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Antibunching of Light in Coherent Anti-Stokes Hyper Raman Scattering

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This paper aims to study the photons antibunching in the fundamental, Stokes and signal modes of CAHRS (coherent anti-Stokes hyper Raman scattering). The generalized interaction Hamiltonian was calculated for different conditions in the Heisenberg picture under short time approximation to observe photons antibunching in the fundamental, Stokes and signal modes of CAHRS. It has been found that antibunching in the fundamental field is dependent upon interaction time, photons number, and coupling strength of field. Third order photon antibunching is found to be nonclassical to a greater extent than second- and first-order photon antibunching for the identical pump photons count. It has been also observed that the effect of photon antibunching in Stokes and signal modes is less prominent than in the fundamental- mode. Furthermore, it is also found that antibunching will increase at short interaction timeswith the increase of depth of nonclassical behavior. Finally, the extent of photons antibunching peaks precisely where the second correlation function is lowest.

Keywords: Photons antibunching, Photon number operator, Coherent anti-stokes hyper-raman scattering, Short-time approximation

1 Introduction

Antibunching of photons[1-10] is a non-classical property[11-14] of current significant interest for Quantum Teleportation[15, 16], Computation[17-21], Cryptography[22-24], and also for the utilization of Single Photon Sources[25-28] due to the unconditional security they provide. Single-photon sources are those that produce photons individually as discrete particles which generate an effective single photon number state. This shows there is a greater probability of producing a photon then there is of producing two, three, four, etc., at the same time. With the source in the antibunched state, it is known that the source of generating two or more photons simultaneously is less than in the single-photon state. Therefore, there is a higher likelihood of finding a source that produces a one photon over one emitting multiplein a bunch. Second-order temporal coherence serves as the most common method used for assessing the quality of a single photon source, with time delay being set to zero[26-28]. Stoler[1] was the first to develop a theory of how to create and observe photon antibunching in his work published in 1976. The majority of previous researchers[2-9] have been studying photon antibunching in various nonlinear optical setups. In 1990, the notion of higher order photons antibunching (HOPA) was developed[29, 30] and proposed the first criteria for higher order antibunching. The criteria for higher-order photon antibunching were later modified and extended in subsequent studies[31,32]. Higher order antibunching occurs frequently, but rather is easily observable in various straightforward nonlinear optical phenomena, including the six-wave mixing, the four-wave mixing and the second-harmonic generation[33]. Theoretical models studied here may be experimentally realized[34,35] and the conditions for achieving higher-order antibunching were determined using factorial moment which may be detected through homodyne photon-counting techniques[34–37]. Thus, all these uncomplicated physical models can be observed and implemented in standard nonlinear optics labs. Higher-order photons antibunching has also been widely investigated and published on over the last few years in several optical processes[38–42]. An extensive analysis of the theoretical and experimental aspects of multiphoton quantum optics was presented[43]. Their work provides a unified treatment of multiphoton events, with a Hamiltonian approach

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to describe higher-order nonlinear multiphoton processes with promising roles in quantum computing, teleportation, and various communication and information challenges. This opens new avenues of investigation into nonclassical effects of higher order.

In earlier study of nonclassical light generation, interaction models such as χ^2 and χ^3 processes have been widely used to study photon antibunching and related quantum statistical properties. These models mainly involve comparatively simpler nonlinear interactions and limited mode coupling which restrict the range of quantum correlations that can be explored. In contrast, the CAHRS process represents a higher order multi-photon interaction where multiple pump photons participate simultaneously in the generation of Stokes and anti-Stokes fields through molecular vibrational transitions. Such higher-order coupling can significantly influence photon statistics and enhance the possibility of observing stronger nonclassical effects. Therefore, analyzing photon antibunching in the CAHRS process provides a platform for studying nonclassicality generation and allows a meaningful comparison with previously studied χ type interaction processes.

2 Definition of Photon Antibunching

"The criterion for higher-order photon antibunching (HOPA) was proposed in earlier studies[29] and is given as:

$$R(q,p)=\left[\frac{\left\langle \hat{n}_y{}^{p-1}\right\rangle\left\langle \hat{n}_y{}^{q+1}\right\rangle}{\left\langle \hat{n}_y{}^{p}\right\rangle\left\langle \hat{n}_y{}^{q}\right\rangle}-1\right]<0 \qquad \dots (1)$$

where $\hat{n}$ is number operator of photons.

$\left\langle (n^{(s)})\right\rangle=\left\langle \{n\}\{n-1\}\{n-2\}............\{n-s+1\}\right\rangle$ is s^{th} number operator of photons. Both (q) and (p) fulfill the condition $(q\le p\le 1)$ and subscript (y) signifies the certain mode. "For $p=1$[31], Eq. (1) reduces to the following form:

$$\frac{\left\langle \hat{n}_y{}^{q+1}\right\rangle}{\left\langle \hat{n}_y{}^{q}\right\rangle\left\langle \hat{n}_y\right\rangle}-1<0 \qquad \dots (2)$$

and

$$\left\langle \hat{n}_y{}^{q+1}\right\rangle<\left\langle \hat{n}_y{}^{q}\right\rangle\left\langle \hat{n}_y\right\rangle \qquad \dots (3)$$

A state that exhibits photons antibunched in q^{th} order must also exhibit photons antibunched in the $(q-1)^{th}$ order. So, Eq. (3) can be simplified as:

$$\begin{gathered}\left\langle (\hat{n}_y{}^{(q+1)})\right\rangle<\left\langle (\hat{n}_y{}^{(q)})\right\rangle\left\langle (\hat{n}_y)\right\rangle<\left\langle (\hat{n}_y{}^{(q-1)})\right\rangle\\ \left\langle (\hat{n}_y)\right\rangle^2<..........<\left\langle (\hat{n}_y)\right\rangle^{(q+1)}\end{gathered} \qquad \dots (4)$$

and obtaining criterion for q^{th} order photons antibunching as:

$$d_y(q)=\left\langle \hat{n}_y{}^{q+1}\right\rangle-\left\langle \hat{n}_y\right\rangle^{q+1}<0 \qquad \dots (5)$$

The simplified condition expressed in Eq. (4) aligns precisely with the physical criterion for HOPA[32]. The expression $\left\langle \hat{a}_y^{\dagger q}\hat{a}_y^{q}\right\rangle=\left\langle \hat{n}_y^{q}\right\rangle$ signifies the likelihood of detecting q photons within the same mode at a given time. Thus, the interpretation of inequality (5) is detecting a single photon in bunch form exceeds that of two photon pulse, which in turn is higher than the detecting a three photon, and so forth. This behavior confirms HOPA criteria outlined in Eq. (5).

