Higher-order squeezing in kth-harmonic generation

M. Kozierowski
Nonlinear Optics Division, Institute of Physics, Adam Mickiewicz University, Grunwaldzka 6, PL-60-780 Poznań, Poland
(Received 13 March 1986)

Hong and Mandel's concept of higher-order squeezing is applied to higher-harmonic generation ($k \geq 2$). The fundamental mode is shown to be intrinsically squeezed to the order k of the process for even k and to the order of $k-1$ for odd k.

I. INTRODUCTION

The 1980s witnessed an intense search for squeezed states of the electromagnetic field.[1] Like photon antibunching, squeezing is a direct manifestation of the quantum nature of light. Moreover, squeezing is a promising phenomenon for optical communication and the detection of gravitational waves.

Squeezing has recently been observed by Slusher *et al.*[2] in four-wave mixing. Also recently, Hong and Mandel[3] introduced the concept of higher-order squeezing and discussed the problem for resonance fluorescence, degenerate parametric down conversion and second-harmonic generation.

It is our aim to extend their results for second-harmonic generation[3] to generation of higher-order harmonics (kth order, $k=2,3,4,\ldots$). We show for the fundamental mode of frequency ω that the existence of intrinsic higher-order squeezing is simply related to the order of the process, and the field of this mode is intrinsically squeezed to the kth order for even k and to the $(k-1)$th order for odd k; Nth-order squeezing ($N>k$) is a manifestation of these intrinsic lower-order squeezings.

II. EQUATION OF MOTION

At perfect phase matching the process of kth harmonic generation in a nondissipative medium is described in the electric dipole approximation by the following Hamiltonian:[4]

$$H=\hbar\omega\hat{a}_f^\dagger\hat{a}_f+k\hbar\omega\hat{a}_k^\dagger\hat{a}_k+\hbar\kappa_k(\hat{a}_k^\dagger\hat{a}_f^k+\hat{a}_f^{\dagger k}\hat{a}_k)\,, \tag{1}$$

where the subscripts k and f denote the mode with frequency $k\omega$ and the fundamental mode, respectively. The factor κ_k stands for the coupling constant and $\hat{a}^\dagger$ and $\hat{a}$ are photon creation an annihilation operators. Owing to nonlinear coupling these operators are functions of the time t not only by way of the oscillating factors. Quite generally,

$$\begin{aligned}\hat{a}_f(t)&=\hat{a}_{fs}(t)e^{-i\omega t}\,,\\ \hat{a}_k(t)&=\hat{a}_{ks}(t)e^{-ik\omega t}\,,\end{aligned} \tag{2}$$

where $\hat{a}_{fs}$ and $\hat{a}_{ks}$ are slowly varying parts of the operators.

The Hamiltonian (1) leads to the following set of coupled equations of motion for the slowly varying parts of the annihilation operators

$$\begin{aligned}\dot{\hat{a}}_{fs}&=-ik\kappa_k\hat{a}_{fs}^{\dagger k-1}\hat{a}_{ks}\,,\\ \dot{\hat{a}}_{ks}&=-i\kappa_k\hat{a}_{fs}^{k}\,.\end{aligned} \tag{3}$$

The time evolution of these operators has been solved as yet in the short-time approximation only, and hence photon antibunching[5] and squeezing[4,6,7] have been predicted solely at the beginning of the process, i.e., for appropriately thin nonlinear media. The short-time approximation is related to the expansion of the operators in a power series in t.

