Cauchy-Schwarz characterization of tripartite quantum correlations in an optical parametric oscillator

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We analyze the three-mode correlation properties of the electromagnetic field in an optical parametric oscillator below threshold. We employ a perturbative expansion of the Itô equations derived from the positive-P representation of the density matrix. Using the generalized Cauchy-Schwarz inequality, we investigate the genuine quantum nature of the triple correlations between the interacting fields, in case this continuous variable entanglement is not detected by the van Loock-Furusawa criterion [Phys. Rev. A 67, 052315 (2003)]. Although not being a necessary condition, these triple correlations are a sufficient evidence of tripartite entanglement. Of course, our characterization of the quantum correlations is applicable to non-Gaussian states, which we show to be the case of the optical parametric oscillator below threshold, provided nonlinear quantum fluctuations are properly taken into account.

The nonlocal character of quantum mechanics has been widely exploited to develop quantum information protocols [1] sometimes implemented in laboratories [2], or even for commercial purposes as has occurred with quantum cryptography [3]. Bipartite entangled states are the simplest resource for such protocols and can be generated by optical parametric oscillators (OPOs) [4–7]. However, more sophisticated protocols may require entanglement of many parts [8, 9]. The next difficulty is trying to find a source of tripartite entanglement. The first idea in this sense came from Greenberger, Horne, and Zeilinger [10], suggesting the GHZ state, a discrete variable (spin variables) tripartite entangled state detectable by triple product spin measurements, but actually very difficult to implement. However the implementation of this idea by using continuous variables such as quadratures of optical fields, seems quite feasible with the available homodyning techniques. Nowadays, there are a few devices that can generate multipartite entangled states, and the OPO has been explored for this task [11].

The nonlinear interaction between the OPO modes suggests the possibility of tripartite entanglement between the three modes, and also the possibility of controlling the bipartite correlations between signal and idler only by handling a third part, the pump mode. The quantum correlations between the different parts are supposed to change depending on statistics, spatial profile and other possible characteristics of the pump input, as observed in many experiments based on parametric down conversion [12]. Indeed, tripartite pump-signal-idler entanglement has been predicted in the intense regime, above the OPO threshold [13] and experimentally generated [14]. Tripartite correlations were analyzed according to the criteria proposed by van Loock and Furusawa [15], based on pair correlations among the three modes. Also, there are other experiments that generate continuous-variable tripartite entangled states obtained by mixing squeezed vacua with linear optical elements [16].

However, there has been little attention to tripartite correlations in the OPO operating below threshold, but it was in this context that triple quadrature correlations were proposed to be measured as means to compare classical and quantum correlations [17]. The experimental difficulty resides in homodyning the weak down converted beams, which is not a problem for intense beams generated above threshold, since a smart technique of self-homodyning [13] can be used. It is also important to notice that it was in the below threshold regime where squeezed vacuum was first produced, and EPR correlations were first detected [18, 19].

In this work, we calculate, using a perturbative approach, all triple quadrature correlations between the interacting fields in the OPO below threshold. These triple correlations should vanish for Gaussian quantum states, whether they are entangled or not, so that the van Loock and Furusawa entanglement criterion is well suited. This is the case, for example, of the OPO above threshold [14]. However, we shall demonstrate that triple quadrature correlations are predicted for below threshold operation when the perturbative approach is taken beyond the linear approximation. While evidencing non-Gaussian behavior, these triple correlations are also sufficient conditions for tripartite entanglement. We also show that, in this case, the van Loock-Furusawa criterion fails to evidence entanglement. Due to this inconclusive condition, and the lack of a criterion for this case, we also show that certain correlations violate the tripartite Cauchy-Schwarz inequality expected for classical c-number variables.

The OPO can be modeled by three modes coupled through a nonlinear crystal inside a lossy Fabry-Perot cavity, described by the following master equation in the Born-Markov approximation [20]:

$$\frac{\partial \hat{\rho}}{\partial t} = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}] + \sum_{i=0}^{2} \gamma_i \left( 2\hat{a}_i \hat{\rho} \hat{a}_i^\dagger - \hat{a}_i^\dagger \hat{a}_i \hat{\rho} - \hat{\rho} \hat{a}_i^\dagger \hat{a}_i \right), \quad (1)$$
where

\[
\hat{H} = \sum_{i=0}^{2} \hbar \omega_i \hat{a}_i^\dagger \hat{a}_i + \hbar \chi \left( \hat{a}_1^\dagger \hat{a}_3^\dagger \hat{a}_0 - \hat{a}_1 \hat{a}_2^\dagger \hat{a}_0^\dagger \right) + \hbar \left( E e^{-i \omega_0 t} \hat{a}_1^\dagger - E^* e^{i \omega_0 t} \hat{a}_0^\dagger \right).
\]  (2)

