SELF-SQUEEZING OF LIGHT PROPAGATING THROUGH NONLINEAR OPTICALLY ISOTROPIC MEDIA ☆

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In an isotropic medium in which light self-induces a cubic optical nonlinearity, the light field is shown to be in squeezed states on traversal of the medium. The dependence of the squeezing effect on the polarization state of the field and the nonlinear molecular parameters is derived explicitly. A comparison is made to the photon antibunching effect in the same phenomenon.

1. Introduction

Recently, considerable activity is directed to the generation of so-called "squeezed states" of the electromagnetic field which allow to reduce the uncertainty in the measurement of one observable at the expense of increased uncertainty in another, non commuting observable. This activity is stimulated by the potential use of squeezed states in the detection of gravitational waves [1]. Squeezing, like photon antibunching, is a purely quantum mechanical phenomenon and both are often encountered together.

A number of nonlinear optical processes have been shown theoretically to exhibit squeezing: degenerate parametric amplification [2,3], resonance fluorescence [4], degenerate four-wave mixing [5], optical bistability [6], free-electron lasers [7], Jaynes–Cummings model [8] and second-harmonic generation [9]. Mandel [9] has shown that in the case of second-harmonic generation the maximum of squeezing in the fundamental mode has the same value as the photon antibunching [10].

As we have shown previously [11], using a perturbative approach, there exists a possibility of photon antibunching in the self-induced rotation of a 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 [12], who used a more general approach and obtained the strict, nonperturbative solution of the problem.

In this paper we will show that the process of light propagation in a nonlinear, optically isotropic medium can also produce squeezed states.

2. Equations of motion

Let us consider the interaction of an intense light beam and an isotropic medium, consisting of N atoms (molecules). The interaction between the light and an individual atom will be described by the following effective hamiltonian (in the electric-dipole approximation):

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

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where $\alpha_{\sigma\tau}(\omega) = \alpha_{\sigma\tau}(-\omega, \omega)$ and $\gamma_{\sigma\tau\nu\rho}(\omega) = \gamma_{\sigma\tau\nu\rho}(-\omega, -\omega, \omega, \omega)$ are polarizability and hyperpolarizability tensors of the molecule [13], and the field operators $E_\sigma^{(+)}$ and $E_\sigma^{(-)}$ are given by

$$E_\sigma^{(+)} = [E_\sigma^{(-)}]^+ = \mathrm{i}(\hbar\omega/2\epsilon_0)^{1/2} \mathrm{e}^{\mathrm{i}kz} e_\sigma a. \tag{2}$$

We assume here that the incoming beam is a single radiation mode of frequency ω, elliptically polarized and propagating in the z-direction of the laboratory reference frame. e_σ is its polarization vector component, a the photon annihilation operator, and $k = \omega/c$. We decompose this mode into two orthogonal modes using the relation

$$e_\sigma a = e_\sigma^{(1)} a_1 + e_\sigma^{(2)} a_2, \tag{3}$$

$e_\sigma^{(1)}$ and $e_\sigma^{(2)}$ being the σ-components of the orthogonal unit polarization vectors $e^{(1)}$ and $e^{(2)}$ of the two modes a_1 and a_2. Applying the orthogonality condition for the polarization vectors we obtain the following formula

$$a = e_1^* a_1 + e_2^* a_2, \tag{4}$$

where $e_1^* = e^* \cdot e^{(1)}$, $e_2^* = e^* \cdot e^{(2)}$ are components of the original polarization vector in the new reference frame and the operators a_1, a_2 are the annihilation operators of the new modes fulfilling the boson commutation rules

$$[a_i, a_j^+] = \delta_{ij} \qquad (i, j = 1, 2). \tag{5}$$

So far the decomposition (3) (or equivalently (4)) is quite general and can be further specified either to two modes of mutually perpendicular linear polarizations or to right and left circularly polarized modes.

