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Original article

Photon antibunching in sixth harmonic generation

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ABSTRACT We studied photon antibunching in the pump and the harmonic modes of the sixth harmonic generation process. The generalized interaction Hamiltonian is solved for several particular cases in the Heisenberg picture, and the possibility of observing photon antibunching is investigated using the short-time approximation technique. It is shown that the photon antibunching in the pump field depends on the number of pump photons, the interaction time, and the coupling of the field between the modes. With the same amount of pump photons, we deduced that third-order photon antibunching is more nonclassical than second- and first-order photon antibunching. In this process, the effect of photon antibunching is not seen in the harmonic mode over pump mode. It is shown that photon antibunching is more noticeable with shorter interaction times as the depth of nonclassicality increases and the second-order correlation function decreases. The first-order Hamiltonian interaction, which stimulates both pump and harmonic fields, is demonstrated to be more nonclassical than the second-order Hamiltonian interaction. It is clear that the coherent state, or vacuum state, of a pump field with a nonzero harmonic field creates photon clusters because the pump field causes interaction, which leads to bunching effects. It is interpreted that the degree of photon antibunching is maximum in the regions where the second correlation function is minimum. Photon antibunching has been shown to be one of the quantum properties of light.

KEYWORDS photon antibunching, higher-order antibunching, sixth harmonic generation, photon number operator, short-time approximation

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1. Introduction

Photon antibunching [1–10], a nonclassical phenomenon [11–14], is currently of considerable interest in the context of quantum teleportation [15,16], quantum computation [17–21], and quantum cryptography [22–24] by utilizing a single-photon source as unconditional security [25–28]. Single-photon sources are photon sources that emit photons as single particles or photons. This gives rise to an effective one-photon number state, which means that the chance of making one photon is higher than the chance of making two, three, four, or more photons at the same time. In the antibunched state, the rate of emitting two or more photons at the same time is lower than in the single-photon state. This means that it is more likely to find a source of a single photon than a source of two or more photons in a bunch. The second-order temporal coherence, $g^{(2)}(\tau)$, is the standard way to measure the quality of a single-photon source when the time delay is set to zero ($\tau = 0$) [26–28]. $g^{(2)}(0) = 0$ for a perfect source that never sends out more than one photon at a time. In his research, Stoler [1] introduced the concept of photon antibunching generation and outlined methods for observing it. Many authors [2–9] have previously centered their research on photon antibunching in various nonlinear optical systems. In addition, Lee created the notion of higher-order antibunching in 1990 [29, 30] when he proposed the first higher-order antibunching criterion. Ba An [31] and Pathak et al. [32] modified Lee's criterion, respectively. As stated by Gupta et al. [33] in detail, higher-order antibunching is not an unusual phenomenon; rather, it can be observed in numerous simple nonlinear optical processes, such as the six-wave mixing process, the four-wave mixing process, and the second harmonic generation. Antibunching occurs in pump modes, which lose energy, whereas bunching manifests in harmonic modes, which gain energy from the pump, and vice versa: as energy decreases, noise increases [34–36]. All the models studied here are experimentally realizable [37, 38], and the criteria for higher-order antibunching appear in terms of the factorial moment, which can be measured using homodyne photon counting experiments [37–40]. As a result, it is straightforward to experimentally validate the theoretical predictions of this study in the sixth harmonic generation process. In essence, all of the aforementioned simple physical models are readily observable and empirically realizable in any nonlinear optics laboratory. In addition, higher-order antibunching has been extensively studied and published in several optical processes [41–45] in the past few years. Dell'Anno et al. [46] gave a study of the theoretical and experimental properties of multiphoton quantum optics. In this research, they offer a consistent generalization of multiphoton events and a Hamiltonian description of higher-order nonlinear multiphoton phenomena. Combining linear and nonlinear optical devices, according to their theory, enables the realization of multiphoton nonclassical states of the electromagnetic field in discrete or continuous variables, which have applications in quantum computation, quantum teleportation, and other quantum communication and information problems. As a result, it creates new avenues for investigating higher-order nonclassical effects.

The objective of this paper is to investigate the feasibility of photon antibunching in sixth harmonic generation. The structure of the paper is as follows: The criterion of photon antibunching is defined in Section 2. In Section 3, the degenerate sixth harmonic generation model and its Hamiltonian are described. In sections 3.1 and 3.2, respectively, we establish the analytic expression of photon antibunching in the pump mode up to first- and second-order Hamiltonian interactions in the sixth harmonic generation. Photon antibunching in the harmonic mode up to first- and second-order Hamiltonian interactions are studied in sixth harmonic generation, respectively, in Sections 3.3 and 3.4. Section 4 describes the second correlation function in terms of the number of photons. Section 5 contains the results and discussion. In Section 6, we finish and summarize the paper.

2. Definition of photon antibunching

A criterion for single-mode photon antibunching, in general, proposed by Lee [29, 30], as

$$\hat{R}(l,m) = \frac{\left\langle \hat{N}_x^{(l+1)} \right\rangle \left\langle \hat{N}_x^{(m-1)} \right\rangle - \left\langle \hat{N}_x^{(l)} \right\rangle \left\langle \hat{N}_x^{(m)} \right\rangle}{\left\langle \hat{N}_x^{(l)} \right\rangle \left\langle \hat{N}_x^{(m)} \right\rangle} < 0. \tag{1}$$

where $\langle ... \rangle$ denotes the quantum average, x denotes modes of the field, l, and m are integers that satisfying the conditions $l \geq m \geq 1$ and $\hat{N}$ is the photon number operator, where $\hat{N}_x^l = \prod_{j=0}^{l-1} \left(\hat{N}_x - j \right)$ is the j^{th} factorial moment of the photon number operator.

Equation (1) reduces by using $l = m = 1$, i.e. for first-order photon antibunching [31], as

$$\left\langle \hat{N}_x^{(2)} \right\rangle < \left\langle \hat{N}_x \right\rangle^2 . \tag{2}$$

and the inequality yields

$$\left\langle \left(\Delta \hat{N}_x \right)^2 \right\rangle \equiv \left\langle \hat{N}_x^{(2)} \right\rangle - \left\langle \hat{N}_x \right\rangle^2 < \left\langle \hat{N}_x \right\rangle . \tag{3}$$

If $l \geq m = 1$ then equation (1) reduces for l^{th}-order photon antibunching for mode x, as

$$\left\langle \hat{N}_x^{(l+1)} \right\rangle < \left\langle \hat{N}_x^{(l)} \right\rangle \left\langle \hat{N}_x \right\rangle \tag{4}$$

Further, Pathak et al. [32] simplified equation (4) as

$$\left\langle \hat{N}_x^{(l+1)} \right\rangle < \left\langle \hat{N}_x^{(l)} \right\rangle \left\langle \hat{N}_x \right\rangle < \left\langle \hat{N}_x^{(l-1)} \right\rangle \left\langle \hat{N}_x \right\rangle^2 < \left\langle \hat{N}_x^{(l-2)} \right\rangle \left\langle \hat{N}_x \right\rangle^3 < \dots\dots\dots\dots\dots < \left\langle \hat{N}_x \right\rangle^{l+1} . \tag{5}$$

and the criterion for l^{th}-order photon antibunching becomes as

$$d(l) = \left[\Delta \hat{N}_x \right]^{l+1} = \left\langle \hat{N}_x^{(l+1)} \right\rangle - \left\langle \hat{N}_x \right\rangle^{l+1} < 0. \tag{6}$$

Criterion in equation (6) is precisely the property required of a probabilistic single photon source in quantum cryptography [41].

3. Degenerate sixth harmonic generation model and its Hamiltonian

The absorption of six pump photons of frequency ω_1 each interacts with a nonlinear medium in this model, as shown in Fig. 1, with the subsequent emission of one harmonic photon of frequency ω_2 with the atomic system to the ground state.

