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Photon antibunching and squeezing

Two non-trivial effects of the nonlinear interaction of laser light with matter

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Abstract. The occurrence of photon antibunching and squeezing is strongly connected with nonlinear optical phenomena in general. Recent results are presented for two-atom resonance fluorescence, the anharmonic oscillator, nonlinear propagation of light and higher-harmonic generation.

1. Introduction

The 25 years since the invention of the laser have witnessed the rapid development of nonlinear and quantum optics, both experimentally and theoretically. Nowadays, the laser is in common use in industry as well as in the research laboratory. Increasingly sophisticated experiments are being performed revealing ever more subtle details of the interaction of laser light with matter. The question of the nature of light, the fundamental problem of quantum electrodynamics (QED), can now be tackled in the laboratory. In the latter half of the 1970s there emerged new arguments in favour of the quantum nature of light, and various investigations have been devoted to the search for the production of antibunched photons. Conventional light sources emit light with bunched photons (i.e. photons arriving at the photodetector in clusters), and such light is commonly called chaotic. On the other hand, lasers operating above the threshold emit coherent light consisting of photons having a random distribution of detection times. Chaotic and coherent light can be described equally well in terms of, either their photon or their classical intensity, but photon antibunching causes a dramatic breakdown of this quantum–classical equivalence; antibunching cannot be described classically. The successful experiment by Kimble *et al.* [1], in which photon antibunching was observed for light of resonance fluorescence from a single two-level atom coherently driven by a laser field, confirmed earlier theoretical predictions [2, 3] and opened the way to investigations of very fine-scale phenomena related to the nonlinear interaction of laser light with matter.

As theoretical predictions show, not only resonance fluorescence but also a wide variety of nonlinear optical phenomena can give rise to non-classical fields (for reviews see [4–10]). All classical field theories (including semiclassical and neoclassical theories) have failed to explain the negative Hanbury Brown–Twiss (HBT) effect related to the intensity correlation function. In the language of QED, the response in the HBT experiment is proportional to the deviation in photon number distribution from the Poisson statistics corresponding to the coherent field. A negative HBT effect indicates that the photon number fluctuates less than it does in a coherent beam; in other words, the variance of the photon number $\langle(\Delta n)^2\rangle$ in a single mode of radiation is less than the mean number of photons $\langle n\rangle$,

$$\langle(\Delta n)^2\rangle<\langle n\rangle, \tag{1}$$

and the photons exhibit sub-poissonian statistics. A deficit in photon count coincidences will in general decrease with the delay time between the signals reaching two photodetectors. If we consider photons as dimensionless particles, the negative HBT effect corresponds to their ordering in the sense of equalizing the distances between them. The greatest antibunching would therefore mean evenly spaced photons, $\langle(\Delta n)^2\rangle=0$, which would occur if the field were in a pure number state.

Another nonclassical effect, the so-called squeezing effect, has recently become the subject of intense study. As with antibunching, the light field in a squeezed state has no classical counterpart. Strictly speaking, squeezing is characterized by a field state in which the variance of one of two non-commuting observables (of the same physical dimension) is less than half the absolute value of their commutator. The quantized electromagnetic field can be decomposed into positive $E^{(+)}$ and negative $E^{(-)}$ frequency parts satisfying the commutation relation

$$[E^{(+)},E^{(-)}]=C, \tag{2}$$

where C is a positive c-number. Defining the in- and out-of-phase components E_1 and E_2 of the field as

$$E_1=E^{(+)}+E^{(-)},\quad E_2=-\mathrm{i}(E^{(+)}-E^{(-)}), \tag{3}$$

we have that

$$[E_1,E_2]=2\mathrm{i}C. \tag{4}$$

For a given mode of the field all irrelevant constants are usually dropped, and instead of E_1 and E_2 are introduced the 'canonical' operators:

$$Q=a+a^+,\quad P=-\mathrm{i}(a-a^+), \tag{5}$$

where a and a^+ are the annihilation and creation operators of the mode fulfilling the boson commutation rules.

