Preparation and characterization of arbitrary states of four-dimensional qudits based on biphotons

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We report interferometric schemes to prepare arbitrary states of four-dimensional qudits (ququarts) based on biphoton states of ultrafast-pumped frequency-nondegenerate spontaneous parametric down-conversion. Preparation and tomographic characterization of a few examples of general single-ququart states, a pure state, a completely mixed state, and a partially-mixed state, are experimentally demonstrated.

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INTRODUCTION

In quantum information, a two-dimensional quantum system is often used as a carrier of the basic information unit, the two-level quantum state or the quantum bit (qubit). Most quantum computing and quantum communication protocols are based on preparation, manipulation, entanglement, distribution, and measurement of multiple qubits [1].

Recently, D-level quantum states (D > 2) or qudits have attracted a lot of attention in the context of quantum communication and experimental tests of quantum mechanics [2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16]. Experimentally, the physical carrier of the qudit can be any D-dimensional quantum systems. Meaningful applications of qudits in quantum information, however, will only be possible if the qudit is encoded in a D-dimensional physical degree of freedom which is easy to handle experimentally. Furthermore, it should be possible to entangle multiple individual qudits in a scalable manner.

In photonic quantum information research, an internal or an external degree of freedom of a photon is used to encode the intended quantum state. For a qubit, the polarization state of a photon is often the obvious choice although it is possible to choose other degrees of freedom. To encode a qudit, it is necessary to choose a multi-dimensional degree of freedom of a single-photon, such as, the angular momentum, transverse momentum-position, time of arrival, etc [10, 11, 12, 13, 14, 15, 16]. These single-photon multi-dimensional photonic degrees of freedom, however, are experimentally difficult to manipulate and there are no scalable schemes to generate multi-qudit entangled states.

The single qudit, however, does not need to be encoded in a single-particle quantum state. In fact, preparation and tomographic characterization of a pure state qutrit (three-dimensional quantum state), see Refs. [17, 18, 19], and a pure state ququart (four-dimensional quantum state), see Refs. [20, 21], have been demonstrated recently using the biphoton polarization states of frequency-degenerate and frequency-nondegenerate spontaneous parametric down-conversion, respectively. In other words, a pair of photons can be used as a carrier of three- or four-dimensional quantum states.

Especially, the ququart based on the biphoton polarization state of frequency-nondegenerate spontaneous parametric down-conversion (SPDC) exhibits a few properties which are important for applications in quantum information research: First, all the ququart basis states can be accessed using only linear optical elements (phase plates) [20, 21]. Second, it is possible to prepare a multi-ququart entangled state starting from multiple individual biphoton ququarts, linear optical elements (beam splitters), and post-selection measurement [22, 23]. So far, only pure state biphoton ququarts have been demonstrated. It is, therefore, of interest and importance to learn how to prepare a general state of a biphoton ququart [24].

In this paper, we report experimental studies on preparation of general states of biphoton ququarts using ultrafast-pumped frequency-nondegenerate spontaneous parametric down-conversion. Methods for preparation and tomographic characterization of some examples of arbitrary ququart states, i.e., pure, mixed, and partially-mixed states of a biphoton ququart, are experimentally demonstrated. We also discuss a couple of alternative experimental schemes which allow to generate arbitrary biphoton-based ququart states.

BIPHOTON QUQUART

Let us start with a brief introduction to the biphoton ququart. In collinear frequency-nondegenerate SPDC, a higher energy pump photon is occasionally split into a pair of co-propagating lower energy photons (signal-idler)
of different frequencies,

\[
\frac{1}{\lambda_p} = \frac{1}{\lambda_1} + \frac{1}{\lambda_2},
\]

where \(\lambda_p\) is the pump wavelength and \(\lambda_1, \lambda_2\) are the wavelengths of the signal (idler) photon. Since each photon of the pair can be horizontally or vertically polarized, the following set of biphoton polarization basis states can be defined\(^{19, 20, 21, 22}\),

\[
|H_{\lambda_1}, H_{\lambda_2}\rangle \equiv |0\rangle, \quad |H_{\lambda_1}, V_{\lambda_2}\rangle \equiv |1\rangle,
\]

\[
|V_{\lambda_1}, H_{\lambda_2}\rangle \equiv |2\rangle, \quad |V_{\lambda_1}, V_{\lambda_2}\rangle \equiv |3\rangle.
\]

The polarization state of the photon pair born in the process of collinear frequency-nondegenerate SPDC, therefore, represents a four-dimensional quantum state or a ququart. Since the states shown in eq. (2) are orthonormal to each other and form a complete basis for a four-dimensional Hilbert space, they form the computational basis for the biphoton ququart.