Since we restrict our attention to the fundamental mode, we present below the power-series solution for $\hat{a}_{fs}$ only:

$$\hat{a}_{fs}(t)=\hat{a}_f(0)-ik\kappa_k t\hat{a}_f^{\dagger k-1}(0)\hat{a}_k(0)+\frac{k}{2}\kappa_k^2t^2\left[\sum_{r=1}^{N-1}r!\begin{pmatrix}k-1\\ r\end{pmatrix}\begin{pmatrix}k\\ r\end{pmatrix}[:\hat{n}_f^{k-1-r}(0):]\hat{a}_f(0)\hat{n}_k(0)-[:\hat{n}_f^{k-1}(0):]\hat{a}_f(0)\right]+\cdots\,, \tag{4}$$

where $\hat{n}=\hat{a}^\dagger\hat{a}$ is the photon-number operator and the symbol : : stands for normal ordering of the photon creation and annihilation operators.

III. DEFINITIONS OF SQUEEZING

Let us introduce, after Hong and Mandel,[3] the two slowly varying, Hermitian, in-phase and out-of-phase, quadrature components $\hat{E}_1$ and $\hat{E}_2$ of the field:

$$\begin{aligned}\hat{E}_1(t)&=\hat{a}_s(t)e^{-i\phi}+\hat{a}_s^\dagger(t)e^{i\phi}\,,\\ \hat{E}_2(t)&=\hat{a}_s(t)e^{-i(\phi+\pi/2)}+\hat{a}_s^\dagger(t)e^{i(\phi+\pi/2)}\,,\end{aligned} \tag{5}$$

where ϕ is some phase angle that may be chosen arbitrarily.

The electromagnetic field is squeezed to the second order if one of the variances of the quadrature components is less than unity,

$$\langle (\Delta \hat{E}_{1,2})^2 \rangle < 1 , \tag{6}$$

or, equivalently, if one of the normally ordered variances satisfies the following inequality:

$$\langle :(\Delta \hat{E}_{1,2})^2: \rangle < 0 . \tag{7}$$

From the inequality (7), it is obvious that negative values of the normally ordered variances cannot have counterparts in the classical description; hence, squeezing has a purely quantum origin.

Hong and Mandel[3] have generalized the definition of the second-order squeezing (6) to Nth-order squeezing. Definitions of higher-order squeezing are, in general, meaningful for even N only, then implying directly its quantum nature. It has been shown by the above authors that, for even N, the field is squeezed to the Nth order if, for the component 1 or 2,

$$\langle (\Delta \hat{E}_{1,2})^N \rangle < (N-1)!! . \tag{8}$$

These moments may be expressed by the normally ordered even moments as follows:

$$\langle (\Delta \hat{E}_{1,2})^N \rangle = \sum_{r=0}^{N/2-1} \left[\binom{N}{2r} \frac{(2r)!}{r!2^r} \langle :(\Delta \hat{E}_{1,2})^{N-2r}: \rangle \right] + (N-1)!! , \tag{9}$$

where the formula (9) is written in shortened form.

As was shown,[3] the inequality (8) may be satisfied even in the case of non-negative terms $\sum_{r=0}^{N/2-2}[\ ]$ if, of course, the dominating term describing second-order squeezing is negative. In this case the field is said to be squeezed to the Nth order but is not intrinsically squeezed to this order. The Nth-order squeezing is then intimately related to the intrinsic second-order squeezing as in the case of resonance fluorescence of a single two-level atom or the fundamental mode in second-harmonic generation.[3]

IV. RESULTS

The kth harmonic mode starts from vacuum fluctuations, so that the following initial conditions are satisfied:

$$\langle 0 | \hat{a}_k(0) | 0 \rangle_k = \langle 0 | \hat{n}_k(0) | 0 \rangle_k = 0 . \tag{10}$$

In turn, let us assume that the input mode is in the coherent state

$$\hat{a}_f(0) | \alpha \rangle = \alpha | \alpha \rangle , \tag{11}$$

and $\alpha = |\alpha| \exp(i\theta)$.

