Higher-order nonclassical properties of atom-molecule Bose-Einstein Condensate

Sandip Kumar Giri¹,², Kishore Thapliyal³, Biswajit Sen⁴, Anirban Pathak³,*

¹Department of Physics, Panskura Banamali College, Panskura-721152, India
²Department of Physics, Vidyasagar University, Midnapore-721102, India
³Jaypee Institute of Information Technology, A-10, Sector-62, Noida, UP-201307, India
⁴Department of Physics, Vidyasagar Teachers’ Training College, Midnapore-721101, India

The transient quantum statistical properties of the atoms and molecules in an atom-molecule BEC system are investigated by obtaining a third-order perturbative solution of the Heisenberg’s equations of motion corresponding to the Hamiltonian of an atom-molecule BEC system where two atoms can collide to form a molecule. Time dependent quantities like two boson correlation, entanglement, squeezing, antibunching, etc., are computed and their properties are compared. It is established that atom-molecule BEC system is highly nonclassical as lower-order and higher-order squeezing and antibunching in pure (atomic and molecular) modes, squeezing and antibunching in compound mode and lower-order and higher-order entanglement in compound mode can be observed in the atom-molecule BEC system. Exact numerical results are also reported and analytic results obtained using the perturbative technique are shown to coincide with the exact numerical results.

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I. INTRODUCTION

Nonclassical properties of radiation field have been investigated since long [1]. However, the interest on nonclassical states has not been decreased over time. Interestingly, interest on nonclassical states has been considerably increased in recent past as several applications of nonclassical states are recently reported in the context of quantum computation and communication [2–5]. Specifically, nonclassical states are shown to be essential for the implementation of all the recently proposed protocols of device independent quantum cryptography and a bunch of traditional protocols of discrete [2] and continuous variable quantum cryptography [3], quantum teleportation [4], dense-coding [5], etc. Further, the focus of study of nonclassical properties has recently been extended beyond quantum optics (i.e., beyond nonclassical properties of radiation field), and nonclassical properties are recently been investigated in several atomic [6] and references therein] and optomechanical systems [7]. To a large extent, these studies are motivated by the atom-optics analogy, and the fact that recently several possibilities of implementation of quantum computing devices using superconductivity-based systems [8, 9], and Bose Einstein condensate (BEC) based systems [10, 11, 12] have been reported. For example, two weakly coupled BECs confined in a double-well trap is shown to produce Josephson charged qubits [10]; possibility of implementation of quantum algorithms using BECs is reported [11]; Josephson qubits that are suitable for implementation of a scalable integrated quantum circuits are realized [9]; quantum state is transferred using cavities containing two-component BECs coupled by optical fiber [12]; schemes for implementing protocols of quantum metrology using two-component BECs are proposed [13, 14]. Thus, many of the recently reported applications of BECs in quantum information processing involve two-mode BECs [12, 13] and references therein]. These facts have motivated us to systematically investigate the nonclassical properties of a specific class of two-mode BEC systems that are usually referred to as atom-molecule BEC system [15]. Here it would be apt to note that the two-mode BECs can be broadly classified into two classes: (i) atom-atom BEC where total number of bosons present in the system is conserved [10–12, 16–23], and (ii) atom-molecule BEC where total number of bosons present in the system is not conserved as two or more bosons of atomic mode can combine to form a boson in molecular mode and equivalently a boson in molecular mode can decompose into two or more bosons in atomic mode [15, 20, 24, 25].

A state is called nonclassical if its Glauber-Sudarshan $P$ function is not a classical probability density, which implies that for a nonclassical state the $P$ function is either negative or more singular than $\delta$ function. This $P$ function based definition of nonclassicality is both necessary and sufficient, but $P$ function is not directly mea-
surable through experiments. Because of this experimental restriction, several other operational criteria of nonclassicality are proposed, which are sufficient but not necessary. For example, zeroes of $Q$ function, negativity of Wigner function, Fano factor, $Q$ parameter, various inseparability criteria, etc., are introduced as operational criteria for detection of nonclassicality. Here we restrict ourselves to a set of nonclassical criteria that are experimentally realizable and can characterize a set of nonclassical characters of practical importance, such as, antibunching, intermodal antibunching, squeezing, inter-modal squeezing, intermodal entanglement, etc. Further, recently higher-order nonclassical properties are experimentally observed in some bosonic systems \cite{26,28}. In these experimental works, it has been clearly observed that it may be easier to detect a weak nonclassicality when we use a criterion of higher-order nonclassicality (cf. Fig. 4 of \cite{24}). Earlier, the existence of higher-order nonclassicality was theoretically studied in a number of quantum optical systems \cite{24,31}. However, except two very recent works \cite{6,32}, not much effort has been made until now to obtain the signatures of higher-order nonclassicalities in the coupled BEC systems. Keeping these facts in mind, we aim to study the possibilities of observing higher-order squeezing, higher-order antibunching and higher-order entanglement in two-mode atom-molecule BEC system. Present study on nonclassicality in atom-molecule BEC is also motivated by the fact that the nonclassical characters investigated here are already shown to be useful for various important tasks related to quantum communication.

