SQUEEZED STATES OF LIGHT IN THE SECOND AND THIRD HARMONIC GENERATED BY SELF-SQUEEZED LIGHT

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CONTENTS

I. INTRODUCTION

Squeezed states in optical fields are at present a very attractive problem for theorists and experimenters (extensive accounts of the literature can be found in review articles [1–4] and in special issues of journals [5, 6]).

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Possibilities of their generation have been found in many nonlinear processes, such as resonance fluorescence [7–11], parametric amplification [12–19], four-wave mixing [20–31], multiphoton absorption [32, 33], the Jaynes-Cummings model [34–37], parametric down-conversion, [38–40], nonlinear propagation of light, and the harmonics generation considered in this paper.

In Section II we give a short review of the research related to light propagation and second- and third-harmonic generation in a nonlinear medium, especially from the point of view of quantum effects. Section III contains fundamental information about the squeezed states of light (also referred to in this paper as "ordinary" squeezed states). Two models of light propagation in a nonlinear medium are discussed in Sections III and IV: an anharmonic oscillator model and a model based on the effective Hamiltonian of the system. We study the squeezing effect in both approaches, analyzing the exact solutions for the quadrature variances. These results are also compared with ordinary squeezing, and the classical description of light propagation is recalled.

In Section V second-harmonic generation is studied. The first part of this section contains the classical treatment of the phenomenon. In the second part squeezing is discussed on the basis of the approximate analytical results holding in the quantum description. Section IV presents the quantum description of third-harmonic generation. Using the approximate solutions for the variances of the quadrature operators we analyze the squeezing effect.

II. HISTORY AND PERSPECTIVES

The first observation of the second harmonic of a laser beam by Franken et al. [41] has become a landmark in nonlinear optics. The classical description of the effect is due to Armstrong et al. [42]. In the next few years scientists concentrated on finding nonclassical properties of light in nonlinear media. In 1970 Walls [43] showed that the intensity of the second-harmonic beam exhibits periodic behavior in the quantum treatment, unlike the classical approach. Two years later Crosignani et al. [44] proved the impossibility of complete vanishing of the fundamental field in second-harmonic generation. Next, the second harmonic [45], higher order harmonics [46] and subharmonic [47] were studied for their quantum statistical properties. Stolarov [48], Kozierowski and Tanaś [49], and Kielich et al. [50] have shown that if the incoming beam is in a coherent state the antibunching effect (see, for example, Refs. 51–53) occurs in harmonics generation. This effect was also studied in Refs. 54 and 55.

For experiments the possibility of obtaining steady states is essential. Hence, theorists have searched for the quantum effect in an optical cavity. McNeil et al. [56] forecasted the "self-pulsing" effect in the intensities of the second-harmonic and fundamental beam in a cavity. The antibunching effect and bistability in the subharmonic and second-harmonic generation in a Fabry-Pérot cavity system were analyzed by Drummond et al. [57–59].

The search for squeezing in harmonics generation began in 1982. The first results were due to Mandel [60]. Kozierowski and Kielich [61] and Kielich et al. [62, 63] proposed a more general description. Lugiato et al. [64] predicted this nonclassical effect in the fundamental and second-harmonic beam, generated in a nonlinear crystal in a cavity. Friberg and Mandel [65] found the possibility of generation of squeezed states via a combination of parametric down-conversion and second-harmonic generation.

At the same time Tanaś [66] proposed the anharmonic oscillator model to describe laser light propagating in a nonlinear medium, which gives a squeezing effect different from ordinary squeezing. Tanaś and Kielich [67, 68] proposed the name *self-squeezing* when analyzing the effective Hamiltonian of the system. Moreover, Kielich et al. [69] applied an external magnetic field along the direction of propagation to achieve control of the self-squeezing of light. The evolution of the field in a nonlinear medium was described by Milburn [70, 71], who used the quasiprobability function $Q(\alpha, \alpha^*, t)$. He succeeded in revealing its periodic behavior and the role of dissipation in the effect and predicted squeezing in this treatment. Yurke and Stoler [72] proved that the state produced in the anharmonic oscillator model can be a superposition of a finite number of coherent states.

In 1985 Hong and Mandel [73] introduced the definition of higher-order squeezing and, in particular, studied second-harmonic generation. Kozierowski [74] searched for this effect in nth-harmonic generation. The squeezing of the square of the amplitude in second-harmonic generation was analyzed by Hillery [75], who predicted this kind of squeezing in the fundamental beam. He also found correlation between ordinary squeezing in the harmonic beam and amplitude-squared squeezing in the fundamental. Amplitude-squared squeezing was observed by Sizman et al. [76], who measured a 40% reduction of noise.

Correlation between the fundamental field and the harmonic beam was searched for in Lukš et al. [77]. The possibility of producing squeezed states in the fundamental stimulated by multiple higher-harmonic generation was proposed in Chmela et al. [78]. Kielich et al. analyzed the second harmonic [79] and third harmonic [80] generated by self-squeezed light.

Ekert and Rzążewski [81] found that the intensity of the second harmonic depends on the kind of fundamental beam. The state of the fundamental field in second-harmonic generation has been studied from the point of view of the initial phase [82]. In 1988 Pereira et al. [83] observed squeezing in the fundamental beam in second-harmonic generation. His result has been compared with the theoretical description [84].

The anharmonic oscillator model has been studied extensively at the same time. Lukš et. al. [85] defined "principal squeezing" related to the geometrical representation of the quadrature components as an ellipse. Loudon [86] proposed a different representation by Booth's elliptical lemniscate. Both representations were compared by Tanaś et al. [87]. They also proved that "crescent" squeezing, introduced by Kitagawa and Yamamoto [88] and Yamamoto et al. [89] (see also [4]), is the same as self-squeezing. Using the quasiprobability function Miranowicz et al. [90] proved the generation of superpositions of coherent states in the anharmonic oscillator model.

Recently light propagation in a nonlinear medium has been studied from the point of view of second- and fourth-order squeezing [91], the saturation effect [92], and the effect of dispersion [93]. It has been found that squeezing decreases with increasing saturation parameter and can be produced only within a limited frequency interval, determined by dispersion in the medium. Tanaś and Kielich [94] considered the role played by higher-order nonlinearity in the self-squeezing of light.

The theoretical basis for experiments has been prepared in recent years. The possibility of producing squeezed states in nth-harmonic generation in a laser resonator was proposed by Gorbaczev and Polzik [95]. Schack et al. [96] described a method of a doubly resonant cavity containing a laser medium as well as a χ^2 nonlinearity. They have predicted more than 60% squeezing in the up-converted mode. The "input–output" theory has been used by Collett and Levien [97] to generate the second harmonic. They obtained 50% squeezing. An analysis of the resonator parameters was given in Ref. 98. In 1991 You-bang Zhan [99, 100] studied in detail amplitude-cubed squeezing in second- and third-harmonic generation and amplitude-squared squeezing in nth-harmonic generation.

Quantum fluctuations in the Stokes operators of elliptically polarized light propagating in a Kerr medium were discussed by Tanaś and Kielich [101], who treated the medium as optically transparent, and by Tanaś and Gantsog [102] for a medium with dissipation. For the two-mode case, the influence of losses and noise was discussed by Horak and Peřina [103]. The influence of dissipation on the dynamics of the anharmonic oscillator, i.e., the one-mode propagation problem, was considered by Milburn and

Holmes [71], and recently the exact solutions of the master equation for the system have been discussed [104–107].

On the basis of the Pegg-Barnett formalism [108–110] the theorists have attempted to search for quantum phase properties in nonlinear processes. Tanaś et al. [111] analyzed the superposition of coherent states in the anharmonic oscillator model, using the quasiprobability function as well as the probability phase distribution $P(\theta)$. Quantum phase fluctuations in nonlinear processes are discussed in Refs. [112–114].

III. SQUEEZED STATES OF LIGHT

A. Minimum Uncertainty States and Coherent States

The Heisenberg uncertainty principle limits the possibility of measuring two observables in the same state. The variances of two observables A, B satisfy the following relation:

$$\left\langle (\Delta \hat{A})^2 \right\rangle \left\langle (\Delta \hat{B})^2 \right\rangle \geq \tfrac{1}{4} \left| \left\langle [\hat{A}, \hat{B}] \right\rangle \right|^2 \tag{1}$$

If the sign of equality holds in (1), the state is referred to as the "minimum uncertainty state."

It is well known that a single mode of an electromagnetic field in a cavity can be treated as a simple harmonic oscillator, described by the "position" and "momentum" operators. These are related to the electric and magnetic components of light. In the simplest case of a one-dimensional cavity with z axis, on the assumption of linear polarization, we can write

$$\begin{aligned} \hat{E}(z,t) &= \left(\frac{2\omega^2}{\varepsilon_0 V} \right)^{1/2} \hat{q}(t) \sin kz \\ \hat{H}(z,t) &= \left(\frac{2\varepsilon_0 c^2}{V} \right)^{1/2} \hat{p}(t) \cos kz \end{aligned} \tag{2}$$

where $\hat{E}(z,t)$ is the electric field operator, $\hat{H}(z,t)$ is the magnetic operator, $\hat{q}(t)$ is the position operator, $\hat{p}(t)$ is the momentum operator, ω is the frequency of the mode under consideration, and k is the wave vector. The commutation relation for $\hat{q}$ and $\hat{p}$ is defined as

$$[\hat{p}(t), \hat{q}(t)] = -i\hbar \tag{3}$$

Let us consider the photon number states space. When describing the electric field in the states it is helpful to introduce the annihilation $\hat{a}$ and creation $\hat{a}^+$ operators, which obey the following relations:

$$\begin{aligned} \hat{a} &= (2\hbar\omega)^{-1/2}(\omega\hat{q} + i\hat{p}) \\ \hat{a}^+ &= (2\hbar\omega)^{-1/2}(\omega\hat{q} - i\hat{p}) \\ [\hat{a}, \hat{a}^+] &= 1 \end{aligned} \tag{4}$$

According to Eqs. (4) the electric field can be written as

$$\hat{E}(z,t) = C[\hat{a}(t) + \hat{a}^+(t)] \tag{5}$$

where $C = (\hbar\omega/\varepsilon_0 V)^{1/2} \sin kz$, $\hat{a}(t) = \hat{a}(0)\exp(-i\omega t)$. The annihilation and creation operators act on the photon number state as follows:

$$\begin{aligned} \hat{a}|n\rangle &= n^{1/2}|n-1\rangle \\ \hat{a}^+|n\rangle &= (n+1)^{1/2}|n+1\rangle \end{aligned} \tag{6}$$

Hence, the number state $|n\rangle$ can be created from the vacuum state $|0\rangle$:

$$|n\rangle = (n!)^{1/2}(\hat{a}^+)^n|0\rangle \tag{7}$$

According to Eqs. (6), the average values of the position and momentum equal zero:

$$\langle\hat{q}\rangle = \langle\hat{p}\rangle = 0 \tag{8}$$

but their variances do not:

$$\begin{aligned} \left\langle(\Delta\hat{q})^2\right\rangle &= \frac{\hbar}{2\omega}\langle(\hat{a} + \hat{a}^+)(\hat{a} + \hat{a}^+)\rangle = \frac{\hbar}{2\omega}(1 + 2n) \\ \left\langle(\Delta\hat{p})^2\right\rangle &= \frac{\hbar\omega}{2}(1 + 2n) \end{aligned} \tag{9}$$

So, using expression (3), we write the uncertainty relation (1) for $\hat{p}$ and $\hat{q}$ as follows:

$$\left\langle(\Delta\hat{q})^2\right\rangle\left\langle(\Delta\hat{p})^2\right\rangle > \frac{\hbar^2}{4} \qquad (n > 0) \tag{10}$$

and

$$\langle(\Delta\hat{q})_0^2\rangle\langle(\Delta\hat{p})_0^2\rangle = \frac{\hbar^2}{4} \qquad (n = 0) \tag{11}$$

These equations mean that only the vacuum state is the minimum uncertainty state among the photon number states.

