Coherent manipulation of a three-dimensional maximally entangled state

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Maximally entangled photon pairs with a spatial degree of freedom is a potential way for realizing high-capacity quantum computing and communication. However, methods to generate such entangled states with high quality, high brightness, and good controllability are needed. Here, a scheme is experimentally demonstrated that generates spatially maximally entangled photon pairs with an orbital angular momentum degree of freedom via spontaneous parametric down-conversion in a nonlinear crystal. Compared with existing methods using post-selection, the technique can directly modulate the spatial spectrum of down-converted photon pairs by engineering the input pump beam. In addition, the relative phase between spatially entangled photon pairs can be easily manipulated by preparing the relative phase of input pump states.

Introduction - Engineering the entangled state is an important direction in quantum technology and forms a basis for quantum information processing. In particular, both the realization of a high-dimensional, maximally entangled state (MES) and its coherent manipulation are indispensable for investigating fundamental quantum physics, such as non-locality1,2. A MES have been fundamental resource for quantum communication protocols such as teleportation3, storage4,5, and secure cryptography6,7. Furthermore, applications to multimode quantum computation, such as high-dimensional quantum gates and Bell basis8,9 will be important.

In photonic systems, a high-dimensional MES can be constructed by entangling two photons in a spatial degree of freedom10,16, or by photon number17, path18,19, frequency20, or temporal modes21–23. High-dimensional MES encoded in orbital angular momentum (OAM) via spontaneous parameter down-conversion (SPDC) has been a well-known and effective method11,24, and much progress has been made2,12–14. Specifically, a three-dimensional MES of the form $|\varphi\rangle = 1/\sqrt{3}(|00\rangle + |11\rangle + |2, 2\rangle)$ was prepared in Ref.13. Recently, the non-locality of a higher-dimensional MES ($d = 12$) was studied in Ref.2 using post-selection of down-converted photons. In addition, a high-dimensional multi-photon entanglement was obtained with two nonlinear crystals14. However, most of OAM-based MES are generated via post-selections of the quantum state ($|\varphi\rangle = \sum_{L=-\infty}^{\infty} c_l |L\rangle_A |l\rangle_B$) produced by using SPDC with a Gaussian pump ($|L\rangle_p = |0\rangle$). An example was reported in Ref.24, which used Procrustean filtering. In fact, the state generated by post-filtering is not truly a maximally entangled state. This type of scheme has several disadvantages. On the one hand, because the amplitude $c_{|l|>0}$, determined by the spiral bandwidth (azimuthal Schmidt number)26, is always less than $c_{|l|=0}$, many useful photon pairs with lower-order modes (i.e. $|00\rangle$) are lost in the process of filtering. This is a more serious problem when constructing a higher-dimensional MES. On the other hand, it is difficult to change the relative phase between different modes, while the phase between them is fundamental and important to increase the freedom of modulations for quantum information processes8. This is especially true for constructing high-dimensional Bell basis9, and quantum dense coding27. Furthermore, to experimentally adjust the spiral bandwidth $c_l$, some parameters must be changed, such as input beam waists or the length of the nonlinear crystal28, which is unwieldy in experiment. Fortunately, utilization of a superposition state as the pump can overcome these shortcomings, and there are several theoretical protocols for engineering a high-dimensional MES28,29. Experimentally, a spatial Bell state with a transverse Hermite-Gaussian mode have been directly generated without spatial filtration via SPDC10. Since the OAM degree of freedom of a photon is easier to manipulate and measure20, it would be very interesting to generate an OAM-based MES without post-selections.

