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On the possibility of almost complete self-squeezing of strong electromagnetic fields

R. TANAŚ and S. KIELICH
Nonlinear Optics Division, Institute of Physics,
A. Mickiewicz University, 60-780 Poznań, Poland

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Abstract. The propagation of quantized electromagnetic fields through optically isotropic media with cubic optical nonlinearity is considered. Analytical solutions are presented in closed form showing that the light field can emerge from the medium in a squeezed quantum state. A detailed numerical analysis of the results is performed and presented graphically. Over 90 per cent of the squeezing permitted by quantum-mechanical theory is achieved in this way. The dependence of the squeezing effect on the polarization state of the field and the nonlinear molecular parameters is also discussed.

1. Introduction

The problem of generating so-called 'squeezed states' of an electromagnetic field, which allows the uncertainty in the measurement of one observable to be reduced at the expense of an increased uncertainty in the measurement of another non-commuting observable, has recently attracted much attention. The possibility of reducing quantum fluctuations in one quadrature component of the field seems to be very promising in overcoming quantum limits in the detection of gravity waves [1] and for the use of such a field in communication systems [2]. These advantages of squeezed states have greatly stimulated research, and in a number of theoretical studies in nonlinear optics, various schemes have been considered as possible sources of such states: degenerate parametric amplification [3, 4], resonance fluorescence [5], degenerate four-wave mixing [6]. optical bistability [7], free electron lasers [8], the Jaynes–Cummings model [9] and harmonic generation of a laser beam [10, 11].

Squeezing, like photon antibunching [12–14], is a purely quantum effect and, although there is no direct connection between the two (field states exist that exhibit the former but not the latter, and vice versa), they are often encountered together. Neither of them has a classical analogue in the sense that they cannot be described by a non-negative diagonal coherent-state representation [15, 16], and they are of fundamental importance on their own. Unlike photon antibunching, however, squeezing has yet to be observed experimentally. Walls and Zoller [5] have asserted that squeezing should be observable under experimental conditions similar to those pertaining when observing photon antibunching in resonance fluorescence from a coherently driven two-level atom [17–19]. Mandel [20] has shown that the detection of the squeezed state by phase-sensitive interference with another optical field in the coherent state, followed by photoelectric detection of the resulting intensity fluctuations, always gives rise to sub-poissonian photon statistics; however, he concluded that this way of detecting squeezing in the resonance fluorescence of a single coherently driven atom is much more difficult than the direct detection of photon antibunching. Generally, the theoretically predicted values of squeezing are very small in the schemes considered so far.

As we have shown previously [21], using a perturbative approach, photon antibunching is possible in the self-induced rotation of the polarization ellipse during the propagation of a strong, elliptically polarized light beam through a nonlinear isotropic medium. Our results have since been confirmed by Ritze [22], who used a different approach and obtained the strict, non-perturbative solution of the problem. (The self-induced rotation of the polarization ellipse was first reported in 1964 by Maker *et al.* [23]: for a review of the results and theoretical developments, see [24, 25].)

In this paper we show that the process of light propagation in a nonlinear, optically isotropic medium can also produce squeezed states of the electromagnetic field. The non-perturbative solution of the problem will be given, and the dependence of squeezing on the polarization of the incoming beam will be discussed; it will be shown that, in our scheme, over 90 per cent of the squeezing permitted by quantum-mechanical theory can be achieved in this way.

2. Interaction hamiltonian and equations of motion

It is convenient to write the electromagnetic field as the sum of positive and negative frequency parts:

$$E_\sigma(\mathbf{r},t)=E_\sigma^{(+)}(\mathbf{r},t)+E_\sigma^{(-)}(\mathbf{r},t), \tag{1}$$

where σ denotes the polarization component of the field. It is also useful to perform a mode decomposition of this field. For plane-wave decomposition of the free field, we have

$$E_\sigma^{(+)}(\mathbf{r},t)=\sum_{\mathbf{k},\lambda} i\left(\frac{2\pi\hbar\omega_k}{V}\right)^{1/2} e_{k\sigma}^{(\lambda)} a_{k\lambda}\exp\left[-i(\omega_k t-\mathbf{k}\cdot\mathbf{r})\right], \tag{2}$$

where $e_{k\sigma}^{(\lambda)}$ is the σ component of the polarization vector associated with the polarization state λ and the propagation vector $\mathbf{k}$, and V is the quantization volume. For the quantized field, $a_{k\lambda}$ is the annihilation operator of the photon with the propagation vector $\mathbf{k}$ and polarization λ, fulfilling the commutation rules

$$[a_{k\lambda}, a_{k'\lambda'}^{+}]=\delta_{kk'}\delta_{\lambda\lambda'}. \tag{3}$$

The polarization vectors fulfil the orthogonality conditions

$$\left.\begin{aligned} e_{k\sigma}^{(\lambda)*}e_{k\sigma}^{(\lambda')}&=\delta_{\lambda\lambda'},\\ e_{k\sigma}^{(\lambda)}k_\sigma&=0,\end{aligned}\right\} \tag{4}$$

and summation in equations (4) is to be performed over the repeated greek indices.

For a monochromatic field of frequency ω propagating along the z axis of the laboratory reference frame, we can drop the index k in the above notation and write

$$E_\sigma^{(+)}(z,t)=i\left(\frac{2\pi\hbar\omega}{V}\right)^{1/2}\exp\left[-i(\omega t-kz)\right]\sum_{\lambda=1,2} e_\sigma^{(\lambda)}a_\lambda, \tag{5}$$

with $k=\omega/c$. In equation (5) the sum over two possible mutually orthogonal polarizations of the field remains, so that we still have a two-mode description of the field. If the field is a coherent superposition of these two modes, however, the two-mode description can be replaced by one mode of a (generally) elliptically polarized field:

$$e_\sigma a = e_\sigma^{(1)} a_1 + e_\sigma^{(2)} a_2, \tag{6}$$

where $e_\sigma^{(1)}$ and $e_\sigma^{(2)}$ are the σ components of the orthogonal unit polarization vectors $\mathbf{e}^{(1)}$ and $\mathbf{e}^{(2)}$ of the modes a_1 and a_2, and e_σ is the σ component of the polarization vector of the mode a. The relation (6) can also be considered in the reverse sense as a decomposition of initially elliptically polarized light into two orthogonal modes. Applying the orthogonality condition (4) for the polarization vectors, we obtain the formula

$$a = e_1^* a_1 + e_2^* a_2 \tag{7}$$

where

$$e_1^* = \mathbf{e}^* \cdot \mathbf{e}^{(1)}, \qquad e_2^* = \mathbf{e}^* \cdot \mathbf{e}^{(2)}.$$

So far the decomposition (6) (or, equivalently, (7)) is quite general and can be further specified—either for two modes with mutually perpendicular linear polarizations or for right- and left-circularly polarized modes.