3 Coherent anti-Stokes Hyper Raman Scattering Process and its Hamiltonian

In a theoretical model for CAHRS, the interaction between the light and the molecule can be viewed in terms of a series of processes involving two pump photons of ω_1 frequency are absorbed with a Stokes photon at ω_2 frequency is emitted. The next step involves two more pump photons of ω_1 frequency are absorbed and an anti-Stokes photon of ω_3 frequency is emitted, ultimately returning molecular system to its original energy state.

The total energy (Hamiltonian) for the above considered system consist of non interacting and interacting energy terms as $H=H_0+H_I$ where H_0 and H_I represents the non interacting (free) and interacting Hamiltonian.

Since the CAHRS process involves several radiation modes such as pump, Stokes and anti-Stokes modes. Thus the non-interacting Hamiltonian describes the independent energy of the radiation modes before the nonlinear interaction takes place.

Thus, $H_0 = \hbar\omega_1 \hat{a}^\dagger \hat{a} + \hbar\omega_2 \hat{b}^\dagger \hat{b} + \hbar\omega_3 \hat{c}^\dagger \hat{c}$ and interaction between the electromagnetic field and the molecular system is defined by electric dipole approximation. The interaction Hamiltonian is given by

$H_I = -P.E$ where P is the polarization of the medium and E is the electric field.

In nonlinear optical processes the polarization of the medium can be expressed as: $P = \varepsilon_0 (\chi^{(1)} E + \chi^{(2)} E^2 + ...)$

For Higher order Raman Processes such as CAHRS, the interaction arises from higher order nonlinear polarization terms involving multiphoton coupling. The quantized electric field operator for each radiation mode can be written as:

$$E \infty a e^{-i\omega t} + a^\dagger e^{i\omega t}$$

By substituting the quantized electric field into the nonlinear polarization and retaining only the resonant terms using the rotating wave approximation. The interaction Hamiltonian describing the multiphoton coupling between pump, Stokes and anti-Stokes modes can be written as:

$$H_I = \hbar g (\hat{a}^2 \hat{b}^\dagger \hat{a}^2 \hat{c}^\dagger + \hat{a}^{\dagger 2} \hat{b} \hat{a}^{\dagger 2} \hat{c})$$

where g is the coupling constant representing the strength of the nonlinear interaction.

Thus total Hamiltonian for the coherent anti-Stokes hyper-Raman scattering is given as:

$$H = H_0 + H_I = \hbar\omega_1 \hat{a}^\dagger \hat{a} + \hbar\omega_2 \hat{b}^\dagger \hat{b} + \hbar\omega_3 \hat{c}^\dagger \hat{c} + \hbar g (\hat{a}^2 \hat{b}^\dagger \hat{a}^2 \hat{c}^\dagger + \hat{a}^{\dagger 2} \hat{b} \hat{a}^{\dagger 2} \hat{c})$$

(taking $\hbar = 1$) … (6)

where the nonlinear interaction term $\hat{a}^2 \hat{b}^\dagger \hat{a}^2 \hat{c}^\dagger$ in the Hamiltonian represents the annihilation of four pump photons of frequency ω_1 (A_1 mode) and the simultaneous creation of one Stokes photon of frequency ω_2 (B_1 mode) and one signal photon of frequency ω_3 (C_1 mode). Therefore, conservation of energy requires that total energy of absorbed pump photons equals the total energy of generated photon, i.e.

$$4\hbar\omega_1 = \hbar\omega_2 + \hbar\omega_3$$

This resonance condition follows from operator structure of the interaction Hamiltonian and ensures that the CAHRS process corresponds to an energy conserving multiphoton transition.

Since the operators represent observables, then the time development of the operators will correspond to the time development of those observables. Hence, using Eq. (6), can be obtained the Heisenberg equation for slowly developing operator in the fundamental mode $\hat{A}_1$ using Heisenberg interaction picture as:

$$\dot{\hat{A}}_1 = \frac{\partial \hat{A}_1}{\partial t} + i \left[\hat{H}, \hat{A}_1 \right]$$

The following expression is obtained:

$$\dot{\hat{A}}_1 = -4ig \hat{A}_1^{\dagger 3} \hat{B}_1 \hat{C}_1 \quad \dots (7)$$

and

$$\ddot{\hat{A}}_1 = 4g^2 (16 \hat{A}_1^{\dagger 2} \hat{A}_1^3 \hat{B}_1^\dagger \hat{B}_1 \hat{C}_1^\dagger \hat{C}_1 + 52 \hat{A}_1^\dagger \hat{A}_1^2 \hat{B}_1^\dagger \hat{B}_1 \hat{C}_1^\dagger \hat{C}_1 + 32 \hat{A}_1 \hat{B}_1^\dagger \hat{B}_1 \hat{C}_1^\dagger \hat{C}_1 - \hat{A}_1^{\dagger 3} \hat{A}_1^4 \hat{C}_1^\dagger \hat{C}_1 - \hat{A}_1^{\dagger 3} \hat{A}_1^4 \hat{B}_1^\dagger \hat{B}_1 - \hat{A}_1^{\dagger 3} \hat{A}_1^4) \quad \dots (8)$$

The present analytical solution is based on the short time approximation in which the field operators are expanded in a Taylor series and terms are retained only up-to second order in interaction time t. Since, the interaction of electromagnetic wave with nonlinear medium takes place for a very short time. Therefore, the obtained expressions are only valid in perturbative region of small interaction duration ($\approx 10^{-10}$ sec)[42, 44] and weak nonlinear coupling such as $|gt| << 1$. Under this condition, higher power of t can be neglected in the Taylor series expansion. A possible limitation of this approximation is that it may not accurately capture long-time evolution, stronger nonlinear coupling effects and higher order corrections that could modify the quantitative extent of photon antibunching.

$$\hat{A}_1[t] = \hat{A}_1[0] + \dot{\hat{A}}_1[t] + \ddot{\hat{A}}_1[\frac{t^2}{2!}] + ... \quad \dots (9)$$

