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Generation of higher-order squeezing of quantum electromagnetic fields

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The concept of Nth-order squeezing and intrinsic Nth-order squeezing is introduced and applied to several physical situations that are known to exhibit second-order squeezing. It is shown that the field produced in degenerate parametric down-conversion is squeezed to all orders, as well as being intrinsically squeezed to orders $2,6,10,\ldots$. The same is true for the fundamental mode in second-harmonic generation within the short-time approximation, but in this case there is only intrinsic second-order squeezing. In resonance fluorescence from a two-level atom squeezing is again found to be intrinsically a second-order phenomenon, although an intrinsic second-order effect can give rise to a weak form of higher-order squeezing.

I. INTRODUCTION

The subject of squeezing, particularly of a quantized electromagnetic field, has generated a good deal of attention in the last few years.[1–4] This interest is related to the possibility of reducing the noise of an optical signal below the vacuum limit, or below that achievable in a completely coherent field. This has obvious attractions for optical communication, and for such purposes as gravitational-wave detection, where signals close to or even below the quantum limit are expected. It has been shown theoretically that squeezing should be realizable in numerous physical processes, such as degenerate parametric processes,[1–11] phase conjugation or four-wave mixing,[12–16] harmonic generation,[17–21] resonance fluorescence,[22–26] the free-electron laser,[27] etc. However, the phase-sensitive nature of the squeezing phenomenon makes it very difficult to observe experimentally.

In the usual approach to squeezing within the context of quantum optics, the real field $\hat{E}$ is decomposed into two quadrature components $\hat{E}_1$ and $\hat{E}_2$ which are canonical conjugates.[28] In a squeezed state the fluctuations $\langle(\Delta\hat{E}_1)^2\rangle$ of one of these components, $\hat{E}_1$ say, are reduced at the cost of a corresponding increase in the fluctuations $\langle(\Delta\hat{E}_2)^2\rangle$ of the other one. This is to be contrasted with the situation in a completely coherent field, where both quadrature components fluctuate equally. If information could be impressed on and extracted from the $\hat{E}_1$ component, this would result in a reduced amount of noise. Up to now the focus has been entirely on quantities like $\langle(\Delta\hat{E}_1)^2\rangle$ that are quadratic in the field. With the development of techniques for making higher-order correlation measurements in quantum optics, it is natural to turn our attention to the higher-order moments of the field also, and to ask if there are circumstances when these also could be reduced below the vacuum limit, or the values for a coherent state. If so, this opens the possibility of extracting information efficiently from an optical signal by some higher-order correlation measurement.

In the following we introduce a natural generalization of the squeezing concept that involves the higher-order moments of the field.[29] Like one that exhibits second-order squeezing, a field that is squeezed to a higher order is in a purely quantum mechanical state, and has no classical description. We show explicitly that the squeezed quantum state studied by Stoler,[2] which is a special case of the more general two-photon coherent states introduced by Yuen,[3] is actually squeezed to all orders. Next, we consider a number of physical situations that are known to generate second-order squeezing, and show that in several cases they also exhibit higher-order squeezing. Indeed, the fractional noise reduction achievable for the higher-order moments can be greater than for the second-order moments. Finally, we show that the process of resonance fluorescence from a two-level atom exhibits the weakest form of higher-order squeezing, in that it is intrinsically a second-order phenomenon.

II. DEFINITION OF HIGHER-ORDER SQUEEZING

Before introducing the definition of higher-order squeezing, it may be useful to summarize the usual second-order properties of a squeezed state. Let $\hat{\mathbf{E}}^{(+)}(\mathbf{r},t)$ and $\hat{\mathbf{E}}^{(-)}(\mathbf{r},t)$ be the positive-frequency and negative-frequency parts of an electromagnetic field vector, such as the electric field. $\hat{\mathbf{E}}^{(+)},\hat{\mathbf{E}}^{(-)}$ may be given a mode expansion in plane-wave modes $\mathbf{k},s$ in the usual manner. In the following we shall restrict this expansion in two ways. Because most detectors are sensitive only to a limited set of frequencies and directions, we limit the mode expansion to the finite set of wave vectors $[\mathbf{k}]$, and also we restrict it to one polarization component. This allows us to represent the measured field by a scalar, and to write the expansion in the form

$$\hat{E}^{(+)}(\mathbf{r},t)=\frac{1}{L^{3/2}}\sum_{[\mathbf{k}]} l(\mathbf{k})\hat{a}_{\mathbf{k}}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} . \quad (1)$$

Here $\hat{a}_{\mathbf{k}}$ is the photon-annihilation operator for the mode of wave vector $\mathbf{k}$, L^3 is the normalization volume, and $l(\mathbf{k})$ is a simple factor that depends on which electromagnetic field operator is being expanded [e.g., $l(\mathbf{k})=(\hbar\omega/2\epsilon_0)^{1/2}$ for the electric field)]. From Eq. (1), the commutator of $\hat{E}^{(+)},\hat{E}^{(-)}$ is given by

$$[\hat{E}^{(+)},\hat{E}^{(-)}]=\frac{1}{L^3}\sum_{[\mathbf{k}]}|l(\mathbf{k})|^2\equiv C\ , \tag{2}$$

where C is finite and positive definite. In the limit $L\to\infty$, the sum is replaceable by an integral in the usual way. In some problems it may be legitimate to focus on just one or two modes of the field, but in other cases we shall have to deal with a continuum of modes.

We now introduce the two slowly varying Hermitian quadrature components $\hat{E}_1,\hat{E}_2$ of the field, defined by

$$\begin{aligned}\hat{E}_1&\equiv\hat{E}^{(+)}e^{i(\omega t-\phi)}+\hat{E}^{(-)}e^{-i(\omega t-\phi)}\ ,\\ \hat{E}_2&\equiv\hat{E}^{(+)}e^{i(\omega t-\phi-\pi/2)}+\hat{E}^{(-)}e^{-i(\omega t-\phi-\pi/2)}\ ,\end{aligned} \tag{3}$$

where ϕ is some phase angle that may be chosen at will and ω is the mid-frequency. Evidently $\hat{E}_2$ can be regarded as just a special case of $\hat{E}_1$, in which ϕ is replaced by $\phi+\pi/2$. From Eqs. (2) and (3) it then follows that

$$[\hat{E}_1,\hat{E}_2]=2iC\ , \tag{4}$$

so that $\hat{E}_1,\hat{E}_2$ are canonical conjugates and obey the uncertainty relation

$$\langle(\Delta\hat{E}_1)^2\rangle\langle(\Delta\hat{E}_2)^2\rangle\geq C^2\ , \tag{5}$$

where $\Delta\hat{E}\equiv\hat{E}-\langle\hat{E}\rangle$. For a coherent state, including the vacuum, each dispersion $\langle(\Delta\hat{E}_1)^2\rangle,\langle(\Delta\hat{E}_2)^2\rangle$ equals C, and the uncertainty product has its minimum value.[30] On the other hand, the state is said to be squeezed if one or the other dispersion is less than its value in the coherent state. In other words, if there exists some phase angle ϕ for which

$$\langle(\Delta\hat{E}_1)^2\rangle<C\ , \tag{6}$$

then the state is squeezed to the second order in $\hat{E}_1$. An alternative definition can be given in terms of the normally ordered variance $\langle:(\Delta\hat{E}_1)^2:\rangle$, which is easily related to $\langle(\Delta\hat{E}_1)^2\rangle$ with the help of Eq. (4),

$$\langle:(\Delta\hat{E}_1)^2:\rangle=\langle(\Delta\hat{E}_1)^2\rangle-C\ . \tag{7}$$