From Fig. 1, the corresponding interaction Hamiltonian is

$$\hat{H} = \hbar\omega_1 \hat{a}^\dagger \hat{a} + \hbar\omega_2 \hat{b}^\dagger \hat{b} + g \left(\hat{a}^6 \hat{b}^\dagger + \hat{a}^{\dagger 6} \hat{b} \right) . \tag{7}$$

where $\hat{a}^\dagger(\hat{a})$ and $\hat{b}^\dagger(\hat{b})$ are the creation (annihilation) operators of the pump field ($\hat{A}$-mode) and harmonic field ($\hat{B}$-mode) respectively. In the interaction Hamiltonian, g is the interaction rate between the modes. It is assumed to be real, depicting the interaction between the two modes at a rate of 10^2–10^4 per second and proportional to the nonlinear susceptibility of the medium and the complex amplitude of the pump field [47, 48].

In this interaction, due to the fact that the time dependence of free modes ($g = 0$) is proportional to exp($i\omega_1 t$) or exp($i\omega_2 t$). The interaction between modes ($g \neq 0$) causes a slower dependence on time and therefore, the field operators

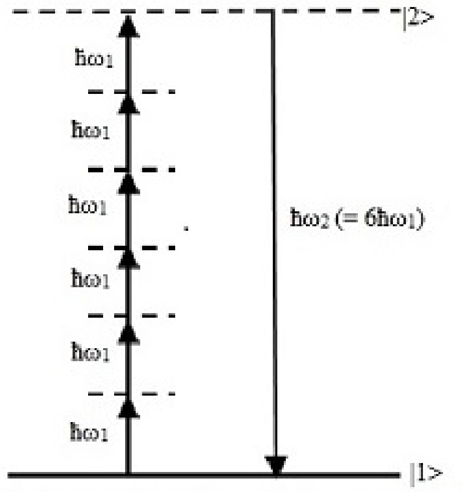

FIG. 1. Sixth harmonic generation model

may be expressed as $\hat{A} = \hat{a}\exp(i\omega_1 t)$ and $\hat{B} = \hat{b}\exp(i\omega_2 t)$ with the relation at frequency $\omega_2 = 6\omega_1$, where the operators $\hat{A}(t)$ and $\hat{B}(t)$ are vary slowly in time. Later, it will be clear that $\hat{A}(t)$ and $\hat{B}(t)$ indeed depend on a scaled time "gt" (rather than on t), thus satisfying their slow variation of the operators $\hat{A}(t)$ and $\hat{B}(t)$ as compared to fast variation of $\hat{a}(t)$ and $\hat{b}(t)$, since $g \ll \omega_1, \omega_2$ usually [47–49].

As operators represent observables, their time evolution directly corresponds to the changes in these observables over time. In order to study the characteristics of nonclassical states of light, like quadrature fluctuations, which are directly tied to observable quantities, utilise the complete Heisenberg picture, where all operators evolve based on the total Hamiltonian of the system. Thus, using equation (7) in Heisenberg equation of motion of slowing varying operator in the fundamental mode $\hat{A}$,

$$\dot{\hat{A}} = \frac{\partial \hat{A}}{\partial t} + i\left[\hat{H}, \hat{A}\right], \quad (\hbar = 1), \tag{8}$$

we obtain

$$\dot{\hat{A}} = -6ig\hat{A}^{\dagger 5}\hat{B}. \tag{9}$$

Furthermore, we have used equation (8) to calculate the second order derivative of the pump mode, as

$$\ddot{\hat{A}} = \frac{\partial \dot{\hat{A}}}{\partial t} + i\left[\hat{H}, \dot{\hat{A}}\right]. \tag{10}$$

Using equations (8) and (9), one transforms equation (10) to the form

$$\ddot{\hat{A}} = 6\,|g|^2\left[30(\hat{A}^{\dagger 4}\hat{A}^5 + 10\hat{A}^{\dagger 3}\hat{A}^4 + 40\hat{A}^{\dagger 2}\hat{A}^3 + 60\hat{A}^{\dagger}\hat{A}^2 + 24A)\hat{B}^{\dagger}\hat{B} - \hat{A}^{\dagger 5}\hat{A}^6\right]. \tag{11}$$

We use the Taylor series in the expansion of $A(t)$ for short-time ($\approx 10^{-10}$ sec), $|gt| \ll 1$ [45, 49] and keep terms up to the second order in 'gt',

$$\hat{A}(t) = \hat{A}(0) + \dot{\hat{A}}(t) + \ddot{\hat{A}}\left(\frac{t^2}{2!}\right) + \ldots\ldots\ldots\ldots \tag{12}$$

Inserting equations (9) and (11) in equation (12), we obtain

$$\hat{A}(t) = \hat{A} - 6i\,|gt|\,\hat{A}^{\dagger 5}\hat{B} + 3\,|gt|^2\left[30(\hat{A}^{\dagger 4}\hat{A}^5 + 10\hat{A}^{\dagger 3}\hat{A}^4 + 40\hat{A}^{\dagger 2}\hat{A}^3 + 60\hat{A}^{\dagger}\hat{A}^2 + 24A)\hat{B}^{\dagger}\hat{B} - \hat{A}^{\dagger 5}\hat{A}^6\right] \tag{13}$$

and in the reversal order,

$$\hat{A}^{\dagger}(t) = \hat{A}^{\dagger} + 6i\,|gt|\,\hat{A}^5\hat{B}^{\dagger} + 3\,|gt|^2\left[30(\hat{A}^{\dagger 5}\hat{A}^4 + 10\hat{A}^{\dagger 4}\hat{A}^3 + 40\hat{A}^{\dagger 3}\hat{A}^2 + 60\hat{A}^{\dagger 2}\hat{A} + 24A^{\dagger})\hat{B}^{\dagger}\hat{B} - \hat{A}^{\dagger 6}\hat{A}^5\right], \tag{14}$$

where $\hat{A}(0) = \hat{A}$ at $t = 0$.

Now, using equation (7) in Heisenberg equation of motion of the harmonic mode $\hat{B}$,

$$\dot{\hat{B}} = \frac{\partial \hat{B}}{\partial t} + i\left[\hat{H}, \hat{B}\right], \tag{15}$$

we obtain

$$\dot{\hat{B}} = -ig\hat{A}^6. \tag{16}$$

For second order derivative of $\hat{B}$ we utilize equation (16) in the Heisenberg differential equation,

$$\ddot{\hat{B}} = \frac{\partial \dot{\hat{B}}}{\partial t} + i\left[\hat{H}, \dot{\hat{B}}\right] \tag{17}$$

Then, we have,

$$\ddot{\hat{B}} = -6\,|g|^2\,[6\hat{A}^{\dagger 5}\hat{A}^5 + 75\hat{A}^{\dagger 4}\hat{A}^4 + 400\hat{A}^{\dagger 3}\hat{A}^3 + 900\hat{A}^{\dagger 2}\hat{A}^2 + 720\hat{A}^{\dagger}\hat{A} + 120]\hat{B} \tag{18}$$

We use the Taylor series expansion in the harmonic mode for short-time (10^{-10} sec) and maintain terms up to the second order in 'gt' ($|gt| \ll 1$) as

$$\hat{B}(t) = \hat{B}(0) + \dot{\hat{B}}(t) + \ddot{\hat{B}}\left(\frac{t^2}{2!}\right) + ... \tag{19}$$

Using equations (16) and (18) in equation (19), we get

$$\hat{B}(t) = \hat{B} - i\,|gt|\,\hat{A}^6 - 3\,|gt|^2\,[6\hat{A}^{\dagger 5}\hat{A}^5 + 75\hat{A}^{\dagger 4}\hat{A}^4 + 400\hat{A}^{\dagger 3}\hat{A}^3 + 900\hat{A}^{\dagger 2}\hat{A}^2 + 720\hat{A}^{\dagger}\hat{A} + 120]\hat{B} \tag{20}$$

and its Hermitian conjugate

$$\hat{B}^{\dagger}(t) = \hat{B}^{\dagger} + i\,|gt|\,\hat{A}^{\dagger 6} - 3\,|gt|^2\,[6\hat{A}^{\dagger 5}\hat{A}^5 + 75\hat{A}^{\dagger 4}\hat{A}^4 + 400\hat{A}^{\dagger 3}\hat{A}^3 + 900\hat{A}^{\dagger 2}\hat{A}^2 + 720\hat{A}^{\dagger}\hat{A} + 120]\hat{B}^{\dagger} \tag{21}$$

3.1. Photon antibunching in the pump field up to first-order Hamiltonian interaction

Now, we assume an initial quantum state as a product of coherent states $|\alpha\rangle$ for the pump mode $\hat{A}$ and $|\beta\rangle$ for the harmonic mode $\hat{B}$, i.e.