Experimentally, the computational basis states for the biphoton ququart can be generated using type-I SPDC (|0\rangle and |3\rangle) and type-II SPDC (|1\rangle and |2\rangle). To prepare an arbitrary superposition state of a ququart,

\[
|\psi\rangle = c_0|0\rangle + c_1|1\rangle + c_2|2\rangle + c_3|3\rangle,
\]

where \(c_l = |c_l|e^{i\theta_l}\) are the complex probability amplitudes which satisfy \(\sum_{l=0}^{3} |c_l|^2 = 1\), one would need to coherently combine two type-I SPDC and two type-II SPDC sources and to control four complex amplitudes \(c_1, c_2, c_3, \text{ and } c_4\). The use of four SPDC sources, however, turns out to be unnecessary since the degree of polarization of the biphoton state of collinear frequency-nondegenerate SPDC is not invariant under SU(2) transformations\(^{23}\). It is, therefore, possible to use linear optical elements (wave plates) to transform one biphoton ququart basis state into any other one as recently demonstrated in Ref.\(^{20, 21}\). As a result, it is possible to prepare an arbitrary superposition state (i.e., pure state) shown in eq. (2) using less than four SPDC sources and we discuss several such schemes in this paper.

The superposition state shown in eq. (2), however, is not the most general quantum state for a ququart. To properly consider mixedness, which comes from controlled or uncontrolled quantum distinguishability among the basis states shown in eq. (2), the ququart state should be expressed as a \(4 \times 4\) density matrix \(\rho\) to describe general single-ququart states: completely mixed states, partially-mixed states, and the pure state shown in eq. (2).

It is interesting to note that, since the ququart state under consideration is in fact formed with two polarization qubits, it is possible to apply the state classification method based on two-qubit entanglement of formation. This method is based on the quantity \(C\), concurrence, which is a measure of two-qubit entanglement\(^{20}\).

For the single biphoton ququart (i.e., biphoton two-qubit state) in eq. (2), it is easy to show that there is no two-qubit entanglement (\(C = 0\), i.e., the two-qubit state is separable) if the amplitudes satisfy the relation \(c_0c_3 = c_1c_2\). Otherwise, the state is an entangled two-qubit state with concurrence \(C = 2|c_0c_3 - c_1c_2| > 0\). (The two-qubit state is non-maximally entangled if \(0 < C < 1\).)

**QUQUART PREPARATION**

In order to prepare a general single-ququart state, including mixed, partially-mixed, and pure states, it is necessary to introduce quantum distinguishability among the biphoton ququart basis states defined in eq. (2) in a controllable manner. Moreover, to establish a confidence bound and to find a reliable method of preparing an intended ququart state, it is required to compare the experimentally reconstructed and the theoretically expected ququart density matrices. Our experiment, therefore, deals with both the ququart preparation as well as the experimental reconstruction of the ququart density matrices using the quantum state tomography.

The schematic of the experimental setup is shown in Fig. 1. In the single-crystal scheme, a type-I BBO crystal, with its optic axis horizontally oriented, is pumped by a train of ultrafast pulses and this scheme is used for preparing pure state ququarts. For preparing mixed and partially-mixed states, we used the double-crystal scheme in which two orthogonally oriented type-I BBO crystals are placed in tandem\(^{27}\). The pump laser had roughly 100 fs pulse width and centered at 390 nm. The polarization of the pump was controlled by using a half-wave plate (Pump HWP). The BBO crystals used on this experiment are 3 mm thick. The SPDC signal-idler photon pair generated at the crystals propagates collinearly with the pump and has different frequencies: the signal pho-
ton is centered at 823.5 nm ($\lambda_1$) and the idler photon is centered at 740.8 nm ($\lambda_2$).

In the single-crystal scheme, the biphoton polarization state generated at the crystal is $|V_{\lambda_1}, V_{\lambda_2}\rangle$ which corresponds to the ququart basis state $|3\rangle$ in eq. (2). This basis state is then unitarily transformed into a superposition state shown in eq. (2) to prepare an arbitrary pure state by using a zero-order wave plate (WP). This is due to the fact that the signal and the idler photons, even though they have the same polarization, acquire different phase shift due to the large difference in the wavelength. As a result, the polarization states of the signal and the idler photons evolve differently, but predictably, for a specific angle of WP: $|V_{\lambda_1}\rangle \rightarrow \alpha|H_{\lambda_1}\rangle + \beta|V_{\lambda_1}\rangle$ and $|V_{\lambda_2}\rangle \rightarrow \gamma|H_{\lambda_2}\rangle + \delta|V_{\lambda_2}\rangle$ with $|\alpha|^2 + |\beta|^2 = 1$ and $|\gamma|^2 + |\delta|^2 = 1$. The final result is the transformation of the initial ququart basis state into a superposition state in eq. (2) which is a pure state. The theoretically expected density matrix $\rho_{\text{theory}}$ for the prepared ququart state, which can be calculated accurately as the settings of WP is in our control, is then compared to the experimentally reconstructed density matrix $\rho_{\text{exp}}$.

The single-crystal scheme, therefore, allows us to prepare one of the four single-ququart basis states in eq. (2) initially. For the case just described, the single-ququart basis state $|3\rangle$ is in fact $|V_{\lambda_1}, V_{\lambda_2}\rangle$, which is a factorizable two-qubit state with $C = 0$. Note now that the degree of entanglement (in this case, between the two polarization qubits) cannot be increased or decreased by local unitary transformations. The unitary transformation due to WP can turn the initial ququart state $|3\rangle$ in to a superposition state in the form of eq. (4). However, the resulting state can always be expressed as a separable two-qubit state of the form $|A_{\lambda_1}, B_{\lambda_2}\rangle$, where $|A_{\lambda_1}\rangle$ and $|B_{\lambda_2}\rangle$ represent arbitrary polarization states of signal and idler photons, respectively.