Using Eqs. (7) and (8) in Eq. (9), the resulting expression is given by

$$\hat{A}_1(t) = \hat{A}_1 - 4ig\hat{A}_1^{\dagger 3}\hat{B}_1\hat{C}_1 + 2\,|\,g^2t^2\,|\,(16\hat{A}_1^{\dagger 2}\hat{A}_1^3\hat{B}_1^\dagger\hat{B}_1\hat{C}_1^\dagger\hat{C}_1$$
$$+52\hat{A}_1^{\dagger}\hat{A}_1^2\hat{B}_1^\dagger\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 + 32\hat{A}_1\hat{B}_1^\dagger\hat{B}_1\hat{C}_1^\dagger\hat{C}_1$$
$$-\hat{A}_1^{\dagger 3}\hat{A}_1^4\hat{C}_1^\dagger\hat{C}_1 - \hat{A}_1^{\dagger 3}\hat{A}_1^4\hat{B}_1^\dagger\hat{B}_1 - \hat{A}_1^{\dagger 3}\hat{A}_1^4) \qquad \ldots (10)$$

using the same approach but in reverse order

$$\hat{A}_1^\dagger(t) = \hat{A}_1^\dagger + 4ig\hat{A}_1^{3}\hat{B}_1^\dagger\hat{C}_1^\dagger + 2\,|\,g^2t^2\,|\,(16\hat{A}_1^{\dagger 3}\hat{A}_1^2\hat{B}_1^\dagger\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 +$$
$$52\hat{A}_1^{\dagger 2}\hat{A}_1\hat{B}_1^\dagger\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 + 32\hat{A}_1^\dagger\hat{B}_1^\dagger\hat{B}_1\hat{C}_1^\dagger\hat{C}_1$$
$$-\hat{A}_1^{\dagger 4}\hat{A}_1^3\hat{C}_1^\dagger\hat{C}_1 - \hat{A}_1^{\dagger 4}\hat{A}_1^3\hat{B}_1^\dagger\hat{B}_1 - \hat{A}_1^{\dagger 4}\hat{A}_1^3) \qquad \ldots (11)$$

where $\hat{A}_1(0) = \hat{A}_1$ at zero time

Substituting Eq. (6) into Heisenberg's interaction picture of mode $\hat{B}_1(t)$

$$\dot{\hat{B}}_1 = \frac{\partial \hat{B}_1}{\partial t} + i\left[\hat{H}, \hat{B}_1\right] \qquad \ldots (12)$$

$$\dot{\hat{B}}_1 = -ig\hat{A}_1^4\hat{C}_1^\dagger \qquad \ldots (13)$$

and

$$\ddot{\hat{B}}_1 = g^2(16\hat{A}_1^{\dagger 3}\hat{A}_1^3\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 - 72\hat{A}_1^{\dagger 2}\hat{A}_1^2\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 -$$
$$96\hat{A}_1^{\dagger}\hat{A}_1\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 - 24\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 + \hat{A}_1^{\dagger 4}\hat{A}_1^4\hat{B}_1) \qquad \ldots (14)$$

Expanding the Taylor series for stokes mode at short times (10^{-10} s) and keeping up to '(g^2t^2)' as follows

$$\hat{B}_1(t) = \hat{B}_1(0) + \dot{\hat{B}}_1(t) + \ddot{\hat{B}}_1\left(\frac{t^2}{2!}\right) + \ldots\ldots\ldots. \qquad \ldots (15)$$

By substituting Eqs. (13) and (14) into Eq. (15) as:

$$\hat{B}_1(t) = \hat{B}_1 - i\,|\,gt\,|\,\hat{A}_1^4\hat{C}_1^\dagger + \frac{|\,g^2t^2\,|}{2}(16\hat{A}_1^{\dagger 3}\hat{A}_1^3\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 -$$
$$72\hat{A}_1^{\dagger 2}\hat{A}_1^2\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 - 96\hat{A}_1^{\dagger}\hat{A}_1\hat{B}_1\hat{C}_1^\dagger\hat{C}_1$$
$$-24\hat{B}_1\hat{C}_1^\dagger\hat{C}_1 + \hat{A}_1^{\dagger 4}\hat{A}_1^4\hat{B}_1) \qquad \ldots (16)$$

and its complex conjugate

$$\hat{B}_1^\dagger(t) = \hat{B}_1^\dagger + i\left|gt\right|\hat{A}_1^{\dagger 4}\hat{C}_1 + \frac{\left|g^2t^2\right|}{2}(16\hat{A}_1^{\dagger 3}\hat{A}_1^3\hat{B}_1^\dagger\hat{C}_1^\dagger\hat{C}_1 -$$
$$72\hat{A}_1^{\dagger 2}\hat{A}_1^2\hat{B}_1^\dagger\hat{C}_1^\dagger\hat{C}_1 - 96\hat{A}_1^{\dagger}\hat{A}_1\hat{B}_1^\dagger\hat{C}_1^\dagger\hat{C}_1$$
$$-24\hat{B}_1^\dagger\hat{C}_1^\dagger\hat{C}_1 + \hat{A}_1^{\dagger 4}\hat{A}_1^4\hat{B}_1^\dagger) \qquad \ldots (17)$$

Similarly, it can be derived $\hat{C}_1(t)$ as

$$\dot{\hat{C}}_1 = \frac{\partial \hat{C}_1}{\partial t} + i\left[\hat{H}, \hat{C}_1\right] \qquad \ldots (18)$$

The resulting expression is given by

$$\dot{\hat{C}}_1 = -ig\hat{A}_1^4\hat{B}_1^\dagger \qquad \ldots (19)$$

and

$$\ddot{\hat{C}}_1 = g^2(-16\hat{A}_1^{\dagger 3}\hat{A}_1^3\hat{B}_1^\dagger\hat{B}_1\hat{C}_1 - 72\hat{A}_1^{\dagger 2}\hat{A}_1^2\hat{B}_1^\dagger\hat{B}_1\hat{C}_1 -$$
$$96\hat{A}_1^{\dagger}\hat{A}_1\hat{B}_1^\dagger\hat{B}_1\hat{C}_1 - 24\hat{B}_1^\dagger\hat{B}_1\hat{C}_1 + \hat{A}_1^{\dagger 4}\hat{A}_1^4\hat{C}_1) \qquad \ldots (20)$$

