Directly produced three-color entanglement by quasi-phase-matched third-harmonic generation

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Abstract: A new scheme is presented to directly produce fundamental, second-, and third-harmonic three-color continuous-variable (CV) entangled beams by cascaded quasi-phase-matched third-harmonic generation (THG) in an optical cavity. THG can be achieved with high efficiency through a coupled sum-frequency process between the second-harmonic and the fundamental fields. It is demonstrated that the three beams (fundamental, second-, and third-harmonic fields) are entangled with each other according to the CV entanglement criterion. In this scheme, only one crystal and one pump field can generate three-color CV entangled beams separated by an octave in frequency through quasi-phase-matched cascaded nonlinear process, which may be very useful for the applications in quantum communication and computation networks.

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References and links
1. Introduction

Quantum entanglement attracts many interests in recent years since it is the central resource in the applications such as quantum communication and computation. Multipartite continuous-variable (CV) entangled beams with different frequencies are necessary to connect different physical systems at the nodes of quantum networks [1], which can facilitate many quantum information protocols of interspecies quantum teleportation. Two-color CV entanglement produced by nondegenerate optical parametric oscillator (OPO) was predicted [2], and experimentally demonstrated both below [3] and above [4] the oscillation threshold. Then, it was predicted that three-color CV entanglement among pump, signal and idler beams can be obtained using an OPO operating above the threshold [5], which also has been realized by the recent experiment [6]. Besides the above schemes, up-conversion, such as second-harmonic generation (SHG), has been suggested as a source of CV entanglement both in an optical cavity [7] and without optical cavity [8]. It was predicted that the perfect entanglement could be produced between the fundamental and the second-harmonic fields by the second-order nonlinear interaction [9], which has been experimentally verified recently [10]. Other suggestions were proposed to generate two-color tripartite entanglement among the fundamental and the second-harmonic fields through the type-II SHG system operating below [11] and above [12] the threshold. In addition, multicolored tripartite CV entanglement generated in a two-port resonator of SHG process was also investigated by applying a necessary and sufficient entanglement criterion [13].

Quasi-phase-matching (QPM) makes nonlinear wave mixing easier in a more general stage through phase compensation provided by the reciprocal vectors of crystal lattice [14]. Efficient SHG was experimentally achieved in a Fibonacci quasi-periodic optical superlattice (QPOS) LiTaO₃ by using QPM method [15]. Third-harmonic generation (THG) can also be obtained with high efficiency through cascaded sum-frequency process between the second-harmonic and the fundamental fields [16]. Based on that previous experiment, here, we propose a new scheme to directly produce three-color CV entangled beams (i.e. fundamental, second-, and
third-harmonic fields) through a coupled nonlinear process of quasi-phase-matched THG. The system consists of a QPOS LiTaO$_3$ placed inside an optical cavity, as shown in Fig. 1(a). The fundamental wave with the frequency of $\omega_0$ is incident upon the QPOS in the cavity. The second-harmonic field with the frequency of $\omega_1$ is generated by the first sum-frequency process. The third-harmonic field with the frequency of $\omega_2$ is generated by the second sum-frequency process between the fundamental and the second-harmonic fields. The phase mismatches in the two nonlinear processes are compensated by two different reciprocal vectors $G_1$ and $G_2$ of the QPOS, respectively. The QPM conditions can be written as

$$k_1 = 2k_0 + G_1$$

for the SHG and

$$k_2 = k_0 + k_1 + G_2$$

for the THG, respectively, which are depicted in Fig. 1(b). $k_0$, $k_1$, and $k_2$ are the wave vectors of the fundamental, second-, and third-harmonic fields, respectively.

![Fig. 1. (a)Sketch of the optical cavity. (b)The sketched momentum geometry for coupled quasi-phase-matching processes.](image)

2. The stationary solutions and output fields

The interaction Hamiltonian for this quasi-phase-matched THG can be written as

$$H_I = i\hbar \kappa_0 a_0^\dagger a_1 + i\hbar \kappa_1 a_0 a_2^\dagger + h.c.,$$

(1)

where $\kappa_0$ and $\kappa_1$ are the dimensionless nonlinear coupling coefficients, which are taken to be real for simplicity.