$$|\psi\rangle = |\alpha\rangle_A\,|\beta\rangle_B \tag{22}$$

Using the condition of equation (22) in equation (13) and (14), we arrive at

$$\langle\psi|\,\hat{A}(t)\,|\psi\rangle = \langle\psi|\left(\hat{A} - 6i\,|gt|\,\hat{A}^{\dagger 5}\hat{B}\right)|\psi\rangle \tag{23}$$

and

$$\langle\psi|\,\hat{A}^{\dagger}(t)\,|\psi\rangle = \langle\psi|\left(\hat{A}^{\dagger} + 6i\,|gt|\,\hat{A}^5\hat{B}^{\dagger}\right)|\psi\rangle \tag{24}$$

Using equation (23) and (24), the expectation values of the photon number operator $\hat{N}_A(t)$ can be written as

$$\begin{aligned}&\langle\psi|\,\hat{N}_A(t)\,|\psi\rangle = \langle\psi|\,\hat{A}^{\dagger}(t)\hat{A}(t)\,|\psi\rangle = \langle\psi|\left(\hat{A}^{\dagger}\hat{A} - 6i\,|gt|\,(\hat{A}^{\dagger 6}\hat{B} - \hat{A}^6\hat{B}^{\dagger})\right)|\psi\rangle \\ &= |\alpha|^2 - 6i\,|gt|\left(\alpha^{*6}\beta - \alpha^6\beta^*\right)\end{aligned} \tag{25}$$

where $\langle N_A\rangle = \left\langle A^{\dagger}A\right\rangle = |\alpha|^2$ and $\langle N_B\rangle = \left\langle B^{\dagger}B\right\rangle = |\beta|^2$ are the photon number operators in A and B respectively and $\alpha = |\alpha|\exp(i\theta_1), \alpha^* = |\alpha^*|\exp(-i\theta_1), \beta = |\beta|\exp(i\theta_2)$ and $\beta^* = |\beta^*|\exp(-i\theta_2)$; θ_1 and θ_2 are the phase angles; α^*, β^* denote the complex conjugate of α and β, respectively.

Furthermore, the expectation values of the second factorial moment and the squared of the expectation values of the photon number operator $\hat{N}_A(t)$ of equation (25) can be represented respectively, as

$$\langle\psi|\left\langle \hat{N}_A^{(2)}(t)\right\rangle|\psi\rangle = \langle\psi|\,\hat{A}^{\dagger 2}(t)\hat{A}^2(t)\,|\psi\rangle = |\alpha|^4 + 12i\,|gt|\left(\alpha^*\alpha^7\beta^* - \alpha^{*7}\alpha\beta\right) + 30i\,|gt|\left(\alpha^6\beta^* - \alpha^{*6}\beta\right) \tag{26}$$

and

$$\langle\psi|\left\langle \hat{N}_A(t)\right\rangle^2|\psi\rangle = \langle\psi|\,\hat{A}^{\dagger}(t)\hat{A}(t)\,|\psi\rangle^2 = |\alpha|^4 + 12i\,|gt|\left(\alpha^*\alpha^7\beta^* - \alpha^{*7}\alpha\beta\right) \tag{27}$$

Therefore, for $l = 1$, the variation of photon number operator between equations (26) and (27) is

$$\begin{aligned}d_A(1) &= \left[\langle\psi|\,\Delta\hat{N}_A(t)\,|\psi\rangle\right]^2 = \langle\psi|\,\hat{N}_A^{(2)}(t)\,|\psi\rangle - \langle\psi|\,\hat{N}_A(t)\,|\psi\rangle^2 \\ &= 30i\,|gt|\left(\alpha^6\beta^* - \alpha^{*6}\beta\right) = -60\,|gt|\left|\alpha^6\beta\right|\{\sin(6\theta_1 - \theta_2)\}\end{aligned} \tag{28}$$

For optimal photon antibunching, let us take $\theta_1 = \dfrac{\pi}{12}$ and $\theta_2 = 0$, then equation (28) reduces to

$$d_A(1) = -60\,|gt|\left|\alpha^6\beta\right| \tag{29}$$

The right-hand side of equation (29) is negative and satisfies the criterion (6), indicating that first-order photon antibunching exists in the pump mode of the sixth harmonic generation.

Similarly, we use the expectation values of the third factorial moment of the photon number operator $\hat{N}_A(t)$ to investigate one of the classes of higher-order antibunching as a function of time, such as second-order antibunching for $l = 2$ of the fundamental mode,

$$\begin{aligned}&\langle\psi|\,\hat{N}_A^{(3)}(t)\,|\psi\rangle = |\alpha|^6 + 18i\,|gt|\,|\alpha|^4\,\alpha^6\beta^* + 90i\,|gt|\,|\alpha|^2\,\alpha^6\beta^* + 120i\,|gt|\,\alpha^6\beta^* \\ &-18i\,|gt|\,|\alpha|^4\,\alpha^{*6}\beta - 90i\,|gt|\,|\alpha|^2\,\alpha^{*6}\beta - 120i\,|gt|\,\alpha^{*6}\beta\end{aligned} \tag{30}$$

and cubed of the expectation values of the photon number operator $\hat{N}_A(t)$ is

$$\langle\psi| \hat{N}_A(t) |\psi\rangle^3 = |\alpha|^6 + 18i\,|gt|\,|\alpha|^4\,\alpha^6\beta^* - 18i\,|gt|\,|\alpha|^4\,\alpha^{*6}\beta \tag{31}$$

Hence, the variance in photon number is

$$\begin{aligned} d_A(2) &= \langle\psi| \hat{N}_A^{(3)}(t) |\psi\rangle - \langle\psi| \hat{N}_A(t) |\psi\rangle^3 = 30i\,|gt|\left(\alpha^6\beta^* - \alpha^{*6}\beta\right)\left(3\,|\alpha|^2 + 4\right) \\ &= -60\,|gt|\,\left|\alpha^6\beta\right|\{\sin(6\theta_1 - \theta_2)\}\left(3\,|\alpha|^2 + 4\right) \end{aligned} \tag{32}$$

Using $\theta_1 = \dfrac{\pi}{12}$ and $\theta_2 = 0$ in equation (32) then the maximum variance in photon number is

$$d_A(2) = -60\,|gt|\,\left|\alpha^6\beta\right|\left(3\,|\alpha|^2 + 4\right) \tag{33}$$

Equation (33) satisfies equation (6) and is always negative, suggesting that the pump mode of the sixth harmonic generation exhibits second-order photon antibunching.

Subsequently, the expectation values of fourth factorial moment of the photon number operator $\hat{N}_A(t)$ is

$$\begin{aligned} \langle\psi| \hat{N}_A^{(4)}(t) |\psi\rangle &= |\alpha|^8 + 24i\,|gt|\,|\alpha|^6\,\alpha^6\beta^* + 180i\,|gt|\,|\alpha|^4\,\alpha^6\beta^* + 480i\,|gt|\,|\alpha|^2\,\alpha^6\beta^* + 360i\,|gt|\,\alpha^6\beta^* \\ &-24i\,|gt|\,|\alpha|^6\,\alpha^{*6}\beta - 180i\,|gt|\,|\alpha|^4\,\alpha^{*6}\beta - 480i\,|gt|\,|\alpha|^2\,\alpha^{*6}\beta - 360i\,|gt|\,\alpha^{*6}\beta \end{aligned} \tag{34}$$

and the fourth power of the expectation values of the photon number operator $\hat{N}_{\hat{A}}(t)$ is

$$\langle\psi| \hat{N}_A(t) |\psi\rangle^4 = |\alpha|^8 + 24i\,|gt|\,|\alpha|^6\,\alpha^6\beta^* - 24i\,|gt|\,|\alpha|^6\,\alpha^{*6}\beta \tag{35}$$

For $l = 3$, the variance in photon number can be calculated as,

$$\begin{aligned} d_A(3) &= \langle\psi| \hat{N}_A^{(4)}(t) |\psi\rangle - \langle\psi| \hat{N}_A(t) |\psi\rangle^4 = 60i\,|gt|\left(\alpha^6\beta^* - \alpha^{*6}\beta\right)\left(3\,|\alpha|^4 + 8\,|\alpha|^2 + 6\right) \\ &= -120\,|gt|\,\left|\alpha^6\beta\right|\{\sin(6\theta_1 - \theta_2)\}\left(3\,|\alpha|^4 + 8\,|\alpha|^2 + 6\right) \end{aligned} \tag{36}$$

Substituting $\theta_1 = \dfrac{\pi}{12}$ and $\theta_2 = 0$ for getting optimal effect of photon antibunching in equation (36) yields:

$$d_A(3) = -120\,|gt|\,\left|\alpha^6\beta\right|\left(3\,|\alpha|^4 + 8\,|\alpha|^2 + 6\right) \tag{37}$$

Equation (37) satisfies the requirements of equation (6) and verifies the existence of the third-order photon antibunching in the pump mode of the sixth harmonic generation.