Although the basic concept of BEC was known since 1924 \cite{33,34}, it has been experimentally observed only in mid-nineties \cite{16}. Since then the interest on BEC has been considerably increased, and it has been observed in various systems. For example, BEC is observed in ultra-cold dilute alkali gases using magneto-optical traps (\cite{19} and references therein). These demonstrations and amplified interest on BEC lead to a set of recent studies on nonclassical properties of two-mode BECs \cite{6,15,17,20–23,32} and references therein]. A two-mode BEC system may be visualized as a combined system of two BECs where each mode is a BEC and a particular boson can only occupy one of the two modes. However, bosons from one mode can shift to the other mode. As described above, mainly two types of two-mode BECs exist, namely atom-atom BEC and atom-molecule BEC. Recently, we have systematically studied nonclassical properties of atom-atom BEC \cite{6}, and the present work aims to extend that to two-mode atom-molecule BEC system. There exist several Hamiltonians for the two-mode atom-molecule BEC system, most of them are equivalent. Here we restrict ourselves to a specific Hamiltonian that was introduced by Vardi \cite{15}, and was recently used by Perinova et al. \cite{32} to investigate the existence of nonclassical states. Equations of motion corresponding to this two-mode BEC Hamiltonian can be solved by using different approaches, such as the short-time approximation \cite{33}, Gross-Pitaevski mean-field theory \cite{36}, Perinova et al.’s invariant subspace method \cite{32}, etc. In the present work, a third order analytic operator solution of the two-mode atom-molecule BEC Hamiltonian of our interest is obtained by using a perturbative technique developed by some of the present authors \cite{37–41}. We have also obtained exact numerical solution. The third order perturbative solutions obtained here as the time evolution of the annihilation and creation operators of different bosonic modes are subsequently used to illustrate the existence of various types of lower-order, and higher-order nonclassicality in the two-mode atom-molecule BEC system. Specifically, existence of lower-order, and higher-order squeezing and antibunching in pure (atomic and molecular) modes, squeezing in compound mode and lower-order, and higher-order entanglement in compound mode are shown in the atom-molecule BEC system.

Remaining part of the present paper is organized as follows. In Sec. \textbf{II} we briefly introduce the model Hamiltonian that describes the two-mode atom-molecule BEC system. We also report a third-order perturbative solution of the Heisenberg’s equations of motion corresponding to the field modes present in this Hamiltonian. In Sec. \textbf{III} we illustrate the existence of squeezing of quadrature variables for the individual and coupled modes of the two-mode atom-molecule BEC system. Existence of higher-order squeezing using Hillery’s criterion of amplitude powered squeezing is also shown. Similarly, in Sec. \textbf{IV} it is shown that the antibunching can be observed for all the individual and coupled modes of the two-mode BEC system studied in this paper. Existence of higher-order antibunching is also shown in the individual modes. In Sec. \textbf{V} lower-order, and higher-order quantum entanglement is studied using a set of inseparability criteria and intermodal entanglement is observed. Finally, we conclude the paper in Sec. \textbf{VI}.

\section*{II. MODEL HAMILTONIAN}

The Hamiltonian of the atom-molecule BEC is given by \cite{15}

\begin{equation}
H = \frac{\hbar}{2} a^\dagger a + \frac{\hbar \Omega}{2} \left( a^\dagger a b + a a b^\dagger \right),
\end{equation}

where $a (a^\dagger)$ and $b (b^\dagger)$ are the annihilation (creation) operators for the atomic and molecular modes, respectively. Bosons present in each mode constitute a BEC. Further, in this model two bosons in atomic mode (i.e., two atoms) can combine to form a boson in molecular mode (i.e., a molecule) without the generation of heat, and the atomic mode is coupled to the molecular mode by the near resonant two boson transition or Feshbach resonance where the detuning is $\Delta$, and the coupling constant is $\Omega$. In order to study the lower-order, and higher-order nonclassicalities in this atom-molecule BEC system, we first construct the Heisenberg’s equations of motion for atomic
and molecular mode as

\[ \dot{a}(t) = -i \left( \frac{\Delta}{\sqrt{2}} a(t) + \Omega a(t) b(t) \right), \]
\[ \dot{b}(t) = -\frac{i}{\sqrt{2}} a^2(t). \]  

(2)

This set of coupled nonlinear differential equations are not exactly solvable in closed analytical form. Here we use a perturbative approach developed by Sen and Mandal [37–41]. It is already established (see [6] and references therein) that the perturbative solutions obtained by Sen-Mandal approach are more general than the solutions obtained by conventional short-time approximation [33]. In this approach the solution of Eq. (2) is assumed as follows

\[ a(t) = f_1 a(0) + f_2 a^*(0) b(0) + f_3 a(0) a^2(0) + f_4 a(0) b(0) + f_5 a^*(0) b(0) + f_6 a(0) b^*(0) b^2(0) + f_7 a^3(0) a(0) b(0) + f_8 a^3(0) b(0) b^*(0), \]
\[ b(t) = g_1 b(0) + g_2 a^2(0) + g_3 b(0) + g_4 a(0) a(0) b(0) + g_5 a^2(0) + g_6 a^2(0) b^2(0) + g_7 a^2(0) b^*(0) b(0) + g_8 a^2(0) a^2(0). \]  