In 1963 Glauber introduced coherent states, which are the eigenstates for the annihilation operator [115]:

$$\begin{aligned} \hat{a}|\alpha\rangle &= \alpha|\alpha\rangle \\ \langle\alpha|\hat{a}^+ &= \langle\alpha|\alpha^* \end{aligned} \tag{12}$$

where $\alpha = |\alpha|\exp(i\phi)$. The coherent state can be constructed from the number states:

$$|\alpha\rangle = \exp\left(-\frac{|\alpha|^2}{2}\right)\sum_{n=0}^{\infty}\frac{\alpha^n}{(n!)^{1/2}}|n\rangle \tag{13}$$

These states are characterized by the Poisson photon-number distribution:

$$|\langle n|\alpha\rangle|^2 = \exp(-|\alpha|^2)\frac{\alpha^{2n}}{n!} \tag{14}$$

The average value of the photon number takes the form

$$\langle\alpha|\hat{n}|\alpha\rangle = |\alpha|^2$$

and its variance

$$\begin{aligned} \langle\alpha|(\Delta\hat{n})^2|\alpha\rangle &= |\alpha|^2 \\ \langle(\Delta\hat{n})^2\rangle &= \langle\hat{n}\rangle \end{aligned} \tag{15}$$

Hence, the variance is equal to the average value of the photon number in this case. Taking into account Eqs. (4) and (12), the expectation values of the position and momentum operators are given as follows:

$$\begin{aligned} \langle\alpha|\hat{q}|\alpha\rangle &= \left(\frac{\hbar}{2\omega}\right)^{1/2}(\alpha + \alpha^*) \\ \langle\alpha|\hat{p}|\alpha\rangle &= -i\left(\frac{\hbar\omega}{2}\right)^{1/2}(\alpha - \alpha^*) \end{aligned} \tag{16}$$

and for their squares we have

$$\begin{aligned}\langle\alpha|\hat{q}^2|\alpha\rangle &= \left(\frac{\hbar}{2\omega}\right)\left[\alpha^2 + (\alpha^*)^2 + 2|\alpha|^2 + 1\right]\\ \langle\alpha|\hat{p}^2|\alpha\rangle &= \left(\frac{\hbar\omega}{2}\right)\left[-\alpha^2 - (\alpha^*)^2 + 2|\alpha|^2 + 1\right]\end{aligned} \tag{17}$$

Thus, we have the following variances:

$$\begin{aligned}\left\langle(\Delta\hat{q})^2\right\rangle &= \frac{\hbar}{2\omega}\\ \left\langle(\Delta\hat{p})^2\right\rangle &= \frac{\hbar\omega}{2}\end{aligned} \tag{18}$$

It is easy to check that the left side of the uncertainty relation (1)

$$\left\langle(\Delta\hat{q})^2\right\rangle\left\langle(\Delta\hat{p})^2\right\rangle = \frac{\hbar^2}{4}$$

is equal to the right side,

$$\frac{1}{4}\left|\left\langle[\hat{p},\hat{q}]\right\rangle\right|^2 = \frac{\hbar^2}{4} \tag{19}$$

This proves that all coherent states are minimum uncertainty states.

The coherent state can be generated from the vacuum [115]

$$\begin{aligned}|\alpha\rangle &= \hat{D}(\alpha)|0\rangle\\ \hat{D}(\alpha) &= \exp(\alpha\hat{a}^+ - \alpha^*\hat{a})\end{aligned} \tag{20}$$

where $\hat{D}(\alpha)$ is the unitary displacement operator. The above operator transforms $\hat{a}$ and $\hat{a}^+$ as follows:

$$\begin{aligned}\hat{D}^+(\alpha)\hat{a}\hat{D}(\alpha) &= \hat{a} + \alpha\\ \hat{D}^+(\alpha)\hat{a}^+\hat{D}(\alpha) &= \hat{a}^+ + \alpha^*\end{aligned} \tag{21}$$

A more detailed discussion of coherent states is to be found in the review papers (for example, [116]).

B. Quadrature Operators

The annihilation and creation operators are non-Hermitian. It is useful to break them down into Hermitian quadrature operators [2, 3]:

$$\begin{aligned} \hat{Q} &= \hat{a} + \hat{a}^{+} \\ \hat{P} &= -\mathrm{i}(\hat{a} - \hat{a}^{+}) \end{aligned} \tag{22}$$

They satisfy the commutation relation

$$\left[\hat{Q}, \hat{P}\right] = 2\mathrm{i} \tag{23}$$

The uncertainty equation for them takes the form

$$\left\langle (\Delta\hat{Q})^{2}\right\rangle \left\langle (\Delta\hat{P})^{2}\right\rangle \geq 1 \tag{24}$$

Considering the minimum uncertainty state (coherent state or vacuum state) we obtain the variances equal to unity:

$$\left\langle (\Delta\hat{Q})^{2}\right\rangle = \left\langle (\Delta\hat{P})^{2}\right\rangle = 1$$

and

$$\left\langle (\Delta\hat{Q})^{2}\right\rangle \left\langle (\Delta\hat{P})^{2}\right\rangle = 1 \tag{25}$$

In terms of the quadrature operators (22), the electric field can be written as

$$\hat{E}(z,t) = C\left[\hat{Q}\cos\omega t + \hat{P}\sin\omega t\right] \tag{26}$$

Hence, $\hat{Q}$ and $\hat{P}$ may be identified with the amplitudes of the two quadrature phases of the electric field [117].

The displacement operator (20) for the quadrature components,

$$\hat{D}(\alpha) = \exp\left[\mathrm{i}\left(\operatorname{Im}\alpha\hat{Q} - \operatorname{Re}\alpha\hat{P}\right)\right] \tag{27}$$

transforms them in the following way:

$$\begin{aligned} \hat{D}^{+}(\alpha)\hat{Q}\hat{D}(\alpha) &= \hat{Q} + 2\operatorname{Re}\alpha \\ \hat{D}^{+}(\alpha)\hat{P}\hat{D}(\alpha) &= \hat{P} + 2\operatorname{Im}\alpha \end{aligned} \tag{28}$$

C. Squeezed States

The variances of the quadrature operators are equal (25) for a coherent state. However, one can imagine that one of them has a value below unity but together with the second variance satisfies the uncertainty relation (25). So the following generalized definition for the quadrature operators can be introduced:

$$\begin{aligned} \hat{Q}_s &= \hat{Q}\exp(-s) \\ \hat{P}_s &= \hat{P}\exp(s) \end{aligned} \tag{29}$$

where s is called the squeezing parameter. According to Eqs. (29), the quadrature variances have the modified form:

$$\begin{aligned} \left\langle \left(\Delta\hat{Q}_s\right)^2 \right\rangle &= \exp(-2s) \\ \left\langle \left(\Delta\hat{P}_2\right)^2 \right\rangle &= \exp(2s) \end{aligned} \tag{30}$$

and the annihilation and creation operators, connected with them (22), take the new form

$$\begin{aligned} \hat{a}_s &= \hat{a}\cosh s - \hat{a}^+ \sinh s \\ \hat{a}_s^+ &= \hat{a}^+\cosh s - \hat{a}\sinh s \end{aligned} \tag{31}$$

These generalized operators remain in commutation relations:

$$\begin{aligned} \left[\hat{Q}_s, \hat{P}_s\right] &= 2\mathrm{i} \\ \left[\hat{a}_s, \hat{a}_s^+\right] &= 1 \end{aligned} \tag{32}$$

The displacement operator has to be redefined:

$$\begin{aligned} \hat{D}_s(\alpha) &= \exp\left[\alpha\hat{a}_s^+ - \alpha^*\hat{a}_s\right] \\ \hat{D}_s(\alpha) &= \exp\left[\mathrm{i}\left(\mathrm{Im}\,\alpha\hat{Q}_s - \mathrm{Re}\,\alpha\hat{P}_s\right)\right] \end{aligned} \tag{33}$$

If $s > 0$ the exponential, in Eqs. (29), compresses the original variance of $\hat{Q}$ and expands the original variance of $\hat{P}$. The squeezing condition for one

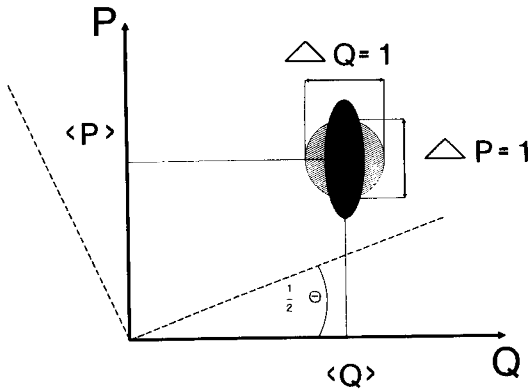

Figure 1. The error countours in the quadrature components plane for the coherent state (circle) and the squeezed state (ellipse) when $\theta = 0$. If $\theta \neq 0$ the error ellipse is inclined at angle $\theta/2$ (dashed axes). We denote $\Delta\hat{X} = \langle(\Delta\hat{X})^2\rangle^{1/2}$ for $\hat{X} = \hat{Q}, \hat{P}$.

of the quadrature components ($\hat{X} = \hat{Q}, \hat{P}$) is given as

$$\left\langle(\Delta\hat{X})^2\right\rangle < 1 \tag{34}$$

In the $\hat{Q}, \hat{P}$ plane the circular error contour for the coherent state is squeezed into an elliptical error contour (Fig. 1).

So far we have discussed the simplest case of squeezing, occurring along the $\hat{Q}$ and $\hat{P}$ component. To obtain squeezed states in general the squeeze operator has to be used [2, 3]:

$$\hat{S}(\zeta) = \exp\left[\tfrac{1}{2}\left(\zeta^*\hat{a}^2 - \zeta\hat{a}^{+2}\right)\right] \tag{35}$$

where $\zeta = s\exp(i\theta)$, $0 \le \theta \le 2\pi$, and $0 \le s \le \infty$. This operator squeezes the error contour in directions inclined at angles $\theta/2$ to the $\hat{Q}$ and $\hat{P}$ axes (dashed line in Fig. 1).

The squeeze operator (35) transforms the $\hat{a}$ and $\hat{a}^+$ operators in the following manner:

$$\begin{aligned} \hat{S}^+(\zeta)\hat{a}\hat{S}(\zeta) &= \hat{a}\cosh s - \hat{a}^+\exp(i\theta)\sinh s \\ \hat{S}^+(\zeta)\hat{a}^+\hat{S}(\zeta) &= \hat{a}^+\cosh s - \hat{a}\exp(-i\theta)\sinh s \end{aligned} \tag{36}$$

Using the squeeze operator we can define the squeezed states,

$$|\alpha,\zeta\rangle = \hat{D}(\alpha)\hat{S}(\zeta)|0\rangle \tag{37}$$

This definition (37) of squeezed states was proposed by Caves [118]. Yuen has given an alternative definition [119]:

$$|\beta,\mu,\nu\rangle = \hat{U}\hat{D}(\beta)\ |0\rangle \tag{38}$$

where $\hat{U}$ is the squeeze operator and $\hat{D}(\beta)$ the displacement operator. The two formalisms of squeezed states are equivalent [2],

$$\begin{aligned} \beta &= \mu\alpha + \nu\alpha^* \\ \mu &= \cosh s \\ \nu &= \exp(i\theta)\sinh s \end{aligned} \tag{39}$$

D. Fundamental Properties of Squeezed States

In this section some useful properties of the one-mode squeezed state are discussed.

- The average values of the annihilation and creation operators are

$$\begin{aligned} \langle\hat{a}\rangle &= \alpha \\ \langle\hat{a}^+\rangle &= \alpha^* \end{aligned} \tag{40}$$

  They do not depend on the squeezing parameter ζ.
- The average photon number is

$$\langle\hat{n}\rangle = \langle\hat{a}^+\hat{a}\rangle = |\alpha|^2 + \sinh^2 s \tag{41}$$

  The second term arises from the process of squeezing the vacuum.
- The eigenvalues of the quadrature operators are

$$\begin{aligned} \langle\hat{Q}\rangle &= \alpha + \alpha^* = 2\,\mathrm{Re}\,\alpha \\ \langle\hat{P}\rangle &= 2\,\mathrm{Im}\,\alpha \end{aligned} \tag{42}$$

  They also are independent of the squeezing parameter ζ.
- The variances of the quadrature operators are as follows:

$$\begin{aligned} \left\langle(\Delta\hat{Q})^2\right\rangle &= \exp(-2s)\cos^2(\tfrac{1}{2}\theta) + \exp(2s)\sin^2(\tfrac{1}{2}\theta) \\ \left\langle(\Delta\hat{P})^2\right\rangle &= \exp(-2s)\sin^2(\tfrac{1}{2}\theta) + \exp(2s)\cos^2(\tfrac{1}{2}\theta) \end{aligned} \tag{43}$$

  The variances are not dependent on the coherent amplitude α.

