Minimizing the frequency correlation of photon pairs in photonic crystal fibers

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Abstract. Based on the dispersion property of a given photonic crystal fiber (PCF), we study how to directly generate photon pairs with minimized frequency correlation via pulse-pumped spontaneous four-wave mixing. After illustrating why the intensity correlation function \( g^{(2)} \) of individual signal (idler) photons can be used to reliably characterize the frequency correlation of photon pairs, we numerically investigate the dependence of \( g^{(2)} \) under various kinds of experimental conditions. The results show that to minimize the frequency correlation, the experimental parameters should be properly optimized by balancing the influences of the high-order dispersion and the intrinsic sinc oscillation of phase matching function, apart from the satisfaction of the specified phase matching condition and the usage of transform-limited pump pulses. To verify the calculated results, we conduct two series of experiments by regulating the pump to respectively satisfy the asymmetric and symmetric group velocity matching conditions in our 0.6 m-long PCF. In both cases, the measured values of \( g^{(2)} \) are less than the calculated results due to the inhomogeneity of the PCF; however, the experimental results qualitatively agree with the numerical simulations. Our investigation is very useful in fiber-based quantum state engineering.

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1. Introduction

For quantum information processing and quantum state characterization involving quantum interference among multiple sources, such as linear optical quantum computing and Fock state tomography, photon pairs in a spectral factorable state are highly desirable [1, 2]. Spontaneous parametric emission (SPE) processes realized by pumping the nonlinear medium with ultra-short pulses, including spontaneous parametric down-conversion (SPDC) in $\chi^{(2)}$-based nonlinear crystals and spontaneous four-wave mixing (SFWM) in $\chi^{(3)}$-based optical fibers, are efficient methods for generating photon pairs. However, the photon pairs usually have a strong spectral correlation, resulting in a timing uncertainty within the pulse duration of the pump. With the usage of commercially available detectors, whose timing resolution is of the order of hundreds of picoseconds and is generally unable to fulfill the single-mode detection [3, 4], the problem is solved either by using narrow band optical filters to eliminate the frequency correlation or by directly generating the photon pairs in a factorable state [5–7]. The former method will result in a huge reduction of the photon counting rate; but the latter will not.

To directly generate the spectral factorable photon pairs, which is in principle filter-free, one should properly control energy and momentum conservation by regulating the pump pulse and by tailoring the dispersion of the nonlinear medium so that the joint spectral amplitude (JSA) of photon pairs can be factorized into $f(\omega_s, \omega_i) = h(\omega_s) \times k(\omega_i)$, where $h(\omega_s)$ ($k(\omega_i)$) is the probability amplitude of the signal (idler) photons having the frequency $\omega_s$($\omega_i$). Recently, the approximately factorable photon pairs generated from the SPDC in $\chi^{(2)}$ crystal and from the SFWM in optical fibers were experimentally realized [8–12]. Among the various kinds of nonlinear media of the pulse-pumped SPEs, the photonic crystal fiber (PCF) is regarded as an ideal one [7]. In addition to its controllable dispersion and its advantage of excellent spatial...
mode purity, PCF-based sources of photon pairs have the ability to realize SFWM with large frequency detuning between signal and idler photons, which not only helps to get rid of the contamination of Raman scattering, but also bridges different optical bands [12, 13].

So far, the influence of the first-order dispersion coefficient of the nonlinear media on factorability has been extensively studied [7, 14], but the impact of other factors still awaits further investigation. In this paper, based on SFWM in a given PCF, we demonstrate that besides the intrinsic sinc oscillation of the phase matching function [10], the frequency correlation of photon pairs is also influenced by the high-order dispersion of PCF and the chirp of the pulsed pump. Our investigation is very useful in generating photon pairs with minimized frequency correlation.

The rest of the paper is organized as follows. In section 2, we briefly introduce the principle of characterizing the factorability of photon pairs, showing that the mutual spectral correlation of photon pairs can be characterized by using the intensity correlation function \( g^{(2)} \) of individual signal (idler) photons. In section 3, we derive the analytical expression of \( g^{(2)} \) when the JSA is simplified by using the linear approximation of dispersion and the Gaussian approximation of sinc function. In section 4, based on the dispersion property of a given PCF, we numerically calculate \( g^{(2)} \) of individual signal photons in different experimental conditions by taking into account the high-order terms of dispersion and the oscillation of sinc function; this illustrates that it is necessary to perform a numerical calculation to understand the detailed dependence of factorability. Section 5 is devoted to the experimental verification, which is performed by regulating the pump to satisfy the asymmetric group velocity matching (AGVM) and symmetric group velocity matching (SGVM) conditions, respectively. The experimental results qualitatively agree with the numerical simulations. Finally, we give a brief conclusion.

2. Theoretical analysis

For the SFWM process in PCF, two pump photons at frequency \( \omega_p \) scatter through the \( \chi^{(3)} \) nonlinearity of PCF to create energy–time-entangled signal and idler photons at frequencies \( \omega_s \) and \( \omega_i \), respectively, such that \( 2\omega_p = \omega_s + \omega_i \). In the theoretical model, the detuning between the signal (idler) and pump photons is much greater than the gain bandwidth of Raman scattering; the Raman effect is therefore negligible. Moreover, the bandwidth of individual signal (idler) photons is naturally narrow band, and the generation of photon pairs is filter-free.

The strong pump pulses propagating along the single-mode PCF (denoted as the \( z \)-direction) remain classical, and can be expressed as

\[
E_p^{(+)} = E_{p0} e^{-i\gamma P_p z} \int d\omega_p \exp \left[ -\frac{(\omega_p - \omega_{pc})^2}{2\sigma_p^2} - i\varphi(\omega_p) \right] e^{i(k_{pz} - \omega_p t')},
\]

where \( \gamma \) is the nonlinear coefficient of PCF, \( P_p \) is the peak power of the pump, \( \varphi(\omega_p) \) is the frequency-dependent spectral phase, \( \omega_{pc} \) and \( \sigma_p \) are the central frequency and bandwidth of the pump, respectively. After omitting the high-order terms of the Taylor expansion \( \varphi(\omega_p) = \varphi_0 + \varphi_1(\omega_p - \omega_{pc}) + \varphi_2(\omega_p - \omega_{pc})^2/2 + \cdots \), with \( \varphi_0, \varphi_1 \) and \( \varphi_2 \) respectively denoting the absolute phase, delay and quadratic phase, the expression of the Gaussian shaped pump is simplified as [15]

\[
E_p^{(+)} = E_{p0} e^{-i\gamma P_p z} \int d\omega_p \exp \left[ -\frac{(\omega_p - \omega_{pc})^2}{2\sigma_p^2} (1 + iC_p) \right] e^{i(k_{pz} - \omega_p t')},
\]

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where \( t = \varphi_1 + t' \) shows that the propagation distance is equivalent to the time delay, the parameter \( C_p = \sigma_p^2 \varphi_2 \) is the linear chirp of the pump and \( E_{p0} \) is related to the peak power of the pump through \( P_p \propto E_{p0}^2 \sigma_p^2 / \sqrt{1 + C_p^2} \). The pulse duration of the pump \( \Delta T_p \) is then expressed as \( \Delta T_p = 2\sqrt{\ln 2} / \sqrt{1 + C_p^2 / \sigma_p} \). For the case of \( C_p = 0 \), the pulsed pump is transform limited. We note that since the pump with a short pulse duration will inevitably acquire a certain amount of chirp during propagation in a transparent medium due to the effect of chromatic dispersion and Kerr nonlinearity, it is necessary to take the chirp of the pump into account.

