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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 waveguide and a nonlinear (quadratic) waveguide 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, and higher-order two-mode and multimode entanglement in the asymmetric nonlinear optical coupler. It is also shown that nonclassical properties of light can transfer from a nonlinear waveguide to a linear waveguide.

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I. INTRODUCTION

Several new applications of nonclassical states have been reported in recent past [1–6]. For example, applications of squeezed states are reported in the implementation of continuous variable quantum cryptography [1], teleportation of coherent states [2], etc.; antibunching is shown to be useful in building single photon sources [3]; and entangled states have 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 [4] and continuous variable quantum cryptography [1], quantum teleportation [5], dense coding [6], 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 [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 the 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 multiphoton entanglement in quantum circuits constructed using waveguides [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. Using similar arrangements of optical couplers the same group has also successfully implemented reconfigurable controlled two-qubit operation [10] and Shor's algorithm [11] on a photonic chip. In another interesting application, Mandal and Midda have shown that a universal irreversible gate library (NAND gate) can be built using nonlinear optical couplers [12]. Mandal and Midda's work essentially showed that, in principle, a classical computer can be built using optical couplers. Further, a directional optical coupler is one of the most important integrated guided wave components [13]. Thus if we can establish the possibility of generation of intermodal entanglement or any other nonclassicality in a directional optical coupler, that would imply the existence of another source of entanglement or other required nonclassical fields in a complex on-chip photonic circuit that can be used to perform a specific task related to quantum computation or quantum communication. In addition to the fact that waveguide based directional couplers can be realized easily, its potential adoptability in the integrated waveguide based photonic circuits provides it an edge over many other systems where nonclassical characters have already been studied as most of the atomic and optomechanical systems cannot be used in integrated quantum optic devices, such as in on-chip photonic circuits. Further, it is already established by several groups that integrated waveguide based structures are a better source of entanglement compared to those based on bulk crystal ([14], and references therein). 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 waveguide and a nonlinear (quadratic) waveguide operated by second harmonic generation. Waveguides interact with each other through evanescent waves and we may say that transfer of nonclassical effect from the nonlinear waveguide to the linear one happens through evanescent waves. The 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 have been studied in the past (see [15] for a review). For example, photon statistics, phase properties,

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and squeezing in codirectional and contradirectional Kerr nonlinear couplers are studied with fixed and varying linear coupling constants [16–20]; photon statistics of Raman and Brillouin couplers [21] and parametric couplers [22] is studied in detail; and photon statistics and other nonclassical properties of asymmetric [23–27] and symmetric [27–29] directional nonlinear couplers is investigated for various conditions such as strong pump [23], weak pump [25], phase mismatching [30] for codirectional [25,28,30] and contradirectional [26,28,30] propagation of classical (coherent) and nonclassical [27,29] input modes. However, almost all the earlier studies were limited to the investigation of lower-order nonclassical effects (e.g., squeezing and antibunching) either under the conventional short-length approximation [26] or under the parametric approximation where a pump mode is assumed to be strong and treated classically as a c number [30]. Only a few discrete efforts have recently been made to study higher-order nonclassical effects and entanglement in optical couplers [31–36], but even these efforts are limited to Kerr nonlinear couplers. For example, in 2004, Leonski and Miranowicz reported entanglement in Kerr nonlinear couplers [33]; subsequently, entanglement in pumped Kerr nonlinear optical couplers [34], entanglement sudden death [31], and thermally induced entanglement [32] are reported in Kerr nonlinear couplers. Amplitude squared (higher-order) squeezing is also reported in Kerr nonlinear couplers [36]. However, neither has any effort yet been made to rigorously study the higher-order nonclassical effects in nonlinear optical couplers in general, nor has a serious effort been made to study entanglement in nonlinear optical couplers other than Kerr nonlinear couplers. 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.

The remaining part of the paper is organized as follows. In Sec. II we briefly describe the momentum operator 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 momentum operator. In Sec. III we list a set of criteria of nonclassicality with special attention to those kinds of nonclassicalities that are never explored for asymmetric nonlinear optical coupler. In Sec. 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 Sec. V.

II. THE MODEL AND THE SOLUTIONS

An asymmetric nonlinear optical coupler is schematically shown in Fig. 1. We are interested in the 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 form 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

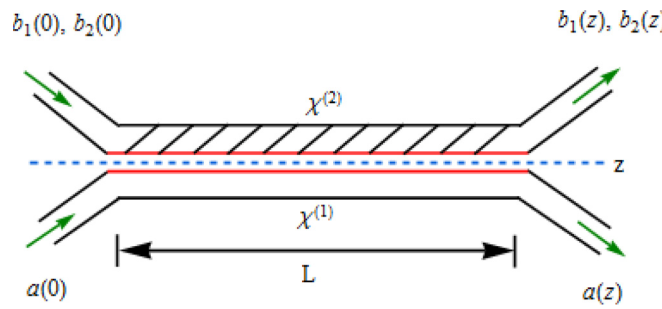

FIG. 1. (Color online) Schematic diagram of a codirectional asymmetric nonlinear optical coupler prepared by combining a linear waveguide with a nonlinear (quadratic) waveguide operated by second harmonic generation. The fields involved are described by the corresponding annihilation operators, as shown; L is the interaction length.

