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Abstract

High-dimensional, frequency-entangled photonic quantum bits (qudits for $d$-dimension) are promising resources for quantum information processing in an optical fiber network and can also be used to improve channel capacity and security for quantum communication. However, up to now, it is still challenging to prepare high-dimensional frequency-entangled qudits in experiments, due to technical limitations. Here we propose and experimentally implement a novel method for a simple generation of frequency-entangled qudits with $d > 10$ without the use of any spectral filters or cavities. The generated state is distributed over 15 km in total length. This scheme combines the technique of spectral engineering of biphotons generated by spontaneous parametric down-conversion and the technique of spectrally resolved Hong-Ou-Mandel interference. Our frequency-entangled qudits will enable quantum cryptographic experiments with enhanced performances. This distribution of distinct entangled frequency modes may also be useful for improved metrology, quantum remote synchronization, as well as for fundamental test of stronger violation of local realism.

Introduction

The study of higher dimensional entangled states has opened a wide range of possibilities in quantum information science and technology. For example, $d$-dimensional quantum bits (qudits) have, in principle, a stronger violation of the local hidden variable hypothesis than a qubits system [1] and are even useful to observe the violation of its more general context including non-contextuality [2]. Entangled qudits are also advantageous in quantum key distribution (QKD) [3–7] and other applications such as quantum bit commitment [8] or spatial imaging [9].

Several experiments have been dedicated to preparing photonic qudit states with photons encoded in different degrees of freedom. For example, qudits have been generated from biphotons in polarization [10], frequency [11], time-bin [12, 13] and spatial modes [14, 15]. Among all these degrees of freedom, frequency may be of great interest in distributing entangled photons in multiplexed modes over long-distance optical fibers for quantum communication. Traditionally, the frequency-entangled qudits can be generated by manipulating the spectra of biphotons through use of narrowband filters and electro-optic modulators [16] or through use of a spatial light modulator (SLM) [11]. Recently, Xie et al prepared a high-dimensional frequency-entangled qudits using external cavity for spectral filtering [17]. However, the separation and measurement of these high-dimensional, frequency-entangled state is limited by optical loss and resolution of the devices.
In this work, we propose a novel method for a simple generation of frequency-entangled qudits with $d > 10$ without using any spectral filters or cavities. We implement it and demonstrate the desired frequency correlation and a partial evidence of entanglement. In addition, the generated state is distributed over 15 km distance. Our scheme is based on two key techniques. The first technique is the quantum state engineering of twin photons from type II spontaneous parametric down-conversion (SPDC). We employ a periodically poled stoichiometric LiTaO$_3$ (PPSLT) crystal [18, 19], which satisfies the group velocity matching (GVM) condition of $V^{-1}_s=V^{-1}_i$ at telecom wavelength, where $V^{-1}_s$, $V^{-1}_i$ is the inverse of the group velocity for the signal (idler) photons from SPDC [20]. This GVM condition allows us to generate broadband twin photons with strong spectral anti-correlation (see Methods). The second technique is the spectrally resolved Hong-Ou-Mandel (HOM) interference [21], where the spectral modes of the twin photons can be separated at different delay positions in the interference [22, 23]. By combining these two techniques, a multi-peak (comb-like) spectral correlation structure can be formed in a 2D diagram of the twin photon frequencies, which is observed by coincidence counts at the two distant nodes after optical fiber transmission.

**Schematic**

The schematic of our experiment is shown in figure 1. It is a simple HOM interference setting with an additional time delay $\tau$ in the idler mode. The quantum state of the photon pair emitted from a SPDC source is [24, 25]

$$|\Psi\rangle = \int_{0}^{\infty} \int_{0}^{\infty} d\omega_1 d\omega_2 f(\omega_1, \omega_2) \hat{a}_s^\dagger(\omega_1) \hat{a}_i^\dagger(\omega_2) |0\rangle,$$

where $\hat{a}_s^\dagger(\omega)$ is the creation operator at angular frequency $\omega$, the subscripts $s$ and $i$ denote the signal and idler photons, respectively, and $f(\omega_1, \omega_2)$ is their joint spectral amplitude (JSA).

