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Higher order antibunching is not a rare phenomenon

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Abstract
Since the introduction of higher order nonclassical effects, higher order squeezing has been reported in a number of different physical systems but higher order antibunching is predicted only in three particular cases. In the present work, we have shown that the higher order antibunching is not a rare phenomenon; rather it can be seen in many simple optical processes. To establish our claim, we have shown it in the six wave mixing process, four wave mixing process and second harmonic generation process.

1. Introduction

Most of the interesting recent developments in quantum optics have arisen through the nonclassical properties of the radiation field. For example, antibunching and squeezing do not have any classical analogue [1–3]. These two nonclassical states have been extensively studied in last 30 years. But the majority of these studies are focused on lowest order nonclassical effects. Higher order extensions of these nonclassical states have only been introduced in the recent past [4–7]. Among these higher order nonclassical effects, higher order squeezing has already been studied in detail [4, 5, 8, 9] but the higher order antibunching (HOA) is not yet studied rigorously. The idea of HOA was introduced by Lee in a pioneering paper [6] in 1990; since then it has been predicted in the two-photon coherent state [6], trio coherent state [10] and in the interaction of an intense laser beam with an inversion symmetric third-order nonlinear medium [11]. From the fact that in the last 15 years HOA has been reported only in three particular cases, HOA appears to be a very rare phenomenon. The present study aims to establish that this apparent rarity is not due to any physical reason. To establish that, we have shown the existence of HOA in the six wave mixing process, four wave mixing process and second harmonic generation process.

Using the negativity of P function [1], Lee introduced the criterion for HOA as

$$R(l, m) = \frac{\langle N_x^{(l+1)}\rangle\langle N_x^{(m-1)}\rangle}{\langle N_x^{(l)}\rangle\langle N_x^{(m)}\rangle} - 1 < 0, \tag{1}$$

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where N is the usual number operator, $\langle N^{(i)}\rangle = \langle N(N-1)\dots(N-i+1)\rangle$ is the ith factorial moment of the number operator, $\langle\,\rangle$ denotes the quantum average, l and m are the integers satisfying the conditions $1 \leqslant m \leqslant l$ and the subscript x denotes a particular mode. An [10] choose $m = 1$ and reduced the criterion of lth-order antibunching to

$$A_{x,l} = \frac{\langle N_x^{(l+1)}\rangle}{\langle N_x^{(l)}\rangle\langle N_x\rangle} - 1 < 0 \tag{2}$$

or

$$\langle N_x^{(l+1)}\rangle < \langle N_x^{(l)}\rangle\langle N_x\rangle. \tag{3}$$

Physically, a state which is antibunched in the lth order has to be antibunched in the $(l-1)$th order. Therefore, we can further simplify (3) as

$$\langle N_x^{(l+1)}\rangle < \langle N_x^{(l)}\rangle\,\langle N_x\rangle < \langle N_x^{(l-1)}\rangle\,\langle N_x\rangle^2 < \langle N_x^{(l-2)}\rangle\,\langle N_x\rangle^3 < \cdots < \langle N_x\rangle^{l+1} \tag{4}$$

and obtain the condition for lth-order antibunching as

$$d(l) = \langle N_x^{(l+1)}\rangle - \langle N_x\rangle^{l+1} < 0. \tag{5}$$

This simplified criterion (5) coincides exactly with the physical criterion of HOA introduced by Pathak and Garica [11]. Here we can note that $d(l) = 0$ and $d(l) > 0$ correspond to higher order coherence and higher order bunching (many photon bunching), respectively. Actually, $\langle a^{\dagger l}a^l\rangle = \langle N^{(l)}\rangle$ is a measure of the probability of observing l photons of the same mode at a particular point in a spacetime coordinate. Therefore, the physical meaning of inequalities (4) is that the probability of detection of a single-photon pulse is greater than that of a two-photon pulse in a bunch and that is greater than the probability of detection of a three-photon pulse in a bunch and so on. This is exactly the characteristic that is required in a probabilistic single-photon source used in quantum cryptography. In other words, all the probabilistic single-photon sources used in quantum cryptography should satisfy the criteria (5) of HOA.