If we assume the new modes to be linearly polarized along the axis x or y, we get from (4):

$$a = e_x^* a_x + e_y^* a_y, \tag{6}$$

with e_x and e_y given by [14]:

$$e_x = \cos\eta \cos\theta - \mathrm{i} \sin\eta \sin\theta, \qquad e_y = \cos\eta \sin\theta + \mathrm{i} \sin\eta \cos\theta, \tag{7}$$

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

For freely orienting molecules, the molecular polarizability and hyperpolarizability tensors should be averaged over all possible orientations Ω of the molecule giving

$$\langle\alpha_{\sigma\tau}(\omega)\rangle_\Omega = \alpha(\omega)\delta_{\sigma\tau}, \qquad \langle\gamma_{\sigma\tau\nu\rho}(\omega)\rangle_\Omega = \gamma_1(\omega)\delta_{\sigma\tau}\delta_{\nu\rho} + \gamma_2(\omega)\delta_{\sigma\nu}\delta_{\tau\rho} + \gamma_3(\omega)\delta_{\sigma\rho}\delta_{\tau\nu}. \tag{8}$$

Above, $\alpha(\omega)$, $\gamma_1(\omega)$, $\gamma_2(\omega)$, $\gamma_3(\omega)$ are the appropriate rotational invariants of the molecular polarizability and hyperpolarizability tensors [13].

According to (2), (3), (6) and (8) the interaction hamiltonian (1) transforms to

$$H_\mathrm{I} = -\tilde{\alpha}(\omega)(a_x^+ a_x + a_y^+ a_y) - \tfrac{1}{2}\{\tilde{\gamma}_1(\omega)(a_x^{+2} + a_y^{+2})(a_x^2 + a_y^2) + [\tilde{\gamma}_2(\omega) + \tilde{\gamma}_3(\omega)]\,(a_x^{+2}a_x^2 + a_y^{+2}a_y^2 + 2a_x^+ a_y^+ a_y a_x)\}, \tag{9}$$

where we have introduced the abreviation $\tilde{\alpha}(\omega) = (\hbar\omega/2\epsilon_0)\,\alpha(\omega)$ and $\tilde{\gamma}_i(\omega) = (\hbar\omega/2\epsilon_0)^2\gamma_i(\omega)$.

Using the hamiltonian (9) and the commutation rules (5) we obtain the quantum equations of motion, in the Heisenberg picture, for the operators a_x and a_y. We perform the interchange $z = -ct$, because the problem under consideration is one of propagation type and not of a field in a cavity. As a result, the quantum equations of motion for the slowly varying parts (free evolution eliminated) of the field operators take the form:

$$(\mathrm{d}/\mathrm{d}z)\,a_x(z) = -(\mathrm{i}N/\hbar c)\{\tilde{\alpha}(\omega)\,a_x(z) + \tilde{\gamma}_1(\omega)\,a_x^+(z)[a_x^2(z) + a_y^2(z)]$$
$$+ [\tilde{\gamma}_2(\omega) + \tilde{\gamma}_3(\omega)]\,[a_x^+(z)\,a_x(z) + a_y^+(z)\,a_y(z)]\,a_x(z)\}. \tag{10}$$

The equation for the operator a_y can be obtained from (10) by way of the interchange $x \Leftrightarrow y$ and the corresponding equations for the creation operators a_x^+, a_y^+ by taking the hermitian conjugate of (10). We have taken into account in (10) that the number of molecules interacting with the field is N.

We have solved eqs. (10) perturbatively in our previous paper [11] showing the possibility of photon antibunching. As Ritze [12] has shown, these equations can be solved strictly provided there is no absorption in the system. On introducing the circular basis associated with the right circular polarization vector $e^{(1)} = (1/\sqrt{2}) \times (\hat{x} + i\hat{y})$ and left circular polarization vector $e^{(2)} = (1/\sqrt{2})(\hat{x} - i\hat{y})$ (with $\hat{x}$ and $\hat{y}$ being the unit vectors along the x- and y-axis, respectively), we have in agreement with (3) and (4):

$$a_1 = (1/\sqrt{2})(a_x - ia_y), \qquad a_2 = (1/\sqrt{2})(a_x + ia_y), \tag{11}$$

and eqs. (1) take the form

$$(d/dz)a_1(z) = -(iN/\hbar c)\{\widetilde{\alpha}(\omega) + [\widetilde{\gamma}_2(\omega) + \widetilde{\gamma}_3(\omega)]a_1^+(z)a_1(z) + [2\widetilde{\gamma}_1(\omega) + \widetilde{\gamma}_2(\omega) + \widetilde{\gamma}_3(\omega)]a_2^+(z)a_2(z)\}a_1(z). \tag{12}$$