The electromagnetic field is in a squeezed state if one of the variances of E_1 or E_2 is less than C [11, 12]:

$$\langle(\Delta E_1)^2\rangle<C\quad\text{or}\quad\langle(\Delta E_2)^2\rangle<C \tag{6}$$

or, equivalently, if the variance of Q or P is less than unity:

$$\langle(\Delta Q)^2\rangle<1\quad\text{or}\quad\langle(\Delta P)^2\rangle<1. \tag{7}$$

On introducing normal ordering of the field operators, the definition (1) of antibunching and the definitions (6) and (7) of squeezing can be rewritten as

$$\left.\begin{array}{l}\langle:(\Delta n)^2:\rangle<0,\\ \langle:(\Delta E_1)^2:\rangle<0\quad\text{or}\quad\langle:(\Delta E_2)^2:\rangle<0,\\ \langle:(\Delta Q)^2:\rangle<0\quad\text{or}\quad\langle:(\Delta P)^2:\rangle<0.\end{array}\right\} \tag{8}$$

From equations (8) it is apparent that fields exhibiting photon antibunching or squeezing, or both, have no classical analogue in the sense that their diagonal coherent state Glauber–Sudarshan P representation cannot be non-negative.

Fields in a squeezed state can also be produced by the nonlinear interaction of laser light with matter. The problem of squeezed states of the electromagnetic field has been reviewed [13, 14].

Both photon antibunching and squeezing are facets of the quantum nature of light, the former a particle effect and the latter a wave effect. These unique properties are related to a decrease of quantum fluctuations in photon number and phase below those of coherent light. Light with reduced quantum noise is of fundamental importance in optical communications [15].

Although photon antibunching (or, more generally, anticorrelation) and squeezing are two distinct aspects of the quantum nature of light (i.e. there is no direct connection between them), they often occur together in nonlinear phenomena. However, fields displaying squeezing but not photon antibunching, and vice versa, can exist.

Photon antibunching and squeezing, non-trivial consequences of the nonlinear interaction of laser light with the medium, are the subject of this paper, in which the authors present their latest contributions to the field. The possibility of photon antibunching and/or squeezing is discussed for two-atom resonance fluorescence, the anharmonic oscillator, nonlinear propagation, the generation of harmonics and light-scattering processes.

Special attention is drawn to self-squeezing as a novel, potent source of quantum fields: the strong beam, in traversing the medium, causes it to become optically nonlinear and thus to undergo a self-squeezing of its own field states at the output. This self-squeezing is universal, occurring to some extent in all nonlinear optical phenomena, even though no attempt has been made to detect it.

Apart from these nonlinear effects, there are many other phenomena capable of producing antibunched photons and squeezed states: multiphoton absorption [16–20], degenerate or non-degenerate parametric amplification [21–27], four-wave mixing [28, 29], the free-electron laser [30–34] or stimulated annihilation of electron–positron pairs [35, 36]. These areas, however, are outside the scope of the present paper.

2. Two-atom resonance fluorescence

Resonance fluorescence is, so far, the only phenomenon in which the non-classical effect of photon antibunching has been observed experimentally by Kimble *et al.* [1] and Leuchs *et al.* [37] (see also [38]), following earlier theoretical predictions by Carmichael and Walls [2] and Kimble and Mandel [3]. Walls and Zoller [11] have shown that squeezing as well as antibunching can occur in one-atom resonance fluorescence, though its detection may be an order of magnitude more difficult, as Mandel [12] has shown, than the already non-trivial task of detecting photon antibunching. The cooperative behaviour of a system of two atoms leads to a significantly less antibunching than occurs for one atom [39]. Two-atom resonance fluorescence has been thoroughly investigated by Ficek *et al.* [40, 41], who examined the production of both photon antibunching and squeezing. These results showed explicitly the dependence of the two effects on the interatomic interaction.

To describe a two-atom system, apart from the Rabi frequency Ω describing the interaction of an individual atom with the resonant field, one needs two additional parameters, γ_{12} and Ω_{12}, which describe collective damping and the collective shift of energy levels (the dipole–dipole interaction). This coupling between the atoms makes the problem of resonance fluorescence from such a system much more complex though not insolvable. A system of 15 equations is usually necessary to describe the time evolution of the system, but sometimes this system may be split into subsystems to make the problem more manageable. Some features of the steady-state resonance fluorescence from such a physical system can be derived analytically within the standard approximations used for the one-atom problem.