For preparation of mixed and partially-mixed ququart states, the double-crystal scheme described earlier is used. Since the two BBO crystals are orthogonally oriented, it is possible to excite two of the ququart basis states shown in eq. (2): $|0\rangle$, $|\langle H_{\lambda_1}, H_{\lambda_2}\rangle\rangle$ and $|3\rangle$, $|\langle V_{\lambda_1}, V_{\lambda_2}\rangle\rangle$. The relative amplitudes between the two can be controlled by changing the pump polarization.

To prepare a completely mixed state of $|0\rangle$ and $|3\rangle$, no further actions are required as the two amplitudes are already distinguishable in time due to the clock effect of the pump pulse [27]. The resulting density matrix is, therefore,

$$\rho_{\text{theory}} = \left(1 - \frac{x}{2}\right)|0\rangle\langle 0| + \frac{x}{2}|3\rangle\langle 3|,$$  (3)

where the parameter $x$ can be varied by changing the polarization of the pump. Adding two additional type-II BBO crystals, which are orthogonally oriented, will allow us to easily prepare a mixture of all four ququart basis states. This, however, is not necessary in principle since it is possible to transform a single basis state into a superposition of all basis states and then to introduce birefringent/dichroic decoherence among the amplitudes.

A more general state, between the completely mixed and the pure states, would exhibit some coherence among the four ququart basis states in eq. (2). In other words, the ququart density matrix has non-zero off-diagonal elements. Such states can be prepared by unitarily transforming the mixed state in eq. (3) using WP. As previously discussed, a ququart basis state can be transformed into a superposition of all basis states linear optically using WP [23]. By subjecting the mixed state in eq. (3) to unitary transformation using WP, it is possible to obtain the following partially-mixed state,

$$\rho_{\text{theory}} = p_1|\psi_1\rangle\langle \psi_1| + p_2|\psi_2\rangle\langle \psi_2|,$$  (4)

where $p_1 + p_2 = 1$ and $|\psi_1\rangle$, for example, is in the form of eq. (2). It is important to note that this unitary transformation process, however, does not actually decrease entropy of the ququart state as we shall show in the next section. To decrease the entropy, it is necessary to erase the temporal distinguishability of the biphoton amplitudes born in the first and the second crystals, for example, by inserting a piece of thick compensating quartz crystal in the pump beam or in the path of the photon pair [27]. Introduction of controllable birefringent/dichroic decoherence will transform eq. (4) into a more complex ququart state with increased entropy.

From the two-qubit perspective, linear optical state transformation from eq. (3) to eq. (4) represents no increase in the degree of two-qubit entanglement as both states exhibit the two-qubit concurrence $C = 0$. This is closely related to the fact that the ququart state entropy remains the same for states in eq. (4) and eq. (1). To be more specific, the two-qubit concurrence $C$ will increase if the single-ququart entropy is decreased by erasing the temporal distinguishing information present in eq. (3) or in eq. (4). Complete erasure of the temporal distinguishability between the biphoton amplitudes from the first ($|H_{\lambda_1}, H_{\lambda_2}\rangle$) and the second ($|V_{\lambda_1}, V_{\lambda_2}\rangle$) type-I BBO crystal, see Fig. 4 will result a pure ququart state with the two-qubit concurrence $C = 1$.

In this paper, we have experimentally demonstrated a pure, a mixed, and a partially mixed biphoton ququart states which are shown in eq. (2), eq. (3), and eq. (4), respectively. For the mixed and partially mixed states, the state entropy can be controlled by inserting a proper compensating crystal before or after the BBO crystals.

**QUQUART TOMOGRAPHY**

The prepared ququart state is characterized experimentally by performing quantum state tomography, a statistical method of reconstructing the quantum state
density matrix based on a set of polarization projection measurement [28, 29].

The experimental schematic for the ququart state tomography is shown in Fig. 1. First, the photon pair that forms the biphoton ququart is split into two spatial modes by using a dichroic beam splitter (DBS), which transmits $\lambda_1 = 823.5$ nm and reflects $\lambda_2 = 740.8$ nm. Each photon, then, undergoes polarization state transformation with the use of a quarter wave plate (QWP) after which the photon is detected at the polarization analyzer (POL) for which the photon is detected at the detector package, which consists of a spectral filter and a multi-mode fiber coupled single-photon counting module (SPCM). Since the ququart is made of a pair of photons, the ququart detection is based on the coincidence counting rates of the two SPCM’s that detects $\lambda_1$ and $\lambda_2$ photons with definite polarizations. The coincidence window used in this experiment was 5 ns.