(3)

The parameters \( f_i \) and \( g_i \) (where \( i \in \{1, 2, \ldots, 8\} \)) are evaluated with the help of the boundary condition \( f_1(0) = g_1(0) = 1 \) and \( f_i(0) = g_i(0) = 0 \) for \( i \in \{2, 3, \ldots, 8\} \). Under these initial conditions, we obtain solutions for \( f_i(t) \), and \( g_i(t) \) as

\[ f_1 = e^{-\frac{\Delta t}{\sqrt{2}}}, \]
\[ f_2 = -\frac{\Delta t}{\sqrt{2}} \sin \frac{\Delta t}{2}, \]
\[ f_3 = -\frac{\Delta t}{2} = \frac{i\sqrt{2}}{2} \left( \sin \frac{\Delta t}{2} - \frac{\Delta t}{2} f_1 \right), \]
\[ f_5 = -\frac{\Delta t}{2} = \frac{\Delta t}{3} \left( \Delta t \cos \frac{\Delta t}{2} - 2 \sin \frac{\Delta t}{2} \right), \]
\[ f_8 = -\frac{\Delta t^3}{2} f_1 (\Delta t - \sin \Delta t), \]
\[ g_1 = 1, \]
\[ g_2 = \frac{\Delta t}{2}, \]
\[ g_3 = \frac{\Delta t}{2} = f_1 f_3^*, \]
\[ g_5 = -\frac{\Delta t}{8} = \frac{\Delta t}{2} = \frac{\Delta t}{2} \]
\[ g_6 = f_1 f_3^*. \]

The above solution is valid up to the third order in \( \Omega \) with \( \Omega t < 1 \), such that the perturbation theory is respected. In what follows, we have used these solutions to study the possibilities of observing various nonclassical effects in the two-mode atom-molecule BEC system described by (1). Further, to investigate the signatures of nonclassical characters of the two-mode atom-molecule BEC system, we consider that initially the atomic mode and the molecular mode are coherent. Thus, at \( t = 0 \), the composite state of the system can be viewed as a product (separable) state \( |\alpha, \beta\rangle = |\alpha\rangle \otimes |\beta\rangle \), which is the product of two coherent states \( |\alpha\rangle \) and \( |\beta\rangle \) that are eigenkets of \( a \) and \( b \), respectively. Therefore, we can write

\[ a(0) |\alpha, \beta\rangle = \alpha |\alpha, \beta\rangle, \quad b(0) |\alpha, \beta\rangle = \beta |\alpha, \beta\rangle. \]  

(5)

In the following sections, we have investigated the existence of various types of lower-order, and higher-order nonclassicalities in the atom-molecule BEC system described by (1) using the solution reported in [3]-[4] and the initial state \( |\alpha, \beta\rangle \).

### III. QUADRATURE SQUEEZING

In order to investigate the quadrature squeezing effect in the atomic and molecular modes in atom-molecule BEC, we use the standard definition of quadrature operators. For example, in atomic mode quadrature operator is defined as

\[ X_a = \frac{1}{\sqrt{2}} (a(t) + a^*(t)), \]
\[ Y_a = \frac{1}{\sqrt{2}} (a(t) - a^*(t)). \]  

(6)

Similarly, we can construct the quadrature operators \( X_b \) and \( Y_b \) for the molecular mode \( b \). Further, the squeezing in the compound mode \( ab \), can be examined using quadrature operators for the compound atomic-molecular mode as

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

(7)

Quadrature squeezing in \( i \)-th mode (where \( i \in \{a, b\} \)) is witnessed if the fluctuation in one of the quadrature operators goes below the minimum uncertainty level, i.e., if we observe

\[ (\Delta X_i)^2 < \frac{1}{4} \quad \text{or} \quad (\Delta Y_i)^2 < \frac{1}{4}. \]  

(8)

Using (3), (4), (5) and (6) we can obtain analytic expressions for the fluctuation of the quadrature operators in atomic mode as

\[ \left( \frac{(\Delta X_a)^2}{(\Delta Y_a)^2} \right) = \frac{1}{4} \left[ 1 + 2 |f_2|^2 |\beta|^2 + (2 f_1 f_3 a^2 \beta^* + \text{c.c.}) \right] \]
\[ \pm \left\{ f_1 f_3 a^2 + (f_1 f_2 + f_1 f_5) \beta + 6 f_1 f_5 |\alpha|^2 \beta \right. \]
\[ + \left. (f_1 f_6 + f_2 f_4) |\beta|^2 \beta + \text{c.c.} \right\}, \]  

(9)

where c.c. stands for the complex conjugate. Similarly, for the pure mode \( b \) and the coupled mode \( ab \) the analytic expressions of quadrature fluctuations can be obtained as follows

\[ \left( \frac{(\Delta X_b)^2}{(\Delta Y_b)^2} \right) = \frac{1}{4} \left[ 1 \pm \left\{ (g_7 + 2 g_2 g_4) a^2 \beta + \text{c.c.} \right\} \right], \]  