After applying the delay and the HOM beamsplitter and postselecting the state such that each output channel (Ch1 and Ch2 in figure 1) contains one photon, and taking into account the structure of $f(\omega_1, \omega_2)$ for our crystal and pumping, the state is approximately described as

$$|\phi(\tau)\rangle \approx \frac{1}{\sqrt{N}} \int_{0}^{\infty} \int_{0}^{\infty} d\omega_1 d\omega_2 h(\omega_1 + \omega_2 - \omega_p)$$

$$\times \delta(\omega_1 + \omega_2 - \omega_p)(1 - e^{-i(\omega_1 - \omega_2)\tau})$$

$$\times \hat{a}_s^\dagger(\omega_1) \hat{a}_i^\dagger(\omega_2) |0\rangle,$$

where $N$ is a normalization factor, $\omega_p$ is the pump frequency, the subscripts 1 and 2 denote modes in Ch1 and Ch2, $\delta(x)$ is the Dirac delta function, and $h(x)$ is a function determined by the phase-matching condition and the pump envelope. In equation (2), one can observe that the amplitude of the photon pair wave function distributes only on $\omega_1 + \omega_2 = \omega_p$ and oscillates with peaks at $\omega_1 - \omega_2 = 2\pi/\tau$, that is, having an entangled structure can be formed in a 2D diagram of the twin photon frequencies, which is observed by coincidence counts at the two distant nodes after optical fiber transmission.
qudit structure $\sim \sum_{j} \epsilon|\omega_j^1 \omega_j^2|$ where $\omega_j^1 = \omega_j^2/2 \pm (2j - 1)\pi/\tau$. See Supplementary Information 1 available at stacks.iop.org/qst/1/015004/mmedia for a more detailed discussion.

Experiment

To generate such a state, one needs (1) high indistinguishability between the signal and idler photons from the SPDC source; and (2) a carefully engineered JSA showing discrete peaks in frequency correlation. These properties are experimentally characterized as follows.

First, the indistinguishability was observed by measuring the coincidence probability $P(\tau)$ in the HOM interference (figure 1, inset) as a function of the delay time $\tau$ [26], where

$$P(\tau) = \frac{1}{4} \int_{0}^{\infty} \int_{0}^{\infty} d\omega_1 d\omega_2 I(\omega_1, \omega_2, \tau),$$

where

$$I(\omega_1, \omega_2, \tau) = |f(\omega_1, \omega_2) - f(\omega_2, \omega_1) e^{-i(\omega_1 - \omega_2)\tau}|^2,$$

is the correlated spectral intensity (CSI) (see Supplementary Information 2 available at stacks.iop.org/qst/1/015004/mmedia for the detailed calculation). The measured HOM dip is shown in figure 2. The net visibility is evaluated as 90.2 ± 0.2%, indicating high indistinguishability between the signal and idler photons from our SPDC source. The FWHM of the dip is 52.9 $\mu$m (176.3 fs). The raw visibility without subtracting the background counts is 68.6 ± 0.4%. The background counts are mainly caused by the double phase-matching condition in PPSLT crystal [19].

Next, we characterize the comb-like structure of frequency-entangled qudits using a one-dimensional (1D) time of arrival (ToA) measurement (see Methods) and two-dimensional (2D) CSI measurement. In both measurements, we employ a fiber-based spectrometer [27, 28] (see Methods), which is constituted of two 7.5-km-long single-mode fibers (SMFs), two superconducting nanowire single photon detectors (SNSPDs) and a time interval analyzer (TIA) as shown in figure 1. The wavelength dispersion effect in optical fibers is used to convert spectral information to arrival time information. We demonstrated clear comb-like structures, not only in the 2D ($\omega_1$, $\omega_2$) diagram, but also in the 1D ToA data $H(\Delta\omega, \tau)$, after integrating the frequency of Ch1 ($\omega_1$):

$$H(\Delta\omega, \tau) = \int_{0}^{\infty} d\omega_1 I(\omega_1, \omega_1 + \Delta\omega, \tau),$$