From (4), at (10) and (11), we arrive at

$$\begin{aligned} \Delta \hat{E}_{1f}(t) = & \left\{ \hat{a}_f - \alpha - i\kappa_k t \hat{a}_f^{\dagger k-1} \hat{a}_k + \frac{k}{2}\kappa_k^2 t^2 \left[\sum_{r=1}^{k-1} r! \binom{k-1}{r} \binom{k}{r} (:\hat{n}_f^{k-1-r}:)\hat{a}_f \hat{n}_k - (:\hat{n}_f^{k-1}:)\hat{a}_f + |\alpha|^{2(k-1)}\alpha \right] \right\} e^{-i\phi} \\ & + \left\{ \hat{a}_f^{\dagger} - \alpha^* + ik\kappa_k t \hat{a}_f^{k-1} \hat{a}_k^{\dagger} + \frac{k}{2}\kappa_k^2 t^2 \left[\sum_{r=1}^{k-1} r! \binom{k-1}{r} \binom{k}{r} \hat{a}_f^{\dagger}(:\hat{n}_f^{k-1-r}:)\hat{n}_k \right. \right. \\ & \left. \left. - \hat{a}_f^{\dagger}(:\hat{n}_f^{k-1}:) + |\alpha|^{2(k-1)}\alpha^* \right] \right\} e^{i\phi} , \end{aligned} \tag{12}$$

where for the sake of brevity we have omitted the argument $t=0$ at the operators.

Second-order squeezing has been obtained in the quadratic approximation.[4–7] We shall calculate here all expectations to within the same accuracy. With respect to Eq. (12) we get for the normally ordered moments,

$$\begin{aligned} \langle :[\Delta \hat{E}_1(t)]^N: \rangle = \sum_{r=0}^{N} \binom{N}{r} \Big\langle & \left[\hat{a}_f^{\dagger} - \alpha^* - \frac{k}{2}\kappa_k^2 t^2 [\hat{a}_f^{\dagger}(:\hat{n}_f^{k-1}:) - |\alpha|^{2(k-1)}\alpha^*] \right]^r \\ & \times \left[\hat{a}_f - \alpha - \frac{k}{2}\kappa_k^2 t^2 [(:\hat{n}_f^{k-1}:)\hat{a}_f - |\alpha|^{2(k-1)}\alpha] \right]^{N-r} \Big\rangle e^{-i(N-2r)\phi} , \end{aligned} \tag{13}$$

where we have neglected all terms containing the operators of the generated kth harmonic mode, since they contribute nothing to the second order in t.

Inspection of Eq. (13) shows further that only terms $r=0$ and $r=N$ can lead to nonvanishing expectations, and hence

$$\langle :[\Delta\hat{E}_1(t)]^N: \rangle = -\frac{k}{2}\kappa_k^2 t^2 \sum_{s=0}^{N-1} \{ \langle (\hat{a}_f-\alpha)^{N-1-s}[(:\hat{n}_f^{k-1}:)\hat{a}_f - |\alpha|^{2(k-1)}\alpha](\hat{a}_f-\alpha)^s \rangle e^{-iN\phi} + \langle (\hat{a}_f^\dagger-\alpha^*)^s[\hat{a}_f^\dagger(:\hat{n}_f^{k-1}:) - |\alpha|^{2(k-1)}\alpha^*](\hat{a}_f^\dagger-\alpha^*)^{N-1-s} \rangle e^{iN\phi} \} . \quad (14)$$

One notes that nonzero expectations in Eq. (14) may be obtained only at $s=0$ and, obviously, for $N\geq 2$.

To evaluate these expectations we use the commutation relation

$$(\hat{a}_f-\alpha)^{N-1}(:\hat{n}_f^{k-1}:) = \sum_{m=0} m! \binom{k-1}{m}\binom{N-1}{m} \times (:\hat{n}_f^{k-1-m}:)(\hat{a}_f-\alpha)^{N-1-m} , \quad (15)$$

where, in general, the upper limit of summation should be equal to the smaller of the numbers $k-1$ or $N-1$. However, the expectation value of Eq. (15) in a coherent state differs from zero only for $m=N-1$ and simultaneously the following condition becomes apparent:

$$N \leq k . \quad (16)$$

In other words, the order of the highest nonzero normally ordered moment may be at the most equal to the order of the process if the latter is caused by coherent input radiation.