For $a_2(z)$ we have to perform the interchange $1 \leftrightarrow 2$. If there is no absorption in the system $\widetilde{\alpha}(\omega)$ and $\widetilde{\gamma}_1(\omega)$, $\widetilde{\gamma}_2(\omega)$, $\widetilde{\gamma}_3(\omega)$ are real and $a_1^+(z)a_1(z)$ as well as $a_2^+(z)a_2(z)$ are constants of motion, permitting the strict solution of eqs. (12) [12], giving

$$a_1(z) = \exp\{i[\varphi(z) + \epsilon(z)(a_1^+(0)a_1(0) + 2da_2^+(0)a_2(0))]\}a_1(0),$$

$$a_2(z) = \exp\{i[\varphi(z) + \epsilon(z)(a_2^+(0)a_2(0) + 2da_1^+(0)a_1(0))]\}a_2(0), \tag{13}$$

where we have introduced the notation

$$\varphi(z) = -(Nz/\hbar c)\widetilde{\alpha}(\omega), \qquad \epsilon(z) = -(Nz/\hbar c)[\widetilde{\gamma}_2(\omega) + \widetilde{\gamma}_3(\omega)], \qquad 2d = 1 + 2\widetilde{\gamma}_1(\omega)/[\widetilde{\gamma}_2(\omega) + \widetilde{\gamma}_3(\omega)]. \tag{14}$$

With this notation clear correspondence is established between our phenomenological molecular parameters and the parameters calculated by Ritze [12] for a particular atomic level structure.

3. Squeezing

Let us introduce the canonical variables

$$Q_\sigma = a_\sigma + a_\sigma^+, \qquad P_\sigma = -i(a_\sigma - a_\sigma^+); \qquad \sigma = 1, 2 \text{ or } x, y, \tag{15}$$

which obey the commutation relation

$$[Q_\sigma, P_{\sigma'}] = 2i\delta_{\sigma\sigma'}. \tag{16}$$

A squeezed state of the electromagnetic field is defined [4] as a quantum state in which the square of the uncertainty of either Q_σ or P_σ is less than unity

$$\langle Q_\sigma^2\rangle - \langle Q_\sigma\rangle^2 < 1 \qquad \text{or } \langle P_\sigma^2\rangle - \langle P_\sigma\rangle^2 < 1. \tag{17}$$

On introducing normal ordering of the operators the definition (17) can be rewritten in the form [4]

$$\langle :Q_\sigma^2:\rangle - \langle Q_\sigma\rangle^2 < 0 \qquad \text{or } \langle :P_\sigma^2:\rangle - \langle P_\sigma\rangle^2 < 0. \tag{18}$$

Having the solutions (13) for the field operators on traversal of the path z in the nonlinear medium, we can insert them into the definition (18) and after taking the expectation value in the quantum state of the incoming beam we are able to answer the question of squeezing in a particular component of the outgoing beam.

Assuming the incoming beam as being in the coherent state $|\alpha\rangle$ defined with respect to the operator $a(0)$ given by (4) i.e.:

$$a(0)|\alpha\rangle = \alpha|\alpha\rangle, \tag{19}$$

we calculated the appropriate expectation values and obtained:

$$\langle :Q_1^2(z): \rangle - \langle Q_1(z) \rangle^2 = \langle :(a_1(z) + a_1^+(z))^2 : \rangle - \langle (a_1(z) + a_1^+(z)) \rangle^2$$

$$= 2\ \mathrm{Re}\{\alpha_1^2 \exp[2\mathrm{i}\varphi(z) + \mathrm{i}\epsilon(z) + (\mathrm{e}^{2\mathrm{i}\epsilon(z)} - 1)|\alpha_1|^2 + (\mathrm{e}^{4\mathrm{i}d\epsilon(z)} - 1)|\alpha_2|^2]$$

