SQUEEZED STATES IN HARMONIC GENERATION OF A LASER BEAM

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Harmonic generation processes are shown to minimize quantum fluctuations of the electromagnetic field in the fundamental as well as in every generated beam. For the harmonics, sqeezing is additionally dependent on the polarisation properties of both beams.

1. Introduction. The quantum description of the electromagnetic field closest to the classical one is its description in the representation of coherent states. If in particular the field is in a pure coherent state, the product of the uncertainties of the momentum P and coordinate Q takes the minimal value unity.

In this paper, we shall be referring to squeezed states of the electromagnetic field as ones with a value of the dispersion of whichever the canonical variables less than unity [1]:

$$\langle(\Delta Q)^2\rangle < 1 \quad \text{or} \quad \langle(\Delta P)^2\rangle < 1 \,. \tag{1}$$

These states by no means imply a more "classical" nature of the field. On the contrary, fields being in such states have no classical counterparts.

For some years the statistical properties of light relevant to obtaining a negative Hanbury-Brown and Twiss effect have been under study. This effect immediately reflects photon antibunching. Light, the field of which is in states with antibunched photons, has no classical analog either. Dispersion of the photon number in the mode is then less than the mean photon number. Maximalisation of photon antibunching is achieved for $\langle(\Delta n)^2\rangle = 0$, i.e. for the field in a pure number state. However, for such light $\langle(\Delta Q)^2\rangle$ and $\langle(\Delta P)^2\rangle$ are greater than unity. There is no general connection between both these purely quantum effects. However, the mechanism of obtaining such fields is identical, namely through nonlinear interaction of coherent light with the medium [1–3].

One of the possibilities in this respect is offered by harmonic-generation processes of laser light. Recent years have brought strong experimental evidence of 3rd, 5th, 7th and 9th harmonic generation of laser light in atomic and molecular gases and, particularly, metal vapours [4]. This stimulates us to undertake an analysis of squeezed states not only with regard to the second harmonic but moreover to higher harmonics of light. In this context the study of odd harmonics, generated in isotropic media by optical nonlinearities induced in the atoms and molecules by electric dipole transtiions, is especially promising [4].

Mandel [1], using our "short optical paths" expansion [5], has shown that the field of the fundamental beam has to be in a squeezed state after traversing the nonlinear medium. Aiming at a more extensive treatment of the matter, we generalize Mandel's results [1] to higher harmonic-generation processes, based on our earlier calculations for the kth harmonic [6,7].

2. Theory. The operators Q and P corresponding to observables are hermitian,

$$Q = a + a^+ \,, \quad P = \mathrm{i}(a^+ - a) \,, \tag{2}$$

where a and a^+ are photon annihilation and creation operators, respectively. Hence we get

$$\langle(\Delta Q)^2\rangle = 1 + 2\langle n\rangle - 2\langle a^+\rangle\langle a\rangle + \langle(\Delta a^+)^2\rangle + \langle(\Delta a)^2\rangle\,, \qquad \langle(\Delta P)^2\rangle = 1 + 2\langle n\rangle - 2\langle a^+\rangle\langle a\rangle - \langle(\Delta a^+)^2\rangle - \langle(\Delta a)^2\rangle\,. \tag{3}$$

The effective interaction hamiltonian between the one-mode fundamental (f) and generated ($k\omega$) beams in the electric dipole approximation has the form [6]:

$$H = \hbar v L_{k\omega} a^+_{k\omega} a^k_{\rm f} + \text{h.c.}. \tag{4}$$

$L_{k\omega}$ denotes the coupling constants, dependent on the nonlinear properties of the medium and the state of polarisation of the incident and generated modes; v is the velocity of light, equal for both beams at phase matching.

The Heisenberg equations of motion for the slowly varying parts of the appropriate operators have the form [6]

$$\mathrm{d}a_{\rm f}(z)/\mathrm{d}z = \mathrm{i}kL^*_{k\omega}\,[a^+_{\rm f}(z)]^{k-1} a_{k\omega}(z)\,, \qquad \mathrm{d}a_{k\omega}(z)/\mathrm{d}z = \mathrm{i}L_{k\omega} a^k_{\rm f}(z)\,. \tag{5}$$

This set of equations is inaccessible to strict solution but accessible to the iteration method only. With accuracy to the approximations of interest to us, we find:

$$a_{\rm f}(z) = a_{\rm f}(0) - \tfrac{1}{2}k|L_{k\omega}|^2 z^2\,[a^+_{\rm f}(0)]^{k-1} a^k_{\rm f}(0) + \dots\,,$$

$$a_{k\omega}(z) = \mathrm{i}L_{k\omega} z a^k_{\rm f}(0) - \tfrac{1}{6}\mathrm{i}kL_{k\omega}|L_{k\omega}|^2 z^3 \sum_{m=0}^{k-1}\sum_{s=0}^{m} s!\binom{m}{s}\binom{k-1}{s}[a^+_{\rm f}(0)]^{k-1-s} a^{2k-1-s}_{\rm f}(0) + \dots\,, \tag{6}$$

where we have omitted all terms involving operators $a_{k\omega}(0)$ or $a^+_{k\omega}(0)$, assuming that at the plane $z = 0$ at the input into the medium there are no photons of the kth harmonic mode and hence its amplitude equals zero. None of these terms affects the final results.