Here, we prepared a two-photon, three-dimensional MES in the Hilbert space formed by OAM degree of freedom of the photon produced via SPDC. The approach was to engineer the pump beam with an arbitrary superposition of Laguerre-Gauss (LG) modes. The state of down-converted photon pairs could then be manipulated independently based on OAM conservations in SPDC. To check the non-classical characteristics of the prepared three-dimensional MES, the Bell-type inequality ($S = 2.3729 \pm 0.0159$) and high-dimensional quantum state tomography ($F = 0.8581 \pm 0.0028$) were performed. In addition, the spatial spectrum of the down-
FIG. 1. Theoretical results of spatial spectrum distributions of down-converted photons and the required intensity profiles of the input beams. (a)-(d): The spatial spectrum distributions for the input pump $|−2\rangle_p + |0\rangle_p + |2\rangle_p$, $\sqrt{1.5} |−2\rangle_p + |0\rangle_p + \sqrt{1.5} |2\rangle_p$, $2.5 |−2\rangle_p + |0\rangle_p + \sqrt{2.5} |2\rangle_p$, $|−2\rangle_p + |2\rangle_p$, respectively, where the beam widths of the pump and detection are $w_p = 1.0, \gamma_s = \gamma_i = 0.5$\[29\]. Actually, the coefficients of input state for MES need to change based on the different system of collections. (e): 2D perspective of Fig. 1(a), where the $x(y)$-label represent the modes of signal and idler photons. (f)-(h): corresponding required intensity-profiles, where the wavelength of the pump is 780 nm.

converted photons was calculated. Comparing with the exist schemes, there were several advantages for these protocols. i) It did not require post selections; ii) the arbitrary phase between different superposition terms could be easily engineered by preparing the input beams. This work provides a convenient and efficient platform to explore potential applications in quantum communications.

Principle. Down-converted photon pairs have the form $|\varphi\rangle_{\text{single}} = \sum_{l=-\infty}^{\infty} c_l |l\rangle_A |l-l\rangle_B$ for a single pure-state pump $|L\rangle_p$; by analogy, it can be written as $|\varphi\rangle_{\text{multi}} = \sum_L \sum_{l=-\infty}^{\infty} c_L c_l |l\rangle_A |L-l\rangle_B$ when the pump is a superposition of multiple LG modes $\sum_L c_L |L\rangle_p$. $C_L$ is a complex number, i.e., $C_L = |C_L|e^{i\theta_L}$. Considering a suitable pump state of $C_{−2} |−2\rangle_p + C_0 |0\rangle_p + C_2 |2\rangle_p$, the down-converted entanglement state can be generated as following based on OAM conservation($l_p = l_s + l_i$): $|\varphi\rangle_{\text{MES}} = \xi_{\text{MES}} (-1 - 1) + 00 + 11 + \sum_L \sum_{l=-\infty}^{\infty} c_l^L \xi_{l} |l\rangle_A |L-l\rangle_B$, where the $\xi_{l}$ are the amplitudes of high order OAM entangled pairs, which can be ignored by changing the pump beam and signal/idler beam widths \[26\][29\]. Then the state is reduced to a three-dimensional, MES in the subspace.

Figure 1 depicts the main principle of the scheme. The effective spatial spectrum is distributed along the corresponding diagonal elements based on the OAM conserved. For example, the green rectangular and red boxes in Fig. 1(c) are the situations for the pump with $|L\rangle_p = |0\rangle_p + |2\rangle_p + |−2\rangle_p$, respectively. If the pump is a superposition state, i.e., $|L\rangle_p = 1/\sqrt{3} (|0\rangle_p + |2\rangle_p + |−2\rangle_p)$, the output spatial spectrum distribution equals the effects of a linear superposition for each pure-state pump \[29\], and the result is presented in Fig. 1(a). By increasing the occupations of pump $|±2\rangle$, the two smaller heights of $|1\rangle_s |1\rangle_i$ and $|−1\rangle_s |−1\rangle_i$ can be grown from Fig. 1(a) to Fig. 1(b). Thus, an arbitrary three-dimensional entangled state can be generated. For an extreme situation with only two pump vortex pumps $|±2\rangle_p$, the output is a two-dimensional, maximally entanglement state (see Fig. 1(d)), which has been widely used in quantum information \[31\]. Fig. 1(c) shows the MES of $|\varphi\rangle_{\text{MES}} = 1/\sqrt{3} (-1)_{s} |−1\rangle_i + |0\rangle_s |0\rangle_i + |1\rangle_s |1\rangle_i$, where the high-order component can be reduced by modulating the beam waist of the pump and optimizing overlaps between distributions of photon pairs and phase holograms in a spatial light modulator (SLM) \[26\][29\]. Figs. 1(f)-(h) indicate the required intensity profiles of the input pump for Figs. 1(b)-(d), respectively.

