual actuator. The full fringe structure yielded an envelope whose temporal FWHM value gives the single-photon autocorrelation time. To choose either the signal or the idler photon, we changed the orientation of the polarizing beam-splitter.
cube before the fiber coupling stage to transmit the selected polarization mode. The input power level was not kept as small as that used for HOM measurements. Since the interference filters at the input rejected the majority of pump spectrum, it was necessary to increase the pump power level to as high as $\sim 1 \text{W}$ to achieve appreciable interference fringes. The multi-pair events for single-photon autocorrelation did not change the fringe shape.

The signal-to-noise ratio for the singles counts was on the order of 1. This was combined with another practical difficulty related to the PZT nonlinearity. The position response of the PZT to a ramp voltage input was far from linear. This was also observed in a classical Mach-Zehnder interferometry with the infrared diode laser input, which displayed irregular fringe patterns for a ramp input to the PZT. Thus, a simple regression of a sinusoidal waveform to the singles counts was not reliable. However, due to the presence of two InGaAs APDs after the 50–50 beam splitter, the singles counts displayed negative correlation, i.e. maximum count level at fringe peak on one detector results in minimum counts on the other detector. This negative correlation was practically independent of the PZT response as long as the measurement time was kept long enough and the PZT ramp input was a slowly-varying signal. We recorded the time series of both detectors’ singles counts in 100 1-s time bins where the ramp input for the PZT in $\sim 10 \mu\text{m}$ dither range was 4 mHz. This procedure was repeated for the entire extent of the interference fringes, where the step size for the air gap translation was 20 $\mu\text{m}$. It can be shown that the estimator for the fringe depth was $\tilde{A}(\tau_i) \propto \sqrt{\text{cov}(\tilde{D}_1(x_i), \tilde{D}_2(x_i))}$, where $\tilde{D}_j(x_i)$ represents the time-series data for detector $j$, recorded at the air gap displacement of $x_i$. $\tau_i$ is the relative time delay with respect to the fixed arm. The fringe depth is assumed to be invariant in our local dithering scheme, since the dither range was only limited to 10 $\mu\text{m}$. The extracted fringe depths were plotted as a function of the air gap displacement and single-photon autocorrelation time was measured as the full-width at half-maximum of the envelope. A sample fringe depth data obtained from the local dithering scheme is shown in Fig. 4-10.

We repeated this procedure for both single-photon polarizations at various input
Figure 4-10: Estimated single-photon interference fringe depths as a function of air gap displacement for vertically-polarized idler photon. The input pump bandwidth was $\sim 2.8$ nm. The single-photon autocorrelation time was 752 fs (FWHM) obtained from a Gaussian fit to the data.

pump bandwidths which were controlled by interference-filters and the laser intracavity prism insertion. The measured single-photon autocorrelation times for both polarizations are plotted against the input pump bandwidth in Fig. 4-11. For comparison, we also plot the two-photon correlation times obtained from the HOM interference as well. It is observed that single-photon autocorrelation times rapidly increased with reduced pump bandwidths, whereas the two-photon correlation time was almost invariant as we have suggested previously. The single-photon correlation time becomes comparable to the two-photon correlation time for $\sim 1.5$ nm pump bandwidth. This points out the ideal range of the pump bandwidth that could generate a factorizable or unentangled two-photon state. However, the comparison of correlation times is a rather crude measurement to quantify the exact amount of entanglement. This comparison can be useful to give an approximate parameter range for generating
unentangled states, which can be used for producing a heralded source of pure-state photons for LOQC [39, 37]. Some of the existing techniques to characterize the purity of the heralded state include the measurement of two-photon joint spectral density [64, 37] and spectral density matrix extraction from HOM interference with an attenuated ultrafast laser pulse [161]. We will discuss the feasibility of these measurements in the following chapter.

![Figure 4-11: Single-photon autocorrelation times and two-photon correlation times are plotted against the input pump bandwidth values. Two-photon correlation times are relatively constant whereas the single-photon correlation times are inversely proportional to the pump bandwidth.](image)

**4.5 Summary**

In this chapter, we presented the experimental generation of a coincident-frequency entangled two-photon state which showed the same output spectra for the signal and the idler. Through extended phase-matching conditions in a 1-cm long PPKTP
crystal, we were able to provide broad phase-matching bandwidth which enabled us to utilize the complete spectrum of an ultrafast laser. We achieved the EPM conditions by inverse group velocity matching, i.e., matching the first-order frequency derivatives in the Taylor expansion for \( \Delta k \). As a limitation of this technique, we note the specific requirements for operating wavelengths due to material dispersion of PPKTP. We speculate that an external dispersion mechanism such as waveguide dispersion can be used as a tool to demonstrate EPM conditions at other wavelengths.

By using a fiber-based HOM interferometer, we were able to show the generation of indistinguishable photon pairs and with single-photon autocorrelation measurements we proved that the two-photon state was highly entangled. Pulsed entanglement provided by this quantum state suits the needs of quantum metrology: The timing and positioning measurements can be potentially improved over the standard quantum limit if coincident-frequency entangled photons are used in time-of-arrival measurements [13].

We further improved the two-photon frequency indistinguishability by finding the exact degeneracy points from difference-frequency generation experiments. Through fine-tuning of the crystal temperature we achieved the optimal operating points for the specific PPKTP crystal. These efforts resulted in improved HOM interference visibilities at low-flux range, without accidental background subtraction. For various input pump bandwidths, we measured the single-photon and two-photon correlation times and found the approximate bandwidth range for obtaining factorizable two-photon states. Such quantum states are useful for the heralded generation of pure single-photon states which are important for LOQC.
Chapter 5

Time-Resolved Characterization of Two-Photon State by Upconversion

5.1 Introduction

As we have motivated in Chapter 1, understanding the spectral—and conversely the temporal—correlations of the two-photon state is critical for many quantum information processing applications. In Chapter 4 we studied the two-photon coincident-frequency entangled state that is potentially useful for quantum metrology. Similar highly-entangled multi-photon states can also be used to test interesting quantum effects by utilizing continuous-variable entanglement [12]. On the other hand, it is just as useful to generate multi-photon states with negligible spectral correlations that can be an important building block for linear optics quantum computation (LOQC) [38, 37, 40]. It is known that the probabilistic operation of LOQC quantum gates requires a large number of ancilla photons that should be spectrally identical in a well-defined spatio-temporal mode. In general, supplying these ancilla photons from a multi-photon event such as SPDC is not ideal for high-fidelity gate operation. To see this, consider the ancilla photons being obtained from an SPDC heralding event,
where a detected photon signals the existence of the conjugate photon. Due to the underlying entanglement between the two photons, the quantum state of the heralded single-photon can only be expressed as a classical mixture of pure states [40, 38]. In order to realize high-fidelity operation for a LOQC quantum gate, it is necessary to generate these ancilla photons as a pure state in a single spatio-temporal mode. Thus, in order to use SPDC as a heralded source of pure state photons, one should eliminate the spectral and consequently temporal correlations of the two-photon state. In other words, it is necessary to generate the joint state as a product state: $|\Psi\rangle_{SPDC} = |\psi\rangle_S \otimes |\phi\rangle_I$, where subscripts $S$ and $I$ stand for signal and idler, respectively.

The necessary conditions to achieve a factorizable state were previously analyzed by other groups [38, 39, 40]. One proposal relied on asymmetric group velocity matching in bulk nonlinear crystals to generate a pure heralded single-photon state [38, 40]. This method was recently demonstrated experimentally by the Walmsley group [37] by measuring the HOM interference of heralded single photons from independent downconverters. In this demonstration, the joint spectral distribution of the photon pairs were measured to monitor the two-photon spectral correlations and to verify that these correlations were eliminated for the pure state heralded photons. The same technique could also be used to characterize the amount of frequency-entanglement for a generic two-photon output [38, 64, 65]. Thus, for any given application, a practical tool to measure the underlying frequency-entanglement would be invaluable. We first look at the joint spectral density method for characterizing the two-photon frequency-entanglement.

The spectral and temporal correlations of the two-photon state can be characterized by the joint spectral amplitude (JSA), $\tilde{A}(\omega_S, \omega_I)$, as given in the joint-state expression in Eq. (5.1) [81, 58, 38]

$$|\Psi\rangle_{SPDC} = \int \int \frac{d\omega_S}{2\pi} \frac{d\omega_I}{2\pi} \tilde{A}(\omega_S, \omega_I) |\omega_S\rangle_S |\omega_I\rangle_I.$$  \hspace{1cm} (5.1)

The JSA is a complex-valued function defined as the product of the pump spectral
amplitude and the phase-matching function. Experimental measurement of the JSA profile is a useful tool to understand and characterize the entanglement in frequency domain. For continuous-variable entanglement, the experimental methods to quantify the underlying nonlocal correlations are different from the discrete variable case. To illustrate this, we can consider the projective measurements outlined in Chapter 3 for the polarization of a photon. Using a half-wave plate and a high extinction-ratio polarizer, one can perform the desired projection to any linear polarization basis. We can also realize a similar projective measurement scheme for a continuous degree of freedom.

In the frequency domain, one can use high-resolution spectrometers or tunable band-pass filters to quantify the probability of detecting a single photon in a narrow spectral window. Experimental schemes for measuring the spectral correlations have traditionally relied on narrowband filtering and coincident detection of the signal and the idler to map the joint spectral density (JSD), $|\tilde{A}(\omega_S, \omega_I)|^2$, which can be viewed as a two-dimensional probability distribution [37, 39, 64]. These measurements reveal important information on whether the heralded single photon state is in a pure or mixed state. A disadvantage of this scheme is that the JSA phase profile cannot be extracted. Unless the joint state is transform-limited, it is not possible to infer the time domain profile of the heralded single photons. As a classical analog, one can consider the optical spectrum analyzer measurement of an ultrafast laser, which cannot discriminate between a transform limited pulse or a chirped pulse with the same spectral content.

A more practical limitation of the JSD measurement concerns the applicability in the wavelength regimes where low detector quantum efficiency or high detector noise may be detrimental. This may impact the data acquisition procedure and necessitate long averaging times [38, 64]. On the other hand, the precise mapping of the JSD requires tunable, sharp, narrowband filters to be used for both signal and idler. In the coincident detection scheme, most of the SPDC output is blocked by these filters. Thus the detected count rate may be exceedingly low.

Let us assess the feasibility of the JSD measurement with the coincident frequency-
entangled two-photon state analyzed in Chapter 4. Using the experimentally measured parameters (generation rates, conditional coupling, etc.), we can estimate the coincidence detection rate for a JSD experiment. With \( \sim 1\% \) mean pair generation rate per pulse, \( \sim 1.5\% \) conditional detection efficiency, 1000:1 duty cycle, 80 MHz pump repetition rate, the expected coincidence rate without filters would be \( \sim 0.3/\text{s} \).

The spectral grid size for the frequency scan is determined by the pump bandwidth. Considering a \( \sim 6\text{ nm} \) input pump bandwidth, the square grid size to be scanned is approximately \( 12\text{ nm} \times 12\text{ nm} \). A typical tunable bandpass filter at L-band range has a 100 GHz bandwidth and 3 dB insertion loss. Hence, the spectral measurements can be performed over a \( 15 \times 15 \) square grid with 6 dB additional loss. Due to this spectral slicing and filter losses, the detected coincidence rate will be reduced to \( \sim 3.6 \times 10^{-4}/\text{s} \).