From the second of eqs. (6) we easily calculate [6,7] that

$$\langle n_{k\omega}(z)\rangle = |L_{k\omega}|^2 z^2 g^{(k)}_{\rm f}(0) - \tfrac{1}{3}k|L_{k\omega}|^4 z^4 \sum_{m=0}^{k-1}\sum_{s=0}^{m} s!\binom{m}{s}\binom{k-1}{s} g^{(2k-1-s)}_{\rm f}(0) + \dots\,, \tag{7}$$

and from the energy conservation principle in nondissipative media we further have:

$$\langle n_{\rm f}(z)\rangle = \langle n_{\rm f}(0)\rangle - k\langle n_{k\omega}(z)\rangle\,. \tag{8}$$

The mean photon number of the fundamental mode, thus calculated, would involve a term of the fourth order which is unnecessary in our further discussion. Restricting ourselves to the quadratic approximation only, we write

$$\langle n_{\rm f}(z)\rangle = \langle n_{\rm f}(0)\rangle - k|L_{k\omega}|^2 z^2 g^{(k)}_{\rm f}(0) + \dots\,, \tag{9}$$

where $g^{(k)}_{\rm f}(0)$ is the kth order correlation function of the incident beam equal to $\langle a^{+k}_{\rm f}(0) a^k_{\rm f}(0)\rangle$, and for coherent light at the same time

$$g^{(k)}_{\rm f}(0) = \langle n_{\rm f}(0)\rangle^k = |\alpha_{\rm f}|^{2k}\,, \tag{10}$$

where $\alpha_{\rm f}$ represents an amplitude-eigenvalue of the photon annihilation operator:

$$\langle[\Delta a_{\rm f}(z)]^2\rangle = \langle[\Delta a_{\rm f}(0)]^2\rangle - \tfrac{1}{2}k(k-1)|L_{k\omega}|^2 z^2 \langle[a^+_{\rm f}(0)]^{k-2} a^k_{\rm f}(0)\rangle\,.$$

$$- k|L_{k\omega}|^2 z^2\,\{\langle[a^+_{\rm f}(0)]^{k-1} a^{k+1}_{\rm f}(0)\rangle - \langle[a^+_{\rm f}(0)]^{k-1} a^k_{\rm f}(0)\rangle\langle a_{\rm f}(0)\rangle\} + \dots\,. \tag{11}$$

The solution for $\langle[\Delta a^+_{\rm f}(z)]^2\rangle$ is obtained from eq. (11) by hermitial coupling:

$$\langle a^+_{\rm f}(z)\rangle\langle a_{\rm f}(z)\rangle = \langle a^+_{\rm f}(0)\rangle\langle a_{\rm f}(0)\rangle - \tfrac{1}{2}k|L_{k\omega}|^2 z^2\,\{\langle a^{+k}_{\rm f}(0) a^{k-1}_{\rm f}(0)\rangle\langle a_{\rm f}(0)\rangle + \langle a^+_{\rm f}\rangle\langle[a^+_{\rm f}(0)]^{k-1} a^k_{\rm f}(0)\rangle\} + \dots\,. \tag{12}$$

For a coherent incident beam and with the approximations used above

$$\langle n_{\rm f}(z)\rangle = \langle a_{\rm f}^{+}(z)\rangle\langle a_{\rm f}(z)\rangle = |\alpha_{\rm f}|^2 - k|L_{k\omega}|^2 z^2|\alpha_{\rm f}|^{2k}, \quad \langle[\Delta a_{\rm f}(z)]^2\rangle = -\tfrac{1}{2}k(k-1)|L_{k\omega}|^2 z^2 |\alpha_{\rm f}|^{2(k-2)}\alpha_{\rm f}^2\,,$$

$$\langle[\Delta a_{\rm f}^{+}(z)]^2\rangle = -\tfrac{1}{2}k(k-1)|L_{k\omega}|^2 z^2|\alpha_{\rm f}|^{2(k-2)}\alpha_{\rm f}^{*2}\,. \tag{13}$$

Taking $\alpha_{\rm f} = |\alpha_{\rm f}|{\rm e}^{i\theta}$, from eqs. (3) and with respect to eqs. (13) we arrive at

$$\langle[\Delta Q_{\rm f}(z)]^2\rangle = 1 - k(k-1)|L_{k\omega}|^2 z^2|\alpha_{\rm f}|^{2(k-1)}\cos 2\theta\,, \quad \langle[\Delta P_{\rm f}(z)]^2\rangle = 1 + k(k-1)|L_{k\omega}|^2 z^2|\alpha_{\rm f}|^{2(k-1)}\cos 2\theta\,. \tag{14}$$

It is easily seen from these dependences that for well-defined phases θ we obtain minimalisation of quantum fluctuations either in $Q_{\rm f}$ or in $P_{\rm f}$. The results (14) go over for $k = 2$ into Mandel's [1] results for second-harmonic generation.