- The uncertainty relation (1) for the quadrature operators is

$$\left\langle (\Delta\hat{Q})^2 \right\rangle \left\langle (\Delta\hat{P})^2 \right\rangle = \cosh^2 2s \sin^2\theta + \cos^2\theta$$

It is obvious that the minimum quantum noise occurs for $\theta = \theta, \pi$:

$$\left\langle (\Delta\hat{Q})^2 \right\rangle \left\langle (\Delta\hat{P})^2 \right\rangle = 1$$

and the maximum for $\theta = \pi/2, 3\pi/2$:

$$\left\langle (\Delta\hat{Q})^2 \right\rangle \left\langle (\Delta\hat{P})^2 \right\rangle = \cosh^2 2s \tag{44}$$

- The condition for squeezing in the $\hat{Q}$ component is

$$\cos\theta > \tanh s$$

Therefore,

$$\left\langle (\Delta\hat{Q})^2 \right\rangle = \exp(-2s) \qquad (\text{for } \theta = 0) \tag{45}$$

- The condition for squeezing in the $\hat{P}$ component is

$$\cos\theta < -\tanh s$$

Therefore

$$\left\langle (\Delta\hat{P})^2 \right\rangle = \exp(-2s) \qquad \left(\text{for } \theta = \frac{\pi}{2}\right) \tag{46}$$

E. Two-Mode Squeezing

Obviously, it is possible to define squeezing not only for the one-mode case. If we consider light in two different frequencies ω_+ and ω_- it is useful to introduce two-mode squeezed states, which can be obtained from the vacuum state [2, 3],

$$|\alpha_+, \alpha_-, \zeta\rangle = \hat{D}_+(\alpha_+)\hat{D}_-(\alpha_-)\hat{S}(\zeta)|0\rangle$$

where the displacement operators are defined as

$$\hat{D}_\pm(\alpha_\pm) = \exp\left(\alpha_\pm \hat{a}_\pm^+ - \alpha_\pm^* \hat{a}_\pm\right)$$

and the squeeze operator is

$$\hat{S}(\zeta) = \exp\left(\zeta^* \hat{a}_+ \hat{a}_- - \zeta \hat{a}_+^+ \hat{a}_-^+\right) \tag{47}$$

This two-mode squeeze operator transforms the annihilation and creation operators [2, 117]:

$$\begin{aligned} \hat{S}^+(\zeta)\hat{a}_\pm \hat{S}(\zeta) &= \hat{a}_\pm \cosh s - \hat{a}_\mp^+ \exp(i\theta)\sinh s \\ \hat{S}^+(\zeta)\hat{a}_\pm^+ \hat{S}(\zeta) &= \hat{a}_\pm^+ \cosh s - \hat{a}_\mp \exp(-i\theta)\sinh s \end{aligned} \tag{48}$$

Similarly to the one-mode case, two-mode quadrature operators can be defined similarly to one-mode operators:

$$\begin{aligned} \hat{Q} &= \frac{1}{\sqrt{2}}\left(\hat{a}_+ + \hat{a}_+^+ + \hat{a}_- + \hat{a}_-^+\right) \\ \hat{P} &= \frac{-i}{\sqrt{2}}\left(\hat{a}_+ - \hat{a}_+^+ + \hat{a}_- - \hat{a}_-^+\right) \end{aligned} \tag{49}$$

Because of the usefulness of two-mode squeezed states, in the next sections we given some of their more important properties:

$$\langle \hat{a}_\pm \rangle = \alpha_\pm \tag{50}$$

$$\langle \hat{n}_\pm \rangle = |\alpha_\pm|^2 + \sinh^2 s \tag{51}$$

$$\left\langle \hat{a}_\pm^+ \hat{a}_\mp \right\rangle = \alpha_\pm^* \alpha_\mp \tag{52}$$

$$\langle \hat{a}_\pm \hat{a}_\pm \rangle = \alpha_\pm^2 \tag{53}$$

$$\langle \hat{a}_\pm \hat{a}_\mp \rangle = \alpha_+ \alpha_- - \exp(i\theta)\sinh s \cosh s \tag{54}$$

and for the quadrature operators:

$$\begin{aligned} \langle \hat{Q} \rangle &= 2^{1/2}(\operatorname{Re}\alpha_+ + \operatorname{Re}\alpha_-) \\ \langle \hat{P} \rangle &= 2^{1/2}(\operatorname{Im}\alpha_+ + \operatorname{Im}\alpha_-) \end{aligned} \tag{55}$$

$$\begin{aligned} \left\langle (\Delta\hat{Q})^2 \right\rangle &= \exp(-2s)\cos^2\left(\tfrac{1}{2}\theta\right) + \exp(2s)\sin^2\left(\tfrac{1}{2}\theta\right) \\ \left\langle (\Delta\hat{P})^2 \right\rangle &= \exp(-2s)\sin^2\left(\tfrac{1}{2}\theta\right) + \exp(2s)\cos^2\left(\tfrac{1}{2}\theta\right) \end{aligned} \tag{56}$$

The variances of the quadrature operators for two-mode states (56) are identical with the variances for one mode. This means that they are independent of the number of modes in the field.

In this section the theory of the squeezed states is only touched on. We have left out an account of higher-order squeezing [3, 73] and the amplitude-squared squeezing defined by Hillery [75].

Squeezed states of light are not considered only theoretically. In recent years many experimental results have been reported [29, 30, 31, 40, 76, 83, 120, 121]. To measure the variance of a quadrature component of the field a special phase-sensitive method is needed. It has been shown that homodyne and heterodyne detections are suitable. The homodyne method is used for a single quadrature measurement and the heterodyne measures both. These methods are based on the interference of squeezed light with a coherent field.

In the next sections we discuss in detail the possibilities of generating squeezed states in the propagation of light and harmonics generation in a nonlinear medium.

IV. ANHARMONIC OSCILLATOR MODEL

The anharmonic oscillator is the simplest model for the description of interaction between quantum light and a nonlinear medium. It was proposed by Tanaś [66]. In spite of its simplicity, this model gives the possibility of obtaining exact analytical results which, among other things, show the dissimilarity between the squeezing process in light propagation and the ordinary squeezing, generated by the squeeze operator (35).

It is assumed that the well-known Hamiltonian of the anharmonic oscillator can describe, for example, a single mode of the field propagating through a nonlinear medium. Then the Hamiltonian takes the form [66]

$$\hat{H} = \hbar\omega\hat{a}^{+}\hat{a} + \tfrac{1}{2}\hbar\kappa\hat{a}^{+2}\hat{a}^{2} \tag{57}$$

where $\hat{a}, \hat{a}^{+}$ are the annihilation and creation operators of the mode, ω is the frequency of the mode, and κ is an anharmonicity parameter (real). It is necessary to know the time evolution of $\hat{a}$ and $\hat{a}^{+}$ to obtain information about the respective quantum effects. To attain this the Heisenberg equation is constructed:

$$\frac{\mathrm{d}\hat{a}}{\mathrm{d}t} = \frac{1}{\mathrm{i}\hbar}[\hat{a}, \hat{H}] \tag{58}$$

According to the Hamiltonian (57) the equation of motion has the following form:

$$\frac{d\hat{a}}{dt} = -i(\omega + \kappa \hat{a}^{+}\hat{a})\hat{a} \tag{59}$$

Since the number-photon operator $\hat{n} = \hat{a}^{+}\hat{a}$ is a constant of motion,

$$[\hat{n}, \hat{H}] = 0 \tag{60}$$

it is possible to obtain the solution in the form

$$\hat{a}(t) = \exp\{-it[\omega + \kappa \hat{a}^{+}(0)\hat{a}(0)]\}\hat{a}(0) \tag{61}$$

where $\hat{a}(0), \hat{a}^{+}(0)$ are the annihilation and creation operators at $t = 0$. The term $\exp(-i\omega t)$ is associated with the free evolution of the system, whereas the second term comes from the nonlinear interaction included in the second part of the Hamiltonian (57). This exact operator solution (61) allows us to give all the characteristics of the field at the time t, if the state of the field at $t = 0$ is known.

We assume that the field is in a coherent state $|\alpha\rangle$ initially ($t = 0$). Since the photon number is a constant of motion, the photon-number distribution retains Poissonian statistics (14). This does not mean that the field has to be in a coherent state throughout its evolution. To search for squeezing we use the quadrature operators defined in (22). As was shown in Section III this effect occurs if one of the variances of the quadrature components has a value below unity:

$$\left\langle (\Delta\hat{Q})^2 \right\rangle < 1 \quad \text{or} \quad \left\langle (\Delta\hat{P})^2 \right\rangle < 1 \tag{62}$$

It is convenient to introduce normal ordering of the operators. Then we can write

$$\begin{aligned} \left\langle :(\Delta\hat{Q})^2: \right\rangle &= \left\langle (\Delta\hat{Q})^2 \right\rangle - 1 \\ \left\langle :(\Delta\hat{P})^2: \right\rangle &= \left\langle (\Delta\hat{P})^2 \right\rangle - 1 \end{aligned} \tag{63}$$

where the colons denote normal ordering. Here, the squeezing conditions take the following form:

$$\left\langle :(\Delta\hat{Q})^2: \right\rangle < 0 \quad \text{or} \quad \left\langle :(\Delta\hat{P})^2: \right\rangle < 0 \tag{64}$$

meaning that squeezing occurs when one of the normally ordered variances takes a negative value. In terms of the annihilation and creation operators these variances can be written as

$$\begin{aligned}\left\langle:(\Delta\hat{Q})^2:\right\rangle &= \left\langle(\Delta\hat{a})^2\right\rangle + \left\langle(\Delta\hat{a}^+)^2\right\rangle + 2(\langle\hat{a}^+\hat{a}\rangle - \langle\hat{a}^+\rangle\langle\hat{a}\rangle)\\ \left\langle:(\Delta\hat{P})^2:\right\rangle &= -\left\langle(\Delta\hat{a})^2\right\rangle - \left\langle(\Delta\hat{a}^+)^2\right\rangle + 2(\langle\hat{a}^+\hat{a}\rangle - \langle\hat{a}^+\rangle\langle\hat{a}\rangle)\end{aligned} \tag{65}$$

Using solution (61) in the equations above, the following results can be derived [66]:

$$\begin{aligned}\left\langle:(\Delta\hat{Q})^2:\right\rangle &= 2\,\mathrm{Re}\left[\alpha^2\exp\left[-i\tau + |\alpha|^2(\exp(-2i\tau) - 1)\right]\right]\\ &\quad - 2\,\mathrm{Re}\left[\alpha^2\exp\left[2|\alpha|^2(\exp(-i\tau) - 1)\right]\right]\\ &\quad + 2|\alpha|^2\left[1 - \exp\left[2|\alpha|^2(\cos\tau - 1)\right]\right]\\ \left\langle:(\Delta\hat{P})^2:\right\rangle &= -2\,\mathrm{Re}[\,\cdots\,] + 2\,\mathrm{Re}[\,\cdots\,] + 2|\alpha|^2[\,\cdots\,]\end{aligned} \tag{66}$$

where $\tau = \kappa t$; the brackets in the second equation (66) contain the same expressions as the first equation; α is the coherent amplitude, and $|\alpha|^2$ is the average number of photons. The variances of the quadrature operators (66) are plotted against $\beta = |\alpha|^2\tau$ in Fig. 2. We assumed that $\tau = 1 \times 10^{-6}$ and chose the initial phase to have α real. Both curves oscillate between

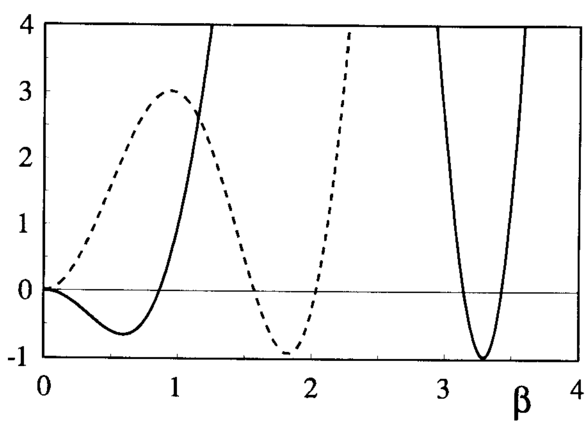

Figure 2. The variance of the $\hat{Q}$ quadrature component (solid line) and the variance of the $\hat{P}$ component (dashed line) are plotted versus $\beta = |\alpha|^2\tau$.

negative and positive values. The first minimum of the $\hat{Q}$ component (solid line) has a value of -0.66 and appears for $\beta = 0.6$. The second minimum is deeper and reaches -0.98. The first minimum of the $\hat{P}$ component (dashed line) occurs for $\beta = 1.82$ and has a value of -0.93. The next minimum is deeper and reaches 0.99. Note that if one of the variances is squeezed then the other is not.

This analysis supposes that a considerable amount of squeezing can be obtained for a large number of photons ($|\alpha|^2 \gg 1$). In this case we can assume that $|\alpha|^2\tau$ takes a value of the order of unity. Moreover, we can make the assumption that $\tau \ll 1$, because of the small value of the anharmonicity parameter κ. These assumptions allow us to expand equations (66) in power series and to retain only the leading terms. Hence we have the following approximate formulas for the quadrature variances:

$$\begin{aligned} \left\langle :(\Delta\hat{Q})^2: \right\rangle &\approx 2\beta[\beta - (\sin 2\beta + \beta\cos 2\beta)] \\ \left\langle :(\Delta\hat{P})^2: \right\rangle &\approx 2\beta[\beta + \sin 2\beta + \beta\cos 2\beta] \end{aligned} \tag{67}$$

These equations are simpler and we shall use them to compare the results obtained in the next section.