The time-dependent amplitude of the signal mode is expanded with respect to short time up to 'g^2t^2' as follows:

$$\hat{C}_1[t] = \hat{C}_1[0] + \dot{\hat{C}}_1[t] + \ddot{\hat{C}}_1[\frac{t^2}{2!}] + \ldots\ldots\ldots. \qquad \ldots (21)$$

Substituting Eqs. (19) and (20) into Eq. (21) yields:

$$\hat{C}_1(t) = \hat{C}_1 - i\,|\,gt\,|\,\hat{A}_1^4\hat{B}_1^\dagger + \frac{|\,g^2t^2\,|}{2}(-16\hat{A}_1^{\dagger 3}\hat{A}_1^3\hat{B}_1^\dagger\hat{B}_1\hat{C}_1 -$$
$$72\hat{A}_1^{\dagger 2}\hat{A}_1^2\hat{B}_1^\dagger\hat{B}_1\hat{C}_1 - 96\hat{A}_1^{\dagger}\hat{A}_1\hat{B}_1^\dagger\hat{B}_1\hat{C}_1$$
$$-24\hat{B}_1^\dagger\hat{B}_1\hat{C}_1 + \hat{A}_1^{\dagger 4}\hat{A}_1^4\hat{C}_1) \qquad \ldots (22)$$

and its conjugate

$$\hat{C}_1^\dagger(t) = \hat{C}_1^\dagger + i\,|\,gt\,|\,\hat{A}_1^{\dagger 4}\hat{C}_1 + \frac{|\,g^2t^2\,|}{2}$$
$$(16\hat{A}_1^{\dagger 3}\hat{A}_1^3\hat{B}_1^\dagger\hat{C}_1^\dagger\hat{C}_1 - 72\hat{A}_1^{\dagger 2}\hat{A}_1^2\hat{B}_1^\dagger\hat{C}_1^\dagger\hat{C}_1 - 96\hat{A}_1^{\dagger}\hat{A}_1\hat{B}_1^\dagger\hat{C}_1^\dagger\hat{C}_1$$
$$-24\hat{B}_1^\dagger\hat{C}_1^\dagger\hat{C}_1 + \hat{A}_1^{\dagger 4}\hat{A}_1^4\hat{B}_1^\dagger) \qquad \ldots (23)$$

3.1 Photons Antibunching in the A_1 Mode

The quantum state $\left|\psi\right\rangle_{A_1}$ is defined as follows:

$$\left|\psi\right\rangle_{A_1} = \left|\alpha'\right\rangle_{A_1}\left|0\right\rangle_{B_1}\left|0\right\rangle_{C_1} \qquad \ldots (24)$$

Using Eq. (24) in Eqs. (10) and (11) to get the average value of photon number operator as:

$$\left\langle\psi\left|\hat{A}_1(t)\right|\psi\right\rangle = \left\langle\psi\left|\hat{A}_1 - 2\,|\,g^2t^2\,|\,(\hat{A}_1^{\dagger 3}\hat{A}_1^4)\right|\psi\right\rangle \qquad \ldots (25)$$

and

$$\left\langle\psi\left|\hat{A}_1^\dagger(t)\right|\psi\right\rangle = \left\langle\psi\left|\hat{A}_1^\dagger - 2\,|\,g^2t^2\,|\,(\hat{A}_1^{\dagger 4}\hat{A}_1^3)\right|\psi\right\rangle \qquad \ldots (26)$$

Using Eqs. (25) and (26), the expectation values for $\hat{n}_{A_1}(t)$ are given as:

$$\left\langle \psi \middle| \hat{n}_{A_1}(t) \middle| \psi \right\rangle = \left\langle \psi \middle| \hat{A}_1^{\dagger}(t)\hat{A}_1(t) \middle| \psi \right\rangle = \left\langle \psi \middle| (\hat{A}_1^{\dagger}\hat{A}_1 - 4\,|gt|^2\,(\hat{A}_1^{\dagger 4}\hat{A}_1^{4})) \middle| \psi \right\rangle = |\alpha|^2 - 4(gt)^2\,|\alpha'|^8 \quad \ldots (27)$$

where $\left\langle \hat{n}_{A_1} \right\rangle = \left\langle \hat{A}_1^{\dagger}\hat{A}_1 \right\rangle = |\alpha'|^2$ represents the photon number operator in A_1 mode $\alpha' = |\alpha'|\exp(i\theta_1)$ and $\alpha'^{*} = |\alpha'^{*}|\exp(-i\theta_1)$ where α'^{*} is the complex conjugate of α'; θ_1 denotes the phase angle of the field.

In addition, using Eq. (27), represent average value of $\hat{n}_{A_1}^2(t)$, as well as the squared of average of $\hat{n}_{A_1}(t)$ as follows:

$$\left\langle \psi \middle| \hat{n}_{A_1}^2(t) \middle| \psi \right\rangle = \left\langle \psi \middle| \hat{A}_1^{\dagger 2}(t)\hat{A}_1^2(t) \middle| \psi \right\rangle = \left\langle \psi \middle| (\hat{A}_1^{\dagger 2}\hat{A}_1^2 - 4(gt)^2(2\hat{A}_1^{\dagger 5}\hat{A}_1^5 + 3\hat{A}_1^{\dagger 4}\hat{A}_1^4)) \middle| \psi \right\rangle = |\alpha|^4 - 4\,|gt|^2\,(2|\alpha'|^{10} + 3|\alpha'|^8) \quad \ldots (28)$$

and

$$\left\langle \psi \middle| \hat{n}_{A_1}(t) \middle| \psi \right\rangle^2 = \left\langle \psi \middle| \hat{A}_1^{\dagger}(t)\hat{A}_1(t) \middle| \psi \right\rangle^2 = |\alpha|^4 - 8\,|gt|^2\,|\alpha'|^{10} \quad \ldots (29)$$

Using Eqs. (28) and (29) in Eq. (5), the expression for normal photon antibunching ($q = 1$) in the A_1 mode is obtained as follows:

$$d_{A_1}(1) = \left[\left\langle \psi \middle| \Delta\hat{n}_{A_1}(t) \middle| \psi \right\rangle\right]^2 = \left\langle \psi \middle| \hat{n}_{A_1}^2(t) \middle| \psi \right\rangle - \left\langle \psi \middle| \hat{n}_{A_1}(t) \middle| \psi \right\rangle^2 = -12\,|g^2t^2|\,|\alpha'|^8 \quad \ldots (30)$$

Hence, the Eq. (5) shows the first-order photons antibunching present in CAHRS process due to negative values obtained in Eq. (30).

