Generation of hexapartite entanglement in a four-wave-mixing process with a spatially structured pump: Theoretical study

Hailong Wang,1,6 Kai Zhang,1 Nicolas Treps,2 Claude Fabre,2 Jun Zhang,3 and Jietai Jing†1,4,5,*

1State Key Laboratory of Precision Spectroscopy, Joint Institute of Advanced Science and Technology, School of Physics and Electronic Science, East China Normal University, Shanghai 200062, China
2Laboratoire Kastler Brossel, Sorbonne Université, CNRS, ENS-PSL Research University, Collège de France, 4 place Jussieu, 75252 Paris, France
3UMich-SJTU Joint Institute and Key Laboratory of System Control and Information Processing (MOE), Shanghai Jiao Tong University, Shanghai, 200240, China
4Collaborative Innovation Center of Extreme Optics, Shannxi University, Taiyuan, Shannxi 030006, China
5Department of Physics, Zhejiang University, Hangzhou 310027, China
6College of Optical and Electronic Technology, China Jiliang University, Hangzhou 310018, China

(Received 26 March 2020; accepted 20 July 2020; published 21 August 2020)

We have experimentally shown in a recent paper [Wang et al., Phys. Rev. A 95, 051802(R) (2017)] that a four-wave mixing (FWM) process in a Rb vapor with a spatially structured pump (SSP), consisting of a coherent combination of two tilted pump beams, can generate six well-separated bright beams that are quantum correlated. The experimental setup is compact, simple, phase insensitive, and easily scalable to a larger number of quantum correlated beams. For applications to quantum information processing, multipartite entanglement is needed in addition to quantum correlations. Very recently, we have also experimentally demonstrated the generation of the hexapartite entanglement from this SSP-based FWM process [Zhang et al., Phys. Rev. Lett. 124, 090501 (2020)]. The purpose of this theoretical paper is to extensively investigate the hexapartite entanglement properties by using different entanglement witnesses such as the van Loock-Furusawa (VLF) criterion and the positivity under partial transposition criterion. We find that hexapartite entanglement is indeed present among the six beams. In addition, we also study the supermodes of this system using basis change.

DOI: 10.1103/PhysRevA.102.022417

I. INTRODUCTION

Multipartite entanglement is a crucial physical resource not only for the tests of fundamental quantum science [1] but also for the development of quantum technologies [2]. The conventional technique for generating continuous variable (CV) multipartite entanglement relies on linearly mixing several single-mode squeezed states using a set of successive beamsplitters [3–10]. This implies that the complexity of the experimental layout will dramatically increase with the number of modes of the generated entangled state. Some strategies utilizing frequency multiplexing [11–14] and time multiplexing [15] have been used to reduce the complexity and ensure the scalability to a very large number of modes. Very recently, the deterministic generation of two-dimensional cluster states in the time domain for universal quantum computation has been experimentally reported [16,17]. Undoubtedly, these schemes are beneficial to measurement-based quantum computation. However, spatially separated multipartite entangled beams are needed for distant quantum communication. Therefore, in the spatial domain, the generation scheme of spatially separated multipartite entangled beams from a parametric downconversion process pumped by a spatially structured pump (SSP) made of multiple symmetrically tilted plane waves has been theoretically proposed [18–20].

The four-wave mixing (FWM) process in a hot atomic vapor cell has been proved to be an efficient technique for generating quantum correlated twin beams and bipartite entangled beams [21–28]. The technique has the following advantages [29]: no need of an optical cavity due to strong nonlinearity of the system and spatial separation of the generated nonclassical beams. Our group has shown that two cascaded FWM processes were able to produce strong quantum correlated triple beams [30]. Inspired by the above-mentioned theoretical proposals of pump shaping [18–20], the six beams generated by a FWM process in a hot atomic vapor using a SSP consisting of a coherent combination of two tilted pump beams have been experimentally demonstrated to be quantum correlated [31], which means that a specific combination of the intensity fluctuations of the generated beams is below the standard quantum limit (SQL), a phenomenon often called intensity-difference squeezing. Therefore, the question arises to know whether the observed quantum correlation is the sign of the existence of entanglement, i.e., the quantum correlations involving conjugate observables such as amplitude and phase quadratures. Very recently, we have experimentally shown that the six beams are in fact entangled using both van Loock-Furusawa (VLF) criterion and positivity under partial transposition (PPT) criterion [32]. In other words, there exists hexapartite entanglement in such SSP-based FWM processes.

*Corresponding author: jjing@phy.ecnu.edu.cn
This paper serves as an extensive theoretical study of the generation of hexapartite entanglement from SSP-based FWM processes. It is organized as follows: In Sec. II, we describe the FWM process with a SSP and exactly solve the corresponding quantum evolution equations; In Sec. III, we evaluate the quantum correlation shared by the six beams; In Sec. IV, we use two different criteria for testing the entanglement likely to exist among the six beams, namely, VLF criterion and PPT criterion; In Sec. V, the supermodes of the six beams are also analyzed; In Sec. VI, we give a brief summary of the results.