Formulas (66) and (67) mean that the states obtained in the anharmonic oscillator model do not preserve minimum uncertainty in the sense of the coherent states, i.e., fluctuations in one of the quadrature components of the field can be reduced.

We would like to emphasize that the solution (61) differs from the transformation (36) which defines ordinary squeezing.

In the next section it is shown that the result derived from the anharmonic oscillator is a particular case of a more general model.

V. SELF-SQUEEZING OF LIGHT IN NONLINEAR MEDIUM

A. Classical Treatment: Self-phase Modulation

Before giving a description of the squeezing effect in light propagation through a nonlinear medium, as proposed by Tanaś and Kielich [67, 68], it may be helpful to recall the classical treatment and some of its more interesting results. The classical approach is based on the assumption that the electric field is described by a vector E, which can be the sum of the positive and negative frequency parts at the time–space point $(\mathbf{r}, t)$:

$$E(\mathbf{r}, t) = E^{+}(\mathbf{r}, t) + E^{-}(\mathbf{r}, t) \tag{68}$$

The positive and negative parts can be written as

$$E^+(\mathbf{r},t) = \sum_i E^+(\omega_i)\exp\left[\mathrm{i}\left(\mathbf{k}_{\omega_i}\cdot\mathbf{r} - \omega_i t\right)\right]$$
$$E^-(\mathbf{r},t) = \sum_i E^-(\omega_i)\exp\left[-\mathrm{i}\left(\mathbf{k}_{\omega_i}\cdot\mathbf{r} - \omega_i t\right)\right] \tag{69}$$

where ω_i is the frequency and $\mathbf{k}_{\omega_i}$ the wave vector of ith mode. On taking into account one mode only, formulas (69) take the following form:

$$E^+(\mathbf{r},t) = E^+(\omega)\exp[\mathrm{i}(\mathbf{k}\cdot\mathbf{r} - \omega t)]$$
$$E^-(\mathbf{r},t) = E^-(\omega)\exp[-\mathrm{i}(\mathbf{k}\cdot\mathbf{r} - \omega t)] \tag{70}$$

We are interested in the interaction between the field and the nonlinear medium. It is contained in the time-averaged free energy [122],

$$F = -\tfrac{3}{4}\chi_{ijkl}E_i^-(\omega)E_j^-(\omega)E_k^+(\omega)E_l^+(\omega) + \text{c.c.} \tag{71}$$

where $E^{\pm}_{i,j,k,l}(\omega)$ are the components of the vector $E^{\pm}$, χ_{ijkl} is the fourth-rank tensor, describing the third-order nonlinear susceptibility, and the summation is defined by Einstein's convention. The free energy is the starting point for the calculation of the components of the nonlinear polarization vector at the frequency ω. They can be obtained from the well-known formula

$$P_i^{\pm}(\omega) = -\frac{\partial F}{\partial E_i^{\mp}} \tag{72}$$

Hence, on using Eq. 71, the vector components take the form

$$P_i^+(\omega) = 3\chi_{ijkl}(-\omega,-\omega,\omega,\omega)E_j^-(\omega)E_k^+(\omega)E_l^+(\omega)$$
$$P_i^-(\omega) = 3\chi_{ijkl}(-\omega,-\omega,\omega,\omega)E_j^-(\omega)E_k^-(\omega)E_l^+(\omega) \tag{73}$$

The above formulas do not include dissipative and resonant processes. In this case the following symmetry relation is fulfilled [122]:

$$\chi^*_{ijkl}(-\omega,-\omega,\omega,\omega) = \chi_{klij}(-\omega,-\omega,\omega,\omega) \tag{74}$$

On the assumption that the medium is isotropic and has a center of symmetry, it is possible to write the third-order susceptibility as follows [123]:

$$\chi_{ijkl}(-\omega,-\omega,\omega,\omega) = \chi_{xxyy}\delta_{ij}\delta_{kl} + \chi_{xyxy}\delta_{ik}\delta_{jl} + \chi_{xyyx}\delta_{il}\delta_{jk} \tag{75}$$

Moreover, this tensor is symmetrical in the pairs of indices i, j and k, l. So, instead of three, we have two independent components: χ_{xxyy}, and $\chi_{xyxy} = \chi_{xyyx}$.

Considering light propagation in an isotropic medium, it is of advantage to introduce a circular basis to describe the field and the polarization of the medium. When the field propagates along the z axis, then the right- and left-polarized components take the forms

$$E^+_{\pm} = 2^{-1/2}\left[E^+_x(\omega) \mp \mathrm{i}E^+_y(\omega)\right] \tag{76}$$

Using formulas (74)–(76) we obtain the following equation for the average free energy (71):

$$\begin{aligned} F = -\tfrac{1}{2}\Big\{ g^{\omega}_1\left[E^-_+(\omega)^2 E^+_+(\omega)^2 + E^-_-(\omega)^2 E^+_-(\omega)^2\right] \\ +4g^{\omega}_2\left[E^-_+(\omega)E^-_-(\omega)E^+_+(\omega)E^+_-(\omega)\right]\Big\} \end{aligned} \tag{77}$$

where the nonlinear coupling parameters $g^{\omega}_1, g^{\omega}_2$ are defined as follows:

$$\begin{aligned} g^{\omega}_1 &= 6\chi_{xyxy}(-\omega, -\omega, \omega, \omega) \\ g^{\omega}_2 &= 3\left[\chi_{xxyy}(-\omega, -\omega, \omega, \omega) + \chi_{xyxy}(-\omega, -\omega, \omega, \omega)\right] \end{aligned} \tag{78}$$

Applying Eqs. (72) and (77), we write the components of the nonlinear polarization vector in the new representation as

$$P^+_{\pm}(\omega) = \left[g^{\omega}_1|E^-_{\pm}(\omega)|^2 + 2g^{\omega}_2|E^-_{\mp}(\omega)|^2\right]E^+_{\pm}(\omega) \tag{79}$$

This expression can be inserted into the Maxwell wave equation. In the slowly varying amplitude approximation the following equation is obtained [122]:

$$\frac{\mathrm{d}E^+_{\pm}}{\mathrm{d}z} = \mathrm{i}\frac{2\pi\omega}{n_{\omega}c}P^+_{\pm} \tag{80}$$

On insertion of (79) into (80) we have

$$\frac{\mathrm{d}E^+_{\pm}(\omega, z)}{\mathrm{d}z} = \mathrm{i}\frac{2\pi\omega}{n_{\omega}c}\left[g^{\omega}_1|E^-_{\pm}(\omega)|^2 + 2g^{\omega}_2|E^-_{\mp}(\omega)|^2\right]E^+_{\pm}(\omega) \tag{81}$$

where $n_\omega = k_\omega c/\omega$ is the refractive index for the frequency ω. Since $|E_\pm^+|^2$ does not depend on z (the derivative vanishes), Eq. (81) has the simple exponential solution

$$E_\pm^+(\omega, z) = \exp(i\phi_\pm z)E_\pm^+(\omega,0) \tag{82}$$

where

$$\phi_\pm = \frac{2\pi\omega}{n_\omega c}\left[g_1^\omega|E_\pm^-(\omega)|^2 + 2g_2^\omega|E_\mp^-(\omega)|^2\right] \tag{83}$$

is the light intensity-dependent phase of the light. This phase is responsible for the emergency of circular birefringence. The refractive index is connected with the nonlinear polarization by the relation [122, 123]

$$n_\omega \delta n_\pm E_\pm(r,t) = 2\pi P_\pm(r,t) \tag{84}$$

According to Eq. (79) one easily finds the variations $\delta n_\pm$:

$$\delta n_\pm(\omega) = \frac{2\pi}{n_\omega}\left[g_1^\omega|E_\pm^-(\omega)|^2 + 2g_2^\omega|E_\mp^-(\omega)|^2\right] \tag{85}$$

So, the difference between the right- and left-polarized indices is determined as

$$\begin{aligned}\delta n_+(\omega) - \delta n_-(\omega) = \frac{2\pi}{n_\omega}\Big[&6\chi_{xxyy}(-\omega,-\omega,\omega,\omega)\\ &\times\left(|E_-^-(\omega)^2| - |E_+^-(\omega)|^2\right)\Big]\end{aligned} \tag{86}$$

The expression above is related to the self-rotation of the polarization ellipse, described by Maker et al. [124]. This effect occurs during interaction between a classical field and a nonlinear medium. To search for squeezing it is necessary to use a quantum description.

B. Quantum Treatment: Self-squeezing

The quantum description is based on the analytical form of the Hamiltonian. Generally, the Hamiltonian can be written as

$$H = H_M + H_{FREE} + H_I \tag{87}$$

where H_M is the Hamiltonian for the nonlinear medium and H_{FREE} is the Hamiltonian for the free field. Our interest bears on H_I, the Hamiltonian describing the interaction between the medium and the propagating light. In nonlinear optics [125], it is useful to construct an effective interaction Hamiltonian. Such a Hamiltonian can be obtained from the averaged free energy of the system,

$$H_I = \int_V F \, dV \tag{88}$$

Formally, in the quantum approach, we replace the field vectors by field boson operators, defined as

$$\hat{E}_{\pm}^{+}(\omega) = i\left(\frac{2\pi\hbar\omega}{n_\omega^2 V}\right)^{1/2} \hat{a}_{\pm} \tag{89}$$

where $\hat{a}_{\pm}, \hat{a}_{\pm}^{+}$ are the annihilation and creation operators, satisfying the commutation relations

$$\begin{aligned} \left[\hat{a}_i, \hat{a}_j\right] &= \left[\hat{a}_i^+, \hat{a}_j^+\right] = 0 \\ \left[\hat{a}_i, \hat{a}_j^+\right] &= \delta_{ij} \end{aligned} \tag{90}$$

On insertion of the averaged free energy (77) into the formula (88) one finds [67, 68]

$$\hat{H}_I = -\frac{\hbar}{2}\left[\bar{g}_1^\omega\left(\hat{a}_+^{+2}\hat{a}_+^2 + \hat{a}_-^{+2}\hat{a}_-^2\right) + 4\bar{g}_2^\omega \hat{a}_+^+ \hat{a}_-^+ \hat{a}_+ \hat{a}_-\right] \tag{91}$$

where the nonlinear coupling parameters have been denoted by

$$\begin{aligned} \bar{g}_1^\omega &= \frac{V}{\hbar}\left(\frac{2\pi\hbar\omega}{n_\omega^2 V}\right)^2 g_1^\omega \\ \bar{g}_1^\omega &= \frac{V}{\hbar}\left(\frac{2\pi\hbar\omega}{n_\omega^2 V}\right)^2 g_2^\omega \end{aligned} \tag{92}$$

We use the Hamiltonian (91) to find the field operator time dependence, according to the Heisenberg equation. Taking into account only the effective interaction Hamiltonian, we write

$$\frac{\mathrm{d}\hat{E}^{+}_{\pm}(\omega)}{\mathrm{d}t} = \frac{1}{\mathrm{i}\hbar}\left[\hat{E}^{+}_{\pm}(\omega), \hat{H}_{\mathrm{I}}\right] \tag{93}$$

The time evolution, generally, is considered in a quantum cavity. Since the propagating field must depend on the path z traversed in the medium, we replace t by $-n_{\omega}z/c$ and obtain the Heisenberg equation in the new form

$$\frac{\mathrm{d}\hat{a}_{\pm}(z)}{\mathrm{d}z} = -\frac{\mathrm{i}n_{\omega}}{\hbar c}\left[\hat{a}_{\pm}, \hat{H}_{\mathrm{I}}\right] \tag{94}$$

On insertion of (91) into (94) and using the commutation relations (90), one easily finds

$$\frac{\mathrm{d}\hat{a}_{\pm}(z)}{\mathrm{d}z} = \mathrm{i}\frac{n_{\omega}}{c}\left[\bar{g}_{1}^{\omega}\hat{a}^{+}_{\pm}\hat{a}_{\pm} + 2\bar{g}_{2}^{\omega}\hat{a}^{+}_{\mp}\hat{a}_{\mp}\right]\hat{a}_{\pm} \tag{95}$$

Since the number of photons in the two circular components $\hat{a}^{+}_{+}\hat{a}_{+}$, $\hat{a}^{+}_{-}\hat{a}_{-}$ are constants of motion, Eq. (95) has the simple exponential solution [67, 68]

$$\hat{a}_{\pm}(z) = \exp\left[\mathrm{i}(\varepsilon\hat{a}^{+}_{\pm}(0)\hat{a}_{\pm}(0) + \delta\hat{a}^{+}_{\mp}(0)\hat{a}_{\mp}(0))\right]\hat{a}_{\pm}(0) \tag{96}$$

with the nonlinear parameters

$$\varepsilon = \frac{n_{\omega}}{c}\bar{g}_{1}^{\omega} \qquad \delta = 2\frac{n_{\omega}}{c}\bar{g}_{2}^{\omega} \tag{97}$$

This exact operator solution (96) for the field propagating in an isotropic nonlinear medium can be used to search for quantum effects.