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

$$G = -\hbar k a b_1^\dagger - \hbar\Gamma b_1^2 b_2^\dagger \exp(i\Delta k z) + \text{H.c.}, \tag{1}$$

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 Γ 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 $\chi^{(2)}/\chi^{(1)} \simeq 10^{-6}$) 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 [15,24,25]. Specifically, in Ref. [24] 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 the short-length solution used in Ref. [24] was strictly restricted by the condition $\Gamma z \ll 1$. Later on Sen and Mandal developed a perturbative solution technique [37] that can solve Heisenberg's equations of motion for $\Gamma z \ll 1$. The Sen-Mandal technique was subsequently used in Ref. [25] to obtain spatial evolution of field operators corresponding to (1) and to study single-mode and intermodal squeezing and antibunching. Interestingly, in [25] some nonclassical characters of asymmetric nonlinear optical coupler were observed which were not observed in the earlier investigations [15,24] performed using short-length solution. This was indicative of the fact that the Sen-Mandal perturbative method provides better solution[1] 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

[1]In fact, short-length (time) solution can be obtained as a special case of Sen-Mandal perturbative solution.

of the higher-order nonclassical properties were studied in earlier papers. Keeping these facts in mind we have used the solution reported in Ref. [25] to study the higher-order nonclassicalities.

In Ref. [25] closed form analytic expressions for evolution of field operators valid up to linear power of the coupling coefficient Γ were obtained as follows:

$$\begin{aligned} a(z) &= f_1 a(0) + f_2 b_1(0) + f_3 b_1^{\dagger}(0) b_2(0) + f_4 a^{\dagger}(0) b_2(0), \\ b_1(z) &= g_1 a(0) + g_2 b_1(0) + g_3 b_1^{\dagger}(0) b_2(0) + g_4 a^{\dagger}(0) b_2(0), \\ b_2(z) &= h_1 b_2(0) + h_2 b_1^2(0) + h_3 b_1(0) a(0) + h_4 a^2(0), \end{aligned} \tag{2}$$

where

$$\begin{aligned} f_1 &= g_2 = \cos|k|z, \quad f_2 = -g_1* = -\frac{ik^*}{|k|}\sin|k|z, \\ f_3 &= \frac{2k^*\Gamma^*}{4|k|^2-(\Delta k)^2}\left[G_- f_1 + \frac{f_2}{k^*}\left\{\Delta k - \frac{2|k|^2}{\Delta k}G_-\right\}\right], \\ f_4 &= \frac{4k^{*2}\Gamma^*}{\Delta k[4|k|^2-(\Delta k)^2]}G_- f_1 + \frac{2k^*\Gamma^*}{[4|k|^2-(\Delta k)^2]}G_+ f_2, \\ g_3 &= \frac{2\Gamma^* k}{[4|k|^2-(\Delta k)^2]}G_+ f_2 - \frac{2\Gamma^*(2|k|^2-(\Delta k)^2) f_1}{\Delta k[4|k|^2-(\Delta k)^2]}G_-, \\ g_4 &= \frac{4\Gamma^*|k|^2}{\Delta k[4|k|^2-(\Delta k)^2]}f_2 - \frac{2\Gamma^*(2|k|^2-(\Delta k)^2)}{\Delta k[4|k|^2-(\Delta k)^2]} \\ &\quad \times (G_+ - 1) f_2 + \frac{2k^*\Gamma^*}{[4|k|^2-(\Delta k)^2]}G_- f_1, \\ h_1 &= 1, \\ h_2 &= \frac{\Gamma G_-^*}{2\Delta k} - \frac{i\Gamma}{2[4|k|^2-(\Delta k)^2]}\{2|k|(G_+^* - 1)\sin 2|k|z \\ &\quad - i\Delta k[1-(G_+^*-1)\cos 2|k|z]\}, \\ h_3 &= \frac{-\Gamma|k|}{k^*[4|k|^2-(\Delta k)^2]}\{i\Delta k(G_+^*-1)\sin 2|k|z \\ &\quad + 2|k|[1-(G_+^*-1)\cos 2|k|z]\}, \\ h_4 &= -\frac{\Gamma|k|^2 G_-^*}{2k^{*^2}\Delta k} - \frac{i\Gamma|k|^2}{2k^{*^2}[4|k|^2-(\Delta k)^2]}\{2|k|(G_+^*-1) \\ &\quad \times \sin 2|k|z - i\Delta k[1-(G_+^*-1)\cos 2|k|z]\}, \end{aligned} \tag{3}$$

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. [25].

III. CRITERIA OF NONCLASSICALITY

A state having negative or highly singular (more singular than δ 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 have been proposed in the 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) and (3) provide us with the sufficient mathematical framework required to study the nonclassical properties of the codirectional asymmetric nonlinear optical coupler. As mentioned above we are interested in the higher-order nonclassical properties of radiation fields. 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 ([38], and references therein). Until recently, past studies on higher-order nonclassicalities were predominantly restricted to theoretical investigations. However, a bunch of exciting experimental demonstrations of higher-order nonclassicalities have been recently reported [39–41]. Specifically, the existence of higher-order nonclassicality in bipartite multimode states produced in a twin-beam experiment has been recently demonstrated by Allevi, Olivares, and Bondani [39] 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 [39]. This observation was consistent with the earlier theoretical observation of Pathak and Garcia [42] that established that the 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 has 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 [43–45]. In the first approach introduced by Hillery in 1987 [43] 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 [44,45], 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

$$Y_{1,a} = \frac{a^k + (a^{\dagger})^k}{2} \tag{4}$$

and

$$Y_{2,a} = i\left(\frac{(a^{\dagger})^k - a^k}{2}\right). \tag{5}$$

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

$$A_{i,a} = \langle(\Delta Y_{i,a})^2\rangle - \left\langle N_a + \tfrac{1}{2}\right\rangle < 0, \tag{6}$$

