QUANTUM FLUCTUATIONS IN SECOND-HARMONIC LIGHT GENERATION*

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Second-harmonic light generation (SHLG) is analyzed from the viewpoint of the photon statistics of the fundamental and generated beams versus the path traversed by the two waves in the medium. The calculations lead to an anti-bunching effect for coherent incident light.

1. Introduction

The problem of the statistical properties of light propagating in various media can be dealt with from the viewpoint of the changes in its statistical properties due to interaction with the medium, or from that of the dependence of the gain of various optical processes on the statistics of the incident light beam.

In the present investigation, we propose a quantum theory of second-harmonic light generation in the aspect of photon statistics. The problem has been dealt with repeatedly [1–7]. Our approach resembles that of ref. [4]; however, we moreover consider the change in statistics of the fundamental beam. Our results differ from those of Dewael [4]. For coherent incident light, our calculations lead to a negative Hanbury-Brown and Twiss effect [8]. This appears to us of considerable interest and worthy of further investigation.

2. Theory

In the cavity type of problem the time evolution of the field operators is considered. In the processes of harmonic generation, however, we are dealing with the propagation type of problem. Substituting $t = -z/v$, where z is the path traversed by a wave in a medium and v its velocity, the cavity problem may be replaced by the problem of the travelling wave (Shen [2]). Starting from the phenomenological hamiltonian given by Shen [2] for slowly varying parts of the photon annihilation operators $a_{\mathrm{f,h}}(z)$ of the fundamental beam f (frequency ω) and generated beam h (frequency 2ω), at phase matching we obtain the following quantum equations of motion:

$$\frac{\mathrm{d}a_{\mathrm{f}}(z)}{\mathrm{d}z} = 2\mathrm{i}K^{*}a_{\mathrm{f}}^{+}(z)\,a_{\mathrm{h}}(z),$$

$$\frac{\mathrm{d}a_{\mathrm{h}}(z)}{\mathrm{d}z} = \mathrm{i}Ka_{\mathrm{f}}^{2}(z), \tag{1}$$

where K is a coupling constant defined by the nonlinear susceptibility tensor of the medium and the polarisation state of the beams [2].

This set of operator differential equations is inaccessible to an exact solution. The most commonly considered case of $\mathrm{d}a_{\mathrm{f}}(z)/\mathrm{d}z = 0$, which can be solved exactly, is irrelevant from the point of view of the problem considered here.

We calculate the field correlation functions: $G^{(1)}(z)$ of the first and $G^{(2)}(z)$ of the second order, for the two beams. Quite generally, for a correlation function of order n, we assume the form:

$$G^{(n)}(z) = \langle [a^{+}(z)]^{n}\,[a(z)]^{n}\rangle, \tag{2}$$

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where the symbol ⟨ ⟩ denotes quantum-mechanical averaging over the states of the field.

Our procedure involves the approximate method [9] of expanding the correlation functions in a power series in z:

$$G^{(n)}(z) = G^{(n)}(z=0) + \sum_{k=1} \frac{z^k}{k!} \left.\frac{\mathrm{d}^k G^{(n)}(z)}{\mathrm{d}z^k}\right|_{z=0} \tag{3}$$

If the variations of the correlation functions along the path z are small, only the first few terms need to be considered. In this paper we include, at the most, the sixth approximation.

By eqs. (1) and assuming no second-harmonic photons to be present at $z = 0$, we obtain:

$$\left.\frac{\mathrm{d}a_\mathrm{f}(z)}{\mathrm{d}z}\right|_{z=0} = 0, \qquad \left.\frac{\mathrm{d}^2 a_\mathrm{f}(z)}{\mathrm{d}z^2}\right|_{z=0} = -2|K|^2 a_{\mathrm{f0}}^{+} a_{\mathrm{f0}}^2,$$

$$\left.\frac{\mathrm{d}^3 a_\mathrm{f}(z)}{\mathrm{d}z^3}\right|_{z=0} = 0, \qquad \left.\frac{\mathrm{d}^4 a_\mathrm{f}(z)}{\mathrm{d}z^4}\right|_{z=0} = 4|K|^4(4a_{\mathrm{f0}}^{+2} a_{\mathrm{f0}}^3$$

$$+ a_{\mathrm{f0}}^{+} a_{\mathrm{f0}} a_{\mathrm{f0}}^{+} a_{\mathrm{f0}}^2), \tag{4}$$

and

$$\left.\frac{\mathrm{d}a_\mathrm{h}(z)}{\mathrm{d}z}\right|_{z=0} = \mathrm{i}K a_{\mathrm{f0}}^2, \qquad \left.\frac{\mathrm{d}^2 a_\mathrm{h}(z)}{\mathrm{d}z^2}\right|_{z=0} = 0,$$

$$\left.\frac{\mathrm{d}^3 a_\mathrm{h}(z)}{\mathrm{d}z^3}\right|_{z=0} = -2\mathrm{i}K|K|^2(a_{\mathrm{f0}} a_{\mathrm{f0}}^{+} a_{\mathrm{f0}}^2 + a_{\mathrm{f0}}^{+} a_{\mathrm{f0}}^3), \tag{5}$$

where, for brevity, we have written $a_\mathrm{f}(z=0) = a_{\mathrm{f0}}$. Above, only the derivatives useful for our further calculations are given, omitting all terms involving a_{h0} and a_{h0}^{+} as these do not affect the shape of the correlation functions with regard to normal ordering within their products.