Obviously, this count rate is too low to be useful, because it will be lower than the expected accidental coincidences resulting from detector dark counts \( (\sim 10^{-3}/\text{s}) \). The InGaAs APD speed is the limiting factor for the coincidence detection scheme. The availability of a faster infrared (IR) single-photon detector, such as a superconducting NbN nanowire detector, would enhance the count rates dramatically and enable the JSD measurement to be performed with the frequency-entangled output. Nevertheless, without having access to a fast and efficient IR single-photon detector, mapping the two-photon spectral correlations proved infeasible.

An alternative characterization scheme of the joint state is in the time domain. The two-photon state can also be expressed in terms of time variables. By using the Fourier relation between frequency and time, the frequency-domain annihilation operator can be written as [162]:

\[
\hat{a}(\omega) = \int dt \, \hat{a}(t) e^{i\omega t}, \quad (5.2)
\]

and we rewrite the joint state expression in Eq. (5.1) in terms of time variables:

\[
|\Psi\rangle_{\text{SPDC}} = \int \int d\tau_S d\tau_I A(\tau_S, \tau_I)|\tau_S\rangle_S |\tau_I\rangle_I \quad (5.3)
\]

where, \( |\tau_j\rangle_j \equiv \hat{a}^\dagger(\tau_j)|0\rangle \), for \( j = S, I \). The Fourier analog of the joint spectral ampli-
tude is the joint temporal amplitude (JTA), $A(\tau_S, \tau_I)$. We define the corresponding probability density, $(|A(\tau_S, \tau_I)|^2)$, as the joint temporal density (JTD). JTD can be measured in a manner analogous to the JSD measurement. Instead of using narrow-band spectral filters, one uses narrow temporal filters that can discriminate individual photon arrivals with a time resolution set by the temporal width of the filters. Given that the single-photon autocorrelation times in the coincident-frequency entanglement experiment range around 400 fs, it is necessary to detect single-photons with a time resolution on the order of $\sim100$ fs.

The current timing resolution capability of single-photon detector technology is sufficient for many practical applications such as light detection and ranging [163] and single quantum-emitters with long coherence times [74, 164]. The characteristic time-scale of these single-photon events associated with the narrow atomic or molecular transitions [74, 164] are much longer than the detector timing resolution. State-of-the-art NbN superconducting nanowire detectors can resolve single photon events with a timing jitter of $\sim30$ ps [165]. On the other hand, the high-quantum-efficiency Tungsten transition-edge-sensors typically have $\sim1 \mu$s timing resolution [166, 167]. Commercially available Si APD single-photon counting modules have a timing resolution range of 40 ps to 300 ps. However, these standard detectors are still much slower than the 100-fs timing resolution that we need for a coincident-frequency entangled two-photon state. Our solution is the time-resolved upconversion scheme that not only provides the needed 100-fs timing resolution but also increases the detection efficiency and duty cycle dramatically.

### 5.2 Single-Photon Upconversion

Single-photon upconversion is a well-known technique to frequency-translate IR photons to the visible range, where they can be detected efficiently with Si APDs. As we reviewed in Chapter 2, a single IR photon can be reliably upconverted by virtue of efficient sum-frequency generation (SFG) in a nonlinear crystal with a strong classical pump. Previous demonstrations for single-photon upconversion have shown near-
unity internal conversion efficiencies \[75, 76, 77, 168, 78, 84\]. We tabulate some of the key parameters from these experiments in Table 5.1.

<table>
<thead>
<tr>
<th>System</th>
<th>System detection efficiency [%]</th>
<th>Pump configuration</th>
<th>Nonlinear crystal type</th>
</tr>
</thead>
<tbody>
<tr>
<td>MIT [75]</td>
<td>34</td>
<td>cw (resonant cavity)</td>
<td>Bulk PPLN</td>
</tr>
<tr>
<td>Stanford [76]</td>
<td>46</td>
<td>cw (single-pass)</td>
<td>Waveguide PPLN</td>
</tr>
<tr>
<td>UIUC [168]</td>
<td>25</td>
<td>Pulsed (nanosecond)</td>
<td>Bulk PPLN</td>
</tr>
<tr>
<td>NIST [84]</td>
<td>20</td>
<td>cw/pulsed</td>
<td>Waveguide (PPLN)</td>
</tr>
</tbody>
</table>

Table 5.1: Previous demonstrations of single-photon upconversion.

A common aspect for all previous demonstrations is that the upconversion process does not discriminate individual arrival times of the photons, by design this fact is obvious for cw pumped upconverters. For pulsed configurations, typically nanosecond-long pulses are used to provide a flat and efficient upconversion window for the single-photon wavepackets. Hence, the timing variation of the photons within the pump pulse window can only be observed in this configuration if it can be resolved by the photodetector. As noted earlier, this is not possible at the \(\sim 100\,\text{fs}\) time scale we need for JTD observation.

**5.3 Time-Resolved Photodetection with Upconversion**

In order to obtain precise timing information for single photons, our plan is to use a classical pump which produces ultrafast pulses shorter than the single-photon time scale. The same idea has been used for time-resolved photoluminescence spectroscopy in which fluorescence signatures of multiple quantum emitters were recorded through ultrafast SFG \[169, 170\]. This method enables the time-stamping of single-photon arrivals by effectively using the ultrafast pump as a sampling probe. Therefore, the timing resolution of the system is determined by the ultrafast pump pulse duration and not by the single-photon counting module. This optical sampling technique using 100-fs pulses provides nearly 3 orders of magnitude better time resolution compared
with the fastest single-photon detectors.

We quantify this assertion with a simple theoretical model adapted from continuous-time photodetection theory. The measurement operator for an infinitely-fast unity quantum-efficiency photodetector with integration time $T$ can be expressed as [162, 171]:

$$
\hat{M} = \int_{-\infty}^{\infty} dt \hat{a}^\dagger_t \hat{a}_t
$$

(5.4)

where $\hat{a}(t)$ is the continuous-field single-mode annihilator operator that satisfies the bosonic commutation relation, $[\hat{a}(t), \hat{a}^\dagger(u)] = \delta(t - u)$. The operator $\hat{M}$ is implicitly normalized by $\hbar \omega_0$ to be measured in photon-number units, where $\omega_0$ is the center frequency. We also assume that the natural bandwidth of the system is much smaller than $\omega_0$ [162]. The measurement operator in Eq. (5.4) can be modified to reflect the photodetection process due to ultrafast single-photon detection via upconversion

$$
\hat{U}_i(u) = \int_{-\infty}^{\infty} dt |E_U(t - u)|^2 \hat{a}^\dagger_t \hat{a}_t
$$

(5.5)

where the operator index $i = S, I$ stands for signal and idler modes, respectively. The parameter $u$ specifies the delay of the upconverting pump pulse with respect to the time origin. We also assume that the crystal nonlinearity and other parameters are implicitly contained in the upconverting pulse profile $E_U(t)$, which is taken to be normalized as $\int dt |E_U(t)|^2 = 1$ for convenience. It is possible to develop a more general treatment with non-unity conversion efficiencies that include the vacuum fields, but this simple model can be insightful to understanding the joint detection statistics of the entangled photon pairs. The joint measurement operator for upconverting photodetection and the corresponding singles and coincidence rates for two independent upconverters can be expressed as:

$$
\hat{U}_{SI}(u, v) = \hat{U}_S(u) \otimes \hat{U}_I(v)
$$

(5.6)

$$
n_S(u) = \text{Tr} \left[ \hat{U}_S(u) \hat{\rho}_S \right]
$$

(5.7)
\begin{align}
n_I(v) &= \text{Tr} \left[ \hat{U}_I(v) \hat{\rho}_I \right] \quad (5.8) \\
C(u, v) &= \text{Tr} \left[ \hat{U}_{SI}(u, v) \hat{\rho}_{SI} \right] \quad (5.9)
\end{align}

where \( \hat{\rho}_{SI} \) is the density operator for the joint two-photon state. The single-photon density operators \( \hat{\rho}_S \) and \( \hat{\rho}_I \) are obtained from partial trace operations:

\begin{align}
\hat{\rho}_S &= \text{Tr}_I [ \hat{\rho}_{SI} ] , \quad (5.10) \\
\hat{\rho}_I &= \text{Tr}_S [ \hat{\rho}_{SI} ] . \quad (5.11)
\end{align}

We now explicitly define the density operator for the joint state in time domain variables:

\begin{align}
\hat{\rho}_{SI} &= |\Psi\rangle_{SI} \langle \Psi| \quad (5.12) \\
&= \int \int \int \int dt_S dt_I dt'_{S'} dt'_{I'} A^*(t_S, t_I) A(t'_{S'}, t'_{I'}) |t_S\rangle |t_I\rangle \langle t_S'| \langle t_I'| \quad (5.13)
\end{align}

where the integral bounds are taken from \(-\infty\) to \(\infty\). The continuous-mode single-photon states are defined as \(|t_i\rangle = \hat{a}^\dagger(t_i) |0\rangle\) and the Hermitian conjugation specifies the bra terms. The joint temporal amplitude, \(A(t_S, t_I)\), is given by the Fourier transform of the joint spectral amplitude, \(\tilde{A}(\omega_S, \omega_I)\):

\begin{align}
A(t_S, t_I) &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{d\omega_S}{2\pi} \frac{d\omega_I}{2\pi} \tilde{A}(\omega_S, \omega_I) e^{-i(\omega_S t_S + \omega_I t_I)} \quad (5.14)
\end{align}

In order to evaluate the integrals analytically, we prefer to use Gaussian shaped downconversion pump amplitude and phase-matching function. We perform the Gaussian approximation for the phase-matching function by matching the full-width at half-maximum of a Gaussian pulse and the \(\sin(x)/x\) function [38]. We consider the downconversion pump separately from the upconversion pump because the bandwidth of each pulse can be controlled independently. The JSA with the Gaussian approximation can be written as:
\[ \tilde{A}(\omega_S, \omega_I) = E_P(\omega_S + \omega_I) \Phi(\omega_S, \omega_I) \] (5.15)

\[ = \exp \left[ -\frac{(\omega_S + \omega_I - 2\omega_0)^2}{2\Omega_p^2} \right] \exp \left[ -\frac{(\omega_S - \omega_I)^2}{2\Omega_F^2} \right] \] (5.16)

where \((\omega_S, \omega_I)\) represent the frequencies of the signal and the idler, respectively, around the center frequency, \(\omega_0\), and we have omitted a normalization constant. \(\Omega_P\) is the bandwidth of the downconversion pump, whereas \(\Omega_F\) is the phase-matching bandwidth. Note that the Gaussian approximation for the phase-matching function is a good approximation for the coincident-frequency entangled state with extended phase-matching conditions. Due to the broad phase-matching characteristics in the EPM scheme, the Gaussian approximation only depends on the difference-frequency term, \(\omega_S - \omega_I\). Therefore, this expression is consistent with the functional form for the EPM phase-matching function given in Eq. (4.2) The Fourier transform in Eq. (5.16) becomes separable once we change the variables for sum-frequency \((\omega_+ = \omega_S + \omega_I)\) and difference frequency \((\omega_- = \omega_S - \omega_I)\) terms.

