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Generation and Manipulation of Nonclassical Photon Sources in Nonlinear Processes

Zhi-Yuan Zhou and Bao-Sen Shi

Abstract

Nonclassical photon sources are key components in quantum information science and technology. Here, the basic principles and progresses for single photon generation and their further manipulation based on second- or third-order nonlinear processes in various degrees of freedom are briefly reviewed and discussed. Based on spontaneous parametric down-conversion and spontaneous four-wave mixing, various nonlinear materials such as quasi-phase-matching crystals, dispersion-shifted fibers, and silicon-on-insulator waveguides are used for single photon generation. The kinds of entanglement generated include polarization, time-energy, time-bin, and orbital angular momentum. The key ingredient for photon pair generation in nonlinear processes is described and discussed. Besides, we also introduce quantum frequency conversion for converting a single photon from one wavelength to another wavelength, while keeping its quantum properties unchanged. Finally, we give a comprehensive conclusion and discussion about future perspectives for single photon generation and manipulation in nonlinear processes. This chapter will provide an overview about the status, current challenge, and future perspectives about single photon generation and processing in nonlinear processes.

Keywords: photon pair, spontaneous parametric down-conversion, spontaneous four-wave mixing, polarization, time-bin, time-energy, orbital angular momentum, quantum entanglement, quantum frequency conversion, quasi-phase-matching, dispersion-shifted fiber, silicon-on-insulator waveguide

1. Introduction

Nonclassical photon sources are fundamental resources for researches in quantum information science and technology (QIST), which are widely used for applications like quantum communications, computations, sensing, and studying fundamental physics of quantum mechanics [1–3]. Therefore the ability to generate and manipulate single photon determines how far we can go in QIST. Generally, there are two distinct methods for generating single photons: one is based on excitation-reemission of photon in a semiconductor quantum dot [4], a single defect in NV center [5], or a single atom [6]; another convenient method is based on spontaneous emission based on a second- [7] or a third-order nonlinear process [8]. In this chapter, we will focus on single photon generation by using nonlinear processes. Usually, there are two nonlinear processes for generating nonclassical photon pairs:

(1) spontaneous parametric down-conversion (SPDC), which is a second-order nonlinear process; (2) spontaneous four-wave mixing (SFWM), which is a third-order nonlinear process. In both SPDC and SFWM, energy, linear momentum, and angular momentum conservations should be fulfilled. Due to these conservation laws and the technology of quantum interference used, two photons in each pair generated in SPDC and SFWM can be correlated in various degrees of freedoms, for example, polarization, energy-time, orbital angular momentum, position-linear momentum, angular momentum, and photon number and path [1]; we can utilize these freedoms in a specific application scenario in QIST.

In the subsequent section, we will first introduce the basic principle of SPDC and SFWM for generating photon pairs and then the various materials used for SPDC and SFWM. In the key part of this chapter, we will review the developments of various entangled photon pair sources and methods for charactering these sources. After that we will introduce a nonlinear method for transducing the wavelength of the photon from one to another while keeping its quantum properties unchanged, which is suitable for building up a quantum interface to connect different quantum systems. Finally, we will give a brief summary in which some future perspectives for nonclassical photon pair generation and potential applications are discussed.

2. Photon pair generation using SPDC or SFWM

SPDC is realized in a second-order nonlinear process (see **Figure 1** left image), in which a pump photon at higher frequency (ω_p) is split into two daughter photons at lower frequencies with certain probability in a nonlinear crystal; these two daughter photons are usually called signal (ω_s) and idler (ω_i) photons. The conservation laws of energy, linear momentum, and angular momentum require that the frequency, linear momentum (k), and angular momentum (l) of the pump, signal, and idler photon fulfill the following conditions: $\omega_p = \omega_s + \omega_i$; $k_p = k_s + k_i$; $l_p = l_s + l_i$. These conservation laws are responsible for the generation of various entangled sources.

In correspondence to SPDC, SFWM is a third-order nonlinear process; a big difference is that there are two pump beams in SFWM (see **Figure 1** right image), in comparison to SPDC in which only one pump beam is used. The conservation laws in SFWM require that the corresponding parameters of the pump, signal, and idler photons have the following relationships: $\omega p_1 + \omega p_2 = \omega_s + \omega_i$; $k p_1 + k p_2 = k_s + k_i$; $l p_1 + l p_2 = l_s + l_i$.