Assuming the signal and idler beams are linearly co-polarized with the pump; then in the low-gain regime, the annihilation operator of the signal (idler) photons at the output port of PCF is given by [16, 17]

\[
\alpha(\omega_s, \omega_i) = a_0(\omega_s, \omega_i) + \frac{G}{\sigma_p} \int d\omega_0 f(\omega_s, \omega_i) a_0^\dagger(\omega_0) + o(G),
\]

where \( a_0(\omega_s, \omega_i) \) and \( a_0^\dagger(\omega_s, \omega_i) \) are the annihilation and creation operators of the vacuum fields at \( \omega_s, \omega_i \), respectively, and the coefficient \( G \propto \gamma P_p L \sqrt{1 - iC_p} \) with \( L \) denoting the length of PCF is proportional to the gain of SFWM. The so-called JSA function \( f(\omega_s, \omega_i) \) in equation (3) is expressed as [7]

\[
f(\omega_s, \omega_i) = \alpha(\omega_s, \omega_i) \times \phi(\omega_s, \omega_i),
\]

where

\[
\alpha(\omega_s, \omega_i) = \exp \left[ -\frac{(\omega_s + \omega_i - 2\omega_p)^2}{4\sigma_p^2} (1 + iC_p) \right]
\]

describes the pump envelope, and

\[
\phi(\omega_s, \omega_i) = \text{sinc} \left( \frac{\Delta k L}{2} \right)
\]

is the phase matching function, with \( \Delta k = 2k(\omega_p) - k(\omega_s) - k(\omega_i) - 2\gamma P_p \) denoting the wave vector mismatch, in which \( k(\omega_l) \) \( (l = p, s, i) \) is the wave vector at \( \omega_l \) and the term \( 2\gamma P_p \) can be neglected due to its smallness. Using the Taylor expansion of \( k(\omega_p), k(\omega_s) \) and \( k(\omega_i) \) at the perfect phase matching frequencies, \( \omega_{pc}, \omega_{s0} \) and \( \omega_{i0} \), respectively, we obtain

\[
\Delta k = \tau_s \Omega_s + \tau_i \Omega_i + \xi_s \Omega_s^2 + \xi_i \Omega_i^2 + \frac{k^{(2)}_{pc}}{2} \Omega_s \Omega_i + O(\Omega^3),
\]

where \( \tau_{(i)} = k_{(i)}^{(1)} - k_{(i0)}^{(1)} \) with \( k_{(j)}^{(1)} = \frac{dk(\omega)}{d\omega}_{|_{\omega_j}} \) \( (j = pc, s0, i0) \) denoting the first-order dispersion coefficient of PCF is the so-called group velocity mismatch between the pump and signal (idler) fields, \( \xi_{(i)} = \frac{k_{(i)}^{(2)}}{k_{(0)}^{(2)}} - \frac{k_{(i0)}^{(2)}}{k_{(00)}^{(2)}} \) is determined by the second-order dispersion coefficient \( k_{(j)}^{(2)} = \frac{d^2k(\omega)}{d\omega^2}_{|_{\omega_j}} \) \( (j = pc, s0, i0) \), \( \Omega_s \) and \( \Omega_i \) are related to \( \omega_s \) and \( \omega_i \) by \( \Omega_s = \omega_s - \omega_{s0} \) and \( \Omega_i = \omega_i - \omega_{i0} \), respectively, and \( O(\Omega^3) \) indicates the dispersion terms of third and higher orders.

It has been proved that the experimentally measurable quantity used to characterize the factorability of photon pairs is the intensity correlation function \( g^{(2)} \) of individual signal or idler.
fields [17, 18]. The value of $g^{(2)}$, measured by utilizing the Hanbury Brown–Twiss setup [19] and single-photon detectors (SPDs), can be calculated and expressed as [17]

$$
| {\mathcal D}^{(2)} | = \frac{\int d\Omega_1 d\Omega_2 |A^{(+)}_{\Omega_1}(t_1)|^2 |A^{(-)}_{\Omega_2}(t_2)|^2 \int d\Omega_3 d\Omega_4 |A^{(-)}_{\Omega_3}(t_1)|^2 |A^{(+)}_{\Omega_4}(t_2)|^2}{\int d\Omega_1 d\Omega_2 |A^{(+)}_{\Omega_1}(t_1)|^2 |A^{(-)}_{\Omega_2}(t_2)|^2 \int d\Omega_3 d\Omega_4 |A^{(-)}_{\Omega_3}(t_1)|^2 |A^{(+)}_{\Omega_4}(t_2)|^2}.
$$

where $A^{(+)}_{\Omega_1}(t) = \frac{1}{\sqrt{2\pi}} \int d\omega_1 a(\omega_1) e^{-i\omega_1 t} \text{ and } A^{(-)}_{\Omega_2}(t) = [A^{(+)}_{\Omega_2}(t)]^*$ are the positive and negative field operators of signal (idler) photons, respectively. Since the response time of each SPD is much longer than the creation time period of signal and idler photon pairs confined within the pump pulse duration, the time integral can be treated as an integral from $-\infty$ to $+\infty$. Note that $g^{(2)} \leq 2$ because of the Schwarz inequality; the equality holds if and only if $f(\omega_1, \omega_2)$ is factorable.

The physical meaning of equation (8) can be understood by thinking of the thermal nature of the individual signal (idler) field [20]. Within the response time of SPDs, $g^{(2)}$ increases with a decrease of the mode number contained in the field. For the photon pairs with a strong spectral correlation, the individual signal (idler) field is in the multi-mode thermal state, and the measured photon statistics is approximately described by the Poisson distribution with $g^{(2)} \rightarrow 1$; while for photon pairs in a spectral factorable state, the individual signal (idler) field with minimized time uncertainty is in the single-mode thermal state, and the measured photon statistics is described by the Bose–Einstein distribution with $g^{(2)} = 2$.

It is worth noting that the visibility of two-photon Hong–Ou–Mandel (HOM) interference of the individual signal (idler) photons between independent sources is closely related to $g^{(2)}$ [8, 17, 21]. However, the visibility of HOM interference not only depends on $g^{(2)}$, but also depends on other factors, such as the reflectivity/transmissivity of the beam splitter and the mode matching [17]. Therefore, $g^{(2)}$ is more suitable for characterizing the factorability of JSA.

In principle, the factorability of photon pairs can be characterized by applying the Schmidt decomposition to the corresponding JSA as well [22, 23]. Indeed, there is a simple relation between $g^{(2)}$ and the Schmidt number $K$: $g^{(2)} = 1 + 1/K$ [24]. However, for the determination of $K$ we need to know detailed information about the JSA function, whose precise description is difficult to obtain because of various kinds of imperfections in realistic experiments, such as the quantity of pump chirp and the unexpected variation of dispersion along the fiber [25]. Therefore, measuring $g^{(2)}$ seems to be more practical for reliably testing the mutual spectral correlation of photon pairs.