Assuming the two modes to be linearly polarized along the x and y axes, we obtain from equation (7)

$$a = e_x^* a_x + e_y^* a_y, \tag{8}$$

with e_x and e_y given by [25]

$$\left. \begin{aligned} e_x &= \cos\eta \cos\theta - i \sin\eta \sin\theta, \\ e_y &= \cos\eta \sin\theta + i \sin\eta \cos\theta, \end{aligned} \right\} \tag{9}$$

where θ denotes the azimuth and η the ellipticity of the polarization ellipse of the incident beam.

On the other hand, on introducing the circular basis associated with the right-circular polarization vector, $\mathbf{e}^{(1)} = (\hat{\mathbf{x}} + i\hat{\mathbf{y}})/\sqrt{2}$ and the left-circular polarization vector, $\mathbf{e}^{(2)} = (\hat{\mathbf{x}} - i\hat{\mathbf{y}})/\sqrt{2}$ (where $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are the unit vectors along x and y, respectively), we have in agreement with equations (6) and (7)

$$\left. \begin{aligned} a_1 &= \frac{1}{\sqrt{2}} (a_x - i a_y), \\ a_2 &= \frac{1}{\sqrt{2}} (a_x + i a_y). \end{aligned} \right\} \tag{10}$$

Both bases can be used alternatively to describe the interaction of elliptically polarized light with the system of molecules to be considered in this paper.

Let us consider the interaction of an intense light beam and an isotropic medium consisting of N atoms or molecules in a cavity of volume V. The interaction between the light and an individual atom in the electric-dipole approximation, discarding non-energy-conserving terms, is described by the effective hamiltonian

$$H_{\mathrm{I}} = -\alpha_{\sigma\tau}(\omega) E_\sigma^{(-)} E_\tau^{(+)} - \tfrac{1}{2}\gamma_{\sigma\tau\nu\rho}(\omega) E_\sigma^{(-)} E_\tau^{(-)} E_\nu^{(+)} E_\rho^{(+)}, \tag{11}$$

where

$$\alpha_{\sigma\tau}(\omega)=\alpha_{\sigma\tau}(-\omega,\omega)$$

$$\gamma_{\sigma\tau\nu\rho}(\omega)=\gamma_{\sigma\tau\nu\rho}(-\omega,-\omega,\omega,\omega)$$

are the polarizability and hyperpolarizability tensors of the molecule [24], and the field operators $E_\sigma^{(+)}=[E_\sigma^{(-)}]^+$ are given by equation (5). Again, the summation is to be performed over repeated greek indices. We assume here that the light beam is initially a single radiation mode of frequency ω, elliptically polarized and propagating in the z direction of the laboratory reference frame (x, y, z).

We are interested in the field evolution produced by nonlinear interaction with the medium. In the Heisenberg picture, the time evolution of any operator A is given by the equation

$$\frac{dA}{dt}=\frac{1}{i\hbar}[A,H], \tag{12}$$

where H is the total hamiltonian which, in our case, is the sum of the free field hamiltonian H_F and the interaction hamiltonian H_I. For A equal to $E_\sigma^{(+)}$, the commutator with H_F gives the 'rapid' time evolution of the free field already expressed in equations (2) and (5). The commutator with H_I gives the 'slow' part of the evolution resulting from interaction with the medium. The 'rapid' evolution of the free field can be eliminated from equation (12) by the introduction of 'slowly varying' operators, the evolution of which are described by H_I only.

Before proceeding to the equations of motion for the field operators, however, we shall simplify our interaction hamiltonian (11). Since we assume that the medium is composed of freely orienting molecules, the molecular polarizability and hyperpolarizability tensors have to be isotropically averaged over all possible orientiations Ω of the molecule, giving

$$\left.\begin{aligned}\langle\alpha_{\sigma\tau}(\omega)\rangle_\Omega&=\alpha(\omega)\delta_{\sigma\tau},\\ \langle\gamma_{\sigma\tau\nu\rho}(\omega)\rangle_\Omega&=\gamma_1(\omega)\delta_{\sigma\tau}\delta_{\nu\rho}+\gamma_2(\omega)\delta_{\sigma\nu}\delta_{\tau\rho}\\ &\quad+\gamma_3(\omega)\delta_{\sigma\rho}\delta_{\tau\nu},\end{aligned}\right\} \tag{13}$$

where $\alpha(\omega)$, $\gamma_1(\omega)$, $\gamma_2(\omega)$ and $\gamma_3(\omega)$ are the rotational invariants of these tensors, given by [24]

$$\left.\begin{aligned}\alpha(\omega)&=\tfrac{1}{3}\alpha_{\alpha\alpha}(\omega),\\ \gamma_1(\omega)&=\tfrac{1}{30}[4\gamma_{\alpha\alpha\beta\beta}(\omega)-\gamma_{\alpha\beta\alpha\beta}(\omega)-\gamma_{\alpha\beta\beta\alpha}(\omega)],\\ \gamma_2(\omega)&=\tfrac{1}{30}[4\gamma_{\alpha\beta\alpha\beta}(\omega)-\gamma_{\alpha\beta\beta\alpha}(\omega)-\gamma_{\alpha\alpha\beta\beta}(\omega)],\\ \gamma_3(\omega)&=\tfrac{1}{30}[4\gamma_{\alpha\beta\beta\alpha}(\omega)-\gamma_{\alpha\alpha\beta\beta}(\omega)-\gamma_{\alpha\beta\alpha\beta}(\omega)];\end{aligned}\right\} \tag{14}$$

$\alpha(\omega)$ is simply the mean polarizability of the molecule. The values taken by $\gamma_1(\omega)$, $\gamma_2(\omega)$ and $\gamma_3(\omega)$ generally depend on the permutation symmetry of the tensor $\gamma_{\alpha\beta\gamma\delta}(\omega)$ as well as the molecular symmetry. If the tensor $\gamma_{\alpha\beta\gamma\delta}(\omega)$ is symmetric with respect to all possible permutations of its indices, then $\gamma_1(\omega)=\gamma_2(\omega)=\gamma_3(\omega)$, regardless of the symmetry point group of the molecule. For permutational symmetry with respect to the first and second pairs of indices only, $\gamma_1(\omega)\neq\gamma_2(\omega)=\gamma_3(\omega)$ and nonlinear asymmetry appears. Here, for later use, we introduce a nonlinear asymmetry parameter d, defined as

$$2d = 1 + \frac{2\gamma_1(\omega)}{\gamma_2(\omega)+\gamma_3(\omega)}. \tag{15}$$

