Squeezing in the third-harmonic field generated by self-squeezed light

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A new mechanism for producing squeezed states in the third-harmonic field generated in isotropic media is discussed. It is shown that considerable squeezing is obtained if the third-harmonic field is generated by self-squeezed light resulting from a nonlinear propagation process. Analytical formulas describing this novel way of producing squeezed states are derived and illustrated graphically, showing strong correlations of squeezing in the fundamental and the third-harmonic beams.

1. INTRODUCTION

Squeezed states of light have become a subject of intense theoretical and experimental research in the past few years. Some of the research results as well as literature on this subject can be found in the review paper by Walls.[1] Theoretical predictions have shown that squeezing of quantum fluctuations can occur in a variety of nonlinear optical processes, such as resonance fluorescence,[2–6] parametric amplification,[7–10] four-wave mixing,[11–14] the Hanle effect,[15] multiphoton absorption,[16,17] the free-electron laser,[18] the Jaynes–Cummings model,[19] harmonics generation,[20–25] nonlinear propagation of light,[24,26] and the Rydberg-atom maser.[27] A number of experiments have already been performed in order to observe squeezed states,[28–32] confirming some theoretical predictions and giving new impetus to additional studies.

In this paper we present a new mechanism for obtaining squeezed states in the third-harmonic field generated in isotropic media. It is well known[20–25] that coupling between the fundamental and harmonic beams in a nonlinear medium leads to squeezing of quantum fluctuations in the in-phase or the out-of-phase quadrature components of the fundamental and/or the harmonic beam. It has also been shown[24,26] that a strong optical field propagating in a nonlinear medium can squeeze its own quantum fluctuations; we have called this effect self-squeezing.[26] In isotropic media, in which third- (but not second-) harmonic generation is permitted by the symmetry of the medium, both processes coexist, i.e., nonlinear propagation of the fundamental beam, leading to self-squeezing, and third-harmonic generation (both of these being third-order effects). Thus, in a sense, the third-harmonic field is generated by the self-squeezed light of the fundamental beam, and the following question is posed: To what extent can squeezing be transferred from the fundamental beam to the third-harmonic beam? The answer to this question is the subject of this paper. It will be shown that a considerable amount of squeezing in the third-harmonic field can be achieved because of this mechanism.

2. SELF-SQUEEZING OF LIGHT PROPAGATING IN A NONLINEAR MEDIUM

It is well known that strong elliptically polarized light propagating through an isotropic nonlinear medium varies its state of polarization. This effect of self-induced rotation of the polarization ellipse was first reported by Maker *et al.*[33] in 1964. Since then, induced birefringence has become a standard topic in textbooks on nonlinear optics.[34,35] Recently it was shown by Tanaś and Kielich[26] that a single monochromatic beam propagating in an isotropic nonlinear medium can become squeezed. This self-induced squeezing effect was called self-squeezing. The details of the self-squeezing effect are given in Ref. 26, and we shall refrain from reproducing them here. In this paper we refer only to those results that will be needed in our subsequent calculations.

Third-order nonlinear polarization (in the electric-dipole approximation) at frequency ω can be written as follows[34,35]:

$$P_i(\omega) = \sum_{jkl} \chi_{ijkl}(-\omega, -\omega, \omega, \omega) E_j^{(-)}(\omega) E_k^{(+)}(\omega) E_l^{(+)}(\omega), \quad (1)$$

where $\chi_{ijkl}(-\omega, -\omega, \omega, \omega)$ is the third-order nonlinear susceptibility tensor of the medium and the electromagnetic field is decomposed into the positive- and negative-frequency parts

$$E_i(\mathbf{r}, t) = E_i^{(+)}(\mathbf{r}, t) + E_i^{(-)}(\mathbf{r}, t), \quad (2)$$

with

$$E_i^{(+)}(\mathbf{r}, t) = \sum_j E_i^{(+)}(\omega_j) \exp[-i(\omega_j t - \mathbf{k}_j \cdot \mathbf{r})]. \quad (3)$$

The above definition of the field gives the following expression for the intensity of the beam at frequency ω:

$$I(\omega) = \frac{cn(\omega)}{2\pi} \sum_i E_i^{(-)}(\omega) E_i^{(+)}(\omega), \quad (4)$$

where $n(\omega)$ is the refractive index of the medium for frequency ω.