3. Analytical expression of the intensity correlation function $g^{(2)}$

To obtain instructive information for generating the factorable photon pairs, we simplify the JSA by applying the Gaussian approximation [6]

$$
sinc \left( \frac{\Delta k L}{2} \right) \approx \exp \left[ - \left( \frac{r \Delta k L}{2} \right)^2 \right] \quad (r = 0.439),
$$

and by using the linear approximation of the wave vector mismatch

$$
\Delta k \approx r_\Omega + r_{\Omega_i}.
$$

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In this situation, the simplified JSA is written as

\[
f(\Omega_s, \Omega_i) = \exp \left[ -\left( \frac{1 + i C_p}{4\sigma_p^2} + \frac{r^2 L^2}{4} \right) \Omega_s^2 \right] \exp \left[ -\left( \frac{1 + i C_p}{4\sigma_p^2} + \frac{r^2 L^2}{4} \right) \Omega_i^2 \right] 
\times \exp \left[ -\left( \frac{1 + i C_p}{2\sigma_p^2} + \frac{r^2 L^2}{2} \right) \frac{\tau_s \tau_i}{\Omega_s \Omega_i} \right].
\] (11)

Equation (11) indicates that any unfactorability resides in the mixed term containing an argument proportional to \(\Omega_s \Omega_i\). Therefore, the conditions to guarantee the elimination of the mixed term are

\[
\frac{1}{\sigma_p^2} = -r^2 L^2 \tau_s \tau_i
\] (12)

and

\[
C_p = 0,
\] (13)

which imply that to factorize the JSA in equation (11), in addition to satisfying the bandwidth requirement in equation (12), the pulsed pump should be transform limited. Obviously, a finite pump bandwidth \(\sigma_p\) can be deduced from equation (12) only if the phase matching condition \(\tau_s \tau_i < 0\) is fulfilled. While under the conditions of \(\tau_i = 0\) or \(\tau_s = 0\), termed the AGVM conditions, equation (12) can be approximately satisfied by using the transform-limited pump with a very broad bandwidth.

Substituting equation (11) into (8), we obtain the analytical expression

\[
g^{(2)} = 1 + \frac{|\tau_s - \tau_i|}{\sqrt{\tau_s^2 + \tau_i^2 + (r \tau_s \tau_i L \sigma_p)^2 + (1 + C_p^2) / (r L \sigma_p)^2}}
\]

\[
= 1 + \frac{1}{\sqrt{1 + \frac{(r \tau_s \tau_i L \sigma_p)^2 + 1 + C_p^2}{(\tau_s - \tau_i)^2 (r L \sigma_p)^2}}}.
\] (14)

Equation (14) clearly shows that \(g^{(2)}\) decreases with an increase of \(C_p\), which means that \(C_p\) in equation (11) refers to the phase contribution to JSA. A quantitative analysis of the influence of pump chirp on the factorability of JSA was not included in the earlier work by U’Ren et al [14].

To further illustrate the dependence of factorability, we first assume that the phase matching condition \(\tau_s \tau_i < 0\) is fulfilled and the pump bandwidth is regulated to satisfy equation (12). In this case, we arrive at

\[
g^{(2)} = 1 + \frac{1}{\sqrt{1 + \frac{r \tau_s \tau_i}{(\tau_s - \tau_i)^2}}},
\] (15)

showing that we have \(g^{(2)} = 2\) for \(C_p = 0\). For the case of \(C_p \neq 0\), there is no way to eliminate the influence of frequency correlation introduced by the pump chirp. We then assume that the AGVM condition \(\tau_i(s) = 0\) is satisfied. In this case, we have

\[
g^{(2)} = 1 + \frac{1}{\sqrt{1 + \frac{1 + C_p^2}{(r \tau_s \tau_i L \sigma_p)^2}}} = 1 + \frac{1}{\sqrt{1 + \frac{\Delta T_p}{(r \tau_s \tau_i L)^2}}}.
\] (16)
which indicates that the presence of $C_p$ causes a reductive $g^{(2)}$, and the ideal factorable state with $g^{(2)} = 2$ cannot be realized unless the pump pulse duration $\Delta T_p$ is infinitely short or the PCF length $L$ is infinitely long, i.e. $\frac{\Delta T_p^2}{(\tau_{\text{real}} L)^2} \rightarrow 0$. Although equation (16) implies that for the case of $C_p \neq 0$, the value of $g^{(2)}$ can be reserved to some extent by enlarging the bandwidth of pump $\sigma_p$ or by increasing the length $L$ of PCF. However, a realistic numerical analysis and an experimental investigation presented in later sections illustrate that the solution to reduce the influence of pump chirp is not practical because the approximations used in deriving the analytical expressions of $g^{(2)}$ often deviate from the real experimental conditions.

4. Numerical simulation of the intensity correlation function $g^{(2)}$

In this section, we numerically calculate $g^{(2)}$ by taking the sinc oscillation of the phase matching function and all order terms of $\Delta k$ into account. Considering that the phase matching function plays a key role in determining the frequency correlation of photon pairs, we first characterize the phase matching contour of a given PCF. Then, under the phase matching condition suitable for generating a factorable state $\tau_s \tau_i \leq 0$, we numerically investigate the dependence of $g^{(2)}$ by varying the experimental parameters, including the central wavelength, bandwidth and chirp of pump and the length of PCF. After respectively comparing the results with those obtained by applying the linear approximation of dispersion and by the analytical expression, we illustrate how to minimize the frequency correlation of photon pairs.

4.1. Characterization of the phase matching contour

We start with predicting the phase matching contour of SFWM in our 0.6 m long PCF (NL-1050-ZERO-2, Crystal Fibre). Using the method proposed in [26] and assuming that the dispersion is homogeneous along the PCF, we first experimentally deduce the effective core radius $r$ and air fraction $f$ of the PCF along the fast axis. By varying the central wavelength of the pump from 1037 to 1047 nm and measuring the spectra of the signal and idler photons via SFWM, the values of $r$ and $f$ are found to be 0.949 $\mu$m and 29.5%, respectively. Based on the accordingly calculated dispersion property, we then compute the phase matching curve of co-polarized SFWM when the pump wavelength $\lambda_{pc} = \frac{2 \pi}{\omega_{pc}}$ is altered, as shown in figure 1(a). Moreover, to clarify the phase matching conditions suitable for generating factorable photon pairs, the calculated group velocity mismatch terms $\tau_s$ and $-\tau_i$, and the product $\tau_s \tau_i$ versus $\lambda_{pc}$ are plotted in figure 1(b). One sees that bounded by the AGVM conditions of $\tau_i = 0$ and $\tau_s = 0$, which are satisfied at $\lambda_{pc} = 1070$ and 987 nm, respectively, the phase matching condition $\tau_s \tau_i \leq 0$ can be fulfilled in the range of 987 nm $\leq \lambda_{pc} \leq 1070$ nm. Additionally, it is worth noting that the so-called SGVM condition $\tau_s = -\tau_i$, which is of interest in generating spectral factorable photon pairs with symmetric spectra [27], is satisfied at $\lambda_{pc} = 1014.5$ nm.