The normalized, equal-time intensity correlation function of the fluorescent light is one of the characteristics that can be calculated exactly. This function, if taking values smaller than unity, describes the photon antibunching effect. For the two-atom problem we have that (for details see [41])

$$g^{(2)}(0)=\lim_{t\to\infty}\frac{G^{(2)}(\mathbf{R}_1,t;\mathbf{R}_2,t)}{G^{(1)}(\mathbf{R}_1,t)G^{(1)}(\mathbf{R}_2,t)}$$
$$=\frac{\{32\beta^4+8(1+\Delta^2)\beta^2+\frac{1}{2}(1+\Delta^2)[(1+a)^2+(\Delta+b)^2]\}\{1+\cos[k\mathbf{r}_{12}\cdot(\hat{\mathbf{R}}_1-\hat{\mathbf{R}}_2)]\}}{\{8\beta^2+(1+\Delta^2)[1+\cos(k\mathbf{r}_{12}\cdot\hat{\mathbf{R}}_1)]\}\{8\beta^2+(1+\Delta^2)[1+\cos(k\mathbf{r}_{12}\cdot\hat{\mathbf{R}}_2)]\}},\qquad(9)$$

where

$$\beta=\Omega/4\gamma,\quad a=\gamma_{12}/\gamma,\quad b=\Omega_{12}/\gamma,\quad \Delta=(\omega_0-\omega)/\gamma,\qquad(10)$$

with 2γ denoting the Einstein A coefficient. $\hat{\mathbf{R}}_1$ and $\hat{\mathbf{R}}_2$ are the unit vectors of the two possible directions of observation of the fluorescent intensity; $\mathbf{r}_{12}=\mathbf{r}_2-\mathbf{r}_1$ is the vector connecting the centres of the two atoms.

The interatomic interaction between the atoms is described by the parameters a and b. If there is no interaction between the atoms ($r_{12}\to\infty$), then $a=b=0$. The dipole–dipole interaction b, which can be quite substantial if the atoms are close together, modifies considerably the values of $g^{(2)}(0)$. It is particularly interesting that there is a sort of interplay between the dipole–dipole interaction b and the detuning Δ of the laser frequency from the atomic transition. A considerable photon antibunching effect ($g^{(2)}(0)$ close to zero) can be obtained in such a two-atom system for certain values of Δ [40–42]. This is illustrated in figure 1, and occurs when the dipole–dipole interaction b and detuning Δ cancel ($\Delta=-b$). Here, however, one can consider the two-atom system as an individual two-level system because the laser is tuned to a particular pair of energy levels that are shifted by the dipole–dipole interaction.

For certain configurations of the two photodetectors for which

$$1+\cos[k\mathbf{r}_{12}\cdot(\hat{\mathbf{R}}_1-\hat{\mathbf{R}}_2)]=0,$$

we have that $g^{(2)}(0)=0$. Thus there is anticorrelation between the photons emitted in the directions $\hat{\mathbf{R}}_1$ and $\hat{\mathbf{R}}_2$, an effect which does not occur in one-atom fluorescence. This anticorrelation effect, being the result of interference between the fields emitted by the two atoms, is nevertheless a quantum effect and cannot be obtained for classical fields [43].

Equation (9) is valid only for $a\neq 1$; if $a=1$ the two-atom system becomes $\mathbf{S}^2$-conserving ($\mathbf{S}$ being the total spin) and the results differ from those for $a\neq 1$ because the singlet state of the system remains unpopulated [40, 44].