As noted in Ref. [20], the ququart based on the collinear frequency-non-degenerate SPDC photon pair is mathematically equivalent to the non-collinear frequency-degenerate SPDC photon pair which is often used in quantum information research. It is thus possible to apply the two-qubit quantum tomography method described in Ref. [28] directly in this experiment to reconstruct the single-ququart density matrix. Sixteen particular joint biphoton polarization state measurements are, therefore, necessary to reconstruct a single-ququart density matrix for the identically prepared ensemble of biphoton ququarts. Table I shows experimental settings of WP to perform the sixteen polarization projection measurements. The set of sixteen coincidence measurement outcomes $n_\nu$ allow the linear tomographic reconstruction of the single-ququart density matrix. It is, however, possible that the mathematically reconstructed density matrix by the linear tomographic reconstruction might violate the physical properties of a density matrix. To avoid this problem, the maximum likelihood method was applied as follows [28]. First, we generate a physical density matrix which satisfies normalization, Hermiticity, and positivity, as a function of sixteen variables. We then introduce the “likelihood function” which quantifies how good physical density matrix is in relation to the experimental data. Finally, using standard numerical optimization techniques, we obtain the best estimate of the density matrix by maximizing the likelihood function. The single-ququart density matrix initially reconstructed by the linear tomography is used as the seed for the iteration algorithm.

The experimentally reconstructed ququart density matrix $\rho_{\text{exp}}$ is then compared to the theoretically expected density matrix $\rho_{\text{theory}}$ which is calculated from the known values of the pump polarization and WP settings. The fidelity $F = (Tr\sqrt{\rho_{\text{theory}}\rho_{\text{exp}}\sqrt{\rho_{\text{theory}}}})^2$ is then calculated to see how closely the two overlap and the state purity is analyzed by calculating the state entropy defined as $S = -\sum_{k=1}^{4} \lambda_k \log_4 \lambda_k$, where $\lambda_k$ are the eigenvalues of the density matrix $\rho$.

### Table I: QWP and HWP (fast axis) settings for ququart tomography

| $\nu$ | HWP1 | QWP1 | HWP2 | QWP2 | basis1 | basis2 |
|-------|------|------|------|------|--------|--------|
| 1     | 45°  | 0    | 45°  | 0    | $|H\rangle$ | $|H\rangle$ |
| 2     | 45°  | 0    | 0    | 0    | $|H\rangle$ | $|V\rangle$ |
| 3     | 0    | 0    | 0    | 0    | $|V\rangle$ | $|V\rangle$ |
| 4     | 0    | 0    | 45°  | 0    | $|V\rangle$ | $|H\rangle$ |
| 5     | 22.5°| 0    | 45°  | 0    | $|R\rangle$ | $|H\rangle$ |
| 6     | 22.5°| 0    | 0    | 0    | $|R\rangle$ | $|V\rangle$ |
| 7     | 22.5°| 45°  | 0    | 0    | $|D\rangle$ | $|V\rangle$ |
| 8     | 22.5°| 45°  | 45°  | 0    | $|D\rangle$ | $|H\rangle$ |
| 9     | 22.5°| 45°  | 22.5°| 0    | $|D\rangle$ | $|R\rangle$ |
| 10    | 22.5°| 45°  | 22.5°| 45°  | $|D\rangle$ | $|D\rangle$ |
| 11    | 22.5°| 0    | 22.5°| 45°  | $|R\rangle$ | $|D\rangle$ |
| 12    | 45°  | 0    | 22.5°| 45°  | $|H\rangle$ | $|D\rangle$ |
| 13    | 0    | 0    | 22.5°| 45°  | $|V\rangle$ | $|D\rangle$ |
| 14    | 0    | 0    | 22.5°| 90°  | $|V\rangle$ | $|L\rangle$ |
| 15    | 45°  | 0    | 22.5°| 90°  | $|H\rangle$ | $|L\rangle$ |
| 16    | 22.5°| 0    | 22.5°| 90°  | $|R\rangle$ | $|L\rangle$ |

#### Pure state ququart

To prepare a pure state ququart as in eq. (2), we used a single type-I BBO crystal generating the initial ququart state $|3\rangle$, which in fact is a factorizable two-qubit state $|V\lambda_1, V\lambda_2\rangle$. This state is then transformed into a superposition state with the help of WP shown in Fig. 1. In this experiment, a zero-order half wave plate designed at 823.5 nm was used in place of WP. As a demonstration of pure state ququart preparation, we set the fast axis of the half-wave plate at 30° from the vertical axis. Since local unitary transformations do not change the degree of entanglement, the final states belong to the factorizable subset of two-qubit (ququart) states with $C = 0$.

The theoretically expected ququart density matrix in this case is calculated to be,
We obtain \( \rho_{\text{theory}} \) for this state, it is easy to see that \( T_r[\rho^2] = 1 \). The entropy of the theoretical density matrix \( \rho_{\text{theory}} \) is calculated to be \( S = 0 \), as it should for a pure state.

To obtain the ququart density matrix, we have performed the sixteen projection measurement described in Table I. The coincidence data (for the accumulation time of 180 s) are \( n_1 = 6118 \), \( n_2 = 1858 \), \( n_3 = 917 \), \( n_4 = 2943 \), \( n_5 = 1565 \), \( n_6 = 477 \), \( n_7 = 2362 \), \( n_8 = 7549 \), \( n_9 = 2395 \), \( n_{10} = 8254 \), \( n_{11} = 1664 \), \( n_{12} = 6653 \), \( n_{13} = 3078 \), \( n_{14} = 2739 \), \( n_{15} = 5817 \), and \( n_{16} = 1398 \). The accidental coincidences, which appear at the same period as the pump pulse period, have been subtracted. In this paper, all the coincidence measurement data show accidental subtracted values.