Figure 1(c) plots the contour map of the phase matching function $\phi(\omega_s, \omega_i)$ in the frequency ($\{\omega_s, \omega_i\}$) and wavelength ($\{\lambda_s, \lambda_i\}$) space. For a pulsed pump with a central wavelength of $\lambda_{pc}$, we characterize the corresponding phase matching contour from the following three aspects: orientation, bandwidth and curvature. We note that the contour map is also calculated by using the approximation of the phase mismatch term in equation (7), and the results indicate that the contribution of dispersion coefficients higher than the second order is negligible. Therefore, we will only discuss how the first- and second-order dispersion coefficients affect the phase matching contour.

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The orientation of the phase matching contour at $\lambda_{\text{pc}}$ is described by using the angle $\theta_{i(s)}$ between the symmetric line of the $\phi(\omega_s, \omega_i)$ and $\omega_i - (\omega_s -)$ axis. According to the expression

$$\theta_i = \arctan(\tau_i / \tau_s),$$

one sees that $\theta_s$ changes from $-\frac{\pi}{2}$ to 0 for $\lambda_{\text{pc}}$ varying from 987 to 1070 nm (figure 1(d)), and it is straightforward to deduce that

$$\theta_s = -\arctan(\tau_s / \tau_i)$$

will accordingly change from 0 to $\frac{\pi}{2}$. In particular, for the AGVM condition $\tau_{s(i)} = 0$ at $\lambda_{\text{pc}} = 987$ (1070) nm, we have $\theta_{s(i)} = 0$, and the orientation of $\phi(\omega_s, \omega_i)$ is parallel to the $\omega_{s(i)}$-axis; while for the SGVM condition $\tau_s = -\tau_i$ at $\lambda_{\text{pc}} = 1014.5$ nm, we have $\theta_s = -\frac{\pi}{4},$ and
are obtained for AGVM conditions (20) (21).

It is clear that for \( \lambda \) frequency region of the pulsed pump, and the correlation shown in figure



Because the experimentally measurable joint spectral intensity (JSI) function

\[ |f(\omega_s, \omega_i)|^2 = \exp \left[ -\frac{(\omega_s + \omega_i - 2\omega_{pc})^2}{2\sigma_p^2} \right] \text{sinc}^2 \left( \frac{\Delta k L}{2} \right) \]  

(22)
gives an intuitive picture of the frequency correlation of photon pairs [12], before numerically studying the dependence of \( g^{(2)} \), we first plot the contour map of JSI under different phase matching conditions by substituting the dispersion coefficient of the 0.6 m PCF into equations (4)–(6) and by varying the bandwidth of the pump. The plots in the top and middle of figure 2 are obtained for AGVM conditions \( \tau_1 = 0 \) (\( \lambda_{pc} = 1070 \text{ nm} \)) and \( \tau_1 = 0 \) (\( \lambda_{pc} = 987 \text{ nm} \)), respectively. It is clear that for \( \Delta \lambda_p = 2.3 \text{ nm} \), the frequencies of signal and idler photons are negatively correlated; while for \( \Delta \lambda_p = 4.6 \text{ nm} \), the frequency correlation between signal and idler photons turns out to be indistinct. However, in contrast to the prediction of equation (14), when \( \Delta \lambda_p \) further increases to 15 nm, frequency correlation becomes observable in the marginal frequency region of the pulsed pump, and the correlation shown in figure 2(f) is more than that in figure 2(c), which clearly illustrates the influence of high-order dispersion.
Figure 2. The contour map of JSI, \( |f(\omega_s, \omega_i)|^2 \), for different pump bandwidths \( \Delta \lambda_p \) when the phase matching condition is varied by changing the central wavelength of pump \( \lambda_{pc} \). The three plots at the top, (a)–(c), are obtained for \( \lambda_{pc} = 1070 \) nm; the three plots in the middle, (d)–(f), are obtained for \( \lambda_{pc} = 987 \) nm; and the three plots at the bottom, (g)–(i), are obtained for \( \lambda_{pc} = 1060 \) nm. The calculation is based on the calculated dispersion of the 0.6 m PCF.

We also plot the contour map of JSI for the pump satisfying the phase matching condition \( \tau_i \tau_s < 0 \). Without losing generality, we choose the case of \( \lambda_{pc} = 1060 \) nm. When \( \Delta \lambda_p = 3.1 \) nm, the bandwidth requirement in equation (12) is fulfilled, and the two symmetric axes of the elliptically featured JSI are respectively parallel to the \( \omega_i \) and \( \omega_s \) axes. In this situation, the frequency correlation mainly originates from the sinc oscillation of \( \phi(\omega_s, \omega_i) \), as shown in figure 2(h). However, if \( \Delta \lambda_p \) is less than or greater than 3.1 nm, negative or positive frequency correlation between signal and idler photons would become distinct, as shown in figure 2(g) or figure 2(i).

Since the JSI is not sensitive to the pump chirp, figure 2 only qualitatively illustrates the dependence of frequency correlation of photon pairs to some extent. To thoroughly analyze the
Influences of high-order dispersion and sinc oscillation of phase matching function

We first numerically calculate $g^{(2)}$ of the individual signal photons by using equation (8) with no approximation involved. In the calculation, the phase matching function is obtained by directly substituting the dispersion of the 0.6 m PCF into equation (6), and the pump is assumed to be transform limited. We compute $g^{(2)}$ as a function of $\Delta \lambda_p$ for the pump with a specified $\lambda_{pc}$ in the range of 987–1070 nm. As shown in figure 3, the changing tendency of each set of data is similar: with the increase of $\Delta \lambda_p$, the graph presents an ascending curve in the beginning and then starts to descend when $g^{(2)}$ reaches the maximum at a certain turning point of $\Delta \lambda_p$. Moreover, with the variation of $\lambda_{pc}$, the values of the maximum $g^{(2)}$ and turning point of $\Delta \lambda_p$ will accordingly change.

As a comparison, based on the same parameters of pump used in plotting figure 3, we calculate $g^{(2)}$ by applying the linear approximation of $\Delta k$ (equation (10)). Figure 4 shows that different from figure 3, for $\lambda_{pc}$ near 1070 or 987 nm, the maximum $g^{(2)}$ and the corresponding turning points of $\Delta \lambda_p$ in figure 4 are greater than those in figure 3. In particular, for $\lambda_{pc} = 1070$ nm.
Figure 5. Calculated $g^{(2)}$ versus $\Delta \lambda_p$ for different $\lambda_{pc}$. The calculation is done by using the analytical expression (equation (14)), obtained by using the linear approximation of $\Delta k$ and the Gaussian approximation of sinc function.

Figure 6. (a) The maximum $g^{(2)}$ and (b) the corresponding turning point of $\Delta \lambda_p$ versus $\lambda_{pc}$. The calculations are performed by using three kinds of methods as labeled in the figure.

or 987 nm, $g^{(2)}$ in figure 4 always increases with $\Delta \lambda_p$, and the descending trend does not appear. These significant differences between figures 3 and 4 reflect the influence of the high-order dispersion.

