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Original article

Pump photons present in a non-linear process as a witnesses of non-classicality of a system

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ABSTRACT We have studied non-classical effects, i.e. higher order photon antibunching (HOA) and higher order sub-Poissonian photon number statistics (HOSPS) in various non-linear optical processes like second harmonic generation, fourth harmonic generation, coherent anti-Stokes Raman scattering (CARS) and coherent anti-Stokes hyper-Raman scattering (CAHRS) using short time interaction techniques. The non-classical effects directly depend on number of photons prior to interaction with non-linear medium has already been studied but we have found that non-linear processes involving equal number of pump photons have same higher order photo antibunching (HOA) and higher order sub-Poissonian photon number statistics (HOSPS) independent of the non-linear process involved.

KEYWORDS Higher order sub-Poissonian photon number statistics, higher order photon antibunching, optical processes.

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1. Introduction

Non-classical states cannot be characterized by a mixture of coherent states and these are often defined by the negative values of Glauber-Sudarshan P-functions [1,2]. In fact, there is no method for determining the P-function experimentally, except for a single proposal [3]. A number of operational criteria for witnessing non classicality have been developed [4,5]. These observers of non-classicality can be communicated as the moments of creation and annihilation operators and a non-classical property seen through a moment-based model that observes a non-classical characteristic through a lower order connection is known as a lower-order non-classical property. Higher-order non-classicality, as the name implies, refers to the non-classical characteristics revealed by higher order correlations. Higher order photon antibunching (HOA) [6], higher-order sub-Poissonian photon number statistics (HOSPS) [7,8], higher order squeezing of Hong-Mandel type [9,10] are the most often researched higher order non-classical characteristics. All of these non-classical characteristics have lower order equivalents that have been well investigated [11, 12]. Due to the successful experimental characterizations of higher-order non-classical states, much attention has been given to these states recently [13–16]. The fact that weak non-classicalities not identified by their lower order equivalents can be recognized by higher order non-classicality criteria has led to a significant number of theoretical studies as well [14, 15]. Indeed, higher order photon antibunching (HOA) and higher order squeezing (HOS) has been accounted in an opto-mechanical like system [17], finite dimensional coherent state [5], optical coupler [18], hyper-Raman process [19] and higher order sub-Poissonian photon number statistics (HOSPS) has been accounted in finite dimensional coherent state [5], photon added and subtracted squeezed coherent states [20]. Previously, research into these non-classical phenomena was mostly for academic interest [21], but their numerous applications in quantum information theory, such as optical communication [14], dense coding [22], quantum teleportation [23], and quantum cryptography [24], are now well-known. Non-classicality has been shown to be a required input for the entangled state [25]. All of the physical systems mentioned above are experimentally feasible and may be easily seen in a non- linear optics laboratory [26, 27]. Photon number statistics may be obtained experimentally using the homodyne detection technique [28, 29]. A number of new opportunities for non-linear optics have arisen due to the rapid growth of nanotechnology and nanoscience. During the last few decades, non-linear optical materials have made significant progress in laser technology and these materials have large non-linear optical properties and a fast non-linear response for various photonic applications such as pulsed laser deposition, laser ablation, optical information processing, optical communication, optical limiters and optical data storage [30–35]. Nanomaterials with large non-linear responses are useful in photocatalysis and optical limiting applications [36] and optical non-linear microscopy [37]. Materials with large third order optical non-linearity and fast response time will be required for future optical device applications [38,39]. Due to their high non-linear optical response of these nonlinear optical materials, these materials are used in fiber optic communication systems such as all digital signal restoration, routing units, de- multiplexing and multiplexing and optical storage media [39] as well as in optical switching [40]. Strong optical non-linearities observed due to the quantum confinement effects such as second and third order optical non-linearities and non-linear optical absorption that can be studied for making laser second and third harmonic generators and optical modulators [38, 39]. There has been a lot of research done on measuring third order nonlinear susceptibility χ^3 to examine optical nonlinearity of nanoparticles may be Z scan technique and degenerate four wave mixing experiments [39]. Hanamura analyzed theoretically the third order optical polarizability χ^3 and the oscillator strength in semiconductor microcrystallites [41].

The state of non-classicality of the non-linear optical system is described in segment 2 of the current study. Segment 3 will introduce a second order solution of equation of motion by means of an illustration of fourth harmonic generation process and show the presence of higher order photon antibunching (HOA) and higher order sub-Poissonian photon number statistics (HOSPS). In segment 4, we have investigated the presence of higher order non-classical effects in second harmonic generation, coherent anti-Stokes Raman scattering process (CARS) and coherent anti-Stokes hyper-Raman scattering (CAHRS) non-linear optical processes, as well as their direct association with pump photons present in the system. In section 5, we used graphs to compare the results and section 6 is devoted to the conclusion.