The cavity pumping is

$$H_{pump} = i\hbar (\varepsilon a_0^\dagger - \varepsilon^* a_0),$$

(2)

where $\varepsilon$ is the classical pumping laser amplitude and taken as a real field here for $\varepsilon = \varepsilon^* = E_0$.

The losses of the three modes are given by

$$\Lambda_i \rho = \gamma_i (2a_i \rho a_i^\dagger - a_i^\dagger a_i \rho - \rho a_i^\dagger a_i),$$

(3)

where $\gamma_i$ ($i = 0, 1, 2$) stand for the damping rates of the three cavity fields.

We proceed by mapping the master equation onto a set of stochastic differential equations by the Fokker-Planck equation in the positive-$P$ representation [17]. We find the Fokker-Planck...
away the noise terms in Eq. (5). The steady-state value should be noted that increment [17].

\[
\frac{dp}{dt} = \left\{ -\frac{\partial}{\partial \alpha_0} [e^{-2\kappa_0 \alpha_0} \alpha_1 - \kappa_1 \alpha_1^* \alpha_2 - \gamma_1 \alpha_0] - \frac{\partial}{\partial \alpha_0'} [e^{-2\kappa_0 \alpha_0'} \alpha_1^* - \kappa_1 \alpha_1 \alpha_2^* - \gamma_0 \alpha_0'] \\
- \frac{\partial}{\partial \alpha_1} [\kappa_0 \alpha_0^2 - \kappa_1 \alpha_0 \alpha_2 - \gamma_1 \alpha_1] - \frac{\partial}{\partial \alpha_1^*} [\kappa_0 \alpha_0'^2 - \kappa_1 \alpha_1^* \alpha_2^* - \gamma_1 \alpha_1^*] \\
- \frac{\partial}{\partial \alpha_2} [\kappa_1 \alpha_0 \alpha_1 - \gamma_2 \alpha_2] - \frac{\partial}{\partial \alpha_2^*} [\kappa_1 \alpha_0^* \alpha_1^* - \gamma_2 \alpha_2^*] \\
+ \frac{1}{2} \frac{\partial^2}{\partial^2 \alpha_0} [-2\kappa_0 \alpha_1] + \frac{1}{2} \frac{\partial^2}{\partial \alpha_0 \alpha_1} [-2\kappa_0 \alpha_1^*] + \frac{1}{2} \frac{\partial^2}{\partial \alpha_0 \alpha_1} [-2\kappa_1 \alpha_2] + \frac{1}{2} \frac{\partial^2}{\partial \alpha_0' \alpha_1^*} [-2\kappa_1 \alpha_2^*] \right\} P_{ij}. \quad (4)
\]

where \( \alpha_i (i = 0, 1, 2) \) correspond to \( a_i \) in the \( P \) representation, respectively. The above Fokker-Planck equation does not possess a positive-definite diffusion matrix. So we must double the phase space and use the positive-\( P \) representation to find the appropriate stochastic differential equations. \( \alpha^* \) will be substituted by \( \alpha^\dagger \) in the stochastic differential equations. However, it should be noted that \( \alpha \) and \( \alpha^\dagger \) are independent complex variables in the positive-\( P \) representation [17]. Following the standard procedure [17], the stochastic differential equations for the cavity fields can be written as