Then the state is squeezed to the second order in $\hat{E}_1$ if there exists some phase ϕ for which $\langle:(\Delta\hat{E}_1)^2:\rangle$ is negative. The negative value of the normally ordered dispersion makes it clear that the field cannot be described classically when it is in a squeezed state, so that we are dealing with an explicitly quantum mechanical phenomenon.[30,31]

It seems natural to generalize the foregoing by describing the state as squeezed to the $(2N)$th order in $\hat{E}_1$ $(N=1,2,3,\ldots)$ if there exists a phase angle ϕ such that $\langle(\Delta\hat{E}_1)^{2N}\rangle$ is smaller than its value in a completely coherent state of the field. Although this definition could of course be extended to moments of odd order as well, it is not very meaningful for odd powers. $\langle(\Delta\hat{E}_1)^{2N+1}\rangle$ vanishes for a coherent state, and for other states the sign of the odd moments depends on the phase. Moreover, only for even moments does squeezing imply that the state is purely quantum mechanical. It is worth noting also that whereas the commutator of $\hat{E}_1$ and $\hat{E}_2$ is a c-number, so that the lower bound on the uncertainty product is independent of the state, this is not so for the higher moments of $\hat{E}_1$ and $\hat{E}_2$.

In order to show the nonclassical character of the squeezed state, it is useful to relate the higher moments of $\Delta\hat{E}_1$ to those in normal order. We make use of Eqs. (3) and (4) and the Campbell-Baker-Hausdorff identity in the form

$$\langle e^{x\Delta\hat{E}_1}\rangle=\langle:e^{x\Delta\hat{E}_1}:\rangle e^{x^2C/2}\ , \tag{8}$$

where x is any c-number. By expanding both sides as a power series in x, and equating coefficients of $x^N N!$, we arrive at the relation

$$\begin{aligned}\langle(\Delta\hat{E}_1)^N\rangle=\langle:(\Delta\hat{E}_1)^N:\rangle&+\frac{N^{(2)}}{1!}(\tfrac{1}{2}C)\langle:(\Delta\hat{E}_1)^{N-2}:\rangle+\frac{N^{(4)}}{2!}(\tfrac{1}{2}C)^2\langle:(\Delta\hat{E}_1)^{N-4}:\rangle+\cdots\\ &+\begin{cases}(N-1)!!C^{N/2} & \text{if } N \text{ is even,}\\ \dfrac{N!C^{N/2-3/2}}{3!2^{N/2-3/2}(\frac{1}{2}N-\frac{3}{2})!}\langle:(\Delta\hat{E}_1)^3:\rangle & \text{if } N \text{ is odd .}\end{cases}\end{aligned} \tag{9}$$

Here $N^{(r)}$ stands for $N(N-1)\cdots(N-r+1)$. Now the normally ordered moments $\langle:(\Delta\hat{E}_1)^N:\rangle$ all vanish for a coherent state. It follows from the definition that the state is squeezed to any even order N if

$$\langle(\Delta\hat{E}_1)^N\rangle<(N-1)!!C^{N/2}\ , \tag{10}$$

in which case the Nth moment is smaller than in the vacuum state of the field. Inspection of Eq. (9) reveals that one or more of the normally ordered even moments $\langle:(\Delta\hat{E}_1)^r:\rangle$ for $r\leq N$ then has to be negative, and this is possible only for a nonclassical state of the field. By limiting our discussion to moments of even order, we retain this important connection with second-order squeezing. As examples of the inequality (10) we list below the conditions for squeezing of the second, fourth, and sixth order, respectively:

$$\langle:(\Delta\hat{E}_1)^2:\rangle<0 \text{ for 2nd order squeezing ,}$$

$$\langle:(\Delta\hat{E}_1)^4:\rangle+6C\langle:(\Delta\hat{E}_1)^2:\rangle<0 \text{ for fourth-order squeezing ,}$$

$$\langle:(\Delta\hat{E}_1)^6:\rangle+15C\langle:(\Delta\hat{E}_1)^4:\rangle+45C^2\langle:(\Delta\hat{E}_1)^2:\rangle<0 \text{ for sixth-order squeezing .}$$

Finally, there is the possibility that condition (10) may be satisfied for some order N beyond 2, even though $\langle :(\Delta\hat{E}_1)^N: \rangle$ is not negative beyond $N=2$, simply because the term $\langle :(\Delta\hat{E}_1)^2: \rangle$ dominates in the series (9). We shall describe that situation as one in which there is only *intrinsic* second-order squeezing, which however gives rise to higher-order squeezing.

A convenient parameter q_N for measuring the degree of Nth order squeezing, which is a natural generalization of one introduced for second-order squeezing,[23] is

$$q_N = \frac{\langle (\Delta\hat{E}_1)^N \rangle - (N-1)!!C^{N/2}}{(N-1)!!C^{N/2}} . \tag{11}$$

q_N is negative whenever there is Nth order squeezing, and it has the maximum negative value -1.