For an isotropic medium with a center of inversion, the nonlinear susceptibility tensor, $\chi_{ijkl}(\omega) = \chi_{ijkl}(-\omega, -\omega, \omega, \omega)$, can be written as[34,35]

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$$\chi_{ijkl}(\omega) = \chi_{xxyy}(\omega)\delta_{ij}\delta_{kl} + \chi_{xyxy}(\omega)\delta_{ik}\delta_{jl} + \chi_{xyyx}(\omega)\delta_{il}\delta_{jk}, \quad (5)$$

with the additional relation

$$\chi_{xxxx}(\omega) = \chi_{yyyy}(\omega) = \chi_{xxyy}(\omega) + \chi_{xyxy}(\omega) + \chi_{xyyx}(\omega). \quad (6)$$

Assuming that the beam propagates along the z axis of the laboratory reference frame, inserting Eq. (5) into Eq. (1) and introducing the circular basis

$$E_{\pm}^{(+)}(\omega) = \frac{1}{\sqrt{2}}[E_x^{(+)}(\omega) \mp iE_y^{(+)}(\omega)], \quad (7)$$

one obtains for the circular components of the nonlinear polarization

$$\begin{aligned} P_{\pm}^{(+)}(\omega) &= [\chi_{xyxy}(\omega) + \chi_{xyyx}(\omega)]|E_{\pm}^{(+)}(\omega)|^2 E_{\pm}^{(+)}(\omega) \\ &\quad + [2\chi_{xxyy}(\omega) + \chi_{xyxy}(\omega) + \chi_{xyyx}(\omega)] \\ &\quad \times |E_{\mp}^{(+)}(\omega)|^2 E_{\pm}^{(+)}(\omega). \end{aligned} \quad (8)$$

Taking into account the permutation symmetry of the tensor χ with respect to the first and second pairs of indices, we have $\chi_{xyxy}(\omega) = \chi_{xyyx}(\omega)$, and formula (8) can be further simplified.

Inserting Eq. (8) as a source term into the Maxwell equations, and using the slowly varying amplitude approximation, one obtains the following equation for the amplitudes of the circular components of the field:

$$\begin{aligned} \frac{dE_{\pm}^{(+)}(\omega)}{dz} &= \frac{i2\pi\omega}{n(\omega)c}\{2\chi_{xyxy}(\omega)|E_{\pm}^{(+)}(\omega)|^2 + 2[\chi_{xxyy}(\omega) \\ &\quad + \chi_{xyxy}(\omega)]|E_{\mp}^{(+)}(\omega)|^2\}E_{\pm}^{(+)}(\omega), \end{aligned} \quad (9)$$

where the amplitudes $E_{\pm}^{(+)}(\omega)$ are assumed to be dependent on z. Equation (9) immediately shows the advantage of the circular basis used here. One can easily check that $|E_{\pm}^{(+)}(\omega)|^2$ does not depend on z, i.e., $(d/dz)|E_{\pm}^{(+)}(\omega)|^2 = 0$, and Eq. (9) has simple exponential solutions.[36]

So far, the field has been treated classically. To describe quantum effects such as squeezing, we need quantum equations of motion for the field operators. Such equations, the Heisenberg equations of motion, can be obtained from the following effective-interaction Hamiltonian[24,26]:

$$H_I = \frac{\hbar}{2}[\kappa_1(a_+^{+2}a_+^2 + a_-^{+2}a_-^2) + 2\kappa_2 a_+^+ a_-^+ a_- a_+], \quad (10)$$

where the nonlinear coupling constants κ_1 and κ_2 are real and are given by

$$\kappa_1 = \frac{V}{\hbar}\left[\frac{2\pi\hbar\omega}{n^2(\omega)V}\right]^2 2\chi_{xyxy}(\omega),$$

$$\kappa_2 = \frac{V}{\hbar}\left[\frac{2\pi\hbar\omega}{n^2(\omega)V}\right]^2 2[\chi_{xyxy}(\omega) + \chi_{xxyy}(\omega)], \quad (11)$$

with V denoting the quantization volume.

The operators $a_{\pm}$ are the annihilation operators for the circularly right- and left-polarized modes satisfying the commutation relations

$$[a_i, a_j^+] = \delta_{ij} \qquad (i, j = + \text{ or } -). \quad (12)$$

The relation between the annihilation operators, which are dimensionless, and the corresponding field operators is given by

$$E_{\pm}^{(+)}(\omega) = i\sqrt{\frac{2\pi\hbar\omega}{n^2(\omega)V}}\, a_{\pm}. \quad (13)$$

Using the interaction Hamiltonian [Eq. (10)] and the commutation rules [Eq. (12)], one can easily write the Heisenberg equations of motion describing the time evolution of the field operators. When t is replaced by $-n(\omega)z/c$, we obtain the following equation:

$$\frac{da_{\pm}(z)}{dz} = i\frac{n(\omega)}{c}[\kappa_1 a_{\pm}^+(z)a_{\pm}(z) + \kappa_2 a_{\mp}^+(z)a_{\mp}(z)]a_{\pm}(z). \quad (14)$$