$$- \alpha_1^2 \exp[2\mathrm{i}\varphi(z) + 2(\mathrm{e}^{\mathrm{i}\epsilon(z)} - 1)|\alpha_1|^2 + 2(\mathrm{e}^{2\mathrm{i}d\epsilon(z)} - 1)|\alpha_2|^2]\}$$

$$+ 2|\alpha_1|^2 \{1 - \exp[2(\cos\epsilon(z) - 1)|\alpha_1|^2 + 2(\cos 2d\epsilon(z) - 1)|\alpha_2|^2\}, \tag{20}$$

where, according to (19) with (3), (7) and (11),

$$\alpha_1 = (1/\sqrt{2})(\cos\eta + \sin\eta)\,\mathrm{e}^{-\mathrm{i}\theta}\alpha, \qquad \alpha_2 = (1/\sqrt{2})(\cos\eta - \sin\eta)\,\mathrm{e}^{\mathrm{i}\theta}\alpha, \tag{21}$$

and $|\alpha|^2 = |\alpha_1|^2 + |\alpha_2|^2$. Similarly,

$$\langle :P_1^2(z): \rangle - \langle P_1(z) \rangle^2 = -2\ \mathrm{Re}\ \{...\} + 2|\alpha_1|^2 \{...\}, \tag{22}$$

where the expressions in parentheses are the same as in eq. (20).

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 (20) and (22) while for the operators $Q_x(z)$ and $P_x(z)$ we have:

$$\langle :Q_x^2(z): \rangle - \langle Q_x(z) \rangle^2 = \langle :(a_x(z) + a_x^+(z))^2 : \rangle - \langle a_x(z) + a_x^+(z) \rangle^2$$

$$= \tfrac{1}{2} [\langle :(a_1(z) + a_2(z) + a_1^+(z) + a_2^+(z))^2 : \rangle - \langle a_1(z) + a_2(z) + a_1^+(z) + a_2^+(z) \rangle^2]$$

$$= \mathrm{Re}\{\alpha_1^2 \exp[2\mathrm{i}\varphi(z) + \mathrm{i}\epsilon(z) + (\mathrm{e}^{2\mathrm{i}\epsilon(z)} - 1)|\alpha_1|^2 + (\mathrm{e}^{4\mathrm{i}d\epsilon(z)} - 1)|\alpha_2|^2]$$

$$- \alpha_1^2 \exp[2\mathrm{i}\varphi(z) + 2(\mathrm{e}^{\mathrm{i}\epsilon(z)} - 1)|\alpha_1|^2 + 2(\mathrm{e}^{2\mathrm{i}d\epsilon(z)} - 1)|\alpha_2|^2]$$

$$+ \alpha_2^2 \exp[2\mathrm{i}\varphi(z) + \mathrm{i}\epsilon(z) + (\mathrm{e}^{2\mathrm{i}\epsilon(z)} - 1)|\alpha_2|^2 + (\mathrm{e}^{4\mathrm{i}d\epsilon(z)} - 1)|\alpha_1|^2]$$

$$- \alpha_2^2 \exp[2\mathrm{i}\varphi(z) + 2(\mathrm{e}^{\mathrm{i}\epsilon(z)} - 1)|\alpha_2|^2 + 2(\mathrm{e}^{2\mathrm{i}d\epsilon(z)} - 1)|\alpha_1|^2]$$

$$+ 2\alpha_1\alpha_2 \exp[2\mathrm{i}\varphi(z) + 2\mathrm{i}d\epsilon(z) + (\mathrm{e}^{\mathrm{i}(1+2d)\epsilon(z)} - 1)|\alpha|^2]$$

$$- 2\alpha_1\alpha_2 \exp[2\mathrm{i}\varphi(z) + (\mathrm{e}^{\mathrm{i}\epsilon(z)} + \mathrm{e}^{2\mathrm{i}d\epsilon(z)} - 2)|\alpha|^2]\}$$

