Nonclassical properties of a contradirectional nonlinear optical coupler

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Abstract

We investigate the nonclassical properties of output fields propagated through a contradirectional asymmetric nonlinear optical coupler consisting of a linear waveguide and a nonlinear (quadratic) waveguide operated by second harmonic generation. In contrast to the earlier results, all the initial fields are considered weak and a completely quantum mechanical model is used here to describe the system. Perturbative solutions of Heisenberg’s equations of motion for various field modes are obtained using Sen-Mandal technique. Obtained solutions are subsequently used to show the existence of single-mode and intermodal squeezing, single-mode and intermodal antibunching, two-mode and multi-mode entanglement in the output of contradirectional asymmetric nonlinear optical coupler. Further, existence of higher order nonclassicality is also established by showing the existence of higher order antibunching, higher order squeezing and higher order entanglement. Variation of observed nonclassical characters with different coupling constants and phase mismatch is discussed.

Keywords: entanglement, higher order nonclassicality, waveguide, optical coupler

1 Introduction

Different aspects of nonclassical properties of electromagnetic field have been studied since the advent of quantum optics. However, the interest on nonclassical states has been considerably escalated with the progress of interdisciplinary field of quantum computation and quantum communication in recent past as a large number of applications of nonclassical states have been reported in context of quantum computation and quantum communication \cite{1, 2, 3, 4, 5, 6}. Specifically, it is shown that squeezed states can be used for the implementation of continuous variable quantum cryptography \cite{1} and teleportation of coherent states \cite{2}, antibunched states can be used to build single photon sources \cite{3}, entangled states are essential for the implementation of a set of protocols of discrete \cite{4} and continuous variable quantum cryptography \cite{1}, quantum teleportation \cite{5}, dense-coding \cite{6}, etc., states violating Bell’s inequality are useful for the implementation of protocols of device independent quantum key distribution \cite{7}. Study of the possibility of generation of nonclassical states (specially, entanglement and nonlocal characters of quantum states) in different quantum systems have recently become extremely relevant and important for the researchers working in different aspects of quantum information theory and quantum optics. Existence of entanglement and other nonclassical states in a large number of bosonic systems have already been reported (See \cite{8, 9, 10} and references therein). However, it is still interesting to study the possibility of generation of nonclassical states in experimentally realizable simple systems. A specific system of this kind is a nonlinear optical coupler which can be easily realized using optical fibers or photonic crystals. Optical couplers are interesting for several reasons. Firstly, in a coupler the amount of nonclassicality can be controlled by controlling the interaction length and the coupling constants. Further, optical couplers are of specific interest as recently Matthews et al. have experimentally

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demonstrated manipulation of multiphoton entanglement in quantum circuits constructed using optical couplers [11]. Mandal and Midda have shown that NAND gate (thus, in principle a classical computer) can be build using nonlinear optical couplers [12]. Motivated by these facts we aim to systematically investigate the possibility of observing nonclassicality in nonlinear optical couplers. As a first effort we have reported lower order and higher order entanglement and other higher order nonclassical effects in an asymmetric codirectional nonlinear optical coupler which is prepared by combining a linear waveguide and a nonlinear (quadratic) waveguide operated by second harmonic generation [13]. Waveguides interact with each other through evanescent wave. Extending the earlier investigation [13] here we study a similar asymmetric nonlinear optical coupler for contradirectional propagation of fields. This type of contradirectional asymmetric optical coupler was studied earlier by some of us [14, 15]. However, in the earlier studies [14, 15] intermodal entanglement and some of the higher order nonclassical properties studied here were not studied. Further, in those early studies second-harmonic mode \( b_2 \) was assumed to be pumped with a strong coherent beam. In other words, \( b_2 \) mode was assumed to be classical and thus it was beyond the scope of the previous studies to investigate single-mode and intermodal nonclassicalities involving this mode. Interestingly, completely quantum mechanical treatment adopted in the present work is found to show intermodal squeezing in a compound mode involving \( b_2 \) mode. In addition, conventional short-length solutions of Heisenberg’s equations of motion were used in earlier studies [14, 15], but recently it is established by some of us that using improved perturbative solutions obtained by Sen-Mandal approach [16, 17, 18] we can observe several nonclassical characters that are not observed using short-length (or short-time approach) [9, 10, 13, 19]. Keeping these facts in mind present paper aims to study nonclassical properties of this contradirectional asymmetric nonlinear optical coupler with specific attention to entanglement using perturbative solution obtained by Sen-Mandal technique.

Nonclassical properties of different optical couplers are extensively studied in past (see [14] for a review). Specifically, signatures of nonclassicality in terms of photon statistics, phase properties and squeezing were investigated in codirectional and contradirectional Kerr nonlinear couplers having fixed and varying linear coupling constant [20, 21, 22, 23, 24], Raman and Brillouin coupler [25] and parametric coupler [26, 27], asymmetric [15, 19, 28, 29, 30] and symmetric [30, 31, 32] directional nonlinear coupler etc. Here it would be apt to note that the specific coupler system that we wish to study in the present paper has already been investigated [15, 33] for contradirectional propagation of classical (coherent) input modes and it is shown that depth of nonclassicality may be controlled by varying the phase mismatching \( \Delta k \) [33].

Existing studies are restricted to the investigation of lower order nonclassical effects (e.g., squeezing and antibunching) under the conventional short-length approximation. Only a few discrete efforts have recently been made to study higher order nonclassical effects and entanglement in optical couplers [13, 34, 35, 36, 37, 38, 39], but except a recent study on codirectional nonlinear optical coupler reported by us [13], all the other efforts were limited to Kerr nonlinear optical coupler. For example, in 2004, Leonski and Miranowicz reported entanglement in Kerr nonlinear optical coupler [36] and pumped Kerr nonlinear optical coupler [39], subsequently entanglement sudden death [34] and thermally induced entanglement [35] were reported in the same system. Amplitude squared (higher order) squeezing was also reported in Kerr nonlinear optical coupler [38]. However, no effort has yet been made to rigorously study the higher order nonclassical effects and entanglement in contradirectional nonlinear optical coupler. Keeping these facts in mind in the present letter we aim to study nonclassical effects (including higher order nonclassicality and entanglement) in contradirectional nonlinear optical coupler.

Remaining part of the paper is organized as follows. In Section 2, the model momentum operator that represents the asymmetric nonlinear optical coupler is described and perturbative solutions of equations of motion corresponding to different field modes present in the momentum operator are reported. In Section 3, we briefly describe a set of criteria of nonclassicality. In Section 4 the criteria described in the previous section are used to investigate the existence of different nonclassical characters (e.g., lower order and higher order squeezing, antibunching, and entanglement) in various field modes present in the contradirectional asymmetric nonlinear optical coupler. Finally, Section 5 is dedicated for conclusions.