The present work aims to show that HOA is not really a rare phenomenon; rather it can be seen in many simple nonlinear optical processes. To establish that we have used the criterion (5) and short-time approximated solutions of equations of motion corresponding to various Hamiltonians (such as six wave mixing process, four wave mixing process and second harmonic generation) and have shown that HOA can be seen in all the physical systems selected for the present study. The theoretical predictions of the present study can be experimentally verified easily because all the models studied here are experimentally realizable [12, 13] and the criteria for HOA appear in terms of factorial moment, which can be measured by using homodyne photon counting experiments [12–15]. In the next section, we present a second-order operator solution of the equation of motion of the six wave mixing process in detail and use that to show the existence of HOA in the six wave mixing process. In sections 3 and 4 we study the possibilities of observing higher order antibunching in the four wave mixing process and second harmonic generation process, respectively. Finally, section 5 is dedicated to conclusions.

2. Six wave mixing process

Six wave mixing is a well-known nonlinear optical process which is experimentally observed in a number of nonlinear media. This process is used for many practical applications, such as image transfer [16] and photon echo detection [17]. The present work opens up the possibility of using it as a higher order sub-Poissonian photon source. This process may occur in different ways. One way is that two photons of frequency ω_1 are absorbed (as pump photon) and three photons of frequency ω_2 and another of frequency ω_3 are emitted. The Hamiltonian representing this particular six wave mixing process is

$$H = a^\dagger a\omega_1 + b^\dagger b\omega_2 + c^\dagger c\omega_3 + g(a^{\dagger 2}b^3c + a^2b^{\dagger 3}c^\dagger), \tag{6}$$

where a and $a^\dagger$ are annihilation and creation operators in the pump mode which satisfy $[a, a^\dagger] = 1$; similarly $b, b^\dagger$ and $c, c^\dagger$ are annihilation and creation operators in stokes mode and signal mode, respectively, and g is the coupling constant. Substituting $A = a\,e^{i\omega_1 t}$, $B = b\,e^{i\omega_2 t}$ and $C = c\,e^{i\omega_3 t}$ we can write the Hamiltonian (6) as

$$H = A^\dagger A\omega_1 + B^\dagger B\omega_2 + C^\dagger C\omega_3 + g(A^{\dagger 2}B^3C + A^2B^{\dagger 3}C^\dagger). \tag{7}$$

Since the Hamiltonian is known, we can use Heisenberg's equation of motion (with $\hbar = 1$)

$$\dot{A} = \frac{\partial A}{\partial t} + i[H, A] \tag{8}$$

and short-time approximation to find out the time evolution of the essential operators. From equation (7) we have

$$[H, A] = -A\omega_1 - 2gA^\dagger B^3C. \tag{9}$$

From (8) and (9) we have

$$\dot{A} = iA\omega_1 - iA\omega_1 - i2gA^\dagger B^3C = -2igA^\dagger B^3C. \tag{10}$$

Similarly

$$\dot{B} = -3igA^2B^{\dagger 2}C^\dagger \tag{11}$$

and

$$\dot{C} = -igA^2B^{\dagger 3}. \tag{12}$$

We can find the second-order differential of A using (8) and (10)–(12) as

$$\begin{aligned}\ddot{A} = \frac{\partial \dot{A}}{\partial t} + i[H, \dot{A}] &= 4g^2AB^{\dagger 3}B^3C^\dagger C - 18g^2A^\dagger A^2B^{\dagger 2}B^2C^\dagger C - 36g^2A^\dagger A^2B^\dagger BC^\dagger C \\ &\quad - 2g^2A^\dagger A^2B^{\dagger 3}B^3 - 18g^2A^\dagger A^2B^{\dagger 2}B^2 - 36g^2A^\dagger A^2B^\dagger B \\ &\quad - 12g^2A^\dagger A^2C^\dagger C - 12g^2A^\dagger A^2.\end{aligned} \tag{13}$$