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

B. Higher-order antibunching

Since 1977 signatures of higher-order nonclassical photon statistics (HONPS) 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. [15] and Chap. 10 of [46], 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 value of the parameter that describes HONPS decreases with increasing n occurring on a shorter time interval in parametric processes, whereas different order HONPS occurs on the same time interval in Raman scattering. A specific criterion for HOA was first introduced by Lee [47] in 1990 using higher-order moments of number operators. 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 [48]. Since then HOA has been reported in several quantum optical systems ([38], and references therein) and atomic systems [49]. However, no effort has yet been made to study HOA in optical couplers. Thus the present study of HOA in asymmetric nonlinear optical couplers is expected to lead to similar observations in other types 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 the following simple criterion of nth-order single-mode antibunching introduced by Pathak and Garcia [42]:

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

Here $n = 2$ corresponds to the usual antibunching and $n \geqslant 3$ refers to the higher-order antibunching. Here it would be apt to note that the term "higher-order antibunching" was coined by Lee in his pioneering work [47] in 1990. In Ref. [47], Lee used $D_a(2) = \langle a^{\dagger 2} a^2\rangle - \langle a^\dagger a\rangle^2 < 0$ as the condition of antibunching and supported his choice by citing a 1959 work of Mandel [50], and Lee stated, "The correspondence between antibunching and sub-Poisson distribution has been established by Mandel through the so-called Poisson transform. Therefore, we consider antibunching and the sub-Poissonian distribution as equivalent. In this paper." This is how the term HOA originated. The same notion of HOA was used in all the future works on this topic [38,42,48,49,51], and in the present work we have also followed the same convention. However, for $n = 2$, Lee's criterion yields a condition of nonclassical state as $\langle a^{\dagger 2}(t) a^2(t)\rangle - \langle a^\dagger(t) a(t)\rangle^2 < 0$, which is more related to sub-Poisson behavior. Technically, it is more appropriate to characterize antibunching by the criterion $g^{(2)}(\tau) > g^{(2)}(0)$ where $g^{(2)}(\tau) = \frac{\langle a^\dagger(t) a^\dagger(t+\tau) a(t+\tau) a(t)\rangle}{\langle a^\dagger(t) a(t)\rangle \langle a^\dagger(t+\tau) a(t+\tau)\rangle}$ and to characterize the sub-Poissonian photon statistics by $g^{(2)}(0) = \frac{\langle a^\dagger(t) a^\dagger(t) a(t) a(t)\rangle}{\langle a^\dagger(t) a(t)\rangle\langle a^\dagger(t) a(t)\rangle} < 1$ [52] or equivalently by the criterion $D_a(2) = \langle a^{\dagger 2}(t) a^2(t)\rangle - \langle a^\dagger(t) a(t)\rangle^2 < 0$ as is used in the present paper. It is interesting to note that following the same convention as was adopted in [47], several other authors [53,54] have also used $D_a(2) < 0$ as the criterion of antibunching. This specific choice of antibunching criterion can be justified if we note that for any finite bandwidth electromagnetic field $g^{(2)}(\tau) \to 1$ for sufficiently long time scale ($\tau \to \infty$) [54]. This is so because if $g^{(2)}(\tau) = 1$, then $g^{(2)}(0) < 1$ or $D_a(2) < 0$ essentially implies $g^{(2)}(\tau) > g^{(2)}(0)$. Thus for any finite bandwidth electromagnetic field which satisfies $g^{(2)}(0) < 1$ or $D_a(2) < 0$ will always show antibunching for some time scale. Further, the criterion of higher-order sub-Poissonian photon statistics (HOSPS) is different from (7) for all values of $n \geqslant 3$ ([38], and references therein). Thus, although (7) reduces to the criterion of sub-Poissonian photon statistics for $n = 2$, it does not reduce to the criterion of HOSPS for $n \geqslant 3$ and consequently it is not a criterion of HOSPS. However, it gives a nonclassical photon statistics as shown by Lee [47], and following Lee the specific nonclassical character revealed by this criterion is traditionally being referred to as HOA [38,42,47–49,51] and in what follows we have also used the same convention.