For quantum optical description of SPDC and SFWM, the Hamiltonian of the two processes can be expressed as [9]:

$$\hat{H} = \hbar\xi(\hat{a}_s^\dagger \hat{a}_i^\dagger + H.C.) \quad (1)$$

where ξ depends on the pump intensity, the nonlinear coefficient of the crystal, crystal length, and focusing parameters. Therefore the photon states generated in SPDC and SFWM can be expressed in Fock state basis as [9]:

$$|\Phi\rangle = \text{Exp}\left[-\frac{i\hat{H}t}{\hbar}\right]|0, 0\rangle = |0, 0\rangle + \kappa|1, 1\rangle + \kappa^2/2|2, 2\rangle + \dots \quad (2)$$

It can be seen from Eq. (2) that we obtain a vacuum state with a high probability if the pump is weak. The second term is the photon pair state we need, and the other terms are multiphoton states which should be avoided. It is clear from Eq. (2) that the pump beam should be at a moderate intensity level in order to eliminate the effects of higher photon number states. The photon pair generated in SPDC and SFWM is of probability and is undetermined, which is a disadvantage for photon

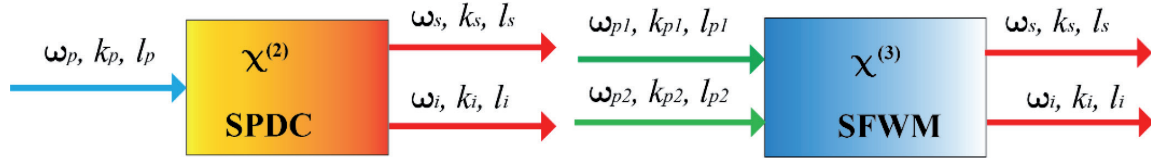


Figure 1. A simple diagram for SPDC and SFWM. The conservation of energy, linear momentum, and angular momentum holds in both nonlinear processes.

sources generated from nonlinear processes. For this reason, one needs to choose a proper pump intensity level in order to balance between different experimental parameters.

All materials have third-order nonlinearity, but only those materials that are central asymmetric have second-order nonlinearity. The commonly used materials for SPDC can be divided into two kinds according to different phase matching: one is birefringent angle phase-matching materials, such as LBO, BBO, KTP, and LN [10]; another is quasi-phase-matching (QPM) crystals such as PPKTP and PPLN [11]. QPM crystals have the advantages of high generation rate and narrow bandwidth, which are frequently used in photon pair generation in modern quantum optics experiments [12–19]. For SFWM, the commonly used materials are hot or cold atomic ensembles [20–24] and guided-wave materials such as dispersion-shifted fibers (DSF) [25–29], photonic crystal fibers (PCF), [30–32], and silicon-on-insulator (SOI) waveguide [33–41]. To look for new kinds of nonlinear materials for generating high-quality photon sources is still a very hot topic in QIST.

3. Various kinds of photon sources generated in SPDC and SFWM

Because of the conservation of energy, linear momentum, and angular momentum in SPDC and SFWM, various kinds of nonclassical sources can be generated; in this section we will review the recent development and key points in generating various kinds of nonclassical photon sources.

3.1 Polarization-entangled photon source

A polarization-entangled photon source (PEPS) is one of the most important entangled photon sources that have been studied for decades of years. In the literatures, people generate PEPS using different materials with different experimental configurations [12–19, 42, 43]. For SPDC, in the early times, PEPSs are created using birefringence phase-matching (BPM) crystals, for example, a type-II phase-matched BBO crystal is used to create a PEPS in the first practical and effective experiment, in which orthogonal polarized photons are emitted at the intersection cones [42]. Later on, a beam-like design is used for high-brightness photon pair generation, which is widely used in multiphoton quantum experiments [44]. The significant progress in nonlinear crystal fabrication makes a QPM crystal a better choice for researchers in many nonlinear optics applications [11]. The most important merit of using QPM crystals in generation photon pairs is its high spectral brightness in contrast to BPM crystals, due to its large effective nonlinear coefficient and long allowable interaction length.