If $\gamma_{\alpha\beta\gamma\delta}(\omega)$ is symmetric in all its indices, d thus defined is always equal to 1; however, for symmetry with respect to the pairs of indices $(\alpha\beta)$ and $(\gamma\delta)$ only, $d \neq 1$ and describes the asymmetry of the nonlinear properties of the molecule. For molecules with high symmetry (point groups O or T), the relations [24]

$$\left.\begin{aligned} \gamma_{\alpha\alpha\beta\beta}(\omega) &= 3[\gamma_{3333}(\omega)+2\gamma_{1133}(\omega)] \\ \gamma_{\alpha\beta\alpha\beta}(\omega) &= \gamma_{\alpha\beta\beta\alpha}(\omega) = 3[\gamma_{3333}(\omega)+2\gamma_{1313}(\omega)] \end{aligned}\right\} \tag{16}$$

hold, and equation (15) may be rewritten in the form

$$2d = 1 + \frac{\gamma_{3333}(\omega)+4\gamma_{1133}(\omega)-2\gamma_{1313}(\omega)}{\gamma_{3333}(\omega)-\gamma_{1133}(\omega)+3\gamma_{1313}(\omega)}. \tag{17}$$

According to equations (5)–(10) and (13), the interaction hamiltonian (11) can be transformed to two alternative, equivalent forms depending on whether the decomposition is made into linearly or circularly polarized modes. For two orthogonal linear polarizations, we have that

$$\begin{aligned} H_{\mathrm{I}} = -\tilde{\alpha}(\omega)(a_x^+ a_x + a_y^+ a_y) - \tfrac{1}{2}\{\tilde{\gamma}_1(\omega)(a_x^{+2}+a_y^{+2})(a_x^2+a_y^2) \\ + [\tilde{\gamma}_2(\omega)+\tilde{\gamma}_3(\omega)](a_x^{+2}a_x^2 + a_y^{+2}a_y^2 + 2a_x^+ a_y^+ a_y a_x)\} \end{aligned} \tag{18 a}$$

and, for two circular polarizations,

$$\begin{aligned} H_{\mathrm{I}} = -\tilde{\alpha}(\omega)(a_1^+ a_1 + a_2^+ a_2) - \tfrac{1}{2}\{4\tilde{\gamma}_1(\omega)a_1^+ a_2^+ a_2 a_1 \\ + [\tilde{\gamma}_2(\omega)+\tilde{\gamma}_3(\omega)](a_1^{+2}a_1^2 + a_2^{+2}a_2^2 + 2a_1^+ a_2^+ a_2 a_1)\}. \end{aligned} \tag{18 b}$$

where we have introduced the abbreviations

$$\tilde{\alpha}(\omega) = \frac{2\pi\hbar\omega}{V}\alpha(\omega) \quad \text{and} \quad \tilde{\gamma}_i(\omega) = \left(\frac{2\pi\hbar\omega}{V}\right)^2 \gamma_i(\omega).$$

Although both forms, (18 *a*) and (18 *b*), of the interaction hamiltonian are equivalent (either may be transformed into the other by using equations (10)), they nevertheless show explicitly an important difference in the choice between a cartesian and a circular basis. It is easy to check that H_{I}, as given by equation (18 *b*), commutes with $a_1^+ a_1$ and $a_2^+ a_2$, whereas H_{I} given by equation (18 *a*) does not commute with $a_x^+ a_x$ or $a_y^+ a_y$. This means that $a_1^+ a_1$ and $a_2^+ a_2$ are constants of motion, but $a_x^+ a_x$ and $a_y^+ a_y$ are not. This is a clear advantage of the circular basis over the cartesian basis. At this point we wish to emphasize that elliptically polarized light which, in an optically linear medium, can be treated as one mode, can no longer be dealt with as one mode when nonlinear interaction occurs, even in an optically isotropic medium. Moreover, since $a_x^+ a_x$ and $a_y^+ a_y$ are not constants of motion, linear polarization is not preserved at such an interaction.

The interaction hamiltonian (18 *a*) or (18 *b*) is now inserted into equation (12) to obtain the time evolution of the operators a_x and a_y or a_1 and a_2. Since the problem is one of propagation rather than of a field in a cavity we perform the transformation $z=-ct$. As a result, the quantum equations of motion for the slowly varying parts (with free evolution eliminated) of the field operators in the two representations take the form

$$\frac{d}{dz}a_x(z)=-\frac{iN}{\hbar c}\{\tilde{\alpha}(\omega)a_x(z)+\tilde{\gamma}_1(\omega)a_x^+(z)[a_x^2(z)+a_y^2(z)] + [\tilde{\gamma}_2(\omega)+\tilde{\gamma}_3(\omega)][a_x^+(z)a_x(z)+a_y^+(z)a_y(z)]a_x(z)\}, \tag{19 a}$$

and

$$\frac{d}{dz}a_1(z)=-\frac{iN}{\hbar c}\{\tilde{\alpha}(\omega)+[\tilde{\gamma}_2(\omega)+\tilde{\gamma}_3(\omega)]a_1^+(z)a_1(z) + [2\tilde{\gamma}_1(\omega)+\tilde{\gamma}_2(\omega)+\tilde{\gamma}_3(\omega)]a_2^+(z)a_2(z)\}a_1(z), \tag{19 b}$$

The equation for the operator $a_y(a_2)$ can be obtained from equations (19) by making the interchange $x\leftrightarrow y(1\leftrightarrow 2)$, and the corresponding equations for the creation operators by taking the hermitian conjugate of equations (19). We have taken into account in equations (19) that the number of molecules interacting with the field is N.