C. Entanglement and higher-order entanglement

There exist several inseparability criteria ([55], and references therein) that are expressed in terms of expectation values of field operators and thus suitable for study of entanglement dynamics within the framework of the present approach. Among these criteria, the criterion of Duan *et al.* [56], which is usually referred to as Duan's criterion, and Hillery-Zubairy criterion I and II (HZ-I and HZ-II) [57–59] have received more attention for various reasons, such as computational simplicity, experimental realizability, and their recent success in detecting entanglement in various optical, atomic, and optomechanical systems ([8,49], and references therein). To begin with we may note that the first inseparability criterion of Hillery and Zubairy, i.e., the HZ-I criterion of inseparability, is described as

$$\langle N_a N_b\rangle - |\langle a b^\dagger\rangle|^2 < 0, \tag{8}$$

whereas the second criterion of Hillery and Zubairy, i.e., the 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 the criterion of Duan *et al.*, which is described as follows [56]:

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

where

$$\begin{aligned} u_{ab} &= \frac{1}{\sqrt{2}}\{(a + a^\dagger) + (b + b^\dagger)\},\\ v_{ab} &= -\frac{i}{\sqrt{2}}\{(a - a^\dagger) + (b - b^\dagger)\}. \end{aligned} \tag{11}$$

Clearly our analytic solution (2)–(3) enable us to investigate intermodal entanglement in asymmetric nonlinear optical coupler using all three inseparability criteria described above and a set of other criteria listed in [60]. It is interesting to note that all the inseparability criteria described above and in the rest of the paper are special cases of the Shchukin-Vogel inseparbility criterion [61]. Miranowicz *et al.* have clearly established this point in Refs. [60,62]. As all three inseparability criteria that are explicitly described here 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 bipartite entanglement of lowest order. As the possibility of generation of entanglement in asymmetric nonlinear optical coupler has not been 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 multipartite entanglement are essentially higher-order criteria [63–65] as they reveal some higher-order correlation. Interestingly, there exist higher-order inseparability criteria for detection of higher-order entanglement in the bipartite case, too. Specifically, Hillery and Zubairy introduced two criteria for intermodal higher-order entanglement [57] as follows:

$$E_{ab}^{m,n} = \langle (a^\dagger)^m a^m (b^\dagger)^n b^n\rangle - |\langle a^m (b^\dagger)^n\rangle|^2 < 0 \quad (12)$$

and

$$E_{ab}^{'m,n} = \langle (a^\dagger)^m a^m\rangle\langle (b^\dagger)^n b^n\rangle - |\langle a^m b^n\rangle|^2 < 0. \quad (13)$$

Here m and n are nonzero positive integers and the 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 a generalized version of the well-known lower-order criteria of Hillery and Zubairy and we may refer to (12) and (13) as the HZ-I criterion and the HZ-II criterion, respectively, in analogy to the lowest-order cases. A quantum state will be referred to as a (bipartite) higher-order entangled state if it is found to satisfy (12) and/or (13) for any choice of integer m and n satisfying $m + n \geqslant 3$. The other type of higher-order entanglement i.e., multipartite entanglement can be detected in various ways. In the present paper we have used a set of multimode inseparability criteria introduced by Li *et al.* [66]. Specifically, Li *et al.* have shown that a three-mode quantum state is not biseparable 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:

$$E_{ab_1|b_2}^{m,n,l} = \left\langle (a^\dagger)^m a^m (b_1^\dagger)^n b_1^n (b_2^\dagger)^l b_2^l\right\rangle - \left|\left\langle a^m b_1^n (b_2^\dagger)^l\right\rangle\right|^2 < 0, \quad (14)$$

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 the $ab_1|b_2$ entangled state. The three-mode inseparability criterion can be written in various alternative forms. For example, an alternative criterion for detection of the $ab_1|b_2$ entangled state is [66]

$$E_{ab_1|b_2}^{\prime m,n,l} = \left\langle (a^\dagger)^m a^m (b_1^\dagger)^n b_1^n\right\rangle\left\langle (b_2^\dagger)^l b_2^l\right\rangle - \left|\left\langle a^m b_1^n b_2^l\right\rangle\right|^2 < 0. \quad (15)$$

Similarly, one can define the criteria for detection of $a|b_1b_2$ and $b_1|ab_2$ entangled states and use them to obtain the criterion for detection of fully entangled tripartite states. For example, using (14) and (15), respectively, we can write that the three modes of our interest are not biseparable in any form if any one of the following two sets of inequalities is satisfied simultaneously:

$$E_{ab_1|b_2}^{1,1,1} < 0, \qquad E_{a|b_1b_2}^{1,1,1} < 0, \qquad E_{b_1|b_2a}^{1,1,1} < 0, \quad (16)$$

$$E_{ab_1|b_2}^{\prime 1,1,1} < 0, \qquad E_{a|b_1b_2}^{\prime 1,1,1} < 0, \qquad E_{b_1|b_2a}^{\prime 1,1,1} < 0. \quad (17)$$

Further, for a fully separable pure state we always have

$$|\langle ab_1b_2\rangle| = |\langle a\rangle\langle b_1\rangle\langle b_2\rangle| \leqslant \left[\langle N_a\rangle\langle N_{b_1}\rangle\langle N_{b_2}\rangle\right]^{1/2}. \quad (18)$$

Thus a three-mode pure state that violates (18) (i.e., satisfies $\langle N_a\rangle\langle N_{b_1}\rangle\langle N_{b_2}\rangle - |\langle ab_1b_2\rangle|^2 < 0$) and simultaneously satisfies either (16) or (17) is a fully entangled state as it is neither fully separable nor biseparable in any form.