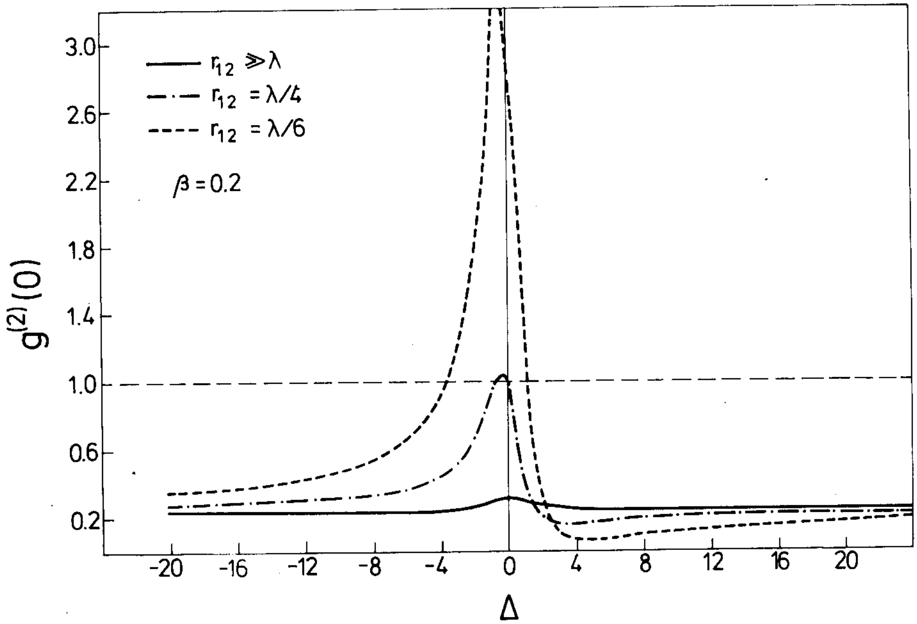

Figure 1. The intensity correlation function $g^{(2)}(0)$ versus the detuning Δ for a field strength of $\beta=0{\cdot}2$, and for various interatomic separations $r_{12}(\hat{\mathbf{R}}_1=\hat{\mathbf{R}}_2\perp\mathbf{r}_{12})$.

The normally ordered variances of the in- and out-of-phase quadrature components of the fluorescent field represent yet another characteristic of steady-state resonance fluorescence. These quantities describe the squeezing effect of the fluorescent field. It has been shown [41] that for the two-atom system these variances are proportional to the respective variances of the collective spin variables of the two-atom system:

$$\left.\begin{aligned}\langle{:}(\Delta E_1)^2{:}\rangle &\simeq \langle(\Delta R_1)^2\rangle-\tfrac{1}{2}|\langle R_3\rangle|\equiv F_1,\\ \langle{:}(\Delta E_2)^2{:}\rangle &\simeq \langle(\Delta R_2)^2\rangle-\tfrac{1}{2}|\langle R_3\rangle|\equiv F_2,\end{aligned}\right\}\qquad(11)$$

where R_1, R_2 and R_3 are Dicke's spin variables. Negative values of F_1 or F_2 give rise to squeezing in the E_1 or E_2 component, respectively, of the fluorescent field. The exact form of F_1 and F_2 for the two interacting atoms has been obtained [41, 45]. From figures 2 and 3, in which these quantities are plotted against the detuning Δ, an interplay similar to that observed in photon antibunching between the detuning Δ and the dipole–dipole interaction b is seen. A considerable amount of squeezing in the F_1 component can be obtained for $\Delta=-b$, although the minima are not so deep as for non-interacting atoms, and in this sense the interatomic interaction has a destructive effect on squeezing. The matter is discussed in more detail elsewhere [41, 45].

The proportionality expressed in equation (11), which holds in the steady-state regime, means that squeezing in the atomic variables implies that the normally ordered variance of the corresponding component of the fluorescent field is negative. This may be untrue in the transient regime of resonance fluorescence [46], in which there are intervals of time during which the atomic squeezing does not proceed in step with the field squeezing.

Loudon [47] has recently given an expression for the two-time photon number correlation function for fluorescent light homodyned with coherent light.