Now by using the Taylor's series expansion

$$f(t) = f(0) + t\left(\frac{\partial f(t)}{\partial t}\right)_{t=0} + \frac{t^2}{2!}\left(\frac{\partial^2 f(t)}{\partial t^2}\right)_{t=0} + \cdots \tag{14}$$

and substituting (10) and (13) in (14), we get

$$\begin{aligned}A(t) = A - 2igtA^\dagger B^3C + g^2t^2[&2AB^{\dagger 3}B^3C^\dagger C - 9A^\dagger A^2B^{\dagger 2}B^2C^\dagger C - 18A^\dagger A^2B^\dagger BC^\dagger C \\ &- A^\dagger A^2B^{\dagger 3}B^3 - 9A^\dagger A^2B^{\dagger 2}B^2 - 18A^\dagger A^2B^\dagger B - 6A^\dagger A^2C^\dagger C - 6A^\dagger A^2]\end{aligned} \tag{15}$$

or

$$\begin{aligned}A(t) = A - 2igtA^\dagger B^3C + g^2t^2[&2AB^{\dagger 3}B^3N_C - 9N_AAB^{\dagger 2}B^2N_C - 18N_AAN_BN_C \\ &- N_AAB^{\dagger 3}B^3 - 9N_AAB^{\dagger 2}B^2 - 18N_AAN_B - 6N_AAN_C - 6N_AA],\end{aligned} \tag{16}$$

where $N_A = A^\dagger A,\ N_B = B^\dagger B,\ N_C = C^\dagger C$.[1] The Taylor series is valid when t is small, so this solution is valid for a short time and that is why it is called short-time approximation. The above calculation is shown in detail as an example. Following the same prescription, we can

[1] A short-time approximated expression of the time evolution of the annihilation operator in the pump mode of the six wave mixing process described by (6) is also derived in [9] but unfortunately their solution contains some mistakes.

find out the time evolution of B and C or any other creation and annihilation operators that appear in the Hamiltonian of matter field interaction. This is a very strong technique since this straightforward prescription is valid for any optical process where interaction time is short. After obtaining the analytic expression for the time evolution of the annihilation operator, we now can use it to check whether it satisfies condition (5) or not.

Let us start with the possibility of observing first-order antibunching. From equation (15), we can derive expressions for $N(t)$ and $N^{(2)}(t)$ as

$$\begin{aligned} N(t) = A^\dagger A &- 2igt(A^{\dagger 2}B^3C - A^2B^{\dagger 3}C^\dagger) + g^2t^2[8A^\dagger AB^{\dagger 3}B^3C^\dagger C - 18A^{\dagger 2}A^2B^{\dagger 2}B^2C^\dagger C \\ &- 36A^{\dagger 2}A^2B^\dagger BC^\dagger C - 2A^{\dagger 2}A^2B^{\dagger 3}B^3 - 18A^{\dagger 2}A^2B^{\dagger 2}B^2 \\ &- 36A^{\dagger 2}A^2B^\dagger B - 12A^{\dagger 2}A^2C^\dagger C + 4B^{\dagger 3}B^3C^\dagger C - 12A^{\dagger 2}A^2] \end{aligned} \tag{17}$$

and

$$\begin{aligned} N^{(2)}(t) = A^{\dagger 2}(t)A^2(t) = A^{\dagger 2}A^2 &- 2igt(2A^{\dagger 3}AB^3C - A^{\dagger 2}B^3C - 2A^\dagger A^3B^{\dagger 3}C^\dagger - A^2B^{\dagger 3}C^\dagger) \\ &+ g^2t^2[24A^{\dagger 2}A^2B^{\dagger 3}B^3C^\dagger C + 32A^\dagger AB^{\dagger 3}B^3C^\dagger C + 4B^{\dagger 3}B^3C^\dagger C \\ &- 36A^{\dagger 3}A^3B^{\dagger 2}B^2C^\dagger C - 72A^{\dagger 3}A^3B^\dagger BC^\dagger C - 18A^{\dagger 2}A^2B^{\dagger 2}B^2C^\dagger C \\ &- 36A^{\dagger 2}A^2B^\dagger BC^\dagger C - 4A^{\dagger 3}A^3B^{\dagger 3}B^3 - 36A^{\dagger 3}A^3B^{\dagger 2}B^2 - 72A^{\dagger 3}A^3B^\dagger B \\ &- 2A^{\dagger 2}A^2B^{\dagger 3}B^3 - 18A^{\dagger 2}A^2B^{\dagger 2}B^2 - 4A^{\dagger 4}B^6C^2 - 4A^4B^{\dagger 6}C^{\dagger 2} - 36A^{\dagger 2}A^2B^\dagger B \\ &- 24A^{\dagger 3}A^3C^\dagger C - 12A^{\dagger 2}A^2C^\dagger C - 24A^{\dagger 3}A^3 - 12A^{\dagger 2}A^2]. \end{aligned} \tag{18}$$