### 2 The model and the solution

A schematic diagram of a contradirectional asymmetric nonlinear optical coupler is shown in Fig. 1. From Fig. 1 one can easily observe that a linear waveguide is combined with a nonlinear (\( \chi^{(2)} \)) waveguide to constitute the asymmetric coupler of our interest. Further, from Fig. 1 we can observe that in the linear waveguide field propagates in a direction opposite to the propagation direction of the field in the nonlinear waveguide. Electromagnetic field characterized by the bosonic field annihilation (creation) operator \( a (a^\dagger) \) propagates through the linear waveguide. Similarly, the field operators \( b_i (b_i^\dagger) \) corresponds to the nonlinear medium. Specifically, \( b_1(k_1) \) and \( b_2(k_2) \) denote annihilation operators (wave vectors) for fundamental and second harmonic modes, respectively. Now the momentum
operator for contradirectional optical coupler is \[ G = -\hbar k a b_1^\dagger - \hbar \Gamma b_1^\dagger b_2^2 \exp (i \Delta k z) + \text{h.c.} \quad (1) \]

where h.c stands for the Hermitian conjugate and \( \Delta k = |2k_1 - k_2| \) represents the phase mismatch between the fundamental and second harmonic beams. The linear (nonlinear) coupling constant, proportional to susceptibility \( \chi^{(1)} (\chi^{(2)}) \), is denoted by the parameter \( k (\Gamma) \). It is reasonable to assume \( \chi^{(2)} \ll \chi^{(1)} \) as in a real physical system we usually obtain \( \chi^{(2)}/\chi^{(1)} \approx 10^{-6} \). As a consequence, in absence of a highly strong pump \( \Gamma \ll k \). Earlier this model of contradirectional optical coupler was investigated by some of the present authors ([14] and references therein).

Using (1) and the procedure described in [14] we can obtain the coupled differential equations for three different modes as follows

\[
\frac{d a}{dz} = i k^* b_1, \quad \frac{d b_1}{dz} = -i k a - 2i \Gamma^* b_1^\dagger b_2 \exp (-i \Delta k z), \quad \frac{d b_2}{dz} = -i \Gamma b_2^\dagger \exp (i \Delta k z). \quad (2)
\]

Here it would be apt to mention that momentum operator for contradirectional asymmetric nonlinear coupler (1) is same as that of codirectional asymmetric nonlinear optical coupler [14]. However, for the contradirectional couplers the sign of derivative in the Heisenberg’s equation of motion of the contra-propagating mode is changed (i.e., in the present case \( \frac{d}{dz} \) is replaced by \( -\frac{d}{dz} \) as mode \( a \) is considered here as the contra-propagating mode). The method used here to obtain (2) is described in Refs. [14, 15]. Further, this particular description of contradirectional coupler is valid only for the situation when the forward propagating waves reach \( z = L \) and the counter (backward) propagating wave reach \( z = 0 \). Thus the coupled equations described by (2) and their solution obtained below are not valid for \( 0 < z < L \) [15]. Earlier these coupled equations (2) were solved under short-length approximation. Here we aim to obtain perturbative solution for these equations using Sen-Mandal method which is already shown to be useful in detecting nonclassical characters not identified by short-length solution [13, 19]. Keeping this in mind, we plan to solve (2) using Sen-Mandal approach.

Using Sen-Mandal approach we have obtained closed form perturbative analytic solutions of (2) as

\[
\begin{align*}
a(0) &= f_1 a(L) + f_2 b_1(0) + f_3 b_1^\dagger(0)b_2(0) + f_4 a^\dagger(L)b_2(0), \\
b_1(L) &= g_1 a(L) + g_2 b_1(0) + g_3 b_1^\dagger(0)b_2(0) + g_4 a^\dagger(L)b_2(0), \\
b_2(L) &= h_1 b_2(0) + h_2 b_1^\dagger(0) + h_3 b_1(0)a(L) + h_4 a^\dagger(L),
\end{align*}
\quad (3)
\]

with

\[
\begin{align*}
f_1 &= g_2 = \text{sech} |k| L, \\
f_2 &= -g_1^* = -\frac{i k^* \tanh |k| L}{|k|}, \\
f_3 &= C k^* \Delta k f_1^2 \{ 4 |k|^2 G_0^+ + \Delta k^2 (1 - 2 G_0^+ - 1 + \cosh 2 |k| L) G_0^+ - 1) \}, \\
f_4 &= 2 C k^* \Delta k f_1^2 \{ \Delta k \sinh |k| L G_0^- - 2 |k| \cosh |k| L G_0^- \}, \\
g_3 &= -2 C |k| f_1^2 \{ (\Delta k^2 + 2 |k|^2) \cosh |k| L G_0^- + i \Delta k |k| \sinh |k| L G_0^- \}, \\
g_4 &= C k^* \Delta k f_1^2 \{ \Delta k \sinh 2 |k| L (G_0^+ - 1) - 2 |k| (1 - \cosh 2 |k| L (G_0^+ - 1))) \}, \\
h_1 &= 1, \\
h_2 &= \frac{C |k|}{2} f_1 \{ 4 |k|^2 G_0^+ + \Delta k^2 (1 - 2 (G_0^+ - 1) + \cosh 2 |k| L) - 2 i \Delta k |k| \sinh 2 |k| L \}, \\
h_3 &= 2 C |k| f_1^2 \{ \Delta k |k| G_0^+ \cosh |k| L + [i \Delta k^2 - 2 i |k|^2 G_0^+] \sinh |k| L \}, \\
h_4 &= \frac{C |k|}{k} f_1 \{ 2 |k|^2 f_1 G_0^+ + \Delta k \sinh |k| L (G_0^+ - 1) [2 i |k| + \Delta k \tanh |k| L] \},
\end{align*}
\quad (4)
\]
where \( C = \frac{\Gamma^*}{|k\Delta k(\Delta k^2 + 3L^2)|} \) and \( G_\pm = (1 \pm \exp(-i\Delta kL)) \). The solution obtained above is verified by ESCR (Equal Space Commutation Relation) which implies \([a(0), a^\dagger(0)] = [b_1(L), b_1^*(L)] = [b_2(L), b_2^*(L)] = 1\) while all other equal space commutations are zero. Further, we have verified that the solutions reported here satisfies constant of motion. To be precise, in Ref. [15], it was shown that the constant of motion for the present system leads to

\[
 a^\dagger(0) a(0) + b_1^\dagger(L) b_1(L) + 2b_2^\dagger(L) b_2(L) = a^\dagger(L) a(L) + b_1^\dagger(0) b_1(0) + 2b_2^\dagger(0) b_2(0).
\]  