The equations for the creation operators are Hermitian conjugates of Eq. (14). When relation (13) is applied, Eq. (14) reverts to the form of Eq. (9). Since the numbers of photons in the two modes $a_+^+a_+$ and $a_-^+a_-$ are constants of motion, [they commute with the Hamiltonian, Eq. (10)]; Eq. (14) has the simple exponential solution[26,37]

$$a_{\pm}(z) = \exp\{iz[\epsilon a_{\pm}^+(0)a_{\pm}(0) + \delta a_{\mp}^+(0)a_{\mp}(0)]\}a_{\pm}(0), \quad (15)$$

where we have introduced the notation

$$\epsilon = \frac{n(\omega)}{c}\kappa_1,$$

$$\delta = \frac{n(\omega)}{c}\kappa_2. \quad (16)$$

The solutions [Eq. (15)] are exact operator solutions for the field operators of light propagating through an isotropic nonlinear medium and can be used for calculations of quantum effects such as photon antibunching[37] and squeezing.[26] It has been shown that more than 90% of the squeezing allowed by quantum mechanics can be achieved in this way, and exact formulas as well as their graphical illustrations have been given in Ref. 26. In this paper we use the solutions [Eq. (15)] as a starting point (the zeroth approximation) in our calculations of squeezing in the third harmonic generated by such a field.

3. THIRD-HARMONIC GENERATION FROM SELF-SQUEEZED LIGHT

Third-harmonic generation in isotropic media is a well-known nonlinear phenomenon; see Refs. 34 and 35 and papers cited therein. To find the quantum equations describing the evolution of the annihilation and creation operators of the third harmonic, one can proceed in the same way as described in Section 2. The interaction Hamiltonian describing third-harmonic generation in isotropic media has the form

$$\begin{aligned} H_I &= \hbar\kappa_3(c_x^+a_x + c_y^+a_y)(a_x^2 + a_y^2) + \text{H.c.} \\ &= 2\hbar\kappa_3(c_+^+a_+^2a_- + c_-^+a_-^2a_+) + \text{H.c.}, \end{aligned} \quad (17)$$

where κ_3 is the nonlinear coupling constant

$$\kappa_3(z) = \frac{V}{\hbar}\sqrt{\frac{2\pi\hbar(3\omega)}{n^2(3\omega)V}}\left[\sqrt{\frac{2\pi\hbar\omega}{n^2(\omega)V}}\right]^3 \chi_{xxxx}(3\omega)\exp(i\Delta kz), \quad (18)$$

with the nonlinear susceptibility tensor component

$$\chi_{xxxx}(3\omega) = \chi_{xxxx}(-3\omega, \omega, \omega, \omega) = \chi_{xxyy}(3\omega) + \chi_{xyxy}(3\omega) + \chi_{xyyx}(3\omega). \quad (19)$$

The interaction Hamiltonian [Eq. (17)] is written in two equivalent forms by using the Cartesian or the circular basis with the annihilation operators a_x, a_y (or a_+, a_-) of the fundamental beam of frequency ω and the operators c_x, c_y (or c_+, c_-) of the third-harmonic beam. We have assumed that both beams propagate along the z axis, and $\Delta k = 3k_1 - k_3 = 3\omega/c[n(\omega) - n(3\omega)]$ is the linear phase mismatch.

Using the interaction Hamiltonian [Eq. (17)] and replacing t by $-n(\omega_j)z/c$, we obtain the following equations of motion (in the circular basis):

$$\frac{\mathrm{d}c_\pm(z)}{\mathrm{d}z} = i\,\frac{n(3\omega)}{c}\,2\kappa_3(0)a_\pm{}^2(z)a_\mp(z)\exp(i\Delta kz), \quad (19'\mathrm{a})$$

$$\frac{\mathrm{d}a_\pm(z)}{\mathrm{d}z} = i\,\frac{n(\omega)}{c}\,2\kappa_3(0)[2c_\pm(z)a_\pm{}^+(z)a_\mp{}^+(z) + c_\mp(z)a_\mp{}^{+2}(z)]\exp(-i\Delta kz). \quad (19'\mathrm{b})$$

