Higher order nonclassicalities in a codirectional nonlinear optical coupler: Quantum entanglement, squeezing and antibunching

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Higher order nonclassical properties of fields propagating through a codirectional asymmetric nonlinear optical coupler which is prepared by combining a linear wave guide and a nonlinear (quadratic) wave guide operated by second harmonic generation are studied. A completely quantum mechanical description is used here to describe the system. Closed form analytic solutions of Heisenberg’s equations of motion for various modes are used to show the existence of higher order antibunching, higher order squeezing, higher order two-mode and multi-mode entanglement in the asymmetric nonlinear optical coupler. It is also shown that nonclassical properties of light can transfer from a nonlinear wave guide to a linear wave guide.

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I. INTRODUCTION

Several new applications of nonclassical states have been reported in recent past \cite{1, 2}. For example, applications of squeezed state are reported in implementation of continuous variable quantum cryptography \cite{1}, teleportation of coherent states \cite{2}, \textit{etc.}, antibunching is shown to be useful in building single photon sources \cite{3}, entangled state has appeared as one of the main resources of quantum information processing as it is shown to be essential for the implementation of a set of protocols of discrete \cite{4} and continuous variable quantum cryptography \cite{5}, quantum teleportation \cite{6}, dense-coding \cite{7}, \textit{etc.} As a consequence of these recently reported applications, generation of nonclassical states in various quantum systems emerged as one of the most important areas of interest in quantum information theory and quantum optics. Several systems are already investigated and have been shown to produce entanglement and other nonclassical states (see \cite{7, 8} and references therein). However, experimentally realizable simple systems that can be used to generate and manipulate nonclassical states are still of much interest. One such experimentally realizable and relatively simple system is nonlinear optical coupler. Nonlinear optical couplers are of specific interest because they can be easily realized using optical fibers or photonic crystals and the amount of nonclassicality present in the output field can be controlled by controlling the interaction length and the coupling constant. Further, recently Matthews et al. have experimentally demonstrated manipulation of multi-photon entanglement in quantum circuits constructed using waveguides \cite{9}. Quantum circuits implemented by them can also be viewed as optical coupler based quantum circuits as in their circuits waveguides are essentially combined to form couplers. In another interesting application, Mandal and Midda have shown that universal irreversible gate library (NAND gate) can be built using nonlinear optical couplers \cite{10}. Mandal and Midda’s work essentially showed that in principle a classical computer can be built using optical couplers. These facts motivated us to systematically investigate the possibility of observation of nonclassicality in nonlinear optical couplers. Among different possible nonlinear optical couplers one of the simplest systems is a codirectional asymmetric nonlinear optical coupler that is prepared by combining a linear wave guide and a nonlinear (quadratic) wave guide operated by second harmonic generation. Waveguides interact with each other through evanescent wave and we may say that transfer of nonclassical effect from the nonlinear wave guide to the linear one happens through evanescent wave. Present paper aims to study various higher order nonclassical properties of this specific optical coupler with specific attention to entanglement.

It is interesting to note that several nonclassical properties of optical couplers are studied in past (see \cite{11} for a review). For example, photon statistics, phase properties and squeezing in codirectional and contradirectional Kerr nonlinear coupler are studied with fixed and varying linear coupling constant \cite{12, 16}, photon statistics of Raman and Brillouin coupler \cite{17} and parametric coupler \cite{18} is studied in detail, photon statistics and other nonclassical properties of asymmetric \cite{19, 24} and symmetric \cite{24, 25} directional nonlinear coupler is investigated for various conditions such as strong pump \cite{19}, weak pump \cite{21}, phase mismatching \cite{26} for codirectional \cite{21, 22, 26} and contradirectional \cite{22, 23, 26} propagation of classical (coherent) and nonclassical \cite{24, 25} input modes. However, almost all the earlier studies were limited to the in-
vestigation of lower order nonclassical effects (e.g., squeezing and antibunching) either under the conventional short-length approximation [23] or under the parametric approximation where a pump mode is assumed to be strong and treated classically as a c-number [26]. Only a few discrete efforts have recently been made to study higher order nonclassical effects and entanglement in optical couplers [27–31], but even these efforts are limited to Kerr nonlinear coupler. For example, in 2004, Leonski and Miranowicz reported entanglement in Kerr nonlinear coupler [29], subsequently entanglement sudden death [27] and thermally induced entanglement [28] are reported in Kerr nonlinear coupler. Amplitude squared (higher order) squeezing is also reported in Kerr nonlinear coupler [31]. However, neither any effort has yet been made to rigorously study the higher order nonclassical effects in nonlinear optical coupler in general nor a serious effort has been made to study entanglement in nonlinear optical couplers other than Kerr nonlinear coupler. Keeping these facts in mind in the present paper we aim to study higher order nonclassical effects (e.g., higher order antibunching, squeezing and entanglement) in codirectional nonlinear optical coupler.

