On four-photon entanglement from parametric down-conversion process

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Abstract
We propose a scheme to generate a family of four-photon polarization-entangled states from the second-order emission of the spontaneous parametric down-conversion (PDC) process. Based on linear optical elements and the coincidence detection, the four indistinguishable photons emitted from PDC source result in the family of states which are so different from the previous. By tuning the orientation of wave plate, we are able to obtain the well-known four-photon Greenberger–Horne–Zeilinger (GHZ) state, superposition of two (three) orthogonal GHZ states and generic superposition of four orthogonal GHZ states. As an application, we analyze quantum nonlocality for the present superposition states. Under particular phase settings, we calculate the local hidden variable (LHV) correlation and the quantum correlation function. As a result, the Bell inequality derived from the LHV correlation is completely violated by these four-photon entangled states. The maximal quantum violation occurs naturally with the four-photon GHZ state, and there exist the quantum violations which are larger than previously reported values over a surprisingly wide range. It means that the present four-photon entangled states are therefore suitable for testing the LHV theory.

Keywords Multiphoton entanglement · Parametric down-conversion process · Quantum nonlocality

1 Introduction
As a quantum resource, multiphoton entanglement [1,2] plays an important role in both theoretical studies and experimental techniques. One of the attractive aspects of this field is how to generate the desired multiphoton entangled states [3–14]. Since a spontaneous parametric down-conversion (PDC) [15–18] source is capable of creating
pairs of entangled photons, in general, a standard method of generating the multiphoton entanglement is to evolve the pairs emitted from respective source with a set of passive optical elements [2]. For this purpose, it is crucial to suppress undesired multipair emissions and enhance the collection efficiency of the entangled photon pairs.

A higher-order emission of the PDC source [19–25] is related to a twin-beam multiphoton entangled state. For the second-order emission, it has been shown that [23], when the duration of the pump pulse is much shorter than the coherence time of the photons, the emitted state can be described as an indistinguishable twin-beam four-photon entangled state (a genuine four-party entanglement) rather than two independent pairs. Based on such four-photon emission, Weinfurter and Žukowski [26] proposed a novel method of generating a superposition state of a four-photon Greenberger–Horne–Zeilinger (GHZ) state with a product state of two Bell states by using several linear optical elements, and this four-photon entangled state can be used to test the local hidden variable (LHV) theory. Later, it leads to the following discussion on four-photon entanglement in two-crystal geometry by Li and Kobayashi [27,28]. Experimentally, by using a wave plate with a tunable angle parameter, Wieczorek et al. [29] and Lanyon et al. [30] reported two schemes for realizing multiphoton entanglement in the laboratory, respectively. One is used to prepare a family of four-photon superposition states of the GHZ state with the product state of two Bell states, and another is used to observe arbitrary four-photon \emph{W}-class states.

In this paper, we at first focus on the generation of a family of the generic superposition states of four orthogonal four-photon GHZ states from the second-order emission of the PDC source. The optical quantum operations can be implemented by using only passive linear optical devices, especially a controllable angle parameter by means of polarization rotation after the PDC source. Then, as an application, we calculate LHV correlation and quantum correlation function for the output states. The result is that the quantum violation of the present four-photon entangled states is larger than the previously reported values [26–28], and clearly, the maximal quantum violation occurs with the four-photon GHZ state.

2 Generation of a family of four-photon polarization-entangled states

In the PDC process, a simplified Hamiltonian [5] of the nonlinear interaction is given by

$$\hat{H}_{\text{int}} = i \kappa (\hat{a}_{1H}^\dagger \hat{a}_{2V}^\dagger - \hat{a}_{1V}^\dagger \hat{a}_{2H}^\dagger) + \text{H.c.},$$

where \(\hat{a}_{1H}^\dagger\) and \(\hat{a}_{1V}^\dagger\) indicate, respectively, the creation operators with horizontal (H) and vertical (V) polarizations in the spatial mode \(a_1\), and \(\hat{a}_{2H}^\dagger\) and \(\hat{a}_{2V}^\dagger\) indicate the corresponding creation operators for the spatial mode \(a_2\). \(\kappa\) is a real-valued coupling constant, related to the nonlinearity of the crystal and the intensity of the pump pulse. The resulting photon state [21,22] is described by

$$|\Psi\rangle = \frac{1}{\cosh^2 \tau} \sum_{n=0}^{\infty} \sqrt{n} + 1 \tan \hbar^n \tau |\psi_n^-\rangle,$$

