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ANTIBUNCHING IN LIGHT HARMONICS GENERATION FROM FIELD QUANTISATION

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1. INTRODUCTION

In quantum optics, one usually deals with light which can be described in terms of a Sudarshan-Glauber phase-space distribution function. Light of this kind exhibits a positive, or at the most, a zero photon correlation (Hanbury Brown-Twiss or bunching) effect. Recently, however, it has been shown that nonlinear processes such as degenerate parametric amplification[1] as well as two-[2] and many-photon absorption,[3] can give rise to light with a negative Hanbury Brown-Twiss (antibunching) effect. Such light has no classical counterpart, and has to be treated by quantum mechanics.

The problem of light statistics in second harmonic generation has been considered in various papers;[4-8] nonetheless, even if based on a quantal approach (dealing with the field as an operator rather than a c-number), their results do not go beyond formal quantum-classical equivalence in their description of the field. Walls[9] drew attention to the fact that, if spontaneous decay of the second harmonic into two photons of the fundamental beam is taken into account, an oscillating solution given by an elliptical Jacobi function is obtained for the number of photons of the second harmonic. Classically, omitting fluctuations one obtains a monotonically increasing solution in the form of a hyperbolic tangent. Dewael[10] first attempted to take into account effectively the influence of the quantal properties of light on the degree of second-order coherence of the generated beam. Albeit his result, a positive value of the Hanbury Brown-Twiss effect, is hardly correct.

In this paper we show that if the field is dealt with quantally, a negative Hanbury Brown-Twiss effect results for processes of harmonics generation also. This amounts to the statement that harmonics generation can be the source of nonclassical fields to the same degree as the above mentioned nonlinear processes.

2. THEORY

Quantum mechanically, k'th harmonic generation can be described starting from the Hamiltonian of interaction between the fundamental and generated beams, in the form:[5]

$$H_I = \hbar c L_{k\omega} a_h^{\dagger} a_f^{k} + \text{h.c.} \tag{1}$$

with $L_{k\omega}$ - the coupling constant - dependent on the nonlinear properties of the medium and the state of polarisation of the incident beam, and $a_f(a_f^{\dagger}), a_h(a_h^{\dagger})$ - annihilation (creation) operators for the fundamental (f) and harmonic (h) beams. Using the interaction Hamiltonian (1), one readily derives the Heisenberg equations of time-evolution for the four operators. However, in processes of harmonics generation, we deal with travelling waves rather than fields in a cavity. By substitution of $t = -z/c$, where z is the path traversed by the wave in the medium, the cavity problem reduces formally to a travelling waves problem.[11,5] After the above substitution, the equations of motion of the slowly-variable part (free evolution is eliminated) of the annihilation operators for both beams, for perfect phase matching, become:

$$\frac{da_h(z)}{dz} = i\, L_{k\omega}\, a_f^{k}(z) \quad ,$$

$$\frac{da_f(z)}{dz} = i\, k\, L_{k\omega}^{*}\, [a_f^{\dagger}(z)]^{k-1}\, a_h(z) \quad . \tag{2}$$

Equations (2), jointly with the Hermitian-conjugate equations of the creation operators, form a set of differential operator equations, inaccessible to strict solution. Applying Eqs.(2) and their operator properties, it is possible to calculate approximately the variations in field correlation function for the generated and fundamental beams on traversal of the path z in the medium.[12]

On expanding the correlation functions in z, one has:

$$G^{(n)}(z) = G_0^{(n)} + \sum_{k=1} \frac{z^k}{k!} \frac{d^k}{dz^k} G^{(n)}(z) \Big|_{z=0} \quad , \tag{3}$$

where the correlation functions $G^{(n)}(z)$ are defined as:

$$G^{(n)}(z) = <[a^{\dagger}(z)]^n \ [a(z)]^n> \quad . \tag{4}$$

The symbol < ... > in (4) stands for the quantum mechanical mean. Provided the correlation functions (4) vary but little along the path z in the medium, it is sufficient to take but the first few terms of (3) in order to approximate the values of these functions satisfactorily. The procedure, in fact, corresponds to that of short-time solutions in a cavity problem. On differentiating Eqs. (2) and putting z=0, one obtains the values of the successive derivatives of the creation and annihilation operators. By insertion of the thus calculated values of the field operator derivatives into the right-hand term of (3) and on reduction of all terms to normal ordering by the use of boson commutation rules, one obtains the successive terms of the expansion of the functions $G^{(n)}(z)$. The expressions thus obtained for $G^{(n)}(z)$ depend, in successive approximations, on higher powers of z and on correlation functions of the incident beam $G_{f0}^{(n)}$ of higher and higher orders.