Recently, to generate PEPS by placing a QPM crystal inside a Sagnac interferometer configuration has been demonstrated to be superior to other configurations (see **Figure 2**). The basic idea for a Sagnac loop-based PEPS is as the following: a pump beam is split into two beams by a double polarized beam splitter (DPBS) and

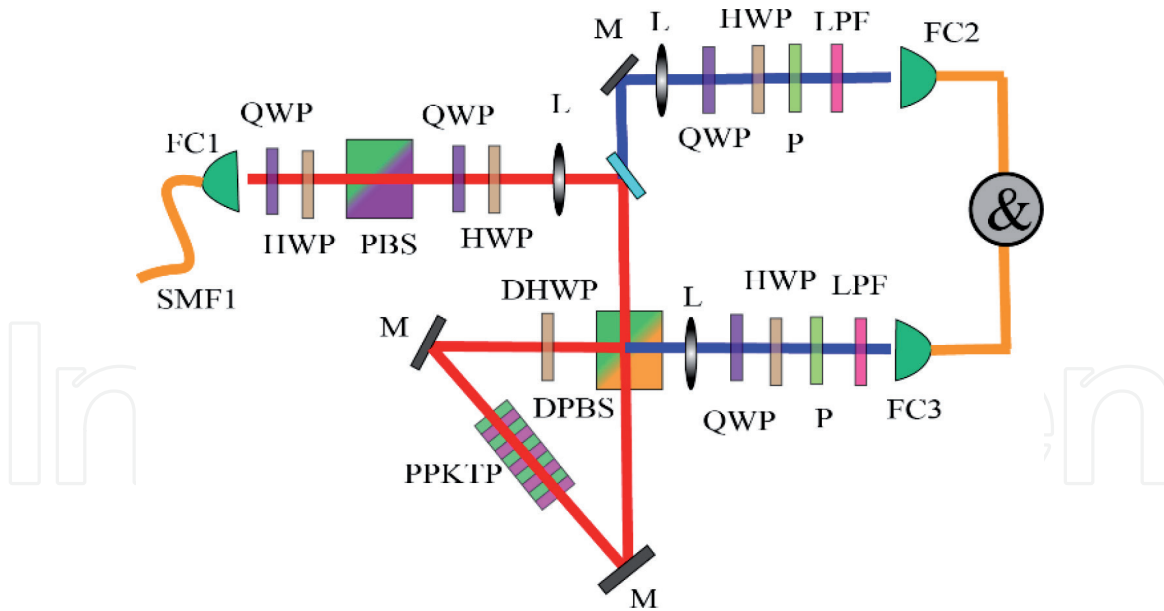


Figure 2. The experimental setup for a typical polarization-entangled source based on Sagnac interferometer (figure cited from [13]).

counter-propagates in the Sagnac loop; each beam generates a pair of photon with orthogonal polarization, in one circulation direction; the photon pair is rotated by a double half wave plate; then two pairs of photons are recombined in the DPBS; and a PEPS with a form of $|\Phi\rangle = 1/\sqrt{2}(|HV\rangle + e^{i\theta}|VH\rangle)$ is generated; the relative phase θ can be tuned by a pair of wave plates placed in the input port of the Sagnac loop. The merits to use the Sagnac interferometer configuration are its compactness, high stability, and high brightness. The original idea of Sagnac loop-based PEPS is from [43] where a BPM crystal is used, and then this idea is generalized to a QPM crystal by Kim in 2006 [45] for a CW pumped photon source. After that, a pulsed PEPS at 780 nm based on this configuration was developed by Kuzucu and Wong in 2008 [46]. In the early experiments, the wavelengths of the photons generated are in visible range; therefore these photons are not suitable for long-distance quantum communications in fiber. Only recently, telecom band PEPS is developed [13, 16]. A pulsed PEPS at 1584 nm based on a type-II PPKTP was demonstrated by Jin et al. in 2014 [16], and Li et al. reported a tunable CW PEPS in 2015 [13]. Now, PEPS based on QPM crystals in a Sagnac configuration has become a basic tool for many experiments [47–49].