(1)
Fig. 1 Generation and detection of the four-photon polarization-entangled states. The parametric downconversion (PDC) source is used to produce four indistinguishable twin-beam entangled photons. \( R \) represents a polarization rotation implemented by a wave plate with a controllable angle parameter \( \theta \). Two 50:50 beam splitters (BSs) and the following polarizing beam splitter (PBS) are used to evolve these photons from the spatial modes \( a_1 \) and \( a_2 \) to \( d_1, d_2, d_3 \) and \( d_4 \). Each \( \text{PA}_i \) \( (i = 1, 2, 3, 4) \) is a polarization analysis used to investigate the entanglement property of the output four-photon entangled states

where \( \tau = \kappa t \) is the interaction parameter with \( t \) being interaction time and

\[
|\psi_n^-\rangle = \frac{1}{\sqrt{n+1}} \frac{1}{n!} \left( \hat{a}^\dagger_{1H} \hat{a}^\dagger_{2V} - \hat{a}^\dagger_{1V} \hat{a}^\dagger_{2H} \right)^n |0\rangle.
\]  

(2)

We now concentrate on the second-order emission of the PDC source, i.e.,

\[
|\Phi\rangle = \frac{1}{2\sqrt{3}} \left( \hat{a}^\dagger_{1H} \hat{a}^\dagger_{2V} - \hat{a}^\dagger_{1V} \hat{a}^\dagger_{2H} \right)^2 |0\rangle.
\]  

(3)

and describe a method of generating a family of four-photon polarization-entangled states, as indicated in Fig. 1. Consider a wave plate \( R \) used to rotate the polarization in spatial mode \( a_2 \) with the evolution in operator form

\[
\hat{a}^\dagger_{2H} \rightarrow \cos \theta \hat{a}^\dagger_{2H} - i \sin \theta \hat{a}^\dagger_{2V},
\]  

(4)

\[
\hat{a}^\dagger_{2V} \rightarrow \cos \theta \hat{a}^\dagger_{2V} - i \sin \theta \hat{a}^\dagger_{2H}.
\]  

(5)

Due to the action of this wave plate, the initial four-photon entangled state evolves

\[
\frac{1}{2\sqrt{3}} \left( \cos \theta \hat{a}^\dagger_{1H} \hat{a}^\dagger_{2V} - i \sin \theta \hat{a}^\dagger_{1H} \hat{a}^\dagger_{2V} - \cos \theta \hat{a}^\dagger_{1V} \hat{a}^\dagger_{2H} + i \sin \theta \hat{a}^\dagger_{1V} \hat{a}^\dagger_{2V} \right)^2 |0\rangle.
\]  

(6)
We here assume that the two polarization independent 50:50 beam splitters (BSs) transform $a_1$ into $(d_1 + D_1)/\sqrt{2}$ and transform $a_2$ into $(d_4 - D_2)/\sqrt{2}$, respectively. Since the interference at two BSs, the twin-beam four-photon entangled state yields

$$|\Phi_{\text{BS}}\rangle = \frac{1}{8\sqrt{3}} \left\{ \cos^2 \theta \left[ (d_{1H}^\dagger + D_{1H}^\dagger) (d_{4V}^\dagger - D_{2V}^\dagger) - (d_{1V}^\dagger + D_{1V}^\dagger) (d_{3H}^\dagger - D_{2H}^\dagger) \right]^2 
- \sin^2 \theta \left[ (d_{1H}^\dagger + D_{1H}^\dagger) (d_{4H}^\dagger - D_{2H}^\dagger) - (d_{1V}^\dagger + D_{1V}^\dagger) (d_{2H}^\dagger - D_{2V}^\dagger) \right]^2 
- i \sin(2\theta) \left[ (d_{1H}^\dagger + D_{1H}^\dagger) (d_{4V}^\dagger - D_{2V}^\dagger) - (d_{1V}^\dagger + D_{1V}^\dagger) (d_{4H}^\dagger - D_{2H}^\dagger) \right] \right\} |0\rangle.$$  \hspace{1cm} (7)

When photons travel through the PBS in the horizontal–vertical basis, the evolution of the photons from the spatial modes $D_1$ and $D_2$ to $d_2$ and $d_3$ obeys

$$|V\rangle_{D_1} \rightarrow |V\rangle_{d_2}, \quad |H\rangle_{D_1} \rightarrow |H\rangle_{d_3}, \quad |V\rangle_{D_2} \rightarrow |V\rangle_{d_3}, \quad |H\rangle_{D_2} \rightarrow |H\rangle_{d_2}. \hspace{1cm} (8)$$