In a previous paper [21] we solved equations (19 *a*) perturbatively, showing the possibility of photon antibunching. As Ritze has shown [22], equations (19 *b*) can be solved strictly provided there is no absorption in the system. The solutions of equation (19 *a*) are then given by

$$\left.\begin{aligned} a_1(z)&=\exp\{i[\varphi(z)+\varepsilon(z)(a_1^+(0)a_1(0)+2da_2^+(0)a_2(0)]\}a_1(0)\\ a_2(z)&=\exp\{i[\varphi(z)+\varepsilon(z)(a_2^+(0)a_2(0)+2da_1^+(0)a_1(0))]\}a_2(0),\end{aligned}\right\} \tag{20}$$

where $2d$ is defined by equation (15) or (17), and use is made of the notation

$$\left.\begin{aligned} \varphi(z)&=-\frac{Nz}{\hbar c}\tilde{\alpha}(\omega),\\ \varepsilon(z)&=-\frac{Nz}{\hbar c}[\tilde{\gamma}_2(\omega)+\tilde{\gamma}_3(\omega)].\end{aligned}\right\} \tag{21}$$

With this notation, clear correspondence is established between our phenomenological molecular parameters and the parameters calculated by Ritze [22] for a particular atomic level structure.

3. Self-squeezing of light propagating in a nonlinear medium

In order to discuss the problem of squeezing, let us introduce the canonical variables 'coordinate' Q and 'momentum' P [10]:

$$Q_\sigma=a_\sigma+a_\sigma^+ \quad \text{and} \quad P_\sigma=-i(a_\sigma-a_\sigma^+), \quad \sigma=1,2 \quad \text{or} \quad x,y \tag{22}$$

which obey the commutation relation

$$[Q_\sigma, P_{\sigma'}]=2i\delta_{\sigma\sigma'}. \tag{23}$$

A squeezed state of the electromagnetic field is defined [5] as a quantum state in which the square of the uncertainty of either Q_σ or P_σ is less than unity:

$$\langle(\Delta Q_\sigma)^2\rangle < 1 \quad \text{or} \quad \langle(\Delta P_\sigma)^2\rangle < 1, \tag{24}$$

where $\Delta Q_\sigma = Q_\sigma - \langle Q_\sigma\rangle$. On introducing normal ordering of the operators, the definition (24) can be rewritten in the form [5, 20]

$$\langle:(\Delta Q_\sigma)^2:\rangle < 0 \quad \text{or} \quad \langle:(\Delta P_\sigma)^2:\rangle < 0. \tag{25}$$

The solutions (20) for the field operators on traversal of the path z in the nonlinear medium are now inserted into the definition (25), and the expectation value in the quantum state of the incoming beam is taken. If one of the resulting values is negative, the corresponding component of the field (the in-phase component Q_σ or the out-of-phase component P_σ) of the outgoing beam is squeezed.

Assuming the incoming beam to be in the coherent state $|\alpha\rangle$ defined with respect to the operator $a(0)$ given by equation (7), i.e.

$$a(0)|\alpha\rangle = \alpha|\alpha\rangle, \tag{26}$$

the appropriate expectation values can be calculated explicitly, giving in circular representation

$$\begin{aligned}\langle:(\Delta Q_1(z))^2:\rangle &= \langle:(a_1(z)+a_1^+(z))^2:\rangle - \langle a_1(z)+a_1^+(z)\rangle^2\\ &= 2\,\mathrm{Re}\,(\alpha_1^2 \exp\{2i\varphi(z)+i\varepsilon(z)+[\exp(2i\varepsilon(z))-1]|\alpha_1|^2\\ &\quad +[\exp(4id\varepsilon(z)-1]|\alpha_2|^2\} - \alpha_1^2\exp\{2i\varphi(z)\\ &\quad +2[\exp(i\varepsilon(z))-1]|\alpha_1|^2 + 2[\exp(2id\varepsilon(z))-1]|\alpha_2|^2\})\\ &\quad +2|\alpha_1|^2\{1-\exp[2(\cos\varepsilon(z)-1)|\alpha_1|^2\\ &\quad +2(\cos 2d\varepsilon(z)-1)|\alpha_2|^2]\}\end{aligned} \tag{27}$$

where, in accordance with equations (26), (6), (9) and (10),

$$\left.\begin{aligned}\alpha_1 &= \frac{1}{\sqrt{2}}(\cos\eta+\sin\eta)\exp(-i\theta)\alpha,\\ \alpha_2 &= \frac{1}{\sqrt{2}}(\cos\eta-\sin\eta)\exp(i\theta)\alpha,\\ |\alpha|^2 &= |\alpha_1|^2+|\alpha_2|^2.\end{aligned}\right\} \tag{28}$$

Similarly,

$$\langle:(\Delta P_1(z))^2:\rangle = -2\,\mathrm{Re}\,(\ldots) + 2|\alpha_1|^2\{\ldots\}, \tag{29}$$

where the expressions in parentheses and braces are the same as in equation (27).

In order to obtain suitable formulae for the operators $Q_2(z)$ and $P_2(z)$, it suffices to interchange the indices 1 and 2 in equations (27) and (29), whereas for the operators $Q_x(z)$ and $P_x(z)$ we have that

$$\begin{aligned}
\langle:(\Delta Q_x(z))^2:\rangle &= \langle:(a_x(z)+a_x^+(z))^2:\rangle - \langle a_x(z)+a_x^+(z)\rangle^2 \\
&= \tfrac{1}{2}[\langle:(a_1(z)+a_2(z)+a_1^+(z)+a_2^+(z))^2:\rangle \\
&\quad -\langle a_1(z)+a_2(z)+a_1^+(z)+a_2^+(z)\rangle^2] \\
&= \mathrm{Re}\,(\alpha_1^2 \exp\{2i\varphi(z)+i\varepsilon(z)+[\exp(2i\varepsilon(z))-1]|\alpha_1|^2 \\
&\quad +[\exp(4id\varepsilon(z))-1]|\alpha_2|^2\} \\
&\quad -\alpha_1^2 \exp\{2i\varphi(z)+2[\exp(i\varepsilon(z))-1]|\alpha_1|^2 \\
&\quad +2[\exp(2id\varepsilon(z))-1]|\alpha_2|^2\}+\alpha_2^2 \exp\{2i\varphi(z)+i\varepsilon(z) \\
&\quad +[\exp(2i\varepsilon(z))-1]|\alpha_2|^2+[\exp(4id\varepsilon(z))-1]|\alpha_1|^2\} \\
&\quad -\alpha_2^2 \exp\{2i\varphi(z)+2[\exp(i\varepsilon(z))-1]|\alpha_2|^2 \\
&\quad +2[\exp(2id\varepsilon(z))-1]|\alpha_1|^2\} \\
&\quad +2\alpha_1\alpha_2 \exp[\![2i\varphi(z)+2id\varepsilon(z)+\{\exp[i(1+2d)\varepsilon(z)]-1\}|\alpha|^2]\!] \\
&\quad -2\alpha_1\alpha_2 \exp\{2i\varphi(z)+[\exp(i\varepsilon(z))+\exp(2id\varepsilon(z))-2]|\alpha|^2\}) \\
&\quad +\{|\alpha|^2-|\alpha_1|^2 \exp[2(\cos\varepsilon(z)-1)|\alpha_1|^2+2(\cos 2d\varepsilon(z)-1)|\alpha_2|^2] \\
&\quad -|\alpha_2|^2 \exp[2(\cos\varepsilon(z)-1)|\alpha_2|^2+2(\cos 2d\varepsilon(z)-1)|\alpha_1|^2] \\
&\quad +2\,\mathrm{Re}\,[\alpha_1^*\alpha_2 \exp(\{\exp[-i(1-2d)\varepsilon(z)]-1\}|\alpha_1|^2 \\
&\quad +\{\exp[i(1-2d)\varepsilon(z)]-1\}|\alpha_2|^2) \\
&\quad -\alpha_1^*\alpha_2 \exp\{[\exp(-i\varepsilon(z))+\exp(2id\varepsilon(z))-2]|\alpha_1|^2 \\
&\quad +[\exp(i\varepsilon(z))+\exp(-2id\varepsilon(z))-2]|\alpha_2|^2\}]\}.
\end{aligned} \tag{30}$$