The parameter $E$ represents the external pump field at frequency $\omega_0$. The pump, signal and idler fields are represented by the annihilation operators $\hat{a}_0$, $\hat{a}_1$ and $\hat{a}_2$, respectively, and satisfy the frequency match condition $\omega_0 = \omega_1 + \omega_2$. $\chi$ is the nonlinear coupling and $\gamma_i$ ($i = 0, 1, 2$) are amplitude damping rates. Now, the density matrix equation of motion can be transformed into c-number Fokker-Planck or stochastic equations by means of operator representation theory.

The quantum evolution of the system can be described by the generalized P-representation \cite{21}, by expanding the density matrix in a off diagonal coherent state basis, defined as

\[
\hat{\rho} = \int_D |\alpha\rangle \langle\alpha| P(\alpha, \alpha^+)^{d\alpha} d^6 \alpha^+, \quad (3)
\]

where $\alpha \equiv (\alpha_0, \alpha_1, \alpha_2)$ and $\alpha^+ \equiv (\alpha_0^+, \alpha_1^+, \alpha_2^+)$ are complex variables that run independently over all complex plane. The function $P(\alpha, \alpha^+)$ can be understood as a positive distribution in a double phase space that satisfies a Fokker-Planck equation. The Itô stochastic equations derived from the Fokker-Planck equation, after transforming to the rotating-wave frame, are

\[
\begin{align*}
d\alpha_0 &= (E - \gamma_0 \alpha_0 - \chi \alpha_1 \alpha_2) \, dt \\
d\alpha_0^+ &= (E^* - \gamma_0 \alpha_0^+ - \chi \alpha_1^+ \alpha_2^+) \, dt \\
d\alpha_j &= (-\gamma_j \alpha_j + \chi \alpha_k \alpha_0) \, dt + (\chi \alpha_0)^{1/2} dW_j \\
d\alpha_j^+ &= (-\gamma_j \alpha_j^+ + \chi \alpha_k \alpha_0^+) \, dt + (\chi \alpha_0^+)^{1/2} dW_j^+, \quad (4)
\end{align*}
\]

where $j = 1, 2$; $k = 1, 2$ and $j \neq k$. The Wiener increments satisfy $\langle dW_j \, dW_k \rangle = \langle dW_j^+ \, dW_k^+ \rangle = dt$, and all other correlations vanish.

In order to analyze the correlations between the modes, we proceed by applying the perturbation theory for the three modes below threshold, where this approach is valid. Around the threshold, the quantum fluctuations become huge and break the perturbative analysis. Following the physical situation found in most experiments, we shall assume a common damping rate for the down converted fields: $\gamma_1 = \gamma_2 = \gamma$. It is also useful to define a dimensionless coupling constant $g = \chi/(\gamma \sqrt{2} \gamma_r)$, where $\gamma_r \equiv \gamma_0/\gamma$. In order to employ a perturbative approach and to decouple the dynamical equations, it is convenient to define the following normalized variables \cite{22}:

\[
\begin{align*}
x &= \sqrt{2} \gamma_r \frac{g}{i} (\alpha_0 + \alpha_2^+) = \sqrt{2} \gamma_r X_0 \\
y &= \sqrt{2} \gamma_r \frac{1}{i} (\alpha_0 - \alpha_2^+) = \sqrt{2} \gamma_r Y_0
\end{align*}
\]

\[
x = g (\alpha_1 + \alpha_2^+) = g X \\
y = \frac{1}{i} (\alpha_1 - \alpha_2^+) = g Y. \quad (5)
\]

Note that in positive-P representation the quadrature variables $x$ and $y$ are not real and their definitions are accompanied by the corresponding phase space conjugates $x^+$ and $y^+$. We now expand these variables in terms of the dimensionless coupling constant \cite{23, 24}.

\[
x_0 = \sum_{n=0}^{\infty} g^n x_0(n) = x_0^{(0)} + g x_0^{(1)} + g^2 x_0^{(2)} + \ldots, \quad (6)
\]

and analogous expansions for $y_0, x, y, x^+$ and $y^+$.