As the commutator C enters explicitly into the equations, it may be useful to have an estimate of its order of magnitude for a continuum of modes. Let $\hat{E}$ be the electric field, so that $l(\mathbf{k})$ in Eq. (1) stands for $(\hbar\omega/2\epsilon_0)^{1/2}$, and let the set $[\mathbf{k}]$ of plane-wave modes correspond to a band $\Delta\nu$ of frequencies centered on ν in some direction within the solid angle $\Delta\Omega$. Then in the limit of large L, where the sum in Eq. (2) becomes an integral, if λ is the mean wave length, we have approximately

$$C \approx \left[\frac{\hbar\omega}{2\epsilon_0} \right] \left[\frac{\nu^2 \Delta\nu}{c^3} \right] \Delta\Omega = \frac{\pi\hbar}{\epsilon_0} \frac{\Delta\nu\Delta\Omega}{\lambda^3} . \tag{12}$$

III. APPLICATION TO THE SQUEEZED STATE $|v,z\rangle$

In 1970 Stoler[2] introduced the state $|v,z\rangle$ that is derived from the coherent state $|v\rangle$ by a unitary transformation involving squares of $\hat{a}$ and $\hat{a}^\dagger$, with the interesting property that the dispersion $\langle (\Delta\hat{E}_1)^2 \rangle$ in this state could be made arbitrarily small. This state, a form of which had already been encountered,[1] is a special case of the more general two-photon coherent state later introduced by Yuen.[3]

For simplicity we limit ourselves to a single-mode field. Let $|v\rangle$ be the right eigenstate of $\hat{a}$ belonging to the eigenvalue v,

$$\hat{a}|v\rangle = v|v\rangle , \tag{13}$$

which is the coherent state[30] $|v\rangle$. Let $\hat{U}(z)$ be the unitary operator

$$\hat{U}(z) \equiv e^{(z\hat{a}^2 - z^*\hat{a}^{\dagger 2})/2}, \quad z \equiv \Theta e^{i\theta} . \tag{14}$$

Then the state $|v,z\rangle$ is obtained from $|v\rangle$ by making the unitary transformation $\hat{U}(z)$, or

$$|v,z\rangle \equiv \hat{U}(z)|v\rangle . \tag{15}$$

It is convenient to introduce new, transformed annihilation and creation operators $\hat{A}, \hat{A}^\dagger$ given by

$$\hat{A}(z) = \hat{U}(z)\hat{a}\hat{U}^\dagger(z) , \tag{16}$$

and its conjugate, which also satisfy the commutation relation

$$[\hat{A}(z), \hat{A}^\dagger(z)] = 1 . \tag{17}$$

By application of the transformation in Eq. (16) it follows that

$$\hat{A}(z) = \hat{a}\cosh\Theta + \hat{a}^\dagger e^{-i\theta}\sinh\Theta , \tag{18}$$

and conversely,

$$\hat{a} = \hat{A}(z)\cosh\Theta - \hat{A}^\dagger(z)e^{-i\theta}\sinh\Theta . \tag{19}$$

Then one finds from Eqs. (15) and (16)

$$\hat{A}(z)|v,z\rangle = v|v,z\rangle , \tag{20}$$

and comparison with Eq. (13) shows that $\hat{A}(z)$ stands in the same relation to the state $|v,z\rangle$ as does $\hat{a}$ to the coherent state $|v\rangle$.

Now from the definition (3) applied to a single-mode field, we have

$$\hat{E}_1 = g\hat{a} + g^*\hat{a}^\dagger , \tag{21}$$

where

$$g \equiv \frac{1}{L^{3/2}} l(\mathbf{k}) e^{i(\mathbf{k}\cdot\mathbf{r} - \phi)} , \tag{22}$$

and

$$C = |g|^2 . \tag{23}$$

Also with the help of Eq. (19) and its conjugate we may write

$$\hat{E}_1 = \hat{A}f + \hat{A}^\dagger f^* , \tag{24}$$

where

$$f \equiv g\cosh\Theta - g^* e^{i\theta}\sinh\Theta . \tag{25}$$

If $l(\mathbf{k})$ is a real factor, then

$$|f|^2 = |g|^2[\cosh(2\Theta) - \sinh(2\Theta)\cos(2\mathbf{k}\cdot\mathbf{r} - 2\phi - \theta)] . \tag{26}$$

Hence $\hat{E}_1$ and $\Delta\hat{E}_1$ can be decomposed equally well into the operators $\hat{a}, \hat{a}^\dagger$ according to Eq. (21), or into the operators $\hat{A}, \hat{A}^\dagger$ according to Eq. (24).

We now apply the Campbell-Baker-Hausdorff identity in the form of Eq. (8), but with the normal ordering applicable to the $\hat{A}, \hat{A}^\dagger$ operators rather than to $\hat{a}, \hat{a}^\dagger$, and with the commutator C therefore identified with $|f|^2$. We then obtain in place of Eq. (9), after equating coefficients of $x^N/N!$,

$$\langle (\Delta\hat{E}_1)^N \rangle = \langle ::(\Delta\hat{E}_1)^N:: \rangle + \frac{N^{(2)}}{1!}(\tfrac{1}{2}|f|^2)\langle ::(\Delta\hat{E}_1)^{N-2}:: \rangle + \frac{N^{(4)}}{2!}(\tfrac{1}{2}|f|^2)^2\langle ::(\Delta\hat{E}_1)^{N-4}:: \rangle + \cdots + (N-1)!!\,|f|^N \quad (N \text{ even}) , \tag{27}$$

where :: :: denotes normal ordering with respect to $\hat{A},\hat{A}^\dagger$ operators and plays the same role for $\hat{A},\hat{A}^\dagger$ as does : : for $\hat{a},\hat{a}^\dagger$. Now for the state $|v,z\rangle$ we have with the help of the eigenvalue Eq. (20) and its conjugate for any $n\neq 0$,

$$\langle v,z|::(\Delta\hat{E}_1)^n::|v,z\rangle=\langle v,z|::(\Delta\hat{A}f+\Delta\hat{A}^\dagger f^*)^n::|v,z\rangle$$
$$=0\,, \tag{28}$$

so that from Eqs. (27) and (28)

$$\langle v,z|(\Delta\hat{E}_1)^N|v,z\rangle=(N-1)!!\,|f|^N$$
$$=(N-1)!!\,|g|^N[\cosh(2\Theta)-\sinh(2\Theta)\cos(2\mathbf{k}\cdot\mathbf{r}-2\phi-\theta)]^{1/2N}\quad(N\text{ even})\,. \tag{29}$$

Actually this result also follows directly from the fact that $\hat{E}_1$ is a Gaussian variable in the state $|v,z\rangle$ (or in any other two-photon coherent state[3]) when we invoke the Gaussian moment theorem. If we compare this result with the definition (10) of Nth-order squeezing, where $C=|g|^2$, we see that the state $|v,z\rangle$ is squeezed to all orders N if there is some phase angle ϕ for which $|\cosh(2\Theta)-\sinh(2\Theta)\cos(2\mathbf{k}\cdot\mathbf{r}-2\phi-\theta)|<1$. In fact, one can always find ϕ to satisfy this inequality. If $\Theta>0$, we choose $2\mathbf{k}\cdot\mathbf{r}-2\phi-\theta=0$, in which case the factor within square brackets in Eq. (29) becomes $e^{-2\Theta}$, and if $\Theta<0$, we choose $2\mathbf{k}\cdot\mathbf{r}-2\phi-\theta=\pm\pi$, when the same factor becomes $e^{2\Theta}$. It follows that the state $|v,z\rangle$ is squeezed not merely to the second order as has long been known, but to all orders. The squeeze parameter $q_N=e^{-N|\Theta|}-1$, and this can be close to -1 when $|\Theta|\gg 1$.