II. FWM PROCESS WITH A SSP

The FWM configuration for generating hexapartite entanglement is shown in Fig. 1(a): two bright pump beams (pump$_1$ and pump$_2$) tuned about 0.85 GHz to the blue of the $^{85}$Rb D1 Line transition (5S$_{1/2} \rightarrow 5P_{1/2}$), which constitute the SSP, are focused and crossed at the center of a hot $^{85}$Rb vapor cell at a small angle. A coherent weak seed beam, 3.04 GHz redshifted from the pump beams (in fact, with a frequency of 377107.21 GHz), is sent into the vapor cell and symmetrically crossed with the two pump beams. The seed beam and the two pump beams are not within the same plane, making the multiple output beams naturally separated in space. The exact energy diagram giving the seed beam, the pump beams, and the Rb energy levels can be seen in Fig. 1(b) [30,31,33]. Because of this geometry, first, each pump beam will interact with the seed beam individually by a single-pump FWM process [21–29]. The seed beam is amplified ($\hat{a}_1$) and two conjugate beams ($\hat{a}_2$ and $\hat{a}_3$) are simultaneously generated. Due to the phase matching condition, a dual-pump FWM process involving both of the two pump beams is also possible, in which each pump beam annihilates one photon, the seed beam gets one photon, and another photon is generated synchronously in a new conjugate beam ($\hat{a}_4$). Three FWM processes contribute to the amplification of the probe beam ($\hat{a}_1$) and the generation of the three conjugate beams ($\hat{a}_2$, $\hat{a}_3$, and $\hat{a}_4$). The new probe beam $\hat{a}_5$ ($\hat{a}_6$) is generated by the coupling between $\hat{a}_1$ and pump$_1$ (pump$_2$). Another type of coupling is possible, in which beam $\hat{a}_4$ ($\hat{a}_2$) couples with pump$_1$ and pump$_2$, resulting in the generation of $\hat{a}_5$ ($\hat{a}_6$).

The interaction structure of the output beams is summarized in Fig. 1(c). The purple dots indicate the spatial location of the pump$_1$ and pump$_2$ at the output of the vapor cell. Similarly, the red (orange) dot indicates the spatial location of the probe (conjugate) beams at the output of the vapor cell. Fig. 1(c) also gives the nonlinear interaction structure of the six beams by the blue lines [34]. The connection with purple dot means the single-pump FWM interaction, and the connection without purple dot means the aforementioned dual-pump FWM interaction. The generation of beams $\hat{a}_5$ and $\hat{a}_6$ includes both the single-pump and dual-pump FWM interactions.

The interaction Hamiltonian in the undepleted and classical coherent pump approximation can be written as

$$\hat{H} = i\hbar(\varepsilon_1 e^{i(\phi_1 - \psi_i - \psi_j)} \hat{a}_1 \hat{a}_1^\dagger + \varepsilon_2 e^{i(\phi_2 + \phi_3 - \psi_j)} \hat{a}_3 \hat{a}_3^\dagger + \varepsilon_3 e^{2i\phi_2} \hat{a}_4 \hat{a}_4^\dagger + \varepsilon_4 e^{2i\phi_1} \hat{a}_5 \hat{a}_5^\dagger + \varepsilon_6 e^{i(\phi_1 + \phi_2)} \hat{a}_6 \hat{a}_6^\dagger + H.c.,$$

where $\phi_1$ and $\phi_2$ are the phases of the two pump beams, $\psi_i$ ($i = 1, 2, 3, 4, 5, 6$, and 7) represents the interaction strength of the various FWM processes, $\hat{a}_j^\dagger$ ($j = 1, 2, 3, 4, 5, 6$, and 6) is the bosonic creation operator of the beam $j$ and H.c. is the Hermitian conjugate.

Now, let us allow for the possibility of changing an angle $\psi_i$ (the phase reference of the phase space of each beam $i$) by using a rotated creation operator $\hat{a}_j^\dagger = \hat{a}_j^\dagger e^{i\phi_i}$, with, of course, $[\hat{a}_j^\dagger, \hat{a}_j^\dagger] = 1$. The new expression of the Hamiltonian is

$$\hat{H} = i\hbar(\varepsilon_1 e^{i(2\phi_1 - \psi_i - \psi_j)} \hat{a}_1 \hat{a}_1^\dagger + \varepsilon_2 e^{i(\phi_1 + \phi_3 - \psi_j)} \hat{a}_3 \hat{a}_3^\dagger + \varepsilon_3 e^{2i\phi_2} \hat{a}_4 \hat{a}_4^\dagger + \varepsilon_4 e^{2i\phi_1} \hat{a}_5 \hat{a}_5^\dagger + \varepsilon_5 e^{i(\phi_1 + \phi_2)} \hat{a}_6 \hat{a}_6^\dagger + H.c.,$$