Below threshold, the zero-order steady state solution for the quadratures are given by

\[
\begin{align*}
x_0^{(0)} &= x_0^{(1)} = y_0^{(0)} = y_0^{(1)} = 0 \\
x_0^{(2)} &= 2 \mu
\end{align*}
\]

where $\mu = \chi E/(\gamma \gamma_0)$ is the dimensionless pump parameter. These are the mean values of the fields below threshold. As expected, at this regime all mean field quadratures are zero, since only fluorescence is present. The pumped quadrature is the only one having a macroscopic value. The first quantum correction comes from the next order contribution:

\[
\begin{align*}
x_1^{(1)}(t) &= \sqrt{2} \mu \int_{-\infty}^{t} e^{-(1-\mu)(t-t')} \xi_x(t') \, dt' \\
y_1^{(1)}(t) &= -i \sqrt{2} \mu \int_{-\infty}^{t} e^{-i(1+\mu)(t-t')} \xi_y(t') \, dt' \\
x_0^{(2)}(t) &= -\gamma_r \int_{-\infty}^{t} e^{-\gamma_r(t-t')} \left( x_1^{(1)}(t') x_0^{(1)}(t') - y_1^{(1)}(t') y_0^{(1)}(t') \right) \, dt' \\
y_0^{(2)}(t) &= -\gamma_r \int_{-\infty}^{t} e^{-\gamma_r(t-t')} \left( x_1^{(1)}(t') y_0^{(1)}(t') + y_1^{(1)}(t') x_0^{(1)}(t') \right) \, dt', \quad (8)
\end{align*}
\]

where $\langle \xi_x(t) \xi_x(t') \rangle = \langle \xi_y(t) \xi_y(t') \rangle = \delta(t-t')$. Note that we kept the second order perturbation term for the pumped mode variables to account for pump depletion, since their first order corrections vanish. Therefore, pump depletion is a second order effect below threshold. It is also interesting to note that the second order corrections in the quadrature variables of the pumped mode depend on the product of two correlated Gaussian processes. This implies that double correlations between the pump and any one of the down converted modes vanish. As a result, a criterion based on pair correlations for tripartite entanglement, like the one by van Loock and Furusawa \cite{13}, fails to detect the existing triple correlations which are quantum mechanical in essence, as we shall see shortly. Moreover, the second order correction
The non vanishing triple correlations up to fourth order since in the positive-P representation the incoming field actually these results are also the output cavity fields, relations calculated in normal order, is presented below. Due to this property, the three-mode correlations unveil most of the important result addressed by this paper.

The results for the steady state time ordered triple correlations calculated in normal order, is presented below. Actually these results are also the output cavity fields, since in the positive-P representation the incoming field (vacuum) is not correlated with the intracavity modes. The non vanishing triple correlations up to fourth order in the coupling constant are:

\[
\begin{align*}
\langle \Delta x \Delta x^+ \Delta x_0 \rangle &= -g^4 \left( \frac{\mu}{1-\mu} \right)^2 \left[ \frac{2}{1+\mu} \right. \\
&\quad + \frac{\gamma_r}{\gamma_r + 2(1-\mu)} \left. \right] \\
\langle \Delta y \Delta y^+ \Delta x_0 \rangle &= g^4 \left( \frac{\mu}{1+\mu} \right)^2 \left[ \frac{2}{1-\mu} \right. \\
&\quad + \frac{\gamma_r}{\gamma_r + 2(1+\mu)} \left. \right] \\
\langle \Delta y \Delta x^+ \Delta y_0 \rangle &= g^4 \left( \frac{\mu^2}{1-\mu^2} \right)^2 \left( \frac{\gamma_r}{2 + \gamma_r} \right) \\
\langle \Delta x \Delta y^+ \Delta y_0 \rangle &= g^4 \left( \frac{\mu^2}{1-\mu^2} \right)^2 \left( \frac{\gamma_r}{2 + \gamma_r} \right), 
\end{align*}
\]

where \( \Delta u \equiv u - \langle u \rangle \). One can easily show that for any kind of separable state of the form: \( \sum p_i \rho_k \otimes \rho_{im} \), where \( \{ k, l, m \} \) is any permutation of \( \{ 0, 1, 2 \} \), at least one of these triple correlations should vanish identically. Therefore, the set of triple correlations given by eq.9 is a sufficient condition for tripartite entanglement. However, it does not exclude classical correlations [23], so that further analysis is required to evidence genuine quantum behavior.