Similarly, for $\hat{n}_{A_1}^3(t)$, the following expression for $q = 2$ in the A_1 mode is obtained:

$$\left\langle \psi \middle| \hat{n}_{A_1}^3(t) \middle| \psi \right\rangle = |\alpha'|^6 - 4\,|gt|^2\,(3|\alpha'|^{12} + 9|\alpha'|^{10} + 6|\alpha'|^8) \quad \ldots (31)$$

and cube of average of $\hat{n}_{A_1}(t)$ is obtain as:

$$\left\langle \psi \middle| \{\hat{n}_{A_1}(t)\} \middle| \psi \right\rangle^3 = |\alpha'|^6 - 12\,|gt|^2\,|\alpha'|^{12} \quad \ldots (32)$$

Using Eqs. (31) and (32) in Eq. (5), the expression for second-order photon antibunching ($q = 2$) in A_1 mode is obtained as follows:

$$d_{A_1}(2) = \left\langle \psi \middle| \hat{n}_{A_1}^3(t) \middle| \psi \right\rangle - \left\langle \psi \middle| \hat{n}_{A_1}(t) \middle| \psi \right\rangle^3 = -12\,|g^2t^2|\,(3|\alpha'|^{10} + 2|\alpha'|^8) \quad \ldots (33)$$

Equation (33) satisfies the condition given in Eq. (5), since a negative value is obtained for the CAHRS process.

The average value of $\hat{n}_{A_1}^4(t)$ is given as:

$$\left\langle \psi \middle| \hat{n}_{A_1}^4(t) \middle| \psi \right\rangle = |\alpha'|^8 - 4\,|gt|^2\,(4|\alpha'|^{14} + 18|\alpha'|^{12} + 24|\alpha'|^{10} + 6|\alpha'|^8) \quad \ldots (34)$$

and the fourth power of the average value of $\hat{n}_{A_1}(t)$ is:

$$\left\langle \psi \middle| \hat{n}_{A_1}(t) \middle| \psi \right\rangle^4 = |\alpha'|^8 - 16\,|gt|^2\,|\alpha'|^{14} \quad \ldots (35)$$

For $q = 3$, substitution of Eqs. (34) and (35) into Eq. (5) yields the expression for higher-order photon antibunching ($q = 3$) in A_1 mode as follows:

$$d_{A_1}(3) = \left\langle \psi \middle| \hat{n}_{A_1}^4(t) \middle| \psi \right\rangle - \left\langle \psi \middle| \hat{n}_{A_1}(t) \middle| \psi \right\rangle^4 = -12\,|g^2t^2|\,(6|\alpha'|^{12} + 8|\alpha'|^{10} + 2|\alpha'|^8) \quad \ldots (36)$$

Here, also the criterion of Eq. (5) for higher order photon antibunching is satisfied for the CAHRS process.

3.2 Photons Antibunching in the B_1 Mode

Initial quantum state is defined by $|\psi\rangle_{B_1}$ as:

$$|\psi\rangle_{B_1} = |0\rangle_{A_1}\,|\beta'\rangle_{B_1}\,|0\rangle_{C_1} \quad \ldots (37)$$

Using Eq. (37) in Eqs (16) and (17) the average values of first and second order are given:

$$\left\langle \psi \middle| [\hat{n}_{B_1}(t)] \middle| \psi \right\rangle = \left\langle \psi \middle| [\hat{B}_1^{\dagger}(t)\hat{B}_1(t)] \middle| \psi \right\rangle = \left\langle \psi \middle| (\hat{B}_1^{\dagger}\hat{B}_1) \middle| \psi \right\rangle = |\beta'|^2 \quad \ldots (38)$$

$$\left\langle \psi \middle| [\hat{n}_{B_1}^2(t)] \middle| \psi \right\rangle = \left\langle \psi \middle| \{\hat{B}_1^{\dagger 2}(t)\}\,\{\hat{B}_1^2(t)\} \middle| \psi \right\rangle = |\beta'|^4 \quad \ldots (39)$$

and

$$\left\langle \psi \middle| \hat{n}_{B_1}(t) \middle| \psi \right\rangle^2 = \left\langle \psi \middle| \hat{B}_1^{\dagger}(t)\hat{B}_1(t) \middle| \psi \right\rangle^2 = |\beta'|^4 \quad \ldots (40)$$

Therefore, first-order photons antibunching in the B_1 mode is:

$$d_{B_1}(1) = \left[\left\langle \psi \middle| \Delta\hat{n}_{B_1}(t) \middle| \psi \right\rangle\right]^2 = \left\langle \psi \middle| \hat{n}^2_{B_1}(t) \middle| \psi \right\rangle - \left\langle \psi \middle| \hat{n}_{B_1}(t) \middle| \psi \right\rangle^2 = 0 \qquad \ldots (41)$$

Additionally, using similar way, one can obtained the $d_{B_1}(2)$ and $d_{B_1}(3)$ in B_1 mode as:

$$d_{B_1}(2) = \left\langle \psi \middle| \hat{n}^3_{B_1}(t) \middle| \psi \right\rangle - \left\langle \psi \middle| \hat{n}_{B_1}(t) \middle| \psi \right\rangle^3 = 0 \qquad \ldots (42)$$

and

$$d_{B_1}(3) = \left\langle \psi \middle| \hat{n}^4_{B_1}(t) \middle| \psi \right\rangle - \left\langle \psi \middle| \hat{n}_{B_1}(t) \middle| \psi \right\rangle^4 = 0 \qquad \ldots (43)$$

here the average values of $\hat{n}^3_{B_1}(t)$ and $\hat{n}^4_{B_1}(t)$ is obtained as:

$$\left\langle \psi \middle| \hat{n}^3_{B_1}(t) \middle| \psi \right\rangle = \left\langle \psi \middle| \hat{B}_1^{\dagger 3}(t)\hat{B}_1^3(t) \middle| \psi \right\rangle = |\beta'|^6 \qquad \ldots (44)$$

$$\left\langle \psi \middle| \hat{n}_{B_1}(t) \middle| \psi \right\rangle^3 = \left\langle \psi \middle| \hat{B}_1^{\dagger}(t)\hat{B}_1(t) \middle| \psi \right\rangle^3 = |\beta'|^6 \qquad \ldots (45)$$

$$\left\langle \psi \middle| \hat{n}^4_{B_1}(t) \middle| \psi \right\rangle = \left\langle \psi \middle| \hat{B}_1^{\dagger 4}(t)\hat{B}_1^4(t) \middle| \psi \right\rangle = |\beta'|^8 \qquad \ldots (46)$$

$$\left\langle \psi \middle| \hat{n}_{B_1}(t) \middle| \psi \right\rangle^4 = \left\langle \psi \middle| \hat{B}_1^{\dagger}(t)\hat{B}_1(t) \middle| \psi \right\rangle^4 = |\beta'|^8 \qquad \ldots (47)$$

Equations (41-43) do not satisfy the criteria of Eq. (5); therefore, there does exist neither first nor higher-order photons antibunching in B_1 mode in CAHRS.