where $\Delta\omega = \omega_2 - \omega_1$. In the ToA measurement, the TIA provides the information of the frequency difference $\Delta\omega = \omega_2 - \omega_1$. Figure 3 shows these comb-like structures, corresponding to frequency-entangled qudits. Graphs in the first row (a.1-f.1) show the experimental results of the ToA data $H(\Delta\omega, \tau)$. The horizontal axis is the arrival time difference $t_2 - t_1$, corresponding to frequency difference $\Delta\omega = \omega_2 - \omega_1$. The ToA data were accumulated for 30 seconds with Ch1 as the stop channel and Ch2 as the start channel for the TIA, at a fixed delay time $\tau$. At the dip center, i.e. zero delay position, there is no comb-like structure, as shown in figure 3(a.1). As the delay position is moved from zero to 40 $\mu$m, 160 $\mu$m, 320 $\mu$m and 600 $\mu$m, the peak numbers increased to

Figure 2. Hong-Ou-Mandel dip. Coincidence counts in one second as a function of the optical path delay. The error bars were evaluated by assuming Poissonian statistics of these coincidence counts.
2, 4, 8 and 14, as shown in figure 3(b.1-e.1). When the delay position is far away from the dip center, the comb-like structure disappears gradually. Figure 3(f.1) shows such a case at the delay position of 2.1 mm.

Graphs in the second row (a.2-f.2) show experimental CSI $I(\omega_1, \omega_2, \tau)$ of the twin photons. The CSI data are measured with three input signals to the TIA: the trigger timing information of the pump laser pulses for synchronization, the channel and Ch2 as the second stop channel. The CSI is reconstructed by analyzing the arrival time of the photon pairs in Ch1 and Ch2. Each CSI data set was accumulated only for 5 seconds, thanks to our highly efficient fiber spectrometer that is constituted of high-efficiency SNSPDs (see Methods) [29–31]. The peak numbers in the ToA data in the first row of figure 3 correspond well to the discrete mode numbers in the diagram $\omega_1, \omega_2$ of the CSI in the second row of figure 3. The mode numbers in figures 3(b.2-e.2) are 2, 4, 8 and 14, respectively. In figure 3(f.2), the correlation diagram does not show any mode structure due to our limited timing resolution. In the CSI data, the comb position is determined by $\cos(\omega_1 - \omega_2) r_1 = -1$ in equation (4). The peak-to-peak distance is equal to $\omega_1 - \omega_2 = 2\pi r / r$. It can be noticed that the full width of the ToA data (≈8 ns in figure 3(f.1)) is about two times the full width of the CSI data (≈4 ns in figure 3(f.2)). This difference is caused by the fact that $\omega_1(\omega_2)$ has an arrival time range of around $[-2 \text{ ns}, 2 \text{ ns}]$, while the difference of $\omega_1$ and $\omega_2$, i.e. $\omega_1 - \omega_2$, has an arrival time range of around $[-4 \text{ ns}, 4 \text{ ns}]$.

We also performed numerical simulation of the CSI using the experimental conditions (40-mm-long PPSLT crystal; 2-ps-long pump pulse with Gaussian shape; filtering by a LPF with experimentally measured transmission spectrum) as shown in figures 3(a.3-f.3). The experimentally measured frequency mode numbers coincide well with our simulation. The experimental sizes of the comb teeth in figures 3(a.2-f.2) are slightly ‘fatter’ than the theoretical sizes in figures 3(a.3-f.3), due to the limited timing resolution of the detection system. By integrating the data in figures 3(a.3-f.3) over Ch2 wavelength, we obtain the marginal spectra of figures 3(a.3-f.3) over Ch1 wavelength, as shown in figures 3(a.4-f.4). The peak numbers in figures 3(a.3-f.3) match well to the peak numbers in figures 3(a.1-f.1). The profile of figure 3(f.4) also agrees well with that of figure 3(f.1).