where $\phi_1$ and $\phi_2$ are the phases of the two pump beams, $\psi_i$ ($i = 1, 2, 3, 4, 5, 6$, and 7) represents the interaction strength of the various FWM processes.
It is easy to see that if one chooses the following phases for the creation operators:
\[
\begin{align*}
\psi_1 &= \phi_1 - \phi_2, \\
\psi_2 &= \phi_1 + \phi_2, \\
\psi_3 &= 2\phi_2, \\
\psi_4 &= 3\phi_2 - \phi_1, \\
\psi_5 &= 2\phi_1 - 2\phi_2, \\
\psi_6 &= 0,
\end{align*}
\]
the Hamiltonian takes the simple form with no explicit phases:
\[
\hat{H} = i\hbar [\varepsilon_1 \hat{a}^\dagger_1 \hat{a}^\dagger_2 + \varepsilon_2 \hat{a}^\dagger_3 \hat{a}^\dagger_4 + \varepsilon_3 \hat{a}^\dagger_5 \hat{a}^\dagger_6 + \varepsilon_4 \hat{a}^\dagger_7 \hat{a}^\dagger_8 + \varepsilon_5 \hat{a}^\dagger_9 \hat{a}^\dagger_{10} + \varepsilon_6 \hat{a}^\dagger_{11} \hat{a}^\dagger_{12} + \varepsilon_7 \hat{a}^\dagger_{13} \hat{a}^\dagger_{14} + \varepsilon_8 \hat{a}^\dagger_{15} \hat{a}^\dagger_{16} + \text{H.c.}]
\]
\]
This means that the physical properties of the present SSP-based FWM process will not depend on these phases, and especially not on the relative phase \(\phi_1 - \phi_2\) between the two pump beams, as long as one considers properties which do not depend on an absolute definition of the phases of the six output beams.

For the convenience of discussion, the Hamiltonian can be simply expressed as the following form:
\[
\hat{H} = i\hbar [\varepsilon_3 \hat{a}^\dagger_1 \hat{a}^\dagger_2 + \varepsilon_4 \hat{a}^\dagger_3 \hat{a}^\dagger_4 + \varepsilon_7 \hat{a}^\dagger_5 \hat{a}^\dagger_6 + \varepsilon_8 \hat{a}^\dagger_7 \hat{a}^\dagger_8 + \varepsilon_6 \hat{a}^\dagger_{13} \hat{a}^\dagger_{14} + \varepsilon_5 \hat{a}^\dagger_9 \hat{a}^\dagger_6 + \varepsilon_9 \hat{a}^\dagger_{13} \hat{a}^\dagger_{14} + \text{H.c.}]
\]
\]
Denote
\[
\begin{align*}
x &= [\hat{a}^\dagger_1, \hat{a}^\dagger_2, \hat{a}^\dagger_3, \hat{a}^\dagger_4, \hat{a}^\dagger_5, \hat{a}^\dagger_6]^T \quad \text{and} \\
y &= [\hat{a}^\dagger_1, \hat{a}^\dagger_2, \hat{a}^\dagger_3, \hat{a}^\dagger_4, \hat{a}^\dagger_5, \hat{a}^\dagger_6]^T.
\end{align*}
\]
\]
\[
T = \begin{bmatrix}
0 & 0 & (v_2 - \theta) / \varepsilon_1 & (v_2 + \theta) / \varepsilon_1 & (v_2 + \theta) / \varepsilon_1 & (v_2 - \theta) / \varepsilon_1 \\
1 & -1 & -1 & -1 & 1 & 1 \\
0 & 0 & (\theta - v_2) / \varepsilon_1 & -(v_2 + \theta) / \varepsilon_1 & (v_2 + \theta) / \varepsilon_1 & (v_2 - \theta) / \varepsilon_1 \\
-1 & 1 & 1 & 1 & 1 & 1 \\
0 & 0 & (\theta - v_2) / \varepsilon_1 & -(v_2 + \theta) / \varepsilon_1 & (v_2 + \theta) / \varepsilon_1 & (v_2 - \theta) / \varepsilon_1 \\
1 & -1 & 1 & 1 & 1 & 1 \\
\end{bmatrix},
\]
\]
\[
T^{-1} = \frac{1}{8\theta} \begin{bmatrix}
0 & 2\theta & 0 & -2\theta & -2\theta & 2\theta \\
-2\varepsilon_1 & -\theta - v_2 & 2\varepsilon_1 & -\theta - v_2 & \theta + v_2 & \theta + v_2 \\
2\varepsilon_1 & -\theta + v_2 & -2\varepsilon_1 & -\theta + v_2 & \theta - v_2 & \theta - v_2 \\
-2\varepsilon_1 & \theta - v_2 & 2\varepsilon_1 & \theta - v_2 & \theta + v_2 & \theta + v_2 \\
\end{bmatrix}.
\]
\]
In these expressions the parameters \(v_1, v_2\), and \(\theta\) are given by
\[
\begin{align*}
v_1 &= (\varepsilon_2 + \varepsilon_6) / 2, \\
v_2 &= (\varepsilon_2 - \varepsilon_6) / 2, \\
\theta &= \sqrt{2\varepsilon_1^2 + v_2^2}.
\end{align*}
\]
Now we can obtain the input-output relation for six beams using Eqs. (10)–(15).
\[
\begin{bmatrix}
x(t) \\
y(t)
\end{bmatrix} = \begin{bmatrix} T & 0 \\
0 & T \end{bmatrix} \begin{bmatrix} \cosh \Lambda t & \sinh \Lambda t \\
\sinh \Lambda t & \cosh \Lambda t \end{bmatrix} \begin{bmatrix} x(0) \\
y(0) \end{bmatrix} \begin{bmatrix} T^{-1} & 0 \\
0 & T^{-1} \end{bmatrix} \begin{bmatrix} x(0) \\
y(0) \end{bmatrix}.
\]
\]
where \(t = \ell / c\), and \(\ell\) is the interaction length. From Eqs. (6)–(16), the detailed expressions of the annihilation operators of the six beams can be expressed as
\[
\hat{a}_1(t) = \frac{1}{8\theta \varepsilon_1} [A_{11} \hat{a}_1(0) + A_{13} \hat{a}_3(0) + A_{15} \hat{a}_5(0) + B_{12} \hat{a}_2(0) + B_{13} \hat{a}_3(0) + B_{14} \hat{a}_4(0)],
\]
\]