There remains the question whether the squeezing is *intrinsically* of higher order, in the sense defined in Sec. II. By using the ordering relation (8) in reverse, we can derive the normally ordered moments $\langle:(\Delta\hat{E}_1)^N:\rangle$ from $\langle(\Delta\hat{E}_1)^N\rangle$ given by Eq. (29). Straightforward summation of a series like that in Eq. (9) leads to the result for $\Theta>0$,

$$\langle:(\Delta\hat{E}_1)^N:\rangle=(N-1)!!\,|g|^N(-1)^{N/2}(1-e^{-2\Theta})^{N/2}\quad(N\text{ even}) \tag{30}$$

and it can readily be verified that this satisfies Eqs. (9) and (29). It follows that there is intrinsic Nth order squeezing in this state for all values of N for which $\frac{1}{2}N$ is odd, viz. for $N=2,6,10,\ldots$.

IV. DEGENERATE PARAMETRIC DOWN-CONVERSION

As an application of these results to a physically realizable situation, we consider the process of degenerate parametric down-conversion, in which a strong, classical field of complex amplitude v and frequency 2ω is incident on a nonlinear crystal. The interaction effectively splits some photons of energy $2\hbar\omega$ into two photons of energy $\hbar\omega$. The simplest Hamiltonian for this problem has the quadratic form

$$\hat{H}=\hbar\omega\hat{n}+\hbar g(ve^{-2i\omega t}\hat{a}^{\dagger 2}+\text{H.c.})\,, \tag{31}$$

in which the interaction energy has the same general structure as appears in the exponent of $\hat{U}(z)$ in Eq. (14). We might therefore expect the foregoing conclusions to apply. The down-converted field is treated as a single mode and only this mode is quantized. The general solution of the Heisenberg equation of motion for $\hat{a}(t)$ has the form[5,32]

$$\hat{a}(t)=\hat{a}(0)\cosh(2g|v|t)e^{-i\omega t}-i\frac{v}{|v|}\hat{a}_1^\dagger(0)\sinh(2g|v|t)e^{-i\omega t}\,. \tag{32}$$

In analogy with Eq. (3), we then define the quadrature component $\hat{E}_1(t)$ by

$$\hat{E}_1(t)=\hat{a}(t)e^{i(\omega t-\phi)}+\hat{a}^\dagger(t)e^{-i(\omega t-\phi)}$$
$$=\hat{a}(0)\left[\cosh(2g|v|t)e^{-i\phi}+i\frac{v^*}{|v|}\sinh(2g|v|t)e^{i\phi}\right]$$
$$+\hat{a}^\dagger(0)\left[\cosh(2g|v|t)e^{i\phi}-i\frac{v}{|v|}\sinh(2g|v|t)e^{-i\phi}\right], \tag{33}$$

which has the structure of Eq. (3), but with an effective commutator

$$C=\cosh(4g|v|t)-\sinh(4g|v|t)\sin(2\phi-\arg v)\,. \tag{34}$$

Now if we take the initial quantum state of the down-converted light to be the vacuum, then all the normally ordered dispersions in Eq. (9) are zero, and if ϕ is chosen so that $2\phi-\arg v=\pi/2$, then $C=e^{-4g|v|t}$, and Eq. (9) leads to the result

$$\langle[\Delta\hat{E}_1(t)]^N\rangle=(N-1)!!e^{-2Ng|v|t},\quad(N\text{ even})\,. \tag{35}$$

As the right-hand side is less than $(N-1)!!$, which is the corresponding Nth order dispersion for a coherent state, we see that the down-converted light is squeezed to all orders. Moreover, for the same reason as in Sec. III, there is again intrinsic squeezing of order $n=2,6,10,\ldots$. This shows that the concept of higher-order squeezing is more than purely mathematical, but should have physically realizable manifestations, because down-conversion is observable experimentally.

V. SECOND HARMONIC GENERATION

To a certain extent this process is the inverse of the degenerate parametric down-conversion treated in the previous section. If the incident light is described as mode 1 of frequency ω, and the harmonically generated light as mode 2 with frequency 2ω, and there is parametric coupling between them provided by a nonlinear crystal, then we may write for the total energy,[17–21]

$$\hat{H}=\hbar\omega\hat{n}_1+2\hbar\omega\hat{n}_2+\hbar g(\hat{a}_2^\dagger\hat{a}_1^2+\text{H.c.})\ . \tag{36}$$

This leads to the coupled Heisenberg equations of motion

$$\dot{\hat{a}}_{1s}=-2ig\hat{a}_{1s}^\dagger\hat{a}_{2s}\ , \tag{37}$$

$$\dot{\hat{a}}_{2s}=-ig\hat{a}_{1s}^2\ , \tag{38}$$

where

$$\hat{a}_{1s}\equiv\hat{a}_1e^{i\omega t},\quad \hat{a}_{2s}\equiv\hat{a}_2e^{2i\omega t} \tag{39}$$

are slowly varying annihilation operators.

The general time-dependent solution of these equations of motion for $\hat{a}_{1s}(t)$ and $\hat{a}_{2s}(t)$ is complicated. However, as the interaction between the modes persists only for a short time t, which is of order of the propagation time of the light through the nonlinear crystal, we are justified in using short-time power-series expansions in t to solve the equations of motion, as was done by Kozierowski and Tanas.[33] If we write

$$\begin{aligned}\hat{a}_{1s}(t)&=\hat{a}_{1s}(0)+t\dot{\hat{a}}_{1s}(0)+\frac{t^2}{2!}\ddot{\hat{a}}_{1s}(0)+O((gt)^3)\ ,\\ \hat{a}_{2s}(t)&=\hat{a}_{2s}(0)+t\dot{\hat{a}}_{2s}(0)+\frac{t^2}{2!}\ddot{\hat{a}}_{2s}(0)+O((gt)^3)\ ,\end{aligned} \tag{40}$$

and substitute in Eqs. (37) and (38), we readily obtain

$$\begin{aligned}\hat{a}_{1s}(t)=\ &\hat{a}_1(0)-2igt\hat{a}_1^\dagger(0)\hat{a}_2(0)\\ &+2(gt)^2[\hat{n}_2(0)\hat{a}_1(0)-\tfrac{1}{2}\hat{n}_1(0)\hat{a}_1(0)]\\ &+O((gt)^3)\ ,\end{aligned} \tag{41}$$

$$\begin{aligned}\hat{a}_{2s}(t)=\ &\hat{a}_2(0)-igt\hat{a}_1^2(0)\\ &-2(gt)^2[\hat{n}_1(0)+\tfrac{1}{2}]\hat{a}_2(0)+O((gt)^3)\ ,\end{aligned} \tag{42}$$

and we can use these to calculate the moments of

$$\hat{E}_1(t)\equiv\hat{a}_{1s}(t)e^{-i\phi}+\hat{a}_{1s}^\dagger(t)e^{i\phi}\ . \tag{43}$$