Here we have verified that the solution proposed here satisfies (5). We have also verified that the solution of contradirectional coupler using short-length solution method reported in [14] can be obtained as a special case of the present solution. Specifically, to obtain the short-length solution we need to expand the trigonometric functions present in the above solution and neglect all the terms beyond quadratic powers of \( L \) and consider phase mismatch \( \Delta k = 0 \). After doing so we obtain

\[
 f_1 = g_2 = (1 - \frac{k}{4}|k|^2 L^2), \quad f_2 = -g_4^* L, \quad f_3 = -g_4 = -\Gamma^* L^2, \\
 g_3 = -2\Gamma^* L, \quad h_2 = -i\Gamma L, \quad h_3 = -\Gamma k L^2 \text{ and } h_1 = 1, \quad f_4 = h_4 = 0,
\]  

which coincides with the short-length solution reported earlier by some of the present authors [14, 15]. Clearly the solution obtained here is valid and more general than the conventional short-length solution as the solution reported here is fully quantum solution and is valid for any length, restricting the coupling constant only. Further, in case of codirectional optical coupler we have already seen that several nonclassical phenomena not identified by short-length solution are identified by the perturbative solution obtained by Sen-Mandal method [13, 19]. Keeping this in mind, in what follows we investigate nonclassical characters of the fields that have propagated through a contradirectional asymmetric nonlinear optical coupler.

### 3 Criteria of nonclassicality

As the criteria for obtaining signatures of different nonclassical phenomena are expressed in terms of expectation values of functions of annihilation and creation operators of various modes, we can safely state that Eqs. (3) and (4) provide us sufficient resource for the investigation of the nonclassical phenomena. To illustrate this point we may note that the criteria for quadrature squeezing in single-mode \((j)\) and compound mode \((j, l)\) are [40]

\[
 (\Delta X_j)^2 < \frac{1}{4} \text{ or } (\Delta Y_j)^2 < \frac{1}{4},
\]  

and

\[
 (\Delta X_{jl;j\neq l})^2 < \frac{1}{4} \text{ or } (\Delta Y_{jl;j\neq l})^2 < \frac{1}{4},
\]  

where \( j, l \in \{a, b_1, b_2\} \) and the quadrature operators are defined as

\[
 X_a = \frac{1}{2}(a + a^\dagger), \quad Y_a = -\frac{1}{2}(a - a^\dagger),
\]  

and

\[
 X_{ab} = \frac{1}{2\sqrt{2}}(a + a^\dagger + b + b^\dagger), \quad Y_{ab} = -\frac{1}{2\sqrt{2}}(a - a^\dagger + b - b^\dagger).
\]

Similarly, the existence of single- and multi-mode nonclassical (sub-Poissonian) photon statistics can be obtained through the following inequalities

\[
 D_a = (\Delta N_a)^2 - \langle N_a \rangle < 0,
\]  

and

\[
 D_{ab} = (\Delta N_{ab})^2 - \langle a^\dagger b^\dagger ba \rangle - \langle a^\dagger a \rangle \langle b^\dagger b \rangle < 0,
\]  

where (11) provides us the condition for single-mode antibunching\(^2\) and (12) provides us the condition for intermodal antibunching. Many of the early investigations on nonclassicality were limited to the study of squeezing and antibunching only, but with the recent development of quantum computing and quantum information it has become very relevant to study entanglement. Interestingly, there exist a large number of inseparability criteria ([41, 42] and

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\ [2]To be precise, this zero-shift correlation is more connected to sub-Poissonian behavior. However, it is often referred to as antibunching [42].
references therein) that can be expressed in terms of expectation values of moments of field operators. For example, Hillery-Zubairy criterion I and II (HZ-I and HZ-II) [43, 44, 45] are described as

\[ \langle N_a N_b \rangle - \langle ab \rangle^2 < 0, \]  
(13)

and

\[ \langle N_a \rangle \langle N_b \rangle - \langle ab \rangle^2 < 0, \]  
(14)

respectively. Another, interesting criterion of inseparability that can be expressed in terms of moments of creation and annihilation operators is Duan et al.'s criterion which is described as follows [46]:

\[ d_{ab} = (\Delta u_{ab})^2 + (\Delta v_{ab})^2 - 2 < 0, \]  
(15)

where

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

\[ v_{ab} = -\frac{1}{\sqrt{2}} \left\{ (a - a^\dagger) + (b - b^\dagger) \right\}. \]  
(16)

Clearly our analytic solution (3)-(4) enables us to investigate intermodal entanglement using these criteria and a bunch of other criteria of nonclassicality that are described in Ref. [42]. Interestingly, all the inseparability criteria described above and in the remaining part of the present work can be viewed as special cases of Shchukin-Vogel entanglement criterion [47]. In Ref. [42, 48], Miranowicz et al. have explicitly established this point.

So far we have described criteria of nonclassicality that are related to the lowest order nonclassicality. However, nonclassicality may be witnessed via higher order criterion of nonclassicality, too. Investigations on higher order nonclassical properties of various optical systems have been performed since long. For example, in late seventies some of the present authors showed the existence of higher order nonclassical photon statistics in different optical systems using criterion based on higher order moments of number operators (cf. Ref. [14] and Chapter 10 of [49] and references therein). However, in those early works, higher order antibunching (HOA) was not specifically discussed, but existence of higher order nonclassical photon statistics was reported for degenerate and nondegenerate parametric processes in single and compound signal-idler modes, respectively and also for Raman scattering in compound Stokes-anti-Stokes mode up to \( n = 5 \). Criterion for HOA was categorically introduced by C. T. Lee [50] in 1990. Since then HOA is reported in several quantum optical systems ([51, 52] and references therein) and atomic systems [10]. However, except a recent effort by us [13] no effort has yet been made to study HOA in optical couplers. The existence of HOA can be witnessed through a set of equivalent but different criteria, all of which can be interpreted as modified Lee criterion. In what follows we will investigate the existence of HOA in the contradirectional optical coupler of our interest using a simple criterion of \( n \)-th order single-mode antibunching introduced by some of us [53]

\[ D_{a}(n) = \langle a^\dagger^n a^n \rangle - \langle a^\dagger a \rangle^n < 0. \]  
(17)

Here \( n = 2 \) and \( n \geq 3 \) refer to the usual antibunching and the higher order antibunching, respectively.