Note that this solution is the general two-mode case of the single-mode solution (61), calculated on the anharmonic oscillator model. If the light is circularly, say right-polarized, then the second term in the exponential vanishes and we get the result for the anharmonic oscillator.

We assume that the incoming beam is in a coherent state with amplitude consisting of two components $|\alpha\rangle = |\alpha_{+}, \alpha_{-}\rangle$. To search for squeezing we introduce the quadrature components of the field (22). On insertion of (96) into the formulas (65) we obtain the following normally ordered quadrature variances [67, 68]:

$$\begin{aligned}\left\langle :\left(\Delta\hat{Q}_+\right)^2: \right\rangle = 2\,\mathrm{Re}\Big[\alpha_+^2 \exp[\mathrm{i}\varepsilon \\ +(\exp(\mathrm{i}2\varepsilon)-1)|\alpha_+|^2 + (\exp(\mathrm{i}2\delta)-1)|\alpha_-|^2\Big] \\ -\alpha_+^2 \exp\Big[2(\exp(\mathrm{i}\varepsilon)-1)|\alpha_+|^2 \\ +(\exp(\mathrm{i}\delta)-1)|\alpha_-|^2\Big]\Big] \\ + 2|\alpha_+|^2\Big[1-\exp\Big[2(\cos\varepsilon-1)|\alpha_+|^2 \\ +2(\cos\delta-1)|\alpha_-|^2\Big]\Big] \\ \left\langle :\left(\Delta\hat{P}_+\right)^2: \right\rangle = -2\,\mathrm{Re}[\cdots] + 2|\alpha_+|^2[\cdots]\end{aligned} \tag{98}$$

where the expressions in brackets in the second equation are the same as in the first equation. On replacing the indices + and − we obtain the variances of the left-polarized quadrature components $\hat{Q}_-$, $\hat{P}_-$. If one of the variances has a value less than zero the field is in the squeezed state.

Because of the complexity of the exact analytical results (98) it is difficult (without numerical analysis) to determine whether they are negative or positive. In real physical processes the nonlinear parameters are very small $\varepsilon \ll 1$, $\delta \ll 1$. This means that significant changes in fluctuations appear for large numbers of photons in the components $|\alpha|^2 \gg 1$, in other words, for strong field. This fact allows us to expand the expressions (98) in power series and to neglect all terms less than $\varepsilon z|\alpha_\pm \pm^2$ or $\delta z|\alpha_\pm|^2$. On the assumption that the phase of the incoming beam is zero, i.e., $|\alpha_\pm| = \alpha_\pm$, we obtain the following simpler formulas for the normally ordered variances:

$$\begin{aligned}\left\langle :\left(\Delta\hat{Q}_\pm\right)^2: \right\rangle = 2(\beta_\pm^2 + \gamma_+\gamma_-) - 2\big[\beta_\pm \sin 2(\beta_\pm + \gamma_\mp) \\ +(\beta_\pm^2 + \gamma_+\gamma_-)\cos 2(\beta_\pm + \gamma_\mp)\big] \\ \left\langle :\left(\Delta\hat{P}_\pm\right)^2: \right\rangle = 2(\cdots) + 2[\cdots]\end{aligned} \tag{99}$$

where the brackets in the second equation include the same expressions as those in the first equation. The parameters are defined as

$$\beta_{\pm} = \varepsilon z |\alpha_{\pm}|^2 \quad \text{and} \quad \gamma_{\pm} = \delta z |\alpha_{\pm}|^2 \tag{100}$$

Considering only one mode of the field, for example $|\alpha_{-}|^2 = 0$, Eqs. (99) go over into formulas (67) obtained on the anharmonic oscillator model.

The numerical results based on the exact solution (98) have been discussed in detail by Tanaś and Kielich [67, 68], showing the possibility of obtaining 98% of squeezing in one of the components for a proper choice of the initial phase. We should note that the canonical nonlinear transformation (96) differs from the transformation (48) for ordinary squeezing and this is the reason why the states obtained in this model also have different properties. Tanaś and Kielich [67, 68] proposed the term "self-squeezing" for the effect, because it depends on the intensity of the mode undergoing it. In 1986, when analyzing the states created due to self-phase modulation in a nonlinear medium, Kitagawa et al. [88, 89] obtained a quasiprobability density with crescent shape. In fact, crescent squeezing is the same as self-squeezing. Tanaś et al. compared the two representations in Ref. 87.

VI. SECOND-HARMONIC GENERATION BY SELF-SQUEEZED LIGHT IN NONLINEAR MEDIUM

A. Second-Harmonic Generation: Classical Treatment

Second-harmonic generation is an important and highly useful nonlinear process. Its first observation by Franken et al. [41] has been the source of much progress in nonlinear optics. Classical effects in second-harmonic generation have been studied extensively [122, 123], and before we describe squeezing we would like to recall some of them.

In the classical approach it is assumed that the field at the space–time point $(\mathbf{r}, t)$ is the superposition of two fields with the fundamental frequency ω and the second-harmonic frequency 2ω:

$$\begin{aligned} E(\mathbf{r}, t) = {} & E^{+}(\omega)\exp[i(\mathbf{k}_{\omega} \cdot \mathbf{r} - \omega t)] \\ & + E^{+}(2\omega)\exp[i(\mathbf{k}_{2\omega} \cdot \mathbf{r} - 2\omega t)] + \text{c.c.} \end{aligned} \tag{101}$$

where $\mathbf{k}_{\omega}, \mathbf{k}_{2\omega}$ are the wave vectors of the fundamental and second-harmonic light waves. We are interested in the interaction between the field and the nonlinear medium. Following Bloembergen [122] and Kielich

[126], the time-averaged free energy can be written in the form

$$\begin{aligned} F = & -\chi_{ijk}(-2\omega, \omega, \omega) E_i^-(2\omega) E_j^+(\omega) E_k^+(\omega) \exp(i\Delta\mathbf{k}_2 \cdot \mathbf{r}) + \text{c.c.} \\ & -\tfrac{3}{4}\left[\chi_{ijkl}(-\omega, -\omega, \omega, \omega) E_i^-(\omega) E_j^-(\omega) E_k^+(\omega) E_l^+(\omega) + \text{c.c.}\right] \\ & -3\left[\chi_{ijkl}(-\omega, -2\omega, \omega, 2\omega) E_i^-(\omega) E_j^-(2\omega) E_k^+(\omega) \right. \\ & \qquad \left. \times E_l^+(2\omega) + \text{c.c.}\right] \\ & -\tfrac{3}{4}\left[\chi_{ijkl}(-2\omega, -2\omega, 2\omega, 2\omega) E_i^-(2\omega) E_j^-(2\omega) \right. \\ & \qquad \left. \times E_k^+(2\omega) E_l^+(2\omega) + \text{c.c.}\right] \end{aligned} \tag{102}$$

where $\Delta\mathbf{k}_2 = 2\mathbf{k}_\omega - \mathbf{k}_{2\omega}$. $E^{\pm}(\omega), E^{\pm}(2\omega)$ are the components of the field vectors. Recall from the preceding section that nonlinear polarization can be obtained from the averaged free energy (72). In this case we get the following form of the polarization components:

$$\begin{aligned} P_i^+(\omega) = & \, 2\chi_{ijk}(-\omega, -\omega, 2\omega) E_j^-(\omega) E_k^+(2\omega) \exp(-i\Delta\mathbf{k}_2 \cdot \mathbf{r}) \\ & + 3\chi_{ijkl}(-\omega, -\omega, \omega, \omega) E_j^-(\omega) E_k^+(\omega) E_l^+(\omega) \\ & + 6\chi_{ijkl}(-\omega, -2\omega, \omega, 2\omega) E_j^-(2\omega) E_k^+(\omega) E_l^+(2\omega) \end{aligned} \tag{103}$$

and for the second-harmonic frequency

$$\begin{aligned} P_i^+(2\omega) = & \, \chi_{ijk}(-2\omega, \omega, \omega) E_j^+(\omega) E_k^+(\omega) \exp(i\Delta\mathbf{k}_2 \cdot \mathbf{r}) \\ & + 6\chi_{ijkl}(-2\omega, -\omega, 2\omega, \omega) E_j^-(\omega) E_k^+(2\omega) E_l^+(\omega) \\ & + 3\chi_{ijkl}(-2\omega, -2\omega, 2\omega, 2\omega) E_j^-(2\omega) \\ & \times E_k^+(2\omega) E_l^+(2\omega) \end{aligned} \tag{104}$$

In Eq. (103) the third-rank tensor $\chi_{ijk}(-\omega, -\omega, 2\omega)$ describing second-order susceptibility is related to the reconversion of part of the second harmonic back into the fundamental beam, $\chi_{ijkl}(-\omega, -\omega, \omega, \omega)$. As we showed in Section V, this is related to self-induced ellipse rotation (86) and $\chi_{ijkl}(-\omega, -2\omega, \omega, 2\omega)$ determines the optical Kerr effect at ω due to the intensity $|E^-(2\omega)|^2$. In Eq. (104) the tensor $\chi_{ijk}(-2\omega, \omega, \omega)$ is responsible for second-harmonic generation [41]; $\chi_{ijkl}(-2\omega, -\omega, 2\omega, \omega)$ determines the variation of the refractive index at 2ω, stimulated by the intensity $|E^-(\omega)|^2$; and $\chi_{ijkl}(-2\omega, -2\omega, 2\omega, 2\omega)$ is connected with the effect of self-induced intensity-dependent refractive index at 2ω.

Since Eqs. (103) and (104) concern a nonresonant, nondissipative process, it is possible to derive the following symmetry relations for the susceptibility tensors:

$$\begin{aligned}
\chi^{*}_{ijkl}(-\omega,-2\omega,\omega,2\omega) &= \chi_{lkji}(-2\omega,-\omega,2\omega,\omega)\\
\chi^{*}_{ijkl}(-\omega,-2\omega,\omega,2\omega) &= \chi_{klij}(-\omega,-2\omega,\omega,2\omega)\\
\chi^{*}_{ijkl}(-\omega,-\omega,\omega,\omega) &= \chi_{klij}(-\omega,-\omega,\omega,\omega)\\
\chi^{*}_{ijk}(-\omega,-\omega,2\omega) &= \chi_{kij}(-2\omega,\omega,\omega)
\end{aligned} \tag{105}$$

Let us consider a nonlinear isotropic medium with a center of symmetry. In this case the tensors $\chi_{ijk}(-2\omega,\omega,\omega)$, which are responsible for the generation of the second harmonic, vanish. To arouse the wave at frequency 2ω an externally dc electric field has to be applied to destroy the center of symmetry. Then the medium becomes capable of generating the second-harmonic beam. Assuming the dc electric field to act along the y axis, the third-rank tensors can be written [127] as follows:

$$\chi^{2\omega}_{ijk}(E^0) = \chi^{2\omega}_{ijk}(0) + \chi^{2\omega}_{xxyy}\delta_{ij}E^0_k + \chi^{2\omega}_{xyxy}\delta_{ik}E^0_j + \chi^{2\omega}_{yxxy}\delta_{jk}E^0_i \tag{106}$$

where E^0 is the external dc field. Since the second harmonic propagates along the z axis, like the fundamental beam, the $\chi_{ijk}(0)$ vanish [79]. Moreover, considering the isotropic medium with center of symmetry we take into account the symmetry relation (75). As was shown in Section V, it is convenient to have recourse to circular components. If the field propagates along the z axis they are defined by formulas (76). On using this basis the averaged free energy takes the form [79]

$$\begin{aligned}
F = -\tfrac{1}{2}\Big[& g^{\omega}_1\big[E^-_+(\omega)^2E^+_+(\omega)^2 + E^-_-(\omega)^2E^+_-(\omega)^2\big]\\
&+4g^{\omega}_2E^-_+(\omega)E^-_-(\omega)E^+_+(\omega)E^+_-(\omega)\\
&+g^{2\omega}_1\big[E^-_+(2\omega)^2E^+_+(2\omega)^2 + E^-_-(2\omega)^2E^+_-(2\omega)^2\big]\\
&+4g^{2\omega}_2E^-_+(2\omega)E^-_-(2\omega)E^+_+(2\omega)E^+_-(2\omega)\Big]\\
&-\mathrm{i}\Big[g^{2\omega}_3\big[E^-_+(2\omega)E^+_-(\omega)^2 - E^-_-(2\omega)E^+_-(\omega)^2\big]\\
&-2g^{2\omega}_4\big[E^-_+(2\omega) - E^-_-(2\omega)\big]E^+_+(\omega)E^+_-(\omega)\Big]\exp(\mathrm{i}\Delta k_2 r) + \text{c.c.}\\
&-g^{2\omega}_5\big[E^-_+(2\omega)E^-_-(\omega)E^+_+(\omega)E^+_-(2\omega)\\
&\qquad+E^-_-(2\omega)E^-_+(\omega)E^+_-(\omega)E^+_+(2\omega)\big]\\
&-g^{2\omega}_6\big[E^-_+(2\omega)E^-_-(\omega)E^+_-(\omega)E^+_+(2\omega)\\
&\qquad+E^-_-(2\omega)E^-_+(\omega)E^+_+(\omega)E^+_-(2\omega)\big]\\
&-g^{2\omega}_7\big[E^-_+(2\omega)E^-_+(\omega)E^+_+(\omega)E^+_+(2\omega)\\
&\qquad+E^-_-(2\omega)E^-_-(\omega)E^+_-(\omega)E^+_-(2\omega)\big]
\end{aligned} \tag{107}$$