In addition, the relative phase between the pump superposition state can be fully transferred to the corresponding OAM entangled photon pairs. For example, the output would be $|\varphi\rangle_{\text{MES}} = 1/\sqrt{3} (-1)_{s} |−1\rangle_i + e^{i\theta_0} |0\rangle_s |0\rangle_i + e^{i\theta_2} |1\rangle_s |1\rangle_i$, under the input pump of $N (\sqrt{2.5} |−2\rangle_p + e^{i\theta_0} |0\rangle_p + \sqrt{2.5} e^{i\theta_2} |2\rangle_p)$. Hence, the coherence was preserved during the overall process, which was useful for engineering an arbitrary entangled state.

Fig. 2 shows the optical paths for generating and coherently modulating a three-dimensional MES. The 780-nm input source was a TA: sapphire continuous laser (MBR110, Coherent). A visible-wavelength spatial light modulator (SLM-V, 512 × 512 SLM, Model P512-0785, Meadowlark, Optics) was used to modulate the input laser beams. The laser beam carrying the acquired phase in SLM-V was exactly imaged on the center of nonlinear crystals (PPKTP, 10 mm) by a lens ($f$=100 mm). The beam waist in the center of the crystal was 31 $\mu$m. The collection of down-converted photon pairs was implemented as follows. First, the generated infrared OAM-
entangled 1550-nm photon was long-pass filtered. Then, it passed through an imaging system with an infrared lens (f = 150 mm) and a polarization beam splitter (PBS) before reaching the plane of SLM-I (PLUTO-2-TELCO-013,1920 × 1080 Pixel). Finally, by a group of coupling lenses, the entangled photons in SLM-I without vortex phase information are imaged that on the surfaces of fibers, which is ported to a superconducting nanowire single-photon detector that had four channels for detections. The imprinted phase profiles in both SLMs were given by amplitude-encoded phase-only holograms (see supplementary materials). In this type of encoded hologram, adjustment of the beam waists for different OAM-modes was required for measurements of optimal mode overlaps.

Results. Results for engineering a three-dimensional arbitrary entangled state are presented in Fig. 3. Two manipulation steps were employed. One was to change the real part of the coefficients $|C_j|$ for three inputs, or named as amplitude-modulations. To prove that this phase was fully converted from input states to quantum entanglement states, the measurement basis $|\varphi_A\rangle \otimes |\varphi_B\rangle = N(|-1_A \rangle + e^{i\alpha} |0_A \rangle + e^{i\theta} |1_A \rangle) \otimes (|-1_B \rangle + e^{-i\alpha} |0_B \rangle + e^{-i\theta} |1_B \rangle)$ was designed and loaded on SLM-I. Then, the coincidences $|\langle \varphi_A | \varphi_B \rangle_{MES}|^2$ were derived as $N(3+2(\cos(\alpha+\theta_0)+\cos(2\alpha+\theta_2)+\cos(\alpha+\theta_2-\theta_0)))$. Fig. 3(c) exhibits the three-dimensional (3-D) surface with variations of the two phase angles. Figs. 3(b,d) present two special interference curves for $\theta_2 = 0 - 3\pi, \theta_0 = \pi/4$ and $\theta_0 = 0 - 3\pi, \theta_2 = 0$. For each of the subfigures, the parameters $\alpha$ were $2\pi/3$, $\pi/3$, respectively, and the solid lines were fits based on theoretical predictions. The visibility $(V = C_{Max} - C_{Min}/C_{Max} + C_{Min})$ of the red and blue solid curves were 0.9387 ± 0.0178, 0.9735 ± 0.0133 for Fig. 3(b), and 0.9126 ± 0.0382, 0.9116 ± 0.0347 for Fig. 3(d), respectively. In Fig. 3(d), the initial coincidence was not a minimum value, which illustrated that, for the prepared MES, there existed a basic constant phase factor between OAM entangled photon pairs.

To test the non-classical characteristics of the OAM-based MES, measurement of the high-dimensional Bell
inequalities was performed\cite{1}. Many groups have successfully demonstrated violation of the high-dimensional Bell inequality of OAM-based entangled photon pairs\cite{2,4,13,31}. Here, the measurement basis $|\theta_{A,B}^{a,b}\rangle$ on SLM-I was designed as a three-dimensional superposition state\cite{11,2}. Details are in the Supplementary Material. For a three-dimensional MES, the coincidence formed a 3-D surface (Fig. 4(a)) with the variation of two angles. To test the Bell inequality, 36 measurements were divided into four groups (four colors in Fig. 4(a)) of nine samplings.