Remaining part of the paper is organized as follows. In Section II we briefly describe the Hamiltonian that describes the model of the asymmetric nonlinear optical coupler studied here and perturbative solutions of equations of motion corresponding to different field modes present in the Hamiltonian. In Section III we list a set of criteria of nonclassicality with special attention to those kind of nonclassicalities that are never explored for asymmetric nonlinear optical coupler. In Section IV we use the criteria described in the previous section to illustrate the nonclassical characters of various field modes present in the asymmetric nonlinear optical coupler. Specifically, we have reported higher order squeezing, antibunching, and entanglement. Finally, the paper is concluded in Section V.

II. THE MODEL AND THE SOLUTIONS

An asymmetric nonlinear optical coupler is schematically shown in Fig. 1. We are interested in nonclassical properties of this coupler. From Fig. 1 we can clearly see that a linear waveguide is combined with a nonlinear one with \( \chi^{(2)} \) nonlinearity to from the asymmetric coupler. As the \( \chi^{(2)} \) medium can produce second harmonic generation, we may say that the coupler is operated by second harmonic generation. The linear waveguide carries the electromagnetic field characterized by the bosonic field annihilation (creation) operator \( a (a^\dagger) \). On the other hand, the field operators \( b_i (b_i^\dagger) \) correspond to the nonlinear medium. Further, \( b_1 (k_1) \) and \( b_2 (k_2) \) denote annihilation operators (wave vectors) for fundamental and second harmonic modes, respectively. Now the nonlinear momentum operator in the interaction picture for this coupler can be written as

\[
G = -\hbar k a b_1^\dagger - \hbar \Gamma b_1^\dagger b_2^\dagger \exp(i\Delta k z) + \text{h.c.}, \tag{1}
\]

where h.c. stands for the Hermitian conjugate and \( \Delta k = |2k_1 - k_2| \), denotes the phase mismatch between the fundamental and second harmonic beams. The parameters \( k \) and \( \Gamma \) are the linear and nonlinear coupling constants and are proportional to the linear \( \chi^{(1)} \) and nonlinear \( \chi^{(2)} \) susceptibilities, respectively. The value of \( \chi^{(2)} \) is considerably smaller than \( \chi^{(1)} \) (typically \( \chi^{(2)} / \chi^{(1)} \approx 10^{-6} \)) and as a consequence \( \Gamma \ll k \) unless an extremely strong pump is present. The model is elaborately discussed by some of the present authors in their earlier publications [11, 20, 21]. Specifically, in Ref. [20] single mode and intermodal squeezing, antibunching and subshot noise was studied using analytic expressions of spatial evolution of field operators obtained by short-length solution of the Heisenberg’s equations of motion corresponding to (1). The validity of short length solution used in Ref. [20] was strictly restricted by the condition \( \Gamma z \ll 1 \). Later on Sen and Mandal developed a perturbative solution technique [32] that can solve Heisenberg’s equations of motion for \( \Gamma z \ll 1 \). Sen-Mandal technique was subsequently used in Ref. [21] to obtain spatial evolution of field operators corresponding to (1) and to study single mode and intermodal squeezing and antibunching. Interestingly, in [21] some nonclassical characters of asymmetric nonlinear optical coupler were observed which were not observed in the earlier investigations [11, 20] performed using short-length solution. This was indicative of the fact that the Sen-Mandal perturbative method provides better solution for the study of nonclassical properties. The same fact is observed in other optical systems, too (8 and references therein). However, neither entanglement nor any of the higher order nonclassical properties were studied in earlier papers. Keeping these facts in mind we have used the solution reported in Ref. [21] to study the higher order nonclassicalities.

In Ref. [21] closed form analytic expressions for evolution of field operators valid up to linear power of the coupling co-

- Figure 1: Schematic diagram of a codirectional asymmetric nonlinear optical coupler prepared by combining a linear wave guide with a nonlinear (quadratic) wave guide operated by second harmonic generation. The fields involved are described by the corresponding annihilation operators, as shown; \( L \) is the interaction length.
efficient $\Gamma$ were obtained as follows:

\[ a(z) = f_1 a(0) + f_2 b_1(0) + f_3 b_1^2(0) b_2(0) + f_4 a^2(0) b_2(0), \]
\[ b_1(z) = g_1 a(0) + g_2 b_1(0) + g_3 b_1^2(0) b_2(0) + g_4 a(0) b_2(0), \]
\[ b_2(z) = h_1 b_2(0) + h_2 b_2(0) a(0) + h_3 a^2(0), \]