\[
\begin{align*}
\frac{d\alpha_0}{dt} &= e^{-2\kappa_0 \alpha_0} \alpha_1 - \kappa_1 \alpha_1^* \alpha_2 - \gamma_1 \alpha_0 + \sqrt{-\kappa_0 \alpha_0} \eta_1 + \sqrt{-\kappa_1 \alpha_0} \eta_2 \\
\frac{d\alpha_1}{dt} &= e^\ast -2\kappa_0 \alpha_0 \alpha_1^\dagger - \kappa_1 \alpha_1 \alpha_2^\dagger - \gamma_1 \alpha_0^\dagger + \sqrt{-\kappa_0 \alpha_0^\dagger} \eta_1^\dagger + \sqrt{-\kappa_1 \alpha_1} \eta_3^\dagger \\
\frac{d\alpha_2}{dt} &= \kappa_0 \alpha_0 - \kappa_1 \alpha_0 \alpha_2 - \gamma_1 \alpha_1 + \sqrt{-\kappa_0 \alpha_0} \eta_1 \\
\frac{d\alpha_2^\dagger}{dt} &= \kappa_0 \alpha_0^\dagger - \kappa_1 \alpha_0^\dagger \alpha_2^\dagger - \gamma_1 \alpha_1^\dagger + \sqrt{-\kappa_0 \alpha_0^\dagger} \eta_1^\dagger \\
\end{align*}
\]

\( \eta_i (i = 1, 2, 3) \) are the complex noise terms, which satisfy the relations \( \langle \eta_i(t) \rangle = 0 \) and \( \langle \eta_i(t) \eta_j(t') \rangle = \delta_{ij} \delta(t-t') \).

The steady-state solutions can be obtained by letting \( \frac{d\alpha_i}{dt} = 0 \) \( (i = 0, 1, 2) \) and throwing away the noise terms in Eq. (5). The steady-state value \( A_0 \) for the fundamental field satisfies the following equation

\[
\Delta_0 \alpha_1^2 + \kappa_0^2 \gamma_2 A_0^3 (3\Delta_1 - \gamma_1 \gamma_2) = 0,
\]

where \( \Delta_0 = \gamma_0 A_0 - E_0, \Delta_1 = \kappa_1^2 A_0^3 + \gamma_1 \gamma_2 \). The other steady-state values are related to \( A_0 \) as

\[
\begin{align*}
A_1 &= A_0 [\kappa_0^2 \gamma_2 A_0 (2\gamma_1 \gamma_2 + 3\Delta_1) - \kappa_1^2 \Delta_0 \Delta_1] / \kappa_0^2 \gamma_2^2, \\
A_2 &= \kappa_1 A_0 A_1 / \gamma_2,
\end{align*}
\]

where \( A_i (i = 1, 2) \) are the steady-state values of the second- and third-harmonic fields, respectively.

In the following, one can decompose the system variables into their steady-state values and small fluctuations around the steady-state values as \( \alpha_i = A_i + \delta \alpha_i \) and use this linearization procedure to rewrite the Eq. (5) as [17, 18]

\[
d \delta \dot{\alpha} = -A \delta \dot{\alpha} dt + B dW, \quad (8)
\]

where \( \delta \dot{\alpha} = [\delta \alpha_0, \delta \alpha_0^\dagger, \delta \alpha_1, \delta \alpha_1^\dagger, \delta \alpha_2, \delta \alpha_2^\dagger]^T; A \) is the drift matrix with the steady-state values; \( B \) contains the steady-state coefficients of the noise terms of Eq. (5); \( dW \) is a vector of Wiener increment [17].
Then, one can obtain the intracavity spectral matrix in the frequency domain as [17, 18]

\[ S(\omega) = (A + i\omega I)^{-1}BB^{\dagger}(A^{\dagger} - i\omega I)^{-1}, \]

where \( I \) is the identity matrix and \( \omega \) is the analysis frequency. The output fields can be obtained by applying the well-known input-output relations [19].

However, the condition for the validity of the linearization process is that the eigenvalues of the drift matrix \( A \) have no negative real parts [18]. In Fig. 2 we plot the real parts of the eigenvalues (RPEA) versus (a) \( \sigma = E_{0}/\epsilon_{th} \), (b) \( \kappa_{1}/\kappa_{0} \), (c) \( \gamma/\gamma_{0} \), and (d) \( \gamma_{2}/\gamma_{1} \), respectively, where \( \gamma = \gamma_{1} = \gamma_{2} \) for simplicity; \( \sigma = E_{0}/\epsilon_{th} \) and \( \epsilon_{th} = \gamma_{0}/\kappa_{0} \) is the well-known threshold of OPO which used here as a terminology to investigate entanglement for different values of \( E_{0} \).