In the present study, we have taken all the expectations with respect to $|\alpha\rangle|0\rangle|0\rangle$ for simplification. This assumption physically means that initially a coherent state (say, a laser) is used as a pump and before the interaction of the pump with the atom, there was no photon in the signal mode (b) or stokes mode (c). Thus, the pump interacts with the atom and causes excitation followed by emissions. Now from (17) and (18), we have

$$\langle N(t)\rangle^2 = |\alpha|^4 - 24g^2t^2|\alpha|^6. \tag{19}$$

$$\langle N^2(t)\rangle = |\alpha|^4 - g^2t^2(24|\alpha|^6 + 12|\alpha|^4), \tag{20}$$

where $A|\alpha\rangle = \alpha|\alpha\rangle$. Now by using (19) and (20) we can show that the six wave mixing process satisfies the criterion of antibunching (5) since

$$\begin{aligned} d(1) &= \langle N^{(2)}(t)\rangle - \langle N(t)\rangle^2 \\ &= [|\alpha|^4 + g^2t^2(-24|\alpha|^6 - 12|\alpha|^4)] - [|\alpha|^4 - 24g^2t^2|\alpha|^6] \\ &= -12g^2t^2|\alpha|^4. \end{aligned} \tag{21}$$

From the last equation it is clear that $d(1)$ is always negative, i.e. it always shows usual antibunching. Essentially, this is a nonclassical state but mere satisfaction of nonclassicality or antibunching is not enough because we are looking for HOA. Let us see what happens in the next higher order.

For the study of second order of antibunching, $A^3(t)$ can be obtained by using (15) and operator ordering techniques

$$\begin{aligned} A^3(t) = A^3 &- 2igt(3A^\dagger A^2B^3C + 3AB^3C) + g^2t^2[6A^3B^{\dagger 3}B^3C^\dagger C - 27A^\dagger A^4B^{\dagger 2}B^2C^\dagger C \\ &- 54A^\dagger A^4B^\dagger BC^\dagger C - 54A^3B^\dagger BC^\dagger C - 27A^3B^{\dagger 2}B^2C^\dagger C - 3A^\dagger A^4B^{\dagger 3}B^3 \\ &- 27A^\dagger A^4B^{\dagger 2}B^2 - 54A^\dagger A^4B^\dagger B - 3A^3B^{\dagger 3}B^3 - 27A^3B^{\dagger 2}B^2 \\ &- 18A^\dagger A^4C^\dagger C - 18A^3C^\dagger C - 54A^3B^\dagger B - 12A^{\dagger 2}AB^6C^2 \\ &- 12A^\dagger B^6C^2 - 18A^\dagger A^4 - 18A^3]. \end{aligned} \tag{22}$$