In SFWM, PEPS is generated using an atomic ensemble with different configurations. The PEPS generated with the atomic ensemble has narrow bandwidth; the wavelength is fixed to specific atomic transition lines [50, 51]. Many works report PEPS generation based on guided-wave materials such as DSF [8, 25, 27, 28], PCF [30], and SOI waveguide [37, 52], the advantages of using guided-wave materials are free of free-space coupling, low loss, low cost, and easy to integrate. The guided-wave platform is very promising in large-scale applications which require hundreds of optical components. It is also convenient for building up a compact, versatile photonic source platform for various kinds of applications in QIST.

3.2 Time-energy and time-bin-entangled photon source

Because of conservation of energy in nonlinear processes, the two photons in each pair generated are correlated in frequency and are also generated simultaneously. Although the uncertainty in time and frequency domain for individual

particle should meet the requirement of uncertainty principle, the sum of the frequency of signal and idler multiplies the difference between arrive times of the two photon should have a very small value, and violates an inequality for two photons existed classical correlations [53]. A two-photon Franson-type interference is used to characterize the correlations between the two photons; the phases between the two unbalanced Michelson interferometers (UMI) are correlated [54, 55]. To generate a time-energy entangled photon pair, a laser with long coherent time is needed (see **Figure 3(a)**); the time difference between two paths in UMI should be much larger than the coherence time of the single photon but much shorter than the coherent time of the pump laser [53]. A similar kind of entangled photon source is a time-bin entangled photon source [56], in which a pulse pump is split into two pulses in an UMI, and then these two pulses have a certain probability to generate a pair of photon separately; the photon pairs generated by these two pulses are indistinguishable after passing through two UMIs (the time difference of the UMI in measurement part is the same as the UMI in pump part, see **Figure 3(b)**). The quantum states for a time-energy or a time-bin entangled photon source can be expressed as $|\Phi\rangle = 1/\sqrt{2}(|SS\rangle + e^{i\theta}|LL\rangle)$, where S and L represent the short and long arm of the UMIs, respectively. A time-bin entangled photon pair is robust for long-distance transmission, which is widely used in demonstrating various quantum communication protocols [57]. A time-energy entangled photon source has been realized in various material systems such as atomic vapor [21], nonlinear crystals [56, 57], and guided-wave platform [26, 28, 34, 35, 39–41]. The differences between various materials are the photon emission bandwidth and spectral ranges. Furthermore, researchers have realized three photon genuine time-energy entangled photon sources, and their nonclassical correlations are verified [58].

3.3 Orbital angular momentum entangled photon source

Another important degree of freedom of photon is orbital angular momentum (OAM), which has been widely investigated since 1992 [59]. OAM has unbounded dimensions, which is very promising for high-capacity communication task in both classical and quantum optical communications [60–62]. OAM entangled photon pairs can be generated in SPDC and SFWM based on crystals [48, 63–69] and atomic vapors [70, 71]. The quantum state for an OAM entangled photon pair generated directly by pumping a nonlinear crystal (**Figure 4**, left image) can be