We also calculate $g^{(2)}$ by substituting parameters of pump into the analytical expression of equation (14). As shown in figure 5, the maximum $g^{(2)}$ of each set of data is always 2, having no relevance to $\lambda_{pc}$. In particular, for a certain $\lambda_{pc}$ within the middle range of 987–1070 nm, the maximum $g^{(2)}$ in figure 4 is obviously less than that in figure 5. The difference between the data in figures 4 and 5 illustrates the influence of sinc oscillation of the phase matching function.

In order to better illustrate the unfactorability induced by the high-order dispersion and the sinc oscillation, based on $g^{(2)}$ calculated by exploiting the methods used in figures 3–5, respectively, we distil the information about maximum $g^{(2)}$ and turning point of $\Delta \lambda_p$ for each specified $\lambda_{pc}$, and the changing step of $\lambda_{pc}$ is much smaller than that in figures 3–5. As shown in figures 6(a) and (b), when the value of the turning points of $\Delta \lambda_p$ obtained by equation (14) is relatively small, the corresponding $\lambda_{pc}$ is in the middle range of 987–1070 nm. In this case, the turning points of $\Delta \lambda_p$ obtained by the three kinds of methods are about the same, but the values of maximum $g^{(2)}$ obtained with no approximation and with linear approximation,
which are almost equal to each other, are distinct from the analytically calculated result: maximum \( g^{(2)} \equiv 2 \). This is because the influence of high-order dispersion, determined by the pump bandwidth, is negligible, and the frequency correlation mainly originates from the sinc oscillation. Moreover, comparing the results in figures 6 and 1(d), one sees that the influence of the sinc oscillation depends on the orientation of the phase matching function, which has a mild effect on the turning point \( \Delta \lambda_p \), but plays an important role in determining the maximum \( g^{(2)} \). For the SGVM condition \( \theta_i = -\frac{\pi}{4} \), the impact of sinc oscillation is maximized, and the achievable maximum \( g^{(2)} \approx 1.82 \) is the smallest; while for the AGVM conditions \( \theta_{s(i)} \to 0 \), the impact of sinc oscillation is minimized. Hence, when \( \lambda_{pc} \) is approaching 1070 or 987 nm, the maximum \( g^{(2)} \) obtained by using the linear approximation is getting close to the analytically calculated result.

On the other hand, when the value of turning points of \( \Delta \lambda_p \) obtained by using equation (14) is large, the corresponding \( \lambda_{pc} \) is around 1070 or 987 nm. In this case, the maximum \( g^{(2)} \) and turning points of \( \Delta \lambda_p \) calculated with no approximation are smaller than those with two kinds of approximations, mainly due to the frequency correlation contributed by the high-order dispersion. Figure 6 shows that a better factorability can be realized for \( \lambda_{pc} \) around 1070 or 987 nm. In the vicinity of 1070 nm, the peak value of maximum \( g^{(2)} \), about 1.94, is obtained for pump with \( \lambda_{pc} = 1069.5 \) nm and \( \Delta \lambda_p = 10.4 \) nm, respectively; while in the vicinity of 987 nm, the peak value of maximum \( g^{(2)} \), about 1.90, is obtained for pump with \( \lambda_{pc} = 988.5 \) nm and \( \Delta \lambda_p = 7.4 \) nm, respectively. Clearly, for the former case, the peak value of maximum \( g^{(2)} \) is greater and less distinct. Therefore, to improve the factorability, in addition to mitigating the influence of the sinc oscillation of \( \phi(\omega_s, \omega_i) \) by satisfying the condition \( \tau_i \approx 0 \) (\( \tau_s \approx 0 \)), the influence of high-order dispersion can be reduced by using PCF with a greater \( \tau_s \) (\( \tau_i \)) and a smaller \( \xi_i \) (\( \xi_s \)), which means that the curvature of the phase matching curve is less tight. Moreover, it is worth pointing out that the difference between the calculated values of maximum \( g^{(2)} \) at \( \lambda_{pc} = 1069.5 \) and 1070 nm is only about 0.001. For a real experiment presented in the next section, \( g^{(2)} \) obtained at \( \lambda_{pc} = 1070 \) nm should be very close to the highest.

4.4. Influences of pump chirp and length of photonic crystal fiber

Having performed the calculation by using a transform-limited pump in the given 0.6 m PCF, we then numerically analyze the influences of the pump chirp \( C_p \) and the PCF length \( L \) on the factorability with no approximation involved. For clarity, we only calculate \( g^{(2)} \) of individual signal photons as a function of \( \Delta \lambda_p \) for the cases of \( \lambda_{pc} = 1014.5 \) nm and \( \lambda_{pc} = 1070 \) nm, respectively. For each \( \lambda_{pc} \), \( L \) is assumed to be 0.6 and 1.5 m, respectively, and \( C_p \) is assumed to be \( C_p = 0 \) and \( |C_p| = 0.75 \), respectively.

Figure 7(a) is obtained under the SGVM condition \( \lambda_{pc} = 1014.5 \) nm, which shows that the introduction of \( C_p \) results in a decreased \( g^{(2)} \), and the decrement declines with an increase of \( \Delta \lambda_p \). Moreover, for a certain \( C_p \), the maximum \( g^{(2)} \) does not vary with \( L \). This is because the optimized pump bandwidth—the turning point of \( \Delta \lambda_p \)—is almost reversely proportional to \( L \) and is relatively narrow. In this case, the dominant factor limiting the factorability is the sinc oscillation, which only depends on the orientation of phase matching function.

Figure 7(b) is obtained under the AGVM condition \( \lambda_{pc} = 1070 \) nm, which also shows that the presence of pump chirp results in a reduced \( g^{(2)} \). However, in contrast to figure 7(a), for a certain \( C_p \), the maximum \( g^{(2)} \) increases with \( L \), and the relation between the turning point of \( \Delta \lambda_p \) and \( L \) is different from that in figure 7(a). To understand the dependence of \( L \), we qualitatively analyze the mechanism of obtaining the factorable state. Generally speaking, \( g^{(2)} \)
Figure 7. $g^{(2)}$ of individual signal photons versus $\Delta \lambda_p$ for (a) SGVM condition ($\lambda_{pc} = 1014.15$ nm) and (b) AGVM condition ($\lambda_{pc} = 1070$ nm), respectively. The calculation is performed by varying $L$ and $C_p$, and no approximation is involved.

increases with a decrease of the time uncertainty of signal photons. In the case of $\tau_i = 0$, the time uncertainty is described by the ratio between the pump pulse duration and coherence time of signal photons $\Delta T_p/\delta T_s$ [28]. Ideally, the factorability can be improved by decreasing $\Delta T_p$. However, limited by the frequency correlation originating from the high-order dispersion, the optimized minimum $\Delta T_p$ cannot be infinitely short. To figure out how to optimize $\Delta T_p$, we need to study the phase mismatch term in equation (7). Because the bandwidth of idler photons is much broader than that of signal photons for $\tau_i = 0$ (see figure 2(c)), the high-order dispersion terms $\xi_s \Omega_s^2$ and $\frac{k_p}{2} \Omega_s \Omega_i$ can be neglected, and equation (7) can be simplified as

$$\Delta k L \approx L \tau_s \Omega_s + L \xi_i \Omega_i^2,$$

where the value of $\Omega_{\text{a(i)}}$ is associated with the bandwidth of signal (idler) photons, and the bandwidth of idler photons is determined by the pump bandwidth $\Delta \lambda_p$. To ensure that the high-order dispersion effect-induced phase mismatch, mainly contributed by the term $L \xi_i \Omega_i^2$ in equation (23), is less than a certain value, i.e. $L \xi_i \Delta \lambda_p^2 < \delta$, the corresponding pump bandwidth, which is almost equivalent to the turning point of $\Delta \lambda_p$, should be reversely proportional to $\frac{1}{\sqrt{L}}$.