The equations of motion in the Cartesian basis can be readily obtained from the alternative form of the Hamiltonian [Eq. (17)]. Here we prefer the circular basis because we use the solutions [Eq. (15)] in our subsequent calculations. It is seen from the form of Eqs. (19′) that, were the field classical, there would be no third-harmonic generation for a circularly polarized fundamental beam (both circular components of the fundamental beam appear on the right-hand side of the equation for the third harmonic). For quantum fields this statement is no longer true. The coupling between the operators of the third-harmonic and fundamental beams described by Eqs. (19′) leads to squeezing in the third-harmonic or the fundamental beam.[21,24] However, the fundamental beam itself varies because of its self-interaction described in Section 2, and this fact should be taken into account when we consider squeezing obtained from the third-harmonic process. In fact, we should add the two Hamiltonians [Eqs. (10) and (17)] when we write the equations of motion for the fundamental beam. Since we have already solved the evolution equations resulting from the Hamiltonian [Eq. (10)] and since the solutions are given by Eq. (15), we simply use these solutions as zero-approximation solutions in solving Eqs. (19′). Equation (19′a) can be integrated formally, giving

$$c_\pm(z) = c_\pm(0) + 2i\kappa\int_0^z a_\pm{}^2(z')a_\mp(z')\exp(i\Delta kz')\mathrm{d}z', \quad (20)$$

where we have used the shortened notation

$$\kappa = \frac{n(3\omega)}{c}\kappa_3(0). \quad (21)$$

If κ is treated as a small parameter, Eq. (20) is easy to iterate. To obtain the lowest-order solution, we insert the solutions [Eq. (15)] into the right-hand side of Eq. (20). In this paper we restrict our considerations to the lowest-order solution and ask the following question: Can the third harmonic be squeezed because it is generated by a self-squeezed fundamental beam?

Thus we completely ignore the well-known mechanism,[21,24] resulting from the coupling of Eqs. (19′), that leads to squeezing, which is, however, of higher order in κ than the effect discussed here. In practice, small κ means that only a small part of the fundamental beam intensity is transferred to the third-harmonic beam. Our approximation is thus justified if the intensity (power) conversion ratio is sufficiently low.

If the third-harmonic beam is initially, i.e., for $z = 0$, in the vacuum state and the fundamental beam is in a coherent state, then, using Eq. (20) together with the solution [Eq. (15)] and the commutation rules [Eq. (12)], one can easily obtain the following expression for the normally ordered variance of the in-phase component of the third-harmonic field component with right circular polarization:

$$\begin{aligned}\langle:[\Delta[c_+(z) + c_+{}^+(z)]]^2:\rangle &= \langle:[c_+(z) + c_+{}^+(z)]^2:\rangle - \langle c_+(z) + c_+{}^+(z)\rangle^2 \\ &= -8\kappa^2\int_0^z \mathrm{d}z'\int_0^z \mathrm{d}z''[\mathrm{Re}\,\alpha_+{}^4\alpha_-{}^2\exp(i(z'+z'')(\Delta k+\epsilon+2\delta) + \{\exp[i(z'+z'')(2\epsilon+\delta)] - 1\}|\alpha_+|^2 \\ &\quad + \{\exp[i(z'+z'')(\epsilon+2\delta)] - 1\}|\alpha_-|^2 + iz''(5\epsilon+4\delta)) - \mathrm{Re}\,\alpha_+{}^4\alpha_-{}^2\exp(i(z'+z'')(\Delta k+\epsilon+2\delta) \\ &\quad + \{\exp[iz'(2\epsilon+\delta)] + \exp[iz''(2\epsilon+\delta)] - 2\}|\alpha_+|^2 + \{\exp[iz'(\epsilon+2\delta)] + \exp[iz''(\epsilon+2\delta)] - 2\}|\alpha_-|^2) \\ &\quad - |\alpha_+|^4|\alpha_-|^2\exp(-i(z'-z'')(\Delta k+\epsilon+2\delta) + \{\exp[-i(z'-z'')(2\epsilon+\delta)] - 1\}|\alpha_+|^2 \\ &\quad + \{\exp - i(z'-z'')(\epsilon+2\delta)] - 1\}|\alpha_-|^2) + |\alpha_+|^4|\alpha_-|^2\exp(-i(z'-z'')(\Delta k+\epsilon+2\delta) \\ &\quad + \{\exp[-iz'(2\epsilon+\delta)] + \exp[iz''(2\epsilon+\delta)] - 2\}|\alpha_+|^2 + \{\exp[-iz'(\epsilon+2\delta)] + \exp[iz''(\epsilon+2\delta)] - 2\}|\alpha_-|^2)],\end{aligned} \quad (22)$$