The nonlinear coupling parameters are defined as ($\Omega = \omega$ or 2ω)

$$
\begin{aligned}
g_1^{\Omega} &= 6\chi_{xyxy}(-\Omega, -\Omega, \Omega, \Omega) \\
g_2^{\Omega} &= 3[\chi_{xxyy}(-\Omega, -\Omega, \Omega, \Omega) + \chi_{xyxy}(-\Omega, -\Omega, \Omega, \Omega)] \\
g_3^{2\omega} &= 2^{1/2}\chi_{xxyy}(-2\omega, \omega, \omega, 0)E_y^0 \\
g_4^{2\omega} &= 2^{-1/2}[\chi_{xxyy}(-2\omega, \omega, \omega, 0) + \chi_{xyxy}(-2\omega, \omega, \omega, 0)]E_y^0 \\
g_5^{2\omega} &= 3[\chi_{xxyy}(-2\omega, -\omega, \omega, 2\omega) + \chi_{xyxy}(-2\omega, -\omega, \omega, 2\omega)] \\
g_6^{2\omega} &= 3[\chi_{xxyy}(-2\omega, -\omega, \omega, 2\omega) + \chi_{xyyx}(-2\omega, -\omega, \omega, 2\omega)] \\
g_7^{2\omega} &= 3[\chi_{xyxy}(-2\omega, -\omega, \omega, 2\omega) + \chi_{xyyx}(-2\omega, -\omega, \omega, 2\omega)]
\end{aligned}
\tag{108}
$$

In accordance with formula (72) we obtain the following components of the nonlinear polarization:

$$
\begin{aligned}
P_{\pm}^{+}(\omega) = {} & [g_1^{\omega}|E_{\pm}^{-}(\omega)|^2 + 2g_2^{\omega}|E_{\mp}^{-}(\omega)|^2]E_{\pm}^{+}(\omega) \\
& -2\mathrm{i}[\pm g_3^{-2\omega}E_{\pm}^{+}(2\omega)E_{\pm}^{-}(\omega) \\
& -g_4^{-2\omega}[E_{+}^{+}(2\omega) - E_{-}^{+}(2\omega)]E_{\mp}^{-}(\omega)] \\
& \times \exp(-\mathrm{i}\mathbf{\Delta k}_2 \cdot \mathbf{r}) + g_5^{2\omega}E_{-}^{-}(2\omega)E_{-}^{+}(\omega)E_{+}^{+}(2\omega) \\
& +[g_6^{2\omega}|E_{\mp}^{-}(2\omega)|^2 + g_7^{2\omega}|E_{\pm}^{-}(2\omega)|^2]E_{\pm}^{+}(\omega)
\end{aligned}
\tag{109}
$$

and at 2ω

$$
\begin{aligned}
P_{\pm}^{+}(2\omega) = {} & [g_1^{2\omega}|E_{\pm}^{-}(2\omega)|^2 + 2g_2^{2\omega}|E_{\mp}^{-}(2\omega)|^2]E_{\pm}^{+}(2\omega) \\
& +\mathrm{i}[\pm g_3^{2\omega}E_{\pm}^{+}(\omega)^2 \mp g_4^{2\omega}E_{+}^{+}(\omega)E_{-}^{+}(\omega)] \\
& \times \exp(\mathrm{i}\mathbf{\Delta k}_2 \cdot \mathbf{r}) + g_5^{2\omega}E_{-}^{-}(\omega)E_{+}^{+}(\omega)E_{-}^{+}(2\omega) \\
& +[g_6^{2\omega}|E_{\mp}^{-}(\omega)|^2 + g_7^{2\omega}|E_{\pm}^{-}(\omega)|^2]E_{\pm}^{+}(2\omega)
\end{aligned}
\tag{110}
$$

On insertion of these expressions into the Maxwell equation (80) and neglecting terms unrelated to the self-induced intensity-dependent effect, one finds

$$
\frac{\mathrm{d}E_{\pm}^{+}(\Omega)}{\mathrm{d}z} = \mathrm{i}\frac{2\pi\Omega}{n_{\Omega}c}[g_1^{\Omega}|E_{\pm}^{-}(\Omega)|^2 + 2g_2^{\Omega}|E_{\mp}^{-}(\Omega)|^2]E_{\pm}^{+}(\Omega) \tag{111}
$$

Since $(\mathrm{d}/\mathrm{d}z)|E_{\pm}^{-}|^2 = 0$, Eq. (111) possesses the simple solution

$$
E_{\pm}^{+}(\Omega, z) = \exp(\mathrm{i}\phi_{\pm}z)E_{\pm}^{+}(\Omega, 0) \tag{112}
$$

where the phase shifts have the form:

$$\phi_{\pm} = \mathrm{i}\frac{2\pi\Omega}{n_{\Omega}c}\left[g_1^{\Omega}|E_{\pm}^{-}(\Omega)|^2 + 2g_2^{\Omega}|E_{\mp}^{-}(\Omega)|^2\right] \tag{113}$$

Equation (112) represents the general solution for the fundamental wave ($\Omega = \omega$) (obtained above in Eq. (82)) and, at the same time, for the second harmonic ($\Omega = 2\omega$).

Similar to the case of light propagation at ω alone (Section V), it is possible to analyze the birefringence effects for the fundamental and second harmonic. Applying formula (84) and inserting the nonlinear polarization of Eqs. (109) and (110), one can easily calculate the variations of the refractive indices:

$$\begin{aligned} \delta n_{\pm}(\omega) &= \frac{2\pi}{n_{\omega}}\left[g_1^{\omega}|E_{\pm}^{-}(\omega)|^2 + 2g_2^{\omega}|E_{\mp}^{-}(\omega)|^2\right. \\ &\qquad \left. + g_6^{2\omega}|E_{\mp}^{-}(2\omega)|^2 + g_7^{2\omega}|E_{\pm}^{-}(2\omega)|^2\right] \\ \delta n_{\pm}(2\omega) &= \frac{2\pi}{n_{2\omega}}\left[g_1^{2\omega}|E_{\pm}^{-}(2\omega)|^2 + 2g_2^{2\omega}|E_{\mp}^{-}(2\omega)|^2\right. \\ &\qquad \left. + g_6^{2\omega}|E_{\mp}^{-}(\omega)|^2 + g_7^{2\omega}|E_{\pm}^{-}(\omega)|^2\right] \end{aligned} \tag{114}$$

Hence, the difference between the two circular components takes the form

$$\begin{aligned} &\delta n_{+}(\omega) - \delta n_{-}(\omega) \\ &= \frac{2\pi}{n_{\omega}}\left[6\chi_{xxyy}(-\omega, -\omega, \omega, \omega)\left[|E_{-}^{-}(\omega)|^2 - |E_{+}^{-}(\omega)|^2\right]\right. \\ &+ 3\left[\chi_{xxyy}(-\omega, -2\omega, \omega, 2\omega) - \chi_{xyyx}(-\omega, -2\omega, \omega, 2\omega)\right] \\ &\left.\times\left(|E_{-}^{-}(2\omega)|^2 - |E_{+}^{-}(2\omega)|^2\right)\right] \end{aligned} \tag{115}$$

and, at 2ω,

$$\begin{aligned} &\delta n_{+}(2\omega) - \delta n_{-}(2\omega) \\ &= \frac{2\pi}{n_{2\omega}}\left[6\chi_{xxyy}(-2\omega, -2\omega, 2\omega, 2\omega)\left[|E_{-}^{-}(2\omega)|^2 - |E_{+}^{-}(2\omega)|^2\right]\right. \\ &+ 3\left[\chi_{xxyy}(-2\omega, -\omega, 2\omega, \omega) - \chi_{xyyx}(-2\omega, -\omega, 2\omega, \omega)\right] \\ &\left.\times\left(|E_{-}^{-}(\omega)|^2 - |E_{+}^{-}(\omega)|^2\right)\right] \end{aligned} \tag{116}$$

The first term of Eq. (115) was discussed in Section V. The second term is responsible for the additional anisotropy caused by the intensity of the second harmonic. The effect determined by expression (116) has not been studied experimentally.

B. Squeezing in Second-Harmonic Generation

As was done in Subsection V.B, the electric field vectors should be replaced by boson operators in the quantum description. The operator for the fundamental field was defined in (89). Similarly, the operator for the second-harmonic field can be determined as

$$\hat{E}_{\pm}^{+}(2\omega) = \mathrm{i}\left(\frac{2\pi\hbar 2\omega}{n_{2\omega}^{2}V}\right)^{1/2}\hat{b}_{\pm} \tag{117}$$

where $\hat{b}_{\pm}, \hat{b}_{\pm}^{+}$ are the boson annihilation and creation operators for photons at the frequency 2ω. These operators obey the boson commutation relations (90) and additionally

$$\left[\hat{a}_i, \hat{b}_j\right] = \left[\hat{a}_i^{+}, \hat{b}_j^{+}\right] = \left[\hat{a}_i, \hat{b}_j^{+}\right] = 0 \tag{118}$$

To consider the squeezing effect it is necessary to have available the form of the field propagating through the nonlinear medium. We get it from the Heisenberg equation (93), taking into account the interaction Hamiltonian (slowly varying amplitude approximation), which can be derived from formula (88). On insertion of the averaged free energy (107) into (88) the interaction Hamiltonian, in our case, takes the form

$$\begin{aligned}
\hat{H}_{\mathrm{I}} = -\frac{\hbar}{2}\Big[&\bar{g}_1^{\omega}\left(\hat{a}_{+}^{+2}\hat{a}_{+}^{2} + \hat{a}_{-}^{+2}\hat{a}_{-}^{2}\right) + 4\bar{g}_2^{\omega}\hat{a}_{+}^{+}\hat{a}_{-}^{+}\hat{a}_{+}\hat{a}_{-} \\
&+\bar{g}_1^{2\omega}\left(\hat{b}_{+}^{+2}\hat{b}_{+}^{2} + \hat{b}_{-}^{+2}\hat{b}_{-}^{2}\right) + 4\bar{g}_2^{2\omega}\hat{b}_{+}^{+}\hat{b}_{-}^{+}\hat{b}_{+}\hat{b}_{-}\Big] \\
&- \hbar\left[\bar{g}_3^{2\omega}\left(\hat{b}_{-}^{+}\hat{a}_{-}^{2} - \hat{b}_{+}^{+}\hat{a}_{+}^{2}\right) + 2\bar{g}_4^{2\omega}\left(\hat{b}_{+}^{+} - \hat{b}_{-}^{+}\right)\hat{a}_{+}\hat{a}_{-}\right] \\
&\times \exp(\mathrm{i}\boldsymbol{\Delta}\mathbf{k}_2\cdot\mathbf{r}) + \mathrm{h.c.} \\
&- \hbar\Big[\bar{g}_5^{2\omega}\left(\hat{b}_{+}^{+}\hat{a}_{-}^{+}\hat{a}_{+}\hat{b}_{-} + \hat{b}_{-}^{+}\hat{a}_{+}^{+}\hat{a}_{-}\hat{b}_{+}\right) \\
&+\bar{g}_6^{2\omega}\left(\hat{b}_{+}^{+}\hat{a}_{-}^{+}\hat{a}_{-}\hat{b}_{+} + \hat{b}_{-}^{+}\hat{a}_{+}^{+}\hat{a}_{+}\hat{b}_{-}\right) \\
&+\bar{g}_7^{2\omega}\left(\hat{b}_{+}^{+}\hat{a}_{+}^{+}\hat{a}_{+}\hat{b}_{+} + \hat{b}_{-}^{+}\hat{a}_{-}^{+}\hat{a}_{-}\hat{b}_{-}\right)\Big]
\end{aligned} \tag{119}$$

where the nonlinear coupling parameters (108) are redefined:

$$
\begin{aligned}
\bar{g}^{\Omega}_{1,2} &= \frac{V}{\hbar}\left(\frac{2\pi\hbar\Omega}{n^2_{\Omega}V}\right)^2 g^{\Omega}_{1,2} \\
\bar{g}^{2\omega}_{3,4} &= \frac{V}{\hbar}\left(\frac{2\pi\hbar 2\omega}{n^2_{2\omega}V}\right)^{1/2}\left(\frac{2\pi\hbar\omega}{n^2_{\omega}V}\right) g^{2\omega}_{3,4} \\
\bar{g}^{2\omega}_{5,6,7} &= \frac{V}{\hbar}\left(\frac{2\pi\hbar 2\omega}{n^2_{2\omega}V}\right)\left(\frac{2\pi\hbar\omega}{n^2_{\omega}V}\right) g^{2\omega}_{5,6,7}
\end{aligned}
\tag{120}
$$