Let us recall at this point that the scaled Hanbury-Brown and Twiss effect $[g^{(2)} - (g^{(1)})^2]/g^{(1)}$ for the fundamental beam amounts to [7,8] $-k(k-1)|L_{k\omega}|^2 z^2|\alpha_{\rm f}|^{2(k-1)}$, so that the deviation from unity in eqs. (14) is of the same order.

On performing similar calculations for the generated harmonic beams, for a coherent incident beam, we get:

$$\langle n_{k\omega}(z)\rangle = \langle a_{k\omega}^{+}(z)\rangle\langle a_{k\omega}(z)\rangle = |L_{k\omega}|^2 z^2|\alpha_{\rm f}|^{2k} - \tfrac{1}{3}k|L_{k\omega}|^4 z^4 \sum_{m=0}^{k-1}\sum_{s=0}^{m} s!\binom{m}{s}\binom{k-1}{s}|\alpha_{\rm f}|^{2(2k-1-s)}\,,$$

$$\langle[\Delta a_{k\omega}(z)]^2\rangle = \tfrac{1}{6}kL_{k\omega}^2|L_{k\omega}|^2 z^4 \sum_{m=0}^{k-1}\sum_{s=0}^{m}\sum_{t=1}^{k-1-s} s!t!\binom{m}{s}\binom{k-1}{s}\binom{k-1-s}{t}\binom{k}{t}|\alpha_{\rm f}|^{2(k-1-s-t)}\alpha_{\rm f}^{2k}\,,$$

$$\langle[\Delta a_{k\omega}^{+}(z)]^2\rangle = \tfrac{1}{6}kL_{k\omega}^{*2}|L_{k\omega}|^2 z^4 \sum_{m=0}^{k-1}\sum_{s=0}^{m}\sum_{t=1}^{k-1-s} s!t!\binom{m}{s}\binom{k-1}{s}\binom{k-1-s}{t}\binom{k}{t}|\alpha_{\rm f}|^{2(k-1-s-t)}\alpha_{\rm f}^{*2k}\,. \tag{15}$$

If we take $L_{k\omega} = |L_{k\omega}|\exp({\rm i}\varphi_k)$, then

$$\langle[\Delta R_{k\omega}(z)]^2\rangle = 1 \pm \tfrac{1}{3}k|L_{k\omega}|^4 z^4 \sum_{m=0}^{k-1}\sum_{s=0}^{m}\sum_{t=1}^{k-1-s} s!t!\binom{m}{s}\binom{k-1}{s}\binom{k-1-s}{t}\binom{k}{t}|\alpha_{\rm f}|^{2(2k-1-s-t)}\cos 2(\varphi_k + k\theta)\,, \tag{16}$$

where the upper + sign refers to the dispersion of $R = Q$ whereas − refers to $R = P$. Again, depending on the sign of the phase, but now given as $\varphi_k + k\theta$, the one or other quadrature component is squeezed. The deviation from unity is of the same order as photon antibunching in the generated beam [6,7].

3. Discussion. In particular, for the second harmonic

$$\langle[R_{2\omega}(z)]^2\rangle = 1 \pm \tfrac{8}{3}|L_{2\omega}|^4 z^4|\alpha_{\rm f}|^4\cos 2(\varphi_2 + 2\theta)\,, \quad R = P, Q\,, \tag{17}$$

while the scaled Hanbury-Brown and Twiss effect is $-\tfrac{8}{3}|L_{2\omega}|^4 z^4|\alpha_{\rm f}|^4$ [5]. Mandel [1] did not carry out calculations for the generated second-harmonic component since minimalisation of quantum fluctuations is not so effective in this case as for the fundamental beam. However, it is worth stressing that for the generated beams the effect under consideration is additionally dependent on the new phase φ_k arising from the complexity of the coupling constant. This originates in the polarisation properties of both beams.

In particular, let us assume that the incident light is linearly polarized. Then we can assume $\varphi_k = 0$, and a simple analysis of eqs. (14) and (16) shows that simultaneous squeezing for $Q_{\rm f}$ and $Q_{k\omega}$ ($k = 2, 3$) is possible for the phases $\theta \in (\pi/4k, \pi/4)$, and for $k \geqslant 4$ at $\theta \in (\pi/4k, 3\pi/4k)$; for $Q_{\rm f}$ and $P_{k\omega}$ for $\theta < \pi/4k$; and for $P_{\rm f}$ and $Q_{k\omega}$ for $\theta \in (\pi/4, 3\pi/8)$ for the second harmonic only. One can never get simultaneous squeezing for $P_{\rm f}$ and $P_{k\omega}$.

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