For the initial state $|v\rangle_1|0\rangle_2$, in which mode 1 is in a coherent state and mode 2 is in the vacuum state, we have for the deviation $\Delta\hat{E}_1(t)\equiv\hat{E}_1(t)-\langle\hat{E}_1(t)\rangle$,

$$\begin{aligned}\Delta\hat{E}_1(t)=\ &[\hat{a}_1-v-2igt\hat{a}_1^\dagger\hat{a}_2+2(gt)^2(\hat{n}_2\hat{a}_1-\tfrac{1}{2}\hat{n}_1\hat{a}_1+\tfrac{1}{2}|v|^2v)]e^{-i\phi}\\ &+[\hat{a}_1^\dagger-v^*+2igt\hat{a}_2^\dagger\hat{a}_1+2(gt)^2(\hat{a}_1^\dagger\hat{n}_2-\tfrac{1}{2}\hat{a}_1^\dagger\hat{n}_1+\tfrac{1}{2}|v|^2v^*)]e^{i\phi}+O((gt)^3)\ ,\end{aligned} \tag{44}$$

where all operators without time arguments are understood to be zero-time operators. Then we obtain for the normally ordered moments, for any even N,

$$\begin{aligned}\langle:[\Delta\hat{E}_1(t)]^N:\rangle&=\sum_{r=0}^{N}\begin{bmatrix}N\\ r\end{bmatrix}\langle[\hat{a}_1^\dagger-v^*+2igt\hat{a}_2^\dagger\hat{a}_1+2(gt)^2(\hat{a}_1^\dagger\hat{n}_2-\tfrac{1}{2}\hat{a}_1^\dagger\hat{n}_1+\tfrac{1}{2}|v|^2v^*)]^r\\ &\qquad\times[\hat{a}_1-v-2igt\hat{a}_1^\dagger\hat{a}_2+2(gt)^2(\hat{n}_2\hat{a}_1-\tfrac{1}{2}\hat{n}_1\hat{a}_1+\tfrac{1}{2}|v|^2v)]^{N-r}\rangle e^{-i(N-2r)\phi}\\ &=-(gt)^2[v^2\langle(\hat{a}_1-v)^{N-1}(\hat{a}_1^\dagger-v^*)\rangle e^{-iN\phi}+v^{*2}\langle(\hat{a}_1-v)(\hat{a}_1^\dagger-v^*)^{N-1}\rangle e^{iN\phi}]+O((gt)^3)\ ,\end{aligned}$$

after making use of the fact that only the terms $r=0$ and $r=N$ make a contribution to the second order in gt. The remaining operator expectations are readily evaluated with the help of the commutation relation

$$[(\hat{a}_1-v),(\hat{a}_1^\dagger-v^*)^m]=m(\hat{a}_1^\dagger-v^*)^{m-1}\ ,$$

whose expectation vanishes unless $m=1$. We then arrive at the result

$$\begin{aligned}\langle:[\Delta\hat{E}_1(t)]^N:\rangle&=-(gt)^2\delta_{N2}|v_2e^{-2i\phi}+v^{*2}e^{2i\phi}|+O((gt)^3)\\ &=-2(gt)^2|v|^2\cos[2(\phi-\theta)]\delta_{N2}+O((gt)^3)\ ,\end{aligned} \tag{45}$$

where we have written $v=|v|e^{i\theta}$.

We now substitute this result in Eq. (9), and obtain for any even N,

$$\begin{aligned}\langle[\Delta\hat{E}_1(t)]^N\rangle&=(N-1)!!+\frac{N^{(N-2)}}{(\frac{1}{2}N-1)!}\left[\frac{1}{2}\right]^{N/2-1}\langle:[\Delta\hat{E}_1(t)]^2:\rangle\\ &=(N-1)!!-2\frac{N^{(N-2)}}{(\frac{1}{2}N-1)!}\left[\frac{1}{2}\right]^{N/2-1}(gt)^2|v|^2\cos[2(\phi-\theta)]+O((gt)^3)\ .\end{aligned} \tag{46}$$

The state is squeezed to order N whenever the second term is negative, and this will be the case if the phase angle ϕ is chosen so that $(\phi-\theta)=n\pi$, where n is any integer. It follows that the fundamental mode becomes squeezed to all even orders in the process of second harmonic generation, within the short time approximation. In particular

$$\begin{aligned}\langle[\Delta\hat{E}_1(t)]^2\rangle &= 1-2(gt)^2|v|^2\cos[2(\phi-\theta)]\\ &\quad +O((gt)^3)\,,\\ \langle[\Delta\hat{E}_1(t)]^4\rangle &= 3-12(gt)^2|v|^2\cos[2(\phi-\theta)]\\ &\quad +O((gt)^3)\,,\\ \langle[\Delta\hat{E}_1(t)]^6\rangle &= 15-90(gt)^2|v|^2\cos[2(\phi-\theta)]\\ &\quad +O((gt)^3)\,.\end{aligned} \tag{47}$$

From Eq. (46), the squeeze parameter q_N defined by Eq. (11) has the value $-N(gt)^2|v|^2$. However, by virtue of Eq. (45), we see that in this case there is no intrinsic squeezing beyond the second order, because all the higher normally ordered moments of $\Delta\hat{E}_1(t)$ are zero. The higher-order squeezing exhibited by Eq. (46) to the order $(gt)^2$ is simply a manifestation of intrinsic second-order squeezing.

VI. RESONANCE FLUORESCENCE FROM AN ATOM

It was first shown by Walls and Zoller[22] that the light emitted spontaneously by a two-level atom that is being coherently excited by a light beam near resonance is squeezed to the second order in $\hat{E}_1$. Here we wish to examine the possibility of higher-order squeezing in the same situation.

We consider a two-level atom with level spacing $\hbar\omega_0$ and atomic lowering operator $\hat{b}(t)$, that is interacting with a quantum field of frequency ω_1 in a coherent state $|\{v\}\rangle$. The atomic Rabi frequency Ω is a convenient measure of the amplitude of the exciting field, and we let 2β be the Einstein A coefficient for the transition. We shall denote the relative detuning $(\delta\omega+\omega_1-\omega_0)/\beta$ by θ, where $\delta\omega$ is the Lamb shift. Because the spontaneous emission from the atom has no well-defined direction or frequency, the radiation field has to be treated as a multimode system in this problem. In the following we shall make considerable use of results obtained in Ref. 34 for the resonance fluorescence of an atom.