$$+ \{|\alpha|^2 - |\alpha_1|^2 \exp[2(\cos\epsilon(z) - 1)|\alpha_1|^2 + 2(\cos 2d\epsilon(z) - 1)|\alpha_2|^2]$$

$$- |\alpha_2|^2 \exp[2(\cos\epsilon(z) - 1)|\alpha_2|^2 + 2(\cos 2d\epsilon(z) - 1)|\alpha_1|^2]$$

$$+ 2\ \mathrm{Re}\{\alpha_1^*\alpha_2 \exp[(\mathrm{e}^{-\mathrm{i}(1-2d)\epsilon(z)} - 1)|\alpha_1|^2 + (\mathrm{e}^{\mathrm{i}(1-2d)\epsilon(z)} - 1)|\alpha_2|^2]$$

$$- \alpha_1^*\alpha_2 \exp[(\mathrm{e}^{-\mathrm{i}\epsilon(z)} + \mathrm{e}^{2\mathrm{i}d\epsilon(z)} - 2)|\alpha_1|^2 + (\mathrm{e}^{\mathrm{i}\epsilon(z)} + \mathrm{e}^{-2\mathrm{i}d\epsilon(z)} - 2)|\alpha_2|^2]\}\} \tag{23}$$

and

$$\langle :P_x^2(z): \rangle - \langle P_x(z) \rangle^2 = -\mathrm{Re}\{...\} + \{...\}, \tag{24}$$

with the same expressions in the parentheses as in eq. (23).

Although the expressions (20)–(24) are exact formulae describing fluctuations in a particular component of the field in the outgoing beam after its traversal of the path z in the medium, it is not easy to say without a detailed numerical analysis whether they are negative or not.

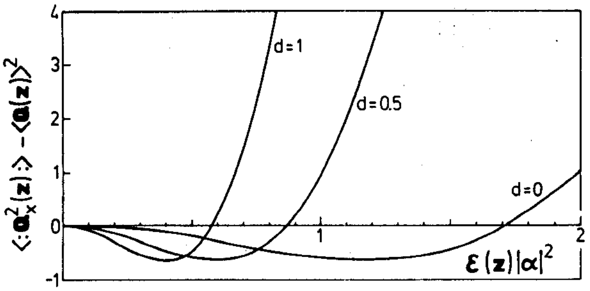

Fig. 1. $\langle :Q_x^2(z): \rangle - \langle Q_x(z) \rangle^2$ versus $\epsilon(z)|\alpha|^2$, as given by eq. (23), for $\theta = \eta = 0$ and $\varphi(z) + \varphi_0 = 0$.

In order to make the analysis a little bit simpler, let us assume the incoming beam to be polarized linearly ($\eta = 0$) with the azimuth $\theta = 0$. On this assumption, according to (21), $\alpha_1 = \alpha_2 = (1/\sqrt{2})\alpha$, and the complex number α can be written in the form

$$\alpha = |\alpha| e^{i\varphi_0}, \tag{25}$$

where φ_0 denotes the initial phase of the incoming beam. Choosing φ_0 in such a way that $\varphi(z) + \varphi_0 = 0$, we further reduce the number of parameters to be dealt with in eq. (23). Making the above assumptions, we have plotted in fig. 1 the expressions given by eq. (23) as a function of $\epsilon(z)|\alpha|^2$ for several values of the molecular parameter d. Fig. 1 shows clear evidence of squeezing in $Q_x(z)$. The maximum squeezing in this component amounting to -0.66 makes 2/3 of the value -1 allowed by quantum mechanics for thus defined operators Q and P. In order to achieve this maximum value, $\epsilon(z)|\alpha|^2$ should be of the order of unity or so, depending on the molecular parameter d. This requires a great number of photons $|\alpha|^2$ if $\epsilon(z)$ is very small, as it usually is in real physical situations. In our numerical calculations we have taken $\epsilon(z) = 10^{-4}$, but even for $\epsilon(z) \approx 10^{-6}$ or less the requirement $\epsilon(z)|\alpha|^2 \approx 1$ is met quite realistically.