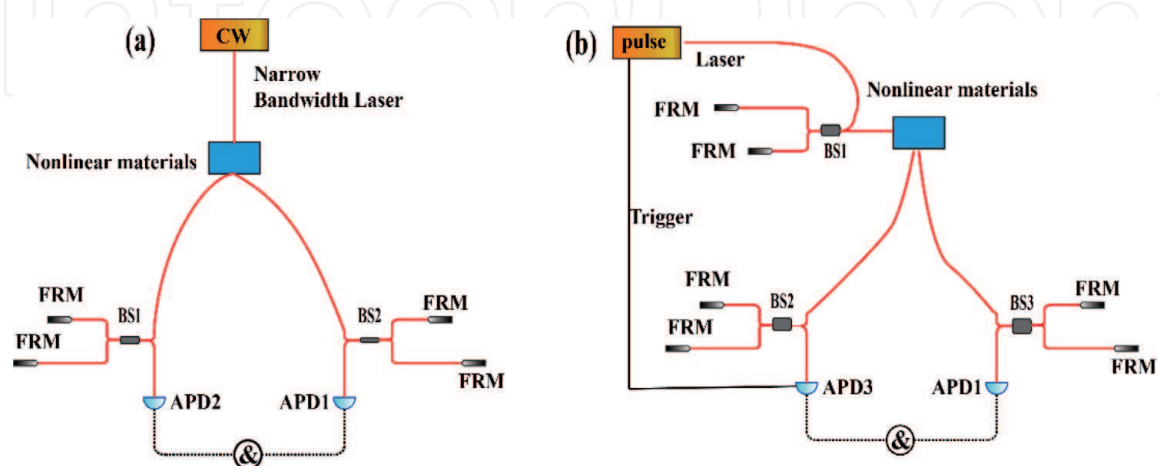


Figure 3. Simplified diagrams for (a) time-energy; (b) time-bin entangled photon generation. A narrow bandwidth CW laser is used for generating of time-energy entangled photon pair, while a pulse laser is used for generating time-bin entangled photon pair.

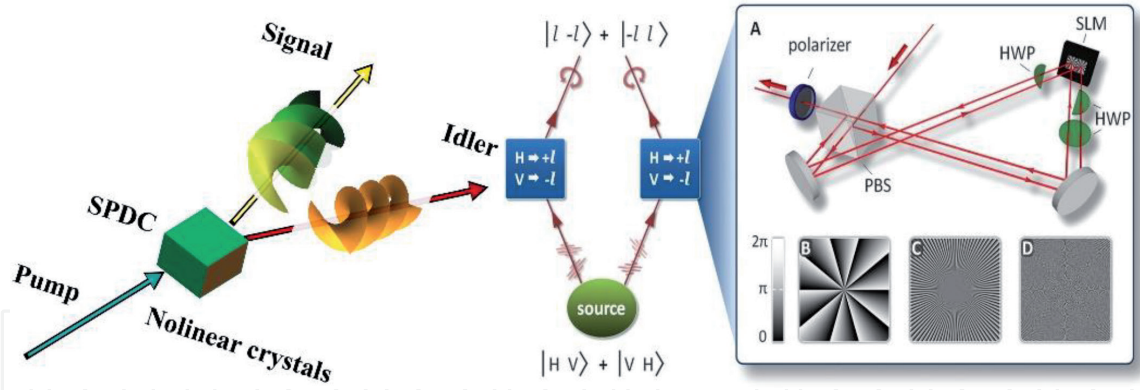


Figure 4.

Generate HD OAM entangled state directly from SPDC (left image); 2D OAM entangled state generation by converting a polarized entangled state into 2D subspace of OAM entangled state (right image, cited from [48]).

expressed as $|\Phi\rangle = \sum C_l |l, -l\rangle$, where C_l is the weight for different OAM modes. One can investigate OAM entanglement in a two-dimensional (2D) subspace [48, 63, 67–70] or in high-dimensional (HD) space [65, 66, 71]. The properties and the methods of characterizing a 2D entangled source in different degrees of freedom are similar and can be converted from one kind to another [48, 68] (please see **Figure 4** (right images)). The post-selected OAM entangled states in a 2D subspace can be expressed as $|\Phi\rangle = 1/\sqrt{2}(|l, -l\rangle + |-l, l\rangle)$. While for a HD entangled source, the properties and the methods of characterization are rather different. [65] reported on the realization of a 11D entangled source, demonstrating the violation of the Bell inequality. Zeilinger's group has demonstrated a 100×100 HD entanglement by measuring the entanglement witness of the generated state [66]. For a 2D OAM entangled photon source, Zeilinger's group converted a polarized entangled photon source into an OAM entangled source with OAM momenta of 300 h in 2D subspace via a spatial light modulator [48]. Later on, a higher OAM momentum of about 10,000 h for a 2D OAM entangled source is realized by using a vortex reflection mirror [68]. A HD OAM entangled photon source is preferred for studying the basic principle of quantum mechanics and for HD quantum communication applications.