3.3 Photons Antibunching in the C_1 Mode up to Second-Order

Here, it can defined $|\psi\rangle_{C_1}$ as

$$|\psi\rangle_{C_1} = |0\rangle_{A_1} |0\rangle_{B_1} |\gamma'\rangle_{C_1} \qquad \ldots (48)$$

The average value of $\hat{n}_{C_1}(t)$ by using Eq. (48) in Eqs. (22) and (23) as:

$$\left\langle \psi \middle| \hat{n}_{C_1}(t) \middle| \psi \right\rangle = \left\langle \psi \middle| \hat{C}_1^{\dagger}(t)\hat{C}_1(t) \middle| \psi \right\rangle = \left\langle \psi \middle| (\hat{C}_1^{\dagger}\hat{C}_1) \middle| \psi \right\rangle = |\gamma'|^2 \qquad \ldots (49)$$

Therefore, the average values of $\hat{n}^2_{C_1}(t)$ and the squared of the average values of $\hat{n}_{C_1}(t)$ of Eq. (49) is obtained as:

$$\left\langle \psi \middle| \hat{n}^2_{C_1}(t) \middle| \psi \right\rangle = \left\langle \psi \middle| \hat{C}_1^{\dagger 2}(t)\hat{C}_1^2(t) \middle| \psi \right\rangle = |\gamma'|^4 \qquad \ldots (50)$$

and

$$\left\langle \psi \middle| \hat{n}_{C_1}(t) \middle| \psi \right\rangle^2 = \left\langle \psi \middle| \hat{C}_1^{\dagger}(t)\hat{C}_1(t) \middle| \psi \right\rangle^2 = |\gamma'|^4 \qquad \ldots (51)$$

Hence, the first-order antibunching in the C_1 mode is:

$$d_{C_1}(1) = \left[\left\langle \psi \middle| \Delta\hat{n}_{C_1}(t) \middle| \psi \right\rangle\right]^2 = \left\langle \psi \middle| \hat{n}^2_{C_1}(t) \middle| \psi \right\rangle - \left\langle \psi \middle| \hat{n}_{C_1}(t) \middle| \psi \right\rangle^2 = 0 \qquad \ldots (52)$$

Similarly, it can be obtained $d_{C_1}(2)$ and $d_{C_1}(3)$ as:

$$d_{C_1}(2) = \left\langle \psi \middle| \hat{n}^3_{C_1}(t) \middle| \psi \right\rangle - \left\langle \psi \middle| \hat{n}_{C_1}(t) \middle| \psi \right\rangle^3 = 0 \qquad \ldots (53)$$

and

$$d_{C_1}(3) = \left\langle \psi \middle| \hat{n}^4_{C_1}(t) \middle| \psi \right\rangle - \left\langle \psi \middle| \hat{n}_{C_1}(t) \middle| \psi \right\rangle^4 = 0 \qquad \ldots (54)$$

where the average values of the $\hat{n}^3_{C_1}(t)$ and $\hat{n}^4_{C_1}(t)$ are:

$$\left\langle \psi \middle| \hat{n}^3_{C_1}(t) \middle| \psi \right\rangle = \left\langle \psi \middle| \hat{C}_1^{\dagger 3}(t)\hat{C}_1^3(t) \middle| \psi \right\rangle = |\gamma'|^6 \qquad \ldots (55)$$

$$\left\langle \psi \middle| \hat{n}_{C_1}(t) \middle| \psi \right\rangle^3 = \left\langle \psi \middle| \hat{C}_1^{\dagger}(t)\hat{C}_1(t) \middle| \psi \right\rangle^3 = |\gamma'|^6 \qquad \ldots (56)$$

$$\left\langle \psi \middle| \hat{n}^4_{C_1}(t) \middle| \psi \right\rangle = \left\langle \psi \middle| \hat{C}_1^{\dagger 4}(t)\hat{C}_1^4(t) \middle| \psi \right\rangle = |\gamma'|^8 \qquad \ldots (57)$$

$$\left\langle \psi \middle| \hat{n}_{C_1}(t) \middle| \psi \right\rangle^4 = \left\langle \psi \middle| \hat{C}_1^{\dagger}(t)\hat{C}_1(t) \middle| \psi \right\rangle^4 = |\gamma'|^8 \qquad \ldots (58)$$

Equations (52-54) do not fulfill the criterion of Eq. (5); hence, first and higher-order photons antibunching does not exist in the C_1 mode in the CAHRS process.

4 The Photons Number Correlation

The second-order correlation functioni.e. $[g^2(0)']$ can be written in terms of photons number[10, 12, 27]:

$$g^2(0)' = 1 + \frac{\left\langle (\Delta n_{A_1})^2 \right\rangle - \left\langle n_{A_1} \right\rangle}{\left\langle n_{A_1} \right\rangle^2} \qquad \ldots (59)$$

Substituting Eqs. (27), (29), and (30), in Eq. (59) as:

$$g^2(0)' = 1 + \frac{-12|g^2t^2||\alpha'|^8 - (|\alpha|^2 - 4|gt|^2|\alpha'|^8)}{|\alpha|^4 - 8(gt)^2|\alpha'|^{10}} \qquad \ldots (60)$$

To make the connection between the antibunching criterion and second-order correlation function more explicit, the first order photon antibunching condition for $q = 1$ from Eq. (5) may be written as:

$$d(1) = \langle n(n-1)\rangle - \langle n\rangle^2 \qquad \dots (61)$$

On the other hand, the zero delay second order correlation function from Eq. (59) is defined as:

$$g^2(0)' = 1 + \frac{\langle(\Delta n)^2\rangle - \langle n\rangle}{\langle n\rangle^2} = \frac{\langle n(n-1)\rangle}{\langle n\rangle^2} \qquad \dots (62)$$

Using Eq. (62) in above Eq. (61) as:

$$d(1) = \langle n^2\rangle\left[g^2(0)' - 1\right] \qquad \dots (63)$$

Since $\langle n\rangle^2 > 0$ for coherent field, the condition $d(1) < 0$ is analytically equivalent to $g^2(0)' < 1$.