and

$$\langle:(\Delta P_x(z))^2:\rangle = -\mathrm{Re}\,(\ldots)+\{\ldots\}, \tag{31}$$

where the expressions in parentheses and braces are the same as in equation (30).

The expressions (27)–(31) are exact analytical formulae describing the fluctuations in each particular component of the field in the outgoing beam after its traversal of the path z in the nonlinear medium. Because of their complexity it is not easy to say without a detailed numerical analysis whether they are negative or not. We have performed such an analysis, and the results are displayed in graphical form in figures 1–8.

Our formulae involve quite a number of parameters. Throughout the numerical calculations, we assume that $\varepsilon(z)$, which is given by equation (21), has the value 10^{-6}. Obviously, $\varepsilon(z)$ depends on the hyperpolarizability of the molecule, the number of molecules N and the length of the medium z; however, it has been shown [26] that it is quite possible to have a medium for which $\varepsilon(z)$ is of the order of 10^{-6}. The choice of a particular value for $\varepsilon(z)$ is not crucial to our calculations because it is $\varepsilon(z)|\alpha|^2$ rather than $\varepsilon(z)$ to which the results are sensitive. The complex number α can be written in the form

$$\alpha = |\alpha| \exp(i\varphi_0), \tag{32}$$

where φ_0 denotes the initial phase of the incoming beam and $|\alpha|^2$ is the mean number of photons in it.

Figure 1 shows the overall behaviour of $\langle:(\Delta Q_x(z))^2:\rangle$ and $\langle:(\Delta P_x(z))^2:\rangle$ as functions of $\varepsilon(z)|\alpha|^2$, where we have assumed the incoming beam to be linearly polarized ($\eta=0$) with azimuth $\theta=0$, whereas the initial phase φ_0 has been chosen in such a way that $\varphi(z)+\varphi_0=0$. The quantity $\varphi(z)$, which is given by equations (21), describes the change in phase of the field produced by linear interaction with the medium (refractive index of the latter). Figure 1 shows the oscillatory nature of the dependence on $\varepsilon(z)|\alpha|^2$ in both components $\langle:(\Delta Q_x(z))^2:\rangle$ and $\langle:(\Delta P_x(z))^2:\rangle$ of the field with positive as well as negative values. The negative values we are searching for are not very clear on the scale used for figure 1 (where the positive values are seen to increase enormously with increasing $\varepsilon(z)|\alpha|^2$). Figures 2 and 3 show the same dependences, albeit on a different scale, for various values of the molecular asymmetry parameter d. These curves, when taking negative values, show clear evidence of squeezing in either the $Q_x(z)$ or $P_x(z)$ component of the outgoing field for certain values of $\varepsilon(z)|\alpha|^2$. For small values of $\varepsilon(z)|\alpha|^2$ the $Q_x(z)$ component is squeezed, while $P_x(z)$ is not. As $\varepsilon(z)|\alpha|^2$ increases from zero, $\langle:(\Delta Q_x(z))^2:\rangle$ decreases to reach its first minimum, the value of which is $-0{\cdot}66$. This means that at this minimum we obtain 66 per cent of the maximum squeezing predicted by quantum mechanics, which is -1 for our definition of the operators Q and P. Note that squeezing occurs where our curves have negative abscissae, and that squeezing is greatest where the curve exhibits a minimum. The second minimum of $\langle:(\Delta Q_x(z))^2:\rangle$ is even deeper than the first, with a value of $-0{\cdot}97$, i.e. 97 per cent of the maximum squeezing predicted by theory. The first minimum of $\langle:(\Delta P_x(z))^2:\rangle$ is $-0{\cdot}92$. The values of $\varepsilon(z)|\alpha|^2$ for which these minima occur depend strongly on the molecular asymmetry parameter d, as is seen from figures 1 and 2. The larger is d, the smaller the value of $\varepsilon(z)|\alpha|^2$ at which the minimum occurs. All in all, $\varepsilon(z)|\alpha|^2$ should be of the order of unity if these minima are to be achieved, a requirement which is quite easy to meet in practice. As Ritze and Bandilla [26] have estimated, a beam intensity of $0{\cdot}4\,\mathrm{W\,cm^{-2}}$ suffices to have $|\alpha|^2=2\times10^6$ (for $\lambda=1\,\mu$m).

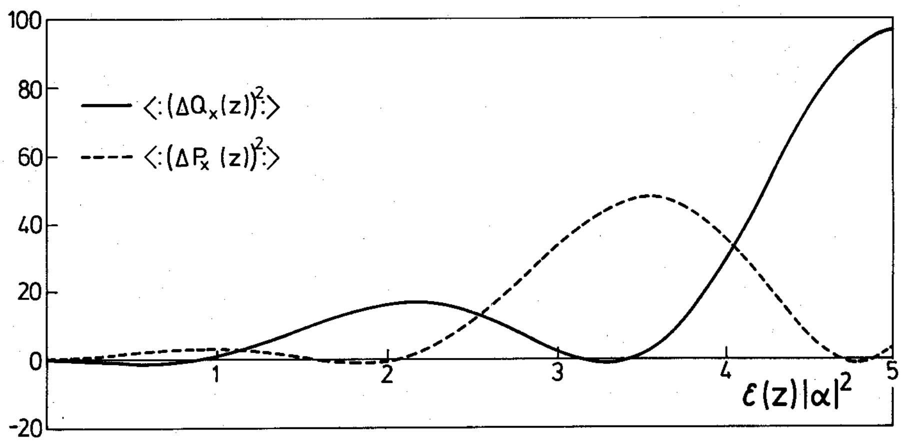

Figure 1. $\langle:(\Delta Q_x(z))^2:\rangle$ and $\langle:(\Delta P_x(z))^2:\rangle$ plotted against $\varepsilon(z)|\alpha|^2$ for $d=0{\cdot}5$, $\eta=0$, $\theta=0$ and $\varphi(z)+\varphi_0=0$.