where

\[ f_1 = g_2 = \cos|k| z, \]
\[ f_2 = -g_1^* = -\frac{i k}{|k|} \sin|k| z, \]
\[ f_3 = -\frac{2k^*}{4|k|^2 - (\Delta k)^2} \left[ G_f - f_1 + \text{exp}\left\{ \Delta k - \frac{2|k|^2}{\Delta k} G_f \right\} \right], \]
\[ f_4 = \frac{4k^*}{4|k|^2 - (\Delta k)^2} G_f - f_1 + \frac{2k^*}{2|k|^2 - (\Delta k)^2} G_f f_2, \]
\[ g_3 = \frac{2k^*}{4|k|^2 - (\Delta k)^2} G_f f_2 - \frac{2k^*}{2|k|^2 - (\Delta k)^2} G_f G_f f_1, \]
\[ g_4 = \frac{4k^*}{4|k|^2 - (\Delta k)^2} G_f f_2 - \frac{4k^*}{2|k|^2 - (\Delta k)^2} G_f G_f f_1 \]
\[ \times (G_f - 1) f_2 + \frac{2k^*}{4|k|^2 - (\Delta k)^2} G_f - f_1, \]
\[ h_1 = 1, \]
\[ h_2 = \frac{G_f}{2s^*} - \frac{2k^*}{4|k|^2 - (\Delta k)^2} \left[ 2|k| \left( G_f^* - 1 \right) \sin 2|k| z \right. \]
\[ + i \Delta k \left( 1 - (G_f^* - 1) \cos 2|k| z \right], \]
\[ h_3 = -\frac{k^*}{4|k|^2 - (\Delta k)^2} \left[ i \Delta k \left( G_f^* - 1 \right) \sin 2|k| z \right. \]
\[ + 2|k| \left( 1 - (G_f^* - 1) \cos 2|k| z \right], \]
\[ h_4 = -\frac{k^*}{4|k|^2 - (\Delta k)^2} \left[ 2|k| \left( G_f^* - 1 \right) \right. \]
\[ \times \sin 2|k| z - i \Delta k \left( 1 - (G_f^* - 1) \cos 2|k| z \right], \]

where $G_{\pm} = (1 / \pm \exp(-i \Delta k z))$. In what follows we will use these closed form analytic expressions of the field operators to investigate the spatial evolution of entanglement and some higher order nonclassical characteristics of the field modes. We will not discuss the usual nonclassical characters such as squeezing and antibunching as they are already discussed in Ref. [21].

### III. CRITERIA OF NONCLASSICALITY

A state having negative or highly singular (more singular than $\delta$-function) Glauber-Sudarshan $P$-function is referred to as a nonclassical state as it cannot be expressed as a classical mixture of coherent states. $P$-function provides us an essential as well as sufficient criterion for detection of nonclassicality. However, $P$-function is not directly experimentally measurable. Consequently, several operational criteria for nonclassicality are proposed in last 50 years. A large number of these criteria are expressed as inequalities involving expectation values of functions of annihilation and creation operators. This implies that Eqs. (2)-(3) provide us the sufficient mathematical framework required to study the nonclassical properties of the codirectional asymmetric nonlinear optical coupler. As mentioned above we are interested in higher order nonclassical properties of radiation field. In quantum optics and quantum information higher order nonclassical properties of bosons (e.g., higher order Hong-Mandel squeezing, higher order antibunching, higher order sub-Poissonian statistics, higher order entanglement, etc.) are often studied (33 and references therein). Until recent past studies on higher order nonclassicalities were predominantly restricted to theoretical investigations. However, a bunch of exciting experimental demonstrations of higher order nonclassicalities are recently reported [34]. Specifically, existence of higher order nonclassicality in bipartite multi-mode states produced in a twin-beam experiment is recently demonstrated by Allevi, Oliveares and Bondani [34] using a new criterion for higher order nonclassicality introduced by them. They also showed that detection of weak nonclassicalities is easier with their higher order criterion of nonclassicality as compared to the existing lower order criteria [33]. This observation was consistent with the earlier theoretical observation of Pathak and Garcia [37] that established that depth of nonclassicality in higher order antibunching increases with the order. The possibility that higher order nonclassicality may be more useful in identifying the weak nonclassicalities have considerably increased the interest of the quantum optics community on the higher order nonclassical characters of bosonic fields. In the remaining part of this section we list a set of criteria of higher order nonclassicalities and in the following section we study the possibility of satisfying those criteria in the codirectional asymmetric nonlinear optical coupler.