Replacing the time t by the path of propagation z, as in Section V, the Heisenberg equations become

$$
\begin{aligned}
\frac{\mathrm{d}\hat{a}_{\pm}(z)}{\mathrm{d}z} &= -\frac{\mathrm{i}n_{\omega}}{\hbar c}\left[\hat{a}_{\pm}, \hat{H}\right] \\
\frac{\mathrm{d}\hat{b}_{\pm}(z)}{\mathrm{d}z} &= -\frac{\mathrm{i}n_{2\omega}}{\hbar c}\left[\hat{b}_{\pm}, \hat{H}\right]
\end{aligned}
\tag{121}
$$

In accordance with formula (119) one obtains the general operator equations of motion for the fundamental and second-harmonic fields:

$$
\begin{aligned}
\frac{\mathrm{d}\hat{a}_{\pm}(z)}{\mathrm{d}z} = \mathrm{i}\frac{n_{\omega}}{c}\Big[&\left(\bar{g}^{\omega}_{1}\hat{a}^{+}_{\pm}\hat{a}_{\pm} + 2\bar{g}^{\omega}_{2}\hat{a}^{+}_{\mp}\hat{a}_{\mp}\right)\hat{a}_{\pm} \\
&+ 2\left[\mp\bar{g}^{-2\omega}_{3}\hat{b}_{\pm}\hat{a}^{+}_{\pm} + \bar{g}^{-2\omega}_{4}\left(\hat{b}_{+} - \hat{b}_{-}\right)\hat{a}^{+}_{\mp}\right]\exp(-\mathrm{i}\Delta\mathbf{k}_2\cdot\mathbf{r}) \\
&+ \left[\bar{g}^{2\omega}_{5}\hat{b}^{+}_{\mp}\hat{a}_{\mp}\hat{b}_{\pm} + \left(\bar{g}^{2\omega}_{6}\hat{b}^{+}_{\mp}\hat{b}_{\mp} + \bar{g}^{2\omega}_{7}\hat{b}^{+}_{\pm}\hat{b}_{\pm}\right)\hat{a}_{\pm}\right]\Big] \\
\frac{\mathrm{d}\hat{b}_{\pm}(z)}{\mathrm{d}z} = \mathrm{i}\frac{n_{2\omega}}{c}\Big[&\left(\bar{g}^{2\omega}_{1}\hat{b}^{+}_{\pm}\hat{b}_{\pm} + 2\bar{g}^{2\omega}_{2}\hat{b}^{+}_{\mp}\hat{b}_{\mp}\right)\hat{b}_{\pm} \\
&+ \left(\mp\bar{g}^{2\omega}_{3}\hat{a}^{2}_{\pm} \pm 2\bar{g}^{2\omega}_{4}\hat{a}_{+}\hat{a}_{-}\right)\exp(\mathrm{i}\Delta\mathbf{k}_2\cdot\mathbf{r}) \\
&+ \left[\bar{g}^{2\omega}_{5}\hat{a}^{+}_{\mp}\hat{a}_{\pm}\hat{b}_{\mp} + \left(\bar{g}^{2\omega}_{6}\hat{a}^{+}_{\mp}\hat{a}_{\mp} + \bar{g}^{2\omega}_{7}\hat{a}^{+}_{\pm}\hat{a}_{\pm}\right)\hat{b}_{\pm}\right]\Big]
\end{aligned}
\tag{122}
$$

The first equation in (122) is a generalization of the expression (96). Since both equations in (122) contain interference terms, they should be solved simultaneously. This is a difficult task and some approximations are needed. To start with we assume that the dominant process resides in self-interaction of the fundamental beam that is described by the parameters $\bar{g}_1^{\omega}$ and $\bar{g}_2^{\omega}$. Hence, this assumption means that the other coupling constants are smaller and can be neglected. We next apply the solution (96) as zero-order solution in solving (122) for the second harmonic perturbatively. On formal integration, we arrive at the following equation [79]:

$$\hat{b}_{\pm}(z) = \hat{b}_{\pm}(0) \mp i\frac{n_{2\omega}}{c}\int_0^z dz' \exp(i\Delta k_2 z')\left[\bar{g}_3^{2\omega}\hat{a}_{\pm}^2(z') - 2\bar{g}_4^{2\omega}\hat{a}_+(z')\hat{a}_-(z')\right] \tag{123}$$

where terms containing the second-harmonic operators $\hat{b}_{\pm}(z)$ have been neglected. Next, we assume that the fundamental field is in a coherent state with circular polarization, for example right. Automatically the term with $\bar{g}_4^{2\omega}$ vanishes ($\hat{a}_-|\alpha_+\rangle = 0$). Moreover, the second harmonic does not exist for $z = 0$ ($\hat{b}_{\pm}|\alpha_+\rangle = 0$). These assumptions enable us to find simple formulas for the variances of the quadrature operators. Inserting (96) into Eq. (123) and using the definitions of the quadrature operators for the second-harmonic field we obtain their normally ordered variances,

$$\left.\begin{matrix}\langle:(\Delta\hat{Q}_+)^2:\rangle \\ \langle:(\Delta\hat{P}_+)^2:\rangle\end{matrix}\right\} = -2\kappa_{2\omega}^2\int_0^z dz'\int_0^z dz''\Big[\pm\alpha_+^4 \cos\big[(\Delta k_2+\varepsilon)(z'+z'') + 4\varepsilon z'' + |\alpha_+|^2 \sin 2\varepsilon(z'+z'')\big]\exp\big[(\cos 2\varepsilon(z'+z'')-1)|\alpha_+|^2\big]$$
$$\mp\alpha_+^4 \cos\big[(\Delta k_2+\varepsilon)(z'+z'') + |\alpha_+|^2\sin 2\varepsilon z' + |\alpha_+|^2 \sin 2\varepsilon z''\big] \times\exp\big[(\cos 2\varepsilon z' + \cos 2\varepsilon z'' - 2)|\alpha_+|^2\big]$$
$$-|\alpha_+|^4\cos\big[(\Delta k_2+\varepsilon)(z'-z'') + |\alpha_+|^2\sin 2\varepsilon(z'-z'')\big] \times\exp\big[(\cos 2\varepsilon(z'-z'')-1)|\alpha_+|^2\big] \tag{124}$$
$$+|\alpha_+|^2\cos\big[(\Delta k_2+\varepsilon)(z'-z'') + |\alpha_+|^2\sin 2\varepsilon z' - |\alpha_+|^2\sin 2\varepsilon z''\big] \times\exp\big[(\cos 2\varepsilon z' + \cos 2\varepsilon z'' - 2)|\alpha_+|^2\big]$$

where the coupling parameter is determined as

$$\kappa_{2\omega} = \frac{n_{2\omega}}{c} \bar{g}_3^{2\omega} \tag{125}$$

The squeezing effect occurs, in the second-harmonic beam, if one of the variances in (124) takes a negative value. These equations are very complicated and difficult to analyze. From Section V, we find that it is possible to make the assumption that $\varepsilon z \ll 1$. Then the variances can be expanded in power series and we retain only terms containing $\varepsilon z|\alpha_+|^2 \approx 1$ for $|\alpha_+|^2 \gg 1$. Moreover, we assume phase matching, i.e., $\Delta k_2 = 0$, and that the phase of the incoming beam is zero, i.e., $|\alpha_+| = \alpha_+$. On these assumptions the following approximate expressions are obtained:

$$\begin{aligned}\left\langle :\left(\Delta \hat{Q}_+\right)^2: \right\rangle \approx 2\frac{\eta}{\beta_2^2}\Big[&2\cos\beta_2 - \cos 2\beta_2 - 1 - \beta_2(\sin 2\beta_2 - \sin\beta_2) \\ &+ (\cos\beta_2 - 1 + \beta_2\sin\beta_2)^2\Big] \\ \left\langle :\left(\Delta \hat{P}_+\right)^2: \right\rangle \approx 2\frac{\eta}{\beta_2^2}\Big[&-2\cos\beta_2 + \cos 2\beta_2 + 1 + \beta_2(\sin 2\beta_2 - \sin\beta_2) \\ &+ (\sin\beta_2 - \beta_2\cos\beta_2)^2\Big]\end{aligned} \tag{126}$$

where we introduce

$$\beta_2 = 2\varepsilon z|\alpha_+|^2 \tag{127}$$

By η we denote the part of the fundamental beam power transferred into the second harmonic,

$$\eta = \frac{2\kappa_{2\omega}^2|\alpha_+|^4 z^2}{|\alpha_+|^2} \approx \frac{I(2\omega)}{I(\omega)} \tag{128}$$

The approximate results (126) are convenient to analyze. The normally ordered variances of the $\hat{P}$ component of the second harmonic is plotted in Fig. 3 against $\varepsilon z|\alpha_+|^2$ together with the $\hat{Q}$ component of the fundamental beam (99) showing that the squeezing effect in the second harmonic (solid line) is correlated with the self-squeezing in the fundamental beam (dashed line), because they are negative for small values of β_2. The squeezing from the $\hat{Q}$ component of the fundamental beam can be said to

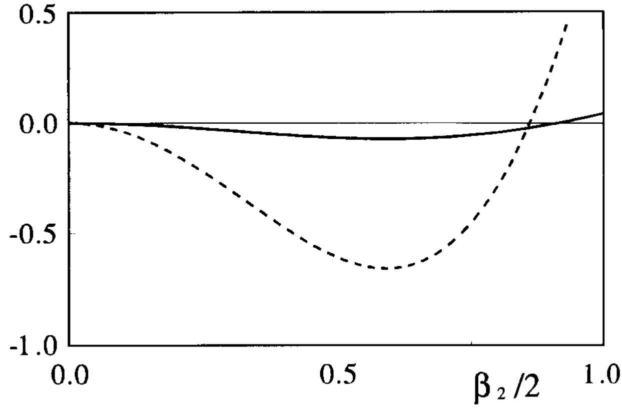

Figure 3. The approximate variance of the $\hat{Q}$ component of the fundamental field (dashed line) and the approximate variance of the $\hat{P}$ component of the second-harmonic field (solid line) are plotted versus $\beta_2/2 = \varepsilon z|\alpha_+|^2 (\eta = 0.1)$.

be transferred, in some sense, into the $\hat{P}$ component of the second harmonic. However, we have to recall that these results (126) have been obtained under the approximation that there was no coupling of the second harmonic back into the fundamental. Hence, if η takes a large value this assumption breaks down.

VII. THIRD-HARMONIC GENERATION BY SELF-SQUEEZED LIGHT IN NONLINEAR MEDIUM

In a nonlinear medium that, with regard to its symmetry, admits third-harmonic (not second-harmonic) generation, two nonlinear processes occur simultaneously: first, nonlinear propagation of the fundamental field, described in Sections IV and V, leading to the self-squeezing effect, and second, third-harmonic generation. In the classical treatment the latter is a well-known phenomenon [123]. Since the self-squeezed light produces the third harmonic we can suppose that, as in the second-harmonic case (Section V), squeezing is transferred to the beam at 3ω.