where α_+ and α_- are the initial complex amplitudes of the circular right and left components of the fundamental beam, which was assumed to be in a coherent state. The corresponding expression for the left circular component can be obtained from Eq. (22) by replacing all the plus subscripts into minus subscripts and vice versa. The expression for the out-of-phase component can be obtained from Eq. (22) by changing the signs of the phase-sensitive Re terms. The variance [Eq. (22)] is equal to zero if there is only one circular component (α_+ or α_-) in the incoming beam. If the incoming beam is linearly polarized along the x axis, we have $\alpha_+ = \alpha_- = \alpha/\sqrt{2}$, where $|\alpha|^2$ is the average number of photons in the incoming beam. In this case formula (22) is considerably simplified but still remains quite complicated because the integrations cannot be performed analytically. To calculate the variance for the x component of the third-harmonic field, we have recourse to the relation

$$\begin{aligned}\langle:[\Delta[c_x(z) + c_x{}^+(z)]]^2:\rangle = \tfrac{1}{2}\{&\langle:[\Delta[c_+(z) + c_+{}^+(z)]]^2:\rangle \\ &+ \langle:[\Delta[c_-(z) + c_-{}^+(z)]]^2:\rangle \\ &+ 2\langle:\Delta[c_+{}^2 + c_+{}^+(z)] \\ &\times \Delta[c_-(z) + c_-{}^+(z)]:\rangle\},\end{aligned} \quad (23)$$

where the colon stands for normal ordering of the operators. To simplify formulas (22) and (23) further, we assume here that the nonlinear coupling parameters ϵ and δ, defined by Eqs. (16), have the same value. This is the case when only the transitions $J = 1 \leftrightarrow 1$ and $0 \leftrightarrow 1$ contribute to the susceptibility tensor of the medium.[37] This assumption, however, is not crucial for squeezing considerations, but it simplifies the formulas, and this is why we use it here. With this assumption, for linear polarization of the incoming beam, we have

$$\begin{aligned}\langle:[\Delta[c_x(z) + c_x{}^+(z)]]^2:\rangle &= 2\langle:[\Delta[c_+(z) + c_+{}^+(z)]]^2:\rangle\\ &= -2\kappa^2|\alpha|^6 \int_0^z \mathrm{d}z' \int_0^z \mathrm{d}z''(\exp\{[\cos 3\epsilon(z' + z'') - 1]|\alpha|^2\}\cos[(\Delta k + 3\epsilon)(z' + z'')\\ &\quad + 9\epsilon z'' + |\alpha|^2 \sin 3\epsilon(z' + z'')] - \exp[(\cos 3\epsilon z' + \cos 3\epsilon z'' - 2)|\alpha|^2]\\ &\quad \times \cos[(\Delta k + 3\epsilon)(z' + z'') + |\alpha|^2(\sin 3\epsilon z' + \sin 3\epsilon z'')] - \exp\{[\cos 3\epsilon(z' - z'') - 1]|\alpha|^2\}\\ &\quad \times \cos[(\Delta k + 3\epsilon)(z' - z'') + |\alpha|^2 \sin 3\epsilon(z' - z'')] + \exp[(\cos 3\epsilon z' + \cos 3\epsilon z'' - 2)|\alpha|^2]\\ &\quad \times \cos[(\Delta k + 3\epsilon)(z' - z'') + |\alpha|^2(\sin 3\epsilon z' - \sin 3\epsilon z'')]). \end{aligned} \tag{24}$$

In a real physical situation, the nonlinear coupling parameter ϵ is very small, and it is safe to assume that $\epsilon z \ll 1$ and that, for a great number of photons in the incoming beam ($|\alpha|^2 \gg 1$), we have $\epsilon z|\alpha|^2$ of the order of unity.

Treating ϵz as a small parameter, we can expand the integrand of Eq. (24) into a power series and retain only the leading terms. We then have ($\epsilon z \ll 1$)

$$\begin{aligned}&\langle:[\Delta[c_x(z) + c_x{}^+(z)]]^2:\rangle\\ &\approx 2\kappa^2|\alpha|^6 \int_0^z \mathrm{d}z' \int_0^z \mathrm{d}z''\{9\epsilon z'' \sin[(\Delta k + 3\epsilon|\alpha|^2)(z' + z'')]\\ &+ 18\epsilon^2|\alpha|^2 z'z'' \cos[(\Delta k + \epsilon|\alpha|^2)z']\\ &\times \cos[(\Delta k + \epsilon|\alpha|^2)z'']\}. \end{aligned} \tag{25}$$