\[ \tilde{A}(\omega_+, \omega_-) = \exp \left[ -\frac{(\omega_+ - 2\omega_0)^2}{2\Omega_p^2} \right] \exp \left[ -\frac{\omega_-^2}{2\Omega_F^2} \right] \] (5.17)

Thus, we can easily evaluate the separable Gaussian integral for Fourier transformation to obtain the unnormalized joint temporal amplitude:

\[ A(t_S, t_I) = \int_{-\infty}^{\infty} \frac{d\omega_+}{4\pi} \exp \left[ -\frac{(\omega_+ - 2\omega_0)^2}{2\Omega_p^2} \right] \exp \left[ -i\omega_+ \left( \frac{t_S + t_I}{2} \right) \right] \times \]

\[ \int_{-\infty}^{\infty} \frac{d\omega_-}{2\pi} \exp \left[ -\frac{\omega_-^2}{2\Omega_F^2} \right] \exp \left[ -i\omega_- \left( \frac{t_S - t_I}{2} \right) \right] \]

\[ = \left[ \frac{e^{-i\omega_0(t_S + t_I)}}{16\pi^3 \Omega_p \Omega_F} \right] \exp \left[ -\frac{\Omega_p^2}{8} (t_S + t_I)^2 \right] \exp \left[ -\frac{\Omega_F^2}{8} (t_S - t_I)^2 \right] \]
We observe that the joint temporal amplitude has different temporal extents for \((t_S + t_I)\) and \((t_S - t_I)\). The sum of the signal and idler arrival times, \(t_S + t_I\), is constrained within a time window of \(\sim 1/\Omega_P\), whereas the difference in their time arrivals can occur within a time interval of \(\sim 1/\Omega_F\). Given that \(\Omega_P > \Omega_F\) in our coincident-frequency entanglement experiment, the timing sum average has a sharper peak than the time difference. This corresponds to temporal anti-correlation, as expected from Fourier duality of the positive correlation in the frequency domain for the coincident-frequency entangled state.

We can express the density operator for the joint state in time variables:

\[
\hat{\rho}_{SI} = N \int \int \int \int \ dt_S \ dt_I \ dt_S' \ dt_I' \left[ e^{i \omega_0 \left( (t_S + t_I) - (t_S' + t_I') \right)} \right] \times \left[ \frac{1}{(16\pi^3 \Omega_P \Omega_F)^2} \right] \times f_1 \left( (t_S + t_I), (t_S' + t_I') \right) \times f_2 \left( (t_S - t_I), (t_S' - t_I') \right) \times |t_S'\rangle \langle t_I'| \langle t_S| \langle t_I|,
\]

(5.19)

where \(N = 4(2\pi \Omega_P \Omega_F)^5\) is the normalization constant to ensure that \(\text{Tr}(\hat{\rho}_{SI}) = 1\).

We also define two-dimensional Gaussian functions \(f_1, f_2\) as:

\[
f_1(\alpha, \beta) = \exp \left[ -\frac{\Omega_P^2}{8}(\alpha^2 + \beta^2) \right], \quad f_2(\gamma, \epsilon) = \exp \left[ -\frac{\Omega_F^2}{8}(\gamma^2 + \epsilon^2) \right].
\]

(5.20)

(5.21)

With the full expression for the density operator, we can evaluate the coincidence rate term given in Eq. (5.9). Substituting the measurement operators for upconversion photodetection in Eqs. (5.5) and (5.6), and using linearity, we get:

\[
\mathcal{C}(u, v) \propto \int_{-\infty}^{\infty} dx |E_U(x - u)|^2 \int_{-\infty}^{\infty} dy |E_U(y - v)|^2 \int \int \int \int \ dt_S \ dt_I \ dt_S' \ dt_I' \times e^{i \omega_0 \left[ (t_S + t_I) - (t_S' + t_I') \right]} \times f_1 \left[ (t_S + t_I), (t_S' + t_I') \right] \times f_2 \left[ (t_S - t_I), (t_S' - t_I') \right] \times
\]

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\[
\text{Tr} \left[ \left( \hat{a}_{S}(t_{S})\hat{a}_{S}(x)\hat{a}_{S}(t_{S'}) |0\rangle_{ss} \langle 0 | \right) \otimes \left( \hat{a}_{I}(t_{I})\hat{a}_{I}(y)\hat{a}_{I}(t_{I'}) |0\rangle_{II} \langle 0 | \right) \right]
\]

(5.22)

The explicit evaluation of the trace operation can be found in Appendix A. Substituting the final value of the trace, \(\delta(x-t_{S})\delta(x-t_{S'})\delta(y-t_{I})\delta(y-t_{I'})\), into Eq. (5.22), we simplify the integrals considerably. For the upconversion pump envelope in Eq. (5.22), we use a Gaussian function with \(|E_{U}(t)|^{2} = \exp\left(-t^{2}\Omega_{U}^{2}/4\right)\), where \(\Omega_{U}\) defines the upconversion pump bandwidth. After substituting the expression for the Gaussian upconversion pump and simplifying the time integrals we obtain:

\[
C(u, v) \propto \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dx \, dy \exp \left[ -\frac{(x-u)^{2}\Omega_{U}^{2}}{4} \right] \exp \left[ -\frac{(y-v)^{2}\Omega_{U}^{2}}{4} \right] \times \exp \left[ -\frac{(x+y)^{2}\Omega_{P}^{2}}{4} \right] \exp \left[ -\frac{(x-y)^{2}\Omega_{F}^{2}}{4} \right].
\]

(5.23)

The Gaussian integral in Eq. (5.23) can be analytically evaluated through a change of variables. The functional form for the coincidence profile in the two-dimensional space parameterized by the signal and idler relative delay with respect to the upconversion pump pulse, \((u, v)\), is:

\[
C(u, v) \propto \exp \left[ -\frac{(u+v)^{2}\Omega_{P}^{2}}{4(1+2\Omega_{P}^{2}/\Omega_{U}^{2})} \right] \exp \left[ -\frac{(u-v)^{2}\Omega_{F}^{2}}{4(1+2\Omega_{F}^{2}/\Omega_{U}^{2})} \right].
\]

(5.24)

We can immediately see that in the limit of infinite upconverting pump bandwidth, or alternatively by setting \(E_{U}(t)\) as a delta function, \(\delta(t)\), the coincidence profile in Eq. (5.24) reduces to \(|A(t_{S}, t_{I})|^{2}\). Thus, we show that the time-resolved detection of single-photons with a classical ultrafast upconverting pulse allows us to measure the JTD distribution accurately and without being limited by the detector speed.

Modifying the traditional single-photon upconversion scheme for the broadband SFG requires several practical considerations:
The phase-matching bandwidth of the nonlinear crystal should be large enough to accommodate the broadband spectra of the upconverting pump and the photons of interest. This requires a short crystal length, which may reduce the peak upconversion efficiency.

The background noise from previous demonstrations originate from pump-induced fluorescence photons at the IR wavelength of interest and subsequent upconversion. Since the output flux of the downconversion source is low (few percent generation probability per pulse), the overall signal-to-noise performance depends on limiting these pump-induced background counts.

In order to monitor the joint statistics of the two-photon state in the time domain, two independent upconverters are needed. This allows coincidence detection using both upconversion modules, where the relative time delays with respect to the upconverting pump pulse can be independently varied.

The upconversion pump pulse should be synchronized to the downconversion pump pulse with minimal timing jitter. This residual timing jitter should be smaller than the single-photon duration.

### 5.4 Experimental Setup

To address the considerations outlined in Sect. 5.3, we chose non-collinear phase-matched SFG configuration with a periodically-poled 1%-mol MgO-doped stoichiometric lithium tantalate (PPMgSLT) crystal as sketched in Figure 5-1. A single PPMgSLT crystal was placed at the focal plane of the intersecting signal, idler and pump beams. Because of the momentum conservation in SFG, the upconverted signal and idler outputs emerge between the input signal-pump and idler-pump beams. The non-collinear phase-matching geometry had several practical advantages. First, the upconverted output would be spatially different from the pump, thus reducing the leakage of the pump light into the detection stage. Second, the non-collinear geometry enables the implementation of two independent upconverters in a single crystal.
with a single pump beam. In this arrangement, the upconverted photons could be easily separated from the pump beam with two pick-up mirrors and detected with two efficient Si APDs to record singles and coincidences. Moreover, the noncollinear geometry permits the two upconverters to be set up in a non-planar configuration to further reduce background coincidences. The PPMgSLT crystal was supplied by the National Institute for Materials Science of Japan at various lengths (5-mm, 10-mm, and 20-mm long). All PPMgSLT samples were poled with 8.5 $\mu$m grating period for type-0 phase-matching process (1580 nm $+$ 790 nm $\rightarrow$ 526.7 nm). The 10-mm sample was cut into 1-mm and 2-mm sections and dual anti-reflection (AR) coated on two sides (input side: 1580 nm & 790 nm, output side: 527 nm & 790 nm). Due to broadband operation requirements, we used a 1-mm PPMgSLT sample.

![Figure 5-1: Non-collinear phase-matching geometry in PPMgSLT crystal.](image)

Figure 5-1: Non-collinear phase-matching geometry in PPMgSLT crystal. The upconverted signal (solid) and idler (dashed) output beams are located between the pump and signal (solid) beams, and the pump and idler (dashed) beams, respectively.

To address the synchronization problem, we drove both the downconversion and upconversion crystals with the same ultrafast pump, centered at 790 nm, thereby removing the timing jitter problem between the two setups. A half-wave plate and a polarizing beam-splitter (PBS) combination tapped a portion of the pump power
for the upconversion setup through the vertical output port of the PBS cube. The transmitted (horizontal) pump polarization was used to drive the downconversion setup.

A detailed sketch of the synchronized downconversion and upconversion setups is shown in Fig. 5-2. The downconversion setup was identical to that used in the $|DB\rangle$-state experiment, where we used the same configuration for focusing the pump beam, collimating the output and filtering the residual pump with dichroic mirrors. Signal and idler were coupled into the polarization-maintaining (PM) single-mode fiber and separated at the fiber PBS. The delays of the signal and idler were adjusted such that they would arrive at the upconversion crystal with the upconversion pump pulse in the same time frame or with the following pump pulse delayed by a full period ($\sim 12.5$ ns). It was optimal to use the pump pulse in the same time frame for upconversion, because this minimized the PM fiber length and therefore reduced the fiber loss and dispersion. In order to provide synchronization with the free-space propagating upconversion pump pulse, the PM fiber lengths of the fiber PBS were set at $\sim 55$ cm. Both PM fiber outputs were collimated with AR-coated aspheric lenses ($f = 11$ mm) for free-space non-collinear SFG alignment. For adjusting the relative delays of the signal and idler with respect to the upconversion pump, we mounted the collimation packages on linear translation stages with manual actuators and a 25-mm travel range.