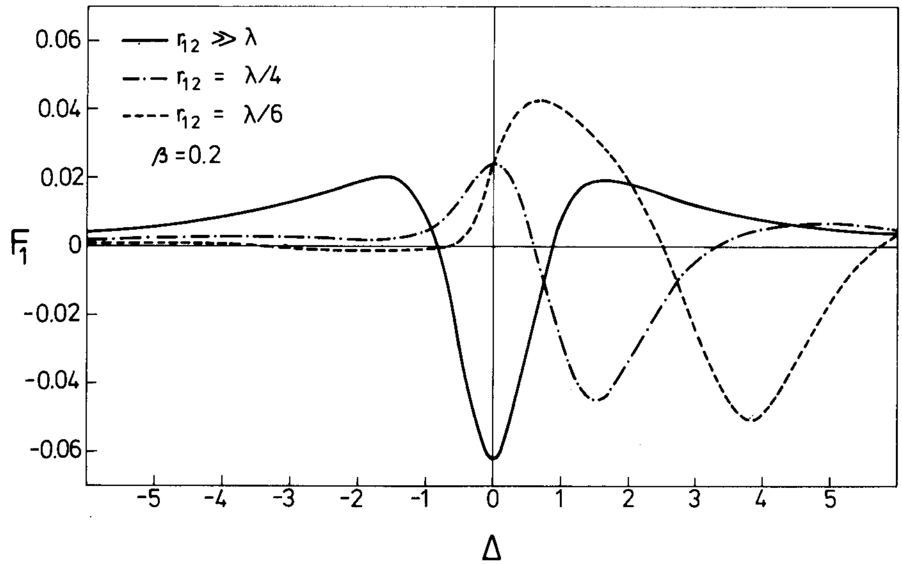

Figure 2. F_1 versus the detuning Δ for a field strength of $\beta = 0{\cdot}2$ and various interatomic separations r_{12}. The transition dipole moment $\boldsymbol{\mu}$ is parallel to $\mathbf{r}_{12}$, and $\hat{\mathbf{R}}$ is perpendicular to $\mathbf{r}_{12}$.

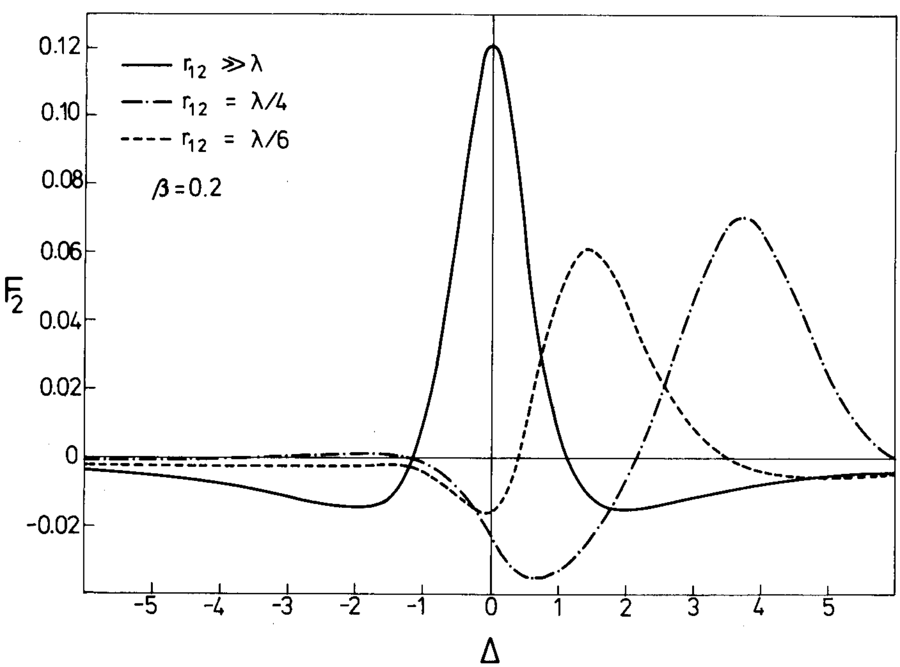

Figure 3. As figure 2, but for F_2.

3. Anharmonic oscillator

The anharmonic oscillator model is probably one of the simplest models used to describe certain features of the nonlinear interaction of laser light with matter. Despite its simplicity, this model is very instructive and provides results which are easy to interpret. In this model, the hamiltonian of the system is taken (discarding the non-energy-conserving terms) to be

$$H=\hbar\omega a^{+}a+\tfrac{1}{2}\hbar\kappa a^{+2}a^{2}, \tag{12}$$

where κ is the anharmonicity parameter, assumed to be real, and the creation and annihilation operators are taken in normal order.

The Heisenberg equation of motion for the annihilation operator a, according to equation (12), has the form [48]

$$\dot{a}=-\mathrm{i}\hbar^{-1}[a,H]=-\mathrm{i}(\omega+\kappa a^{+}a)a. \tag{13}$$

Since $a^{+}a$ is a constant of motion, equation (13) has the simple exponential solution

$$a(t)=\exp\{-\mathrm{i}t[\omega+\kappa a^{+}(0)a(0)]\}a(0). \tag{14}$$

The $\exp(-\mathrm{i}\omega t)$ part of the time evolution describes the free evolution of the system, whereas the remaining part of the exponential arises from the nonlinear term in the hamiltonian.