2. Condition of non-classicality of a non-linear optical system

2.1. Condition for higher order photon antibunching(HOA)

Lee presented the higher order photon antibunching (HOA) criteria as follows [42]:

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

where number operator is represented by N.

$\left\langle N^{(k)}\right\rangle = \langle N(N-1)(N-2)\cdots(N-k+1)\rangle$ is the k^{th} factorial moment of the number operator. Integers l and p fulfilling condition $l \leq p \leq 1$ and x addendum indicates specific mode. $p = 1$ is picked by Ba An [43] and condition of the l^{th} order photon antibunching is reduced to

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

And:

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

Physically, a state which is photon antibunched in the l^{th} order must be photon antibunched in the $(l-1)^{th}$ order. Therefore, we can simplify (3) as:

$$\langle N_x^{l+1}\rangle < \langle N_x^{l}\rangle \langle N_x\rangle < \langle N_x^{l-1}\rangle \langle N_x^{2}\rangle < \cdots < \langle N_x\rangle^{l+1},$$

and obtain condition of l^{th} order photon antibunching as:

$$d(l) = \langle N_x^{l+1}\rangle - \langle N_x\rangle^{l+1} < 0. \tag{4}$$

From equation (4), we can see that for sub-Poissonian state $d(l) < 0$. Along these lines, we can say that a single photon source utilized in quantum cryptography should fulfill the criteria given in equation (4) of higher order photon antibunching (HOA).

2.2. Criteria for higher order sub-poissonian photon number statistics (HOSPS)

Mishra and Prakash [44] provide the criteria of $(l-1)^{th}$ order higher order sub-Poissonian photon number statistics (HOSPS) is given as:

$$D(l-1) = \sum_{k=0}^{l}\sum_{i=0}^{l-k} {}^{l}C_k(-1)^k S_2(l-k,i)\langle N^i\rangle \langle N\rangle^k - \sum_{k=0}^{l}\sum_{i=0}^{l-k} {}^{l}C_k(-1)^k S_2(l-k,i)\langle N\rangle^{k+i} < 0, \tag{5}$$

where $S_2(l,k)$ is the second-order Stirling number. For $l = 3$, the condition of second order sub-Poissonian photon number statistics (HOSPS) is described as:

$$D(2) = \langle N^3\rangle + 2\langle N\rangle^3 - 3\langle N^2\rangle\langle N\rangle + 3\langle N^2\rangle - 3\langle N\rangle^2 < 0. \tag{6}$$

3. Fourth harmonic generation process

To examine higher order photon antibunching (HOA) and higher order sub-Poissonian photon number statistics (HOSPS), we used the fourth harmonic generation process, which involves absorption of four photons, each with a frequency ω_1 and the emission of one photon with frequency ω_2 where $\omega_2 = 4\omega_1$. For this process, the Hamiltonian is:

$$H = \omega_1 a^\dagger a + \omega_2 b^\dagger b + g(a^4 b^\dagger + a^{\dagger 4} b), \tag{7}$$

where $a^\dagger$ (a), $b^\dagger$ (b) are the creation (annihilation) operators and g is coupling constant. $A = a \exp i\omega_1 t$, $B = b \exp i\omega_2 t$ are the gradually varying operators at frequencies ω_1 and ω_2.

3.1. Time evolution of pump mode A

The Heisenberg equation of motion for the time evolution of operator in mode A is given as:

$$\frac{dA}{dt} = \frac{\partial A}{\partial t} + i\left[H, A\right]. \tag{8}$$

We obtain:

$$\dot{A} = -4igA^{+3}B, \tag{9}$$

and

$$\dot{B} = -igA^{4}. \tag{10}$$

We now expand $A\left(t\right)$ using Taylor series expansion and treating terms up to g^2t^2 by using short time approximation as:

$$A(t) = A - 4igtA^{\dagger 3}B + 2g^2t^2(12A^{\dagger 2}A^3N_B + 36A^{\dagger}A^2N_B + 24AN_B - A^{\dagger 3}A^4). \tag{11}$$