#### A. Higher order squeezing

Higher order squeezing is usually studied using two different approaches [38, 40]. In the first approach introduced by Hillery in 1987 [38] reduction of variance of an amplitude powered quadrature variable for a quantum state with respect to its coherent state counterpart reflects nonclassicality. In contrast in the second type of higher order squeezing introduced by Hong and Mandel in 1985 [33, 40] higher order squeezing is reflected through the reduction of higher order moments of usual quadrature operators with respect to their coherent state counterparts. In the present paper we have studied higher order squeezing using Hillery’s criterion of amplitude powered squeezing. Specifically, Hillery introduced amplitude powered quadrature variables as

\[ Y_{1,a} = \frac{a^k + (a^1)^k}{2}, \]
\[ Y_{2,a} = i \left( \frac{(a^1)^k - a^k}{2} \right). \]

As $Y_{1,a}$ and $Y_{2,a}$ do not commute we can obtain uncertainty relation and a condition of squeezing. For example, for $k = 2$, Hillery’s criterion for amplitude squared squeezing is described as

\[ A_{i,a} = \left\langle (\Delta Y_{i,a})^2 \right\rangle - \left\langle N_a + \frac{1}{2} \right\rangle < 0, \]

where $i \in \{1, 2\}$. 

B. Higher order antibunching

Since 1977 signatures of higher order nonclassical photon statistics in different optical systems of interest have been investigated by some of the present authors using criterion based on higher order moments of number operators (cf. Ref. [11] and Chapter 10 of [41] and references therein). However, higher order antibunching (HOA) was not specifically discussed, but it was demonstrated there for degenerate and nondegenerate parametric processes in single and compound signal-idler modes, respectively and for Raman scattering in compound Stokes-anti-Stokes mode up to \(n = 5\). Further, it was shown that the HOA is deeper with increasing \(n\) occurring on a shorter time interval in parametric processes whereas different order HOA occurs on the same time interval in Raman scattering. A specific criterion for HOA was first introduced by C. T. Lee [42] in 1990 using higher order moments of number operator. Initially, HOA was considered to be a phenomenon that appears rarely in optical systems, but in 2006, some of the present authors established that it is not really a rare phenomenon [43]. Since then HOA is reported in several quantum optical systems (see [43] and references therein) and atomic systems [44]. However, no effort has yet been made to study HOA in optical couplers. Thus the present study of HOA in asymmetric nonlinear optical coupler is first of its kind and is expected to lead to similar observations in other type of optical couplers. Before we proceed further, we would like to note that signature of HOA can be observed through a bunch of equivalent but different criteria, all of which can be interpreted as modified Lee criterion. In what follows we will use following simple criterion of \((n - 1)^{th}\) order single mode antibunching introduced by Pathak and Garcia [47]

\[
D_a(n - 1) = \langle a^\dagger a^n \rangle - \langle a^\dagger a \rangle^n < 0. \tag{7}
\]

Here \(n = 2\) corresponds to the usual antibunching and \(n \geq 3\) refers to the higher order antibunching.

C. Entanglement and higher order entanglement

There exist several inseparability criteria ([45] and references therein) that are expressed in terms of expectation values of field operators and thus suitable for study of entanglement dynamics within the frame-work of the present approach. Among these criteria Duan et al.’s criterion [46] which is usually referred to as Duan’s criterion, Hillery-Zubairy criterion I and II (HZ-I and HZ-II) [47,49] have received more attention because of various reasons, such as computational simplicity, experimental realizability and their recent success in detecting entanglement in various optical, atomic and optomechanical systems (8,44) and references therein). To begin with we may note that the first inseparability criterion of Hillery and Zubairy, i.e., HZ-1 criterion of inseparability is described as

\[
\langle N_a N_b \rangle - |\langle ab \rangle|^2 < 0, \tag{8}
\]

whereas the second criterion of Hillery and Zubairy, i.e., HZ-II criterion is given by

\[
\langle N_a \rangle \langle N_b \rangle - |\langle ab \rangle|^2 < 0. \tag{9}
\]

The other criterion of inseparability to be used in the present paper is Duan et al.’s criterion which is described as follows [46]:

\[
d_{ab} = \left(\langle \Delta u_{ab} \rangle^2 \right) + \left(\langle \Delta v_{ab} \rangle^2 \right) - 2 < 0, \tag{10}
\]

where

\[
u_{ab} = \frac{1}{\sqrt{2}} \left\{ \left( a + a^\dagger \right) + \left( b + b^\dagger \right) \right\}, \quad \langle a^\dagger a \rangle^n \quad 2 \leftrightarrow 0.
\]

\[
u_{ab} = - \frac{1}{\sqrt{2}} \left\{ \left( a - a^\dagger \right) + \left( b - b^\dagger \right) \right\}.
\]