The upconverter pump height was adjusted with a periscope stage and its polarization was rotated with a half-wave plate for type-0 phase-matched SFG in the PPMgSLT crystal. The pump beam was passed through a non-magnifying telescope stage with a pair of 5-cm focal length AR-coated plano-convex lenses. The purpose of the telescope stage was to modify the upconversion pump beam for optimal mode-matching in the PPMgSLT. We aligned the upconversion pump for normal incidence into the 1-mm long PPMgSLT crystal. The signal and idler beams were aligned parallel to the pump by corner mirrors. The lateral spacing between signal (idler) and pump was approximately 3 mm. All beams were focused by a single plano-convex lens ($f = 50.2$ mm) dual-AR coated for 1580 nm and 790 nm. The collimated 1580 nm
input mode was focused to a beam diameter of $\sim 58\,\mu\text{m}$ at the crystal plane. The off-axis incidence of the 1580 nm inputs to the plano-convex lens induced a slight comatic aberration. The focused pump beam diameter was measured to be $\sim 40\,\mu\text{m}$. With the telescope stage adjustment, we collocated the focal planes of the pump and IR beams at the center of the PPMgSLT crystal to optimize the three-wave mixing efficiency.

We used corner mirrors to separate the upconverted outputs at $\sim 527$ nm from the pump beam. The residual pump in the output beam was further filtered by two dichroic mirrors. Plano-convex lenses with 125-mm focal length and broadband visible AR-coating were used to collimate the upconverted outputs. We also used 10-nm interference-filters centered at 530 nm to provide additional blocking for the pump and the parasitic SHG of the pump at 395 nm. The transmittance of the interference-filters were measured to be $\sim 50\%$. Additional spatial filtering was provided by inserting a pair of 2-mm diameter irises into both beam paths. Using 6.2-mm focal length aspheric lenses, the upconverted outputs were coupled into single-mode fibers with
a \( \sim 3.3 \mu\text{m} \) mode-field diameter. These fibers were connected to Si single-photon counters (or standard Si photodetectors during the testing phase of upconversion measurements).

### 5.4.1 PPMgSLT Characterization with Sum-Frequency Generation

For efficient single-photon upconversion, it is useful to first characterize and optimize the experimental setup in the classical regime, using a tunable IR diode laser with a PM fiber-coupled output. With a fiber polarization controller to adjust the polarization, we injected the diode laser output into the unused input port of the fiber PBS. This gave us the flexibility to adjust the power ratio between the two IR collimator outputs and to achieve proper alignment and power optimization for both upconverters. We adjusted the polarization at the output of each collimator with half-wave plates to be vertical, aligned with the \( z \)-axis of the PPMgSLT crystal. A custom-made crystal oven housed the 1 mm \( \times 5 \) mm \( \times 0.5 \) mm PPMgSLT crystal with a custom-made temperature controller that achieved \( \sim 10 \) mK temperature stability. We positioned the crystal at the focal plane of three intersecting beams with a manual 3-axis translation stage.

For classical sum-frequency generation, we individually tested each IR port with \( \sim 4 \) mW average power from the cw diode laser at 1580 nm. The pump was operated in pulsed mode with 1 W average power. The approximately Gaussian pump spectrum was centered at \( \sim 790 \) nm with a 3-dB bandwidth of \( \sim 6 \) nm. In order to couple the upconverted light into the single-mode fiber efficiently, we connected the output of the fiber into an amplified Si photodetector for alignment with improved sensitivity. After applying a 10 kHz modulation to the diode laser, the rf-output of the photodetector was detected with a lock-in amplifier. Using the filtered lock-in signal, we optimized the fiber coupling efficiency to \( \sim 65\% \). We also found the optimal crystal temperature for the given phase-matching geometry to be \( \sim 22^\circ\text{C} \).

We measured the upconversion phase-matching bandwidth and tuning range by
sending the upconverted light to an optical spectrum analyzer. We tuned the cw IR
diode laser wavelength between 1572.5 nm and 1587.5 nm while the pump was oper-
ated in the pulsed regime. We observed efficient upconversion for the entire \( \sim 15 \) nm
tuning range of the diode laser, which is broader than the single-photon bandwidth
from downconversion. The recorded upconverted spectra at various IR probe wave-
lengths are plotted in Fig. 5-3. For each recorded peak, the 3-dB bandwidth was ap-
proximately 1.4 nm, in good agreement with the upconversion bandwidth predictions
from the MgSLT Sellmeier equations [87]. The 1.4 nm phase-matching bandwidth
indicates that only \( \sim 3.2 \) nm wide portion of the pulsed pump spectrum is utilized for
a given IR probe wavelength. This calculation is consistent with the two-dimensional
bandwidth profile for 1-mm PPMgSLT, sketched in Fig. 2-5. However, the entire
spectrum of the pump was utilized for a broadband IR input as indicated by the
tuning curves in Fig. 5-3. The fluctuations in the peak values of the SFG spectra
were mainly caused by the power variations in the diode laser and spectral ripples in
the pump spectrum.

5.4.2 Time-Resolved Single-Photon Upconversion

The classical SFG experiment enabled us to optimize the crystal parameters for effi-
cient upconversion of single photons from SPDC. Pulsed upconversion at the single-
photon level has subtle differences as compared to classical SFG characterization.
The broadband downconverted photon pairs were coincident-frequency entangled as
discussed in Sect. 5.1 with a well-defined temporal sum location, \( t_s + t_i \), relative to the
ultrafast pump. For proper synchronization between the arrival times of the down-
converted light and the upconversion pump, it was necessary to match the average
timing of the downconverted photons, \( (t_s + t_i)/2 \), to the upconversion pump pulse.
For this purpose, free-space paths and fiber lengths were accurately measured to bring
all beams within 5 mm of each other. We were able to finely tune the timing of signal,
idler and pump independently by using 3 translation stages, which enabled precise
synchronization for all input beams.

For detection of the upconverted photons, the single-mode fibers were connected
to a pair of fiber-coupled Si APDs. In addition to singles counts, we also recorded the coincidence counts between the two Si APDs within a 1.8 ns coincidence window. The ultrafast pump power was initially split evenly between upconversion and downconversion setups and the input pump bandwidth was kept at ∼6 nm. We first monitored the singles counts for matching the upconversion and downconversion beam paths. As we varied the relative timing between the pump and the downconverted photons with the delay stages, we observed the peaks in the singles counts. After synchronization of both upconverters, we optimized the beam alignment and pump power-ratio (∼360 mW for downconversion, ∼580 mW for upconversion) for maximum singles detection rate (∼3500/s at peak, with the background subtracted).

The background level in singles counts were ∼1900/s for the optimal pump powerratio, where the detector dark count rate was only 100/s. The background level cor-
responded to a background probability per pulse of $\sim 2.4 \times 10^{-5}$. This background was still present when the downconverter beam path was blocked. Through different filtering schemes we verified that the observed background was not caused by the parasitically generated 395 nm ultraviolet output from PPMgSLT or the pump beam leaking into the upconverter detection port. We also observed that the coincidence count level due to background was beyond the level that would have resulted from independent noise photons. This suggested that the background was possibly due to the non-phase-matched, non-collinear downconversion of the pump in PPMgSLT. These downconverted photons can be subsequently upconverted by the pump and detected by the Si APDs. The same type of background light was observed in collinear upconversion using cw or pulsed pumping [75, 76, 84, 78, 168]. The non-phase-matched spontaneous emission is expected to be broadband and over a large emission angle, and therefore it should affect both collinear and non-collinear configurations. In order to verify this claim, we measured the pump power dependency of the background counts. Since downconversion and upconversion in the low conversion-efficiency limit scale linearly with the input pump power, the cascaded process should result in quadratic power dependency. The background count rate as a function of the upconversion pump power with the detector dark counts subtracted is plotted in Fig. 5-4. The data points are fitted with a quadratic polynomial, which shows excellent agreement. We also measured the crystal temperature dependence of the background. The non-phase-matched downconversion was observed to be a broadband process with minimal sensitivity to the pump wavelength or crystal temperature. However, the upconversion process has a strong crystal temperature dependence. As we varied the oven temperature, we observed that the background count rate followed the exact temperature profile for the upconversion. This validated our hypothesis on the source of background counts with non-random coincidence statistics.

An important question that relates to the background performance of our source concerns whether the non-collinear phase-matching geometry provides any significant advantage against the collinear geometry used in previous upconverting single-photon detectors [75, 76, 84, 172, 78, 168]. We first observe that the non-phase-matched down-
conversion due to the pump beam in 1-mm PPMgSLT was spatially broad for the frequency bandwidth of interest, where the emission cones of the non-phase-matched parametric output overlapped with the detection modes. This overlap prevents the reduction of the background counts as compared to the collinear configuration. To analyze this, we can pursue a qualitative comparison for the background performance of the time-resolved upconversion scheme with non-collinear phase-matching geometry against previous collinear upconversion schemes. In this comparison, we need to perform appropriate scaling for the conversion efficiency and detection bandwidth to understand the contribution of the background in similar conditions. The first benchmark system we chose was the cw-pumped resonant cavity upconverter with collinear phase-matching geometry from our group [75]. This configuration yielded about 90% internal conversion efficiency, with the detected background rate of $5 \times 10^5$/s. The system detection efficiency was $\sim$34%. The second benchmark system was a pulsed
PPLN waveguide upconverter with near-unity internal conversion efficiency [84, 172]. In this implementation, the pump wavelength (1550 nm) was longer than the single-photon wavelength (1310 nm). The detected background rate and the system detection efficiency with ~100% conversion efficiency was 2400/s and 20%, respectively. We tabulate the detected background rates and subsequent normalization steps in Table 5.2:

<table>
<thead>
<tr>
<th>System</th>
<th>CW system (MIT) [75]</th>
<th>Pulsed system (NIST) [84, 172]</th>
<th>Time-resolved upconverter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Background detection rate [/s]</td>
<td>$5 \times 10^5$</td>
<td>$2.4 \times 10^3$</td>
<td>$1.9 \times 10^3$</td>
</tr>
<tr>
<td>Background generation rate [/s]</td>
<td>$1.4 \times 10^6$</td>
<td>$1.2 \times 10^4$</td>
<td>$1.2 \times 10^4$</td>
</tr>
<tr>
<td>Background generation rate @ 90% conversion efficiency [/s]</td>
<td>$1.4 \times 10^6$</td>
<td>$6 \times 10^3$</td>
<td>$3.5 \times 10^5$</td>
</tr>
<tr>
<td>Background generation rate @ 90% conv. eff. per unit conversion bandwidth [/s/nm]</td>
<td>$4.9 \times 10^6$</td>
<td>$1.2 \times 10^4$</td>
<td>$2.4 \times 10^4$</td>
</tr>
<tr>
<td>Background generation probability @ 90% conv. eff. per unit bandwidth per pulse [/s/nm/pulse]</td>
<td>$4.9 \times 10^{-3}$</td>
<td>$1.9 \times 10^{-5}$</td>
<td>$2.9 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

Table 5.2: Background comparison of time-resolved upconverter against previous collinear single-photon upconverter results.