The solution (14) is the exact operator solution which allows us to calculate all the characteristics of the field at the time t, provided the state of the field at the time $t=0$ is known.

We see immediately that

$$\langle a^{+2}(t)a^{2}(t)\rangle=\langle a^{+2}(0)a^{2}(0)\rangle, \tag{15}$$

and that there is no photon antibunching at the time t if none existed at the time $t=0$. Moreover, any function of the photon number operator $n=a^{+}a$ is also a constant of motion, so that the photon number distribution $p(n)$ in the mode does not change in the course of evolution. This means that the photon number distribution remains poissonian if the field was in a coherent state $|\alpha\rangle$ at the time $t=0$, but it does not mean that the field is always in a coherent state. Using the solution (14) and the definition (7), one obtains the following result, due to Tanaś [48]:

$$\begin{aligned}\langle:[\Delta Q(\tau)]^{2}:\rangle=2\,\mathrm{Re}\{&\alpha^{2}\exp[-\mathrm{i}\tau+\langle n\rangle(\exp(-2\mathrm{i}\tau)-1)]\\&-\alpha^{2}\exp[2\langle n\rangle(\exp(-\mathrm{i}\tau)-1)]\}+2\langle n\rangle\{1-\exp[2\langle n\rangle(\cos\tau-1)]\},\end{aligned} \tag{16}$$

$$\langle:[\Delta P(\tau)]^{2}:\rangle=-2\,\mathrm{Re}\{\dots\}+2\langle n\rangle\{\dots\}, \tag{17}$$

where $\tau=\kappa t$. The obvious $\exp(-\mathrm{i}\omega t)$ dependence arising from free evolution has been dropped, and the two pairs of braces in equation (17) contain the same expressions as in equation (16).

Equations (16) and (17) are both strict solutions. They are illustrated graphically in figure 4 as functions of the product $\langle n\rangle\tau(\langle n\rangle=|\alpha|^{2})$ for $\tau=10^{-6}$. The initial phase

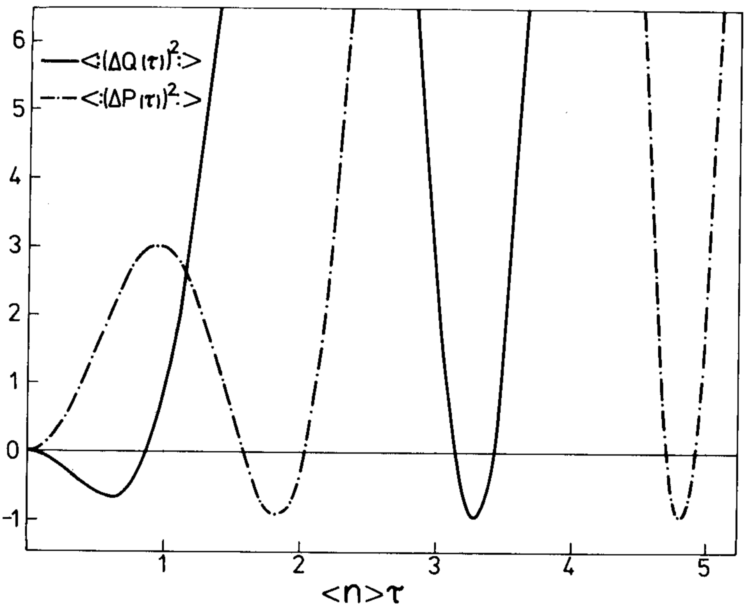

Figure 4. Fluctuations of the in- and out-of-phase components of the field versus $\langle n\rangle\tau$.

is chosen so as to have α real. Both curves show an oscillatory behaviour, with positive as well as negative values. For small $\langle n\rangle\tau$ the in-phase component Q is squeezed, while the out-of-phase component P is not. The first minimum of $\langle:[\Delta Q(\tau)]^2:\rangle$ occurring for $\langle n\rangle\tau \simeq 0{\cdot}6$ has the value $-0{\cdot}66$, two-thirds of the limiting value of -1 allowed by quantum mechanics. The successive minima are even deeper, amounting to as much as $-0{\cdot}92$ and $-0{\cdot}97$, which corresponds to a considerable amount of squeezing.