The second term in expression (25) can be written as the square of a single integral and thus is always positive. So it is the first term that plays the crucial role in obtaining squeezing in the third harmonic. This term, proportional to 9ϵ, appears because the commutation rules [Eq. (12)] were applied; it would not appear if the fields were classical. For the out-of-phase component the sign of this term is changed. After performing the integrations in expression (25), we obtain for the in-phase component (upper sign) and the out-of-phase component (lower sign) of the third-harmonic field the following expressions for the normally ordered variances:

$$\begin{aligned}&\pm\langle:[\Delta[c_x(z) \pm c_x{}^+(z)]]^2:\rangle\\ &= \frac{18\kappa^2|\alpha|^6\epsilon z^3}{\beta^3}\left\{\pm[2\cos\beta - \cos 2\beta - 1 + \beta(\sin\beta - \sin 2\beta)]\right.\\ &\qquad \left. + \frac{2\epsilon|\alpha|^2 z}{\beta}[\cos\beta - 1 + \beta\sin\beta]^2\right\}, \end{aligned} \tag{26}$$

where we have used the notation

$$\beta = (\Delta k + 3\epsilon|\alpha|^2)z. \tag{27}$$

Within the same approximations, for the average number of photons of the third harmonic we have

$$\langle c_x{}^+(z)c_x(z)\rangle = \kappa^2|\alpha|^6 z^2 \sin^2\frac{\beta}{2}\bigg/\left(\frac{\beta}{2}\right)^2, \tag{28}$$

with intensity-dependent mismatch given by Eq. (27). The linear phase mismatch Δk can be compensated for by tuning the laser to the anomalous dispersion region and adding an appropriate amount of buffer gas.[34,35] Even if perfect linear phase matching is achieved, the nonlinear intensity-dependent mismatch appears, thereby lowering the third-harmonic intensity.[38] For perfect linear phase matching, according to Eq. (27), we have $\beta = 3\epsilon|\alpha|^2 z$, and Eq. (26) takes the form

$$\begin{aligned}\pm\langle:[\Delta[c_x(z) \pm c_x{}^+(z)]]^2:\rangle &= \frac{2\eta}{\beta^2}\{\pm 3[2\cos\beta - \cos 2\beta - 1 + \beta\\ &\quad \times (\sin\beta - \sin 2\beta)] + 2[\cos\beta - 1 + \beta\sin\beta]^2\}, \end{aligned} \tag{29}$$

where

$$\eta = \frac{3\kappa^2|\alpha|^6 z^2}{|\alpha|^2} \approx \frac{I(3\omega)}{I_0(\omega)} \tag{30}$$

is the dimensionless power-conversion ratio, the value of which determines the fraction of the initial power that has been transferred to the third harmonic. So, finally, we have derived a relatively simple formula [Eq. (29)] for the third-harmonic field variances. For comparison, we refer here to the formula for the variances of the fundamental beam's field operators, as derived from the exact formulas given in Ref. 26, when the approximations used in this paper are applied. The formula reads as

$$\begin{aligned}&\pm\langle:[\Delta[a_x(z) \pm a_x{}^+(z)]]^2:\rangle\\ &\qquad \approx \frac{2\beta}{3}\left[\frac{\beta}{3} \mp \left(\sin\frac{2\beta}{3} + \frac{\beta}{3}\cos\frac{2\beta}{3}\right)\right], \end{aligned} \tag{31}$$

where, again, $\beta = 3\epsilon|\alpha|^2 z$.

In Fig. 1 we have plotted the normally ordered variances of the in-phase component of the fundamental beam [upper sign in expression (31)] and of the out-of-phase component of the third-harmonic beam [lower sign in Eq. (29)], assuming for the conversion ratio [formula (30)] a value of $\eta = 0.1$ (10% power conversion). Negative values of these variances signify squeezing in the corresponding components of the field. A considerable amount of squeezing in the third-harmonic field can be achieved in this way, and the strong correlation between squeezing in the fundamental beam and squeezing in the third-harmonic beam is clearly visible in Fig. 1. This is a new mechanism by which squeezed states can be produced in the third-harmonic field. It consists of the generation of a third-harmonic field by the self-squeezed light of the fundamental beam. One can say, looking at Fig. 1, that squeezing from the in-phase component of the fundamental field is, in a sense, transferred to the out-of-phase component of the third-harmonic field. However, there is