In order to compare the background rates for the upconverters, we first calculate the background generation rate, which is simply found by dividing the detected background rate with the system detection efficiencies. For the time-resolved upconverter, the system detection efficiency was found to be ~16%. We then approximate the background generation rates for 90% internal conversion efficiency which was the recorded efficiency figure for the cw upconverter. The time-resolved upconverter requires about 5.5 times more pump power to achieve this conversion efficiency, which increases the background generation rate by ~30 times. Following this, we normalize the generation rates by the upconversion bandwidths to adjust for the spectral
content of the detected background. The effective signal conversion bandwidths for
the collinear cw and pulsed upconverters are 0.3 nm and 0.5 nm, respectively. In
comparison, the time-resolved upconverter signal bandwidth is approximately 15 nm.
Thus the broadband non-phase-matched background within this bandwidth can be
efficiently converted to the visible wavelengths. Finally, we compare the bandwidth-
adjusted background generation probability per pulse for all three cases. For the cw
upconverter, we assumed a detection pulse duration of 1 ns which is a typical tempo-
ral width to discriminate independent detection events [168]. The pulsed upconverter
was driven by a pulsed pump with 625 MHz repetition rate. From the adjusted
background rates in Table 5.2, we observe that the non-collinear configuration yields
comparable background generation rates with previous collinear implementations.
Hence, the non-collinear phase-matching geometry does not necessarily improve the
signal-to-noise ratio of the upconverting detector, as we expected.

For the given phase-matching geometry, it is quite difficult to reduce the back-
ground count rates due to the cascaded downconversion and upconversion processes.
One alternative to remedy the pump-induced background level involves shifting the
pump wavelength to be longer than the signal wavelength [84, 76], because Raman
photons from the anti-Stokes process for fluorescence photons are expected to be fewer
than the Stokes photons and significantly fewer than the non-phase-matched output
of the current setup. In a waveguide PPLN-based cw upconversion demonstration,
moving the pump to a longer wavelength and the signal to a shorter wavelength re-
sulted in reduced dark counts by almost a factor of 50 compared to the opposite case,
with high conversion efficiency for both scenarios [76]. Another demonstration with
a pulsed upconversion scheme showed that a longer wavelength upconversion pump
improved the dark counts by a factor of 50 as compared to the cw long-wavelength
upconversion [84]. In our case, a similar approach would necessitate a synchronized
ultrafast optical parametric oscillator (OPO) to drive the upconversion rather than
the 790 nm pump source. If the broadband OPO output for upconversion can be
centered at a wavelength in the (1.6 μm–2 μm) range, then single photon detection
can still be performed with Si APDs.
The accidental coincidence measurements were made with a planar geometry for the upconversion setup of Fig. 5-1 with signal and idler modes being symmetrically tilted with respect to the pump beam. This arrangement, as it turned out, permitted simultaneous coupling and detection of the conjugate background photons. Due to momentum conservation, the non-phase-matched downconverted photons in PP-MgSLT emerged at the opposite points of an emission cone (see Fig. 5-5(a)) and after upconversion they would be detected by the two Si APDs in coincidence.

It should be obvious that the matching of the non-phase-matched photon pair to the detectors can be avoided if we break the planar symmetry of the phase-matching geometry. In this scenario, we raised the height of the collimated single-photon outputs before the focusing lens by \(\sim 1.5\) mm with respect to the pump beam. When the signal and idler photons were injected from the top plane, the upconverted outputs would emerge below the pump beam (as sketched in Fig. 5-5(b)). Since the upconverted signal and idler were no longer positioned at the antipodal points, the
detectors would not simultaneously detect the conjugate background photons. In
other words, for a detected background photon, the conjugate photon was not cou-
ppled to the single-mode fiber of the other detector. The background coincidence rate
after this modification was at a negligible level because the rate would be due to two
independent detection events. After we altered the phase-matching geometry of the
incoming beams, it was necessary to reduce the crystal temperature to 17°C for op-
timal conversion efficiency. We did not observe any appreciable change in the singles
rate under the new configuration.

With this new arrangement we proceeded to study the detailed temporal profile of
the singles rates. In this scheme, we effectively used the upconversion pump pulse as a
sampling probe to monitor the arrival times of SPDC photons. By scanning the pump
pulse through the arrival windows of signal and idler, we obtained the single-photon
histograms for both upconversion channels. In Fig. 5-6 we compare the histogram data
for the SPDC output generated from two distinct gratings of the same PPKTP crystal.
The main grating (Grating 1) with 46.1 μm period was used for the generation of
coincident-frequency entanglement. The auxiliary grating (Grating 2) was originally
designed for a shorter poling period of 46.0 μm, however the characterization with
difference-frequency generation revealed that the effective poling period was very
close to that of the main grating. We plot in Fig. 5-6 the normalized histogram data
for signal and idler from both gratings as a function of the relative pump delay. Each
data point was averaged over 60 seconds. The input pump bandwidth was ∼6 nm
(FWHM) and we subtracted the constant singles background for both channels. The
pump power-ratio between the two setups was adjusted for maximum singles detection
rate (∼360 mW for downconversion, ∼580 mW for upconversion). The signal and idler
arrival time windows were slightly displaced by moving one of the translation stages
to prevent full overlap of the signal and idler histograms.

In general, the timing of a signal photon generated at the beginning and that of
one at the end of the nonlinear crystal are not the same due to crystal birefringence
(or alternatively different group delays for pump and signal). From the Sellmeier
equations for PPKTP [156, 158, 72], the group indices for pump, signal and idler can
be easily calculated, from which we compute the group delay between pump and signal (or idler) for 1-cm long PPKTP crystal as \( \sim 1.47 \) ps. For the coincident-frequency entangled state, the histogram width is given by the two-photon coherence time, because this defines the uncertainty for the relative delay between the downconversion pump and signal (idler). The measured temporal widths for both gratings are approximately 1.3 ps, consistent with the two-photon coherence times of \( \sim 1.4 \) ps obtained from the HOM measurements in Chapter 4.

Since the downconversion pump and signal (idler) have different group velocities in PPKTP, a given time location in the histogram corresponds to a specific position along the crystal. Consider the case where the PPKTP crystal was uniformly poled. Intuitively, one would expect a boxcar shaped singles histogram because it would be
equally probable to detect the generated photon from any location along the crystal. Note that the weakly focused pump would not affect the histogram slope because there would be no clipping and also the generation rate was not dependent on the beam size. Experimentally, as shown in Fig. 5-6, the signal and idler histograms display non-uniform features due to the local inhomogeneities in the crystal grating structure. Primarily the non-uniform generation rate reflect the local poling quality of the grating. For the first time, our time-resolved measurements reveal the quality of the nonlinear crystal at a timing resolution of $\sim 150$ fs or equivalently a crystal location resolution of $\sim 1.1$ mm. Moreover, we found that the poling qualities of two separate gratings on the same crystal were significantly different.

Another observation for the histogram relates to the time-asymmetric structure of the singles profiles. This is not surprising given that coincident-frequency entanglement generates time anti-correlated photon pairs, where signal and idler photons move in opposite directions in the pump time frame. This observed asymmetry is also consistent with the local inhomogeneity hypothesis of a non-uniform histogram profile. The pair generation rate at a specific point on the grating can be simply modeled as carrying an efficiency weighting due to the local poling quality. Once generated, the signal and idler photons acquire equal but opposite-sign delays with respect to the pump as they propagate through the PPKTP crystal. Thus, the spatial variation of the downconversion efficiency was reflected in the signal and idler upconversion profiles that show a mirror symmetry relative to the center of the crystal. The singles histogram measurement therefore is an indirect but simple method for evaluating the grating quality of QPM crystals. It can be used as an in situ diagnostic tool for the poling process. The time-resolved upconversion flux can be monitored in real time to track the progress of local domain reversal. The spatial resolution of this scheme is set by the upconversion pump pulse width and the time span is determined by the two-photon coherence time. Therefore, this characterization scheme only applies to nonlinear crystals that can accommodate ultrafast pump inputs for broadband downconversion.

We applied the same pump scanning scheme to record the coincidence counts
from the two upconverters. Similar to the previous measurement, the data points for the singles and coincidences were averaged over a 60 s acquisition period. The delay stages for signal and idler ports were adjusted to overlap with the arrival windows for both channels. We plot the normalized profiles for singles and coincidences with respect to their peak values in Fig. 5-7, without any background subtraction. The maximum singles (coincidence) rate at the center of the distribution was $\sim 5300/s$ ($\sim 17/s$), including the background. The sharp peak for the coincidence profile had a $\sim 165$ fs FWHM width, which was significantly narrower than the singles histograms. Note that the narrow coincidence peak was a direct consequence of the time anti-correlated generation of signal and idler. As the temporally-short upconversion pulse was scanned through the arrival windows of both photons, the only instance where the two upconverters could simultaneously detect photons was around the time origin. At other times, simultaneous upconversion of the two photons was not possible: when the upconversion pump pulse detected one photon, it would miss the conjugate photon because it was asymmetrically displaced in time.

When the arrival windows of signal and idler are collocated in time, we observed the coincidences of those downconverted photons from the center of the distribution. We were also able to observe coincidences from the photon pairs that were offset from the center. For this purpose, we displaced the signal and idler arrival windows, where the signal was delayed or advanced by 500 fs with respect to the idler. Without changing other experimental parameters, we recorded the singles and coincidences histograms as plotted in normalized form in Figs. 5-8, clearly showing the time location of the anti-correlated photon pairs. For both cases we observed the sharp coincidence peaks with the same $\sim 165$ fs temporal width. The coincidence peaks were equidistant from the center points of the signal and idler distributions, which indicates that the coincident counts were due to time anti-correlated two-photon events.

In the time-resolved upconversion scheme, the upconversion efficiency for single-photon detection is inherently low. Due to the large timing uncertainty of the single-photon timing, only a fraction of the incoming photons overlapping with the pump envelope is upconverted. The mismatch between the pump pulse width and arrival
Figure 5-7: The normalized singles and coincidence histograms by time-resolved upconversion. The pump pulse was scanned through collocated signal and idler arrival windows. The sharp coincidence peak as the center (165 fs FWHM) was a consequence of the temporally anti-correlated two-photon state.

The window size imposes an effective duty cycle that we calculated to be 1:7. We estimated the upconversion efficiency as follows. First, the single-photon histogram is integrated to yield the expected count rate for upconversion if the pump pulse spanned the entire arrival window. This estimate is compared with the detected count rate from a single-photon counting InGaAs APD, whose quantum efficiency (≈20%) and duty cycle (10^{-3}) was previously measured. The filtering and path losses that are not common to both detection schemes (fiber-coupling efficiency: 65%, interference-filter transmission: 50%, Si APD quantum efficiency: ≈50%) and the upconversion duty cycle are also included in the calculation. From the InGaAs APD count rates, we infer the single-photon flux before the upconverter as 5 \times 10^5/s. After the fixed loss factors of 16% and the upconversion duty cycle of 14% were factored out, the effective internal upconversion efficiency per pump pulse is estimated to be ≈25%.
Figure 5-8: Normalized singles and coincidence histograms for offset signal and idler arrival windows. The histograms were obtained by (a) delaying and (b) advancing signal with respect to the idler by 500 fs. Symmetrically displaced sharp coincidence peaks resulted from off-center SPDC photons.

This efficiency was mainly limited by the available pump power.

5.4.3 Joint Temporal Density Measurement

Through time-resolved upconversion for signal and idler, we were able to qualitatively verify time anti-correlated generation. However, with a one-dimensional pump scan we cannot fully characterize the joint temporal statistics. The scanning of the sampling probe corresponds to the measurement of $C(u, u)$, for collocated signal and idler arrival windows. A more complete characterization is achievable by varying the signal and idler relative delays independently. In this section, we will explore the measurement capability to map the joint temporal density of the two-photon state, as discussed in Sect. 5.3.