Applying the single-ququart tomography algorithm, the experimentally reconstructed ququart density matrix is obtained to be,

\[
\rho_{\text{exp}}^\text{pure} = \begin{pmatrix}
0.5138 & 0.2749 + 0.0523i & 0.3236 + 0.1308i & 0.1643 + 0.1026i \\
0.2749 - 0.0523i & 0.1590 & 0.1887 + 0.0418i & 0.1004 + 0.0379i \\
0.3236 - 0.1308i & 0.1887 - 0.0418i & 0.2463 & 0.1259 + 0.0224i \\
0.1643 - 0.1026i & 0.1004 - 0.0379i & 0.1259 - 0.0224i & 0.0777
\end{pmatrix}
\]

\[ (5) \]

We obtain \( T_r[\rho_{\text{exp}}^2] = 0.962 \) which means that the experimentally reconstructed state is not an entirely pure state. The entropy of the experimentally reconstructed density matrix is found to be \( S = 0.052 \).

Clearly, the experimentally prepared ququart state is somewhat different from what we initially intended to prepare and this is reflected in the state fidelity \( F = 0.938 \). Figure 2 shows the graphical representations of different degrees of mixedness for a biphoton ququart, we
The theoretical ququart density matrix results for this case, as in eq. (5), takes a very simple form,

\[
\rho_{\text{mix}}^{\text{theory}} = \begin{pmatrix}
0.2500 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0.7500
\end{pmatrix}.
\] (8)

We obtain \(\text{Tr}[\rho_{\text{mix}}^{\text{theory}}] = 0.625\) and \(S = 0.406\) for the experimentally reconstructed ququart density matrix. The fidelity is calculated to be \(F = 0.987\) and Fig. 3(a) shows the graphical representation of the real part of the experimentally reconstructed mixed state ququart shown in eq. (9).

The experimentally reconstructed density matrix has the following inherent errors due to the fluctuations of the coincidence count rate.

\[
\Delta \rho_{\text{mix}}^{\text{exp}} = \begin{pmatrix}
0.0053 & 0.0027 - 0.0027i & 0.0030 + 0.0023i & 0.0071 + 0.0044i \\
0.0027 + 0.0027i & 0.0008 & 0.0044 - 0.0070i & 0.0044 + 0.0053i \\
0.0030 - 0.0023i & 0.0044 + 0.0070i & 0.0007 & 0.0048 - 0.0048i \\
0.0071 - 0.0044i & 0.0044 - 0.0053i & 0.0048 + 0.0048i & 0.0096
\end{pmatrix}.
\] (10)

Let us now discuss the case in which the pump polarization is 45°. In this case, the two ququart basis states \(|3\rangle\) and \(|0\rangle\) are equally excited. Therefore, the theoretical ququart density matrix is given as,

\[
\rho_{\text{mix}}^{\text{theory}} = \begin{pmatrix}
0.5000 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0.5000
\end{pmatrix}.
\] (11)

The experimental ququart density matrix results in \(\text{Tr}[\rho_{\text{mix}}^{\text{theory}}] = 0.5\) and \(S = 0.5\). Note that for a complete mixture of all four ququart basis states, \(\text{Tr}[\rho^2] = 1/4\) and \(S = 1\).

For the equal mixture of \(|3\rangle\) and \(|0\rangle\), the tomographic coincidence measurement outcomes are (for 180 s) \(n_1 = 3442, n_2 = 30, n_3 = 3983, n_4 = 23, n_5 = 1621, n_6 = 2358, n_7 = 1950, n_8 = 1895, n_9 = 1906, n_{10} = 1973, n_{11} = 1959, n_{12} = 1840, n_{13} = 2040, n_{14} = 2026, n_{15} = 1809, and n_{16} = 1909\).

The experimentally reconstructed ququart density matrix for this case is found to be,
The experimentally reconstructed ququart density matrix is characterized by $\rho_{\text{exp}}^\text{mix}$ in eq. (12). The fidelity is calculated to be $F = 0.989$ and $S = 0.504$. The experimental reconstructed mixed state ququart shown in eq. (12) is given by

$$
\rho_{\text{exp}}^\text{mix} = \begin{pmatrix}
0.4584 & 0.0142 + 0.0048i & 0.0253 + 0.0162i & 0.0158 - 0.0097i \\
0.0142 - 0.0048i & 0.0041 & 0.0012 + 0.0004i & -0.0406 + 0.0118i \\
0.0253 - 0.0162i & 0.0012 - 0.0004i & 0.0031 & 0.0024 + 0.0029i \\
0.0158 + 0.0097i & -0.0406 - 0.0118i & 0.0024 - 0.0029i & 0.5313
\end{pmatrix}. \quad (12)
$$