Similarly, we can also investigate the existence of higher order squeezing, but definition of higher order squeezing is not unique. To be precise, it is usually investigated using two different criteria [54, 58, 59]. The first criterion was introduced by Hong and Mandel in 1985 [58, 59] and a second criterion was subsequently introduced by Hillery in 1987 [54]. In Hong and Mandel criterion [58, 59], the reduction of higher order moments of usual quadrature operators with respect to their coherent state counterparts are considered as higher order squeezing. However, in Hillery’s criterion, reduction of variance of an amplitude powered quadrature variable for a quantum state with respect to its coherent state counterpart is considered as higher order squeezing. In what follows we have restricted our study on higher order squeezing to Hillery’s criterion of amplitude powered squeezing. Specifically, Hillery introduced amplitude powered quadrature variables as

\[ Y_{1,a} = \frac{a^n + (a^\dagger)^n}{2} \]  
(18)

and

\[ Y_{2,a} = i \frac{(a^\dagger)^n - a^n}{2}. \]  
(19)

It is easy to check that \( Y_{1,a} \) and \( Y_{2,a} \) do not commute and consequently we can obtain an uncertainty relation and thus a criterion of \( n \)-th order amplitude squeezing as

\[ A_{i,a} = (\Delta Y_{i,a})^2 - \frac{1}{2} |\langle [Y_{1,a}, Y_{2,a}] \rangle| < 0. \]  
(20)
Thus, for the specific case, \( n = 2 \), Hillery’s criterion for amplitude squared squeezing can be obtained as

\[
A_{i,a} = (\Delta Y_{i,a})^2 - \langle N_a + 1 \rangle / 2 < 0, \tag{21}
\]

where \( i \in \{1, 2\} \). Similarly, we can obtain specific criteria of amplitude powered squeezing for other values of \( n \). Further, there exists a set of higher order inseparability criteria. To be precise, all the criteria that are used for witnessing the existence of multi-partite (multi-mode) entanglement are essentially higher order criteria [60, 61, 62] as they always uncover some higher order correlation. Interestingly, even in bipartite (two-mode) case one can introduce operational criterion for detection of higher order entanglement. For example, Hillery-Zubairy introduced two criteria of higher order intermodality entanglement [43]

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

and

\[
E_{ab}^{m,n,l} = \langle (a^\dagger)^m a^m (b^\dagger)^n b^n \rangle - \langle (a^m b^n) \rangle^2 < 0.
\]

Here \( m \) and \( n \) are non-zero positive integers and the lowest possible values of \( m \) and \( n \) are \( m = n = 1 \) which reduces (22) and (23) to usual HZ-I criterion (i.e., (13)) and HZ-II criterion (i.e., (14)), respectively. Thus these two criteria may be viewed as generalized versions of the well known lower order criteria of Hillery and Zubairy (i.e., the criteria described in (13) and (14)). However, these generalized criterion can also be obtained as special cases of more general criterion of Shchukin and Vogel [47]. For the convenience of the readers, we refer to (22) and (23) as HZ-I criterion and HZ-II criterion respectively in analogy to the lowest order cases. In what follows, a quantum state will be called (bipartite) higher order entangled state if it is found to satisfy (22) and/or (23) for any choice of integers \( m \) and \( n \) satisfying \( m + n \geq 3 \). Existence of higher order entanglement can also be viewed through the criteria of multi-partite entanglement. For example, Li et al. [63] proved that a three-mode (tripartite) 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

\[
E_{ab_1|b_2}^{m,n,l} = \langle (a^\dagger)^m a^m (b_1^\dagger)^n b_1^n (b_2^\dagger)^l b_2^l \rangle - \langle (a^m b_1^n) (b_2^l) \rangle^2 < 0,
\]

where \( m, n, l \) are positive integers and annihilation operators \( a, b_1, b_2 \) correspond to the three modes. A quantum state that satisfy (24) is referred to as \( ab_1|b_2 \) entangled state. The three-mode inseparability criterion mentioned above is not unique. There exist various alternative criteria of three-mode entanglement. For example, an alternative criterion for detection of \( ab_1|b_2 \) entangled state is [63]

\[
E_{ab_1|b_2}^{m,n,l} = \langle (a^\dagger)^m a^m (b_1^\dagger)^n b_1^n (b_2^\dagger)^l b_2^l \rangle - \langle (a^m b_1^n b_2^l) \rangle^2 < 0.
\]

For \( m = n = l = 1 \), this criterion coincides with Miranowicz et al.’s criterion [48]. Using (24) and (25), we can easily obtain criteria for detection of \( ab_1|b_2 \) and \( b_1|ab_2 \) entangled states and use them to obtain a simple criterion for detection of fully entangled tripartite state. Specifically, using (24) and (25) respectively we can conclude that the three modes of our interest are not bi-separable in any form if one of the following two sets of inequalities are satisfied simultaneously

\[
E_{ab_1|b_2}^{1,1,1} < 0, E_{a|b_1 b_2}^{1,1,1} < 0, E_{b_1|b_2 a}^{1,1,1} < 0,
\]

\[
E_{ab_1|b_2}^{1,1,1} < 0, E_{a|b_1 b_2}^{1,1,1} < 0, E_{b_1|b_2 a}^{1,1,1} < 0.
\]

Further, for a fully separable pure state we always have

\[
|\langle ab_1 b_2 \rangle| = |\langle a \rangle \langle b_1 | b_2 \rangle| \leq \left[ \langle N_a \rangle \langle N_{b_1} \rangle \langle N_{b_2} \rangle \right]^{\frac{1}{2}}.
\]

Thus, a three-mode pure state that violates (28) (i.e., satisfies \( \langle N_a \rangle \langle N_{b_1} \rangle \langle N_{b_2} \rangle - |\langle ab_1 b_2 \rangle|^2 < 0 \)) and simultaneously satisfies either (26) or (27) is a fully entangled state as it is neither fully separable nor bi-separable in any form.
Spatial evolution of different operators that are relevant for witnessing nonclassicality can be obtained using the perturbative solutions (3)-(4) reported here. For example, using (3)-(4) we can obtain the following closed form expressions for number operators of various field modes

\[
N_a = a^\dagger a = |f_1|^2 a^\dagger (L) (aL) + |f_2|^2 b_1^\dagger (0) b_1(0) + \left[ f_1^* f_2 a^\dagger (L) b_1(0) + f_3^* f_4 a^\dagger (L) b_1^\dagger (0) b_2(0) \right] + f_4^* a^\dagger (L) b_2(0) + f_2^* f_3^* b_1^\dagger (0) b_2(0) + f_2^* f_4^* b_1(0) a^\dagger (L) b_2(0) + \text{h.c.}, \tag{29}
\]

\[
N_{b_1} = b_1^\dagger b_1 = |g_1|^2 a^\dagger (L) (aL) + |g_2|^2 b_1^\dagger (0) b_1(0) + \left[ g_1^* g_2 a^\dagger (L) b_1(0) + g_3^* g_4 a^\dagger (L) b_1^\dagger (0) b_2(0) \right] + g_4^* a^\dagger (L) b_2(0) + g_2^* g_3^* b_1^\dagger (0) b_2(0) + g_2^* g_4^* b_1(0) a^\dagger (L) b_2(0) + \text{h.c.}, \tag{30}
\]

\[
N_{b_2} = b_2^\dagger b_2 = b_2^\dagger (0) b_2(0) + \left[ h_2 b_2^\dagger (0) b_2^\dagger (0) + h_3 b_2^\dagger (0) b_1(0) a(L) + h_4 b_2^\dagger (0) a^\dagger (L) + \text{h.c.} \right]. \tag{31}
\]