If $\epsilon(z)|\alpha|^2 \ll 1$, however, we are justified in expanding our formulae (20)–(24) in power series retaining the terms linear in $\epsilon(z)|\alpha|^2$ only. This gives us approximate analytical formulae which are much simpler than the exact ones and make it possible to compare the present results with our earlier results [11] for photon antibunching. In this approximation we arrive at:

$$\langle :Q_1^2(z): \rangle - \langle Q_1(z) \rangle^2 \approx -\epsilon(z)|\alpha|^2 (1 + \sin 2\eta) \sin 2(\varphi_0 + \varphi(z) - \theta). \tag{26}$$

For the operator $P_1(z)$ we obtain the same value as in (25) albeit with the opposite sign. Similarly, from (23) we get

$$\langle :Q_x^2(z): \rangle - \langle Q_x(z) \rangle^2 \approx -\epsilon(z)|\alpha|^2 [(\cos 2\theta + 2d \cos 2\eta) \sin 2(\varphi_0 + \varphi(z))$$

$$- \sin 2\theta \sin 2\eta \cos 2(\varphi_0 + \varphi(z))], \tag{27}$$

and on changing the sign of (27) we obtain the result for the operator $P_x(z)$. So, in this approximation, either $Q(z)$ or $P(z)$ is negative, signifying squeezing in either the variable Q or P of the outgoing beam. Explicit dependence of the squeezing effect on the polarization parameters θ and η of the incoming beam and its initial phase φ_0 is also inherent in (25)–(27). $\varphi(z)$ describes the change in phase of the field due to linear interaction with the medium (refractive index of the latter). The approximation used in deriving the formulae (26) and (27) is valid for $\epsilon(z)|\alpha|^2 \ll 1$ and thus the squeezing effect is also small within the range of validity of this approximation. Unlike second harmonic generation [9], in this case the relation between photon antibunching [11,12] and squeezing is not so simple. The photon antibunching effect which is proportional to $\widetilde{\gamma}_1(\omega) \sin 4\eta$ necessarily requires the beam to be elliptically polarized and attains its maximum value for $\eta = \pi/8$ whereas the squeezing effect is not so sensitive to the polarization state of the field and can exist for any polarization. For the parameters $\theta = \pi/4$ and $\eta = \pi/8$, for which the antibunching has its maximum value, the squeezing effect in the variables Q_x or P_x depends according to (27) on the molecular parameter $\widetilde{\gamma}_1(\omega)$ alone only if $2(\varphi_0 + \varphi(z)) = \pi/4 + n\pi$ and its value is then $Nz(\hbar c)^{-1}|\alpha|^2\tilde{\gamma}_1(\omega)$. If the incoming beam is circularly polarized the amount of squeezing is $Nz(\hbar c)^{-1}|\alpha|^2[\tilde{\gamma}_2(\omega)+\tilde{\gamma}_3(\omega)]\sin 2(\varphi_0+\varphi(z)\mp\theta)$ with the upper sign for the right sense. In the case of a linearly polarized beam with $\theta = 0$ (along the x-axis) the effect is equal to $2Nz(\hbar c)^{-1}|\alpha|^2[\tilde{\gamma}_1(\omega)+\tilde{\gamma}_2(\omega)+\tilde{\gamma}_3(\omega)]\sin 2(\varphi_0+\varphi(z))$.

Particularly interesting is the case when the incident field is polarized perpendicularly to the observed field i.e. for $\theta = \pi/2$. In this case we still have a non-zero effect with the value $2Nz(\hbar c)^{-1}|\alpha|^2\tilde{\gamma}_1(\omega)\sin 2(\varphi_0+\varphi(z))$ and with the same molecular parameter $\tilde{\gamma}_1(\omega)$ which contributed to the photon antibunching effect. It should be noted at this point that the appearance of the field with polarization perpendicular to the linear polarization of the incoming field is a purely quantum effect [12]; moreover, this field exhibits squeezing but does not exhibit photon antibunching.

As we have shown in this paper, light propagating through a nonlinear optically isotropic medium can emerge from the medium in a squeezed quantum state which is produced by the light itself. Accordingly, we refer to this new effect as self-squeezing.

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