\[ H(\Delta \omega, \tau) = \begin{cases} 0 & \text{if } \Delta \omega = 0 \text{ and } \tau = 0 \\ 1 & \text{otherwise} \end{cases} \]
We find that the comb-like structure only exists in the coincidence counts between Ch1 and Ch2, and does not exist in single counts of Ch1 or Ch2 (counts with only one detector at Ch1 or Ch2). To further investigate the relationship between the single counts and coincidence counts, we measured the arrival time of single counts of Ch1 (Ch2) heralded by the trigger from the pump laser. We also measured the arrival time of coincidence counts (i.e., the arrival time of Ch1 heralded by Ch2). The results are shown in figures 4(a)(b), with each data set accumulated for one section. It is clear that the comb-like structure does not exist in the single counts. This phenomenon can be explained as follows: equation (4) suggests that HOM interference phenomena are difference-frequency oscillations between the two photons. When the constituent photons are detected as a single photon at both output ports of the BS in the inset of figure 1 (11 state detection), we observe the anti-phase oscillation structures determined by equation (4). Assume $f(\omega_1, \omega_2) = f(\omega_2, \omega_1) = 1$ for simplicity, equation (4) can be simplified as $1 - \cos[(\omega_1 - \omega_2)\tau]$, which is the anti-phase oscillation. In contrast, a comb-like structure with in-phase oscillations $1 + \cos[(\omega_1 - \omega_2)\tau]$ would appear when two photons are detected at either output ports of the BS (02 or 20 state detection). The interference in single counts is smeared out because the 11 state cannot be distinguished from the 02 or 20 states by our detectors. Therefore, no comb-like structure is observed in the arrival time of the single counts SC1(SC2) in figure 4.

**Discussion**

To intuitively understand the phenomenon of comb-like structure of frequency-entangled qudits in this work, we compare the HOM interference with the classical Young’s double slit interference, as shown in figures 5(a)(b). Equation (4) can be rewritten as

$$I(\omega_1 - \omega_2, \tau) = I_1 + I_2 - 2\sqrt{I_1 I_2} \cos(\omega_1 - \omega_2)\tau,$$

Figure 4. Arrival time of single counts and coincidence counts. Arrival time of the single counts SC1(SC2): single counts of Ch1 (Ch2) heralded by the trigger, and coincidence counts (CC) between Ch1 and Ch2, at the position of 200 μm (a) and 600 μm (b). Note that as the synchronization triggers for single counts and coincidence counts are different, so the horizontal axes for SC and CC are not comparable here.

Figure 5. Analogy between the HOM interference (a) and the Young’s double slit interference (b). The phenomenon of the comb-like structure in the HOM interference (a) can be intuitively compared with the Young’s double slit interference (b).
where $I_k = |f(\omega_1, \omega_2)|^2$ and $I_2 = |f(\omega_2, \omega_1)|^2$. In the classical Young’s double slit interference, as shown in figure 5(b), the fringes can be described by

$$\tilde{I}(n, r_2, k) = \tilde{I}_1 + \tilde{I}_2 - 2\sqrt{\tilde{I}_1 \tilde{I}_2} \cos(n - r_2)k,$$

where $\tilde{I}$ is the overall intensity, $\tilde{I}_1$ and $\tilde{I}_2$ are the intensity of light from the double slit, and $k$ is the wave number of the light. Equation (6) and equation (7) have the same mathematical structure. The physics of HOM interference and double slit interference are different: the former is a two-photon effect which reflects the indistinguishability of photons, while the latter is the light wave interference in the classical sense and also the interference of a single photon itself in the quantum domain. However, the manipulation of comb teeth structure in the interference pattern can be done in the similar way in which one can find clear correspondence between the parameters in the two schemes; by shifting the frequency difference $\omega_1 - \omega_2$ (optical path difference of $n - r_2$), we can adjust the dark or bright points of the interference pattern; by changing the delay time $\tau$ (wave vector $k$), we can vary the teeth numbers in the comb-like structure. Thus, the mechanism of our comb-like structure can be intuitively explained along with the simple and fundamental phenomena in optics.

It is interesting to compare this work with the previous work on biphoton frequency comb [17], which was created by adding an external cavity on a SPDC source from a 10-mm-long PPKTP waveguide pumped by a cw laser at 658 nm. In this case, the external cavity functions as a filter and cuts the biphoton spectral distribution into discrete frequency modes. In our case, the HOM interference functions as a filter and cuts the biphoton into a frequency comb. Without the using of external cavity, our method is simpler.