Using equation (11), number operator $N_A(t) = A^{\dagger}(t)A(t)$ is given as:

$$N_A(t) = A^{\dagger}A - 4igt(A^{\dagger 4}B - A^4B^{\dagger}) + 4g^2t^2(16A^{\dagger 3}A^3N_B + 72A^{\dagger 2}A^2N_B + 96A^{\dagger}AN_B + 24N_B - A^{\dagger 4}A^4). \tag{12}$$

To investigate photon antibunching, we start with a quantum state that is the product of coherent state $|\alpha\rangle$ for pump mode A and vacuum state $|0\rangle$ for stokes mode B i.e.:

$$|\psi\rangle = |\alpha\rangle_A\,|0\rangle_B\,. \tag{13}$$

Using equation (13) in equation (12), we get average value of $\langle N_A(t)\rangle_\alpha = \left\langle A^{\dagger}(t)A(t)\right\rangle$ as:

$$\langle N_A(t)\rangle_\alpha = |\alpha|^2 - 4g^2t^2\,|\alpha|^8\,, \tag{14}$$

where $A\,|\alpha\rangle = \alpha\,|\alpha\rangle$. Now, the average value of $N_A^4(t)$ is given as:

$$\left\langle N_A^4(t)\right\rangle_\alpha = \left\langle A^{\dagger 4}(t)A^4(t)\right\rangle = |\alpha|^8 - 8g^2t^2(2\,|\alpha|^{14} + 9\,|\alpha|^{12} + 12\,|\alpha|^{10} + 3\,|\alpha|^8).$$

Now, using equations (14) in equation (4), we get third order photon antibunching as

$$d_A(3)_\alpha = -24g^2t^2(3\,|\alpha|^{12} + 4\,|\alpha|^{10} + |\alpha|^8). \tag{15}$$

Equation (15) shows that fourth harmonic generation process satisfies the criteria of higher order photon antibunching.

To examine higher order photon antibunching, we used the initial state which is equal to the product of vacuum state $|0\rangle$ for pump mode A and $|\beta\rangle$ for stokes mode B i.e.:

$$|\psi\rangle = |0\rangle_A\,|\beta\rangle_B\,. \tag{16}$$

Now, taking average values of $N_A^4(t)$ in pump mode A with respect to condition (16) is given as:

$$\left\langle N_A^4(t)\right\rangle_\beta = 0. \tag{17}$$

Using equation (17) in equation (4), we obtain third order photon antibunching in mode A in relation to $|0\rangle\,|\beta\rangle$ is given as:

$$d_A(3)_\beta = 0. \tag{18}$$

Equation (18) shows that higher order photon antibunching is absent in mode A with respect to the quantum state $|0\rangle\,|\beta\rangle$.

Now, using equations (12, 13 and 16) in equation (6), we obtain:

$$D(2)_\alpha = -60g^2t^2\,|\alpha|^8\,, \tag{19}$$

$$D(2)_\beta = 0,$$

we obtain a negative value in equation (19), which indicates that higher order sub-Poissonian photon number statistics exists in mode A with respect to quantum state $|\psi\rangle = |\alpha\rangle_A\,|0\rangle_B$.

4. Non-classicality in other non-linear optical processes

We have analyzed higher order photon antibunching (HOA) and higher order sub-Poissonian photon number statistics (HOSPS) in various non-linear optical processes and all the results that we have obtained is mentioned in Table 1 and Table 2 respectively.

In Table 1 and Table 2, $*$ represents the average values are taken with regards to $|\alpha\rangle\,|0\rangle\,|0\rangle$ in mode A.

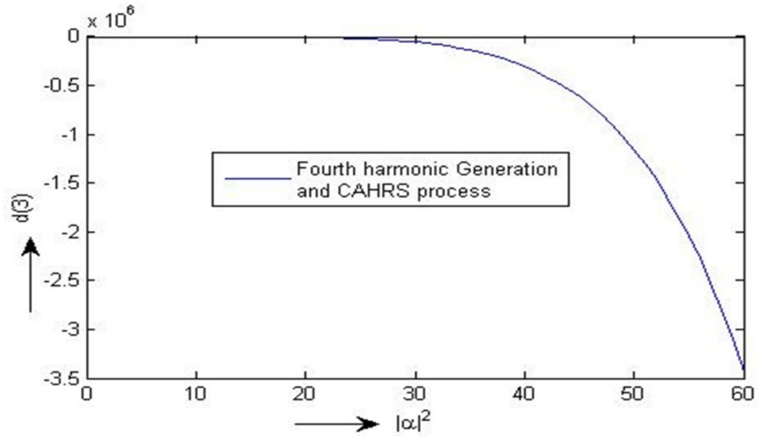

FIG. 1. The plot of third order photon antibunching $d(3)$ versus $|\alpha|^2$ in fourth harmonic generation and coherent anti-Stokes hyper-Raman scattering process (CAHRS) (taking $g^2t^2 \approx 10^{-6}$)