3.2. Photon antibunching in the pump field up to second-order Hamiltonian interaction

Now, we assume an initial quantum state as a product of coherent states $|\alpha\rangle$ for the pump mode $\hat{A}$ and $|0\rangle$ for the harmonic mode $\hat{B}$, i.e.

$$|\psi\rangle = |\alpha\rangle_A\,|0\rangle_B \tag{38}$$

Using the condition of equation (38) in equation (13) and (14), we obtain

$$\langle\psi| \hat{A}(t) |\psi\rangle = \langle\psi| \left(\hat{A} - 3\,|gt|^2\,\hat{A}^{\dagger 5}\hat{A}^6\right) |\psi\rangle \tag{39}$$

and

$$\langle\psi| \hat{A}^\dagger(t) |\psi\rangle = \langle\psi| \left(\hat{A}^\dagger - 3\,|gt|^2\,\hat{A}^{\dagger 6}\hat{A}^5\right) |\psi\rangle \tag{40}$$

Using equations (39) and (40), the expectation values of the photon number operator $\hat{N}_A(t)$ can be expressed as

$$\langle\psi| \hat{N}_A(t) |\psi\rangle = \langle\psi| \hat{A}^\dagger(t)\hat{A}(t) |\psi\rangle = \langle\psi| \left(\hat{A}^\dagger\hat{A} - 6\,|gt|^2\,(\hat{A}^{\dagger 6}\hat{A}^6)\right) |\psi\rangle = |\alpha|^2 - 6\,|gt|^2\,|\alpha|^{12} \tag{41}$$

Furthermore, the expectation values of the second factorial moment and the squared of the expectation values of the photon number operator $\hat{N}_A(t)$ of equation (41) can be represented respectively, as

$$\langle\psi| \left\langle \hat{N}_A^{(2)}(t) \right\rangle |\psi\rangle = \langle\psi| \hat{A}^{\dagger 2}(t)\hat{A}^2(t) |\psi\rangle = |\alpha|^4 - 12\,|gt|^2\,|\alpha|^{14} - 30\,|gt|^2\,|\alpha|^{12} \tag{42}$$

and

$$\langle\psi| \left\langle \hat{N}_A(t) \right\rangle^2 |\psi\rangle = \langle\psi| \hat{A}^\dagger(t)\hat{A}(t) |\psi\rangle^2 = |\alpha|^4 - 12\,|gt|^2\,|\alpha|^{14} \tag{43}$$

Hence, the fluctuation of photon number is

$$d'_A(1) = \left[\langle\psi| \Delta\hat{N}_A(t) |\psi\rangle\right]^2 = \langle\psi| \hat{N}_A^{(2)}(t) |\psi\rangle - \langle\psi| \hat{N}_A(t) |\psi\rangle^2 = -30\,|gt|^2\,|\alpha|^{12} \tag{44}$$

The fact that the right-hand side of equation (44) is negative and meets the condition of equation (6) indicates that first-order photon antibunching exists in the pump mode of the sixth harmonic generation.

Similarly, we use the expectation values of the third factorial moment of the photon number operator $\hat{N}_A(t)$ to analyze one of the classes of higher-order antibunching, such as second-order antibunching for $l = 2$ of the fundamental/pump mode as a function of time,

$$\langle\psi| \hat{N}_A^{(3)}(t) |\psi\rangle = |\alpha|^6 - 18 |gt|^2 |\alpha|^{16} - 90 |gt|^2 |\alpha|^{14} - 120 |gt|^2 |\alpha|^{12} \tag{45}$$

and cubed of the expectation values of the photon number operator $\hat{N}_A(t)$ is

$$\langle\psi| \hat{N}_A(t) |\psi\rangle^3 = |\alpha|^6 - 18 |gt|^2 |\alpha|^{16} \tag{46}$$

Hence, the variance in photon number is

$$d'_A(2) = \langle\psi| \hat{N}_A^{(3)}(t) |\psi\rangle - \langle\psi| \hat{N}_A(t) |\psi\rangle^3 = -30 |gt|^2 |\alpha|^{12} \left(3 |\alpha|^2 + 4\right) \tag{47}$$

Equation (47) is always negative and meets the condition of equation (6), suggesting that the second-order photon antibunching occurs in the pump mode of the sixth harmonic generation.

Subsequently, the expectation values of the fourth factorial moment of the photon number operator $\hat{N}_A(t)$ is

$$\langle\psi| \hat{N}_A^{(4)}(t) |\psi\rangle = |\alpha|^8 - 24 |gt|^2 |\alpha|^{18} - 180 |gt|^2 |\alpha|^{16} - 480 |gt|^2 |\alpha|^{14} - 360 |gt|^2 |\alpha|^{12} \tag{48}$$

and the fourth power of the expectation values of the photon number operator $\hat{N}_A(t)$ is

$$\langle\psi| \hat{N}_A(t) |\psi\rangle^4 = |\alpha|^8 - 24 |gt|^2 |\alpha|^{18} \tag{49}$$

As a result, for $l = 3$, the variance in photon number for third-order antibunching can be calculated as,

$$d'_A(3) = \langle\psi| \hat{N}_A^{(4)}(t) |\psi\rangle - \langle\psi| \hat{N}_A(t) |\psi\rangle^4 = -60 |gt|^2 |\alpha|^{12} \left(3 |\alpha|^4 + 8 |\alpha|^2 + 6\right) \tag{50}$$

The criterion of equation (6) is satisfied by equation (50), indicating the presence of the third-order photon antibunching in the pump mode of the sixth harmonic generation.

3.3. Photon antibunching in the harmonic field up to first-order Hamiltonian interaction

Now, we use the condition of equation (22) in equations (20) and (21) for the photon number in the harmonic mode as

$$\hat{N}_B(t) = \hat{B}^\dagger(t)\hat{B}(t) = \left(\hat{B}^\dagger + igt\hat{A}^{\dagger 6}\right) \left(\hat{B} - igt\hat{A}^6\right) \tag{51}$$

and

$$\langle\psi| \hat{N}_B(t) |\psi\rangle_\alpha = \langle\psi| \hat{B}^\dagger(t)\hat{B}(t) |\psi\rangle_\alpha = \langle\psi| \left(\hat{B}^\dagger\hat{B} + igt(\hat{A}^{\dagger 6}\hat{B} - \hat{A}^6\hat{B}^\dagger)\right) |\psi\rangle_\alpha = |\beta|^2 + igt \left(\alpha^{*6}\beta - \alpha^6\beta^*\right) \tag{52}$$

In addition, the expectation values of the second factorial moment and the squared of the expectation values of the photon number operator $\hat{N}_B(t)$ of equation (52) can be represented respectively, as

$$\langle\psi| \left\langle \hat{N}_B^{(2)}(t) \right\rangle |\psi\rangle_\alpha = \langle\psi| \hat{B}^{\dagger 2}(t)\hat{B}^2(t) |\psi\rangle_\alpha = |\beta|^4 + 2igt |\beta|^2 \left(\alpha^{*6}\beta - \alpha^6\beta^*\right) \tag{53}$$

and

$$\langle\psi| \left\langle \hat{N}_B(t) \right\rangle^2 |\psi\rangle_\alpha = \langle\psi| \hat{B}^\dagger(t)\hat{B}(t) |\psi\rangle_\alpha^2 = |\beta|^4 + 2igt |\beta|^2 \left(\alpha^{*6}\beta - \alpha^6\beta^*\right) \tag{54}$$