The Heisenberg equations governing the time evolution of the six beams can then be written in a compact form as
\[
\frac{d}{dt} \begin{bmatrix} x \\
y \end{bmatrix} = \begin{bmatrix} 0 & A \\
A & 0 \end{bmatrix} \begin{bmatrix} x \\
y \end{bmatrix}.
\]
\]
where
\[
A = \begin{bmatrix}
0 & \varepsilon_1 & \varepsilon_2 & \varepsilon_3 & 0 & 0 \\
\varepsilon_1 & 0 & 0 & 0 & 0 & \varepsilon_7 \\
\varepsilon_2 & 0 & 0 & 0 & \varepsilon_4 & \varepsilon_5 \\
\varepsilon_3 & 0 & 0 & \varepsilon_4 & 0 & 0 \\
0 & \varepsilon_4 & \varepsilon_6 & 0 & 0 & 0 \\
0 & \varepsilon_7 & \varepsilon_5 & 0 & 0 & 0 \\
\end{bmatrix}.
\]
\]
The solution of Eq. (8) is given by
\[
\begin{bmatrix} x(t) \\
y(t) \end{bmatrix} = \begin{bmatrix} \cosh \Lambda t & \sinh \Lambda t \\
\sinh \Lambda t & \cosh \Lambda t \end{bmatrix} \begin{bmatrix} x(0) \\
y(0) \end{bmatrix}.
\]
\]
For the sake of symmetry and simplicity, we assume that \(\varepsilon_1 = \varepsilon_3 = \varepsilon_4 = \varepsilon_5\) and \(\varepsilon_6 = \varepsilon_7\). Now we can diagonalize \(A\) as
\[
A = T \Lambda T^{-1},
\]
\]
where
\[
\Lambda = \text{diag} [\varepsilon_6, -\varepsilon_6, \theta - v_1, -\theta - v_1, \theta + v_1, -\theta + v_1],
\]
\]

022417-3
\[ \dot{\alpha}_2(t) = \frac{1}{8\theta \varepsilon_1} [A_{22}\dot{\alpha}_2(0) + A_{23}\dot{\alpha}_3(0) + A_{24}\dot{\alpha}_4(0) + B_{21}\dot{\alpha}_1(0) + B_{25}\dot{\alpha}_5(0) + B_{26}\dot{\alpha}_6(0)], \]
\[ \dot{\alpha}_3(t) = \frac{1}{8\theta \varepsilon_1} [A_{32}\dot{\alpha}_2(0) + A_{33}\dot{\alpha}_3(0) + A_{34}\dot{\alpha}_4(0) + B_{31}\dot{\alpha}_1(0) + B_{35}\dot{\alpha}_5(0) + B_{36}\dot{\alpha}_6(0)], \]
\[ \dot{\alpha}_4(t) = \frac{1}{8\theta \varepsilon_1} [A_{42}\dot{\alpha}_2(0) + A_{43}\dot{\alpha}_3(0) + A_{44}\dot{\alpha}_4(0) + B_{41}\dot{\alpha}_1(0) + B_{45}\dot{\alpha}_5(0) + B_{46}\dot{\alpha}_6(0)], \]
\[ \dot{\alpha}_5(t) = \frac{1}{8\theta \varepsilon_1} [A_{51}\dot{\alpha}_1(0) + A_{52}\dot{\alpha}_2(0) + A_{53}\dot{\alpha}_3(0) + A_{56}\dot{\alpha}_6(0) + B_{52}\dot{\alpha}_2(0) + B_{53}\dot{\alpha}_3(0) + B_{54}\dot{\alpha}_4(0)], \]
\[ \dot{\alpha}_6(t) = \frac{1}{8\theta \varepsilon_1} [A_{61}\dot{\alpha}_1(0) + A_{65}\dot{\alpha}_5(0) + A_{66}\dot{\alpha}_6(0) + B_{62}\dot{\alpha}_2(0) + B_{63}\dot{\alpha}_3(0) + B_{64}\dot{\alpha}_4(0)], \]

where

\begin{align*}
A_{11} &= A_{33} = -4\varepsilon_1(v_2 - \theta) \cosh(\theta - v_1) \tau + 4\varepsilon_1(v_2 + \theta) \cosh(v_1 + \theta) \tau, \\
A_{15} &= A_{16} = A_{32} = A_{34} = 2(v_2^2 - \theta^2) \cosh(\theta - v_1) \tau + 2(\theta^2 - v_2^2) \cosh(v_1 + \theta) \tau, \\
A_{22} &= A_{44} = A_{55} = A_{66} = 4\varepsilon_1 \cosh \epsilon \theta \tau + 2(1(\nu) + 2 + 2(1(\nu) + 2 + v_1 + \theta) \cosh(\theta - v_1) \tau + 2(1(\nu) + 2 + v_1 + \theta) \cosh(\theta - v_1) \tau, \\
A_{23} &= A_{43} = A_{51} = A_{63} = -4\varepsilon_1 \cosh(\theta - v_1) \tau + 4\varepsilon_1 \cosh(v_1 + \theta) \tau, \\
A_{24} &= A_{42} = A_{56} = A_{65} = -4\varepsilon_1 \cosh \epsilon \theta \tau + 2(1(\nu) + 2 + 2(1(\nu) + 2 + v_1 + \theta) \cosh(\theta - v_1) \tau + 2(1(\nu) + 2 + v_1 + \theta) \cosh(\theta - v_1) \tau.
\end{align*}