Hence, the optimized pump pulse duration $\Delta T_p \propto \frac{1}{\Delta \lambda_p}$ is proportional to $\sqrt{L}$. On the other hand, when the influence of high-order dispersion is negligible, $\delta T_s$ determined by the bandwidth of phase matching function is proportional to $\tau_s L$. However, when $\Delta \lambda_p$ is greater than a certain value, say, the turning point in figure 7(b), the frequency correlation between signal and idler photons in the marginal frequency region will become distinct and the bandwidth of signal photons will increase, resulting in a decreased $\delta T_s$ and $g^{(2)}$. So the longest $\delta T_s$ is proportional to $L$. Thus, we have $\Delta T_p/\delta T_s \propto \frac{1}{\sqrt{L}}$, which implies that the maximum $g^{(2)}$ increases with $L$.

4.5. Improving factorability by optimizing experimental parameters

Our numerical calculation and analysis indicate that in addition to the usage of a transform-limited pump and the satisfaction of phase matching condition $\tau_i \tau_s \leqslant 0$, the key factors affecting the factorability of photon pairs are: (i) the frequency correlation originating from the sinc oscillation of phase matching function and (ii) the high-order dispersion of PCF. The two factors cannot be eliminated simultaneously, because the suppression of the former leads to $\tau_i \tau_s \rightarrow 0$.
and $\sigma_p \to \infty$, which will enhance the negative influence of the latter (see equations (7) and (12)).

Therefore, to maximize $g^{(2)}$, the side effects originating from the two factors should be properly balanced.

5. Experiment

To verify the numerical simulations, we present two series of experiments by regulating the spectrum of the pump to satisfy the AGVM and SGVM conditions in the 0.6 m PCF, respectively. The experimental setup is shown in figure 8. A compact mode-locked femtosecond fiber laser based on Yb-doped PCF is employed as the pump source [29], whose linearly polarized output is a 54.4 MHz pulse train. The central wavelength and full-width at half-maximum (FWHM) of the laser output are 1042 and 8 nm, respectively. To obtain the specified pump wavelengths for realizing the AGVM and SGVM conditions and to ensure that the power spectrum is high enough for conducting the experiments, we first send the 400 mW laser output into a 0.4 m long single-mode fiber (SMF) to expand the spectrum to $\sim 70$ nm via the self-phase modulation effect. By properly adjusting the grating $G$, we are able to carve out the pump pulses with the desired central wavelengths, whose FWHM can be tuned from 0.6 to 7 nm. After passing through the polarization beam splitter (PBS), the polarization of the pump is adjusted along the fast axis of the PCF by using a half-wave-plate (HWP). To ensure the reliable detection of the photon pairs via SFWM, a prism is placed at the output port of the PCF to reject the residual pump and to separate signal and idler photons.

In order to figure out a fine spectral property of the photon pairs, the prism is followed by tunable narrow band-pass filters (NBFs) to achieve high resolution. The NBFs with Gaussian-shaped spectra are realized by gratings, and the FWHMs of NBF1 in the idler band and NBF2 in the signal band are 0.45 and 0.8 nm, respectively. To visualize the spectral property of photon pairs generated from the PCF, we not only record single counts of individual idler and signal photons by scanning the central wavelengths of NBF1 and NBF2, respectively, but also make a series of coincidence measurements by fixing the spectra of the pump and idler photons and scanning the central wavelength of NBF2 to distil the key features of JSI [30].

Before being respectively detected by SPD1 (silicon-based SPD, SPCM-AQRH-15) and SPD2 (InGaAs/InP based SPD, id200), the idler and signal photons are coupled into single-mode fibers. SPD1 is working in the active quench mode, while SPD2 is working in the gated-Geiger mode, whose 2.5 ns-wide gate pulses arrive at a rate of about 3.4 MHz, 1/16 of the repetition rate of the pump pulses. The detection signals of both SPD1 and SPD2 are fed into the counting system so that single counts and coincidence of signal and idler photons in different time slots can be recorded. The total detection efficiencies, including the filtering system and the SPDs, are about $\sim 4$ and $\sim 10\%$ in signal and idler bands, respectively.

When $g^{(2)}$ of the individual signal field is measured (see the inset of figure 8), the NBF2 is then replaced with a long pass filter (LPF) with a cut-off wavelength of 1200 nm. In this situation, the FWHM in the signal band increases to $\sim 45$ nm, which is exclusively provided by the prism and is at least ten times broader than the natural bandwidth of individual signal photons. Therefore, the measurement can be approximated as a filter-free case. In the measurement, a 50/50 beam splitter (BS) is used to split the signal field, and the two outputs of BS are fed into SPD2 and SPD3 (id200), respectively. The value of $g^{(2)}$ is the ratio between the measured coincidence and accidental coincidence rates, which are the coincidences of signal photons originating from the same pulse and adjacent pulses, respectively.
Figure 8. The experimental setup. The inset is the setup for measuring the intensity correlation function \( g^{(2)} \) of individual signal photons. SMF, single-mode fiber; G, grating; PBS, polarization beam splitter; HWP, half-wave plate; NBF1–NBF2, narrow band filters; SPD1–SPD3, single-photon detectors; LPF, long pass filter; BS, beam splitter.

Table 1. \( g^{(2)} \) of individual signal fields for \( \lambda_{pc} = 1070 \) nm.

| \( \Delta \lambda_p \) (nm) | \( \Delta T_p \) (ps) | \( C_p \) | Measured | Theoretical |
|--------------------------|-----------------|---------|----------|-------------|
| 2.3                      | 0.73            | 0       | 1.60 ± 0.02 | 1.81        |
| 4.6                      | 0.45            | 0.72    | 1.65 ± 0.02 | 1.90        |
| 6.9                      | 0.30            | 0.72    | 1.76 ± 0.01 | 1.94        |

5.1. Generation of photon pairs under the asymmetric group velocity matching condition

According to the calculation in section 4, in the first series of experiments, we adjust the central wavelength of the pump \( \lambda_{pc} \) to 1070 nm. Because the factorability of the photon pairs highly relies on the bandwidth of the pump, the measurements are made by using a pump with different \( \Delta \lambda_p \). Additionally, the pulse duration \( \Delta T_p \) of each kind of pump is measured by using an autocorrelator, and the corresponding chirp \( C_p \) is accordingly deduced, as listed in table 1.

We first record the single count rates of SPD1 and SPD2 by varying the central wavelength of NBF1 and NBF2, respectively. We find that the central wavelengths of signal and idler photons are 1408 and 862.9 nm, respectively, which only slightly deviates from the calculated result, 1407.5 and 862 nm (figure 1). The results indicate that the calculation in section 4 is valid, and the AGVM condition is approximately satisfied.