Applying the above procedure for the magnitude of the Hanbury Brown-Twiss effect, proportional to $G^{(2)}(z) - [G^{(1)}(z)]^2$, in the experimentally most highly relevant case of second-harmonic generation, we have obtained[13] the expressions:

$$G_{2\omega}^{(2)}(z) - \left[G_{2\omega}^{(1)}(z)\right]^2 = \left|L_{2\omega}\right|^4 \left\{G_{f0}^{(4)} - \left[G_{f0}^{(2)}\right]^2\right\} z^4$$

$$- \frac{4}{3}\left|L_{2\omega}\right|^6 \left\{2\left[G_{f0}^{(5)} - G_{f0}^{(3)}\ G_{f0}^{(2)}\right] + 3\ G_{f0}^{(4)} - \left[G_{f0}^{(2)}\right]^2\right\} z^6 + \dots ,$$

$$G_{f}^{(2)}(z) - \left[G_{f}^{(1)}(z)\right]^2 = G_{f0}^{(2)} - \left[G_{f0}^{(1)}\right]^2$$

$$- 2\left|L_{2\omega}\right|^2 \left\{2\left[G_{f0}^{(3)} - G_{f0}^{(2)}\ G_{f0}^{(1)}\right] + G_{f0}^{(2)}\right\} z^2 + \dots . \tag{5}$$

In (5), $G_{f0}^{(n)} = <a_{f0}^{\dagger n}\ a_{f0}^{n}>$ are correlation functions of the incident beam. The absence of second-harmonic photons at the input (z=0) is assumed, $G_{2\omega 0}^{(n)} = 0$.

Similarly, expressions for the second-order coherence variations in arbitrary k'th harmonic generation processes can be derived. The formulae, however, are rather bulky. We restrict ourselves to adducing the formula of the generated beam:

$$G^{(2)}_{k\omega}(z) - \left[G^{(1)}_{k\omega}(z)\right]^2 = |L_{k\omega}|^4 \, z^4 \, \left\{G^{(2k)}_{f0} - \left[G^{(k)}_{f0}\right]^2\right\}$$

$$- \frac{k}{3} \, |L_{k\omega}|^6 \, z^6 \, \left\{\sum_{p=0}^{k-1} \sum_{s=0}^{p} s! \binom{p}{s} \binom{k-1}{s} \left[G^{(3k-s-1)}_{f0} - 2\, G^{(2k-s-1)}_{f0} \, G^{(k)}_{f0}\right]\right.$$

$$\left. + \sum_{p=0}^{k-1} \sum_{s=0}^{k-1} s! \, \binom{p+k}{s} \binom{k-1}{s} \, G^{(3k-s-1)}_{f0}\right\} , \tag{6}$$

where the $\binom{p}{s}$ are Newton binomial coefficients.

3. DISCUSSION

The expressions (5) and (6), obtained by us for the variations in second-order coherence functions, take a particularly simple and interesting form if the incident beam is coherent, i.e., if its field is given by the coherent state: $a|\alpha\rangle = \alpha|\alpha\rangle$. One then has $G^{(n)}_{f0} = \langle n_{f0}\rangle^n$, where $\langle n_{f0}\rangle$ is the mean number of photons of the incident beam. Formulae (5), in the lowest non-vanishing approximation, now reduce to:

$$G^{(2)}_{2\omega}(z) - \left[G^{(1)}_{2\omega}(z)\right]^2 = - \frac{8}{3} \, |L_{2\omega}|^6 \, \langle n_{f0}\rangle^4 \, z^6 + \ldots ,$$

$$G^{(2)}_{f}(z) - \left[G^{(1)}_{f}(z)\right]^2 = - 2 \, |L_{2\omega}|^2 \, \langle n_{f0}\rangle^2 \, z^2 + \ldots , \tag{5a}$$

yielding a negative Hanbury Brown-Twiss effect, both for the beam generated at the frequency 2ω and for the fundamental beam of frequency ω , on traversal by them of the path z in the medium.

If the incident beam is coherent, formula (6) takes the form:

$$G^{(2)}_{k\omega}(z) - \left[G^{(1)}_{k\omega}(z)\right]^2 = - \frac{k}{3} \, |L_{k\omega}|^6 \, z^6 \sum_{p=0}^{k-1} \left\{\sum_{s=0}^{k-1} s! \binom{p+k}{s} \binom{k-1}{s}\right.$$

$$\left. - \sum_{s=0}^{p} s! \binom{p}{s} \binom{k-1}{s}\right\} \langle n_{f0}\rangle^{3k-s-1} . \tag{6a}$$

Since the expression in parentheses $\{\ldots\} \geqslant 0$, the right-hand term of (6a) is negative. Thus, for arbitrary $k \geqslant 2$ we obtain a negative correlation of photons, if the incident beam is coherent. It is worth stressing that for classical fields, i.e., if a_{f0} and $a_{f0}^{\dagger}$ are dealt with as c-numbers, s takes but the one value s=0 and the right-hand term of Eq.(6a) vanishes. All the terms with $s \geq 1$ emerged owing to our application of boson commutation rules to the field operators. Consequently, the negative photon correlation reflects the quantal properties of the fields. The effect is negative, as apparent from (5a), not only for the generated beam but, as well, for the fundamental beam on traversal of the medium.

One easily finds from (5) and (6) that in the case of a chaotic incident beam, $G_{f0}^{(n)}=n!\langle n_{f0}\rangle^{n}$, the photon correlation is positive. Quantisation of the fields, however, reduces the effect in magnitude.

It is to be expected that all nonlinear interactions of quantized electromagnetic fields and matter have the ability to produce fields having no classical counterpart.

Quite recently, Mišta and Peřina[14] have discussed the problem of photon statistics in nondegenerate, parametric amplification process. Their results point to a negative correlation effect as well, and, in particular, go over into ours, (5a), for the fundamental beam.

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