The error in the experimentally reconstructed density matrix, due to the count fluctuations, is found to be,

$$
\Delta \rho_{\text{exp}}^\text{mix} = \begin{pmatrix}
0.0078 & 0.0040 - 0.0039i & 0.0043 + 0.0036i & 0.0075 + 0.0047i \\
0.0040 + 0.0039i & 0.0007 & 0.0047 - 0.0074i & 0.0038 + 0.0047i \\
0.0043 - 0.0036i & 0.0047 + 0.0074i & 0.0006 & 0.0042 - 0.0042i \\
0.0075 - 0.0047i & 0.0038 - 0.0047i & 0.0042 + 0.0042i & 0.0084
\end{pmatrix}. \quad (13)
$$

**Partially-mixed state ququart**

A more general state of a ququart, as in eq. (14), can be prepared by transforming an initial mixed state of the form shown in eq. (3). The same double-crystal scheme was used for generating the SPDC photon pair and, in this experiment, the pump polarization was 45°. A zero-order half-wave plate designed at 823.5 nm was used at the fast-axis angle of 22.5° from the vertical axis.

The ququart density matrix calculated for this setting is,

$$
\rho_{\text{theory}}^{\text{int}} = \begin{pmatrix}
0.2500 & 0 & -0.0086 & 0.2414 + 0.0644i \\
0 & 0.2500 & 0.2414 - 0.0644i & 0.0086 \\
-0.0086 & 0.2414 + 0.0644i & 0.2500 & 0 \\
0.2414 - 0.0644i & 0.0086 & 0 & 0.2500
\end{pmatrix}. \quad (14)
$$

The above theoretical ququart density matrix results $Tr[\rho_{\text{theory}}^{\text{int}}] = 0.5$ and $S = 0.5$ which are equal to the values for the mixed state in eq. (11). This means that, even though the ququart state has been unitarily transformed to a new one with the help of a wave plate, the entropy of the system has not been changed. To actually reduce the entropy of the system, it is necessary to remove the temporal distinguishability between the amplitudes $|0\rangle$ and $|3\rangle$ which is introduced due to ultrafast pumping of the BBO crystals [27]. This can be accomplished, for example, by inserting a properly oriented quartz plate of the exact thickness in the path of the pump laser.

For the partially-mixed biphoton ququart state, the projection measurement resulted the following coincidence counts (for 180 s): $n_1 = 1760$, $n_2 = 1730$, $n_3 = 1733$, $n_4 = 1839$, $n_5 = 1687$, $n_6 = 1630$, $n_7 = 1758$, $n_8 = 1961$, $n_9 = 817$, $n_{10} = 3029$, $n_{11} = 1008$, $n_{12} = 1701$, $n_{13} = 1940$, $n_{14} = 1944$, $n_{15} = 1692$, and $n_{16} = 1192$.

The experimentally reconstructed density matrix is found to be,
which shows $\text{Tr}[\rho_{\text{exp}}^2] = 0.483$ and $S = 0.551$.

The inherent fluctuations of the count rate introduce the following error in the reconstructed density matrix,

$$
\Delta \rho_{\text{exp}}^{\text{int}} = \begin{pmatrix}
0.0059 & 0.0042 - 0.0042i & 0.0046 + 0.0039i & 0.0098 + 0.0036i \\
0.0042 + 0.0042i & 0.0059 & 0.0065 - 0.0054i & 0.0043 + 0.0040i \\
0.0046 - 0.0039i & 0.0065 + 0.0054i & 0.0061 & 0.0042 - 0.0042i \\
0.0098 - 0.0036i & 0.0043 - 0.0040i & 0.0042 + 0.0042i & 0.0059
\end{pmatrix}.
$$

Figure 4 shows the real parts of the experimentally reconstructed and the theoretical density matrices. The calculated fidelity is somewhat low $F = 0.878$ in this case.

**DISCUSSION**

The fidelity $F$ quantifies how close the experimentally prepared ququart state is to the one we intended to prepare, i.e., $F$ quantifies the overlap between the theoretical density matrix and the experimentally reconstructed density matrix. In section 4, we have analyzed errors introduced to the experimentally reconstructed density matrices due to the fluctuations of the count rates. As we have seen in eqs. (8), (11), (14), and (17), however, these fluctuations contribute very small errors to the reconstructed density matrices.

There are a number of external experimental factors which could strongly affect the fidelity. First, errors in the angular settings of the wave plates and polarizers used for the projection measurement. Significant improvement is possible by moving from hand-operated optic holders that are graduated in $1^\circ \sim 2^\circ$ increment to motorized holders. Second, less-than-ideal spatial mode matching for the photon pairs coming from the two different crystals. Mode matching can be implemented by adding a spatial-filter or a short-piece of single-mode fiber and this should improve the fidelity substantially. Third, inaccurate transformation matrices of the DBS. For the ququart tomography discussed in the previous section, accurate experimental reconstruction of the ququart density matrix requires the full knowledge of the polarization state change induced by all optical elements. In our experiment, the custom-made dichroic beam splitter (DBS) exhibited unexpected polarization-changing behaviors. It was found that the polarization states were changed both for the transmitted and the reflected beams. To account for the DBS behaviors, we carried out the Stokes parameter measurements for the transmitted and the reflected beams for six different input polarization states ($|H\rangle$, $|V\rangle$, $|45^\circ\rangle$, $|135^\circ\rangle$, $|R\rangle$, and $|L\rangle$). From these measurements, it was possible to deduce the $2 \times 2$ DBS transformation matrices for the transmitted and the reflected modes. The experimental ququart density matrices shown in the previous section were reconstructed using the DBS matrices and therefore experimental errors introduced to the DBS matrices should have slightly affected the fidelity.