(10)

and
Eqs. (9)-(11) indicate that within the domain of validity of the approximate solution reported here, quadrature squeezing may exist in atomic, molecular and compound modes. To illustrate this, plots of right-hand sides of Eqs. (9), (10), and (11) are shown in Fig. 1a, 1b and 1c, respectively. Regions of these plots with variance less than $\frac{1}{4}$ explicitly illustrate the existence of single mode quadrature squeezing in atomic and molecular modes, and also intermodal squeezing in atom-molecule coupled mode. Interestingly, when we consider $\alpha$ and $\beta$ as real and $\alpha = 5$ and $\beta = 2$, quadrature squeezing is dominantly observed in only one quadrature $X_a (X_{ab})$ in atomic (compound) mode, and it is weakly observed in a small region in the other quadrature, i.e., $Y_a (Y_{ab})$ (see Fig. 1a (c)). However, it is clearly observed in both the quadratures in molecular mode (see Fig. 1b). Interestingly, by increasing the value of $\alpha$, amount of quadrature squeezing in $Y_a$ and $Y_{ab}$, and corresponding region of nonclassicality can be considerably increased. For example, for $\alpha = 10$ and $\beta = 2$ we can observe squeezing in $X_a$, $Y_a$, $X_{ab}$ and $Y_{ab}$ (not shown in the figure). Even for lower values of $\alpha$, large squeezing in $Y_a (Y_{ab})$ can be obtained by using appropriate choice of phase of initial coherent states. This fact is illustrated in the Fig. 1d, where we have considered $\alpha = 5$ and $\beta = -2$ (i.e., $\beta = 2 \exp(i\pi)$). In Fig. 1d, squeezing in atomic mode (thin blue lines) and intermodal squeezing in atom-molecule coupled mode (thick red lines) are plotted together to show squeezing in $Y_a$ and $Y_{ab}$ with change of phase of molecular mode. Further, the boxes and circles shown in the plots depict exact numerical results obtained by integrating the time-dependent Schrödinger equation corresponding to the Hamiltonian (11) by defining the operators as matrices. The excellent coincidence of the exact numerical results with the analytic results obtained using the perturbative solution obtained here strongly establishes the accuracy and validity of our perturbative solution. Interestingly, similar coincidence of perturbative results with the exact numerical results is also observed in other signatures of nonclassicalities that are discussed in the present work. However, we have not shown the numerical results in all cases.

A. Higher-order squeezing

So far, we have examined the existence of lower-order squeezing in the two-mode atom-molecule BEC using (3), (4) and (8). The same may be easily extended to the higher-order squeezing that is usually studied using two alternative approaches (12)-(14). Specifically, higher-order squeezing is studied either using Hillery’s criterion of amplitude powered squeezing (12), that provides witness for the existence of higher-order nonclassicality through the reduction of variance of an amplitude powered quadrature variable for a quantum state with respect to its coherent state counterpart, or using the criterion of Hong and Mandel (13)-(14), which reflects the existence of higher-order nonclassicality through the reduction of higher-order moments of the usual quadrature operators with respect to their coherent state counterparts. In the present paper we have restricted ourselves to Hillery’s criterion. Specifically, Hillery introduced amplitude powered quadrature variables as

$$Y_{1,a} = \frac{a^k + (a^{\dagger})^k}{2}$$ (12)

and

$$Y_{2,a} = i \left( \frac{(a^k)^{\dagger} - a^k}{2} \right).$$ (13)

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

$$A_{i,a} = \left< (\Delta Y_{i,a})^2 \right> - \left< N_a + \frac{1}{2} \right> < 0,$$ (14)

where $i \in \{1, 2\}$. Now using (3), (4), (12) and (13) we obtain
In the present study, we have restricted ourselves to parts of the plots depict signature of higher-order squeezing. In (a) and (b) smooth (dashed) line denotes variance in $X_i$ $(Y_j)$ quadrature where $i \in \{a, b\}$, and in (c) smooth (dashed) line represents variance in $X_{ab}$ $(Y_{ab})$ quadrature using approximate analytic solution. Circle (square) represents the variance using exact numerical solution for respective modes. In (d), quadrature fluctuation of atomic mode $a$ (thin blue lines) and compound atom-molecule mode $ab$ (thick red lines) are shown for $\Omega = 10^2$, $\frac{\Delta}{\Omega} = 10^2$, $\alpha = 5$, and $\beta = 2$. (d) shows that the phase of the input coherent state can be used to control the amount of squeezing.

\[
\left( \begin{array}{c} A_{1,a} \\ A_{2,a} \end{array} \right) = 2 |f_2|^2 |\alpha|^2 |\beta|^2 + \left\{ 2 (f_1 f_5 + f_7 f_8) |\alpha|^2 \alpha^2 \beta^* + 2 f_1 f_2 |f_2|^2 \alpha^2 \beta^* |\beta|^2 - f_4 f_3 \alpha^2 \beta^* + c.c. \right\} \pm \left\{ f_3^2 \alpha^4 \right\}
\]

\[
+ f_1 f_2^2 \alpha^2 \beta^3 + \frac{1}{2} f_1^2 f_2^2 \left( 1 + 4 |\alpha|^2 \right) \beta^2 + f_1^2 (f_1 f_2 + 7 f_1 f_5 + f_2 f_3) \alpha^2 \beta
\]

\[
+ 2 f_1^2 (2 f_2 f_3 + 3 f_1 f_5) |\alpha|^2 \alpha^2 \beta + f_1^2 (3 f_2 f_4 - 2 f_1 f_3) \alpha^2 |\beta|^2 + c.c \}
\]

and

\[
\left( \begin{array}{c} A_{1,b} \\ A_{2,b} \end{array} \right) = \pm \left\{ (g_7 + 2 g_2 g_4) \alpha^2 \beta^3 + c.c. \right\}.
\]