We then make a series of coincidence measurements to extract the key features of JSI. For a certain \( \Delta \lambda_p \), when the central wavelength of NBF1 \( \lambda_{iF} \) is fixed at 862, 862.9 and 863.7 nm, respectively, we record the coincidences of signal and idler photons by scanning the central wavelength of NBF2 \( \lambda_{sF} \). For each set of data, the signal wavelength, \( \lambda'_{s0} \), which corresponds to the peak coincidence, is accordingly deduced. Figure 9(a), obtained by using the pump with bandwidth \( \Delta \lambda_p \), chirp \( C_p \) and average power \( P_a \) of about 2.3 nm, 0 and 1.7 mW, respectively, exhibits that the wavelength \( \lambda'_{s0} \) increases with a decrease of \( \lambda_{iF} \), indicating that the frequencies of signal and idler photons are slightly anti-correlated. While figure 9(b), obtained by using the pump with \( \Delta \lambda_p \), \( C_p \) and \( P_a \) of about 4.6 nm, 0.74 and 1 mW, respectively, shows that the wavelength \( \lambda'_{s0} \) does not vary with \( \lambda_{iF} \), indicating that the frequency correlation of signal and idler photons is almost unobservable.

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Figure 9. (a) Coincidences versus the central wavelength of NBF2 in signal band $\lambda_{SF}$ for (a) $\Delta \lambda_p = 2.3$ nm and (b) $\Delta \lambda_p = 4.6$ nm, respectively, as the central wavelength of NBF1 in idler band $\lambda_{IF}$ is fixed at different wavelengths. In the measurement, $\lambda_{pc} = 1070$ nm. The insets in plots (a) and (b) are the calculated results. The solid lines in the main plots are only for guiding the eyes.

As a comparison, we deduce the expression of coincidence

$$C(\omega_{SF}, \omega_{IF}) \propto \int d\omega_s d\omega_i |f(\omega_s, \omega_i)|^2 \exp \left[ -\frac{(\omega_s - \omega_{SF})^2}{\sigma_{SF}^2} \right] \exp \left[ -\frac{(\omega_i - \omega_{IF})^2}{\sigma_{IF}^2} \right],$$

(24)

by taking the NBFs into account [31], where $\omega_{SF}$ ($\sigma_{SF}$) and $\omega_{IF}$ ($\sigma_{IF}$) are the central frequency (bandwidth) of NBFs in signal and idler bands, respectively. To ensure that the step change of $\lambda_{IF}$ is the same as that in the experiments, we perform the calculation when the wavelength $\lambda_{IF}$ is 861.1, 862 and 862.8 nm, respectively. The results are shown in the insets of figures 9(a) and (b), which illustrates that the theoretical predictions qualitatively agree with the experimental results. We note that, for each set of experimental data, there is a small peak sitting next to the main peak of coincidence, which can be ten times greater than that of the theoretically predicted sinc oscillation. Since the coincidence patterns measured under the AGVM condition reflect the shape of the phase matching function [25], and the calculation is performed by assuming that the PCF has homogeneous dispersion, we believe that the observation of the theoretically unexpected small peak is because the dispersion of the PCF used in experiment is not homogeneous.

Next, we quantitatively test the factorability of photon pairs by measuring $g^{(2)}$ of the signal photons when $\Delta \lambda_p$ is adjusted to be 2.3, 4.6 and 6.9 nm, respectively. We first make a measurement when the spectrum broadening of the pump due to self-phase modulation in the 0.6 m PCF is not obvious. In this case, the chirp of the pump propagating along the PCF can be viewed as a constant because the group velocity dispersion of the PCF is flattened and near zero around 1050 nm. The measured results (with the dark counts of SPDs subtracted) are shown in table 1. For the sake of comparison, we also numerically calculate the corresponding $g^{(2)}$ by substituting the experimental parameters into equations (4)–(6) and (8), and list the results in table 1 as well. One sees that the measured results of $g^{(2)}$ are less than the theoretical predictions. Considering that the Raman scattering at such a large detuning is negligible [16, 32], we believe that the observed reduction of $g^{(2)}$ is caused by the inhomogeneity of PCF [25]. However, it is obvious that the measured $g^{(2)}$ increases with a decrease of pump pulse duration $\Delta T_p$, indicating that the changing tendency observed in experiment agrees with the theoretical analysis.

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Table 2. \( g^{(2)} \) of the signal field for the AGVM condition with pump power \( P_a \) varied.

| \( \Delta \lambda_p' \) (nm) | \( \Delta T_p' \) (ps) | Production rate (pairs per pulse) | \( P_a \) (mW) | Measured \( g^{(2)} \) |
|-------------------------|------------------|-------------------------------|-----------|------------------|
| 6.9                     | 0.30             | 0.026                         | 1.0       | 1.76 ± 0.01      |
| 9.2                     | 0.30             | 0.071                         | 1.7       | 1.78 ± 0.01      |
| 14                      | 0.30             | 0.16                          | 2.4       | 1.73 ± 0.01      |

In order to verify the dependence of high-order dispersion upon the factorability of photon pairs, we then measure \( g^{(2)} \) of the signal photons by fixing the FWHM of the incident pump at 6.9 nm and changing the average pump power \( P_a \). In this case, with an increase of \( P_a \), the bandwidth of residual pump \( \Delta \lambda_p' \) becomes broader due to the self-phase modulation, but the change in the pulse duration of residual pump \( \Delta T_p' \) is not observable. When \( P_a \) is 1.0, 1.7 and 2.4 mW, respectively, \( \Delta \lambda_p' \) is 6.9, 9.2 and 14 nm. The accordingly measured \( g^{(2)} \) and deduced production rates of photon pairs are listed in table 2. We find that when \( \Delta \lambda_p' \) is less than 9.2 nm, \( g^{(2)} \) is mainly determined by the pump pulse duration and the gain of SFWM. However, when \( \Delta \lambda_p' \) reaches 14 nm, we observe a decreased \( g^{(2)} \). We believe that this is evidence for demonstrating the unfactorability induced by the high-order dispersion of the PCF.

5.2. Generation of photon pairs under the symmetric group velocity matching condition

In the second series of experiments, we adjust the central wavelength of pump \( \lambda_{pc} \) to 1014.5 nm, at which the SGVM condition is satisfied. Moreover, according to the calculation, the pump bandwidth \( \Delta \lambda_p \) is set to 2.3 nm, so that it matches the bandwidth of phase matching function, i.e. equation (12) is fulfilled. When the average power of pump is 1.3 mW, the pulse durations for both the incident and residual pumps are 0.66 ps, indicating that the pump propagating along the PCF is about transform limited. In this case, we first record the single count rates of SPD1 and SPD2 by varying the central wavelengths of NBF1 and NBF2, respectively. The accordingly plotted spectra of individual signal and idler photons are shown in figures 10(a) and (b), respectively. It is clear that the signal and idler photons are centering at 1364 and 807.5 nm, respectively, which is only slightly deviating from the numerically calculated results (1363.2 and 807.9 nm). Moreover, the measured FWHMs of signal and idler photons, 0.72 and 0.76 THz, are almost equal, which is one of the signatures of the factorable state under the SGVM condition.