Thus, the negativity of the factorial moment based photon antibunching criterion in the present analysis corresponds to zero-delay antibunching condition expressed in terms of the second order correlation function.

5 Results and Discussion

Figure 1 illustrates the variation of the left-hand side of Eqs. (30), (33) and (35) say $d_{A_1}(l)$ versus $|\alpha'|^2$ with $|g^2t^2| \square 10^{-10}$ demonstrates an exponential decrease as $|\alpha'|^2$ is increases. This trend indicates that photon antibunching increases nonlinearly as the number of pump photons $|\alpha'|^2$ rises. Therefore, photon antibunched states emerge in conjunction with high number of pump photons. The comparison between the $d_{A_1}(3)$ curve shown in Fig. 1 with those of second- and first-order $d_{A_1}(2)$ and $d_{A_1}(1)$ respectively, indicates that for an equal number of photons the degree of photon antibunching of the third-order exceeds those of the first- and second-order. Therefore, the third-order photon antibunching $d_{A_1}(3)$ has a greater nonclassicality than $d_{A_1}(2)$ and $d_{A_1}(1)$ photon antibunching. Hence, higher order i.e. second and third photon antibunching allows for a significantly greater noise reduction than the first order photon antibunching.

Figure 2 displays plots of $d_{A_1}(l)$ with $|gt|^2$ for the Eqs. (30), (33) and (35). These illustrate that photon antibunching varies and increases with greater interaction of the pump field and strength of the field. In addition to the plots corresponding to Eq. (35), Fig. 3 also illustrates the variation of $d_{A_1}(l)$ with $|\alpha'|^2$ for varying values of $|gt|$. This plot indicates that there is an upper limit to the maximum antibunching, which varies with interaction time, and this limit is reached at short interaction duration. In addition, the plot of Fig. (3) illustrates the fact that with decreasing interaction time, the effect of antibunching of photons increases[45-47]. The photon antibunching is not observed in the Stokes and signal modes up to second-order Hamiltonian interactions in the CAHRS process. The observed asymmetry primarily arises from the intrinsic nature of the CAHRS interaction rather than solely from the coherent-state input condition. The observed asymmetry primarily arises from the intrinsic nature of the CAHRS interaction

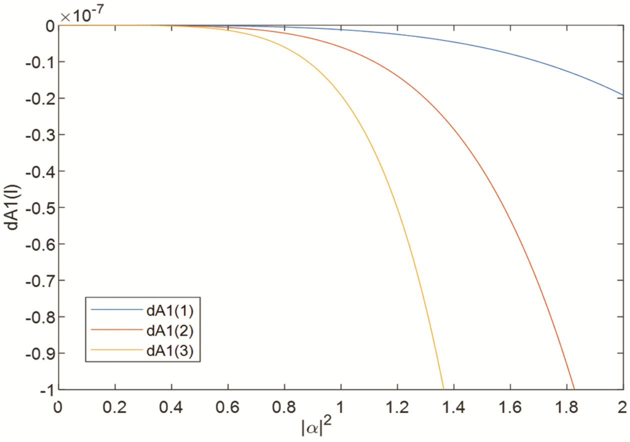

Fig. 1 — Variation of $d_{A_1}(l)$ with $|\alpha'|^2$ (when $|g^2t^2| = 10^{-10}$)

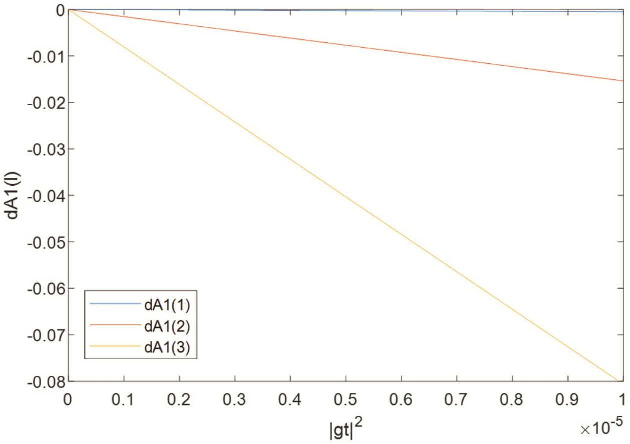

Fig. 2 — Variation of $d_{A_1}(l)$ with $|gt|^2$ (when $|\alpha'|^2 = 2$ (arbitrarily value))

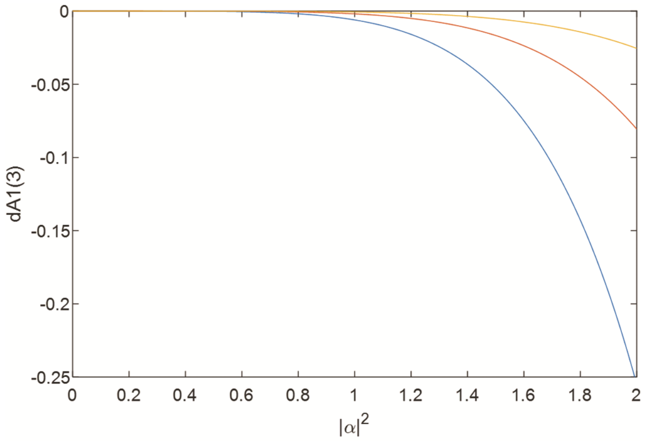

Fig. 3 — Variation of $d_{A_1}(3)$ with $|\alpha'|^2$ ($|gt| = 10^{-4.5}, 10^{-5}, 10^{-5.5}$)