At last, under the condition of detecting one photon in each of the spatial modes $d_1, d_2, d_3$ and $d_4$, we have

$$|\Phi(\theta)\rangle = \frac{1}{\sqrt{2}}\left[ |HVVH\rangle_{d_1d_2d_3d_4} + |VHHV\rangle_{d_1d_2d_3d_4} \right] + 2 \sin^2 \theta (|HHHH\rangle_{d_1d_2d_3d_4} + |VVVV\rangle_{d_1d_2d_3d_4})
- i \sin(2\theta) (|HVVV\rangle_{d_1d_2d_3d_4} + |VHHH\rangle_{d_1d_2d_3d_4})
- |HHHV\rangle_{d_1d_2d_3d_4} - |VVHV\rangle_{d_1d_2d_3d_4}), \hspace{1cm} (9)$$

where $N = [2 \cos^2(2\theta) + 8 \sin^4 \theta + 4 \sin^2(2\theta)]^{-1/2}$. Indeed, the present state is the superposition of four orthogonal GHZ states with unequal probabilities. Furthermore, as it enables one to observe a range of four-photon entangled states, the present scheme is quite versatile in this sense. Clearly, for $\theta = 0$, it yields the canonical four-photon polarization-entangled GHZ state

$$|\Phi(0)\rangle = \frac{1}{\sqrt{2}} \left[ |HVVH\rangle_{d_1d_2d_3d_4} + |VHHV\rangle_{d_1d_2d_3d_4} \right]; \hspace{1cm} (10)$$

for $\theta = \pi/2$, it yields the superposition of two orthogonal GHZ states

$$|\Phi(\pi/2)\rangle = \frac{1}{\sqrt{10}}\left[ 2(\langle HHHH\rangle_{d_1d_2d_3d_4} + \langle VVVV\rangle_{d_1d_2d_3d_4})
- (\langle HVVH\rangle_{d_1d_2d_3d_4} + \langle VHVV\rangle_{d_1d_2d_3d_4}) \right]; \hspace{1cm} (11)$$
for $\theta = \pi/4$, it yields the superposition of three orthogonal GHZ states with equal probability

$$|\Phi(\pi/4)\rangle = \frac{1}{\sqrt{6}}[(|HHHH\rangle_{d_1d_2d_3d_4} + |VVVV\rangle_{d_1d_2d_3d_4})$$
$$- i(|HVVV\rangle_{d_1d_2d_3d_4} + |HHHH\rangle_{d_1d_2d_3d_4})$$
$$+ i(|HVVV\rangle_{d_1d_2d_3d_4} + |VVVV\rangle_{d_1d_2d_3d_4})];$$  \hspace{1cm} (12)

and for $\theta = \pi/6$, one can obtain the superposition of four orthogonal GHZ states

$$|\Phi(\pi/6)\rangle = \frac{1}{4}[(|HVVH\rangle_{d_1d_2d_3d_4} + |VHHV\rangle_{d_1d_2d_3d_4})$$
$$+ (|HHHH\rangle_{d_1d_2d_3d_4} + |VVVV\rangle_{d_1d_2d_3d_4})$$
$$- i\sqrt{3}(|HVVV\rangle_{d_1d_2d_3d_4} + |HVVH\rangle_{d_1d_2d_3d_4})$$
$$+ i\sqrt{3}(|HHHV\rangle_{d_1d_2d_3d_4} + |VVHV\rangle_{d_1d_2d_3d_4})].$$  \hspace{1cm} (13)

### 3 Quantum nonlocality of the four-photon entangled states

As is well known, Bell inequalities can be used to investigate the constitutive relations for multiparticle entanglement [31–37] and quantum nonlocality [38–45]. We now turn to the discussion of quantum nonlocality of the present four-photon entangled state. Four-photon GHZ state maximally violates Mermin inequality [40]. Especially for a four-particle system, it has been shown that [45] a compact Mermin-type inequality is also maximally violated by four-qubit GHZ state with a certain constant visibility 2. Next, as an interesting application, we investigate quantum nonlocality of the present superposition states.

For a four-photon system, for the sake of simplicity, we suppose that each location of the photon is allowed to choose independently between two dichotomic observables and each outcome can take one of the values +1 or −1. In order to investigate quantum nonlocality of the present four-photon entangled states, we here introduce a polarization analysis basis [26]

$$|m_x, \phi_x\rangle = \frac{1}{\sqrt{2}}(|V\rangle_x + m_x e^{-i\phi_x}|H\rangle_x),$$  \hspace{1cm} (14)

where $\phi_x$ is a local phase setting connecting with location $d_x$ ($x = 1, 2, 3, 4$) and $m_x = \pm 1$ are two possible measurement results.