and

\begin{align*}
B_{12} &= B_{14} = B_{35} = B_{36} = 2(\theta^2 - v_2^2) \sinh(\theta - v_1) \tau + 2(\theta^2 - v_2^2) \sinh(v_1 + \theta) \tau, \\
B_{13} &= B_{31} = 4\varepsilon_1(1(\nu) - \theta) \sinh(\theta - v_1) \tau + 4\varepsilon_1(1(\nu) + 2 + \theta) \sinh(\theta + v_1) \tau, \\
B_{21} &= B_{41} = B_{33} = B_{63} = 4\varepsilon_1 \sinh(\theta - v_1) \tau + 4\varepsilon_1 \sinh(v_1 + \theta) \tau, \\
B_{25} &= B_{62} = B_{64} = 2\varepsilon_1(1(\nu) + 2 + \theta) \sinh(v_1 + \theta) \tau + 2\varepsilon_1(1(\nu) + 2 + v_1 + \theta) \sinh(\theta - v_1) \tau + 2\varepsilon_1(1(\nu) + 2 + v_1 + \theta) \sinh(\theta - v_1) \tau, \\
B_{26} &= B_{45} = B_{34} = B_{62} = 4\varepsilon_1(1(\nu) + 2 + v_1 + \theta) \sinh(v_1 - \theta) \tau + 4\varepsilon_1(1(\nu) + 2 + v_1 + \theta) \sinh(v_1 - \theta) \tau.
\end{align*}

### III. QUANTUM CORRELATION

Let us first evaluate the degree of intensity-difference squeezing (DS) between the six beams. For the quantum correlated twin beams, the DS is given by the ratio of the variance on the intensity difference \(\text{Var}[\hat{N}_1 - \hat{N}_2]\) to the same variance at the SQL \(\text{Var}[\hat{N}_1 - \hat{N}_2]_{\text{SQL}} = \langle \hat{N}_1 + \hat{N}_2 \rangle\) [35]. In the hexapartite scenario, it is given by the ratio \(\text{DS}_6\) of the variance of a specific combination \(\delta\hat{N}\) to the same variance at the SQL. In the present configuration given by Fig. 1 the specific combination is \(\delta\hat{N} = \hat{N}_1 - \hat{N}_2 - \hat{N}_3 - \hat{N}_4 + \hat{N}_5 + \hat{N}_6\), so

\[
\text{DS}_6 = \frac{\text{Var}[\delta\hat{N}]_{\text{FWM}}}{\text{Var}[\delta\hat{N}]_{\text{SQL}}} = \frac{\text{Var}[\hat{N}_1 - \hat{N}_2 - \hat{N}_3 - \hat{N}_4 + \hat{N}_5 + \hat{N}_6]_{\text{FWM}}}{\langle \hat{N}_1 + \hat{N}_2 + \hat{N}_3 + \hat{N}_4 + \hat{N}_5 + \hat{N}_6 \rangle}. 
\]

Quantum correlation between the six beams will be present only when \(\text{DS}_6 < 1\).

Using the results of the previous section, we can find that

\[
\text{DS}_6 = \frac{4\theta^2}{\left[\langle (v_2 + \theta) \cosh(v_1 + \theta) \tau + (v_2 + \theta) \cosh(v_1 + \theta) \tau \rangle^2 \right. \\
\left. + 8\varepsilon_1^2 [\cosh^2(v_1 \tau) \sinh^2(\theta \tau) + \sinh^2(v_1 \tau) \sinh^2(\theta \tau)] \\
\left. + \langle (v_2 + \theta) \sinh(v_1 + \theta) \tau - (v_2 + \theta) \sinh(v_1 + \theta) \tau \rangle^2 \right]}
\]

As shown in Fig. 2, the quantum noise of \(\delta\hat{N}\) for six beams versus the interaction time \(t\) for the case of \(\varepsilon_1 = 1, \varepsilon_2 = 5,\) and \(\varepsilon_6 = 0.4,\) is always below the SQL since the value of DS6 is always smaller than 1. This can be simply understood as follows: It can be proved that the linear combination of the photon number operators of the six beams \(\delta\hat{N} = \hat{N}_1 - \hat{N}_2 - \hat{N}_3 - \hat{N}_4 + \hat{N}_5 + \hat{N}_6\) commute with the Hamiltonian in Eq. (5), and therefore can be as a constant of motion and the Casimir operator of the problem. That means that the fluctuations of this quantity involving the intensities of the six output beams
are exactly the same as the intensity fluctuations of the input seed beam. However, as many photons have been added to the six beams due to the FWM amplification process, the total optical power has been increased at the denominator \( \text{Var}[\delta N]_{\text{FWM}} \), without increasing the intensity-difference fluctuations at the numerator \( \text{Var}[\delta N]_{\text{FW}M} \). Therefore, the quantum correlation between the intensities of the six beams will increase with the parametric gain.

**IV. QUANTUM ENTANGLEMENT**

Now we turn to use our theoretical expressions to test the entanglement among the six beams. Among the various CV multipartite entanglement witnesses that have been proposed in the literature, the VLF and PPT criterion are most popular. Here we will use them successively in the following part of the present paper.