4. Methods for characterizing the properties of a nonclassical photon source

Nonclassical photon sources can be characterized from different aspects. For characterizing the properties of a heralded single photon, the heralded efficiency [72], the coincidence to accidental coincidence ratio (CAR) [73], and the single photon Glauber function [74, 75] are important parameters. The heralded efficiency is the probability of detecting the second photon when the first photon is detected. It is a measurement of the photon collection efficiency, filter and transmission losses, and the single photon detector efficiency. The heralded efficiency is the ratio of the coincidence count to the single count rate of the first detected photon. CAR is a measurement of the signal to background noise ratio for a two-photon experiment; high CAR can ensure the quantum nature existed between the two photons. CAR depends on pump power and detector performance. Usually, CAR will increase when the pump power is increased in the low pump power regime. After reaching the maximum value, CAR will decrease with the increase of the pump power [76]. The single photon Glauber function can be measured as shown in [74]. The measured photon is firstly split by a beam splitter, and then by measuring the three party coincidence, single count and two-photon coincidence, we can

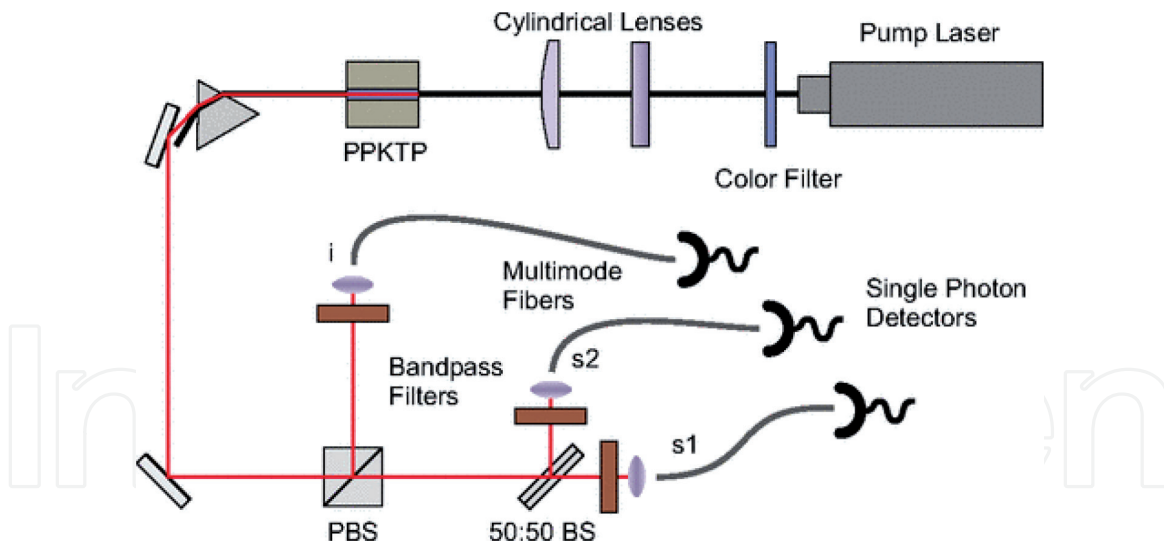


Figure 5.
 Experimental setup for measuring heralded single photon autocorrelation function for single photon generated from SPDC (image cited from [74]).

calculate the single photon $g^{(2)}$ function (see **Figure 5**). A near zero $g^{(2)}(0)$ indicates the high quality of single photon nature. For a pulse pumped photon source, the single photon purity is also an important parameter [17, 77]. The purity of photon is a measurement of spectral correlations between two photons; the purity is determined by the Schmitt number in the Schmitt decomposition. The unity single photon purity indicates that the two-photon spectral can be factorized into product of two separate functions of the signal and idler photons. The high single photon purity is very important for realizing high visibility HOM interference between two independent single photon sources, which is the key technique for realizing high photon number entangled states.

There are various available and faithful methods to characterize the quality of entanglement of an entangled two-photon source, including two-photon interference fringes [65, 78], Bell CHSH inequality [79, 80], and quantum state tomography (QST) [81]. Two-photon interference fringe is much easier to measure; through calculating the interference visibility from the measured data, we can evaluate the quality of an entangled source. A high visibility indicates a high quality of the generated state by comparing the ideal maximum Bell states. When the visibility is greater than a threshold value, the two photons have Bell nonlocality; the threshold value is different for two-photon states in different dimension. For two photons existing in classical correlation, the Bell CHSH parameter S is not greater than a certain value. The violation of this value indicates a nonclassical correlation between the two-photon states. Bell CHSH parameter S is an indicator of whether the two-photon state has Bell nonlocality and how strong this kind of nonlocal correlation is. The violation of Bell inequality has been widely studied in literatures for a 2D and a HD entangled state. To fully know the content of a generated quantum state, QST can be used to reconstruct the density matrix of a certain quantum state. By the density matrix of a quantum state, all the properties of the quantum state can be predicted.