It is now straightforward to compute the average values of the number of photons in different modes with respect to a given initial state. In the present work we consider that the initial state is 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. Thus,

\[
a(L)|\alpha\rangle|\beta\rangle|\gamma\rangle = a(\alpha)|\beta\rangle|\gamma\rangle, \tag{32}
\]

and $|\alpha|^2$, $|\beta|^2$, $|\gamma|^2$ are the number of input photons in the field modes $a$, $b_1$ and $b_2$, respectively. For a spontaneous process, $\beta = \gamma = 0$ and $\alpha \neq 0$. Whereas, for a stimulated process, the complex amplitudes are not necessarily zero and it seems reasonable to consider $\alpha > |\beta| > |\gamma|$. 

### 4.1 Single-mode and intermodal squeezing

Using (3)-(4) and (7)-(10) we obtain analytic expressions for variance in single-mode and compound mode quadratures as

\[
\begin{align*}
\frac{\langle \Delta X_a^2 \rangle}{\langle \Delta Y_a \rangle^2} &= \frac{1}{L} \left[ 1 \pm \{(f_1 f_4 + f_2 f_3) \gamma + \text{c.c.} \} \right], \\
\frac{\langle \Delta X_{b_1} \rangle^2}{\langle \Delta Y_{b_1} \rangle^2} &= \frac{1}{4} \left[ 1 \pm \{(g_1 g_4 + g_2 g_3) \gamma + \text{c.c.} \} \right], \tag{33} \\
\frac{\langle \Delta X_{b_2} \rangle^2}{\langle \Delta Y_{b_2} \rangle^2} &= \frac{1}{4},
\end{align*}
\]

and

\[
\begin{align*}
\frac{\langle \Delta X_{ab_1} \rangle^2}{\langle \Delta Y_{ab_1} \rangle^2} &= \frac{1}{4} \left[ 1 \pm \frac{1}{2} \{(f_1 + g_1)(f_4 + g_4) + (f_2 + g_2)(f_3 + g_3) \gamma + \text{c.c.} \} \right], \\
\frac{\langle \Delta X_{ab_2} \rangle^2}{\langle \Delta Y_{ab_2} \rangle^2} &= \frac{1}{4} \left[ 1 \pm \frac{1}{2} \{(f_1 + f_3)(f_4 + f_2) \gamma + \text{c.c.} \} \right] = \frac{1}{4} \left[ \frac{\langle \Delta X_a \rangle^2}{\langle \Delta Y_a \rangle^2} \right] + \frac{\gamma}{8}, \\
\frac{\langle \Delta X_{b_1 b_2} \rangle^2}{\langle \Delta Y_{b_1 b_2} \rangle^2} &= \frac{1}{4} \left[ 1 \pm \frac{1}{2} \{(g_1 + g_2) \gamma + \text{c.c.} \} \right] = \frac{1}{4} \left[ \frac{\langle \Delta Y_{b_1} \rangle^2}{\langle \Delta Y_{b_2} \rangle^2} \right] + \frac{\gamma}{8}, \tag{34}
\end{align*}
\]

respectively. From Eq. (33) it is clear that no squeezing is observed in $b_2$ mode. However, squeezing is possible in $a$ mode and $b_1$ mode as illustrated in Fig. 2 a-b, d-e. Further, we observed intermodal squeezing in quadratures $X_{ab_2}$ and $Y_{ab_2}$ by plotting right hand sides of Eq. (34) in Fig. 2 c and Fig. 2 f. Variation of amount of squeezing in different modes with phase mismatch $\Delta k$ and nonlinear coupling constant $\Gamma$ are shown in Fig. 2 a-c and Fig. 2 d-f, respectively. We have also studied the effect of linear coupling constant $k$ on the amount of squeezing, but its effect is negligible in all other quadratures except the quadratures of compound mode $ab_1$. In compound mode $ab_1$, after a short distance depth of squeezing is observed to increase with the decrease in the linear coupling constant $k$ (this is not illustrated through figure). Intermodal squeezing in compound mode quadratures $Y_{ab_2}$ and $X_{b_1 b_2}$ can be visualized from the last two rows of Eq.(34). Specifically, we can see that variance in compound mode quadrature $X_{j b_2}$ and $Y_{j b_2}$ have bijective (both one-to-one and onto) correspondence with the variance in $X_j$ and $Y_j$, respectively, where $j \in \{a, b_1\}$. To be precise, quadrature squeezing in single-mode $X_j(\gamma_j)$ implies quadrature squeezing in $X_{j b_2}(\gamma_{j b_2})$ and vice versa. For example, $\langle \Delta X_{j b_2} \rangle^2 < \frac{1}{4} \Rightarrow \frac{1}{4} \langle \Delta X_j \rangle^2 + \frac{\gamma}{8} < \frac{1}{4}$ or, $\langle \Delta X_j \rangle^2 < 2 \left( \frac{1}{4} - \frac{\gamma}{8} \right) = \frac{1}{4}$. This
is why we have not explicitly shown the variance of compound mode quadratures $X_{ab}$ and $Y_{ab}$ with different parameters as we have done for the other cases. As we have shown squeezing in $Y_a, X_{b_1}$, and $Y_{b_1}$ through Fig. 2 a-b, and c-d this implies the existence of squeezing in quadrature $Y_{ab_2}, X_{b_1b_2}$, and $Y_{b_1b_2}$. Thus we have observed intermodal squeezing in compound modes involving $b_2$. This nonclassical feature was not observed in earlier studies [14] as in those studies $b_2$ mode was considered classical. The plots do not show quadrature squeezing in $X_a, X_{ab_2}$, and $X_{ab_1}$ (for some specific values of $\Gamma$). However, a suitable choice of phase of the input coherent state would lead to squeezing in these quadratures. For example, if we replace $\gamma$ by $-\gamma$ (i.e., if we chose $\gamma = \exp(i\pi)$ instead of present choice of $\gamma = 1$) then we would observe squeezing in all these quadratures, but the squeezing that is observed now with the original choice of $\gamma$ would vanish. This is so as all the expressions of variance of quadrature variables that are $\neq \frac{1}{4}$ have a common functional form: $\frac{1}{4} \pm \gamma F(f_i, g_i)$ (c.f., Eqs. (33) and (34)).