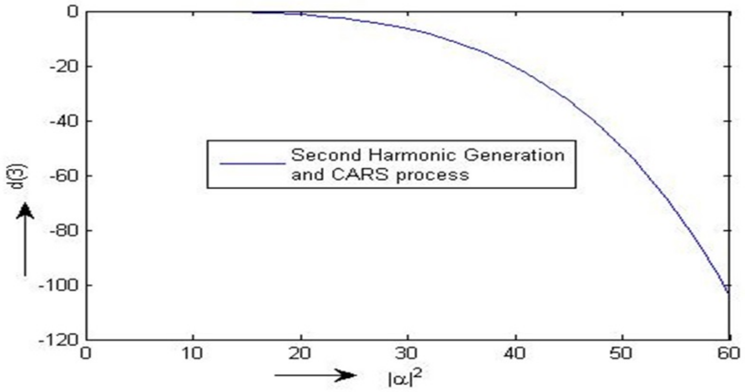

FIG. 2. The plot of third order photon antibunching $d(3)$ versus $|\alpha|^2$ in second harmonic generation and coherent anti-Stokes Raman scattering process (CARS) (taking $g^2t^2 \approx 10^{-6}$)

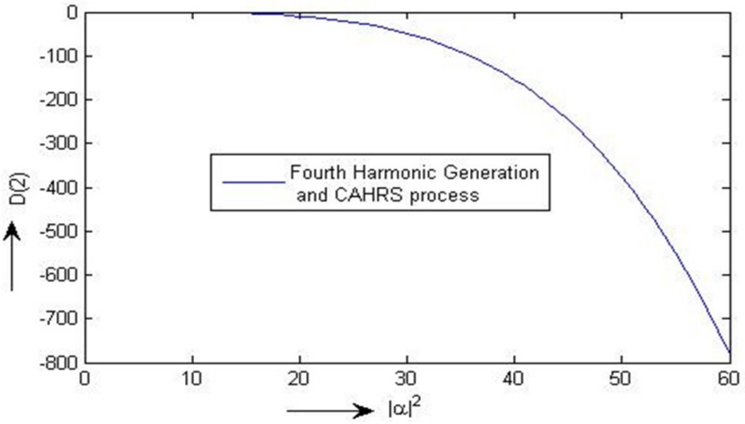

FIG. 3. The plot of higher order sub-Poissonian photon number statistics $D(2)$ versus $|\alpha|^2$ in fourth harmonic generation and coherent anti-Stokes hyper-Raman scattering process (CAHRS) (taking $g^2t^2 \approx 10^{-6}$)

TABLE 1. Results obtained for higher order photon antibunching (HOA) in non-linear optical processes

Sr. no.	Optical processes	Interaction term	Parameter $d(3)$ Expectation value w. r. t. $*$, $\vert\alpha\rangle\,\vert 0\rangle\,\vert 0\rangle$
1	Second harmonic generation	$A^{\dagger 2}B$	$d(3) = -8g^2t^2\,\vert\alpha\vert^8$
2	Coherent anti-Stokes Raman scattering (CARS) process	$A^{\dagger}BA^{\dagger}C$	$d(3) = -8g^2t^2\,\vert\alpha\vert^8$
3	Fourth harmonic generation	$A^{\dagger 4}B$	$d(3) = -24g^2t^2(3\,\vert\alpha\vert^{12} + 4\,\vert\alpha\vert^{10} + \vert\alpha\vert^8)$
4	Coherent anti-Stokes hyper-Raman scattering (CAHRS) process	$A^{\dagger 2}BA^{\dagger 2}C$	$d(3) = -24g^2t^2(3\,\vert\alpha\vert^{12} + 4\,\vert\alpha\vert^{10} + \vert\alpha\vert^8)$

TABLE 2. Results obtained for higher order sub-Poissonian photon number statistics in non-linear optical processes