For a 10-mm-long type-II Periodically Poled Potassium Titanyl Phosphate (PPKTP), the spatial parameters $\tau_a, \tau_b$ associated with the spacing of the measurement basis $|\theta_{A,B}^{a,b}\rangle$ were $4.2/2\pi$, by fitting the Bell-type interference curves in Figs. 4(b,c). By assuming that the coincidences followed Poisson’s distribution, the Bell inequality $S_3 = 2.3735 \pm 0.0159$ in $3 \times 3$ dimensions, which was more than 24 standard deviations. This violation of a Bell inequality directly indicated that the generated state was entangled.

In the next test, density matrices of three-dimensional MES were constructed via high-dimensional quantum-state tomography via mutually unbiased measurements. The combination of SLM-I, single-mode fibers, and coincidence-counting electronics enabled projective measurements of the three-dimensional MES\cite{32,33}. The corresponding reconstructed density matrices were $\rho = N \sum_{n,\lambda,j,k} (A^{nk}_{\lambda j})^{-1} n_{\lambda j} \lambda_j \otimes \lambda_k$. Details on parameters are described in the Supplementary Material. To ensure that the density matrices were ‘physical’, i.e., had the property of positive semi-definiteness\cite{35}, maximum likelihood estimation methods were performed during the reconstructions. The reconstructed density matrices are shown in Fig. 4 with a 3D perspective, where Figs. 4(d,e) represented the real and imaginary parts, respectively. The fidelity $F = Tr[\sqrt{\sqrt{\rho_{\text{exp}}} \rho_{\text{exp}} \sqrt{\rho}}]^{2}$ was $0.8581 \pm 0.0028$, where 0.0028 was the standard deviation obtained by statistical simulations that assumed that each photonic coincidence followed Poisson’s distribution. There were two relatively high imaginary parts in Fig. 4(e), which was the result of a constant phase between low-(|00\rangle) and high-order (|11\rangle) entanglement states. The constant phase could be concluded by the interference curves in Fig. 3(d). Furthermore, calculations of the linear entropy $S_{out} = 1 - Tr(\rho_{\text{exp}}^{2})$ yielded $S_{out} = 0.2451 \pm 0.0174$. These measurement parameters of MES had a little gap in contrast to the theoretical ideal, which were attributed to imperfect mode overlaps between distributions of photons and the phase hologram in SLM-I, and cross talk between different OAM modes. Nevertheless, the main MES features exhibited here were close to other schemes\cite{13,32}. Hence, the method was reliable and practical.

**Discussion** An effective scheme to engineer a three-dimensional MES with arbitrary relative phase was demonstrated experimentally. The results were in good agreement with theoretical predications. A well-controlled and high-quality source could thus provide a platform for exploring the multi-mode quantum information. Recently, we realized a OAM-based Schrödinger cat state $|\alpha\rangle = \sum_{L=0}^{\infty} c_{L} |L\rangle$ with plenty of OAM modes\cite{30}, which ensures that we can explore the high-dimensional MESs under the arbitrary and complex input OAM superposition state. The scheme to prepare an infrared bi-photon three-dimensional MES had some advantages compared with other techniques. An arbitrary modulation could be made in amplitude and phase, which could be an attractive and potential platform for multi-mode quantum information processing. For this method, the brightness of the high-dimensional MES depended on the pump generation rates of high-order OAM modes. For example, the brightness of $|1\rangle_1 |1\rangle_1$ was $1.0 \times 10^{7} / (s \cdot mW)$ for the pump $|2\rangle_p$. The brightness will naturally decrease with the increase of the topologic charges, which is a bottleneck restricting the increase in dimensions. In addition, it would be an interesting and valuable challenge to decrease the noise component of high-orders, or to decrease the spiral bandwidth (Schmidt number) when the single high-order pure state acts as a pump. One solution would be modifications of the input and collection parameters, but that will lead to decreases in source brightness\cite{29,37}. Another solution would be to increase the spatial Schmidt numbers (spatial purity) by modulating the frames of nonlinear crystals. In the frequency domain, much progress has been made by modulating the poling period or duty cycles\cite{35,40}. However, in the spatial domain, there has been little experimental progress.

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