4.2 Higher order squeezing

After establishing the existence of squeezing in single-modes and compound modes we now examine the possibility of higher order squeezing using Eqs. (3)-(4), (29)-(31) and (21). In fact, we obtain

$$\begin{bmatrix} A_{1,a} \\ A_{2,a} \end{bmatrix} = \pm \frac{\gamma}{4} \left[ \gamma (f_1f_4 + f_2f_3)(f_1\alpha + f_2\beta)^{2n-2} + c.c. \right],$$

(35)

$$\begin{bmatrix} A_{1,b_1} \\ A_{2,b_1} \end{bmatrix} = \pm \frac{\gamma}{4} \left[ \gamma (g_1g_4 + g_2g_3)(g_1\alpha + g_2\beta)^{2n-2} + c.c. \right],$$

(36)

and

$$\begin{bmatrix} A_{1,b_2} \\ A_{2,b_2} \end{bmatrix} = 0.$$  

(37)

Figure 2: (color online) Existence of quadrature squeezing in modes $a$ and $b_1$ and intermodal squeezing in mode $ab_1$, is illustrated with $k = 0.1, \alpha = 5, \beta = 2, \gamma = 1$ for various values of phase mismatching $\Delta k$ and nonlinear coupling constant $\Gamma$. In Fig. (a)-(c) squeezing and intermodal squeezing is plotted with rescaled interaction length $\Gamma L$ with $\Gamma = 0.001$ for $\Delta k = 10^{-1}$ (thin blue lines) and $\Delta k = 10^{-2}$ (thick red lines). In Fig. (d)-(f) squeezing and intermodal squeezing is plotted with interaction length $L$ with $\Delta k = 10^{-4}$ for $\Gamma = 0.001$ (thin blue lines) and $\Gamma = 0.01$ (thick red lines). In all the sub-figures a solid (dashed) line represents $X_i (Y_i)$ where $i \in \{a, b_1\}$ or $X_{ab_1}, (Y_{ab_1})$ quadrature. Parts of the plots that depict values of variance $< \frac{1}{4}$ in (a) and (d) show squeezing in quadrature variable $Y_a$, that in (b) and (c) show squeezing in quadrature variable $X_{b_1}, Y_{b_1}$ and intermodal squeezing in quadrature variable $X_{ab_1}, Y_{ab_1}$, respectively. Similarly, (e) and (f) show squeezing and intermodal squeezing in quadrature variable $X_{b_1}$ and $X_{ab_1}$ respectively. Squeezing in the other quadrature variables (say $X_a$) can be obtained by suitable choice of phases of the input coherent states.
Thus, we do not get any signature of amplitude powered squeezing in $b_2$ mode using the present solution. In contrary, mode $a$ ($b_1$) is found to show amplitude powered squeezing in one of the quadrature variables for any value of interaction length as both $A_{1,a}$ and $A_{2,a}$ ($A_{1,b_1}$ and $A_{2,b_1}$) cannot be positive simultaneously. To study the possibilities of amplitude powered squeezing in further detail we have plotted the spatial variation of $A_{i,a}$ and $A_{i,b_1}$ in Fig. 3. Negative regions of these two plots clearly illustrate the existence of amplitude powered squeezing in both $a$ and $b_1$ modes for $n = 2$ and $n = 3$. Extending our observations in context of single-mode squeezing and intermodal antibunching we can state that the appearance of amplitude powered squeezing in a particular quadrature can be controlled by suitable choice of phase of input coherent state $\gamma$ as the expressions for amplitude powered squeezing reported in (35) and (36) have a common functional form $\pm \gamma F(f_i, g_i)$.

4.3 Lower order and higher order antibunching

The condition of HOA is already provided through the inequality (17). Now using this inequality along with Eqns. (3)-(4) and (29)-(31) we can obtain closed form analytic expressions for $D_i(n)$ for various modes as follows

$$D_a(n) = nC_2\gamma |(f_1 \alpha + f_2 \beta)|^{2n-4} \left\{ (f_1 \alpha + f_2 \beta)^2 (f_2^* f_3^* + f_1^* f_4^*) + c.c. \right\},$$  \hspace{1cm} (38)

$$D_{b_1}(n) = nC_2\gamma |(g_1 \alpha + g_2 \beta)|^{2n-4} \left\{ (g_1 \alpha + g_2 \beta)^2 (g_2^* g_3^* + g_1^* g_4^*) + c.c. \right\},$$  \hspace{1cm} (39)

$$D_{b_2}(n) = 0.\hspace{1cm} (40)$$

Further, using the condition of intermodal antibunching described in (12) and Eqns. (3)-(4) we obtain following closed form expressions of $D_{ij}$

$$D_{ab_1} = \left\{ (|g_1|^2 f_1^* f_4 + f_1^* f_3 g_1^* g_2) \alpha^* \gamma + (|g_2|^2 f_2^* f_3 + f_2^* f_4 g_2^* g_1) \beta^* \gamma + (|g_1|^2 - |g_2|^2) (f_2^* f_4 - f_1^* f_3) \alpha \beta \gamma^* + c.c. \right\},$$  \hspace{1cm} (41)

$$D_{ab_2} = 0,$$  \hspace{1cm} (42)

$$D_{b_1b_2} = 0.\hspace{1cm} (43)$$

From the above expressions it is clear that neither the single-mode antibunching nor the intermodal antibunching is obtained involving $b_2$ mode. As the expressions obtained in the right hand sides of (38), (39) and (41) are not simple, we plot them to investigate the existence of single-mode and compound mode antibunching. The plots for usual antibunching and intermodal antibunching are shown in Fig. 4. Existence of antibunching is obtained in single-mode $a$ for $\gamma = 1$ and the same is illustrated through Fig. 4 a. However, in an effort to obtain antibunching in $b_1$ mode, we do not observe any antibunching in $b_1$ mode for $\gamma = 1$. Interestingly, from (39) it is clear that if we replace $\gamma = 1$ by $\gamma = \exp(i\pi) = -1$ as before and keep $\alpha, \beta$ unchanged, then we would observe antibunching for
all values of rescaled interaction length \( \Gamma L \). This is true in general for all values of \( \gamma \). To be precise, if we observe antibunching (bunching) in a mode for \( \gamma = c \) we will always observe bunching (antibunching) for \( \gamma = -c \), if we keep \( \alpha, \beta \) unchanged. This fact is illustrated through Fig. 4b where we plot variation of \( D_{b_i} \) with \( \Gamma L \) and have observed the existence of antibunching. In compound mode \( ab_1 \), we can observe existence of antibunching for both \( \gamma = 1 \) and \( \gamma = -1 \). However, in Fig. 4c we have illustrated the existence of intermodal antibunching in compound mode \( ab_1 \) by plotting variation \( D_{ab_i} \) with rescaled interaction length \( \Gamma L \) for \( \gamma = -1 \) as region of nonclassicality is relatively larger (compared to the case where \( \gamma = 1 \) and \( \alpha, \beta \) are same) in this case.