The joint spectra in figures 3(b, 2-e, 2) show the strong anti-correlation in frequency of the photon pairs. This anti-correlation suggests that the generated state is either frequency-entangled pure state

$$\langle \psi \rangle = \sum_{i} a_i |\omega_i \omega_{n-i}\rangle = a_1 |\omega_1 \omega_0\rangle + a_2 |\omega_2 \omega_{n-1}\rangle + \ldots + a_n |\omega_n \omega_1\rangle$$

or frequency-correlated classical mixed state, or some quantum–classical mixed state. One extreme example is

$$\rho = \sum_{i} |a_i|^2 |\omega_i \omega_{n-i}\rangle \langle \omega_i \omega_{n-i}|,$$

where $a_i$ is a probability amplitude and $|\omega_i \omega_j\rangle$ represents a two-photon state where the signal and idler photons have frequency $\omega_i$ and $\omega_j$, respectively. However, the possibility of this example can be excluded by the HOM result in figure 2. Theoretically, equation (8) is straightforwardly implied when we apply a delay line and a beamsplitter operations onto the state in equation (1). To have the statistically mixed state (equation (9)) at the output of the beamsplitter, on the other hand, the state before the beamsplitter should also be in a statistical mixture of $|\omega_i \omega_j\rangle$ $|\omega_i \omega_j\rangle$ rather than the superposition in equation (1). As shown in detail in the Supplementary Information available at stacks.iop.org/qst/1/015004/mmedia, such a mixed state in equation (9) does not show any HOM interference, i.e., the HOM visibility becomes 0. This fact should be sharply contrasted to that of the state in equation (8), which, in principle, shows 100% of the HOM interference visibility. The statistically mixed state in equation (9) is only a typical example. Some other mixed states may still explain the results of figure 3. However, this possibility is unlikely because the extreme example in equation (9) shows that the statistically mixed state components always act to suppress the visibility of the HOM interference. Therefore, our experimental results shown in figure 2 and figure 3 strongly support that the state in our experiment is a frequency-entangled pure state, as described by equation (8).

Now we discuss the strong and weak points of our method for the generation of high-dimensional frequency qudits. One strong point of our method is that we used the HOM interferometer as an ‘interferometric frequency filter’, which is simpler than the previous methods using filtering devices [11, 16, 17]. The weak point of our method is that the coefficients of the each frequency bins in the qudit state cannot be easily adjusted, but this point may be overcome in the future by inserting some simple modulating devices.

**Conclusion**

We proposed and demonstrated the generation of frequency-entangled qudits in the spectrally resolved HOM interference using the downconverted photons from a PPSLT crystal. The observed maximal spectral mode number is 14 (figure 3) after transmitting 15 km. It is worth noting that the number can be improved simply by removing the LPFs in figure 1. By doing this, we are able to observe more than 20 spectral modes where the FWHM of the downconverted photons is measured to be $\pm 35$ nm and the net visibility of the HOM dip is around 75%.

There are several interesting future issues. First, the dimensionality of entangled qudits could be further improved by using more broadband twin photon sources [32]. Another important challenge is more rigorous characterization of entanglement for the generated frequency-entangled qudit states [33–35]. To rigorously ‘prove’ entanglement without modeling (assuming) physical system of the setup, one has to experimentally...
access some entanglement measure or separability criteria which requires state tomography or similar kind of detection. Such experimental techniques have been well established for standard entangled qubit photons, such as polarization entanglement [36]. However, its application to our high-dimensional frequency-entangled qudits is not straightforward as it is a higher dimensional system and, more importantly, the measurement basis should include the superposition of many different frequency modes which is technically nontrivial to implement. Recent progress on frequency mixed detection [11] might be a promising way if it is extendable to higher dimensional systems including more than 10 frequency modes.

Also, our techniques developed here have various potential applications. For example, the spectrally resolved HOM interference technique used in this experiment would be useful to extend the dynamic range of the phase shift measurement and timing synchronization because this technique allows one to see the interference patterns for a longer delay position or delay time than that in the standard HOM interference. Actually, even when the twin photons have no interference pattern in the region with the delay position larger than 160 μm in figure 2 for the standard time-domain HOM interference, we can still see the interference pattern in frequency domain, as shown in figures 3(c.1–e.1). This property may be used for quantum remote synchronization [37–39]. The comb-like structure may also be useful in metrology [40, 41].