IV. NONCLASSICALITY IN CODIRECTIONAL OPTICAL COUPLER

Using the perturbative solutions (2)–(3) we can obtain spatial evolution of various operators that are relevant for the detection of nonclassical characters. For example, we may use (2)–(3) to obtain the number operators for various field modes as follows:

$$\begin{aligned} N_a = a^\dagger a &= |f_1|^2 a^\dagger(0)a(0) + |f_2|^2 b_1^\dagger(0)b_1(0) \\ &+ \big[f_1^* f_2 a^\dagger(0)b_1(0) + f_1^* f_3 a^\dagger(0)b_1^\dagger(0)b_2(0) \\ &+ f_1^* f_4 a^{\dagger 2}(0)b_2(0) + f_2^* f_3 b_1^{\dagger 2}(0)b_2(0) \\ &+ f_2^* f_4 b_1^\dagger(0)a^\dagger(0)b_2(0) + \text{H.c.}\big], \end{aligned} \quad (19)$$

$$\begin{aligned} N_{b_1} = b_1^\dagger b_1 &= |g_1|^2 a^\dagger(0)a(0) + |g_2|^2 b_1^\dagger(0)b_1(0) \\ &+ \big[g_1^* g_2 a^\dagger(0)b_1(0) + g_1^* g_3 a^\dagger(0)b_1^\dagger(0)b_2(0) \\ &+ g_1^* g_4 a^{\dagger 2}(0)b_2(0) + g_2^* g_3 b_1^{\dagger 2}(0)b_2(0) \\ &+ g_2^* g_4 b_1^\dagger(0)a^\dagger(0)b_2(0) + \text{H.c.}\big], \end{aligned} \quad (20)$$

$$\begin{aligned} N_{b_2} = b_2^\dagger b_2 &= b_2^\dagger(0)b_2(0) + \big[h_2 b_2^\dagger(0)b_1^2(0) \\ &+ h_3 b_2^\dagger(0)b_1(0)a(0) + h_4 b_2^\dagger(0)a^2(0) + \text{H.c.}\big]. \end{aligned} \quad (21)$$

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 the initial state is a product of three coherent states: $|\alpha\rangle|\beta\rangle|\gamma\rangle$, where $|\alpha\rangle, |\beta\rangle$, and $|\gamma\rangle$ are eigenkets of annihilation operators a, b_1, and b_2, respectively. Field operator $a(0)$ operating on such a multimode coherent state yields a complex eigenvalue α. Specifically,

$$a(0)|\alpha\rangle|\beta\rangle|\gamma\rangle = \alpha|\alpha\rangle|\beta\rangle|\gamma\rangle, \quad (22)$$

where $|\alpha|^2, |\beta|^2, |\gamma|^2$ is the number of input photons in the field modes a, b_1, and b_2, respectively. For a spontaneous process,

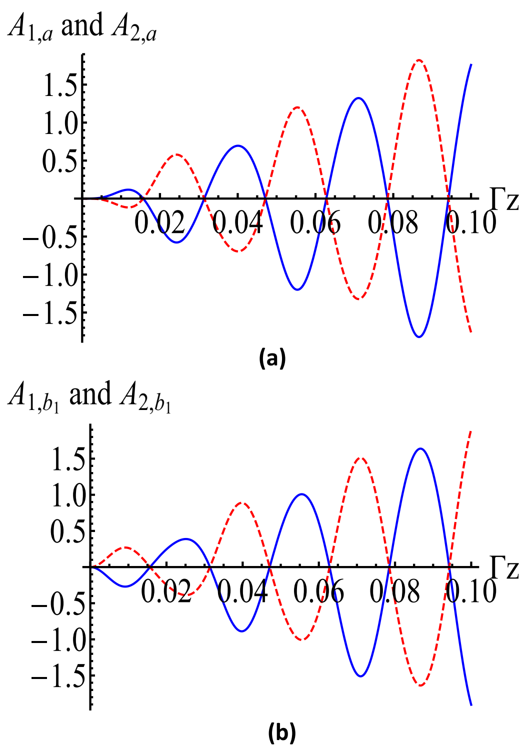

FIG. 2. (Color online) Amplitude squared squeezing is observed in modes (a) a and (b) b_1 for the initial state $|\alpha\rangle|\beta\rangle|\gamma\rangle$ with $k = 0.1$, $\Gamma = 0.001$, $\Delta k = 10^{-4}$, $\alpha = 5$, $\beta = 2$, and $\gamma = 1$. Negative parts of the solid line represent squeezing in quadrature variable $Y_{1,a}$ (Y_{1,b_1}) and that of the dashed line represent squeezing in quadrature variable $Y_{2,a}$ (Y_{2,b_1}).

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. 3) that illustrate the existence of nonclassical character in asymmetric nonlinear optical coupler we have chosen $\alpha = 5$, $\beta = 2$, and $\gamma = 1$.