Clearly our analytic solution (2-3) enables us to investigate intermodal entanglement in asymmetric nonlinear optical coupler using all the three inseparability criteria described above. As all these three inseparability criteria are only sufficient (not necessary), a particular criterion may fail to identify entanglement detected by another criterion. Keeping this fact in mind, we use all these criteria to study the intermodal entanglement in asymmetric nonlinear optical coupler. The criteria described above can only detect bi-partite entanglement of lowest order. As possibility of generation of entanglement in asymmetric nonlinear optical coupler is not discussed earlier we have studied the spatial evolution of intermodal entanglement using these lower order inseparability criteria. However, to be consistent with the focus of the present paper, we need to investigate the possibility of observing higher order entanglement, too. For that purpose we require another set of criteria for detection of higher order entanglement. All criteria for detection of multi-partite entanglement are essentially higher order criteria [51,52] as they reveal some higher order correlation. Interestingly, there exist higher order inseparability criteria for detection of higher order entanglement in bipartite case, too. Specifically, Hillery-Zubairy introduced two criteria for intermodal higher order entanglement [47] as follows

\[
E_{ab}^{m,n} = \left| \langle a^\dagger \rangle^m a^m \langle b^\dagger \rangle^n b^n \right| - \left| \left\langle a^m (b^\dagger) \right\rangle^n \right|^2 < 0, \tag{12}
\]

and

\[
E_{ab}^{m,n} = \left| \langle a^\dagger \rangle^m a^m \langle b^\dagger \rangle^n b^n \right| - |\langle a^m b^n \rangle|^2 < 0. \tag{13}
\]

Here \(m\) and \(n\) are non-zero positive integers and lowest possible values of \(m\) and \(n\) are \(m = n = 1\) which reduces (12) and (13) to usual HZ-I criterion (i.e., (8)) and HZ-II criterion (i.e., (9)), respectively. Thus these two criteria are generalized version of well known lower order criteria of Hillery and Zubairy and we may refer to (12) and (13) as HZ-I criterion and HZ-II criterion respectively in analogy to the lowest order cases. A quantum state will be referred to as (bipartite) higher order entangled state if it is found to satisfy (12) and/or (13) for any choice of integer \(m\) and \(n\) satisfying \(m + n \geq 3\). The other type of higher order entanglement i.e., multi-partite entanglement can be detected in various ways. In the present
In this paper we have used a set of multi-mode inseparability criteria introduced by Li et al. Specifically, Li et al. have shown that a three-mode quantum state is not bi-separable in the form\(ab_1|b_2\) (i.e., compound mode\(ab_1\) is entangled with the mode\(b_2\)) if the following inequality holds for the three-mode system

\[
E_{ab_1|b_2} = \langle a|a|b_1|b_2]\rangle - \langle ab_1|b_2\rangle|\langle ab_1|b_2\rangle|^2 < 0,
\]

where \(m, n, l\) are positive integers and annihilation operators\(a, b_1, b_2\) correspond to the three modes. A quantum state satisfying the above inequality is referred to as \(E_{ab_1|b_2}\) entangled state. Three mode inseparability criterion can be written in various alternative forms. For example, an alternative criterion for detection of \(ab_1|b_2\) entangled state is

\[
E_{ab_1|b_2}^{mn,l} = \langle \hat{a} \hat{a}\rangle |\langle ab_1|b_2\rangle|^2 - \langle ab_1|b_2\rangle^2 < 0.
\]

Similarly, one can define criteria for detection of \(a|b_1|b_2\) and \(b_1|ab_2\) entangled states and use them to obtain criterion for detection of fully entangled tripartite state. For example, using \(\langle 16\rangle\) and \(\langle 15\rangle\) respectively we can write that the three modes of our interest are not bi-separable in any form if any one of the following two sets of inequalities are satisfied simultaneously

\[
E_{a|b_1|b_2}^{11,1} < 0, \quad E_{a|b_1|b_2}^{1,11} < 0, \quad E_{b_1|b_2}^{1,11} < 0,
\]

Further, for a fully separable pure state we always have

\[
|\langle ab_1|b_2\rangle|^2 < 0.
\]

Thus a 3-mode pure state that violates \(\langle 18\rangle\) (i.e., satisfies \(\langle N_a\rangle\langle N_{b_1}\rangle\langle N_{b_2}\rangle\langle 18\rangle\) and simultaneously satisfies either \(\langle 16\rangle\) or \(\langle 17\rangle\) is a fully entangled state as it is neither fully separable nor bi-separable in any form.

IV. NONCLASSICALITY IN CODIRECTIONAL OPTICAL COUPLER

Using the perturbative solutions \(\langle 12\rangle\)–\(\langle 13\rangle\) we can obtain time evolution of various operators that are relevant for the detection of nonclassical characters. For example, we may use \(\langle 12\rangle\)–\(\langle 13\rangle\) to obtain the number operators for various field modes as follows

\[
N_a = a^\dagger a = |f_1|^2 a^\dagger(0)a(0) + |f_2|^2 b_1^\dagger(0)b_2(0) + \left[ f_1^2 f_2 a^\dagger(0)b_1(0) + f_1 f_3 b_1^\dagger(0)b_2(0) + f_1 f_4 b_2^\dagger(0)b_1(0) + f_2 f_4 b_2^\dagger(0)b_1(0) \right].
\]

\[
N_{b_1} = b_1^\dagger b_1 = |g_1|^2 a^\dagger(0)a(0) + |g_2|^2 b_1^\dagger(0)b_2(0) + \left[ g_1^2 g_2 b_1^\dagger(0)\hat{b}_1(0) + g_1^2 g_3 a^\dagger(0)b_2(0) + g_1^2 g_4 b_2^\dagger(0)b_1(0) + g_2^2 g_4 b_2^\dagger(0)b_1(0) \right].
\]

\[
N_{b_2} = b_2^\dagger b_2 = b_2^\dagger(0)b_2(0) + \left[ h_2 b_2^\dagger(0)\hat{b}_2(0) + h_3 b_2^\dagger(0)b_2(0) + h_4 b_2^\dagger(0)b_2(0) \right].
\]