As a result of the first-order antibunching, the fluctuation of photon number in the harmonic mode over pump mode is

$$d_{B\alpha}(1) = \langle\psi| \hat{N}_B^{(2)}(t) |\psi\rangle_\alpha - \langle\psi| \hat{N}_B(t) |\psi\rangle_\alpha^2 = 0 \tag{55}$$

Similarly, the result of the second-order antibunching in the harmonic mode over pump mode is as follows

$$d_{B\alpha}(2) = \langle\psi| \hat{N}_B^{(3)}(t) |\psi\rangle_\alpha - \langle\psi| \hat{N}_B(t) |\psi\rangle_\alpha^3 = 0 \tag{56}$$

where the expectation values of the third factorial moment and cubed of the expectation values of the photon number operator in the harmonic mode $\hat{N}_{\hat{B}}(t)$ are

$$\langle\psi| \hat{N}_B^{(3)}(t) |\psi\rangle_\alpha = |\beta|^6 + 3igt |\beta|^4 \left(\alpha^{*6}\beta - \alpha^6\beta^*\right), \tag{57}$$

and

$$\langle\psi| \hat{N}_B(t) |\psi\rangle_\alpha^3 = |\beta|^6 + 3igt |\beta|^4 \left(\alpha^{*6}\beta - \alpha^6\beta^*\right) \tag{58}$$

For equations (55) and (56) there are no normal and the higher-order photon antibunching in the harmonic mode over pump mode in the sixth harmonic generation up to first-order Hamiltonian interaction. In contrast, the coherent state of a pump field with a nonzero harmonic field generates photon clusters. This is due to the fact that the coherent state of the pump field causes interaction, which results in the bunching effects.

3.4. Photon antibunching in the harmonic field up to second-order Hamiltonian interaction

Now, if we assume an initial quantum state as a product of vacuum state $|0\rangle$ for the pump mode $\hat{A}$ and $|\beta\rangle$ for the harmonic mode $\hat{B}$, i.e.

$$|\psi\rangle_\beta = |0\rangle_A |\beta\rangle_B \tag{59}$$

Using condition of equation (59) in equations (20) and (21) leads to the following result

$$\hat{N}_B(t) = \hat{B}^\dagger(t)\hat{B}(t) = \left(\hat{B}^\dagger - 360\,|gt|^2\,\hat{B}^\dagger\right)\left(\hat{B} - 360\,|gt|^2\,\hat{B}\right) \tag{60}$$

The expectation value of equation (60) is as follows

$$\langle\psi|\,\hat{N}_B(t)\,|\psi\rangle_\beta = \langle\psi|\,\hat{B}^\dagger(t)\hat{B}(t)\,|\psi\rangle_\beta = |\beta|^2\left(1 - 720\,|gt|^2\right) \tag{61}$$

In addition, the expectation values of the second factorial moment and the squared of the expectation values of the photon number operator $\hat{N}_B(t)$ of equation (61) can be represented, respectively, as

$$\langle\psi|\left\langle \hat{N}_B^{(2)}(t)\right\rangle|\psi\rangle_\beta = \langle\psi|\,\hat{B}^{\dagger 2}(t)\hat{B}^2(t)\,|\psi\rangle_\beta = |\beta|^4\left(1 - 1440\,|gt|^2\right) \tag{62}$$

and

$$\langle\psi|\left\langle \hat{N}_B(t)\right\rangle^2|\psi\rangle_\beta = \langle\psi|\,\hat{B}^\dagger(t)\hat{B}(t)\,|\psi\rangle_\beta^2 = |\beta|^4\left(1 - 1440\,|gt|^2\right) \tag{63}$$

As a result, the variance in photon number i.e. first-order antibunching in the harmonic mode over pump mode is

$$d_{B\beta}(1) = \langle\psi|\,\hat{N}_B^{(2)}(t)\,|\psi\rangle_\beta - \langle\psi|\,\hat{N}_B(t)\,|\psi\rangle_\beta^2 = 0 \tag{64}$$

Similarly, the result of the second-order antibunching in the harmonic mode over pump mode is as follows

$$d_{B\beta}(2) = \langle\psi|\,\hat{N}_B^{(3)}(t)\,|\psi\rangle_\beta - \langle\psi|\,\hat{N}_B(t)\,|\psi\rangle_\beta^3 = 0 \tag{65}$$

where the expectation values of the third factorial moment and cubed of the expectation values of the photon number operator $\hat{N}_B(t)$ are

$$\langle\psi|\,\hat{N}_B^{(3)}(t)\,|\psi\rangle_\beta = |\beta|^6\left(1 - 2160\,|gt|^2\right), \tag{66}$$

and

$$\langle\psi|\,\hat{N}_B(t)\,|\psi\rangle_\beta^3 = |\beta|^6\left(1 - 2160\,|gt|^2\right) \tag{67}$$

Equations (64) and (65) do not meet the criteria of equation (6); hence, there is no existence of normal and higher-order photon antibunching in the harmonic mode over pump mode in the sixth harmonic generation up to second-order Hamiltonian interaction. On the other hand, the vacuum state of a pump field with a nonzero harmonic field, results in the photon bunching. This is due to the fact that the vacuum state of the pump field stimulates interaction, producing bunching effects.

4. The photon number correlation $g^2(0)$

Now, consider the second-order correlation function for zero-time delay $g^2(0)$ in terms of photon number. The expression is as follows [10, 12, 27]:

$$g^2(0) = 1 + \frac{\left\langle (\Delta n)^2\right\rangle - \langle n\rangle}{\langle n\rangle^2} \tag{68}$$

Substituting the values of equations (25), (27), and (28) with $\theta_1 = \dfrac{\pi}{12}$ and $\theta_2 = 0$ in equation (68) yields the second-order correlation function or photon number correlation up to the first-order Hamiltonian interaction i.e., dimensionless interaction constant gt, as

$$g^2(0) = 1 + \frac{-60\,|gt|\,\left|\alpha^6\beta\right| - \left(|\alpha|^2 - 12\,|gt|\,\left|\alpha^6\beta\right|\right)}{|\alpha|^4 - 24\,|gt|\,|\alpha|^2\,\left|\alpha^6\beta\right|} \tag{69}$$

Similarly, using equations (41), (43), and (44), in equation (68), we have the second-order correlation function or photon number correlation up to the second-order Hamiltonian interaction i.e., dimensionless interaction constant $|gt|^2$, as

$$g^2(0)' = 1 + \frac{-30\,|gt|^2\,|\alpha|^{12} - \left(|\alpha|^2 - 6\,|gt|^2\,|\alpha|^{12}\right)}{|\alpha|^4 - 12\,|gt|^2\,|\alpha|^{14}} \tag{70}$$

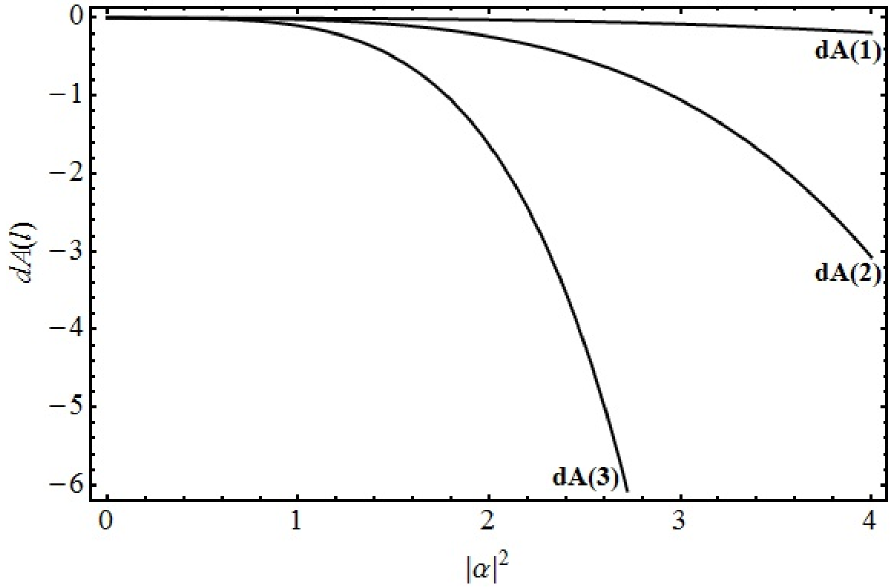

FIG. 2. Degree of photon antibunching $d_A(l)$ with $|\alpha|^2$ of the pump field up to first-order Hamiltonian interaction in sixth harmonic generation (when $|\beta| = 5$ (arbitrarily value) and $|gt| = 10^{-5}$)

5. Results and discussion

We plot a graph (Fig. 2) between the left-hand side of equations (29), (33) and (37) say $d_A(l)$vs. $|\alpha|^2$ having $|gt| = 10^{-5}$ and $|\beta| = 5$ (any arbitrarily value).