Now we may extend the discussion to HOA and plot right hand sides of (38) and (39) for various values of \( n \). The plots are shown in Fig. 5 which clearly illustrates the existence of HOA and also demonstrate that the depth of nonclassicality increases with \( n \). This is consistent with earlier observations on HOA in other systems [53].

### 4.4 Lower order and higher order intermodal entanglement

We first examine the existence of intermodal entanglement in compound mode \( ab_1 \) using HZ-I criterion (13). To do so we use Eqns. (3)-(4) and (29)-(31) and obtain

\[
E_{ab_1}^{1,1} = \langle N_a N_{b_1} \rangle - |\langle ab_1 \rangle|^2 = \langle |g_1|^2 f_1^* f_1 + f_2^* f_1 g_2^* g_2 \rangle \alpha^2 \gamma^* + \langle |f_1|^2 g_1^* g_4 + f_2^* g_2^* g_4 \rangle \alpha^2 \gamma - \langle |f_1|^2 g_2^* g_3 + f_2^* f_1^* g_3 \rangle \beta^2 \gamma^* + \langle |g_1|^2 - |g_2|^2 \rangle \{ (f_2^* f_3 - f_3^* f_2) \alpha \beta \gamma^* - (g_2^* g_4 - g_3^* g_4) \alpha^* \beta \gamma \}.
\]
Similarly, using (3)-(4) and (23) we can produce an analytic expression for $\alpha$\textsuperscript{1,1} and $\alpha'$\textsuperscript{1,1} (blue lines) and $\Delta\Gamma L$ for mode $ab_1$ with $k = 0.1$, $\Gamma = 0.001$, $\Delta k = 10^{-4}$, $\beta = 2$, $\gamma = 1$ for $\alpha = 3$ (thin blue lines) and $\alpha = 5$ (thick red lines).

Similarly, using HZ-II criterion (14) we obtain

$$E^{\prime1,1}_{ab_1} = \langle N_a \rangle - \langle N_b \rangle - |(ab_1)|^2$$

$$= - \left( (|g_1|^2 f_2 f_1 + f_3 f_1 g_2 g_1) \alpha^2 \gamma + (|f_1|^2 g_1^* g_4 + f_3^* f_2 g_1^* g_3) \alpha^2 \gamma ight)$$

$$+ \left( (g_1^* f_2 f_3 + f_3 f_2 g_1 g_2) \beta^2 \gamma + (|f_2|^2 g_2^* g_4 + f_3^* f_2 g_2^* g_3) \beta^2 \gamma ight)$$

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

(45)

It is easy to observe that Eqs. (44) and (45) provide us the following simple relation that is valid for the present case: $E^{\prime1,1}_{ab_1} = -E^{\prime1,1}_{ab_1}$, which implies that for any particular choice of rescaled interaction length $\Delta\Gamma L$ either HZ-I criterion or HZ-II criterion would show the existence of entanglement in contradirectional asymmetric nonlinear optical coupler as both of them cannot be simultaneously positive. Thus the compound mode $ab_1$ always entangled. The same is explicitly illustrated through Fig. 6. Similar investigations using HZ-I and HZ-II criteria in the other two compound modes (i.e., $ab_2$ and $b_1b_2$) failed to obtain any signature of entanglement in these cases. Further, signature of intermodal entanglement was not witnessed using Duan et al. criterion as using the present solution and (15) we obtain

$$d_{ab_1} = d_{ab_2} = d_{b_1b_2} = 0.$$  

(46)

However, it does not ensure separability of these modes as HZ-I, HZ-II and Duan et al. inseparability criteria are only sufficient and not necessary.

We may now study the possibilities of existence of higher order entanglement using Eqs. (22)-(28). To begin with, we use (3)-(4) and (22) to yield

$$E^{mn}_{ab_1} = \langle a^{m}b_{1}^{n}\rangle - \langle |a_{1}|^{2n}b_{1}\rangle$$

$$= \left( f_1 \alpha + f_2 \beta \right)^{2n-2} \left( |g_1 \alpha + g_2 \beta| \right)^2 E^{1,1}_{ab_1}.$$  

(47)

Similarly, using (3)-(4) and (23) we can produce an analytic expression for $E^{mn}_{ab_1}$ and observe that

$$E^{mn}_{ab_1} = -E^{mn}_{ab_1}.$$  

(48)

From relation (48), it is clear that the higher order entanglement between a mode and $b_1$ mode would always exist for any choice of $\Gamma L$, $m$ and $n$. This is so because $E^{mn}_{ab_1}$ and $E^{mn}_{ab_1}$ cannot be simultaneously positive. Using (47) and (48), it is a straightforward exercise to obtain analytic expressions of $E^{mn}_{ab_1}$ and $E^{mn}_{ab_1}$ for specific values of $m$ and $n$. Such analytic expressions are not reported here as the existence of higher order entanglement is clearly observed through (48). However, in Fig. 7 we have illustrated the variation of $E^{1,1}_{ab_1}$ and $E^{2,1}_{ab_1}$ with the rescaled interaction length, $\Gamma L$. Negative parts of the plots shown in Fig. 7 illustrate the existence of higher order intermodal entanglement in compound mode $ab_1$. As expected from (48), we observe that for any value of $\Gamma L$ compound mode $ab_1$ is higher order entangled. Further, it is observed that Hillery-Zubairy’s higher order entanglement criteria (22)-(23) fail to detect any signature of higher order entanglement in compound modes $ab_2$ and $b_1b_2$.