The average value of the number of photons in the modes \(a\), \(b_1\) and \(b_2\) may now be calculated with respect to a given initial state. We assume that initial state is a product of three coherent states: \(|\alpha\rangle|\beta\rangle|\gamma\rangle\), where \(|\alpha\rangle\), \(|\beta\rangle\) and \(|\gamma\rangle\) are eigen kets of annihilation operators \(a\), \(b_1\) and \(b_2\), respectively. Field operator \(a(0)\) operating on such a multi-mode coherent state yields a complex eigenvalue \(\alpha\). Specifically,

\[
a(0)|\alpha\rangle|\beta\rangle|\gamma\rangle = \alpha|\alpha\rangle|\beta\rangle|\gamma\rangle,
\]

where \(|\alpha|^2\), \(|\beta|^2\), \(|\gamma|^2\) is the number of input photons in the field mode \(a\), \(b_1\) and \(b_2\), respectively. For a spontaneous process, the complex amplitudes should satisfy \(\beta = \gamma = 0\) and \(\alpha \neq 0\). Whereas, for a stimulated process, the complex amplitudes are not necessarily zero and it would be physically reasonable to choose \(\alpha > \beta > \gamma\). In what follows, in all the figures (except Fig. 13) that illustrate the existence of nonclassical character in asymmetric nonlinear optical coupler we have chosen \(\alpha = 5\), \(\beta = 2\), \(\gamma = 1\).
A. Higher order squeezing

Using Eqs. (2)-(3), (19), (21) in the criterion of amplitude squared squeezing (6), we obtain

\[
\begin{bmatrix}
A_{1,a} \\
A_{2,a}
\end{bmatrix} = \pm \left[ \left( f_1 f_4 + f_2 f_3 \right) \left( f_1^2 \alpha^2 \gamma + f_2^2 \beta^2 \gamma \right) \\
+ f_1 f_2 \alpha \beta \gamma \right] + c.c.,
\]

(23)

and

\[
\begin{bmatrix}
A_{1,b_1} \\
A_{2,b_1}
\end{bmatrix} = \pm \left[ \left( g_1 g_4 + g_2 g_3 \right) \left( g_1^2 \alpha^2 \gamma + g_2^2 \beta^2 \gamma \right) \\
+ g_1 g_2 \alpha \beta \gamma \right] + c.c.,
\]

(24)

Clearly we don’t obtain any signature of amplitude squared squeezing in \(b_2\) mode using the present solution and mode \(a\) (\(b_1\)) should always show amplitude squared squeezing in one of the quadrature variables as both \(A_{1,a}\) and \(A_{2,a}\) (\(A_{1,b_1}\) and \(A_{2,b_2}\)) cannot be positive simultaneously. To investigate the possibility of amplitude squared squeezing in further detail in modes \(a\) and \(b_1\) we have plotted the spatial variation of \(A_{1,a}\) and \(A_{1,b_1}\) in Fig. [2]. Negative regions of these two plots clearly illustrate the existence of amplitude squared squeezing in both \(a\) and \(b_1\) modes.

B. Higher order antibunching

We have already described the condition of HOA as (7). Now using Eqs. (2)-(3), (7) and (19)-(21) we can obtain closed form analytic expressions for \(D_i(n)\) for various modes as follows

\[
D_a(n) = n C_2 \gamma \left( f_1 \alpha + f_2 \beta \right) |2n-4 \left\{ f_1 \alpha + f_2 \beta \right\}^2 \left( f_3 f_4 + f_1 f_2 + f_1 f_3 + f_2 f_4 \right) + c.c. \right),
\]

(26)

\[
D_b_1(n) = n C_2 \gamma \left( g_1 \alpha + g_2 \beta \right) |2n-4 \left\{ g_1 \alpha + g_2 \beta \right\}^2 \left( g_3 g_4 + g_1 g_2 \right) + c.c. \right),
\]

(27)

\[
D_{b_2}(n) = 0.
\]

(28)

Clearly, the perturbative solution used here cannot detect any signature of higher order antibunching for \(b_2\) mode. However, in the other two modes we observe HOA for different values of \(n\) as illustrated in Fig. [8]. In Fig. [8] we have plotted right hand sides of (26) and (27) along with the exact numerical results obtained by integrating the time dependent Schrödinger equation corresponding to given Hamiltonian by using the matrix form of the operators. Close resemblance of the exact numerical result with the perturbative result even for higher order case clearly validates the perturbative solution used here.