3. Discussions on the entanglement characteristic among three beams

There are two nonlinear sum-frequency processes in quasi-phase-matched THG. In the first sum-frequency process, when two fundamental photons are destroyed, one second-harmonic photon is created. So the intensity of the fundamental field is anticorrelated to that of the second-harmonic field. Similarly, in the second cascaded sum-frequency process, when one second-harmonic photon and one fundamental photon are destroyed, one third-harmonic photon is created. So the intensity of the third-harmonic field is anticorrelated to that of the second-harmonic field, as well as that of the fundamental field. Finally, the three fields are entangled with each other and bright three-color entanglement can be obtained in this scheme.

A necessary and sufficient criterion for bipartite Gaussian states CV entanglement was proposed [20] based on the positivity of the partial transpose (PPT) [21, 22]. Then, necessary and sufficient criteria for \( 1 \times N \) bipartite multi-mode Gaussian states entanglement [23] and tripartite three-mode Gaussian states [24] were also developed, respectively. According to these criteria, the correlation matrix (CM) can be defined as [25] \( \sigma_{k’k} = \mu_{k} \mu_{k’} + \mu_{k}^{*} \mu_{k’} > /2 \), where

\[ \mu = (X_{0}, Y_{0}, X_{1}, Y_{1}, X_{2}, Y_{2}) \] and

\[ \tilde{X}_{i} = (\alpha_{i}^{\text{out}} + \alpha_{i}^{\text{out}}^{*}) (i = 0, 1, 2) \] represent the amplitude quadratures of the fields; \( \tilde{Y}_{i} = -i(\alpha_{i}^{\text{out}} - \alpha_{i}^{\text{out}}^{*}) \) stand for their phase quadratures. The Heisenberg uncertainty
principle is [20]

\[ \sigma + i\Omega \geq 0, \]  

(10)

where

\[ \Omega = \left( \begin{array}{ccc} J & 0 & 0 \\ 0 & J & 0 \\ 0 & 0 & J \end{array} \right), J = \left( \begin{array}{cc} 0 & 1 \\ -1 & 0 \end{array} \right). \]  

(11)

Above uncertainty relation (10) can be replaced by [26]

\[ \nu_i \geq 1, \]  

(12)

where \( \nu_i \) is the symplectic eigenvalues. We denote \( \tilde{\sigma}_A \) as the partial transpose on the CM \( \sigma \) for subsystem \( A \) and \( E_A \) is the symplectic eigenvalues of the matrix \( |i\Omega\tilde{\sigma}| \). Then the subsystems \( A \) and \( B \) will be inseparable (i.e. entangled with each other) when [26]

\[ E_A < 1. \]  

(13)

According to the criterion for tripartite entanglement [24], the three fields are entangled when all conditions for two parties entanglement are satisfied. In present three-mode system, we denote \( E_i \) \((i = 0, 1, 2)\) as the symplectic eigenvalues of the bipartite system between the field \( a_i \) and the subsystem including the remaining fields \((1 \times 2 \) bipartition). When \( E_i < 1 \) are all satisfied simultaneously, the three fields are entangled with each other. In the following, we will employ this necessary and sufficient CV entanglement criterion to discuss the quantum entanglement features among the three fields. Smaller of the symplectic eigenvalue is, larger of the degree of entanglement will be. Figure 3(a) depicts the symplectic eigenvalues \( E_i \) versus analysis frequency \( \omega \) with \( \gamma_0 = 0.01, \gamma = 1.5\gamma_0, \kappa_0 = 0.1, \kappa_1 = 1.5\kappa_0, \) and \( \sigma = 2 \). One can see in Fig. 3(a) that the three values of \( E_i \) are all below 1 simultaneously in a wide range of the analysis frequency which clearly indicates that the fundamental, second-, and third-harmonic fields are CV entangled with each other.