Then $A^{\dagger 3}(t)$ can simply be written as

$$\begin{aligned}A^{\dagger 3}(t) = A^{\dagger 3} &+ 2igt(3A^{\dagger 2}AB^{\dagger 3}C^{\dagger} + 3A^{\dagger}B^{\dagger 3}C^{\dagger}) + g^2t^2[6A^{\dagger 3}B^{\dagger 3}B^3C^{\dagger}C \\ &- 27A^{\dagger 4}AB^{\dagger 2}B^2C^{\dagger}C - 54A^{\dagger 4}AB^{\dagger}BC^{\dagger}C - 54A^{\dagger 3}B^{\dagger}BC^{\dagger}C \\ &- 27A^{\dagger 3}B^{\dagger 2}B^2C^{\dagger}C - 3A^{\dagger 4}AB^{\dagger 3}B^3 - 27A^{\dagger 4}AB^{\dagger 2}B^2 - 54A^{\dagger 4}AB^{\dagger}B \\ &- 3A^{\dagger 3}B^{\dagger 3}B^3 - 27A^{\dagger 3}B^{\dagger 2}B^2 - 18A^{\dagger 4}AC^{\dagger}C - 18A^{\dagger 3}C^{\dagger}C \\ &- 54A^{\dagger 3}B^{\dagger}B - 12A^{\dagger}A^2B^{\dagger 6}C^{\dagger 2} - 12AB^{\dagger 6}C^{\dagger 2} - 18A^{\dagger 4}A - 18A^{\dagger 3}].\end{aligned} \tag{23}$$

The last two equations can be used to calculate the third factorial moment $(N^{(3)}(t))$ of the number operator N as

$$\begin{aligned}N^{(3)}(t) = A^{\dagger 3}A^3 &- 2igt(3A^{\dagger 4}A^2B^3C + 3A^{\dagger 3}AB^3C - 3A^{\dagger 2}A^4B^{\dagger 3}C^{\dagger} - 3A^{\dagger}A^3B^{\dagger 3}C^{\dagger}) \\ &+ g^2t^2[48A^{\dagger 3}A^3B^{\dagger 3}B^3C^{\dagger}C - 54A^{\dagger 4}A^4B^{\dagger 2}B^2C^{\dagger}C - 54A^{\dagger 3}A^3B^{\dagger 2}B^2C^{\dagger}C \\ &- 108A^{\dagger 3}A^3B^{\dagger}BC^{\dagger}C + 108A^{\dagger 2}A^2B^{\dagger 3}B^3C^{\dagger}C + 36A^{\dagger}AB^{\dagger 3}B^3C^{\dagger}C \\ &- 108A^{\dagger 4}A^4B^{\dagger}BC^{\dagger}C - 54A^{\dagger 4}A^4B^{\dagger 2}B^2 - 108A^{\dagger 4}A^4B^{\dagger}B - 6A^{\dagger 3}A^3B^{\dagger 3}B^3 \\ &- 6A^{\dagger 4}A^4B^{\dagger 3}B^3 - 54A^{\dagger 3}A^3B^{\dagger 2}B^2 - 108A^{\dagger 3}A^3B^{\dagger}B - 12A^{\dagger 5}AB^6C^2 \\ &- 12A^{\dagger}A^5B^{\dagger 6}C^{\dagger 2} - 12A^{\dagger 4}B^6C^2 - 12A^4B^{\dagger 6}C^{\dagger 2} - 36A^{\dagger 3}A^3C^{\dagger}C \\ &- 36A^{\dagger 4}A^4C^{\dagger}C - 36A^{\dagger 4}A^4 - 36A^{\dagger 3}A^3].\end{aligned} \tag{24}$$

Taking the expectation value with respect to the initial state we obtain

$$\langle N^{(3)}(t)\rangle = |\alpha|^6 - g^2t^2(36|\alpha|^8 + 36|\alpha|^6). \tag{25}$$

On the other hand,

$$\langle N(t)\rangle^3 = |\alpha|^6 - 36g^2t^2|\alpha|^8. \tag{26}$$

By using the last two equations we can see that the pump mode of the six wave mixing process satisfies the criteria of antibunching of second order (5), since

$$\begin{aligned}d(2) &= \langle N^{(3)}(t)\rangle - \langle N(t)\rangle^3 \\ &= [|\alpha|^6 - g^2t^2(36|\alpha|^8 + 36|\alpha|^6)] - [|\alpha|^6 - 36g^2t^2|\alpha|^8] \\ &= -36g^2t^2|\alpha|^6\end{aligned} \tag{27}$$

is always negative.