C. Intermodal entanglement

To apply HZ-I criterion to investigate the existence of intermodal entanglement between modes \(a\) and \(b_1\) i.e., compound mode \(ab_1\) we use Eqs. (2)-(3) and (19)-(21) and obtain

\[
E_{ab_1}^{1,1} = \left( \left| n_a \right| \left| n_{b_1} \right| - \left| \langle ab_1 \rangle \right| \right)^2
\]

\[
= \left( |g_1|^2 f_1^* f_1 + f_2^* f_2 g_2^* g_1 \right) \alpha^2 \gamma^* + \left( |f_1|^2 g_1^* g_4 + f_2^* f_2 g_1^* g_3 \right) \alpha^2 \gamma^* + \left( |g_2|^2 f_3^* f_2 + f_1^* f_1 g_3^* g_2 \right) \beta^2 \gamma^* + \left( |f_2|^2 g_3^* g_3 + f_3^* f_3 g_4^* g_4 \right) \beta^2 \gamma^* + \left( |g_1|^2 - |g_2|^2 \right) \left( f_1^* f_2 - f_3^* f_1 \right) \alpha \beta \gamma^* - \left( g_2^* g_4 - g_1^* g_3 \right) \alpha^* \beta^* \gamma^*.
\]

(29)

Similarly, applying HZ-II criterion to the compound mode \(ab_1\) we obtain

\[
E_{ab_1}^{1,1} = \left( \left| n_a \right| \left| n_{b_1} \right| - \left| \langle ab_1 \rangle \right| \right)^2
\]

\[
= -\left( |g_1|^2 f_1^* f_1 + f_2^* f_2 g_2^* g_1 \right) \alpha^2 \gamma^* + \left( |f_1|^2 g_1^* g_4 + f_2^* f_2 g_1^* g_3 \right) \alpha^2 \gamma^* + \left( |g_2|^2 f_3^* f_2 + f_1^* f_1 g_3^* g_2 \right) \beta^2 \gamma^* + \left( |f_2|^2 g_3^* g_3 + f_3^* f_3 g_4^* g_4 \right) \beta^2 \gamma^* + \left( |g_1|^2 - |g_2|^2 \right) \left( f_1^* f_2 - f_3^* f_1 \right) \alpha \beta \gamma^* - \left( g_2^* g_4 - g_1^* g_3 \right) \alpha^* \beta^* \gamma^*.
\]

(30)

From Eqs. (29) and (30) we can easily observe that in the present case \(E_{ab_1}^{1,1} = -E_{ab_1}^{1,1}\), which implies that at any
Amplitude squared squeezing is observed in modes \(a\) (top) and \(b_1\) (bottom) for the initial state \(|\alpha\rangle|\beta\rangle|\gamma\rangle\) with \(k = 0.1, \Gamma = 0.001, \Delta k = 10^{-4}, \alpha = 5, \beta = 2, \gamma = 1\). Negative parts of the solid line represents squeezing in quadrature variable \(Y_{1,a}\) \((Y_{1,b_1})\) and that of the dashed line represents squeezing in quadrature variable \(Y_{2,a}\) \((Y_{2,b_1})\).

Following the same approach we investigated the existence of entanglement in other compound modes \((e.g., \ ab_2\) and \(b_1 b_2\)), but both HZ-I and HZ-II criteria failed to detect any entanglement in these cases. However, it does not indicate that the modes are separable as both HZ-I and HZ-II inseparability criteria are only sufficient and not essential. Further, the perturbative analytic solution used here is an approximate solution and in recent past we have seen several examples where the existence of entanglement not detected by HZ criteria is detected by Duan et al.’s criterion or vice versa [8, 44]. Keeping these facts in mind, we studied the possibilities of observing intermodal entanglement using Duan et al.’s criterion, too, but it failed to detect any entanglement in the present case as we obtained

\[ d_{ab_1} = d_{ab_2} = d_{b_1 b_2} = 0. \tag{31} \]

We may now investigate the existence of higher order entanglement using Eqns. (12)-(18). To begin with we may use

\[ E_{ab_1}^{1,1} \text{ and } E_{ab_1}^{1,1} \]