Figure 1: (Color online) Quadrature fluctuation with rescaled interaction time $\Omega t$ of (a) mode $a$ (b) mode $b$, and (c) coupled mode $ab$ for $\Omega = 10^2$, $\frac{\Delta}{\Omega} = 10^2$, $\alpha = 5$, and $\beta = 2$. In (a) and (b) smooth (dashed) line denotes variance in $X_i$ $(Y_j)$ quadrature where $i \in \{a, b\}$, and in (c) smooth (dashed) line represents variance in $X_{ab}$ $(Y_{ab})$ quadrature using approximate analytic solution. Circle (square) represents the variance using exact numerical solution for respective modes. In (d), quadrature fluctuation of atomic mode $a$ (thin blue lines) and compound atom-molecule mode $ab$ (thick red lines) are shown for $\Omega = 10^2$, $\frac{\Delta}{\Omega} = 10^2$, $\alpha = 5$, and $\beta = -2$. (d) shows that the phase of the input coherent state can be used to control the amount of squeezing.

Variation of right hand sides of (15) and (10) are plotted in Fig. 2. Existence of amplitude squared squeezing in both the quadratures of atomic (molecular) mode can be clearly observed in Fig. 2a (b) where negative parts of the plots depict signature of higher-order squeezing. In the present study, we have restricted ourselves to the study of amplitude squared squeezing. However, it is possible to investigate the existence of Hong-Mandel type higher-order squeezing, and amplitude $k$-th power squeezing using the time evolution of the field operators obtained here and the approach adopted above to study lower-order, and higher-order squeezing.
where \( D_{ab} = (\Delta N_{ab})^2 \). As \( \langle N_j(t) \rangle \) is a nonnegative quantity \( g^{(2)}_{ab}(0) < 1 \) implies \( D_{ab} < 0 \) and vise versa. Consequently, the condition of antibunching as well as the sub-Poissonian boson statistics for the couple mode is usually expressed as

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

Now using the solution reported here and considering \(|\alpha|/|\beta|\) as the initial input state, it is easy to obtain closed form analytic expressions of \( D_j \) and \( D_{ab} \) as follows

\[
D_a = |f_a|^2 \left(|\beta|^2 + 6 |\alpha|^2 |\beta|^2 - \frac{1}{2} |\alpha|^4\right) \\
+ \left\{ (f_a^* f_1 + f_a^* f_1 + f_a^* f_3) \alpha^2 \beta^* \right\} \\
+ \left\{ f_a^* f_1 + f_a^* f_4 + 4 |f_a|^2 f_a^* f_1 \right\} |\beta|^2 \alpha^2 \beta^* \\
+ 6 \left( f_a^* f_1 + f_a^* f_3 \right) |\alpha|^2 \alpha^2 \beta^* + c.c.,
\]

\[
D_b = 2 \left( f_b^* g_\Omega |\beta|^2 \alpha^2 \beta^* + c.c.\right)
\]

and

\[
D_{ab} = -|f_a|^2 |\alpha|^2 |\beta|^2 - \left\{ 2 f_a^* g_\Omega |\beta|^2 \alpha^2 \beta^* \\
+ (f_a^* f_1 + f_a^* f_3) |\alpha|^2 \alpha^2 \beta^* + c.c. \right\}.
\]