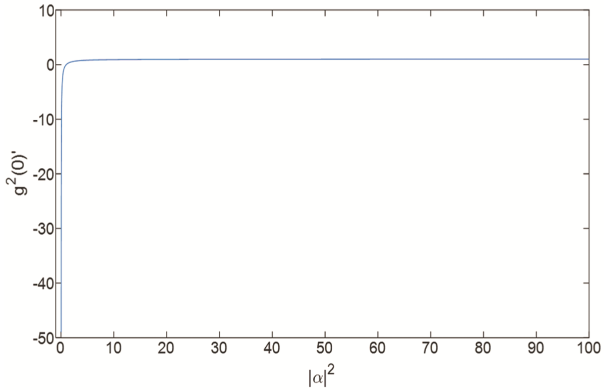

Fig. 4 — Variation of $g^2(0)'$ versus $|\alpha'|^2$ (when $|gt|^2 = 10^{-5}$)

rather than solely from the coherent-state input condition. Physically, this behavior arises because the pump mode acts as the depletion mode in the CAHRS interaction, where four pump photons are annihilated simultaneously in each nonlinear event. Such multiphoton depletion enhances the suppression of simultaneous photon occupancy and can therefore generate sub-Poissonian fluctuations and antibunching in the fundamental mode. By contrast, the Stokes and signal modes behave as generated modes because they are populated through creation operators and acquire photons through correlated emission from the pump field. In the present short time regime, these generated modes do not develop sufficiently strong negative fluctuation, instead their leading contributions remain positive and are associated with photon buildup rather than photon suppression. Consequently, the antibunching inequalities for the B_1 and C_1 modes are not satisfied up to second order in the interaction time. The coherent-state initial condition used in the present analysis mainly affects the quantitative depth of nonclassicality because it provides Poissonian initial photon statistics. Different initial states may modify the strength of the effect and could alter the nonclassical behavior of the generated modes. Nevertheless, within the present coherent-state and short-time approximation, the observed asymmetry should be regarded mainly as an intrinsic property of the CAHRS process**.**

To further analyze the nonclassical behavior of the system, Eq. (60) has been plotted as a function of the pump photon number $|\alpha'|^2$ as shown in Fig. 4 for the parameter $|g^2t^2| \Box 10^{-5}$. Figure 4 illustrates that the photon antibunching condition $g^2(0)' < 1$ exists [10.12.27] and therefore serves as a definitive criterion to confirm the quantum nature of light. In addition, the second-order correlation function attained its minimum value in the regions where the antibunching of photons is maximum.

There is considerable evidence that, in general, higher order photon antibunching reduces the simultaneous detection of multiple photons relative to classical light source. Higher order photon antibunching extends the concept of photons antibunching to encompass both higher-order correlations and pair-wise ones. In addition, the level of nonclassical behavior exhibited by a light source can be quantitatively expressed mathematically using the correlation function or $g^2(n)$, where n denotes the correlation order and this function measures the probability of simultaneous detection of n photons at various time. Therefore, degree of nonclassical behavior exhibited by the light source as indicated by the $g^2(n)$ function indicates how photon correlations deviate from classical predictions, and thus the degree to which the light source exhibits normal and higher order photon antibunching.

The present figures also illustrate the consistency between the factorial moment criterion and the correlation-function-based criterion**.** In Fig. 1, the negativity of Eq. (30) confirms first-order antibunching in the fundamental mode, while in Fig. 4 the corresponding condition $g^2(0)' < 1$ from Eq. (60) is satisfied in the same parameter region. Thus, both criteria lead to the same qualitative conclusion for normal antibunching. However, the factorial moment criterion remains more general in the present analysis because it also describes higher-order photon antibunching through Eqs. (33) and (36), whereas $g^2(n)$ is limited to first-order antibunching. Stronger photon antibunching has been obtained that can be used in generating single photon sources by using the factorial moment criteria than the second order correlation function for the same parameters which can be seen by comparing photon antibunching from Figs. 1 and 4.

For experimentally realistic nonlinear optical systems, the effective coupling strength may typically be of the order of $10^5 - 10^7$ per second[34, 35] while the interaction time may range from $10^{-12} - 10^{-9}$ second [34, 35] so that $|gt| << 1$ remain satisfied within the validity domain of the present short time approximation. Such conditions may be realized in experimental systems such as bulk nonlinear crystals for example, beta-borium borate (BBO) and lithium niobate (LiNbO3), highly nonlinear optical fibres such as photonic crystals fibres and cavity-enhanced nonlinear optical systems like micro-ring generators or Fabry-Perot cavities where multiphoton nonlinear interactions and nonclassical photon statistics can be investigated.In the present work, only $g^2(0)'$ has been considered as the standard correlation function criteria for normal antibunching while higher order photon antibunching has been analyzed using the factorial moment criterion of Eq. (5). Higher order $g^n(0)'$ functions were not evaluated explicitly and may be considered in future work.

6 Conclusion

This study investigates antibunching of photons in the CAHRS process, examining the influence of interaction time, field strength between the modes and pump photon numbers on first-, second- and third-order antibunching effect. It is concluded that interaction duration critically governs the role in determining the achievable degree of antibunching. As interaction time decreases, the highest degree of antibunching is achieved. The number of pump photons is demonstrated to be an effective way to control the nonclassicality and it is further noted that larger pump photon numbers yield higher level of antibunching. Additionally, it is found that higher order antibunching (i.e. second and third order antibunching) have a greater nonclassicality depth than the lower order photon antibunching. However, no evidence emerges photon antibunching in stokes or signal modes relative to the pump mode in CAHRS process. The second-order correlation function is found to attain its minimum value in the regions where photon antibunching is maximum. In addition, the higher-order photons antibunching lowers the likelihood of observing multiple photons at the same time in contrast to normal light sources. Therefore, the concept of antibunching can now include higher order correlations as well as pair correlations of particles. Additionally, the depth of nonclassicality quantitatively describes how much the observed higher order photon correlations differ from what would be expected classically, thereby providing an indication of the degree of nonclassical behavior of the light source.

These results can be easily reproduced using the majority of physical systems available in lab settings. Therefore, these results open the door to the experimental verification of higher order photons antibunching and advancing probabilistic single photon sources for use in quantum teleportation and cryptography applications. Further, the ability to create larger and better nonclassical multiphotons will provide the tools need to develop new devices such as nanosensors and quantum dots that will facilitate advancements in fields of quantum information processing. An extension of the present work would be to go beyond the short-time approximation by including higher-order terms in the operator expansion. Such an analysis may provide more complete information of the long time dynamics, stronger nonlinear coupling effects and possible quantitative modifications in the depth of photon antibunching in the CAHRS process to have potential application in generating single photon source which is useful for quantum cryptography, quantum sensing and quantum imaging etc.

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