Figure 3: (color online) HOA with rescaled interaction length \(\Gamma z\) in mode \(a\) (top panel) and mode \(b_1\) (bottom panel) for \(n = 3\) (smooth line) and \(n = 4\) (dashed line) and square and circle are for the corresponding numerical results with the initial state \(|\alpha\rangle|\beta\rangle|\gamma\rangle\) and \(k = 0.1, \Gamma = 0.001, \Delta k = 10^{-4}, \alpha = 0.5, \beta = 0.2, \gamma = 0.1\).
Similarly, using (23)-(3) and (13) we can obtain a closed form analytic expression for $E_{ab_2}$. To be precise, all multi-mode entanglement are essentially higher order entanglement. To study higher order entanglement, it is necessary to detect lower order entanglement present in compound modes $ab_2$ and $b_1$. In Fig. 5 we have illustrated the spatial evolution of $E_{ab_2}^{1.1}$ and $E_{ab_2}^{0.1}$. Negative regions of this figure clearly show the existence of higher order entanglement in compound mode $ab_1$. As expected from (33), we observe that for any value of $\Gamma z$ compound mode $ab_1$ is higher entangled. However, Hillery-Zubairy’s higher order entanglement criteria (12), (13) could not show any signature of higher order entanglement in compound modes $ab_2$ and $b_1$ and $b_2$. This is not surprising as Hillery-Zubairy’s criteria are only sufficient not necessary and we have already seen that these criteria fail to detect lower order entanglement present in compound modes $ab_2$ and $b_1$. There exists another way to study higher order entanglement. To be precise, all multi-mode entanglement are essentially higher order entanglement. As there are 3 modes in the coupler studied here, we may also investigate the existence of three-mode entanglement. We have already noted that a 3-mode pure state that violates (13) (i.e., satisfies $\langle N_\alpha \rangle \langle N_\beta \rangle \langle N_\gamma \rangle - |\langle ab_2 \rangle^2| < 0$) and simultaneously satisfies either (16) or (17) is a fully entangled state. Now using (2)-(3) and (13)-(18) we obtain following relations for $m=n=l=1$:}

$$E^{1.1,1}_{a \mid b_1, b_2} = -E^{1.1,1}_{a \mid b_2, b_1} = E^{1.1,1}_{a \mid b_2, b_1} = |\gamma|^2 E^{1.1,1}_{ab_1},$$

$$E^{1.1,1}_{ab_1} = E^{1.1,1}_{ab_1} = 0,$$

From (34) we can see that three modes of the coupler is not bi-separable in the form $a \mid b_1 b_2$ and $a \mid b_2 b_1$ for any value of $\Gamma z > 0$. Further, Eqn. (36) and positive regions of $E^{1.1}_{ab_1}$ shown in Fig. 4 show that the three modes of the coupler are not fully separable. However, present solution does not show signature of fully entangled 3-mode state as (35) does not show entanglement between coupled mode $ab_1$ and mode $b_2$. To be specific, we observed 3-mode (higher order) entanglement, but could not observe signature of fully entangled 3-mode state. However, here we cannot conclude whether the three modes of the coupler are fully entangled or not as the criteria used here are only sufficient.

We have already observed different signatures of nonclassicality in asymmetric nonlinear optical coupler of our interest. If we now closely look into all the analytic expressions of signatures of nonclassicality provided here through Eqs. (23), (36) we can find an interesting symmetry: the nonvanishing expressions of signatures of nonclassicality are proportional to $|\gamma|$. Thus we may conclude that within the domain of validity of the present solution, in the spontaneous process we would not observe any of the nonclassical characters that are observed here in stimulated case.

**V. CONCLUSIONS**

We have observed various types of higher order nonclassicality in fields propagating through a codirectional asymmetric nonlinear optical coupler prepared by combining a linear wave guide and a nonlinear (quadratic) wave guide operated by second harmonic generation. The observations are elaborated in Section IV. In brief, we have observed higher order (amplitude squared) squeezing, higher order antibunching and higher order entanglement. None of these higher order nonclassical phenomena were reported in earlier studies on the codirectional asymmetric nonlinear optical coupler ([21] and references therein). In fact, till date neither entanglement nor higher order nonclassicalities are systematically studied in optical couplers other than the Kerr coupler. The method followed in the present paper is quite general and it can be extended easily to the other type of couplers, such as contradirectional asymmetric nonlinear coupler, codirectional and contradirectional Raman and Brillouin coupler [17] and parametric coupler [18]. Further, it is even possible to investigate the existence of Hong-Mandel [39, 40] type higher order squeezing and Agarwal-Tara parameter $\Lambda_m$ [54] for higher order nonclassicality using the present approach. It is also possible to study lower order and higher order steering using...
the present approach and the strategy adopted in Ref. [55]. However, we have not investigated steering as recently it is shown that every pure entangled state is maximally steerable [56]. Since the combined states of three modes of asymmetric codirectional optical coupler is a pure state, the findings of Ref. [56] and the intermodal entanglement observed in the present paper implies that the compound modes \( ab_1 \) is maximally steerable. The importance of entanglement and steering in various applications of quantum computing and quantum communication and the easily implementable structure of the coupler studied here indicate the possibility that the entangled states generated through the coupler of the present form would be useful in various practical purposes.

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