A. Higher-order squeezing

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

$$\begin{bmatrix} A_{1,a} \\ A_{2,a} \end{bmatrix} = \pm\big[(f_1 f_4 + f_2 f_3)\big(f_1^2\alpha^2\gamma + f_2^2\beta^2\gamma + 2 f_1 f_2 \alpha\beta\gamma\big) + \text{c.c.}\big], \tag{23}$$

$$\begin{bmatrix} A_{1,b_1} \\ A_{2,b_1} \end{bmatrix} = \pm\big[(g_1 g_4 + g_2 g_3)\big(g_1^2\alpha^2\gamma + g_2^2\beta^2\gamma + 2 g_1 g_2 \alpha\beta\gamma\big) + \text{c.c.}\big], \tag{24}$$

and

$$\begin{bmatrix} A_{1,b_2} \\ A_{2,b_2} \end{bmatrix} = 0. \tag{25}$$

Clearly we do not 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_1}) 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 $A_{i,a}$ and A_{i,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:

$$D_a(n) = {}^nC_2\gamma|(f_1\alpha + f_2\beta)|^{2n-4} \times \{(f_1\alpha + f_2\beta)^2(f_2^* f_3^* + f_1^* f_4^*) + \text{c.c.}\}, \tag{26}$$

$$D_{b_1}(n) = {}^nC_2\gamma|(g_1\alpha + g_2\beta)|^{2n-4} \times \{(g_1\alpha + g_2\beta)^2(g_2^* g_3^* + g_1^* g_4^*) + \text{c.c.}\}, \tag{27}$$

$$D_{b_2}(n) = 0. \tag{28}$$

Clearly, the perturbative solution used here cannot detect any signature of higher-order antibunching for the b_2 mode. However, in the other two modes we observe HOA for different values of n as illustrated in Fig. 3. In Fig. 3, we have plotted the right-hand sides of (26) and (27) along with the exact numerical results obtained by integrating the z-dependent Schrödinger equation corresponding to a given momentum operator by using the matrix form of the operators. Close resemblance of the exact numerical result with the perturbative result even for the higher-order case clearly validates the perturbative solution used here.

C. Intermodal entanglement

To apply the 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

$$\begin{aligned} E_{ab_1}^{1,1} &= \langle N_a N_{b_1}\rangle - |\langle a b_1^\dagger\rangle|^2 \\ &= (|g_1|^2 f_4^* f_1 + f_3^* f_1 g_2^* g_1)\alpha^2\gamma^* \\ &\quad + (|f_1|^2 g_1^* g_4 + f_1^* f_2 g_1^* g_3)\alpha^{*2}\gamma \\ &\quad + (|g_2|^2 f_3^* f_2 + f_4^* f_2 g_1^* g_2)\beta^2\gamma^* \\ &\quad + (|f_2|^2 g_2^* g_3 + f_2^* f_1 g_2^* g_4)\beta^{*2}\gamma \\ &\quad + (|g_1|^2 - |g_2|^2)((f_4^* f_2 - f_3^* f_1)\alpha\beta\gamma^* \\ &\quad - (g_2^* g_4 - g_1^* g_3)\alpha^*\beta^*\gamma). \end{aligned} \tag{29}$$

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

$$\begin{aligned} E_{ab_1}^{\prime 1,1} &= \langle N_a\rangle\langle N_{b_1}\rangle - |\langle a b_1\rangle|^2 \\ &= -[(|g_1|^2 f_4^* f_1 + f_3^* f_1 g_2^* g_1)\alpha^2\gamma^* \\ &\quad + \{|f_1|^2 g_1^* g_4 + f_1^* f_2 g_1^* g_3)\alpha^{*2}\gamma \end{aligned}$$

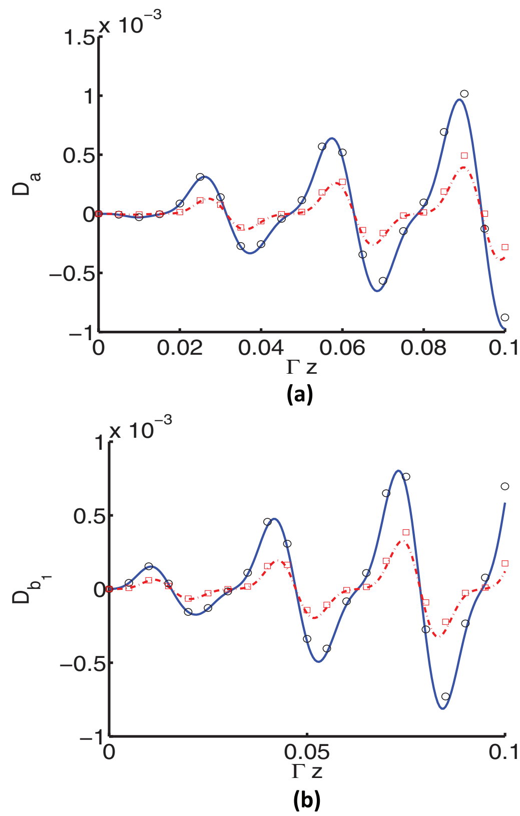

FIG. 3. (Color online) HOA with rescaled interaction length Γz in mode (a) a and (b) mode b_1 for $n=3$ (smooth line) and $n=4$ (dashed line), and squares and circles are for the corresponding numerical results with the initial state $|\alpha\rangle|\beta\rangle|\gamma\rangle$ and $k=0.1$, $\Gamma=0.001$, $\Delta k=10^{-4}$, $\alpha=0.5$, $\beta=0.2$, and $\gamma=0.1$.

$$
\begin{aligned}
&+(|g_2|^2 f_3^* f_2 + f_4^* f_2 g_1^* g_2)\beta^2\gamma^* \\
&+(|f_2|^2 g_2^* g_3 + f_2^* f_1 g_2^* g_4)\beta^{*2}\gamma \\
&+(|g_1|^2 - |g_2|^2)[(f_4^* f_2 - f_3^* f_1)\alpha\beta\gamma^* \\
&-(g_2^* g_4 - g_1^* g_3)\alpha^*\beta^*\gamma]\}. \quad (30)
\end{aligned}
$$