Let us first consider the VLF criterion. Hexapartite entanglement will be present if all the following inequalities are violated (see Eq. (43) in Ref. [4]):

\[
\begin{align*}
V_{12} &= V[X_1 - X_2] + V[Y_1 + Y_2 + g_3 Y_3 + g_4 Y_4 \\
&\quad + g_5 Y_5 + g_6 Y_6] \geq 4, \\
V_{26} &= V[X_2 - X_6] + V[Y_2 + Y_6 + g_1 Y_1 + g_3 Y_3 \\
&\quad + g_4 Y_4 + g_5 Y_5] \geq 4, \\
V_{63} &= V[X_6 - X_3] + V[Y_6 + Y_3 + g_1 Y_1 + g_2 Y_2 \\
&\quad + g_4 Y_4 + g_5 Y_5] \geq 4, \\
V_{35} &= V[X_3 - X_5] + V[Y_3 + Y_5 + g_1 Y_1 + g_2 Y_2 \\
&\quad + g_4 Y_4 + g_6 Y_6] \geq 4, \\
V_{54} &= V[X_5 - X_4] + V[Y_5 + Y_4 + g_1 Y_1 + g_2 Y_2 \\
&\quad + g_3 Y_3 + g_6 Y_6] \geq 4.
\end{align*}
\]

(22)

In the above inequalities, \( V_{ij} \) \((i = 1, \ldots, 6; j = 1, \ldots, 6)\) characterizes the correlation between the output beams, \( X_i = \hat{a}_i + \hat{a}_i^\dagger \) and \( Y_j = -i(\hat{a}_j - \hat{a}_j^\dagger) \) \((i = 1, \ldots, 6)\) are the amplitude and phase quadratures of the output beams, respectively. \( g_i \) \((i = 1, \ldots, 6)\) are any arbitrary real scaling factors, and appropriate \( g_i \) can be chosen to obtain the minimum value of the correlations.

As shown in Fig. 3, all the VLF correlations drop below 4 in the range of \( 0 < t < 0.17 \), thus hexapartite entanglement is present in such a range. Surprisingly, the criterion is not satisfied at the high parametric gain, where the intensity-difference correlation studied in the previous section is maximum. However, this does not mean that there is no entanglement in this range, as the VLF criterion is only sufficient but not necessary.

We now use a necessary and sufficient criterion for Gaussian states under certain conditions, i.e., the PPT criterion, to characterize the many possibilities of bipartite entanglement potentially existing in the system [36–41]. Following the PPT criterion, the symplectic eigenvalues can be computed as the eigenvalues of the matrix \( |\Omega \sigma | \), where \( \Omega = \Theta^T \xi \) is the symplectic transformation matrix and \( \sigma \) is the partial transposed (PT) covariance matrix (CM). Accordingly, there are 31 possible bipartitions for the hexapartite scenario needed to be tested. If the smallest symplectic eigenvalues for each of the 31 PT CMs are all smaller than 1, then the genuine hexapartite entanglement is present. Generally, the CM for the six beams can be written as

\[
\sigma = \begin{bmatrix}
\langle \hat{X}_1^2 \rangle & \langle \hat{X}_1 \hat{Y}_1 \rangle & \langle \hat{X}_1 \hat{X}_2 \rangle & \cdots & \cdots & \cdots \\
\langle \hat{Y}_1 \hat{X}_1 \rangle & \langle \hat{Y}_1^2 \rangle & \cdots & \cdots & \cdots & \cdots \\
\langle \hat{X}_2 \hat{X}_1 \rangle & \langle \hat{Y}_2 \hat{Y}_1 \rangle & \cdots & \cdots & \cdots & \cdots \\
\vdots & \vdots & \ddots & \ddots & \ddots & \cdots \\
\langle \hat{X}_6 \hat{Y}_6 \rangle & \langle \hat{Y}_6 \hat{X}_6 \rangle & \cdots & \cdots & \cdots & \langle \hat{Y}_6^2 \rangle \\
\end{bmatrix}
\]

(23)

And it can also be shown in Fig. 4(a) under the condition of \( \epsilon_1 = 1, \epsilon_2 = 5, \epsilon_6 = 0.4, \) and \( t = 0.1 \). As shown in Fig. 4(a), the term \( \langle \hat{X}_i \hat{Y}_j \rangle \) is equal to zero within the same or different beams, so the amplitude-phase correlation is nonexistent. Moreover, \( \langle \hat{X}_i \hat{X}_j \rangle = -\langle \hat{Y}_i \hat{Y}_j \rangle \) exists between any pairwise beams \( \hat{a}_i \) and \( \hat{a}_j \) with the same frequency, while \( \langle \hat{X}_i \hat{X}_j \rangle = -\langle \hat{Y}_i \hat{Y}_j \rangle \) exists between any pairwise beams \( \hat{a}_i \) and \( \hat{a}_j \) with the different frequencies. To test the entanglement properties of the system, the PT operation is applied to the partition made of any one beam, any two beams, and then any three beams, respectively.
FIG. 4. The CM for the cases of (a) $t = 0.1$, (c) $t = 0.4$, and (e) $t = 1$ with $\epsilon_1 = 1$, $\epsilon_2 = 5$, and $\epsilon_3 = 0.4$. The corresponding smallest symplectic eigenvalues of the 31 bipartitions are displayed in Figs. 4(b), 4(d), and 4(f), respectively. Region A, B, and C in Figs. 4(b), 4(d), and 4(f) display the smallest symplectic eigenvalues of all the 1 × 5, 2 × 4, and 3 × 3 bipartitions, respectively.