For the JTD measurements, we varied the signal and idler delay stages independently, while keeping the upconversion pump delay constant. The pump bandwidth was set to $\sim 6\text{nm}$, with an optimal pump power-ratio between upconversion and downconversion setups. We recorded the coincidence counts over a two-dimensional measurement grid, $2\text{ps} \times 2\text{ps}$ with 133 fs delay steps for each channel. Each data point was averaged over a 60-second measurement interval. The normalized coincidence data is shown as a surface plot over the two-dimensional time grid in Fig. 5-
9. The two-dimensional coincidence profile exhibits clear time anti-correlation, thus verifying the coincident-frequency entanglement in time domain. We also plot the theoretical joint temporal density profile in the same figure, analytically calculated from the inverse Fourier transformation of the joint spectral amplitude. We assumed a Gaussian pump spectrum and a $\sin(x)/x$ profile for the phase-matching function. In time domain, the inverse Fourier transform of the phase-matching function causes the boxcar variation along $t_S - t_I$ axis, where $t_S$ and $t_I$ indicate the signal and idler relative delays with respect to the time origin, respectively. Similarly, we observe a Gaussian variation along $t_S + t_I$ axis due to the assumption of the Gaussian pump spectrum. This calculation did not include the effect of finite upconversion pump bandwidth. We observe that the experimental JTD data exhibits good agreement with the theoretical JTD profile.

Figure 5-9: Left panel: Joint temporal density for $\sim$6 nm downconversion pump bandwidth. Strong temporal anti-correlation was observed with the highly eccentric elliptical distribution. Right panel: Theoretical calculation for joint temporal density with $\sim$6 nm pump bandwidth.

As explained earlier, the JTD measurement is the Fourier analog of the often used JSD measurement. In JSD measurements one uses narrowband filters with associated fixed insertion losses. For the JTD measurement that we have realized for the first time, we used narrow temporal filters in the form of ultrashort upconversion pump pulses. Moreover, the temporal filtering in the JTD measurement can be made nearly
lossless because the internal upconversion efficiency can be made to approach unity, as
demonstrated in previous upconversion experiments. Thus, JTD measurements and
JSD measurements are complementary techniques to yield a more complete charac-
terization of two-photon states. JTD measurements can also be much more flexible
because the temporal filter width can be easily adjusted. We further note that the
JTD data can readily reveal some important temporal metrics that are alternatively
measured by other techniques. For example, the single photon autocorrelation time
for signal can be obtained from the two-dimensional JTD data by tracing over the
idler subspace. For the given JTD profile, this operation can be approximated by
reading the temporal width of the distribution along the constant idler time variable,
i.e., along \( C(u, v_0) \) line since the temporal width of the projection does not exhibit
a significant variation for different values of \( v_0 \). From the measured JTD profile in
Fig. 5-9, we can estimate a single photon autocorrelation time of \( \sim 340 \) fs, which agrees
well with the Mach-Zehnder interference measurements from Chapter 4.

Because the downconversion and upconversion setups were separately driven, we
had the freedom of filtering the downconversion pump bandwidth to manipulate the
joint temporal amplitude without affecting the upconversion timing performance.
By using an interference-filter bank before the PPKTP crystal, we first observed
the variation of the singles and coincidence histograms for various downconversion
pump bandwidths (3.6 nm, 2.1 nm, and 1.1 nm), following the procedure outlined in
Sec. 5.4.2. The additional timing delay induced by insertion of the interference filters
was compensated with delay stages for the signal and idler. Also, the pump power-
ratio for each bandwidth setting was optimized.

The normalized histograms for three different bandwidths are plotted in Fig. 5-10(a-c). Since the filters blocked a significant portion of the pump spectrum, the
available pump power for downconversion was reduced with narrower filter band-
widths. This was compounded by insertion losses for the filters, ranging between
20% and 40%. Hence, we observed that the signal-to-noise ratio in the singles pro-
file degraded as the interference bandwidth decreased. Because of the poor signal
level, the 0.5-nm interference-filter was not used. Another important parameter that
changed with the filter bandwidth was the temporal width of the coincidence peak. When the downconversion pump bandwidth was reduced, the single-photon coherence time increased. The coincidence profile width was proportional to the single-photon coherence time, resulting in wider coincidence peaks for reduced filter bandwidths. For comparison, we plot the coincidence profiles for different downconversion pump bandwidths in Fig. 5-10(d).

Note that with the experimental values for the phase-matching bandwidth, the upconversion and downconversion bandwidths, the theoretical predictions from Eq. (5.24) can be confirmed. The upconversion pump scan corresponds to tracing the $C(u, u)$ curve with the relative delay, $u$, as the free parameter. Hence, after substituting the bandwidth values into Eq. (5.24), we produce the theoretically predicted coincidence profiles in Fig. 5-10(d). The temporal FWHM values from fits to experimental data and theoretical curves are tabulated in Table 5.3. We observe an excellent agreement between experimental data and theoretical predictions. Note that the necessary parameters for calculating the theoretical coincidence profile were experimentally verified with our previous measurements. For instance, the two-photon coherence bandwidth, which is given by the phase-matching bandwidth, was measured by the HOM setup in Chapter 4. The upconversion and downconversion pump bandwidths were also separately measured. Moreover, in our calculation of the joint temporal amplitude we did not assume a quadratic spectral phase profile (or chirp) on the two-photon joint spectral amplitude. Therefore, the temporal widths of the coincidence profiles predicted by our JTD represent transform-limited durations. Hence, the agreement with the experimental data in the temporal widths shows that the single photons were nearly transform-limited. This observation was only possible with time-domain measurement techniques since any spectral filtering scheme would be insensitive to dispersive broadening of the single photons.

We extended the filtered downconversion pump scheme to the JTD measurements. For 3.6 nm and 2.1 nm downconversion pump bandwidths, the grid size for signal and idler time delays was maintained at 2 ps × 2 ps (with a 133 fs step size). We increased the grid size to 4 ps × 4 ps (266 fs step size) for 1.1 nm pump bandwidth. As before,
the pump power-ratio was optimized for each filter. Also, the coincidence data points were obtained from 60 second averaging. In Fig. 5-11(a-c), we map each coincidence profile onto a surface plot and also sketch the contour plots over the measurement grid. The theoretical JTD profiles calculated for each pump bandwidth are plotted separately in Fig. 5-11(d-f). We observe that the variation of the JTD profile for three distinct pump bandwidths are consistent with the theoretical JTD curves.

Note that, with the reduced pump bandwidth, the eccentricity of the JTD elliptic contour plots becomes closer to zero, resulting in a more symmetric distribution. We expect that when the JTD profile becomes more symmetric, the two-photon temporal and spectral correlations would be reduced. Given transform-limited single photons, the rotationally-symmetric (in time) JTD profile would correspond to an unentangled two-photon state [40]. The JSD measurement would be insensitive to dispersive phase shifts acquired by the downconversion pump, signal or idler. Thus, the single-photon wavepackets that are broadened beyond the transform limit cannot be effectively characterized only by JSD measurements. The JTD measurement technique outlined in this chapter would be a useful tool when used alongside JSD measurement, which can detect dispersively broadened single-photon pulses.

### 5.5 Quantifying Continuous-Variable Entanglement

In the previous chapter, we showed that ultrafast pumped downconversion in PPKTP produces frequency-entangled two-photon states. Our approach was comparing the time scales of single-photon and two-photon coherence times obtained from interfero-

<table>
<thead>
<tr>
<th>SPDC Pump Bandwidth [nm]</th>
<th>Experimental FWHM [fs]</th>
<th>Theoretical FWHM from Eq. 5.24 [fs]</th>
</tr>
</thead>
<tbody>
<tr>
<td>6</td>
<td>165 ±25</td>
<td>185</td>
</tr>
<tr>
<td>3.6</td>
<td>278 ±25</td>
<td>281</td>
</tr>
<tr>
<td>2.1</td>
<td>456 ±27</td>
<td>425</td>
</tr>
<tr>
<td>1.1</td>
<td>932 ±43</td>
<td>874</td>
</tr>
</tbody>
</table>

Table 5.3: FWHM values from experimental and theoretical coincidence profiles.
metrical measurements. In this chapter, we have verified the two-photon entanglement by JTD measurement and further analyzed the variation of this entanglement under different input pump bandwidths. However, we need a quantitative tool to characterize this continuous-variable entanglement which can enable us to assign a “measure” of entanglement for different observations of joint spectral or temporal statistics.

Analysis of entanglement in frequency is different from that of a discrete-variable entanglement such as polarization entanglement. Conventionally, entanglement quantification in position, momentum and frequency degrees of freedom are analyzed in the framework of Schmidt decomposition \([173, 174, 175, 176, 177]\). This technique can be viewed as a quantum analog of the singular value decomposition. In this formal-

Figure 5-10: Normalized singles and coincidence histograms for various downconversion pump 3-dB bandwidths: (a) 3.6 nm, (b) 2.2 nm, (c) 1.1 nm. (d) The coincidence peaks are plotted separately for comparison.
Figure 5-11: Experimental joint temporal densities for various downconversion pump 3-dB bandwidths: (a) 3.6 nm, (b) 2.2 nm, (c) 1.1 nm. Theoretical joint temporal densities for the given pump bandwidths are also plotted for comparison: (d) 3.6 nm, (e) 2.2 nm, (f) 1.1 nm.
ism, the joint spectral amplitude, $\tilde{A}(\omega_s, \omega_i)$, or joint temporal amplitude, $A(\tau_s, \tau_i)$, is expressed as a discrete sum with products of frequency or time eigenmodes, which is called Schmidt decomposition:

$$\tilde{A}(\omega_s, \omega_i) = \sum_n \sqrt{\lambda_n} \tilde{\psi}_{S,n}(\omega_s) \tilde{\phi}_{I,n}(\omega_i),$$

$$A(\tau_s, \tau_i) = \sum_n \sqrt{\lambda_n} \psi_{S,n}(\tau_s) \phi_{I,n}(\tau_i). \quad (5.25)$$

The frequency eigenmodes for signal and idler are $\{\tilde{\psi}_{S,n}(\omega_s)\}$ and $\{\tilde{\phi}_{I,n}(\omega_i)\}$ and the temporal eigenmodes $\{\psi_{S,n}(\tau_s)\}$ and $\{\phi_{I,n}(\tau_i)\}$ can be obtained from the Fourier transforms of the frequency eigenmodes [173]:

$$\psi_{S,n}(\tau_s) = \int d\omega \tilde{\psi}_{S,n}(\omega) \exp(-i\omega \tau_s),$$

$$\phi_{I,n}(\tau_i) = \int d\omega \tilde{\phi}_{I,n}(\omega) \exp(-i\omega \tau_i). \quad (5.26)$$

From now on, we consider the analysis of continuous-variable entanglement with the temporal eigenmodes. These eigenmodes are obtained from the self-adjoint operators defined by the joint temporal amplitude using the spectral theorem [173, 177]:

$$\lambda_n \psi_{S,n}(\tau_s) = \iint d\tau' d\tau'' A(\tau_s, \tau'') A^*(\tau', \tau'') \psi_{S,n}(\tau_s),$$

$$\lambda_n \phi_{I,n}(\tau_i) = \iint d\tau' d\tau'' A(\tau'', \tau_i) A^*(\tau'', \tau') \phi_{I,n}(\tau_i). \quad (5.27)$$

These eigenvalue equations can be numerically solved to reveal the temporal eigenmodes and corresponding eigenvalues. In certain cases, such as the Gaussian approximation for the joint temporal amplitude, it is possible to obtain analytical expressions for the eigenvalues [38]. The presence of more than one nonzero eigenvalue is sufficient to show the inseparability of the two-photon state. As a quantitative measure of the
continuous variable entanglement we can investigate how many significantly nonzero eigenvalues are present in the decomposition. For well-behaved joint distributions, the first few largest eigenmodes are sufficient to express the joint temporal amplitude with high accuracy [173, 177]. From these significant eigenvalues, we can compute the von Neumann entropy of the state, which is sometimes called the entanglement entropy [173, 177]:

\[
S = - \sum_{k=1}^{n} \lambda_k \log_2 \lambda_k.
\] (5.28)

Note that an \(S\) value of zero indicates a factorizable state, with single temporal eigenmodes for signal and idler. With larger \(S\) values, the two-photon state is more entangled. For a continuous variable two-photon state, there is no theoretical upper limit for \(S\), since there is a possibility to have infinitely many frequency eigenmodes.