In our experiment, preparation of general states of a ququart was demonstrated using the double-crystal scheme. This scheme, although easy to setup, has potential difficulties in complete control of the ququart states. For example, independent decoherence control for amplitudes $|1\rangle$ ($|H_{\lambda_1}, V_{\lambda_2}\rangle$) and $|2\rangle$ ($|V_{\lambda_1}, H_{\lambda_2}\rangle$) using a birefringent medium is difficult as both amplitudes contain horizontal and vertical polarization components.

For a complete control of the ququart state, i.e., to prepare a ququart state with arbitrary values of $\text{Tr}[\rho^2]$ and $S$, we can envision a four-crystal scheme in which a SPDC source is placed in each arm of a four-path Mach-Zehnder interferometer as shown in Fig. 5. This scheme requires four crystals (two type-I SPDC sources and two type-II SPDC sources) and the four-path interferometer...
must be made stable for an accurate phase control. De-
coherence control can be accomplished by controlling the
effective beam paths of the interferometer arms using the
tunable delays installed at each of the beam path. (Note
that arbitrary pure ququart states can be prepared using
just two crystals, see Ref. [20, 21].)

It is, however, possible to design experimental schemes,
for preparing arbitrary (mixed, partially-mixed, and
pure) ququart states, which are less complicated than
the four-path interferometric scheme shown in Fig. 5. In
the following, we discuss two such experimental schemes,
each of which are suited for particular ququart states in
need.

### Scheme based on Mach-Zehnder interferometer

The basic idea for the new ququart preparation scheme
is based on the observation that the pure ququart state
in eq. (2) can be re-written as,

\[
|\Psi\rangle = c_0|H_{\lambda_1}, H_{\lambda_2}\rangle + c_1|H_{\lambda_1}, V_{\lambda_2}\rangle + c_2|V_{\lambda_1}, H_{\lambda_2}\rangle + c_3|V_{\lambda_1}, V_{\lambda_2}\rangle
\]

\[
= |c_0||H_{\lambda_1}, H_{\lambda_2}\rangle + |c_3|e^{i\phi_{03}}|V_{\lambda_1}, V_{\lambda_2}\rangle + (|c_1||H_{\lambda_1}, V_{\lambda_2}\rangle + |c_2|e^{i\phi_{12}}|V_{\lambda_1}, H_{\lambda_2}\rangle) e^{i\phi_{01}},
\]

(17)

where \(\phi_{ij} (i, j = 0, 1, 2, 3)\) is the relative phase for the \(i\)
and \(j\) ququart basis states. Here, we have put together
the two ququart amplitudes that can be prepared with
type-I SPDC (the first two terms) and the other two
ququart amplitudes that can be prepared with type-II
SPDC (the last two terms).

Experimentally, the first two terms in eq. (17) can
be prepared with two orthogonally oriented type-I BBO
crystals placed in tandem as in Fig. 1. The second two
terms can then be prepared with two similarly placed
type-II BBO crystals. Therefore, coherently or incoher-
ently combining these two experimental schemes will al-
low us to prepare an arbitrary ququart states.

The experimental scheme realizing this idea is shown
in Fig. 6. A Glan-Tompson prism (GP), transmitting
the horizontally polarized component of the UV pump
and reflecting the vertically polarized component, serves
as the input beam splitter for the Mach-Zehnder inter-
erferometer. The reflected pump laser, after passing the
birefringent compensator (Comp.) and a half-wave plate
(HWP2), pumps a set of two orthogonally oriented type-
I BBO crystals. A quartz plate (QP1) compensates the
group velocity delay between the ordinary and the extra-
ordinary polarized photons emitted from the pair of BBO
crystals. The residual pump laser is removed by a uv
mirror (UVM) and two quartz plates QP2 can be tilted along
their optical axes to introduce a phase shift (\(\phi_{03}\)) be-
tween horizontally and vertically polarized type-I biphon-
tons [27]. The dichroic mirror (DM) is designed to trans-
mit the biphoton wavelengths but to reflect the uv pump
beam which comes from the upper path of the Mach-
Zehnder interferometer.

The uv pump beam in the upper path of the Mach-
Zehnder interferometer goes through a birefringent com-
ponsator, phase plates (QP3) for adjusting the phase
(\(\phi_{12}\)) between horizontal and vertical components of the
pump beam, and a piezoelectric translator (PZT) which
introduces a relative phase shift (\(\phi_{01}\)) between the up-
ner and the lower paths of the interferometer. The uv
beam from the upper path, upon reflection at the DM,
serves as the pump for the set of two orthogonally orien-
ted type-II BBO crystals. The group velocity delay
between biphoton amplitudes from the first and the sec-
type-II BBO crystals are then compensated by the
crystal compensator (QP4).