Results are shown in region A in Fig. 4(b). Numbers 1–6 of the horizontal axis label the bipartitions:

- $\tilde{a}_1(\tilde{a}_2, \tilde{a}_3, \tilde{a}_4, \tilde{a}_5, \tilde{a}_6)$,
- $\tilde{a}_2(\tilde{a}_1, \tilde{a}_3, \tilde{a}_4, \tilde{a}_5, \tilde{a}_6)$,
- $\tilde{a}_3(\tilde{a}_1, \tilde{a}_2, \tilde{a}_4, \tilde{a}_5, \tilde{a}_6)$,
- $\tilde{a}_4(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3, \tilde{a}_5, \tilde{a}_6)$,
- $\tilde{a}_5(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3, \tilde{a}_4, \tilde{a}_6)$,

and $\tilde{a}_6(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3, \tilde{a}_4, \tilde{a}_5)$, respectively. The smallest symplectic eigenvalues shown in region A in Fig. 4(b) are all smaller than 1, which means that all the six $1 \times 5$ bipartitions are fully inseparable.

Second, when the PT operation is applied to two beams [42], the results are shown in region B in Fig. 4(b). Numbers 7–21 of the horizontal axis label the bipartitions:

- $(\tilde{a}_1, \tilde{a}_2)(\tilde{a}_3, \tilde{a}_4, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_1, \tilde{a}_3)(\tilde{a}_2, \tilde{a}_4, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_1, \tilde{a}_4)(\tilde{a}_2, \tilde{a}_3, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_1, \tilde{a}_5)(\tilde{a}_2, \tilde{a}_3, \tilde{a}_4, \tilde{a}_6)$,
- $(\tilde{a}_2, \tilde{a}_3)(\tilde{a}_1, \tilde{a}_4, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_2, \tilde{a}_4)(\tilde{a}_1, \tilde{a}_3, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_2, \tilde{a}_5)(\tilde{a}_1, \tilde{a}_3, \tilde{a}_4, \tilde{a}_6)$,
- $(\tilde{a}_3, \tilde{a}_4)(\tilde{a}_1, \tilde{a}_2, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_3, \tilde{a}_5)(\tilde{a}_1, \tilde{a}_2, \tilde{a}_4, \tilde{a}_6)$,
- $(\tilde{a}_3, \tilde{a}_6)(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_4, \tilde{a}_5)(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3, \tilde{a}_4, \tilde{a}_6)$,
- $(\tilde{a}_4, \tilde{a}_6)(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3, \tilde{a}_4, \tilde{a}_5)$,
- $(\tilde{a}_5, \tilde{a}_6)(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3, \tilde{a}_4, \tilde{a}_5)$,

and $(\tilde{a}_5, \tilde{a}_6)(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3, \tilde{a}_4, \tilde{a}_5)$, respectively. As we can see, the smallest symplectic eigenvalues shown in region B in Fig. 4(b) are all smaller than 1, which means that all 15 $2 \times 4$ bipartitions are fully inseparable.

Finally, when the PT operation is applied to three beams, the results are shown in region C in Fig. 4(b). Numbers 22–31 of the horizontal axis label the bipartitions:

- $(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3)(\tilde{a}_4, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_1, \tilde{a}_2, \tilde{a}_4)(\tilde{a}_3, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_1, \tilde{a}_2, \tilde{a}_5)(\tilde{a}_3, \tilde{a}_4, \tilde{a}_6)$,
- $(\tilde{a}_1, \tilde{a}_3, \tilde{a}_4)(\tilde{a}_2, \tilde{a}_5, \tilde{a}_6)$,
- $(\tilde{a}_1, \tilde{a}_3, \tilde{a}_5)(\tilde{a}_2, \tilde{a}_4, \tilde{a}_6)$,
- $(\tilde{a}_1, \tilde{a}_4, \tilde{a}_5)(\tilde{a}_2, \tilde{a}_3, \tilde{a}_6)$,
- $(\tilde{a}_1, \tilde{a}_4, \tilde{a}_6)(\tilde{a}_2, \tilde{a}_3, \tilde{a}_5)$,
- $(\tilde{a}_2, \tilde{a}_3, \tilde{a}_5)(\tilde{a}_1, \tilde{a}_4, \tilde{a}_6)$,
- $(\tilde{a}_2, \tilde{a}_4, \tilde{a}_6)(\tilde{a}_1, \tilde{a}_3, \tilde{a}_5)$,

and $(\tilde{a}_1, \tilde{a}_2, \tilde{a}_3, \tilde{a}_4, \tilde{a}_5, \tilde{a}_6)$, respectively. Here again, the smallest symplectic eigenvalues shown in region C in Fig. 4(b)
are all smaller than 1, which means that all ten $3 \times 3$ bipartitions are fully inseparable.