Let us now turn to a reliable classicality criterion based on Cauchy-Schwarz inequalities. First, we calculate the average product of the down converted squared fluctuations:

\[
\begin{align*}
\langle (\Delta x^2 + \Delta y^2) (\Delta x^+)^2 + (\Delta y^+)^2 \rangle &= 2 g^4 \mu^2 \left( \frac{1}{(1-\mu)^2} + \frac{1}{(1+\mu)^2} \right). 
\end{align*}
\]

The fluctuations on \( x_0 \) are given by

\[
\begin{align*}
\langle (\Delta x_0)^2 \rangle &= g^4 \left( \frac{\mu}{1-\mu} \right)^2 \left( \frac{2}{1+\mu} \right)^2 \\
&+ \left[ 1 + \left( \frac{1-\mu}{1+\mu} \right)^2 \right] \frac{\gamma_r^2}{\gamma_r^2 + 4(1-\mu)^2},
\end{align*}
\]

while \( \langle (\Delta y_0)^2 \rangle \) is negligible. Now we establish a relation between correlations (9) and intensity correlations. To begin with, we assume that for any three complex numbers the following relation holds

\[
\langle \alpha_1 \alpha_2 + \lambda^* \alpha_0 \rangle^2 \geq 0,
\]

so that

\[
\langle \alpha_1^* \alpha_1 \alpha_2^* \alpha_2 \rangle + \lambda^2 \langle \alpha_1^* \alpha_0 \rangle + \lambda \langle \alpha_1 \alpha_2 \alpha_0 \rangle + \lambda^* \langle \alpha_1^* \alpha_2^* \alpha_0^* \rangle \geq 0.
\]

This expression is minimized for \( \lambda = \langle \alpha_1^* \alpha_2^* \alpha_0^* \rangle / \langle \alpha_0^* \alpha_0 \rangle \), giving rise to the following inequality:

\[
\langle \alpha_1^* \alpha_1 \alpha_2^* \alpha_2 \rangle \geq \langle \alpha_1 \alpha_2 \alpha_0 \rangle^2. \tag{14}
\]

In the same way, similar inequalities can also be derived by permutation of the indices in the complex variables \( \alpha_j \). After a straightforward algebra, it is easy to show that the triple correlation appearing in the right hand side of inequality (14) is simply the squared sum of all triple correlations given by eq.9, taking the first one \( \langle (\Delta x \Delta x^+ \Delta x_0) \rangle \) with an inverted sign. Also, the left hand side of the inequality is simply the product of the averages given by eqs.10 and 11.

Using the results given by eqs.9, 10, and 11 on both sides of the Cauchy-Schwarz inequality (14), we can verify its violation for the three-mode state produced by the OPO operating below threshold, which means that these triple correlations are essentially quantum mechanical. The reliability of the analytical calculations can be checked by numerical simulation of the full set of stochastic equations 1, with any perturbative approximation. Fig.1 compares both sides of inequality (14), showing a clear violation of the inequality for large \( \gamma_r \). The numerical results are in very good agreement with the analytical calculations. In our numerical calculations we used \( g = 0.0071 \) (\( \chi = 0.01 \)) whose order of magnitude corresponds to most nonlinear media used for parametric down conversion. Note that the violation of Cauchy-Schwarz inequality occurs with very small correlation values (\( \sim 10^{-4} \)). This is due to the rather small value of the coupling constant. Since these correlations scale as \( g^2 \), this result can be substantially improved with new materials providing significant values of the nonlinear susceptibility. Indeed, we checked this numerically.

In conclusion, we investigated the triple correlations produced in an OPO below threshold, evidencing tripartite entanglement not detected using the well known van
Loock-Furusawa criterion. In order to circumvent this limitation, we employed a non classicality criterion based on general Cauchy-Schwartz inequalities for third order moments. This approach was applicable due to the non Gaussian character of the quantum fluctuations. Moreover, these triple correlations rule out separable states, being therefore a sufficient condition for entanglement. Although these inequalities have already been used for discrete variables [10], this is the first time, to our knowledge, they are used to characterize non classical triple correlations in continuous variables. Whether these triple correlations can be useful for quantum information protocols remains to be investigated.

This work was supported by the Instituto Nacional de Ciência e Tecnologia de Informação Quântica (INCT/IQ - CNPq - Brazil), Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES - Brazil) and Fundação de Amparo à Pesquisa do Estado do Rio de Janeiro (FAPERJ).

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