The conversion efficiency of sum-frequency process is much higher than that of the parametric process and the sum-frequency generation does not have threshold. In the single pass experiment in Ref. [16], the conversion efficiency of THG is about 23%. If we put this setup into a cavity, the efficiency will be farther increased. So the threshold for the cavity is very smaller than that of OPO. Bright three-color entanglement can be easy obtained in this scheme. Equation (6) is a fifth-order equation, it is well known, which has no analytic solution. Therefore, we can not obtain the threshold for the cavity. However, we will continue to use the threshold of OPO as a terminology to investigate entanglement among the three fields for different values of \( E_0 \). In Fig. 3(b) the symplectic eigenvalues \( E_i \) are presented versus \( \sigma \) with \( \gamma_0 = 0.01, \gamma = 1.5\gamma_0, \kappa_0 = 0.1, \kappa_1 = 1.5\kappa_0, \) and \( \omega = \gamma_0 \). Form Fig. 3(b) one can see that the three eigenvalues are below 1 which indicates the three fields are entangled with each other below and above the threshold. From Fig. 2 one can see that the linearization is valid and the three fields are entangled with each other in this range.

In quasi-phase-matched THG process, the nonlinear coupling parameters are related to the structure parameters of QPOS which can be adjusted in the design. Figure 3(c) depicts the symplectic eigenvalues versus the nonlinear coupling parameters \( \kappa_1/\kappa_0 \) with \( \gamma_0 = 0.01, \gamma = 1.5\gamma_0, \kappa_0 = 0.1, \sigma = 2, \) and \( \omega = \gamma_0 \). From Fig. 3(c) one can see that the three symplectic eigenvalues are all below 1 in this range and the best three-color entanglement can be obtained when \( \kappa_1 > \kappa_0 \). In fact, the previous experimental results indicated that a small mismatch for the first sum-frequency process (i.e. \( \kappa_1 > \kappa_0 \)) will increase the conversion efficiency of the THG [16].
We have presented a new scheme to directly produce fundamental, second-, and third-harmonic entangled beams by quasi-phase-matched THG. Three-color CV entanglement among the fundamental, second-, and third-harmonic beams are demonstrated by applying a necessary and sufficient tripartite CV entanglement criterion. In this scheme, only one crystal and one pump field can generate bright three-color CV entangled beams separated by an octave in frequency, which is very different from the scheme reported in Ref. [6] and may be very useful for the applications in quantum communication and computation networks.

4. Conclusions

Figure 3(d) depicts the symplectic eigenvalues versus the damping rates $\gamma/\gamma_0$ with $\gamma_0 = 0.01$, $\kappa_0 = 0.1$, $\kappa_1 = 1.5\kappa_0$, $\sigma = 2$, and $\omega = \gamma_0$. One can see that the three fields are entangled with each other and better three-color entanglement can be obtained at about $\gamma/\gamma_0 = 3.1$. Actually, better squeezed state of the fundamental field can be obtained when the damping rate $\gamma_0$ is smaller [18].

In the above calculations, we set $\gamma = \gamma_1 = \gamma_2$ for simplicity. In Fig. 3(e) we plot the symplectic eigenvalues versus different values of $\gamma_1$ and $\gamma_2$ with $\gamma_0 = 0.01$, $\gamma_1 = 1.5\gamma_0$, $\kappa_0 = 0.1$, $\kappa_1 = 1.5\kappa_0$, $\sigma = 2$, and $\omega = \gamma_0$. One can see that the three fields are also entangled for different values of $\gamma_1$ and $\gamma_2$ and better three-color entanglement can be obtained at about $\gamma_2 = 1.6\gamma_1$.

Figure 3. The symplectic eigenvalues versus (a) $\omega$, (b) $\sigma$, (c) $\kappa_1/\kappa_0$, (d) $\gamma/\gamma_0$, and (e) $\gamma_2/\gamma_1$, respectively.
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