If the atom is located at the origin, the positive frequency part $\hat{E}^{(+)}(\mathbf{r},t)$ of one polarization component of the electric field at position $\mathbf{r}$ at time t (with $r\gg c/\omega_0$) is given by[34,35]

$$\hat{E}^{(+)}(\mathbf{r},t)=K(\mathbf{r})\hat{b}(t-r/c)+\hat{E}^{(+)}_{\text{free}}(\mathbf{r},t)\,. \tag{48}$$

$\hat{E}^{(+)}_{\text{free}}(\mathbf{r},t)$ is the free-field or external part of $\hat{E}^{(+)}(\mathbf{r},t)$, whereas the first term is contributed by the atomic source. $K(\mathbf{r})$ is a geometric factor independent of the quantum state, that is given by[34]

$$K(\mathbf{r})=\frac{\omega_0^2}{4\pi\epsilon_0 c^2 r}\left[\boldsymbol{\mu}-\frac{(\boldsymbol{\mu}\cdot\mathbf{r})}{r^2}\mathbf{r}\right]\cdot\boldsymbol{\epsilon}^*\,, \tag{49}$$

where $\boldsymbol{\mu}$ is the atomic transition dipole moment, and $\boldsymbol{\epsilon}^*$ is the unit polarization vector characterizing the polarization that is being singled out. In the neighborhood of $\mathbf{r}$, $\hat{E}^{(+)}$ can be given a mode expansion as in Eq. (1), with $l(k)=(\hbar\omega/2\epsilon_0)^{1/2}$, and we define the measured real field $\hat{E}_1$ as in Eq. (3). We shall suppose that the exciting field, or the initial coherent state $|\{v\}\rangle$, is so chosen that the right eigenvalue of $\hat{E}^{(+)}_{\text{free}}$ vanishes at $\mathbf{r}$, t, or

$$\hat{E}^{(+)}_{\text{free}}(\mathbf{r},t)|\{v\}\rangle=0\,. \tag{50}$$

In calculating moments of $\hat{E}_1(\mathbf{r},t)$ with help of Eqs. (3) and (48), we encounter products of atomic and field operators at different times. It is then convenient to make use of the following commutation relation derived by Mollow[36]

$$[\hat{b}(t-r/c),\hat{E}^{(\pm)}_{\text{free}}(\mathbf{r},t)]=0\,, \tag{51}$$

which allows us to evaluate normally ordered moments of $\hat{E}_1$ as if the free-field operator were absent. We shall also use the result for the expectation of $\hat{b}(t)$ in the steady state[34] (χ is an arbitrary phase angle),

$$\langle\hat{b}(t)\rangle=-\frac{\frac{1}{2}(\Omega/\beta)(1+i\theta)}{\Omega^2/2\beta^2+1+\theta^2}e^{-i(\omega_1 t+\chi)}\,. \tag{52}$$

Then with the help of Eqs. (3), (48), and (50)–(52), we have for the deviation

$$\begin{aligned}\Delta\hat{E}_1(\mathbf{r},t)&=K\hat{b}\left[t-\frac{r}{c}\right]e^{i[\omega_1(t-r/c)-\phi]}+K^*\hat{b}^\dagger\left[t-\frac{r}{c}\right]e^{-i[\omega_1(t-r/c)-\phi]}+\left[K\frac{\frac{1}{2}(\Omega/\beta)(1+i\theta)}{\Omega^2/2\beta^2+1+\theta^2}e^{-i(\chi+\phi)}+\text{c.c.}\right]\\ &=|K|\left[\hat{b}\left[t-\frac{r}{c}\right]e^{i[\omega_1(t-r/c)-\phi+\arg K]}+\hat{b}^\dagger\left[t-\frac{r}{c}\right]e^{-i[\omega_1(t-r/c)-\phi+\arg K]}+\frac{(\Omega/\beta)(1+\theta^2)^{1/2}}{\Omega^2/2\beta^2+1+\theta^2}\cos\psi\right]\,,\end{aligned} \tag{53}$$

in which

$$\psi=\chi+\phi-\arg K-\tan^{-1}\theta\,. \tag{54}$$

It follows from the multinomial expansion that

$$\langle :[\Delta\hat{E}_1(\mathbf{r},t)]^N:\rangle = |K|^N \sum_{\substack{n=0 \\ (n+m \leq N)}} \sum_{m=0} \frac{N!}{n!m!(N-n-m)!} \left\langle \hat{b}^{\dagger n}\left[t-\frac{r}{c}\right]\hat{b}^{m}\left[t-\frac{r}{c}\right]\right\rangle e^{i(m-n)\omega_1(t-r/c)}$$

$$\times e^{-i(m-n)(\phi-\arg K)}\left[\frac{(\Omega/\beta)(1+\theta^2)^{1/2}}{\Omega^2/2\beta^2+1+\theta^2}\cos\psi\right]^{N-n-m} .$$

Now repeated products of $\hat{b}$ and $\hat{b}^\dagger$ operators vanish, so that the only nonzero contributions come from terms with $n,m=0,1$, and

$$\langle :[\Delta\hat{E}_1(\mathbf{r},t)]^N:\rangle = (N-1)|K|^N\left[\frac{(\Omega/\beta)(1+\theta^2)^{1/2}\cos\psi}{\Omega^2/2\beta^2+1+\theta^2}\right]^N\left[-1+N\left[\frac{\Omega^2/2\beta^2+1+\theta^2}{(\Omega/\beta)(1+\theta^2)^{1/2}\cos\psi}\right]^2\right.$$
$$\left.\times\left\langle \hat{b}^\dagger\left[t-\frac{r}{c}\right]\hat{b}\left[t-\frac{r}{c}\right]\right\rangle\right] .$$

The expected excited-state population in the steady state has been shown to be given by[34]

$$\left\langle \hat{b}^\dagger\left[t-\frac{r}{c}\right]\hat{b}\left[t-\frac{r}{c}\right]\right\rangle = \frac{\Omega^2/4\beta^2}{\Omega^2/2\beta^2+1+\theta^2} , \tag{55}$$

so that we obtain

$$\langle :[\Delta\hat{E}_1(\mathbf{r},t)]^N:\rangle = (N-1)|K|^N\left[\frac{(\Omega/\beta)(1+\theta^2)^{1/2}\cos\psi}{\Omega^2/2\beta^2+1+\theta^2}\right]^N\left[-1+\tfrac{1}{4}N\left[\frac{\Omega^2/2\beta^2+1+\theta^2}{(1+\theta^2)\cos^2\psi}\right]\right] . \tag{56}$$

Finally, we substitute this result in Eq. (9) in order to obtain $\langle[\Delta\hat{E}_1(\mathbf{r},t)]^N\rangle$ for even N. If each of the even normally ordered moments $\langle :(\Delta\hat{E}_1)^N:\rangle$ up to some value $N_{\max}$ were negative, then it is apparent from Eq. (9) that $\hat{E}_1$ would be squeezed to all orders up to $N_{\max}$. Then by choosing the phase angle ϕ so that $\psi=n\pi$ ($n=0,\pm1,\pm2,\ldots$) and making

$$\tfrac{1}{4}N\frac{\Omega^2/2\beta^2+1+\theta^2}{1+\theta^2}<1 ,$$

or

$$\Omega^2/2\beta^2<(4/N-1)(1+\theta^2) , \tag{57}$$

we ensure squeezing up to order N.