3. Four wave mixing process

The four wave mixing process is often observed in resonant media. Specially, it is easy to achieve in a third-order nonlinear $(\chi^{(3)})$ medium under the phase matching condition [12]. This process has long been used as a tool to produce squeezed states. In fact, first experimental realization of squeezing [18] was achieved using this process. Wide domain of applications of this process includes coherent anti-Stokes Raman spectroscopy (CARS) to the generation of 1.5 μm band time bin entanglement [19]. One simple way to describe four wave mixing is as a process in which two photons of frequency ω_1 are absorbed (as pump photon) and one photon of frequency ω_2 and another of frequency ω_3 are emitted. The Hamiltonian representing this particular four wave mixing process is

$$H = a^{\dagger}a\omega_1 + b^{\dagger}b\omega_2 + c^{\dagger}c\omega_3 + g(a^{\dagger 2}bc + a^2b^{\dagger}c^{\dagger}). \tag{28}$$

Following the same prescription as it is used in the six wave case we can write the solution as

$$A(t) = A - 2igtA^{\dagger}BC + \frac{g^2t^2}{2!}[4AB^{\dagger}BC^{\dagger}C - 2A^{\dagger}A^2B^{\dagger}B - 2A^{\dagger}A^2C^{\dagger}C - 2A^{\dagger}A^2] \tag{29}$$

or

$$A(t) = A - 2igtA^{\dagger}BC + g^2t^2[2AN_BN_C - N_AAN_B - N_AAN_C - N_AA]. \tag{30}$$

The respective values of the first-order antibunching ($d(1)$) and second-order antibunching ($d(2)$) can similarly be calculated as we have done for six wave mixing and that yields

$$\begin{aligned} d(1) &= \langle N^{(2)}(t)\rangle - \langle N(t)\rangle^2 \\ &= [|\alpha|^4 + g^2t^2(-4|\alpha|^6 - 2|\alpha|^4)] - [|\alpha|^4 - 4g^2t^2|\alpha|^6] \\ &= -2g^2t^2|\alpha|^4, \end{aligned} \tag{31}$$

and

$$\begin{aligned} d(2) &= \langle N^{(3)}(t)\rangle - \langle N(t)\rangle^3 \\ &= [|\alpha|^6 - g^2t^2(6|\alpha|^8 + 6|\alpha|^6)] - [|\alpha|^6 - 6g^2t^2|\alpha|^8] \\ &= -6g^2t^2|\alpha|^6, \end{aligned} \tag{32}$$

which are negative and thus they satisfy our criterion for antibunching and HOA, respectively.

4. Second harmonic generation

The second harmonic generation or frequency doubling can be seen in all $\chi^{(2)}$ media. This process is used to reliably generate light with high intensity and good noise suppression [12]. The lowest order nonclassical effects are already observed in this process [12]. This process is described by the Hamiltonian

$$H = \hbar\omega N_1 + 2\hbar\omega N_2 + hg\left(a_2^{\dagger}a_1^2 + a_1^{\dagger 2}a_2\right). \tag{33}$$

Following the same procedure the second-order expression of the time evolution of the annihilation operator in the pump mode of the second harmonic generation can be obtained as

$$A(t) = a_1 - 2igta_1^{\dagger}a_2 + 2g^2t^2\left(a_2^{\dagger}a_2a_1 - \tfrac{1}{2}a_1^{\dagger}a_1^2\right). \tag{34}$$

Using the last equation along with the method used in section 2 we obtain

$$\begin{aligned} d(1) &= \langle N^{(2)}(t)\rangle - \langle N(t)\rangle^2 \\ &= [|\alpha|^4 + g^2t^2(-4|\alpha|^6 - 2|\alpha|^4)] - [|\alpha|^4 - 4g^2t^2|\alpha|^6] \\ &= -2g^2t^2|\alpha|^4 \end{aligned} \tag{35}$$

and

$$\begin{aligned} d(2) &= \langle N^{(3)}(t)\rangle - \langle N(t)\rangle^3 \\ &= [|\alpha|^6 - g^2t^2(6|\alpha|^8 + 6|\alpha|^6)] - [|\alpha|^6 - 6g^2t^2|\alpha|^8] \\ &= -6g^2t^2|\alpha|^6, \end{aligned} \tag{36}$$

which are both negative and hence satisfy the criterion (5) for antibunching and HOA, respectively.