In Fig. 2, the steady fall of the curve shows that the effect of photon antibunching increases nonlinearly with the increase in the number of pump photons $|\alpha|^2$.

This confirms that the photon antibunched states are associated with a large number of pump photons. When the plots in Fig. 2 are compared, we see that the third-order photon antibunching $d_A(3)$ is more nonclassical than the second-order $d_A(2)$ and the first-order $d_A(1)$ photon antibunching with the same number of pump photons. Hence, we inferred that higher-order (third- and second-order) photon antibunching makes it possible to achieve a significantly larger noise reduction than ordinary or the first-order photon antibunching.

Fig. 3,4,5 show the variations of $d_A(1)$, $d_A(2)$, and $d_A(3)$ with $|\alpha|^2$ of equations (29), (33) and (37), respectively, for various harmonic mode $|\beta|$ values.

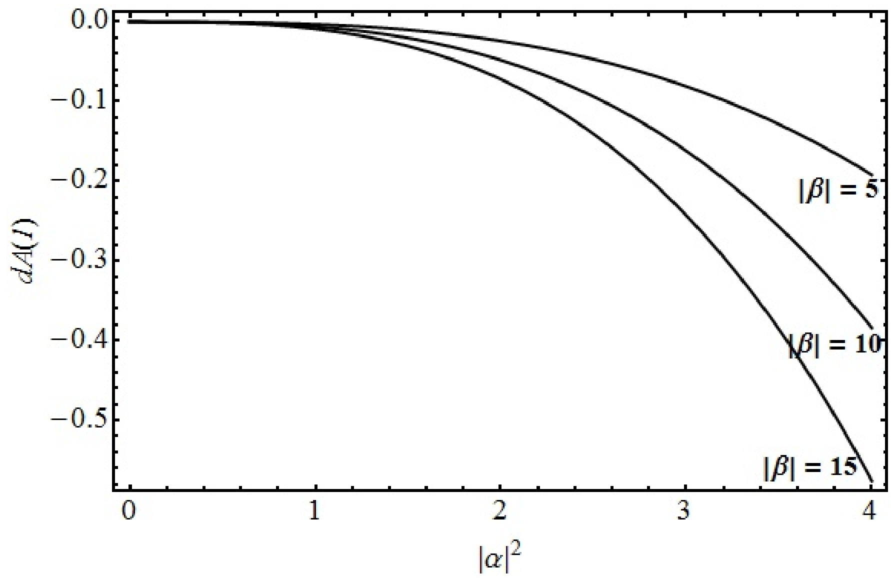

FIG. 3. First-order photon antibunching $d_A(1)$ with $|\alpha|^2$ in the pump field up to first-order Hamiltonian interaction of sixth harmonic generation (when $|\beta| = 5, 10, 15$ and $|gt| = 10^{-5}$)

The steady fall of plots in Fig. 3,4,5 show that degree of photon antibunching occurs in the sixth harmonic generation and responds nonlinearly to the number of pump photons. It demonstrates that when the photon number in pump mode $|\alpha|^2$ is increasing, the degree of photon antibunching $d_A(l)$ increases. In a comparison among Fig. 3,4,5, it is observed that higher the value of $|\beta|$, the effect of photon antibunching increases and lowers the depth of classicality of the field amplitude. It is also shown that the photon antibunching directly depends on the photon number of the fundamental mode as well as the harmonic mode.

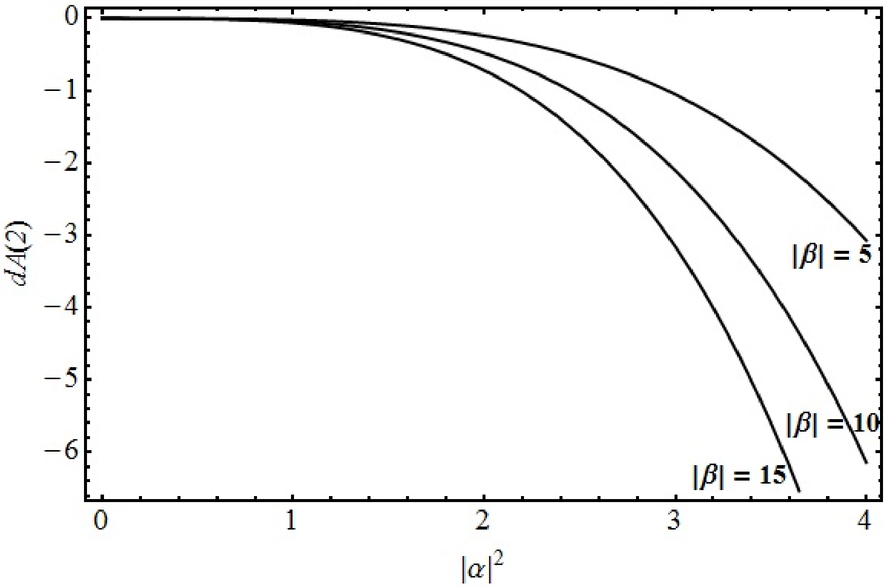

FIG. 4. Second-order photon antibunching $d_A(2)$ with $|\alpha|^2$ in the pump field up to first-order Hamiltonian interaction of sixth harmonic generation (when $|\beta| = 5, 10, 15$ and $|gt| = 10^{-5}$

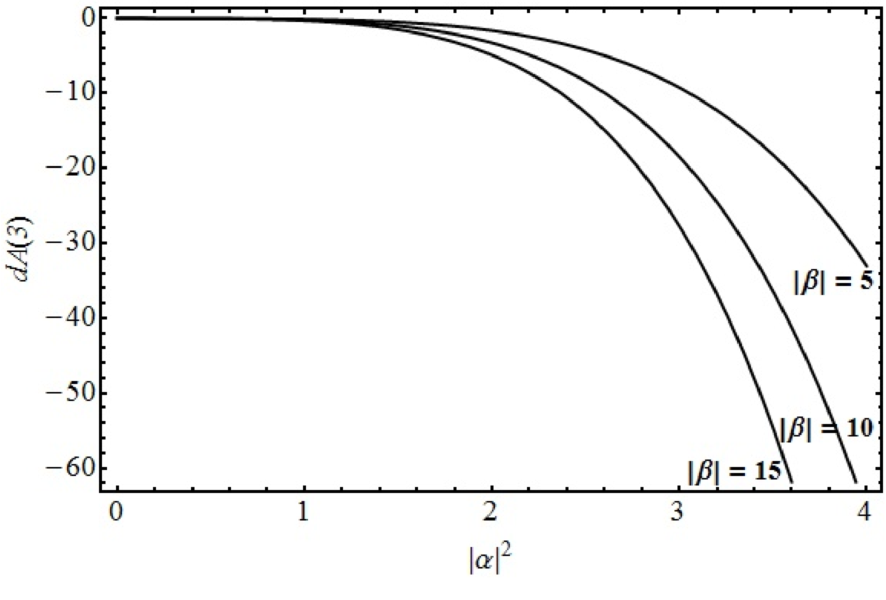

FIG. 5. Third-order photon antibunching $d_A(3)$ with $|\alpha|^2$ in the pump field up to first-order Hamiltonian interaction of sixth harmonic generation (when $|\beta| = 5, 10, 15$ and $|gt| = 10^{-5}$)

The variations of $d_A(l)$ and $|gt|$ of equations (29), (33) and (37) are shown in Fig. 6. The curves show that the degree of photon antibunching increases and directly depends upon the coupling of the field and the interaction time of the pump field. Fig. 7 depict the variation of $d_A(l)$ and $|\alpha|^2$ of equation (37) with different values of $|gt|$, demonstrating that the maximum reachable degree of antibunching is dependent on interaction time and is limited by a short interaction time. It is also worth noting that as the interaction time becomes shorter, the effect of photon antibunching increases [50–52].