Finally, our frequency-entangled source is clearly applicable to the recent proposals of the high-dimensional QKD with the time-frequency-entangled states [4, 5]. The time-frequency encoding provides numerous advantages over other coding methods in the sense of dense multiplexing transmission in long haul, single-mode fibers.

**Methods**

**Quantum state engineering in a PPSLT crystal**

In this experiment, the SPDC source is based on a periodically poled MgO-doped stoichiometric LiTaO$_3$ (PPSLT) crystal [18–20, 42], which has a low birefringence. At 1584 nm, the type II PPSLT crystal satisfies the group velocity matching (GVM) condition of $V_{s}^{-1} = V_{i}^{-1}$, where $V_{s(i)}^{-1}$ is the inverse of the group velocity for the signal (idler) [20]. Under this GVM condition, the phase-matching function $\phi(\omega_s, \omega_i)$ is distributed along the anti-diagonal direction, the same as the pump envelop function $\alpha(\omega_p + \omega_i)$. Therefore, the joint spectral distribution $f(\omega_s, \omega_i) = \phi(\omega_s, \omega_i)\alpha(\omega_p + \omega_i)$ has a broad wavelength distribution (more than 30 nm) for the signal and idler photons with spectral anti-correlation, similar as the data shown in figure 3 (f.3).

**The fiber spectrometer**

The two 7.5-km-long SMFs, the two SNSPDs and the TIA constitute a fiber spectrometer [27, 28]. By using such a fiber spectrometer, the spectral information of the downconverted photons is transferred to the ToA information. In this experiment, the single photons with a FWHM of 22 nm is transferred to a FWHM of 4.5 ns by 7.5-km-long SMFs. The dispersion of the SMFs is calibrated as 27.3 ps/km/nm at 1584 nm. Given an estimated 100 ps FWHM jitter of the detection system, the resolution of this fiber spectrometer is calculated as 0.5 nm. The measured insertion losses of these two 7.5-km-long SMFs (including the loss of fiber connectors) are 2.1 dB for one channel (Ch1), 1.9 dB for the other channel (Ch2).

**The ToA measurement**

In the ToA measurement with Ch1 as the start and Ch2 as the stop channel, the TIA records the arrival time of the photons in Ch1 ($t_i$) and Ch2 ($t_f$), and outputs a distribution of the photon events as a function of the arrival time difference ($t_i - t_f$). After transmission through dispersion fibers, the spectral information ($\omega_1, \omega_2$) is converted to arrival time information ($\omega_1(t_1), \omega_2(t_2)$). Therefore, $I(\omega_1, \omega_2, \tau)$ and $H(\Delta \omega, \tau)$ can be converted from $I(\omega_1(t_1), \omega_2(t_2), \tau)$ and $H(\Delta \omega(\Delta t), \tau)$.

**The SNSPDs**

Our superconducting nanowire single photon detectors (SNSPDs) are fabricated with 5–9 nm thick and 80–100 nm wide niobium nitride (NbN) or niobium titanium nitride (NbTiN) meander nanowires on thermally oxidized silicon substrates [29, 30]. The nanowire covers an area of 15 μm x 15 μm. The SNSPDs are installed in a Gifford–McMahon cryocooler system and are cooled to 2.1 Kelvin. The SNSPDs have a system detection efficiency (SDE) of around 70% with a dark count rate (DCR) less than 1 kcps. The SNSPD also has a wide spectral response range [31]. The measured timing jitter and dead time (recovery time) were 68 ps [29] and 40 ns [43].
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Competing interests

The authors declare that they have no competing financial interests.

Author Contributions

R J, R S and M S conceived the idea. R J performed the experiment. R J, R S, M T, T G and M S analyzed the data. R J, M F, R W and T G developed the fiber spectrometer. M F, T Y, S M, H T and M S developed the SNSPDs. R J, M T and M S wrote the manuscript. M S supervised the whole project. All of the authors contributed to revision of the manuscript.

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