On insertion of eqs. (5) into (3) and by having recourse to the commutation rule for operators, $[a_{\mathrm{f0}}, a_{\mathrm{f0}}^{+}] = 1$, we obtain the following results for the first and second order correlation functions for the generated beam:

$$G_\mathrm{h}^{(1)}(z) = |K|^2 G_{\mathrm{f0}}^{(2)} z^2 - \tfrac{2}{3}|K|^4(2G_{\mathrm{f0}}^{(3)} + G_{\mathrm{f0}}^{(2)})z^4 + \dots,$$

$$G_\mathrm{h}^{(2)}(z) = |K|^4 G_{\mathrm{f0}}^{(4)} z^4 - \tfrac{4}{3}|K|^6(2G_{\mathrm{f0}}^{(5)} + 3G_{\mathrm{f0}}^{(4)})z^6 + \dots, \tag{6}$$

with $G_{\mathrm{f0}}^{(n)} = \langle a_{\mathrm{f0}}^{+n} a_{\mathrm{f0}}^{n}\rangle$. Our expression for $G_\mathrm{h}^{(1)}(z)$ differs from that of [4] in the second term. Although the functions $G_\mathrm{h}^{(1)}(z)$ and $G_\mathrm{h}^{(2)}(z)$ are of a form involving terms of order 4 and 6, their calculation – on the assumption that second harmonic photons are absent at the input – requires but the third derivatives of the creation and annihilation operators of eqs. (5).

For the fundamental beam, we have:

$$G_\mathrm{f}^{(1)}(z) = G_{\mathrm{f0}}^{(1)} - 2G_\mathrm{h}^{(1)}(z),$$

$$G_\mathrm{f}^{(2)}(z) = G_{\mathrm{f0}}^{(2)} - 2|K|^2(2G_{\mathrm{f0}}^{(3)} + G_{\mathrm{f0}}^{(2)})z^2$$

$$+ \tfrac{4}{3}|K|^4(7G_{\mathrm{f0}}^{(4)} + 12G_{\mathrm{f0}}^{(3)} + G_{\mathrm{f0}}^{(2)})z^4 + \dots, \tag{7}$$

The first of eqs. (7), on multiplication by the energy of the photons of the fundamental beam, expresses the energy conservation principle for SHLG.

Were the fields treated classically [$a(z)$ would then be a number, not an operator], the round parentheses of (6) and (7) would contain only the correlation function $G_{\mathrm{f0}}^{(n)}$ of the highest orders. The other terms in these parentheses result from the application of boson commutation rules and thus express the quantum properties of the field.

3. Discussion

The results reported above for the correlation functions (6) and (7) make it possible to calculate the magnitude of the Hanbury-Brown and Twiss effect [8], which is proportional to the difference $G^{(2)}(z) - [G^{(1)}(z)]^2$.

The quantities in question are given, for the two beams, by the following expressions:

(i) in the case of a coherent incident beam ($G_{\mathrm{f0}}^{(n)} = \langle n_{\mathrm{f0}}\rangle^n$, where $\langle n_{\mathrm{f0}}\rangle$ is the mean number of photons incident on the medium),

$$G_\mathrm{h}^{(2)}(z) - [G_\mathrm{h}^{(1)}(z)]^2 = -\tfrac{8}{3}|K|^6 \langle n_{\mathrm{f0}}\rangle^4 z^6 + \dots, \tag{8}$$

$$G_\mathrm{f}^{(2)}(z) - [G_\mathrm{f}^{(1)}(z)]^2 = -2|K|^2 \langle n_{\mathrm{f0}}\rangle^2 z^2 + \dots. \tag{9}$$

(ii) In the case of a chaotic incident beam ($G_{\mathrm{f0}}^{(n)} = n!\langle n_{\mathrm{f0}}\rangle^n$), and restricting ourselves to the lowest non-vanishing approximations in z,

$$G_\mathrm{h}^{(2)}(z) - [G_\mathrm{h}^{(1)}(z)]^2 = 20|K|^4\langle n_{\mathrm{f0}}\rangle^4 z^4$$

$$- \tfrac{16}{3}|K|^6\{54\langle n_{\mathrm{f0}}\rangle^5 + 17\langle n_{\mathrm{f0}}\rangle^4\}z^6 + \dots,$$

$$G_f^{(2)}(z) - [G_f^{(1)}(z)]^2 = \langle n_{f0}\rangle^2 - 4|K|^2 \{4\langle n_{f0}\rangle^3 + \langle n_{f0}\rangle^2\} z^2 + \dots \quad (10)$$

Thus, for coherent incident light we obtain a negative Hanbury-Brown and Twiss effect (anti-bunching), due solely to the quantum properties of the field. The terms responsible for this effect have arisen as a result of applying the commutation rules for the annihilation and creation operators of photons. In the process of harmonic generation, the radiation field from a classical source can go over into a radiation field having no classical counterpart. For a chaotic incident beam, the quantum nature of the field causes but a decrease in the bunching effect.

Experimental studies of photon statistics in second-harmonic generation of light will surely provide guidelines regarding the need to quantize the electromagnetic field. Our investigation of third-harmonic light generation now under way leads to similar results. The quantum properties of the field lead to anti-bunching also in two- [10] and, generally, in multi-photon absorption [11] and in processes involving degenerate parametric amplification [12].

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