One can also investigate higher order entanglement using criterion of multi-partite (multi-mode) entanglement as all the multi-mode entangled states are essentially higher order entangled. Here we have only three modes in the coupler and thus we can study higher order entanglement by investigating the existence of three-mode
entanglement. A three-mode pure state that violates (28) (i.e., satisfies $\langle N_a \rangle \langle N_{b_1} \rangle \langle N_{b_2} \rangle - |\langle ab_1 b_2 \rangle|^2 < 0$) and simultaneously satisfies either (26) or (27) is a fully entangled state. Using (3)-(4) and (24)-(28) we obtain following set of interesting relations for $m = n = l = 1$:

$$E_{ab}^{1,1,1}|a\rangle|b_1 b_2\rangle = -E_{a|b_1 b_2}^{1,1,1} = E_{ab_2|b_1}^{1,1,1} = -E_{ab_1|b_2}^{1,1,1} = |\gamma|^2 E_{ab_1}^{1,1,1},$$  \hspace{1cm} (49)

$$E_{ab_1|b_2}^{1,1,1} = E_{ab_2|b_1}^{1,1,1} = 0,$$  \hspace{1cm} (50)

and

$$\langle N_a \rangle \langle N_{b_1} \rangle \langle N_{b_2} \rangle - |\langle ab_1 b_2 \rangle|^2 = -|\gamma|^2 E_{ab_1}^{1,1,1}.$$  \hspace{1cm} (51)

From (49), it is easy to observe that three modes of the coupler are not bi-separable in the form $a|b_1 b_2$ and $ab_2|b_1$ for any choice of rescaled interaction length $\Gamma L > 0$. Further, Eqn. (51) shows that the three modes of the coupler are not fully separable for $E_{ab_1}^{1,1} > 0$ (c.f. positive regions of plot of $E_{ab_1}^{1,1}$ shown in Fig. 6). However, (50) illustrate that the present solution does not show entanglement between coupled mode $ab_1$ and single-mode $b_2$. Thus the three modes present here are not found to be fully entangled. Specifically, three-mode (higher order) entanglement is observed here, but signature of fully entangled three-mode state is not observed. Further, we have observed that in all the figures depth of nonclassicality increases with $\alpha$.

In the present paper, various lower order and higher order nonclassical phenomena have been observed in contradirectional asymmetric nonlinear optical coupler. However, so far we have discussed only the stimulated cases as no nonclassical phenomenon is expected to be observed in spontaneous case. This is so because all the useful non-vanishing expressions for witnessing nonclassicality (i.e., Eqs. (35)-(51)) are proportional to $|\gamma|$. Thus all these expressions would vanish for $\gamma = 0$. 

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**Figure 7:** (color online) Higher order entanglement is observed using Hillery-Zubairy criteria. Solid lines show spatial variation of $E_{ab_1}^{2,1}$ and dashed lines show spatial variation of $E_{ab_1}^{2,1}$ with $k = 0.1$, $\Gamma = 0.001$, $\Delta k = 10^{-4}$, $\beta = 2$, $\gamma = 1$ for $\alpha = 3$ (thin blue lines) and $\alpha = 5$ (thick red lines). It is observed that depth of nonclassicality increases with increase in $\alpha$.

**Table 1:** Nonclassicalities observed in a contradirectional asymmetric nonlinear optical coupler that were not observed in earlier studies [15, 33].

| S.No. | Nonclassical phenomenon | Modes | Short-length approximation [15, 33] | Present work |
|-------|-------------------------|-------|------------------------------------|--------------|
| 1.    | Squeezing               | $b_2$ | Not investigated                    | Not observed |
| 2.    | Intermodal squeezing    | $b_1 b_2, ab_2$ | Not observed | Observed |
| 3.    | Amplitude squared squeezing | $a, b_1$ | Not investigated | Observed |
| 4.    | Amplitude squared squeezing | $b_2$ | Not investigated | Not observed |
| 5.    | Lower order and higher order intermodal entanglement | $ab_1$ | Not investigated | Observed |
| 6.    | Lower order and higher order intermodal entanglement | $b_1 b_2, ab_2$ | Not investigated | Not observed |
| 7.    | Three-mode (higher order) bi-separable entanglement | $a|b_1 b_2, ab_2|b_1$ | Not investigated | Observed |
| 8.    | Three-mode (higher order) bi-separable entanglement | $a|b_1 b_2$ | Not investigated | Not observed |
5 Conclusions

In the present study we report lower order and higher order nonclassicalities in a contradirectional asymmetric nonlinear optical coupler using a set of criteria of entanglement, single-mode squeezing, intermodal squeezing, antibunching, intermodal antibunching etc. Variation of nonclassicality with various parameters, such as number of input photon in the linear mode, linear coupling constant, nonlinear coupling constant and phase mismatch is also studied and it is observed that amount of nonclassicality can be controlled by controlling these parameters. The contradirectional asymmetric nonlinear optical coupler studied in the present work was studied earlier using a short-length solution and considering $b_2$ mode as classical [15, 33]. In contrast, a completely quantum mechanical solution of the equations of motion is obtained here using Sen-Mandal approach which is not restricted by length. The use of better solution and completely quantum mechanical treatment led to the identification of several nonclassical characters of a contradirectional asymmetric nonlinear optical coupler that were not reported in earlier studies. All such nonclassical phenomena that are observed here and were not observed in earlier studies are listed in Table 1. Further, there exist a large number of nonclassicality criteria that are not studied here and are based on expectation values of moments of annihilation and creation operators (c.f. Table I and II of Ref. [42]). As we already have compact expressions for the field operators it is a straightforward exercise to extend the present work to investigate other signatures of nonclassicality, such as, photon hyperbunching [64], sum and difference squeezing of An-Tinh [65] and Hillery [66], inseparability criterion of Manicini et al. [55], Simon [56] and Miranowicz et al. [57] etc. Further, the present work can be extended to investigate the nonclassical phenomena in other types of contradirectional optical couplers (e.g., contradirectional parametric coupler, contradirectional Raman coupler, etc.) that are either not studied till date or studied using short-length solution. Recently, Allevi et al. [67, 68] and Avenhaus et al. [69] have independently reported that they have experimentally measured $\langle a_{k1}^\dagger a_{k2}^\dagger a_{j1} a_{j2} \rangle$ which is sufficient to completely characterize bipartite multi-mode states. It is easy to observe that ability to experimentally measure $\langle a_{j1}^\dagger a_{k1}^\dagger a_{j2} a_{k2} \rangle$ ensures that we can experimentally detect signatures of nonclassicalities reported here. It is true that most of the nonclassicalities reported here can be observed in some other bosonic systems, too. However, the present system has some intrinsic advantages over most of the other systems as it can be used as a component in the integrated waveguide based structures in general and photonic circuits in particular [70, 71] and references therein. Thus the nonclassicalities reported in this easily implementable waveguide based system is expected to be observed experimentally. Further, the system studied here is expected to play important role as a source of nonclassical fields in the integrated waveguide based structures.

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