5. Quantum frequency conversion for nonclassical quantum state

There are many quantum systems for QIST based on different materials, including atomic ensembles, trapped ions, solid-state materials, and fibers for transmission [82–86]. Each quantum system has some advantages in QST, and these

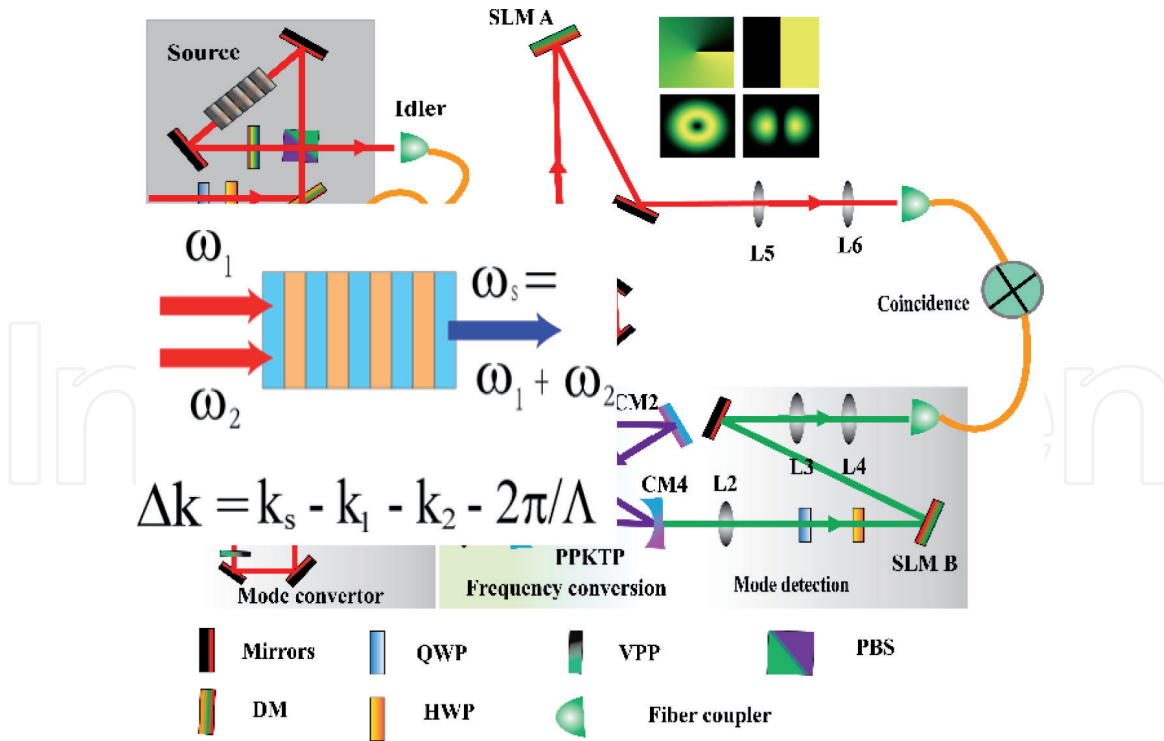


Figure 6. Left image: simple diagram for sum frequency generation in QPM crystal. Right image: experimental setup of QFC for an OAM qubit, an OAM-polarization hybrid entangled state, and an OAM-OAM entangled state (cited from [99]).

systems usually work at different frequencies, which may have a big frequency mismatching. To build up a quantum network consisting of various quantum systems for information encoding, storage, transmission, and processing, a quantum frequency converter (QFC) to link different quantum systems is indispensable. Such a frequency transducer can be realized by utilizing nonlinear processes such as sum frequency generation (SFG) and Bragg reflection in four-wave mixing. The theory of quantum frequency conversion for SFG was first proposed by P. Kumar in 1990 [87]. In SFG, a strong pump laser can convert a weak signal beam with high quantum efficiency; the unity quantum efficiency can be reached when the pump beam is strong enough (see **Figure 6** left image), and the quantum correlations are unchanged after frequency conversion. Since it has been proposed, some significant progresses have been made in this field; researchers have realized that frequency up-conversion and down-conversion for a single photon generated from quantum dot, and various qubit states or entangled states such as polarization, time-energy, and OAM entangled state [39, 88–101] have been up- or down-converted. A typical setup for QFC of an OAM qubit, an OAM-polarization hybrid entangled state, and an OAM-OAM entangled state is shown in **Figure 6** (right image). For frequency down-conversion, the visible single photons generated from atomic ensemble, trapped ions, or NV centers have been converted to telecom band successfully [102–105]. In all these demonstrations, the single photon properties and entanglement are preserved in the conversion processes, which ensures that quantum information can be coupled to different quantum systems by using a quantum frequency interface.