Condition (57) can readily be satisfied for $N=2$. However, it cannot be satisfied at all for larger even integers N, so that the squeezing in resonance fluorescence is intrinsically a second-order phenomenon. The higher normally ordered moments of $\Delta\hat{E}_1$ are all positive.

Nevertheless, higher-order squeezing is not necessarily ruled out if the second-order squeezing is strong enough. To illustrate this we calculate $\langle[\Delta\hat{E}_1(\mathbf{r},t)]^4\rangle$ from Eqs. (9) and (56). We find

$$\langle[\Delta\hat{E}_1(\mathbf{r},t)]^4\rangle = 3C^2+6C\langle :[\Delta\hat{E}_1(\mathbf{r},t)]^2:\rangle+\langle :[\Delta\hat{E}_1(\mathbf{r},t)]^4:\rangle$$
$$=3C^2+6C|K|^2\left[\frac{(\Omega/\beta)(1+\theta^2)^{1/2}\cos\psi}{\Omega^2/2\beta^2+1+\theta^2}\right]^2\left[-1+\frac{\Omega^2/2\beta^2+1+\theta^2}{2(1+\theta^2)\cos^2\psi}\right]$$
$$+3|K|^4\left[\frac{(\Omega/\beta)(1+\theta^2)^{1/2}\cos\psi}{\Omega^2/2\beta^2+1+\theta^2}\right]^4\left[-1+\frac{\Omega^2/2\beta^2+1+\theta^2}{(1+\theta^2)\cos^2\psi}\right] . \tag{58}$$

Let us choose $\psi=n\pi$, as before, and put

$$\Omega^2/2\beta^2=\alpha(1+\theta^2) , \tag{59}$$

with $\alpha<1$ to ensure that the second term is negative, although the third term is positive. Then

$$\langle[\Delta\hat{E}_1(\mathbf{r},t)]^4\rangle = 3C^2+6C|K|^2\frac{\alpha(\alpha-1)}{(\alpha+1)^2}+12|K|^4\frac{\alpha^3}{(\alpha+1)^4}$$
$$=3C^2+\frac{6|K|^2\alpha}{(\alpha+1)^2}\left[C(\alpha-1)+2|K|^2\frac{\alpha^2}{(\alpha+1)^2}\right] . \tag{60}$$

If α is sufficiently small, the positive term $2|K|^2\alpha^2/(\alpha+1)^2$ can be numerically smaller than the negative term $(\alpha-1)C$, with the result that $\langle[\Delta\hat{E}_1(\mathbf{r},t)]^4\rangle < 3C^2$. The field is then squeezed to the fourth order in $\hat{E}_1$. In general, the condition for fourth-order squeezing is

$$\frac{\alpha^2}{(1+\alpha)^2(1-\alpha)} < \frac{C}{2|K|^2} , \tag{61}$$

and the squeeze parameter q_4 is given by

$$q_4 = \frac{2|K|^2\alpha}{C(\alpha+1)^2}\left[\alpha-1+\frac{2|K|^2}{C}\frac{\alpha^2}{(\alpha+1)^2}\right] . \tag{62}$$

It is interesting to examine how easy it is to satisfy condition (61), by making an order-of-magnitude estimate of the right-hand side, with Eq. (49) for K and Eq. (12) for C. We may write approximately

$$|K|^2 \approx \frac{\omega_0^4|\mu|^2}{(4\pi\epsilon_0)^2c^4r^2} ,$$

and with the help of the well-known expression for the Einstein A coefficient

$$2\beta = \frac{1}{4\pi\epsilon_0}\frac{4|\mu|^2\omega_0^3}{3\hbar c^3}$$

we obtain

$$|K|^2 \approx \frac{1}{4\pi\epsilon_0}\frac{3}{2}\frac{\hbar\omega_0\beta}{cr^2} .$$

Hence

$$\frac{C}{2|K|^2} \approx \frac{2}{3}\pi\frac{\Delta\nu}{\beta}\left[\frac{r^2\Delta\Omega}{\lambda^2}\right] = \frac{2}{3}\pi\left[\frac{\Delta\nu}{\beta}\right]\left[\frac{A}{\lambda^2}\right] , \tag{63}$$

where A is the area of the detector aperture. As $A \gg \lambda^2$ usually and $\Delta\nu$ is typically much greater than β, the right-hand side is usually a large number, so that the condition (61) for fourth-order squeezing is easy to satisfy with almost any $\alpha < 1$. However, the degree of fourth-order squeezing is small, for we have approximately $q_4 \approx (2|K|^2/C)\alpha(\alpha-1)/(\alpha+1)^2$, which is very small numerically.

In a similar manner we find from Eq. (9), with $\psi = n\pi$,

$$\langle[\Delta\hat{E}_1(\mathbf{r},t)]^6\rangle = 15C^3 + |K|^6\left[45\frac{C^2}{|K|^4}\frac{\alpha(\alpha-1)}{(\alpha+1)^2} + 180\frac{C}{|K|^2}\frac{\alpha^3}{(\alpha+1)^4} + 20\frac{\alpha^3(3\alpha+1)}{(\alpha+1)^6}\right] , \tag{64}$$

and since $C/|K|^2 \gg 1$ the term in parentheses is again dominated by the negative first term with $\alpha < 1$, although the remaining terms are positive. It is therefore apparent that a small amount of squeezing is achievable to large even order N when α or Ω^2/β^2 is sufficiently small, because of the dominant effect of the negative $\langle:[\Delta\hat{E}_1(\mathbf{r},t)]^2:\rangle$ term. However, $\langle:[\Delta\hat{E}_1(\mathbf{r},t)]^2:\rangle$ is the only negative term in the expansion (9) for $\langle[\Delta\hat{E}_1(\mathbf{r},t)]^N\rangle$, so that squeezing in resonance fluorescence is intrinsically a second-order phenomenon.