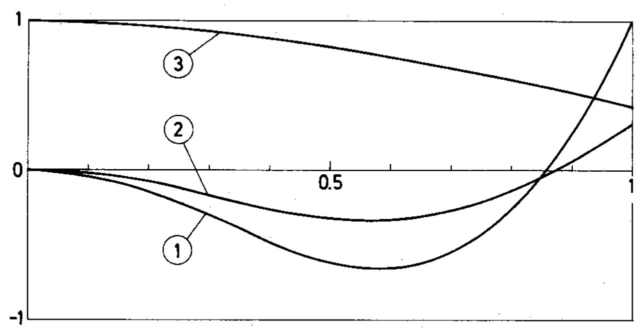

Fig. 1. The plots represent the following: 1, the normally ordered variance of the in-phase quadrature component of the fundamental beam; 2, the normally ordered variance of the out-of-phase component of the third-harmonic beam; and 3, the mismatch function $\sin^2(\beta/2)/(\beta/2)^2$. All curves are plotted versus $\beta/3 = \epsilon|\alpha|^2 z$.

no such simple correlation for the other components of the fundamental and third-harmonic fields. According to Eq. (29), the efficiency of squeezing in the third-harmonic field is proportional to the conversion ratio η and increases if η increases. We must bear in mind, however, that we have ignored the coupling of the third harmonic back to the fundamental field [Eqs. (19′b)], and our approximation breaks down for large η. We have also plotted in Fig. 1 the intensity-dependent mismatch function appearing in formula (28) for the third-harmonic intensity. Figure 1 shows that, for the values of β for which squeezing has its maximum, the intensity still retains 75% of its value for perfect phase matching.

When the linear mismatch is much greater than the intensity-dependent nonlinear mismatch, i.e., $3\epsilon|\alpha|^2 \ll |\Delta k|$, then, according to Eq. (26), the amount of squeezing is proportional to $3\epsilon|\alpha|^2/|\Delta k| \ll 1$, and only weak squeezing can be produced in the third-harmonic field.

4. CONCLUSIONS

In this paper we have considered a new way of producing squeezed states in the third-harmonic field generated in isotropic media. In such media, two nonlinear processes, both of third order, take place simultaneously: (1) nonlinear propagation of the optical field at frequency ω and (2) third-harmonic generation. It has been shown[24,26] that nonlinear propagation of the fundamental beam can lead to self-squeezing of light. On the other hand, it has also been shown[21,24] that squeezing can be obtained in the third-harmonic field without taking into account the changes in the fundamental beam caused by nonlinear propagation. In this paper we have shown that nonlinear propagation of the optical field at frequency ω can be a source of squeezing in the third-harmonic field thereby generated. This mechanism of producing squeezed states in the third-harmonic field is completely different from the one already known. The fundamental beam becomes self-squeezed in the nonlinear propagation process, and this squeezing is transferred to the third-harmonic beam during the generation process. We have obtained analytical formulas that describe the normally ordered variances of the in-phase and out-of-phase quadrature components of the third-harmonic field. We have shown that a considerable amount of squeezing in the third harmonic can be obtained in this way. If 10% of the initial power is transferred to the third harmonic, the amount of squeezing in the third-harmonic field reaches 34% of the value allowed by quantum mechanics. This amount can be even higher if the conversion ratio increases. For higher conversion ratios, however, the approximation used in this paper breaks down, and Eqs. (19′) should be used to solve the problem; however, this is a difficult task. It was shown[21,24] that the iterative solution of Eqs. (19′), when nonlinear propagation effects are not considered, leads to squeezing of the order of κ^4 in the third harmonic. So the nonlinear propagation effect considered here, which is of the order of κ^2, should be dominant whenever κ is small.

Squeezing, which is a phase-sensitive effect, will generally depend on the initial phase of the field and may not attain its maximum value for our choice of the phases (in-phase and out-of-phase components). For the propagation effect, the initial phase dependence was discussed in Ref. 26. To keep the formulas as simple as possible we do not discuss this dependence here.

We should also emphasize that, to obtain considerable squeezing in the third harmonic, the linear mismatch that is due to dispersion of the refractive index of the medium should be made much smaller than the intensity-dependent nonlinear mismatch. On the other hand, a large nonlinear mismatch lowers the third-harmonic intensity[38] and does not permit us to obtain conversion ratios close to unity. This difficulty may be overcome by using beam focusing to compensate for the nonlinear phase mismatch.[39]