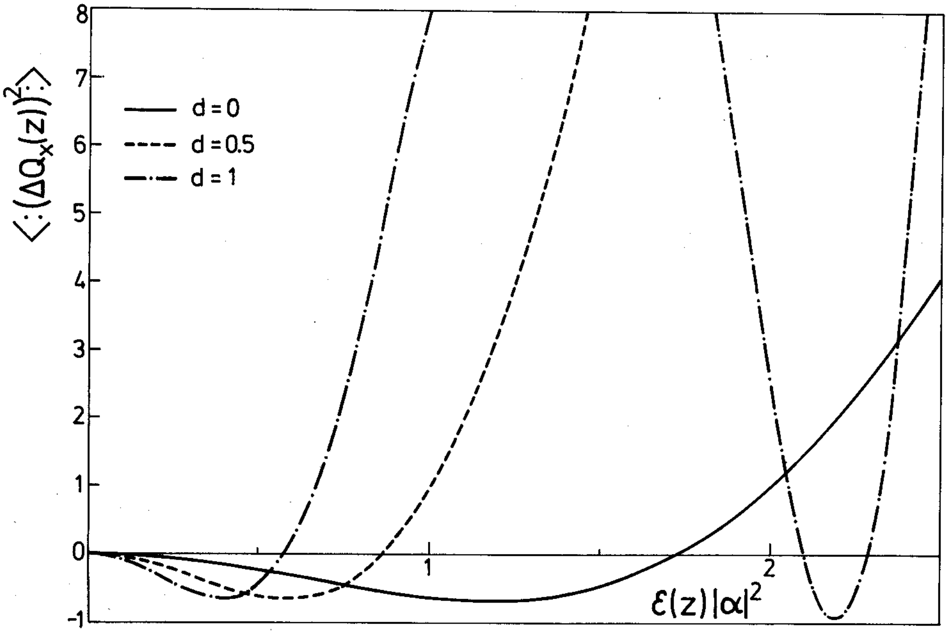

Figure 2. $\langle:(\Delta Q_x(z))^2:\rangle$ plotted against $\varepsilon(z)|\alpha|^2$ for $\eta=0$, $\theta=0$, $\varphi(z)+\varphi_0=0$ and various values of d.

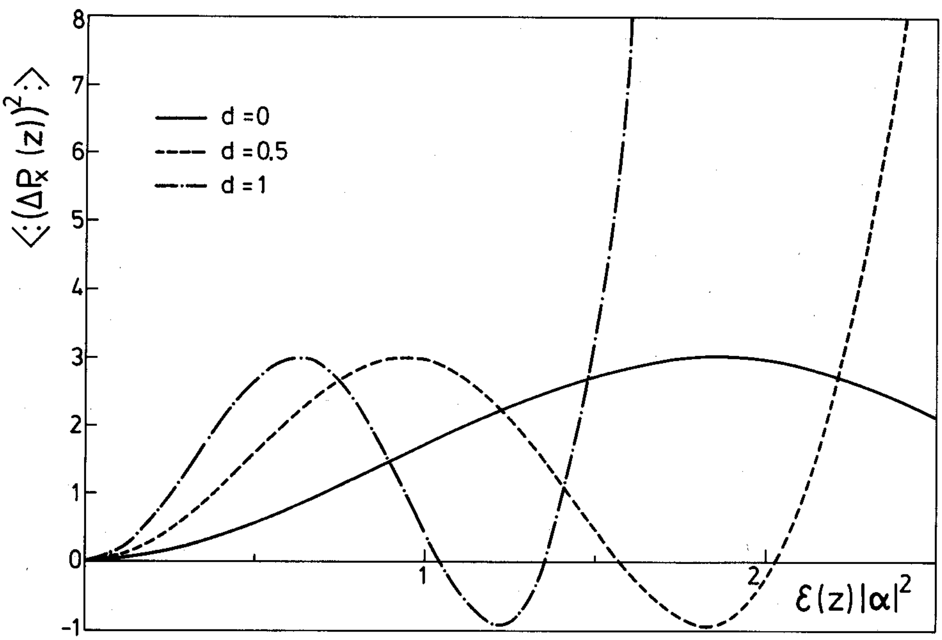

Figure 3. As figure 2, but for $\langle:(\Delta P_x(z))^2:\rangle$.

In figures 4 and 5 the curves for $\langle:(\Delta Q_x(z))^2:\rangle$ and $\langle:(\Delta P_x(z))^2:\rangle$ are plotted for different values of the ellipticity η of the incoming beam, and figures 6 and 7 show the same functions for different values of the aximuth θ. Figures 4 to 7 show that any

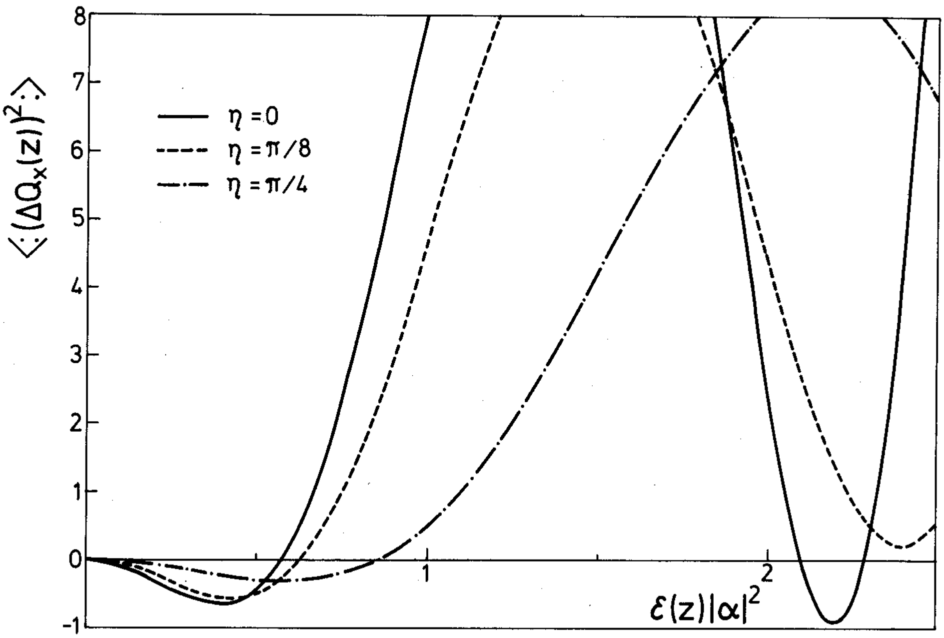

Figure 4. $\langle:(\Delta Q_x(z))^2:\rangle$ plotted againast $\varepsilon(z)|\alpha|^2$ for $d=1$, $\theta=0$, $\varphi(z)+\varphi_0=0$ and various values of η.