In QFC, there are four parameters to evaluate the quality of a converter: quantum conversion efficiency, noise level, spectral bandwidth, and spatial bandwidth. These parameters are not independent; therefore in practical applications, one needs to balance between different parameters [98]. A longer nonlinear crystal is preferred to reach maximum conversion efficiency when the pump power is

limited, but a longer nonlinear crystal would lead to a smaller spectral bandwidth and spatial bandwidth; a proper crystal length should be chosen to balance conversion efficiency and spectral bandwidth. The noise in QFC comes from SPDC and SRS of the intense pump beam; therefore a longer pump wavelength is preferred to reduce the noise photon in QFC [106]. The noise photon can also be dramatically reduced by using a narrow bandwidth filters to filter out the converted photon.

6. Discussions and conclusion

Nonclassical photon sources are used in almost all fields of QIST; the ability to generate and control its properties is at the heart for applications in QIST. Though many progresses have been made in single photon generation and manipulation in nonlinear processes, lots of further techniques should be developed to harness the quality of single photon generated in SPDC or SFWM. The detailed techniques for optimizing the parameter of the photon source depend on the specific applications. For a pulsed heralded single photon source, the heralded efficiency and total photon count are important parameters, but the probability of single photon generation per pulse is very low, which limits the flux of photon pair generation. These defects can be overcome by using time, frequency, and OAM multiplexing to enhance the photon generation probability per pulse and total count rate [41, 107–112]. When the optical elements for multiplexing have low losses, the heralded efficiency and rate can be increased substantially [112]. For applications taking advantages of the sharp time correlations in SPDC, a broadband spectrum of the photon pair is needed. Such a broadband photon pair can be realized with an ultrathin nonlinear crystal or using a chirp quasi-phase-matching crystal; the bandwidth of the photon pair generated can be greater than 100 nm, which has a time correlation of sub-femtosecond [113]. For quantum information applications, a multiplexed time-energy and polarized entangled photon pair is preferred for high-capacity quantum communication by using dense-wave division multiplexing technique. The multiplexed entangled sources are easier to be realized using a waveguide platform such as a PPLN waveguide, a DSF, or SOI waveguide. A SOI ring cavity is also preferred in generating frequency comb entangled states [114]. For generating HD entangled states, by shaping the profile of a pump beam, a much greater Hilbert space can be reached [115, 116]. For QFC of OAM entangled states, the mode-dependent conversion efficiency has not been solved yet. We recently proposed and demonstrated that if we use a flat-top beam to pump the SFG crystal, then we can solve the problem of mode-dependent conversion efficiency by using a Gaussian pump beam [117].

For a compact application, integrated optics will offer a great advantage over free-space implementation; the trends of modern optics are to convert a free-space module to an equivalent integrated optical device, which will be of high compact, robust to environment fluctuations and much easier for larger amounts of fabrication [118, 119].

In conclusion, most of the advances and progresses for generation and manipulation of single photon sources in nonlinear processes are briefly reviewed in this chapter; this review will provide a glance at the current status, and challenges remain to be solved in this field. The general properties for single photon generation in nonlinear processes are introduced firstly; then we introduce the development of various entangled states and the methods to characterize nonclassical photonic states. Next, we review the progresses for frequency conversion of a photonic state in nonlinear processes. Finally, we give comprehensive discussions about

the remaining challenges in generating high-quality and HD entangled states, the unsolved problems for QFC of HD OAM photonic states, and the development of integrated optics for small footprint optical devices and large-scale quantum information processing on chip. This book chapter should be helpful for new researchers working in this field.

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