To find the squeezed states in this process we have to find the equation describing the evolution of the field. As in the preceding sections, we use the Heisenberg equation (93). In quantum description, using a spherical basis, the third harmonic can be written as

$$\hat{E}^+_\pm(3\omega) = \mathrm{i}\left(\frac{2\pi\hbar 3\omega}{n_{3\omega}V}\right)^{1/2} \hat{c}_\pm \tag{129}$$

where $\hat{c}_{\pm}, \hat{c}_{\pm}^{+}$ are the annihilation and creation operators for a photon at the frequency 3ω. The interaction Hamiltonian, in terms of these operators, takes the form [80]

$$\hat{H}_{\mathrm{I}} = 2\hbar \bar{g}^{3\omega}\left(\hat{c}_{+}^{+}\hat{a}_{+}^{2}\hat{a}_{-} + \hat{c}_{-}^{+}\hat{a}_{-}^{2}\hat{a}_{+}\right)\exp(\mathrm{i}\Delta k_3 z) + \mathrm{h.c.} \tag{130}$$

where the nonlinear coupling parameter is determined as

$$\bar{g}^{3\omega} = \frac{V}{\hbar}\left(\frac{2\pi\hbar 3\omega}{n_{3\omega}V}\right)^{1/2}\left(\frac{2\pi\hbar\omega}{n_{\omega}V}\right)^{3/2}\chi_{xxxx}(-3\omega,\omega,\omega,\omega) \tag{131}$$

The susceptibility tensor obeys relation (75). We have assumed that both beams propagate along the z axis with the linear phase mismatch $\Delta k_3 = 3k_{\omega} - k_{3\omega}$.

Applying the Heisenberg equation (93) and replacing t by z, one easily finds the following relations [80]:

$$\begin{aligned}\frac{\mathrm{d}\hat{c}_{\pm}(z)}{\mathrm{d}z} &= 2\mathrm{i}\frac{n_{3\omega}}{c}\bar{g}^{3\omega}\hat{a}_{\pm}^{2}(z)\hat{a}_{\mp}(z)\exp(\mathrm{i}\Delta k_3 z)\\ \frac{\mathrm{d}\hat{a}_{\pm}(z)}{\mathrm{d}z} &= 2\mathrm{i}\frac{n_{\omega}}{c}\bar{g}^{3\omega}\big[2\hat{c}_{\pm}(z)\hat{a}_{\pm}^{+}(z)\hat{a}_{\mp}^{+}(z)\\ &\qquad + \hat{c}_{\mp}(z)\hat{a}_{\mp}^{+2}(z)\big]\exp(-\mathrm{i}\Delta k_3 z)\end{aligned} \tag{132}$$

Equations (132) show the coupling between the third harmonic and fundamental beam. On the assumption that the main process is the self-squeezing described by (95) it is possible to use the solution (96) as the zero-approximation solution solving the first equation of (132). On formal integration the following formula is obtained:

$$\hat{c}_{\pm}(z) = \hat{c}_{\pm}(0) + 2\mathrm{i}\kappa_{3\omega}\int_0^z \hat{a}_{\pm}^{2}(z')\hat{a}_{\mp}(z')\exp(\mathrm{i}\Delta k_3 z')\,dz' \tag{133}$$

where we denote

$$\kappa_{3\omega} = \frac{n_{3\omega}}{c}\bar{g}^{3\omega} \tag{134}$$

To say whether squeezing occurs in the third-harmonic beam it is necessary to analyze the quadrature variances (65) defined for the operators $\hat{c}_{\pm}, \hat{c}_{\pm}^{+}$. We assume that the incoming field is a coherent state at $z = 0$.

Using Eq. (105) and taking into account the solution (96) the normally ordered variances are found [80]:

$$\begin{aligned}
&\left\langle :\left(\Delta \hat{Q}_{+}\right)^{2}: \right\rangle \\
&= -8\kappa_{3\omega}^{2}\int_{0}^{z} dz' \int_{0}^{z} dz'' \Big[\mathrm{Re}\, \alpha_{+}^{4}\alpha_{-}^{2} \exp\big[i(z'+z'')(\Delta k_{3}+\varepsilon+2\delta) \\
&\quad + (\exp[i(z'+z'')(2\varepsilon+\delta)]-1)|\alpha_{+}|^{2} \\
&\quad + (\exp[i(z'+z'')(\varepsilon+2\delta)]-1)|\alpha_{-}|^{2} \\
&\quad + iz''(5\varepsilon+2\delta)\big] \\
&\quad - \mathrm{Re}\, \alpha_{+}^{4}\alpha_{-}^{2} \exp\big[i(z'+z'')(\Delta k+\varepsilon+2\delta) \\
&\quad + (\exp[iz'(2\varepsilon+\delta)] + \exp[iz''(2\varepsilon+\delta)] - 2)|\alpha_{+}|^{2} \\
&\quad + (\exp[iz'(\varepsilon+2\delta)] + \exp[iz''(\varepsilon+2\delta)] - 2)|\alpha_{-}|^{2}\big] \\
&\quad - |\alpha_{+}|^{4}|\alpha_{-}|^{2} \exp\big[-i(z'-z'')(\Delta k_{3}+\varepsilon+2\delta) \qquad (135)\\
&\quad + (\exp[-i(z'-z'')(2\varepsilon+\delta)]-1)|\alpha_{+}|^{2} \\
&\quad + (\exp[-i(z'-z'')(\varepsilon+2\delta)]-)|\alpha_{-}|^{2}\big] \\
&\quad + |\alpha_{+}|^{4}|\alpha_{-}|^{2} \exp\big[-i(z'-z'')(\Delta k_{3}+\varepsilon+2\delta) \\
&\quad + (\exp[-iz'(2\varepsilon+\delta)] \\
&\quad + \exp[iz''(2\varepsilon+\delta)] - 2)|\alpha_{+}|^{2} + (\exp[-iz'(\varepsilon+2\delta)] \\
&\quad + \exp[iz''(\varepsilon+2\delta)] - 2)|\alpha_{-}|^{2}\big]\Big]
\end{aligned}$$

The variance of the left-polarized component is obtained by replacing all plus subscripts by minus subscripts in Eq. (135). The expressions for the quadrature operators $\hat{P}_{\pm}$ differ in the signs of their Re terms from Eq. (135). It is obvious that the variances are zero if only one of the circular components exists in the incoming beam. Hence, we take into account a beam linearly polarized along the x axis, i.e., $\alpha_{+} = \alpha_{-} = \alpha/\sqrt{2}$. Moreover, we assume that the parameters ε and δ defined in formula (97) are equal. Using the x, y, z basis, as the simplest in this case, one easily obtains the following equation:

$$\begin{aligned}\left\langle:\left(\Delta\hat{Q}_x\right)^2:\right\rangle = &-2\kappa_{3\omega}^2|\alpha|^6\int_0^z dz'\int_0^z dz''\Big[\exp\left[(\cos 3\varepsilon(z'+z'')-1)|\alpha|^2\right]\\&\times\cos\left[(\Delta k_3+3\varepsilon)(z'+z'')+9\varepsilon z''+|\alpha|^2\sin 3\varepsilon(z'+z'')\right]\\&-\exp\left[(\cos 3\varepsilon z'+\cos 3\varepsilon z''-2)|\alpha|^2\right]\\&\times\cos\left[(\Delta k_3+3\varepsilon)(z'+z'')+|\alpha|^2(\sin 3\varepsilon z'+\sin 3\varepsilon z'')\right]\\&-\exp\left[(\cos 3\varepsilon(z'-z'')-1)|\alpha|^2\right]\cos\left[(\Delta k_3+3\varepsilon)(z'-z'')\right.\\&\left.+|\alpha|^2\sin 3\varepsilon(z'-z'')\right]+\exp\left[(\cos 3\varepsilon z'+\cos 3\varepsilon z''-2)|\alpha|^2\right]\\&\times\cos\left[(\Delta k_3+3\varepsilon)(z'-z'')+|\alpha|^2(\sin 3\varepsilon z'-\sin 3\varepsilon z'')\right]\Big]\end{aligned} \tag{136}$$

This equation is still too complicated. Since the parameter ε is very small in real physical situations it is possible to expand formula (136) in a power series and to neglect all terms less than $|\alpha|^2\varepsilon z \approx 1$. On the assumption of nonlinear mismatch $\Delta k_3 = 0$, the approximate equations for the quadrature variances of the third harmonic can be written in the form

$$\begin{aligned}\left\langle:\left(\Delta\hat{Q}_x\right)^2:\right\rangle &\approx 2\frac{\eta}{\beta_3^2}\Big\{3[2\cos\beta_3-\cos 2\beta_3-1-\beta_3(\sin 2\beta_3-\sin\beta_3)]\\&\qquad+(\cos\beta_3-1+\beta_3\sin\beta_3)^2\Big\}\\\left\langle:\left(\Delta\hat{P}_x\right)^2:\right\rangle &\approx 2\frac{\eta}{\beta_3^2}\Big\{3[-2\cos\beta_3+\cos 2\beta_3+1+\beta_3(\sin 2\beta_3-\sin\beta_3)]\\&\qquad+(\sin\beta_3-\beta_3\cos\beta_3)^2\Big\}\end{aligned} \tag{137}$$

where

$$\beta_3 = 3\varepsilon z|\alpha|^2 \tag{138}$$

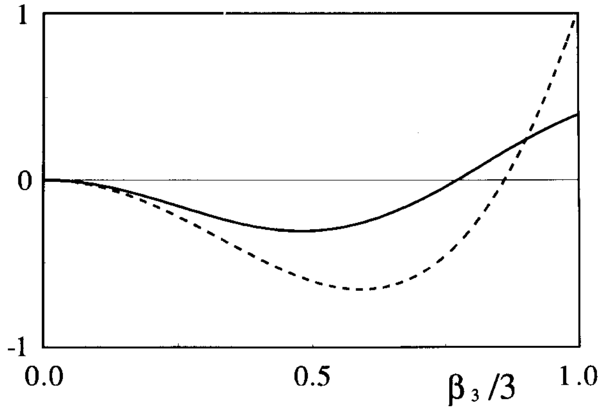

Figure 4. The approximate variance of the $\hat{Q}$ component of the fundamental field (dashed line) and the approximate variance of the $\hat{P}$ component of the third-harmonic field (solid line) are plotted versus $\beta_3/3 = \varepsilon z|\alpha|^2(\eta = 0.1)$.

and

$$\eta = \frac{3\kappa_{3\omega}^2|\alpha|^6 z^2}{|\alpha|^2} \approx \frac{I(3\omega)}{I(\omega)} \tag{139}$$

is the power-conversion ratio describing the part of the power of the fundamental that is transferred to the third harmonic.

The normally ordered variance (139) for the $\hat{P}$ component of the third-harmonic beam is plotted in Fig. 4 in comparison with the variance of the $\hat{Q}$ component of the fundamental (99). Squeezing occurs when the variances have negative values. The curves in Fig. 4 show a correlation between squeezing in the $\hat{P}$ component of the third harmonic (solid line) and self-squeezing in the $\hat{Q}$ component of the fundamental (dashed line) for small z. We can say that squeezing is transferred, in some sense, from the fundamental to the third-harmonic beam. The squeezing effect in the third harmonic depends on the conversion ratio. If η increases, then the squeezing increases too. However, we have to recall that the coupling of the third harmonic back to the fundamental beam has been ignored in our considerations. Hence, the approximation is not true for large η. Comparing Fig. 3 with Fig. 4 one can say that the correlation between the self-squeezed fundamental beam and the third harmonic is stronger than that between the fundamental and second harmonic.

VIII. CONCLUSION

In this paper we have considered the light squeezing at propagation in a nonlinear isotropic medium. The exact operator results obtained in the two quantum descriptions show a dissimilarity between the squeezing occurring in propagating light and the ordinary squeezing, briefly recalled in Section II. This effect has been named self-squeezing by Tanaś and Kielich [67, 68]. We have also analyzed the second- and third-harmonic beams, generated by self-squeezed light in an isotropic medium with a center of symmetry.

To make the medium capable of generating a wave at double frequency, an external dc field has to be applied. The classical and quantum equations describing the time evolution of the fundamental and second-harmonic beams have been derived under the assumption that the main process resided in self-interaction of the fundamental field. To discuss the squeezing in the second-harmonic beam we used the analytical form of normally ordered variances of the quadrature components. Some correlation between the squeezing in the second-harmonic and self-squeezing in the fundamental beam is found. One can say that squeezing is transferred from the fundamental into the second-harmonic beam.

In the same way, third-harmonic generation has been described (obviously without assuming an external dc electric field). The results obtained in our approach, similarly to the second-harmonic case, show correlation between squeezing in the third harmonic and self-squeezing in the fundamental. It is seen from Fig. 3 and Fig. 4 that this correlation is stronger in third-harmonic generation.

The normally ordered variances of the quadrature components of the second- and third-harmonic beams obtained in our treatment are directly proportional to the conversion ratio η. In our discussion we have taken into account that only 10% of the power of the fundamental beam is transferred into the harmonics field. For higher conversion ratio our assumption breaks down and the pairs of equations (122) and (132) have to be solved simultaneously.

We should emphasize that to obtain a considerable amount of squeezing in the second and third harmonic by the mechanism discussed in this paper, the linear mismatch should be much smaller than the intensity-dependent nonlinear mismatch.

The past few years show that interest in the optical phenomena, especially quantum phenomena, occurring in nonlinear media has been increasing steadily [128–169]. One can expect great advances in the research.

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