To further demonstrate the key features of JSI, we record the coincidence rate by scanning the wavelength \( \lambda_{sF} \) when \( \lambda_{iF} \), is fixed at 806.9, 807.5 and 808.1 nm, respectively. As shown in figure 10(c), the values of wavelengths \( \lambda_{s0} \), corresponding to the highest coincidences, do not vary with \( \lambda_{iF} \), indicating that no obvious frequency correlation between the signal and idler photons is observed in this way.

\(^2\) Our latest investigation indicates that when the high-order terms of \( G \propto \gamma P_a L \) in equation (3) are included, for the pump satisfying the AGVM condition, \( g^{(2)} \) calculated by using the linear and Gaussian approximations will increase with \( G \), i.e. \( g^{(2)} \) will increase with the pair production rate.
Figure 10. Spectral property of signal and idler photon pairs for pump with $\lambda_{pc} = 1014.5$ nm and $\Delta \lambda_p = 2.3$ nm. Panels (a) and (b) are the single counts of SPD2 and SPD1 as a function of the central wavelength of NBFs in signal and idler bands, $\lambda_{sF}$ and $\lambda_{iF}$, respectively, showing the spectrum of individual signal and idler photons, respectively. $f_{iF}$ is related to $\lambda_{iF}$ through the relation $f_{iF} = 1/\lambda_{iF}$. (c) Coincidences versus $\lambda_{sF}$ for $\lambda_{iF}$ fixed at different wavelengths. The insets of plots (a), (b) and (c) are the calculated results. The solid lines in the main plots are only for guiding the eyes.

Table 3. $g^{(2)}$ of individual signal fields for $\lambda_{pc} = 1014.5$ nm.

| $\Delta \lambda_p$ (nm) | $\Delta T_p$ (ps) | $C_p$ | Measured | Theoretical |
|------------------------|-------------------|-------|----------|-------------|
| 2.3                    | 0.66              | 0     | 1.60 ± 0.02 | 1.82         |
| 2.3                    | 1.70              | 2.4   | 1.26 ± 0.03 | 1.55         |

To compare the experimental results with theoretical predictions, we then deduce the expression of single counts in the signal (idler) band [25]

$$S_{s(i)}(\omega_{sF(iF)}) \propto \int d\omega_s d\omega_i |f(\omega_s, \omega_i)|^2 \exp \left[-\frac{(\omega_s - \omega_{sF(iF)})^2}{\sigma_{sF(iF)}^2}\right],$$

(25)

under the experimental conditions, and the calculated results of single counts in signal and idler bands are plotted in the insets of figures 10(a) and (b), respectively. We find that the spectra of the calculated results are symmetric, but the spectra of measured signal and idler photons are asymmetric. Moreover, by substituting the experimental parameters into equation (24), we find that the calculated pattern of coincidence (see the inset of figure 10(c)) does not show obvious oscillations in the wing. However, the experimental results show that there is a small peak that sits next to the main coincidence peak (the main plot of figure 10(c)). Again, we believe that the difference between the experimental observations and theoretical predictions is because of the inhomogeneity of PCF.

As a quantitative verification of the factorability, we also measure the $g^{(2)}$ of the individual signal field (see table 3). When the pump propagating along the PCF is about transform limited, $g^{(2)}$ is measured to be $1.60 \pm 0.02$. Moreover, in order to demonstrate the dependence of pump chirp $C_p$, we repeat the measurements by replacing the transform-limited pump with a chirped.
The pump chirp $C_p \approx 2.4$, which is almost constant along the PCF, is introduced by passing the pump pulses through a grating pair (600 lines mm$^{-1}$) twice to stretch the pulse duration to 1.7 ps. In this situation, the measured $g^{(2)}$ is decreased to $1.26 \pm 0.03$, owing to the extra chirp-induced frequency correlation. Additionally, for the sake of comparison, we also numerically calculate $g^{(2)}$ by substituting the experimental parameters into equations (4)–(6) and (8), and list it in table 3. The results show that, similar to the case of the AVGM condition, the changing tendency observed in experiments is consistent with the theoretical calculations, but the measured results are less than the calculated results due to the inhomogeneity of PCF.

Finally, considering that the photon pairs with a higher production rate are desirable for some applications [33], we also measure $g^{(2)}$ by increasing the average power $P_a$ of the transform-limited pump. When $P_a$ is increased from 1.3 to 2.4 mW, the production rate of photon pairs is increased from $\sim 0.02$ to $\sim 0.08$ pair per pulse. In this case, the pulse duration of the residual pump does not change, but the spectrum is broadened to 2.9 nm. Accordingly, the measured $g^{(2)}$ is decreased to $1.56 \pm 0.02$. Comparing with the results in table 2, one sees that under the SGVM condition, generating frequency de-correlated photon pairs with a high production rate is more challenging because the broadened pump spectrum not only introduces chirp via Kerr nonlinearity, but also spoils the required matching between the bandwidths of pump and phase matching function, which is more critical than that in AGVM condition (see figure 3).

6. Conclusion

In conclusion, our investigation indicates that it is necessary to conduct the numerical simulation to optimize the experimental parameters for directly generating photon pairs with a minimized frequency correlation via pulse-pumped SFWM. The simulation illustrates that it is impossible to realize an ideal factorable state due to the influence of the high-order dispersion and the intrinsic sinc oscillation of the phase matching function. In particular, when the SGVM condition $-\tau_i = \tau_s$ is fulfilled, limited by the sinc oscillation, the achievable maximum $g^{(2)}$ is only about 1.82, which is usually independent of the PCF length. In this case, an ideal factorable state with $g^{(2)} = 2$ can be created in principle if $\phi(\omega_s, \omega_i)$ can be engineered to be Gaussian-shaped [34]; however, the solution is not practical at the current stage. While when the AGVM condition $\tau_i = 0$ ($\tau_s = 0$) is fulfilled, the factorability is limited by the high-order dispersion-induced correlation. To minimize the frequency correlation, in addition to the satisfaction of $\tau_i = 0$ ($\tau_s = 0$), the curvature of the contour of phase matching function, determined by the first- and second-order dispersion coefficients, should be as small as possible. In this case, $g^{(2)} \rightarrow 2$ is realizable by optimizing the bandwidth of the transform-limited pump and by increasing the length of PCF. However, at present, the achievable maximum length $L$ is restricted by the inhomogeneity of the PCF [25].

Our theoretical analysis is verified by two series of experiments realized by regulating the spectrum of the pump to respectively satisfy the AGVM and SGVM conditions in 0.6 m PCF. The experimental results quantitatively agree with the theoretical predictions, and the deviation is due to the inhomogeneity of PCF. Moreover, our experimental results show that it is challenging to generate frequency de-correlated photon pairs with a high rate due to spectrum broadening and chirp induced by self-phase modulation of the pulsed pump, particularly for the SGVM condition. We believe our study can be extended to other pulse-pumped SPE processes; therefore, it is not only helpful in generating spectral factorable photon pairs, but also useful in generating pulsed squeezed light [2, 24, 35].
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