With this preparation, then, in output stations $d_1, d_2, d_3$ and $d_4$, the probability of getting a particular set of results $m_1, m_2, m_3$ and $m_4$ with local phase settings $\phi_1, \phi_2, \phi_3$ and $\phi_4$, respectively, is given by

$$\left(\frac{N}{4}\right)^2 [\cos^2(2\theta)|\alpha|^2 + 4\sin^4\theta|\beta|^2 + \sin^2(2\theta)|\gamma|^2$$
$$+ 4\cos(2\theta)\sin^2\theta \text{Re}(\alpha\beta^*) - \sin(4\theta)\text{Im}(\alpha\gamma^*) - 4\sin(2\theta)\sin^2\theta \text{Im}(\beta\gamma^*)],$$  \hspace{1cm} (15)
where

\[
\alpha = m_1 m_4 e^{i(\phi_1 + \phi_4)} + m_2 m_3 e^{i(\phi_2 + \phi_3)},
\]

\[
\beta = 1 + m_1 m_2 m_3 m_4 e^{i(\phi_1 + \phi_2 + \phi_3 + \phi_4)},
\]

and

\[
\gamma = m_1 e^{i\phi_1} + m_2 m_3 m_4 e^{i(\phi_2 + \phi_3 + \phi_4)} - m_1 m_2 m_3 e^{i(\phi_1 + \phi_2 + \phi_3)} - m_4 e^{i\phi_4}.
\]

In general, the correlation function of measurement results is defined as the average of the product of the local values. In this way, the quantum correlation function for the present four-photon superposition states yields

\[
E(\theta, \phi_1, \phi_2, \phi_3, \phi_4) = 2N^2 \{ \cos^2(2\theta) \cos(\phi_1 - \phi_2 - \phi_3 + \phi_4) \\
+ 4\sin^4\theta \cos(\phi_1 + \phi_2 + \phi_3 + \phi_4) \\
+ \sin^2(2\theta) [\cos(\phi_1 - \phi_2 - \phi_3 - \phi_4) \\
+ \cos(\phi_1 + \phi_2 + \phi_3 - \phi_4)] \}.
\]

(19)

On the other hand, in terms of the LHV model, the photons are locally but realistically correlated. A four-photon correlation function for two alternative dichotomic measurements can be described as a four-index tensor [26]

\[
\hat{E}_{\text{LHV}} = \sum_{k_1,k_2,k_3,k_4=1,2,3,4} p_{k_1,k_2,k_3,k_4} v_{k_1}^{x_1} \otimes v_{k_2}^{x_2} \otimes v_{k_3}^{x_3} \otimes v_{k_4}^{x_4},
\]

(20)

where \( v_{k_x} = (I_x(\phi_{k_x}^1, \lambda), I_x(\phi_{k_x}^2, \lambda)) \) \((x = 1, 2, 3, 4)\) represents a two-dimensional real vector, \( I_x(\phi_{k_x}^1, \lambda), I_x(\phi_{k_x}^2, \lambda) = \pm 1 \) are the predetermined outcomes of the measurements, \( p_{k_1,k_2,k_3,k_4} \) is probability of any possible measurement result and thus \( \sum_{k_1,k_2,k_3,k_4=1,2,3,4} p_{k_1,k_2,k_3,k_4} = 1, \lambda \) is a hidden variable.

Let us now suppose that \( v_1^x = (1, 1), v_2^x = (1, -1), v_3^x = (-1, -1) \) and \( v_4^x = (-1, 1) \). Then, we have \( v_3^x = -v_1^x \) and \( v_4^x = -v_2^x \), or clearly \( v_{k_x}^{x+2} = -v_{k_x}^x \) with \( k_x = 1, 2 \). Therefore, we can rewrite expression (20) as

\[
\hat{E}_{\text{LHV}} = \sum_{k_1,k_2,k_3,k_4=1,2} c_{k_1,k_2,k_3,k_4} v_{k_1}^{x_1} \otimes v_{k_2}^{x_2} \otimes v_{k_3}^{x_3} \otimes v_{k_4}^{x_4},
\]

(21)

where the correlation coefficients \( c_{k_1,k_2,k_3,k_4} \) must satisfy

\[
\sum_{k_1,k_2,k_3,k_4=1,2} |c_{k_1,k_2,k_3,k_4}| \leq 1,
\]

(22)

since \( c_{k_1,k_2,k_3,k_4} = p_{k_1,k_2,k_3,k_4} - p_{k_1,+2,k_2,-k_3,k_4} - \cdots p_{k_1,-2,k_2,+k_3,k_4} + p_{k_1,+2,k_2,+k_3,k_4} + p_{k_1,-2,k_2,-k_3,k_4} - p_{k_1,+2,k_2,-k_3,k_4} - p_{k_1,-2,k_2,+k_3,k_4} - p_{k_1,+2,k_2,+k_3,k_4} \). This inequality is derived from
LHV model and generically called Bell inequality, which can be used to test the LHV theory.