So far, we have shown that the 31 smallest symplectic eigenvalues for the 31 bipartitions are all smaller than 1 under a certain interaction condition, which is a clear manifestation of hexapartite entanglement. To find how the interaction strength affects the hexapartite entanglement, we also study the cases of $t = 0.4$ and $t = 1.0$. The longer the interaction time is, the stronger the interaction strength will be. The CM and smallest symplectic eigenvalues for the case of $t = 0.4$ ($t = 1.0$) are shown in Figs. 4(c) and 4(d) (Figs. 4(e) and 4(f), respectively. From Fig. 4, the absolute values of each nonzero element including diagonal and off-diagonal parts in CMs increase with the increasing of the interaction time $t$. This is due to the fact that more interaction time means more amplification difference will become more significant with increasing interaction time. Therefore, the values of $(X_{2,3,4,5,6})$ and $(Y_{2,3,4,5,6})$ in Fig. 4(c) are larger than the corresponding ones in Fig. 4(a) but are much smaller than the ones of $(X_{1,3})$ and $(Y_{1,3})$ in Fig. 4(c). In ideal case without considering the experimental imperfections, the hexapartite entanglement should become stronger with increasing interaction time. In general, the stronger entanglement corresponds to the smaller symplectic eigenvalues. Therefore, with the increasing of the interaction time, the eigenvalues become smaller. In other words, the hexapartite entanglement is present even at high parametric gain, where the VLF criterion cannot witness the produced hexapartite entanglement.

V. SUPERMODES

The above PPT criterion gives the overall entanglement analysis of the SSP-based FWM process, while the underlying entanglement structure involved in the system is not disclosed very well. To further study the entanglement structure, it is worth studying the eigenmode decomposition [43] for the six beams. From the CM, the eigenmodes and eigenvalues for the case of $\epsilon_1 = 1$, $\epsilon_2 = 4$, $\epsilon_6 = 0.8$, and $t = 0.1$ can be obtained, which is shown in Fig. 5. It can be clearly seen that the six output beams can be decomposed into six independent supermodes, i.e., three squeezed modes and three antisqueezed modes. As shown in Fig. 5(a), the squeezing level of −3.94 dB can be obtained by the first squeezed eigenmode of $(0.661X_1 - 0.177X_2 - 0.661X_3 - 0.177X_4 + 0.177X_5 + 0.177X_6)$, which is almost in agreement with our previous experimental results [31]. Meanwhile, the squeezing level of −0.69 dB can also be obtained by the second squeezed eigenmode of $(0.500X_1 - 0.500X_2 - 0.500X_4 - 0.500X_5)$ as shown in Fig. 5(b). This is due to the existence of bipartite entanglement between beams $\hat{a}_2$ and $\hat{a}_6$ ($\hat{a}_4$ and $\hat{a}_5$). The eigenmode of $(0.500X_1 - 0.500X_2 + 0.500X_4 - 0.500X_5)$ is equivalent to the eigenmode of $(0.500X_1 - X_2 - X_4)$. The third squeezed eigenmode $-0.250X_1 - 0.468X_2 + 0.250X_3 - 0.468X_4 + 0.468X_5 + 0.468X_6$ gives a squeezing level of −0.23 dB. However, Figs. 5(d)–5(f) give the same squeezing level as Figs. 5(b) and 5(a), respectively, but on the phase quadrature. Their corresponding eigenmodes are $-0.250X_1 + 0.468X_2 - 0.250X_3 + 0.468X_4 + 0.468X_5 + 0.468X_6$, $0.500X_2 - 0.500X_3 - 0.500X_4 + 0.500X_5$, and $-0.661X_1 - 0.177X_2 - 0.661X_3 - 0.177X_4 - 0.177X_5 - 0.177X_6$, respectively. Similarly, to show a more richer supermode structure, the eigenvalues and eigenmodes for the case of $\epsilon_1 = 1$, $\epsilon_2 = 3.6$, $\epsilon_6 = 0.9$, and $t = 0.1$ are also displayed in Fig. 6.

VI. SUMMARY

We have theoretically characterized the quantum correlation and entanglement properties of the six beams generated by SSP based FWM process. Intensity-difference correlation exists in the whole parameter space and it is especially strong at high parametric gain. The hexapartite entanglement can be witnessed only in a certain interaction strength range with the
VLF criterion, whereas it is predicted at the high parametric gain with the PPT criterion. In addition, the eigenmode decomposition of the six beams are also analyzed and can be decomposed into six independent supermodes. The scheme we report here can also be readily extended to a much larger number of beams by changing the angle between the two pump beams [44,45] or shining more pump beams. Therefore, our results here will be useful not only to be confronted to future experimental data produced by an experimental setup, but also to investigate the use of such multipartite entanglement for scalable measurement-based quantum computing [16,17,46].

ACKNOWLEDGMENTS

This work was funded by the National Natural Science Foundation of China (NSFC) (Grants No. 11874155, No. 11804323, No. 91436211, No. 61673264, No. 61553012), the Natural Science Foundation of Shanghai (Grant No. 17ZR1442900), the Minhang Leading Talents (Grant No. 201971), Major Scientific Research Project of Zhejiang Lab (2019DE0KF01), the Program of Scientific and Technological Innovation of Shanghai (Grant No. 17JC1400401), Shanghai Municipal Science and Technology Major Project (Grant No. 2019SHZDZX01), the National Basic Research Program of China (Grant No. 2016YFA0302103), the 111 project (Grant No. B12024), the Fundamental Research Funds for the Central Universities, ECNU Academic Innovation Promotion Program for Excellent Doctoral Students (YBNL TS2019-012). C.F. and N.T. are members of the Institut Universitaire de France.

H.W. and K.Z. contributed equally to this work.