For the coincident-frequency entangled state, the entanglement entropy can be computed to study the effects of important parameters such as the input pump bandwidth or the phase-matching bandwidth. In particular, we are interested in calculating theoretical predictions for the entanglement entropy to compare with the same entanglement metric derived from the measured joint temporal densities. We first calculate the entanglement entropy from the Schmidt decomposition formalism as outlined earlier. The eigenvalue problem is solved for two cases of interest. In the first case, the joint spectral amplitude is approximated by a two-dimensional Gaussian distribution given in Eq. (5.16). This requires the phase-matching function to be replaced by a Gaussian function with the same FWHM spectral width. The pump amplitude is also taken as a Gaussian function, whose spectral width is kept as a free parameter in our calculation. Once we solve the eigenvalue problem, the resulting eigenvalues can be used to compute the entanglement entropy from Eq. (5.28). For the second case, we keep the phase-matching function in its original \(\sin(x)/x\) form, where the argument is given by the phase mismatch, \(\Delta kL/2\). We numerically compute this phase mismatch to second order in accuracy. The resulting joint spectral amplitude is used for computing the entanglement entropy as a function of the input
pump bandwidth. It has to be noted that we assume the two-photon state to be transform-limited, i.e. the two-dimensional phase profile for the joint spectral amplitude is taken as constant. For comparison with the theoretical predictions, we also calculate the entanglement entropy from the experimentally obtained joint temporal densities. The calculated entropy values are plotted against the respective down-conversion pump bandwidths. These values are plotted in Fig. 5-12 along with the theoretical curves for the two scenarios.

![Figure 5-12: Entanglement entropy for the two-photon state with different pump bandwidths. The theoretical entropy variation as a function of input bandwidth was calculated for both Gaussian approximation and sin(x)/x form for the phase-matching function. The experimental JTD distribution was used to calculate the entropy for various downconversion pump bandwidths.](image)

We observe a good qualitative agreement between the theoretical values and the calculated values derived from the experimental JTD distributions. Note that the
Gaussian approximation results in a fully-factorizable two-photon state for a pump bandwidth of $\sim 1.2\,\text{nm}$. This limit corresponds to the joint state being a product of two Gaussian envelopes for the frequency and time domain representations of the signal and idler photons. These two photons are spectrally or temporally uncorrelated and can be treated as two independent photons. The entropy values for the theoretical $\sin(x)/x$ form of the phase-matching function does not reach zero, yet the theoretical two-photon joint state is close to a factorizable form for $\sim 1.2\,\text{nm}$ pump bandwidth. We also note that the experimental JTD distributions yield smaller entanglement entropy values as compared to the theoretical entropy curve for $\sin(x)/x$ form of the phase-matching function. This is mainly due to the sharp boundary of the theoretical JTD profile along $t_S + t_I = 0$ axis, as sketched in Fig. 5-11(d-f). The boxcar shape of the ideal phase-matching function in time-domain necessitates inclusion of higher order Schmidt modes and increases the entanglement entropy. However, as indicated in the singles histogram measurements of Fig. 5-7, the actual time-domain profile of the phase-matching function is smoother. Therefore, the experimental JTD distributions can be expressed with a smaller number of Schmidt modes, resulting in lower entanglement entropy.

Besides entanglement entropy, we can also calculate the purity of the single photon output, which can be readily obtained from the eigenvalues of the Schmidt decomposition [173, 39]:

$$p = \text{Tr}(\hat{\rho}_S^2) = \sum_{n=0}^{\infty} \lambda_n^2. \quad (5.29)$$

The purity of the heralded single photon can show a great variation depending on the selected downconversion pump bandwidth. For example, the broadband downconversion output with 6 nm pump bandwidth yields a purity of $\sim 0.38$, which is a consequence of the high degree of frequency entanglement. As we reduce the pump bandwidth to generate a symmetric JTD profile, the purity increases. Also, the entropy values for the JTD distributions exhibit a monotonic variation with the pump bandwidth, with the distribution obtained for 1.1 nm bandwidth being closest to a factorizable form. At this pump bandwidth, the calculated purity from the experi-
mental JTD profile is $\sim 0.88$, which indicates a nearly unentangled two-photon state.

We also note that, our highest purity value compares well with other implementations for factorizable two-photon states. For instance, in Ref. [37] a purity of 0.95 was reported for heralded pure-state single-photon generation via asymmetric group velocity matching. We observe that the fluctuations in the measured coincidence profiles gave rise to higher order eigenmodes and consequently increased the entanglement entropy and reduced the output state purity. This condition was more pronounced for low-bandwidth JTD measurements, where the downconversion pump power was reduced due to tight spectral filtering and the signal-to-noise ratio for coincidence measurement was degraded. Note that a smoother JTD distribution would exhibit lower entropy values and higher purities for factorizable output state generation. A more precise control over the pump bandwidth would also facilitate future efforts to generate single photons in pure quantum states.

The comparison for the entanglement entropy and single-photon purity emphasized our capability to engineer temporal and spectral correlations for the two-photon state. We believe that the JTD measurement technique is a powerful tool to monitor these correlations as it complemented our interferometric measurements to characterize and control two-photon frequency entanglement.

5.6 Summary

This chapter investigated a new measurement tool to characterize multi-photon temporal correlations. We designed and implemented a pulsed upconversion experiment for single-photon detection with sub-picosecond timing resolution capability. The non-collinear phase-matching geometry in a PPMgSLT crystal enabled us to realize two independent upconverters for signal and idler and eliminated the background noise in coincidences resulting from pump-induced non-phase-matched parametric fluorescence photons. This upconversion setup was easily integrated with the down-conversion experiment from the previous chapter. We used the same ultrafast pump source to drive both experiments synchronously, which gave us the capability to mea-
sure the joint temporal density without residual pump timing jitter. This measurement was done by recording coincidence statistics from the signal and idler upconverters with independently varied time delays. The resulting distributions for the joint temporal density verifies the continuous-variable entanglement in our experiment. We were able to vary the shape of JTD by controlling the downconversion pump bandwidth independently from the upconversion part. This controllable frequency-entanglement was also verified when we observed a remarkable agreement between the entanglement entropies from the experimentally obtained JTD data and the theoretical models. Such a characterization technique in the time domain would complement the frequency-domain techniques to understand and manipulate multi-photon entanglement for quantum information processing applications.
Chapter 6

Conclusions

In this thesis, we focused on several practical applications of entanglement which, as a novel resource, has already had a significant impact on modern-day computer science, cryptography, metrology, communication and many other quantum information processing (QIP) applications. The question of what physical modality constitutes the ideal platform for building a quantum computer or a quantum memory still remains open today. However, it is certain that the transmission of quantum information can be most reliably achieved with photons. Therefore, engineering the quantum state and quantum correlations of multi-photon systems is of utmost importance to QIP applications such as quantum key distribution (QKD) and precision metrology.

QKD is already a commercially active field with many successful demonstrations on various platforms. However, there are still open questions that relate to the performance and security of various QKD protocols under different practical conditions. We focused on an application specific solution, namely QKD over free-space line-of-sight communication channels. Our approach was to evaluate the restrictions imposed by the free-space channel and to adaptively design the building blocks of a QKD cryptosystem. We considered the Ekert protocol for QKD which relied on entanglement. For the physical means to generate entanglement, we used spontaneous parametric downconversion (SPDC). SPDC is a well-established, practical technique to produce high-purity, high output-flux entangled photon pairs where internal degrees of freedom (frequency, polarization, momentum) can be accurately controlled. With the
capability to tailor two-photon correlations, we focused on ultrafast pumped SPDC which offered a well-defined timing for the generation of the photon pairs.

The spectral and temporal filtering requirements due to ambient solar background have dictated the downconversion source design to be both pulsed and narrowband. For optimal key-generation rate, the transparency of the free-space channel and quantum efficiency of the silicon APDs were taken into account and this determined a degenerate operation for the pulsed downconversion source at \( \sim 780 \text{ nm} \). For a compact and self-contained system we also had to consider the integration of a pulsed, narrowband pump source at \( \sim 390 \text{ nm} \).

Our modular design comprised a pulsed narrowband high power ultraviolet (UV) source and a polarization-entangled photon source based on a polarization Sagnac interferometer. The high power UV source design utilized a narrowband passively modelocked fiber laser in a master-oscillator power amplifier configuration. The amplified infrared output was frequency-quadrupled with two efficient second harmonic generation (SHG) stages. Throughout the design and testing of the UV source, we addressed and resolved several experimental challenges. These included the mitigation of self-phase modulation in erbium-doped fiber amplifier and growth-dependent UV-induced absorption in PPKTP. The output power and the stability of the UV source was sufficient to drive multiple SPDC systems for high-flux output. In light of experimental results from our downconversion source, a future design impetus can focus on increasing the pulse repetition rate of the UV source which can be achieved by various schemes such as active-passive modelocking, repetition rate multiplication with an external cavity, etc. We achieved \( \sim 400 \text{ mW} \) average output power from the UV source and with a better quality SHG stage 1 W pulsed output power is within reach. This power level would enable future stand-alone utilization of the UV source for micro-machining or fluorescence spectroscopy. We also note that the modular UV source design is suitable for other quantum optics applications. For instance, the narrowband, high power (\( \sim 3.5 \text{ W} \)) first SHG output was suitable to be used in a phase-conjugate optical coherence tomography system [178]. Alternatively, the UV source can also be used in conjunction with an array of downconverters for on-demand
single-photon generation [179].