Finally, we have

$$\langle :[\Delta\hat{E}_1(t)]^N: \rangle = -k\kappa_k^2 t^2 \frac{(k-1)!}{(k-N)!} \times |\alpha|^{2k-N}\cos[N(\phi-\theta)] , \quad (17)$$

and, at $k=2$, Hong and Mandel's[3] result is recovered.

By the same procedure

$$\langle :[\Delta\hat{E}_2(t)]^N: \rangle = -k\kappa_k^2 t^2 \frac{(k-1)!}{(k-N)!} \times |\alpha|^{2k-N}\cos\left[N\left(\phi-\theta+\frac{\pi}{2}\right)\right] . \quad (18)$$

Let us restrict our discussion to moments of even order. From Eqs. (17) and (18) it is obvious that, depending on the phase, either the component $\hat{E}_1$ ($\phi-\theta=n\pi$, n any integer) or $\hat{E}_2$ [$\phi-\theta=(n+\frac{1}{2})\pi$] is squeezed intrinsically for all allowed N.

With respect to the inequality (16), in the case of the even-order process the field of the fundamental mode may be intrinsically squeezed up to the kth order $(2,4,6,\ldots,k)$ while in the case of the odd-order process intrinsic squeezing is to the $(k-1)$th order $(2,4,6,\ldots,k-1)$. In other words, the concept of higher-order squeezing is more than purely mathematical.

On substitution of Eq. (17) into Eq. (9) we have for the even-order process

$$\langle [\Delta\hat{E}_1(t)]^N \rangle = (N-1)!! - k(k-1)!\kappa_k^2 t^2 \sum_{r=(N-k)/2}^{N/2-1} \left[\binom{N}{2r} \frac{(2r)!\,|\alpha|^{2k-N+2r}}{r!2^r(k-N+2r)!} \cos[(N-2r)(\phi-\theta)] \right], \quad N \text{ even} \quad (19)$$

and for odd-order process

$$\langle [\Delta\hat{E}_1(t)]^N \rangle = (N-1)!! - k(k-1)!\kappa_k^2 t^2 \sum_{r=(N-k+1)/2}^{N/2-1} [\ \ ], \quad N \text{ even} \quad (20)$$

where the expression inside the large square brackets in Eq. (20) is the same as in Eq. (19). The Nth-order squeezing (now, in general, N may exceed k) is a manifestation of intrinsic lower-order squeezings.

It is worth noting that the moments (17) and (18) can be nonzero also for odd N and, in the case of harmonics generation, this is a purely quantum effect too, as arising from the commutation relation (15). The sign of the odd normally ordered moments may be the same or opposite to that of the even normally ordered moments.

ACKNOWLEDGMENTS

This work was supported by Research Project CPBP 01.06.

[1]D. F. Walls, Nature (London) **306**, 141 (1983). S. Kielich, M. Kozierowski, and R. Tanaś, Opt. Acta **32**, 1023 (1985), and references therein.
[2]R. E. Slusher, L. W. Hollberg, B. Yurke, J. C. Mertz, and J. F. Valley, Phys. Rev. Lett. **54**, 2409 (1985).
[3]C. K. Hong, and L. Mandel, Phys. Rev. A **32**, 974 (1985); Phys. Rev. Lett. **54**, 323 (1985).
[4]M. Kozierowski, and S. Kielich, Phys. Lett. A **94**, 213 (1983).
[5]M. Kozierowski, and R. Tanaś, Opt. Commun. **21**, 229 (1977).
[6]L. Mandel, Opt. Commun. **42**, 437 (1982).
[7]J. Peřina, V. Peřinova, and J. Koďousek, Opt. Commun. **49**, 210 (1984).