We further plot a graph (Fig. 8) between the left-hand side of equations (44), (47) and (50) say $d'_A(l)$ and $|\alpha|^2$, when dimensionless interaction constant, $|gt|^2 = 10^{-10}$. The steady fall of the curves infer that the photon antibunching exists

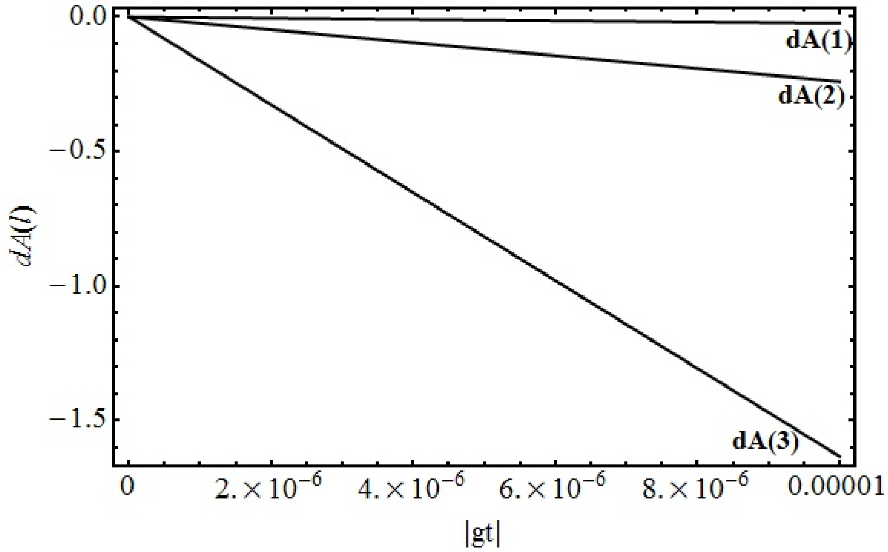

FIG. 6. Degree of photon antibunching $d_A(l)$ with $|gt|$ in the pump field up to first-order Hamiltonian interaction of sixth harmonic generation (when $|\alpha| = 2$ and $|\beta| = 5$ have arbitrarily value)

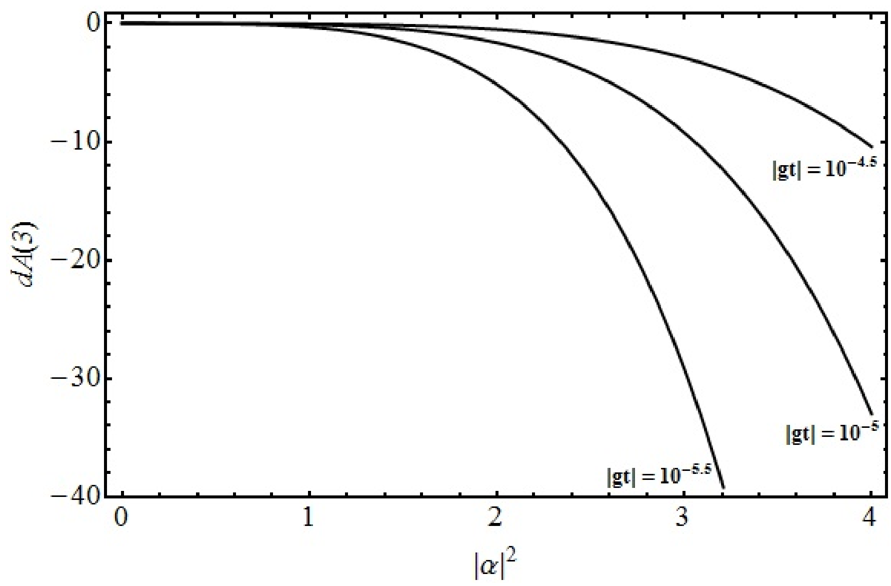

FIG. 7. Third-order photon antibunching $d_A(3)$ with $|\alpha|^2$ in the pump field up to first-order Hamiltonian interaction of sixth harmonic generation (when $|\beta| = 5$ (arbitrarily value) and $|gt| = 10^{-4.5}, 10^{-5}, 10^{-5.5}$)

in the sixth harmonic generation and responds nonlinearly to the number of pump photons. It demonstrates that when $|\alpha|^2$ is increasing the degree of photon antibunching increases. When the plots in Fig. 8 are compared, the third-order plot $d'_A(3)$ shows more photon antibunching than the second-order $d'_A(2)$ and the first-order $d'_A(1)$ plots, despite the fact that the number of photons is the same. It implies that the third-order photon antibunching is more nonclassical than the first- and the second-order photon antibunching.

The variations of $d'_A(l)$ and $|gt|^2$ of equations (44), (47) and (50) are shown in Fig. 9. The curves show that the degree of photon antibunching increases nonlinearly and directly depends upon the coupling of the field and the interaction time of the pump field. It also infers that the third-order photon antibunching is more nonclassical at shorter-time than the first- and the second-order photon antibunching.

It also shows that the maximum reachable degree of antibunching depends upon the interaction time and will be limited by short interaction time. It is worth noting that as the interaction time becomes shorter, the effect of photon antibunching increases [50–52].

Hence, it is inferred that the depth of nonclassicality is increasing with an increase of $|\alpha|^2$ and the maximum reachable degree of photon antibunching is dependent upon the interaction time.

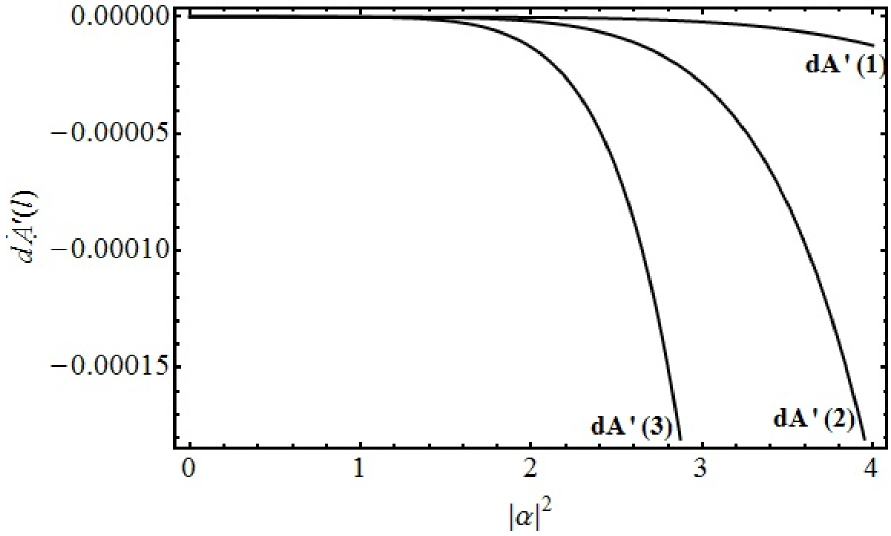

FIG. 8. Degree of photon antibunching $d'_A(l)$ with $|\alpha|^2$ in the pump field up to second-order Hamiltonian interaction of sixth harmonic generation (when $|gt|^2 = 10^{-10}$)

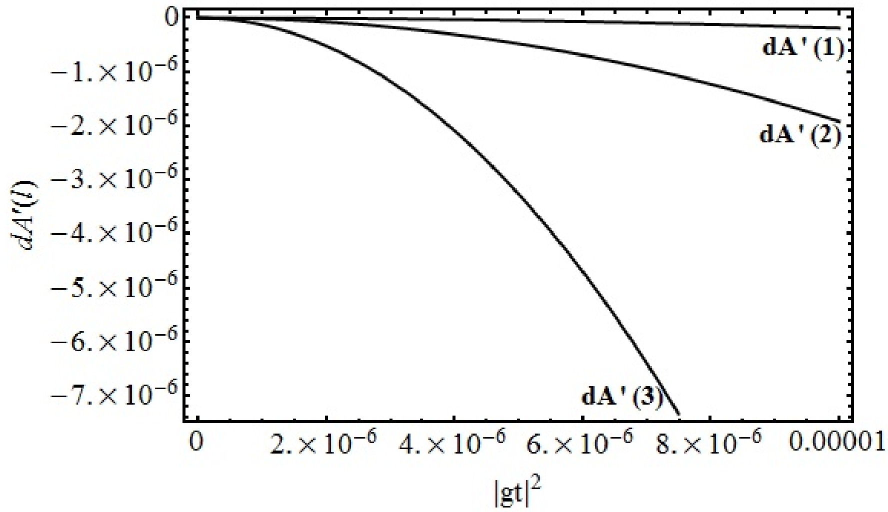

FIG. 9. Degree of photon antibunching $d'_A(l)$ with $|gt|^2$ in the pump field up to second-order Hamiltonian interaction of sixth harmonic generation (when $|\alpha|^2 = 2$ (arbitrarily value))

By comparing figures (2) and (8), it is clear that the first-order Hamiltonian interaction is more nonclassical than the second-order Hamiltonian interaction since it stimulates both pump and harmonic fields. It is observed that a powerful pump field induces a stronger interaction and is helpful for obtaining stronger photon antibunching.