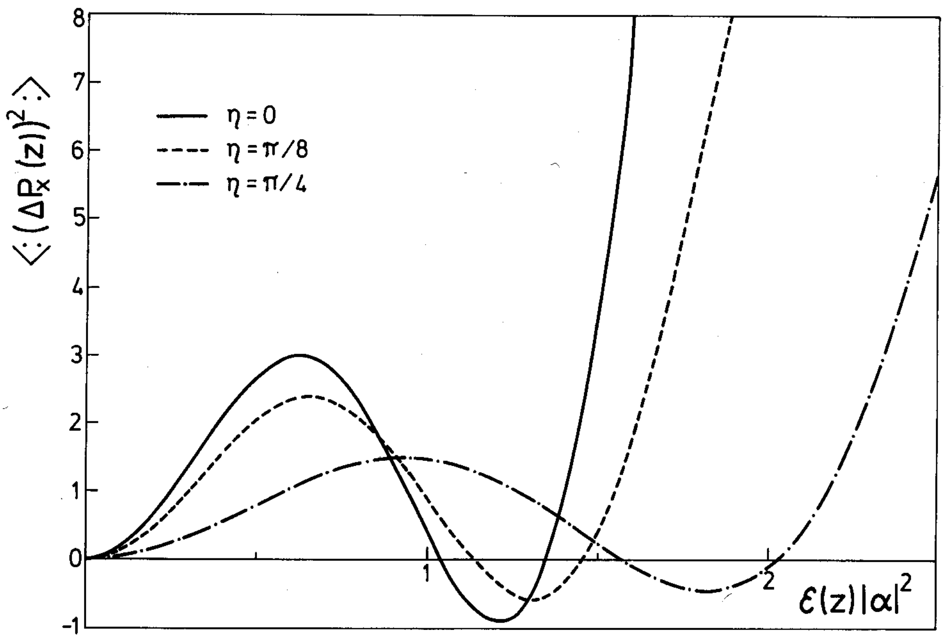

Figure 5. As figure 4, but for $\langle:(\Delta P_x(z))^2:\rangle$.

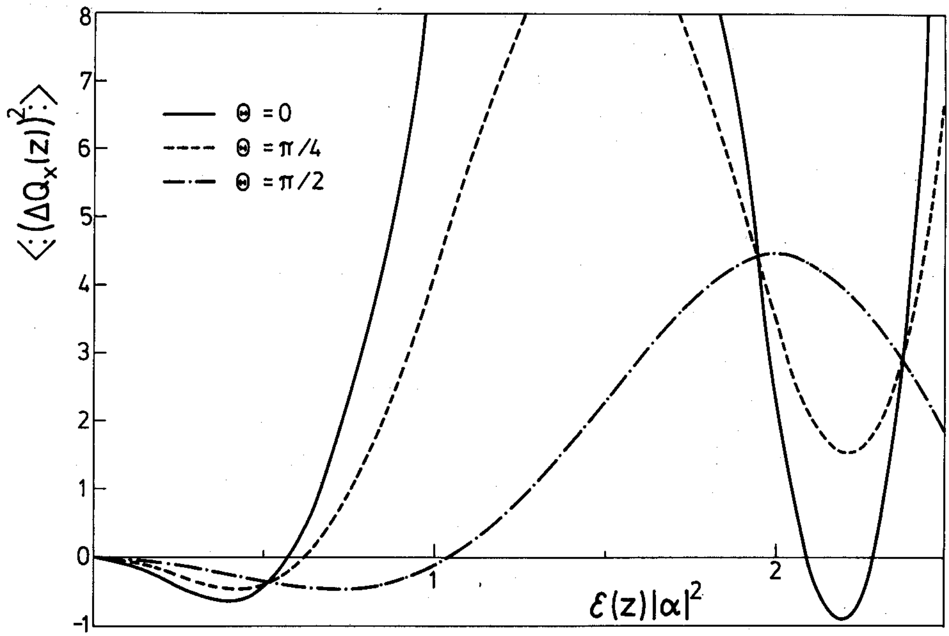

Figure 6. $\langle:(\Delta Q_x(z))^2:\rangle$ plotted against $\varepsilon(z)|\alpha|^2$ for $d=1$, $\eta=0$, $\varphi(z)+\varphi_0=0$ and various values of θ.

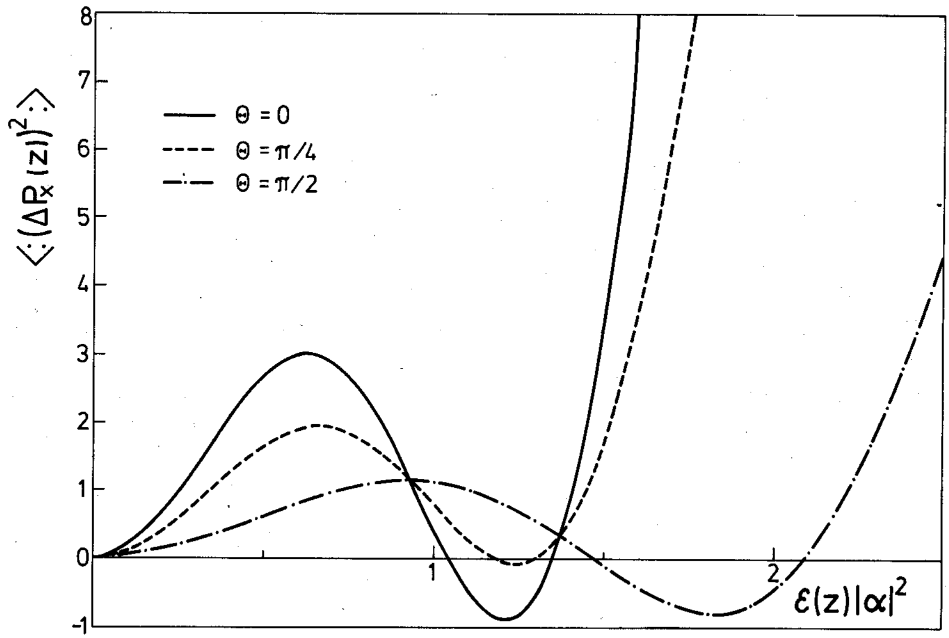

Figure 7. As figure 6, but for $\langle:(\Delta P_x(z))^2:\rangle$.