Using [21]-[24], we can easily investigate the signatures of nonclassical boson statistics in atomic \( a \), molecular \( b \), and compound \( ab \) modes of the atom-molecule BEC of our interest. Specifically, we have clearly observed antibunching in molecular mode \( b \) and compound mode \( ab \), for \( \alpha = 10, \beta = 2 \) for reasonably large regions (See Fig. 3 b-c), but we have observed nonclassical boson statistics in atomic mode \( a \) only in a small region for this specific choice of \( \alpha, \beta \) (See Fig. 3 a). Further, for \( \alpha \leq 6 \) we have not observed nonclassical boson statistics in atomic mode. However, antibunching (nonclassical boson statistics) for all values of rescaled interaction time \( \Omega > 0 \) in atomic mode \( a \) for \( \alpha \leq 6 \) can be observed by modifying the phase of the input coherent state of the molecular mode \( b \) (i.e., using \( \beta = -2 \) instead of \( \beta = 2 \)). Using \( \beta = -2 \), we can also observe antibunching in the molecular mode \( b \) for those values of rescaled interaction time for which we could not observe it for our previous choice of \( \beta \) (i.e., for \( \beta = 2 \)). When the phase of the molecular mode \( b \) is modified (i.e., if we use \( \beta = -2 \) instead of \( \beta = 2 \)) then the phase of all the terms except the first term in Eq. (21) is changed as the first term \( \left( |f_a|^2 \left(|\beta|^2 + 6 |\alpha|^2 |\beta|^2 - \frac{1}{2} |\alpha|^4\right) \right) \) contains only \(|\beta|\), it is independent of phase of \( \beta \). To be precise, for \( \frac{|\alpha|^4}{(1+6|\alpha|^2)} > 2 |\beta|^2 \), this term gives finite negative contribution to the antibunching in atomic mode \( a \) (as for \( \alpha = 10, \beta = 2 \) we obtain \( \frac{|\alpha|^4}{(1+6|\alpha|^2)} = 16.6 > 2 |\beta|^2 = 8 \)). Now if we obtain \( D_a > 0 \) for some values of \( \alpha \) and \( \beta \) (say
\(\alpha = 10, \beta = 2\), then that would mean that the sum of all the terms other than the first term is positive and its absolute value is greater than that of the first term. Interestingly, this sum can be made negative by changing the phase of \(\beta\) (say by choosing \(\beta = -2\)) as it is directly proportional to \(\beta\) and clearly that choice will lead to \(D_a < 0\) or antibunching in mode \(a\). Further, even when \(D_a < 0\) for some values of \(\alpha\) and \(\beta\) that satisfy
\[
\frac{\alpha^4}{(1 + 6|\alpha|^2)} > 2|\beta|^2,
\]
and the sum of all the terms other than the first term is positive, but its amplitude is smaller than that of the first term, then by changing phase of \(\beta\) we can increase the depth of nonclassicality. In brief, by controlling the phase of the input coherent state of the molecular mode, we can control the nonclassical properties of the atomic mode \(a\). Plots of the analytic results described by Eqs. (21)-(23) are shown in Fig. 3 a-c. Fig. 3 d illustrates that the antibunching in atomic mode \(a\) can be observed by modifying the phase of the input coherent state of the molecular mode.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3.png}
\caption{(Color online) Plot of \(D_i\) with rescaled interaction time \(\Omega t\) for (a) mode \(a\), (b) mode \(b\), and (c) coupled mode \(ab\) for \(\Omega = 10^2, \Delta \Omega = 10^2, \alpha = 10,\) and \(\beta = 2,\) and (d) mode \(a\) for \(\Omega = 10^2, \Delta \Omega = 10^2, \alpha = 10,\) and \(\beta = -2.\) Negative regions of the plots in (a)-(d) show antibunching.}
\end{figure}

### A. Higher-order antibunching

In order to investigate the higher-order antibunching of the pure modes, Lee introduced the criterion \(^{15}\)
\[
R(l, m) = \frac{\langle N^{(l+1)} \rangle \langle N^{(m-1)} \rangle}{\langle N^{(l)} \rangle \langle N^{(m)} \rangle} - 1 < 0,
\]
where \(N\) is the usual number operator and \(\langle N^{(i)} \rangle = \langle N (N - 1) .... (N - i + 1) \rangle\) is the \(i\)th factorial moment of the number operator. \(l\) and \(m\) are the integers satisfying the condition \(1 \leq m \leq l\). The subscript \(x\) denotes the particular mode. Pathak and Garcia simplified this criterion for \((n - 1)^{th}\) order antibunching \(^{20}\) as
\[
\langle N_x^{(n)} \rangle - \langle N_x \rangle^{(n)} < 0,
\]
where \(\langle N_x^{(n)} \rangle = \langle a^{\dagger n} a^n \rangle\) is the measure of probability of observing \(n\) bosons of the same mode at a particular point in space time coordinate. Now, for the atomic mode \(a\) we can obtain \((n - 1)^{th}\) order antibunching as
\[
\langle a^{n_1}a^{n_2} \rangle - \langle a^1a \rangle^n = |f_2|^2 \left\{ (nC_2)^2 |\alpha|^2(n-2) |\beta|^2 + (n^3 - n) |\alpha|^{2(n-1)} |\beta|^2 - \frac{1}{2} nC_2 |\alpha|^{2n} \right\} \\
+ \left\{ (n^3 - 3n^2 + 2n) f_1^2 f_2 f_3 + \frac{n}{3} (n^2 - n) f_1 f_2 f_3 + (n^3 - n) f_3^2 f_2 \right\} |\alpha|^{2(n-1)} \alpha^2 \beta \\
+ \left\{ nC_2 f_1^2 + \frac{n}{4} (n^4 - 6n^3 + 11n^2 - 6n) f_1^2 f_2 f_3 + (nC_2 - 6nC_3) f_1 f_2 f_3 + (nC_2)^2 f_3^2 f_2 \right\} |\alpha|^{2(n-2)} \alpha^2 \beta \\
+ \left\{ 3nC_3 f_1^2 f_2 f_3 |\alpha|^{2(n-3)} \alpha^2 \beta + 3nC_4 f_1^2 f_2 f_3 |\alpha|^{2(n-4)} \alpha^2 \beta + 6nC_4 f_1^2 f_2 f_3 |\alpha|^{2(n-4)} \alpha^2 \beta \\
+ \frac{1}{4} nC_5 n (3n-1) f_2^2 f_1^2 f_2 f_3 |\alpha|^{2(n-3)} \alpha^2 \beta + 3nC_2 n C_4 f_2^2 f_1^2 f_2 f_3 |\alpha|^{2(n-4)} \alpha^2 \beta + c.c. \right\}.
\]