VII. DISCUSSION

We have considered several different examples of systems that exhibit Nth-order squeezing with $N > 2$. They differ substantially in the degree of squeezing q_N achievable, and also in the extent to which the higher-order squeezing is intrinsic, or merely a manifestation of second-order squeezing. In degenerate parametric down-conversion there is squeezing to all orders N, with a squeeze parameter q_N that can be close to -1, and the squeezing is intrinsic to orders $2,6,10,14,\ldots$. In second harmonic generation within the short-time approximation [to order $(gt)^2$], there is a small amount of squeezing to all orders N, but it is intrinsic only for $N=2$. The higher moments $\langle:(\Delta\hat{E}_1)^N:\rangle$ all vanish within this approximation. Finally, in resonance fluorescence, there is again a small amount of higher-order squeezing, which is intrinsic only to order $N=2$. But this time the higher even moments $\langle:(\Delta\hat{E}_1)^N:\rangle$ are actually positive for $N > 2$. In a certain sense resonance fluorescence therefore exhibits the weakest form of higher-order squeezing.

Mathematically it is possible to construct states that are not squeezed to the second order, but are squeezed to higher order, although it is not at all clear whether they are physically realizable. For example, let us consider a single-mode field, let $|E_1\rangle$ be the eigenstate of $\hat{E}_1$,

$$\hat{E}_1|E_1\rangle = E_1|E_1\rangle , \tag{65}$$

and let $|\psi\rangle$ be the state defined by

$$|\psi\rangle = \int_{-\infty}^{\infty}\frac{(E_1-E_0)^{m/2}e^{-(E_1-E_0)^2/4\sigma^2}}{[\sqrt{2\pi}(m-1)!!\sigma^{m+1}]^{1/2}}|E_1\rangle dE_1 . \tag{66}$$

Here m is an even integer, and E_0 is the expectation of $\hat{E}_1$ in the state $|\psi\rangle$, which is normalized to unity. Then we readily find that for even N

$$\langle(\Delta\hat{E}_1)^N\rangle = \frac{(N+m-1)!!}{(m-1)!!}\sigma^N . \tag{67}$$

If we choose σ so that

$$(m+1)\sigma^2 = C , \tag{68}$$

where C is the commutator given by Eq. (2), then

$$\langle(\Delta\hat{E}_1)^2\rangle = C , \tag{69}$$

and by definition the state $|\psi\rangle$ is not squeezed to the second order in $\hat{E}_1$. However, the fourth moment is given by

$$\langle(\Delta\hat{E}_1)^4\rangle = (m+3)(m+1)\sigma^4 = \frac{(m+3)}{(m+1)}C^2 , \tag{70}$$

and this is smaller than $3C^2$, as required for fourth-order squeezing, for all even m. The squeeze parameter q_4 is given by $-2m/(3m+3)$, and ranges from $-\frac{4}{9}$ for $m=2$

to $-\frac{2}{3}$ for $m \rightarrow \infty$. In fact, it is not difficult to see that $\hat{E}_1$ is squeezed to all even orders other than 2 in the state $|\psi\rangle$. However, the physical significance of this state is obscure.

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[1]Squeezed states (not under that name) were encountered in connection with the problem of parametric amplification by B. R. Mollow and R. J. Glauber, Phys. Rev. **160**, 1076 (1967); **160**, 1097 (1967).

[2]D. Stoler, Phys. Rev. D **1**, 3217 (1970); **4**, 1925 (1971).

[3]H. P. Yuen, Phys. Rev. A **13**, 2226 (1976); H. P. Yuen and J. H. Shapiro, IEEE Trans. Inf. Theory **24**, 657 (1978); **26**, 78 (1980).

[4]For a recent review of squeezing see D. F. Walls, Nature **306**, 141 (1983).

[5]B. R. Mollow, Phys. Rev. **162**, 1256 (1967).

[6]G. J. Milburn and D. F. Walls, Opt. Commun. **39**, 401 (1981); Phys. Rev. A **27**, 392 (1983).

[7]K. Wódkiewicz and M. S. Zubairy, Phys. Rev. A **27**, 203 (1983).

[8]S. Friberg and L. Mandel, Opt. Commun. **48**, 439 (1984).

[9]G. Scharf and D. F. Walls, Opt. Commun. **50**, 245 (1984).

[10]J. Peřina, V. Peřinová, and J. Kadousec, Opt. Commun. **49**, 210 (1984); V. Peřinová and D. F. Tiebel, *ibid.* **50**, 401 (1984).

[11]H. J. Carmichael, G. J. Milburn, and D. F. Walls, J. Phys. A **17**, 469 (1984).

[12]H. P. Yuen and J. H. Shapiro, Opt. Lett. **4**, 334 (1979).

[13]R. S. Bondurant, P. Kumar, J. H. Shapiro, and M. Maeda, Phys. Rev. A **30**, 343 (1984).

[14]J. Peřina, V. Peřinová, C. Sibilia, and M. Bertolotti, Opt. Commun. **49**, 285 (1984).

[15]J. Jansky and Y. Yushin, Opt. Commun. **49**, 290 (1984).

[16]M. D. Reid and D. F. Walls (unpublished).

[17]L. Mandel, Opt. Commun. **42**, 437 (1982).

[18]L. A. Lugiato, G. Strini, and F. De Martini, Opt. Lett. **8**, 256 (1983).

[19]M. Kozierowski and S. Kielich, Phys. Lett. **94A**, 213 (1983).

[20]R. Tanaś and S. Kielich, Opt. Commun. **45**, 351 (1983); Opt. Acta **31**, 81 (1984).

[21]R. Loudon, Opt. Commun. **49**, 67 (1984).

[22]D. F. Walls and P. Zoller, Phys. Rev. Lett. **47**, 709 (1981).

[23]L. Mandel, Phys. Rev. Lett. **49**, 136 (1982).

[24]Z. Ficek, R. Tanaś, and S. Kielich, Opt. Commun. **46**, 23 (1984); Phys. Rev. A **29**, 2004 (1984).

[25]R. Loudon, Opt. Commun. **49**, 24 (1984).

[26]P. A. Lakshmi and G. S. Agarwal (unpublished).

[27]W. Becker, M. O. Scully, and M. S. Zubairy, Phys. Rev. Lett. **48**, 475 (1982).

[28]We label all Hilbert-space operators by the caret ^.

[29]A preliminary account of this work was given in Phys. Rev. Lett. **54**, 323 (1985).

[30]R. J. Glauber, Phys. Rev. **130**, 2529; **131**, 2766 (1963).

[31]See for example, J. R. Klauder and E. C. G. Sudarshan, *Fundamentals of Quantum Optics* (Benjamin, New York, 1968).

[32]See for example A. Yariv, *Quantum Electronics* (Wiley, New York, 1975).

[33]M. Kozierowski and R. Tanaś, Opt. Commun. **21**, 229 (1977).

[34]H. J. Kimble and L. Mandel, Phys. Rev. A **13**, 2123 (1976).

[35]B. R. Mollow, Phys. Rev. **188**, 1969 (1969).

[36]B. R. Mollow, J. Phys. A **8**, L130 (1975).