At the output of the experimental setup shown in
Fig. 6 an arbitrary ququart states based on the biphoton
polarization states of frequency-nondegenerate SPDC is
prepared. We note that the scheme is loosely based
on the biphoton qutrit setup demonstrated in Ref. [18],
where three SPDC crystals are exploited for generation
of frequency-degenerate collinear biphotons.

### Scheme based on frequency non-degenerate and
non-collinear regime of SPDC

The interferometric scheme proposed in the previous
section, although straightforward, might not be practi-
cal as the scheme inherits high phase sensitivity of the
The biphoton pure state in eq. (2) can be expressed in the form of orthonormal states of the two subsystems (two polarization qubits). We can make use of the fact that a unique set of orthonormal states of the two subsystems (two polarization qubits) can be formed with two polarization qubits as defined in eq. (2), characterized by the Schmidt coefficients $\chi_i$. The Schmidt decomposition is given by

$$\rho_i = \chi_1 |H_i\rangle\langle H_i| + \chi_2 |V_i\rangle\langle V_i|, \quad (20)$$

where the relative magnitude of the coefficients is controlled by WP and the relative phase is controlled by tilting QP. For this biphoton state, the state of individual subsystem (qubit) is then given as

$$|\psi\rangle = \sqrt{\lambda_1} |H_1\rangle |A_i\rangle + \sqrt{\lambda_2} |V_1\rangle |B_i\rangle, \quad (19)$$

where $\lambda_i$ are the Schmidt coefficients and the Schmidt basis states may be different. Equation (21) represents a general form of a pure biphoton polarization state or a pure biphoton ququart state.

The scheme shown in Fig. 7 is interferometrically more stable than the one in Fig. 6 due to the fact that the photon pair from each type-I BBO crystal goes through the same temporal Hilbert space. In other words, in eq. (20), if $|H_i\rangle \leftrightarrow |A_i\rangle$, then $|V_i\rangle \leftrightarrow |B_i\rangle$ with $\langle A_i | B_i \rangle = 0$. As a result, the state eq. (19) becomes

$$|\psi\rangle \xrightarrow{U_1 \otimes U_2} \sqrt{\lambda_1} |A_1\rangle |A_2\rangle + \sqrt{\lambda_2} |B_1\rangle |B_2\rangle, \quad (21)$$

where $U_1$, for example, refers to the unitary polarization transformation for photon 1. Equation (21) represents a general form of a pure biphoton polarization state or a pure biphoton ququart state.

The proposed setup to implement this idea is shown in Fig. 7. A set of two orthogonally oriented type-I BBO crystals, cut for frequency-nondegenerate non-collinear type-I SPDC, are pumped by a uv laser whose polarization is controlled by a half-wave plate (WP). A set of quartz plates (QP) introduces the relative phase between the horizontal and the vertical components of the uv pump. The initial polarization state of the biphoton generated in this process can be written as

$$|\psi\rangle = \sqrt{\lambda_1} |H_{\lambda_1}\rangle |H_{\lambda_2}\rangle + \sqrt{\lambda_2} |V_{\lambda_1}\rangle |V_{\lambda_2}\rangle, \quad (19)$$

where the subsystem index $i = 1, 2$.

So far we have produced a state with a given set of Schmidt coefficients (i.e., a given concurrence), but in a fixed (H-V) basis. The use of a quarter-wave plate (QWP) and a half-wave plate (HWP) on each of the photons then locally and unitarily transforms the polarization states of the individual photon. Since the unitary transformation conserves the inner product, there always exists a state orthogonal to a given one in the two-dimensional Hilbert space. In other words, in eq. (20), if $|H_i\rangle \leftrightarrow |A_i\rangle$, then $|V_i\rangle \leftrightarrow |B_i\rangle$ with $\langle A_i | B_i \rangle = 0$. As a result, the state eq. (19) becomes

The scheme shown in Fig. 7 is interferometrically more stable than the one in Fig. 6 due to the fact that the photon pair from each type-I BBO crystal goes through the same temporal Hilbert space. In principle, to prepare the state in eq. (21), the two paths in Fig. 7 should only be equal up to the coherence length of the pump laser. If the pump is broadband, however, it becomes necessary to further erase the temporal distinguishability between amplitudes from the first and the second BBO crystals, for example, by using a set of compensating crystals [27].

**CONCLUSION**

We have demonstrated that an arbitrary general single-ququart state can be prepared in a simple and controllable way by using the biphoton polarization state of ultrashort-pumped collinear frequency-nondegenerate SPDC. In addition, we have proposed two additional schemes which can be applied for arbitrary (pure and mixed) ququart state preparation.

Compared to other multi-dimensional quantum systems, the biphoton ququart is easier to prepare and characterize and states other than pure states can be prepared more easily. Furthermore, it is possible to prepare a multi-ququart entangled state linearly [22, 23]. We, therefore, believe that the general ququart state preparation scheme analyzed in this paper will find applications in quantum key distribution and quantum information processing.
Acknowledgments

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