It is found that, from sections 3.3 and 3.4, there is no evidence of photon antibunching in the harmonic mode over pump mode up to first- and second-order Hamiltonian interaction in the sixth harmonic generation process.

In contrast, the coherent state of a pump field with a nonzero harmonic field generates photon clusters. This is due to the fact that the coherent state of the pump field causes interaction, which results in bunching effects. On the other hand, the vacuum state of a pump field with a nonzero harmonic field, results in photon bunching. This is due to the fact that the vacuum state of the pump field stimulates interaction, producing bunching effects.

In order to discuss the depth of nonclassicality, we consider equations (69) and (70) in terms of pump photon number and plot a graph (Fig. 10) between $g^2(0)$ and $g^2(0)'$ versus $|\alpha|^2$ having when $|\beta| = 5$ (any arbitrarily constant value) and $|gt| = 10^{-5}$.

Fig. 10 shows that the condition of antibunching $g^2(0) < 1$ exists [10, 12, 27] and is, thus, a conclusive signature of the quantum nature of light. We also infer that the fall of plots of the second-order correlation function for zero time delay or the photon number correlation decreases, i.e., the depth of nonclassicality increases with the increase of photons in the sixth harmonic generation.

Fig. 10 between $\left(g^2(0)\right)'$ and $|\alpha|^2$ of the pump field up to the second-order Hamiltonian interaction in sixth harmonic generation (when $|gt|^2 = 10^{-10}$) reveals that the second-order correlation function for zero time delay or the photon number correlation decreases, i.e., the depth of nonclassicality increases, and hence it is more nonclassical in a shorter interaction time. We see that the second correlation function is minimum in regions where the degree of photon antibunching is maximum.

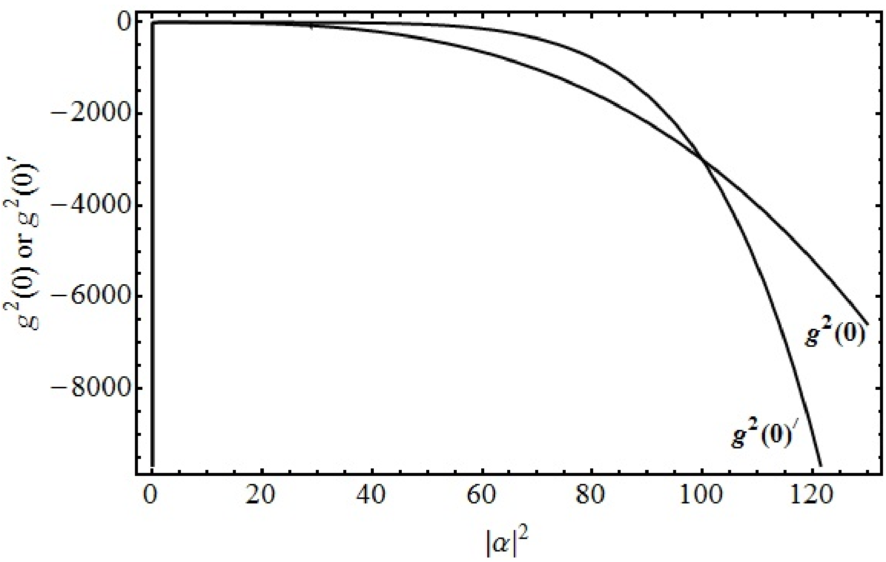

FIG. 10. Second-order correlation function $g^2(0)$ or $g^2(0)'$ versus $|\alpha|^2$ in the pump field up to first-order or second-order Hamiltonian interaction of sixth harmonic generation respectively (when $|\beta| = 5$ (arbitrarily value) and $|gt|^2 = 10^{-10}$)

It is evident that, in general, higher-order antibunching is a phenomenon that reduces the probability of detecting multiple photons simultaneously compared to a classical light source. This effect extends the concept of antibunching beyond pairs of photons to encompass higher-order correlations. Mathematically, the depth of non-classicality in higher-order antibunching can be defined using correlation functions, or $g^2\,(n)$ functions, where n represents the order of the correlation. These functions measure the probability of detecting n photons simultaneously at different times. The depth of non-classicality provides a quantitative measure of the extent to which the observed higher-order photon correlations deviate from classical predictions, indicating the degree of non-classical behaviour exhibited by the light source beyond first-order (normal) antibunching effects.

6. Summary and conclusions

The present manuscript investigates photon antibunching in the sixth harmonic generation. We demonstrate that the number of pump photons, the field coupling between the modes, and the interaction time closely influence the first-, second-, and third-order photon antibunching. Therefore, the interaction time, limited by short interaction times, governs the highest degree of antibunching achievable. It is worth noting that as the interaction time becomes shorter, the effect of photon antibunching increases.

The number of photons present in pump mode appears to be a good way to regulate the depth of nonclassicality. We conclude that the photon-antibunched states are associated with a large number of pump photons. We also demonstrate that the photon numbers of the fundamental mode and the harmonic mode directly influence the photon antibunching. We found that the third-order antibunching, with the same number of pump photons, has the most nonclassical features, followed by the second- and the first-order antibunching. We observe that the higher-order antibunching (second and third-order) achieves nonclassicality depth more effectively than ordinary (normal) antibunching.

We found that the first-order Hamiltonian interaction, which stimulates both pump and harmonic fields, is more nonclassical than the second-order Hamiltonian interaction. A powerful pump field creates a stronger interaction and is useful for achieving stronger photon antibunching.

It was found that there is no evidence of photon antibunching in the harmonic mode over pump mode up to the first- and second-order Hamiltonian interactions in the sixth harmonic generation process. In contrast, the coherent state of a pump field with a nonzero harmonic field generates photon clusters. This is due to the fact that the coherent state of the pump field causes interaction, resulting in bunching effects. On the other hand, the vacuum state of a pump field with a nonzero harmonic field results in photon bunching. This is due to the fact that the vacuum state of the pump field stimulates interaction, producing bunching effects.

We observe that in the sixth harmonic generation, as the number of photons in the pump mode increases, the second-order correlation function with zero time delay, or photon number correlation, declines, and the depth of nonclassicality increases. Consequently, we argued that the second correlation function becomes minimal in locations where photon antibunching is at its highest. We revealed photon antibunching as conclusive evidence of the quantum character of light. Overall, higher-order antibunching is a phenomenon that diminishes the likelihood of concurrently detecting multiple particles in comparison with the standard light source. This phenomenon broadens the definition of antibunching to include higher-order correlations as well as pairs of particles. Beyond normal antibunching effects, the depth of non-classicality quantifies the degree to which observed higher-order photon correlations deviate from classical predictions, thereby indicating the light source's degree of non-classical behaviour.

The results of this paper are simple and easy to reproduce in most physical systems labs. This opens the door for the experimental observation of higher-order antibunching and the development of probabilistic single-photon sources for quantum teleportation and quantum cryptography. More than these basic and abstract ideas, making larger and better nonclassical multiphoton states possible could help in creation of new devices like quantum dots, nanolasers, and on-chip integrated photonic circuits. These devices leverage the quantum properties of light and enable advancements in areas like on-chip quantum information processing and quantum communication.

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