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REFERENCES

1. D. F. Walls, Nature **306**, 141 (1983), and references therein.
2. D. F. Walls and P. Zoller, Phys. Rev. Lett. **47**, 709 (1981).
3. L. Mandel, Phys. Rev. Lett. **49**, 136 (1982).
4. Z. Ficek, R. Tanaś, and S. Kielich, Opt. Commun. **46**, 23 (1984); Phys. Rev. A **29**, 2004 (1984).
5. W. Vogel and D. G. Welsch, Phys. Rev. Lett. **54**, 1802 (1985).
6. A. Heidmann and S. Reynaud, J. Phys. (Paris) **46**, 1937 (1985).
7. D. Stoler, Phys. Rev. Lett. **33**, 1397 (1974).
8. G. J. Milburn and D. F. Walls, Opt. Commun. **39**, 401 (1981); Phys. Rev. A **27**, 392 (1983).
9. H. J. Carmichael, G. J. Milburn, and D. F. Walls, J. Phys. A **17**, 469 (1984).
10. C. M. Caves and B. L. Schumaker, Phys. Rev. A **31**, 3068 (1985).
11. H. P. Yuen and J. H. Shapiro, Opt. Lett. **4**, 334 (1979).
12. R. S. Bondurand, P. Kumar, J. H. Shapiro, and M. Maeda, Phys. Rev. A **30**, 343 (1984); P. Kumar and J. H. Shapiro, Phys. Rev. A **30**, 1568 (1984).
13. J. Peřina, V. Peřinova, C. Sibilia, and M. Bertolotti, Opt. Commun. **49**, 285 (1984).
14. M. D. Reid and D. F. Walls, Phys. Rev. A **31**, 1622 (1985).
15. P. A. Lakshmi and G. S. Agarwal, Phys. Rev. A **32**, 1643 (1985).
16. M. S. Zubairy, M. S. K. Razmi, S. Iqbal, and M. Idress, Phys. Lett. A **98**, 168 (1983).
17. R. Loudon, Opt. Commun. **49**, 67 (1984).
18. W. Becker, M. O. Scully, and M. S. Zubairy, Phys. Rev. Lett. **48**, 475 (1982).
19. P. Meystre and M. S. Zubairy, Phys. Lett. A **89**, 390 (1982).
20. L. Mandel, Opt. Commun. **42**, 437 (1982).
21. M. Kozierowski and S. Kielich, Phys. Lett. A **94**, 213 (1983).
22. L. A. Lugiato, G. Strini, and F. de Martini, Opt. Lett. **8**, 256 (1983).

23. J. Peřina, V. Peřinova, and J. Kodousek, Opt. Commun. **49,** 210 (1984).
24. S. Kielich, M. Kozierowski, and R. Tanaś, Opt. Acta **32,** 1023 (1985).
25. M. J. Collett and D. F. Walls, Phys. Rev. A **32,** 2887 (1985).
26. R. Tanaś and S. Kielich, Opt. Commun. **45,** 351 (1983); Opt. Acta **31,** 81 (1984).
27. A. Heidmann, J. M. Raimond, S. Reynaud, and N. Zagury, Opt. Commun. **54,** 189 (1985).
28. M. D. Levenson, R. M. Shelby, A. Aspect, M. D. Reid, and D. F. Walls, Phys. Rev. A **32,** 1550 (1985).
29. M. W. Maeda, P. Kumar, and J. H. Shapiro, Phys. Rev. A **32,** 3803 (1985).
30. R. E. Slusher, L. W. Hollberg, B. Yurke, J. C. Mertz, and J. F. Valley, Phys. Rev. Lett. **55,** 2409 (1985).
31. M. D. Levenson, R. M. Shelby, R. G. DeVoe, and S. H. Perlmutter, in *Digest of International Quantum Electronics Conference* (Optical Society of America, Washington, D.C., 1986), p. 56.
32. H. J. Kimble and J. L. Hall, in *Digest of International Quantum Electronics Conference* (Optical Society of America, Washington, D.C., 1986), p. 86.
33. P. D. Maker, R. W. Terhune, and C. M. Savage, Phys. Rev. Lett. **12,** 57 (1964).
34. S. Kielich, *Nonlinear Molecular Optics* (Nauka, Moscow, 1981).
35. Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1985).
36. P. L. Kelley, Phys. Rev. Lett. **15,** 1005 (1965).
37. H. H. Ritze, Z. Phys. B **39,** 353 (1980).
38. J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127,** 1918 (1962).
39. G. V. Mironov, A. K. Popov, and V. V. Slabko, Opt. Quantum Electron. **17,** 435 (1985).

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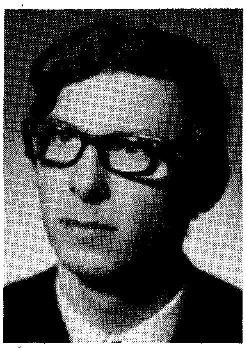

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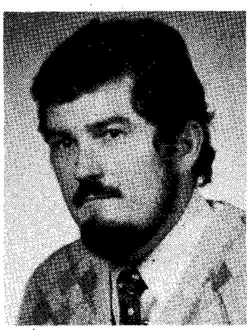

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