The UV source was successfully integrated with the downconversion setup. The polarization entanglement from the downconversion in type-II PPKTP was based on an earlier cw-pumped design, which utilized a polarization Sagnac interferometer. This design was shown to be superior to previous interferometric schemes, since phase stability and output indistinguishability conditions were automatically satisfied. Through various characterization techniques, we verified the generation of high-purity polarization entanglement in the low output-flux regime. We observed the degradation in the quantum-interference visibility due to generation of multiple pairs of photons in a single pump pulse. We rigorously characterized this visibility degradation and established its connection with the mean photon pair generation probability per pulse. This characterization was essential for comparison between a cw-pumped and a pulsed-pumped system. By increasing the repetition rate of the pump source, the boundary for multiple-pair generation can be pushed to higher photon-flux levels, only to be limited by the speed of the single-photon detectors.

The downconversion setup was highly efficient where only 1 mW pump power was needed to produce $\sim 0.01$ pair per pulse at the repetition rate of 31.1 MHz. At this rate, the quantum-interference visibility was $\sim 97\%$ in the $+45^\circ/-45^\circ$ basis. With the available pump power, we observed a pair generation probability as high as 0.7 at the expense of degraded quantum-interference visibility due to multiple pairs. This pronounced generation of multiple pairs opens up other avenues to study higher-dimensional entanglement and realization of novel quantum states for linear optics quantum computation (LOQC). Given the efficiency of the downconversion source, it is possible to generate and detect higher-dimensional entanglement without resorting to kilohertz-rate regeneratively-amplified pump configurations. If the damage thresholds of the downconversion crystal can be scaled up, it should be possible to generate these multi-partite entangled states efficiently with megahertz repetition-rate pump source such as the UV source presented here.

In the second half of the thesis, we adopted a different perspective for the pulsed SPDC. We transitioned from narrowband output to broadband output and from
polarization entanglement to frequency entanglement. The premise of the novel frequency-entangled quantum state was to perform precision measurements beyond the standard quantum limit. Previous theoretical work in quantum metrology suggested that a positive frequency-entangled two-photon state may show a factor of $\sqrt{2}$ improvement in the accuracy of time-of-flight measurements. However, such type of entanglement does not naturally arise from femtosecond-pulse pumped SPDC. In order to generate such high-purity frequency-entangled state, it was necessary to eliminate the frequency distinguishability of the SPDC output. This was a well-known problem in quantum optics community which was previously mitigated by narrow-band filtering schemes that reduced the output flux.

Our work was built upon the previous theoretical studies on the generation of frequency-indistinguishable two-photon state directly from the downconversion process. This was achieved by using extended phase-matching (EPM) conditions in a 1-cm long PPKTP crystal. These conditions were better known as inverse group velocity matching (GVM) in ultrafast nonlinear optics. Application of EPM for a PPKTP crystal resulted in producing signal and idler with identical spectra for broadband degenerate type-II downconversion at $\sim1584$ nm. We were able to demonstrate the frequency-indistinguishable generation of the signal and idler photons by using a fiber-based Hong-Ou-Mandel (HOM) interferometer. Using both cw and broadband ($\sim6$ nm bandwidth) pulsed inputs, we observed a high visibility HOM interference (95% for cw pump, 85% for pulsed pump) without any spectral filtering. We further experimented with the PPKTP crystal to find the exact degenerate conditions for downconversion. This resulted in higher interference visibility (>90%) in HOM interference for various input pump bandwidths and without background subtraction.

The application of EPM conditions to PPKTP extends to classical nonlinear optics. For instance, an experimental setup similar to the one presented in this thesis can be used to implement spectral phase conjugation for dispersion compensation [180]. Also, a recent experimental demonstration that used the EPM conditions in PPKTP in the high gain regime realized cavity-free optical parametric oscillation [181, 182].

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The validation of frequency entanglement was first achieved by observation of the dissimilar single-photon and two-photon correlation times. However, another experimental method was needed to quantify the degree of entanglement. We were motivated to characterize the quantum state in the time domain, since the frequency domain techniques would be impaired by the low detection efficiencies and slow detector speed at infrared wavelengths. We proposed and implemented a time-resolved single-photon detection technique that used sum-frequency generation (SFG) at the single-photon level. A direct-detection scheme with any single-photon detector technology would not be able to resolve the temporal features of two-photon state. We achieved the required timing resolution capability by using an ultrafast pulse as the classical pump in a single-photon upconversion experiment. Therefore, timing resolution was not set by the speed of the single-photon detectors, but by the ultrafast pulse duration.

The noncollinear phase-matching geometry in a 1-mm PPMgSLT crystal was utilized to provide broad phase-matching bandwidth and to contain two independent upconverters for signal and idler in a single crystal. The upconversion setup was used synchronously with the downconversion experiment, eliminating the pump induced timing jitter. We characterized the upconversion system by classical SFG and then used it in conjunction with the downconversion source. Using the ultrafast pump as a sampling probe, we were able to resolve fine features in time-domain profile of single photons. Specifically, we observed the amplitude statistics of the single photons within their arrival window which was set by the timing uncertainty in the generation of the photon pairs. This technique also enabled us to assess the grating quality of the PPKTP crystal with high spatial-resolution. We note that the time-resolved upconversion technique can be used as a non-invasive diagnostic tool to evaluate the grating quality of quasi-phase matched nonlinear crystals that can accommodate a broadband pump spectrum.

The high timing-resolution capability of our single-photon upconversion method also permitted us to measure the joint temporal statistics of the two-photon state. This was accomplished by controlling the pump relative delays of both upconverters
independently and performing coincidence detection. For the first time, we measured
the joint temporal density (JTD) of a two-photon quantum state. This technique was
shown to be the Fourier-dual of the widely-used joint spectral density measurement.
With the JTD measurement, we observed strong time anti-correlation associated
with the coincident-frequency entangled two-photon state. The experimental char-
acterization of the two-photon joint temporal structure was in good agreement with
previous HOM interference and single-photon autocorrelation measurements. We also
compared the experimental data to a theoretical model developed for time-resolved
photodetection and observed remarkable agreement.

We showed that the time-resolved upconversion realizes a narrowband temporal
filtering with a potential to be nearly lossless. A higher pump power and better
mode-matching can realize this potential. Moreover, it is also possible to use a long
upconversion pump pulse to deterministically upconvert every downconverted photon
pair. This could be used to frequency-translate the infrared downconverted photon
pairs to visible wavelengths with high conversion efficiency. It would be of interest to
see whether the frequency translation can preserve the frequency-indistinguishability
of the signal and idler. To verify this, a HOM interferometer can be easily devised
for visible wavelengths.

Another important objective in our study of frequency-entangled two-photon
states was to control and monitor the degree of two-photon frequency entanglement.
For this purpose, we performed a series of time-resolved measurements where the
downconversion pump bandwidth was independently varied to change the joint spec-
tral (and temporal) amplitude. We quantified the continuous-variable entanglement
by calculating the entanglement entropy from the Schmidt decomposition for the
bi-partite state. The experimentally obtained JTD distributions were used to com-
pute the entanglement entropy which were shown to be in good agreement with the
theoretical predictions.

Through SPDC pump bandwidth variation and JTD measurements, we were able
to monitor and control the frequency-entanglement between signal and idler. This
enables us to use the two-photon frequency-entangled state in a diverse set of ap-
plications. For example, a highly frequency-entangled two-photon state can be used to test the fundamental limits on precision measurements whereas the unentangled form is suitable for the heralded generation of a pure single-photon quantum state for LOQC.

The JTD measurement scheme offers many practical advantages for QIP applications. We have emphasized that a more complete characterization of multi-photon frequency-entangled state can use the joint spectral and temporal density measurements together. This could be instrumental in studying the effects of dispersion and spectral phase of the two-photon state, where a joint spectral density measurement alone is not sufficient. For instance, both joint characterization techniques used in conjunction can verify the generation of transform-limited two-photon states. We believe that the JTD measurement can become a mainstream characterization tool for multi-photon entangled states in the near future.
Appendix A

Evaluation of the Bipartite Trace Operation

In Chapter 5, we derived the coincidence count rate expression through density operator formalism. In Eq. (5.22), we find a non-normally ordered trace operation for the bipartite state, which can be evaluated by arranging the field operators in normal order by using commutation relation \([\hat{a}_i(t), \hat{a}_j^\dagger(u)] = \delta_{ij}\delta(t - u)\).

We need to find:

\[
\text{Tr} \left[ \left( \hat{a}_S(t_S)\hat{a}_S^\dagger(x)\hat{a}_S(t_S')|0\rangle_S\langle 0| \right) \otimes \left( \hat{a}_I(t_I)\hat{a}_I^\dagger(y)\hat{a}_I(t_I')|0\rangle_I\langle 0| \right) \right] \\
= S \langle 0|\hat{a}_S(t_S)\hat{a}_S^\dagger(x)\hat{a}_S(t_S')|0\rangle_S \otimes I \langle 0|\hat{a}_I(t_I)\hat{a}_I^\dagger(y)\hat{a}_I(t_I')|0\rangle_I.
\]

(A.1)

We proceed with the evaluation of the signal term only, since the idler dependent term is similar. Using the commutation relation, we achieve the normal ordering in two steps:

\[
s \langle 0|\hat{a}_S(t_S)\hat{a}_S^\dagger(x)\hat{a}_S(t_S')|0\rangle_S \\
= s \langle 0| \left( \hat{a}_S^\dagger(x)\hat{a}_S(t_S) + \delta(x - t_S) \right) \left( \hat{a}_S^\dagger(t_S')\hat{a}_S(x) + \delta(x - t_S') \right) |0\rangle_S
\]
\[= S\langle 0|\hat{a}_S^\dagger(x)\hat{a}_S(t_S)\hat{a}_S^\dagger(t_{S'})\hat{a}_S(x) + \delta(x - t_{S'})\hat{a}_S^\dagger(x)\hat{a}_S(t_S) + \delta(x - t_S)\hat{a}_S^\dagger(t_{S'})\hat{a}_S(x) + \delta(x - t_S)\delta(x - t_{S'})|0\rangle_s\]
\[= S\langle 0|\left(\hat{a}_S^\dagger(x)\hat{a}_S^\dagger(t_{S'})\hat{a}_S(x) + \delta(t_s - t_{S'})\hat{a}_S^\dagger(x)\hat{a}_S(x) + \delta(x - t_S)\hat{a}_S^\dagger(t_{S'})\hat{a}_S(x) + \delta(x - t_S)\delta(x - t_{S'})\right)|0\rangle_s\]

(A.3)

All the normally-ordered terms in the above expression are automatically zero, because \(\hat{a}_i(\tau)|0\rangle = 0\) by definition. The only non-zero term comes from the delta functions, yielding \(\delta(x - t_S)\delta(x - t_{S'})\) since \(S\langle 0|0\rangle_s = 1\). The idler part can be evaluated in a similar way. Finally, the trace expression reads:

\[
\text{Tr}\left[\left(\hat{a}_S(t_S)\hat{a}_S^\dagger(x)\hat{a}_S(x)\hat{a}_S^\dagger(t_{S'})|0\rangle_s\langle ss|0\right) \otimes \left(\hat{a}_I(t_I)\hat{a}_I^\dagger(y)\hat{a}_I(y)\hat{a}_I^\dagger(t_{I'})|0\rangle_{sI}\langle I|0\rangle_I\right)\right] = \delta(x - t_S)\delta(x - t_S')\delta(y - t_I)\delta(y - t_{I'}). \tag{A.4}
\]
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