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 point inside the coupler either the HZ-I criterion or the HZ-II criterion would show the existence of entanglement as both of them cannot be simultaneously positive. Thus compound mode ab_1 is always entangled inside a codirectional asymmetric optical coupler. The same is explicitly illustrated through Fig. 4. Following the same approach we investigated the existence of entanglement in other compound modes (e.g., ab_2 and b_1b_2), but both the 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 the 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 the criterion of Duan *et al.* or vice versa [8,49]. Keeping these facts in mind, we studied the possibilities of observing intermodal entanglement using the criterion of Duan *et al.*, too, but it failed to detect

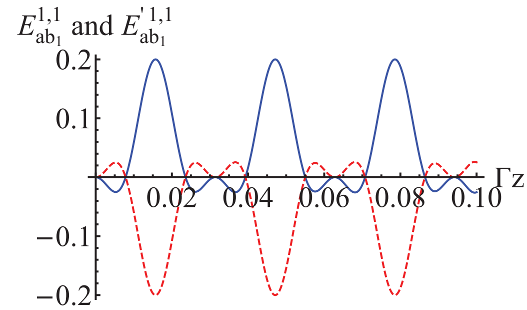

FIG. 4. (Color online) Hillery-Zubairy criterion I (solid line) and criterion II (dashed line) for entanglement are showing intermodal entanglement between modes a and b_1. Here $E_{ab_1}^{1,1}$ (solid line) and $E_{ab_1}^{\prime 1,1}$ (dashed line) are plotted with rescaled interaction length Γz for mode ab_1 with the initial state $|\alpha\rangle|\beta\rangle|\gamma\rangle$ and $k=0.1$, $\Gamma=0.001$, $\Delta k=10^{-4}$, $\alpha=5$, $\beta=2$, and $\gamma=1$.

any entanglement in the present case as we obtained

$$d_{ab_1} = d_{ab_2} = d_{b_1b_2} = 0. \quad (31)$$

We may now investigate the existence of higher-order entanglement using Eqs. (12)–(18). To begin with we may use (2)–(3) and (12) to obtain

$$
\begin{aligned}
E_{ab_1}^{m,n} &= \langle a^{\dagger m} a^m b_1^{\dagger n} b_1^n\rangle - |\langle a^m b_1^{\dagger n}\rangle|^2 \\
&= mn|(f_1\alpha + f_2\beta)|^{2m-2}|(g_1\alpha + g_2\beta)|^{2n-2} E_{ab_1}^{1,1}. \quad (32)
\end{aligned}
$$

Similarly, using (2)–(3) and (13) we can obtain a closed form analytic expression for $E_{ab_1}^{\prime m,n}$ and easily observe that

$$E_{ab_1}^{\prime m,n} = -E_{ab_1}^{m,n}. \quad (33)$$

Equation (33) clearly shows that higher-order entanglement between the a mode and b_1 mode would always be observed for any choice of m and n as $E_{ab_1}^{m,n}$ and $E_{ab_1}^{\prime m,n}$ cannot be simultaneously positive. Using (32) and (33) we can easily obtain analytic expressions of $E_{ab_1}^{2,1}, E_{ab_1}^{\prime 2,1}$, $E_{ab_1}^{\prime 1,2}$, etc. Such analytic expressions are not reported here as existence of higher-order entanglement is clearly seen through (33). However, in Fig. 5 we have illustrated the spatial evolution of $E_{ab_1}^{2,1}$ and $E_{ab_1}^{\prime 2,1}$. Negative regions of this figure clearly show the existence of higher-order intermodal entanglement in

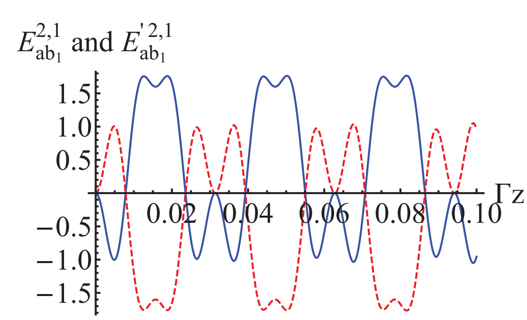

FIG. 5. (Color online) Higher-order entanglement is observed using the Hillery-Zubairy criteria. The solid line shows spatial variation of $E_{ab_1}^{2,1}$ and the dashed line shows spatial variation of $E_{ab_1}^{\prime 2,1}$ with the initial state $|\alpha\rangle|\beta\rangle|\gamma\rangle$ and $k=0.1$, $\Gamma=0.001$, $\Delta k=10^{-4}$, $\alpha=5$, $\beta=2$, and $\gamma=1$.

compound mode ab_1. As expected from (33), we observe that for any value of Γz compound mode ab_1 is higher-order 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_1b_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_1b_2.