Sr. no.	Optical processes	Interaction term	Parameter $D(2)$ Expectation value w. r. t. $*$, $\vert\alpha\rangle\,\vert 0\rangle\,\vert 0\rangle$
1	Second harmonic generation	$A^{\dagger 2}B$	$D(2) = -6g^2t^2(\vert\alpha\vert^4)$
2	Coherent anti-Stokes Raman scattering (CARS) process	$A^{\dagger}BA^{\dagger}C$	$D(2) = -6g^2t^2(\vert\alpha\vert^4)$
3	Fourth harmonic generation	$A^{\dagger 4}B$	$D(2) = -60g^2t^2\,\vert\alpha\vert^8$
4	Coherent anti-Stokes hyper-Raman scattering (CAHRS) process	$A^{\dagger 2}BA^{\dagger 2}C$	$D(2) = -60g^2t^2\,\vert\alpha\vert^8$

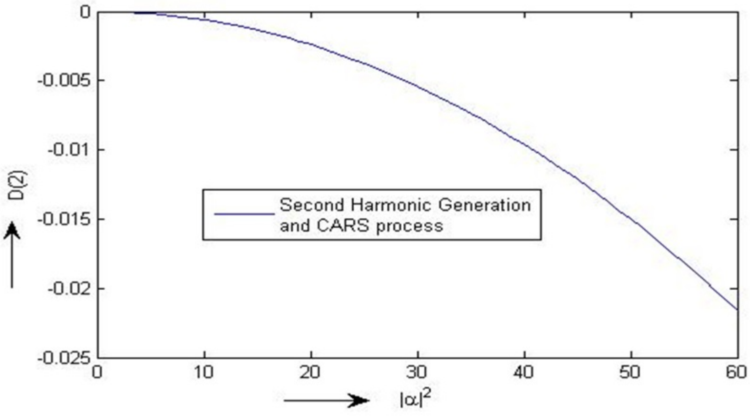

FIG. 4. The plot of higher order sub-Poissonian photon number statistics$D(2)$ versus $|\alpha|^2$ in second harmonic generation and coherent anti-Stokes Raman scattering process (CARS) (taking $g^2t^2 \approx 10^{-6}$)

5. Result

Tables 1 and Tables 2 illustrate the existence of higher order sub-Poissonian photon number statistics (HOSPS) and higher order photon antibunching (HOA) in different non-linear processes. If we plot a graph connecting higher order photon antibunching (HOA) say $d(3)$ and higher order sub-Poissonian photon number statistics (HOSPS) say D(2) with photon number in pump mode A i.e. $|\alpha|^2$, it is obvious that higher order sub-Poissonian photon number statistics (HOSPS) and higher order photon antibunching (HOA) increase non-linearly as $|\alpha|^2$ increases. Fig. 1 of higher order photon antibunching (HOA) and Fig. 3 of higher order sub-Poissonian photon number statistics (HOSPS) of fourth harmonic generation and coherent anti-Stokes hyper-Raman scattering (CAHRS) show the same values of higher order photon antibunching (HOA) as well as higher order sub-Poissonian photon number statistics (HOSPS). Similarly, Fig. 2 and Fig. 4 of second harmonic generation and coherent anti-Stokes Raman scattering process (CARS) also show the same values of higher order photon antibunching (HOA) as well as higher order sub-Poissonian photon number statistics (HOSPS). In all these non-linear optical processes, we have observed higher order photon antibunching (HOA) and higher order sub-Poissonian photon number statistics (HOSPS) only with respect to a quantum state which is the product of a coherent state $|\alpha\rangle$ for pump mode A and vacuum state $|0\rangle$ for Stokes mode B and signal mode C.

6. Conclusion

Higher order non-classical effects i.e. higher order photon antibunching (HOA) and higher order sub-Poissonian photon number statistics (HOSPS) in pump mode have been observed in a variety of non-linear optical processes. We have found that the non-linear processes having the same number of pump photons present prior to interaction have the same value of higher order photon antibunching and higher order sub-Poissonian photon number statistics, which we have demonstrated using examples of fourth harmonic generation with coherent anti-Stokes hyper-Raman scattering (CAHRS) and second harmonic generation with coherent anti-Stokes Raman scattering process (CARS). Further, we are obtaining the maximum value of non-classicality in fourth harmonic generation and coherent anti-Stokes hyper-Raman scattering process (CAHRS), as it has the maximum number of pump photons as compared to other non-linear optical processes which we have taken into consideration. As a result, we can conclude that non-classicality of a system can be revealed by the number of pump photons present in the system prior to interaction irrespective of the non-linear process involved. To study the higher order non-classical effects in various non-linear optical processes, we need the non-linear materials having higher order non-linear susceptibility which can be studied for making optical modulators and higher harmonic generators.

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