It is easy to check that for \( n = 2 \), Eq. (26) reduces to Eq. (21). Similarly, for molecular mode, we obtain

\[
\langle b^{n_1}b^{n_2} \rangle - \langle b^1b \rangle^n = \left[ n (n - 1) f_1^2 g_6 |\beta|^{2(n-1)} \alpha^2 \beta^* + c.c. \right].
\]

(V. Entanglement)

In order to study the two-mode entanglement, we use the Hillery-Zubairy (HZ) criteria (1 and 2) and Duan et al.'s criterion \[50\]. Lower-order HZ-1 and HZ-2 criteria are expressed as

\[
\langle N_a(t)N_b(t) \rangle - |\langle a(t)b^1(t) \rangle|^2 < 0
\]

and

\[
\langle N_a(t) \rangle \langle N_b(t) \rangle - |\langle a(t)b(t) \rangle|^2 < 0,
\]

respectively. Using (4), (14), (15) and (28), we obtain

\[
\langle N_a(t)N_b(t) \rangle - |\langle a(t)b^1(t) \rangle|^2 = |f_2|^2 \left\{ |\beta|^4 - |\alpha|^2 |\beta|^2 \right\} + \left\{ (3f_2^2 f_3^2 f_1^2 + 4f_2^2 f_3 - g_7) |\beta|^2 \alpha^2 \beta^* \\
- (f_3^5 f_1^2 + f_2^2 f_3) |\alpha|^2 \alpha^2 \beta^* + c.c. \right\},
\]

Similarly, using (4), (14), (15) and (28), we obtain

\[
\langle N_a(t) \rangle \langle N_b(t) \rangle - |\langle a(t)b(t) \rangle|^2 = |f_2|^2 \left\{ |\beta|^4 + |\alpha|^2 |\beta|^2 \right\} + \left\{ (2f_2^2 f_1 - f_2^4 f_3 f_1^2) |\beta|^2 \alpha^2 \beta^* \\
- (g_6 + g_4^2 g_2) |\alpha|^2 \alpha^2 \beta^* + c.c. \right\}.
\]

As both HZ-1 and HZ-2 criteria are only sufficient and not essential, we also investigate the existence of entanglement using Duan et al.'s criterion \[50\] which can be written as

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

where

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

\[
u_{ab} = -\frac{i}{\sqrt{2}} \left\{ (a - a^1) + (b - b^1) \right\}.
\]
Here we would like to note that all the inseparability criteria described above and in the rest of the paper can be obtained as special cases of Shchukin-Vogel entanglement criteria \[5, 1]. Using Eqs. \[3, 4, 5\] and criterion \[36\], we obtain

\[
d_{ab} = 2 \left\{ |f_{2}^{*}|^{2} |\beta|^{2} + (f_{3}^{*} f_{3} |\alpha|^{2} \beta^{*} + 2 g_{2}^{*} |\beta|^{2} \alpha^{*} + \text{c.c.}) \right\}.
\]

Temporal evolution of the parameters that indicate the existence of entanglement are shown in the Fig. 5a using HZ-1 and HZ-2 criterion and in the Fig. 5b using Duan et al.’s criteria, respectively. Fig. 5a shows that the atomic and molecular modes are entangled for any value of \(\Omega > 0\) for the specific values of parameters chosen here.

A. Higher-order entanglement

In order to investigate the higher-order entanglement for the coupled mode \(ab\), we use the two criteria of Hillery-Zubairy \[15\]. These are

\[
\langle a^{n} a^{*} b^{m} b^{*} \rangle - |\langle a^{n} b^{m} \rangle|^{2} < 0
\]

and

\[
\langle a^{l} a^{*} b^{m} b^{*} \rangle - |\langle a^{n} b^{m} \rangle|^{2} < 0,
\]

where \(m\) and \(n\) are positive integers and for higher-order entanglement \(m + n \geq 3\). It is easy to observe that for \(m = 1\) and \(n = 1\), criteria \[35\] and \[36\] reduces to \[28\] and \[29\], respectively. This is why criteria \[35\] and \[36\] are usually referred to as higher-order HZ-1 and HZ-2 criteria, respectively. Now using Eqs. \[4, 6\] and criterion \[35\] for a specific case \(n = 1, m = 2\), we obtain

\[
\langle a^{1} a^{*} b^{2} b^{*} \rangle - |\langle a^{2} b^{*} \rangle|^{2} = |f_{2}^{*}|^{2} \left( |\beta|^{6} - 2 |\alpha|^{4} |\beta|^{2} + \left\{ f_{2}^{*} f_{3} + 6 |f_{2}|^{2} g_{2} - 2 g_{2}^{*} \right\} |\beta|^{4} |\alpha|^{2} \beta^{*} + \left\{ \frac{1}{2} |f_{2}|^{2} g_{2} + g_{2}^{*} \right\} |\alpha|^{2} |\beta|^{2} |\alpha|^{2} \beta^{*} + \text{c.c.} \right).
\]