5. Conclusions

In essence all the simple physical models described above are experimentally achievable and quite easily observed in any nonlinear optics laboratory. Photon statistics (factorial moment) of a particular mode after the interaction can be obtained experimentally by using a homodyne detection (photon counting) technique. These facts open up the possibility of experimental observation of HOA in many simple nonlinear optical processes. From (27), (32) and (36), it is clear that all the physical systems selected for the present study show second-order antibunching, i.e. higher order sub-Poissonian photon statistics. Thus, the present work strongly establishes the fact that HOA is not a rare phenomenon. In the case of an interaction of an intense electromagnetic field with a third-order nonlinear medium it was reported [11] that the degree of antibunching ($d(l)$) can be tuned because they depend strongly on the phase of the input field which can be tuned. This is not the case with any of the physical systems studied in the present work. It is also clear that the higher order antibunching would not have been observed if we have considered first-order operator solutions (first order in g); on the other hand, if we use second-order operator solutions, then the depth of nonclassicality is found to increase monotonically with the increase of the input photon number ($|\alpha|^2$). Possibly this monotonic increment will be ceased by the higher order perturbation terms. It is also observed that if we assume that the anharmonic constant and number of photon initially present in the pump mode are the same for all three cases, then the depth of nonclassicality is the same in the four wave mixing and second harmonic generation process and it is more in the six wave mixing process.

The prescription followed in the present work is easy and straightforward and it can be used to study the possibilities of observing higher order antibunching in other physical systems. Thus it opens up the possibility of studying higher order nonclassical effects from a new perspective. This is also important from the application point of view because any probabilistic single-photon source used for quantum cryptography has to satisfy the condition for higher order antibunching. Therefore, the simple prescription followed in the present work may help us to compare the existing sources of a single photon.

References

[1] Meystre P and Sargent M III 1991 *Elements of Quantum Optics* 2nd edn (Berlin: Springer)
[2] Dodonov V V 2002 *J. Opt. B: Quantum Semiclass. Opt.* **4** R1
[3] Hanbury-Brown R and Twiss R Q 1956 *Nature* **177** 27
[4] Hong C K and Mandel L 1985 *Phys. Rev. Lett.* **54** 323
[5] Hong C K and Mandel L 1985 *Phys. Rev.* A **32** 974
[6] Lee C T 1990 *Phys. Rev.* A **41** 1721
[7] Lee C T 1990 *Phys. Rev.* A **41** 1569
[8] Hillery M 1987 *Phys. Rev.* A **36** 3796
[9] Giri D K and Gupta P S 2003 *Opt. Commun.* **221** 135
[10] An N B 2002 *J. Opt. B: Quantum Semiclass. Opt.* **4** 222–7
[11] Pathak A and Garcia M submitted
[12] Bachor H A 1998 *A Guide to Experiments in Quantum Optics* (Weinheim: Wiley-VCH) chapters 8 and 10
[13] Vogel W, Welsch D and Wallentowitz S 2001 *Quantum Optics: an Introduction* 2nd edn (Berlin: Wiley-VCH) chapter 6
[14] Erenso D, Vyas R and Singh S 2002 *J. Opt. Soc.* B **19** 1471
[15] Vyas R and Singh S 1989 *Phys. Rev.* A **40** 5147
[16] Goran M *et al* 1993 *J. Opt. Soc.* B **10** 454
[17] Duppen K and Wiersma D A 1988 *Opt. Lett.* **13** 318
[18] Slusher R E *et al* 1985 *Phys. Rev. Lett.* **55** 2409
[19] Takesue H and Inoue K 2005 *Opt. Express* **13** 7832