There exists another way to study higher-order entanglement. To be precise, all multimode entanglements are essentially higher-order entanglement. As there are three modes in the coupler studied here, we may also investigate the existence of three-mode entanglement. We have already noted that a three-mode pure state that violates (18) (i.e., satisfies $\langle N_a\rangle\langle N_{b_1}\rangle\langle N_{b_2}\rangle - |\langle ab_1b_2\rangle|^2 < 0$) and simultaneously satisfies either (16) or (17) is a fully entangled state. Now using (2)–(3) and (14)–(18) we obtain the following relations for $m = n = l = 1$:

$$E^{1,1,1}_{a|b_1b_2} = -E'^{1,1,1}_{a|b_1b_2} = E^{1,1,1}_{ab_2|b_1} = -E'^{1,1,1}_{ab_2|b_1} = |\gamma|^2 E^{1,1}_{ab_1}, \quad (34)$$

$$E^{1,1,1}_{ab_1|b_2} = E'^{1,1,1}_{ab_1|b_2} = 0, \quad (35)$$

and

$$\langle N_a\rangle\langle N_{b_1}\rangle\langle N_{b_2}\rangle - |\langle ab_1b_2\rangle|^2 = -|\gamma|^2 E^{1,1}_{ab_1}. \quad (36)$$

From (34) we can see that three modes of the coupler are not biseparable in the form $a|b_1b_2$ and $ab_2|b_1$ for any value of $\Gamma z > 0$. Further, Eq. (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, the present solution does not show signature of a fully entangled three-mode state as (35) does not show entanglement between coupled mode ab_1 and mode b_2. To be specific, we observed three-mode (higher-order) entanglement, but could not observe signature of a fully entangled three-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 couplers 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: All 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 the stimulated case.

V. CONCLUSIONS

We have observed various types of higher-order nonclassicalities in fields propagating through a codirectional asymmetric nonlinear optical coupler prepared by combining a linear waveguide and a nonlinear (quadratic) waveguide operated by second harmonic generation. The observations are elaborated in Sec. 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 ([25], 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 other types of couplers, such as contradirectional asymmetric nonlinear couplers, codirectional and contradirectional Raman and Brillouin couplers [21], and parametric couplers [22]. It is possible to experimentally verify the existence of higher-order nonclassicalities reported here as higher-order nonclassical effects can generally be detected using higher numbers of detectors correlating their outcomes. Alternatively, higher-order quantities can be calculated from measured distributions. Specifically, all the criteria of higher-order nonclassicalities reported here are expressed as the expectation values of moments of annihilation and creation operators and these expectation values can be measured using different variants of homodyne measurements and time multiplexing. For example, Shchukin and Vogel clearly showed that amplitude squared squeezing [67] and amplitude nth power squeezing [67] can be detected using a technique based on balanced homodyne correlation measurement [67,68]. Using Shchukin and Vogel's approach one can measure $\langle a^{\dagger k}a^l\rangle$ for any values of k and l (cf., Fig. 1 of Ref. [67]). Thus if we replace the source S in Fig. 1 of Ref. [67] by a field of specific frequency (i.e., a field representing a particular mode) obtained at the output of one of the waveguides that constitute the coupler studied here, it would be possible to measure all the single-mode correlations (including higher-order antibunching and higher-order squeezing) reported in the present work. However, with the increase in k and l, the requirement of number of beam splitters, photodetectors, and measurements increases considerably. This technical limitation of Shchukin and Vogel's approach is considerably circumvented in later works of Prakash and Yadav [69] for nth-order amplitude power squeezing and Prakash, Kumar, and Mishra's work on amplitude squared squeezing [70] where only one beam splitter and one photodetector were used and the number of measurements required was also reduced. In these interesting works [69,70], higher-order moments of number operators were obtained by using standard homodyne technique. Following an independent approach Prakash and Mishra [71] showed that higher-order sub-Poissonian statistics can also be used for detection of amplitude squared squeezing. The proposals of detection of higher-order moments of the form $\langle a^{\dagger k}a^l\rangle$ is relatively new, but schemes for measurement of higher-order moments of number operator (and thus higher-order antibunching) was long in existence [72]. Beyond these new and old schemes of experimental detection of the results reported here what is more exciting is the fact that our control over the field, quality of source, detector, and other devices required for the experimental detection have been considerably improved in recent past, and as a consequence a set of very interesting experimental works demonstrating higher-order nonclassicality have been recently reported [39–41]. Specifically, using a hybrid photodetector Allevi *et al.* [39,40] experimentally measured $\langle a_1^{\dagger j}a_1^j a_2^{\dagger k}a_2^k\rangle$, which can be used to fully characterize bipartite multimode states. Clearly their method can be directly used to detect higher-order entanglement using the HZ-I and HZ-II criteria described by Eqs. (12)–(18), and the higher-order antibunching and higher-order squeezing reported in the present work. Further, Avenhaus *et al.* [41] have also experimentally measured $\langle a_1^{\dagger j} a_1^j a_2^{\dagger k} a_2^k \rangle$ using time multiplexing. Thus, in brief there exist a large number of alternative paths that may be used to experimentally verify the existence of nonclassical states reported in the present work.

It is even possible to investigate the existence of Hong-Mandel [44,45] type higher-order squeezing and Agarwal-Tara parameter A_n [73] 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. [74]. However, we have not investigated steering as recently it is shown that every pure entangled state is maximally steerable [75]. Since the combined states of three modes of the asymmetric codirectional nonlinear optical coupler is a pure state, the findings of Ref. [75] and the intermodal entanglement observed in the present paper imply that the compound mode 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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