In order to compare the LHV prediction and the quantum prediction, we now rewrite the quantum correlation function as a tensor

$$\hat{E}_Q = \sum_{k_1, k_2, k_3, k_4 = 1, 2} q_{k_1, k_2, k_3, k_4} v_1^{k_1} \otimes v_2^{k_2} \otimes v_3^{k_3} \otimes v_4^{k_4}. \quad (23)$$

Consider the experimental settings $\phi_1^1 = 0, \phi_1^2 = \pi/2, \phi_{2,3,4}^1 = -\pi/4$ and $\phi_{2,3,4}^2 = \pi/4$, as described in [26]. The quantum correlation coefficients $q_{k_1, k_2, k_3, k_4}$ can be straightforwardly obtained according to result (19), and then, we have

$$\sum_{k_1, k_2, k_3, k_4 = 1, 2} |q_{k_1, k_2, k_3, k_4}| = \frac{1}{\sqrt{2}} [1 + F(\theta)], \quad (24)$$

where

$$F(\theta) = \frac{|\cos^2(2\theta) + 4\sin^4\theta - 2\sin^2(2\theta)| + 2|\cos^2(2\theta) - 4\sin^4\theta|}{\cos^2(2\theta) + 4\sin^4\theta + 2\sin^2(2\theta)}. \quad (25)$$

For the sake of definiteness, we plot the quantum prediction as a function of angle parameter $\theta$ ranging from $\theta = 0$ to $\theta = \pi/2$, as shown in Fig. 2. It is clear that the Bell inequality derived from the LHV theory is completely violated. For example, the states $|\Phi(0)\rangle, |\Phi(\pi/6)\rangle, |\Phi(\pi/4)\rangle$ and $|\Phi(\pi/2)\rangle$ violate the Bell inequality.
with the values $2\sqrt{2}$, $3\sqrt{2}/4$, $\sqrt{2}$ and $8\sqrt{2}/5$, respectively. More concretely, for $0 \leq \theta \leq \pi/6$, the value of quantum violation decreases as the angle parameter $\theta$ increases; for $\pi/6 \leq \theta < \arctan[(\sqrt{5} + 1)/\sqrt{10} - 2\sqrt{5}]$, the value of quantum violation increases and then decreases with the relative maximum value $\sqrt{2}$, and for $\arctan[(\sqrt{5} + 1)/\sqrt{10} - 2\sqrt{5}] \leq \theta \leq \pi/2$, the value of quantum violation increases smoothly with increasing angle parameter.

4 Discussion and summary

In summary, we have shown a simple and fruitful scheme of generating a family of four-photon polarization-entangled states. The present optical circuit is quite different from the one that yields a single superposition of a four-photon GHZ state and a tensor product of two Bell states [26], or the experimental observation of the entire family of them [29]. In our configuration, based on the evolution of polarization rotation, the interference effects at optical elements and the fourfold coincidence detection, one can obtain the desired four-photon entangled states from the second-order emissions of the PDC source. In particular, due to tunable angle parameter the present approach permits the preparation of various superposition states of orthogonal GHZ states, as well as the canonical GHZ state. Also, although there may be some slight disadvantages in experimental realization because of mismatched pump light with higher-order emissions and imperfect interference of linear optical elements [1,2,46], it is feasible since only a few available optical elements are employed in the present scheme, just like it is in the case of the experimental observations for multiphoton entangled states [29,30].

Besides the exploration of the generic four-photon entangled states, as an application, we have analyzed quantum nonlocality of the present superposition states. By calculating the quantum correlation function, we reveal the relationship between quantum prediction and angle parameter $\theta$. Notably, compared with the previously reported values $4\sqrt{2}/3 \simeq 1.886$ [26] and $3\sqrt{2}/2 \simeq 2.121$ [27,28], the present quantum violations are larger than the two outcomes over a surprisingly wide range. The final result is that the present four-photon entangled states always violate the Bell inequality. This means that the present four-photon entangled states are well suited to testing the quantum formalism against the LHV model in experiments.

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