departure from linear polarization ($\eta=0$) with azimuth $\theta=0$, i.e. from polarization along the x axis, causes a decrease in the values of squeezing. The squeezing is thus the most pronounced when detecting the same polarization component of the outgoing field as that of the incoming beam. This is also true for circular polarization, according to equations (27) and (29).

Another interesting feature of the propagation of the quantized field in the nonlinear medium is to be found in figures 6 and 7. For $\theta=\pi/2$, i.e. when detecting the polarization component perpendicular to that of the incoming beam, one can still observe a non-zero field. This is a purely quantum effect [22] that cannot appear for classical fields. Moreover, this field can be squeezed.

Figure 8 shows the dependence of squeezing on the total phase $\varphi(z)+\varphi_0$ of the field. The minima are seen to be slightly shifted from $\varphi(z)+\varphi_0=0$, and actually a little deeper (squeezing is enhanced). Thus the first minimum of $\langle:(\Delta Q_x(z))^2:\rangle$ improves to $-0{\cdot}68$ for $\varphi(z)+\varphi_0\approx-0{\cdot}1$. Figure 8 also shows to what extent the squeezing is sensitive to a change in the phase of the field.

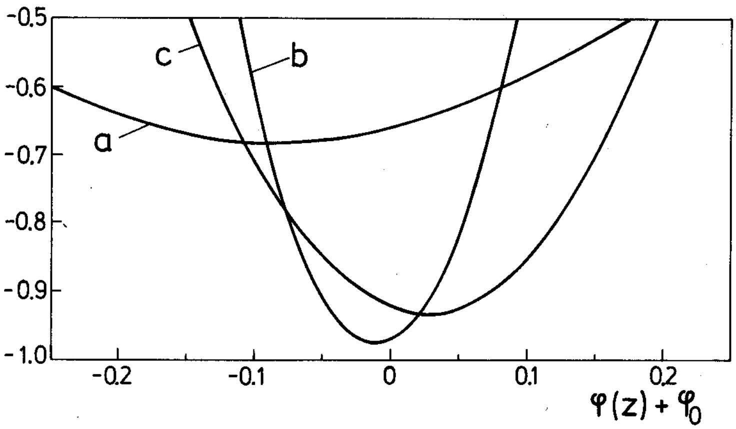

Figure 8. $\langle:(\Delta Q_x(z))^2:\rangle$ and $\langle:(\Delta P_x(z))^2:\rangle$ plotted against $\varphi(z)+\varphi_0$ around the first (curve *a*) and second (curve *b*) minima of $\langle:(\Delta Q_x(z))^2:\rangle$, and the first (curve *c*) minimum of $\langle:(\Delta P_x(z))^2:\rangle$.

4. Conclusions

We have proposed an exact analytical solution for the propagation of a quantized electromagnetic field in a nonlinear, optically isotropic medium, and have shown that the field can emerge from the medium in a squeezed quantum state which is produced by the field itself. We refer to this new way of producing squeezed states of the electromagnetic field as *self-squeezing*.

Our results are illustrated graphically for various values of the parameters on which they depend. It is shown that, for sufficiently strong fields, a considerable amount of squeezing can be achieved with the proper choice of these parameters. The first minimum in $\langle:(\Delta Q_x(z))^2:\rangle$ gives as much as 68 per cent of squeezing, and the second minimum more than 97 per cent. This second minimum is, however, rather narrow and, to reach it in practice, it will be necessary to traverse regions of relatively high positive values of the fluctuations, making it rather difficult to tune exactly to this minimum. The first minimum in $\langle:(\Delta P_x(z))^2:\rangle$, which lies at lower values of $\varepsilon(z)|\alpha|^2$, is broader than the second minimum of $\langle:(\Delta Q_x(z))^2:\rangle$ and is separated from zero by not a very high maximum, should be easier to achieve. This minimum still offers over 90 per cent of squeezing, i.e. almost complete squeezing. It seems to us that at least the first minimum in $\langle:(\Delta Q_x(z))^2:\rangle$ should be accessible, allowing squeezing to be detected in the process under consideration.

To detect the squeezed states generated in the process described in this paper, homodyne schemes are needed, especially when detecting the circularly polarized fields associated with the a_1 and a_2 Bose variables. Since the photon number in these variables is a constant of motion, the photon number distribution cannot change, i.e. it remains poissonian for a coherent initial state. This means that squeezed states with poissonian photon statistics may exist.

The results of this paper provide also an example of a nonlinear canonical transformation which does not preserve the minimum uncertainty of the states, as all linear canonical transformations do.

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On considère la propagation des champs électromagnétiques quantifiés à travers un milieu optiquement isotrope à non-linéarité cubique. Des solutions analytiques explicites sont obtenues, montrant que le champ lumineux peut émerger du milieu dans un état quantique resserré†. On donne une analyse numérique détaillée des résultats, et ceux-ci sont présentés graphiquement. D'après la théorie, le resserrement peut dépasser 90 pour cent. L'effet de resserrement dépend de l'état de polarisation du champ ainsi que des paramètres moléculaires non-linéaires.

† Nous employons 'resserrement' afin de rendre le mot 'squeezing'.

Die Ausbreitung quantisierter elektromagnetischer Felder durch isotrope Medien mit kubischen Nichtlinearitäten wird betrachtet. Analytische Lösungen werden in geschlossener Form präsentiert und zeigen, daß das Lichtfeld aus dem Medium in einem "squeezed" Quantenzustand austreten kann. Eine detaillierte numerische Analyse der Ergebnisse wird durchgeführt und graphisch präsentiert. Über 90 Prozent des "squeezing" wird theoretisch vorhergesagt. Die Abhängigkeit des "Squeezing"-Effekts von Polarisationszuständen des Felds und den nichlinearen Molekülparametern wird ebenfalls diskutiert.

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