Right hand side of \[37\] is plotted in Fig. 6a and we can clearly see the existence of higher-order entanglement through the negative regions of the plot. In this case we have investigated the existence of higher-order entanglement using a particular value of \(m\) and \(n\), but it is possible to obtain a general expression for arbitrary values of \(m\) and \(n\) using the present framework. Just to illustrate this point, we use Eqs. \[3, 4, 5\] and criterion \[36\] in general to obtain

\[
\langle a^{1} a^{*} b^{m} b^{*} \rangle - |\langle a^{n} b^{m} \rangle|^{2} = |f_{2}^{*}|^{2} \left( mn |\alpha|^{2n} |\beta|^{2m} + n^{2} |\alpha|^{2(n-1)} |\beta|^{2(m+1)} \right) + \left\{ \frac{1}{2} mn^{2} f_{2}^{*} f_{3} f_{3} + mnf_{3}^{*} f_{5} + mn^{2} f_{3}^{*} f_{4} f_{2} \right\} |\alpha|^{2n} |\beta|^{2(m-1)} + \alpha^{*2} \beta^{*} - \left\{ f_{2}^{*} f_{3} + 6 |f_{2}|^{2} g_{2} - 2 g_{2}^{*} \right\} |\beta|^{4} |\alpha|^{2} \beta^{*} + \left\{ \frac{1}{2} |f_{2}|^{2} g_{2} + g_{2}^{*} \right\} |\alpha|^{2} |\beta|^{2} |\alpha|^{2} \beta^{*} + \text{c.c.} \right).
\]

Figure 4: (Color online) Variation of \((n - 1)^{th}\) order antibunching with rescaled time \(\Omega t\) for different values of \(n\) for \(\Omega = 10^{2}, \frac{1}{2} = 10^{2}, \alpha = 10, \text{ and } \beta = 2\) for (a) atomic mode \(a\), and (b) molecular mode \(b\) with \(n = 3\) (smooth blue line) and \(n = 4\) (dashed red line). To display the plots in the same scale, the Y-axis of the plots for \(n = 3\) is amplified by 100 in (a), and by 5 in (b). Negative regions of the plots show higher-order antibunching in respective modes.
VI. CONCLUSIONS

Lower-order and higher-order nonclassical properties of a two-mode atom-molecule BEC system is investigated here with the help of a third order perturbative solution of the Heisenberg’s equations of motion corresponding to the Hamiltonian of the BEC system. The investigation established that even if we start with classical (i.e., coherent and separable) input state, the interaction introduces nonclassicality. Thus, the interaction between the atomic and molecular modes leads to a superposition in tensor product space and phase space. Specifically, we considered a separable initial state as we assumed it to be a product of two coherent states \(|\alpha\rangle\langle\beta|\). Now, using lower-order, and higher-order inseparability criteria of Hillery and Zubairy, and Duan et al.’s criterion, we have shown the existence of lower-order, and higher-order intermodal entanglement. As an entangled state can always be viewed as a superposition of separable states in the tensor product space (for example, Bell state is an equal superposition of \(|0\rangle \otimes |0\rangle \) and \(|1\rangle \otimes |1\rangle \)), we may conclude that the interaction between the atomic mode...
and molecular mode of the two-mode BEC system leads to a superposition in tensor product space and the existence of this superposition in tensor product space is reflected here when we observed entanglement through the inseparability criteria mentioned above. In a similar fashion, a traditional nonclassical state such as squeezed state or a Fock state may be viewed as superposition of coherent states. Thus, the lower-order, and higher-order squeezing and antibunching observed here is essentially a manifestation of superposition in phase (Hilbert) space due to interaction between atomic and molecular modes. Interestingly, amount of superposition (i.e., interference in phase space and/or tensor product space) can be controlled by varying various parameters, such as, interaction time, coupling constant $\Omega$, and detuning $\Delta$, boson number of the input modes and the phase of the input coherent states. The effects of these parameters are illustrated through Figs. 1-6.

The methodology adopted here and in our earlier work on atom-atom two-mode BEC is quite general, and is applicable to other bosonic systems, too. Further, in Table I and II of Ref. \cite{PRA85}, a large number of nonclassical criteria based on the expectation values of the moments of the annihilation and creation operators of the field modes is listed. As we already have compact expressions for the field operators, it is possible to extend the present work to investigate other signatures of nonclassicality using the criteria of nonclassicality listed in \cite{PRA85}. For example, we can easily extend the present work to study hyperbunching \cite{PRA85}, sum and difference squeezing of An-Tinh \cite{PRA85} and Hillery \cite{PRA85}, the existence of entanglement using the inseparability criterion of Manicini et al. \cite{PRA85}, Simon \cite{PRA85} and Miranowicz et al. \cite{PRA85}, etc. In addition, recent experimental successes in realizing two-mode BEC systems and observing higher-order nonclassicality indicate that the observations of the present theoretical work can be verified experimentally. We conclude the paper with a hope that the results presented in this work will be useful in the future development of quantum information processing in particular and nonclassical states in general.

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