This simple example shows that squeezed states exist for which the photon distribution remains poissonian. Such states cannot, of course, be detected by using a direct photon counting technique. The exponential form of the solution (14) contributes to squeezing but not to photon antibunching, which is phase-insensitive. In contrast, Quattropani *et al.* [49] have shown that the same anharmonic oscillator, when interacting with a thermal reservoir at thermal equilibrium, leads to sub-poissonian photon statistics. In this case, however, the field is described by the density operator

$$\rho = N\exp\left[-(kT)^{-1}(\hbar\omega a^+ a + \tfrac{1}{2}\hbar\kappa a^{+2}a^2)\right],$$

which is diagonal in the number state representation, and there is no squeezing in such a field.

The solution (14) provides an example of the nonlinear canonical transformation. This particular example shows that nonlinear canonical transformations do not conserve minimum uncertainty. Applied to a coherent state (which is a minimum uncertainty state), this transformation gives a state which is no longer a minimum uncertainty state.

4. Self-squeezing of light propagating in nonlinear media

The anharmonic oscillator model described above can be realized in practice when a strong electromagnetic field propagates through a nonlinear medium without absorption [50].

If the medium is macroscopically isotropic and consists of N molecules which are non-interacting molecules but can be optically active, the interaction between the light mode of frequency ω and an individual molecule can be described by the effective hamiltonian [50, 51]

$$
\begin{aligned}
H_{\rm I} = & -\tilde{\alpha}(\omega)(a_+^{+}a_+ + a_-^{+}a_-) - \tilde{\rho}(\omega)(a_+^{+}a_+ - a_-^{+}a_-) \\
& - [2\tilde{\gamma}_1(\omega) + \tilde{\gamma}_2(\omega) + \tilde{\gamma}_3(\omega)]a_+^{+}a_-^{+}a_-a_+ \\
& - \tfrac{1}{2}[\tilde{\gamma}_2(\omega) + \tilde{\gamma}_3(\omega)](a_+^{+2}a_+^2 + a_-^{+2}a_-^2) \\
& - \tfrac{1}{2}[\tilde{\sigma}_2(\omega) + \tilde{\sigma}_3(\omega)](a_+^{+2}a_+^2 - a_-^{+2}a_-^2),
\end{aligned} \tag{18}
$$

where a_+ and a_- are the annihilation operators for the modes associated with right- and left-circular polarization of the light beam, and

$$
\left.
\begin{aligned}
\tilde{\alpha}(\omega) &= \frac{2\pi\hbar\omega}{V}\alpha(\omega), \quad \tilde{\gamma}_i(\omega) = \left(\frac{2\pi\hbar\omega}{V}\right)^2 \gamma_i(\omega), \\
\tilde{\rho}(\omega) &= \frac{2\pi\hbar\omega}{V}\rho(\omega), \quad \sigma_i(\omega) = \frac{1}{c}\left(\frac{2\pi\hbar\omega}{V}\right)^2 \sigma_i(\omega),
\end{aligned}
\right\} \tag{19}
$$

$\alpha(\omega)$ and $\gamma_i(\omega)$ $(i=1,2,3)$ denoting the rotational invariants of the molecular polarizability and hyperpolarizability tensors, and $\rho(\omega)$ and $\sigma_i(\omega)$ $(i=1,2,3)$ denoting the rotational invariants of the tensors describing the linear and nonlinear optical activity of the molecules [8].

From the circular representation of the interaction hamiltonian (18), it is evident that $a_+^{+}a_+$ and $a_-^{+}a_-$ commute with this hamiltonian. This means that the numbers of photons in both circular components are constants of motion (in the absence of absorption). The same, however, is not true in the cartesian representation for the operators $a_x^{+}a_x$ and $a_y^{+}a_y$, showing that in a nonlinear medium (even if it is isotropic) the linear polarization of the field is not preserved.

Using the interaction hamiltonian (18), we can easily write the Heisenberg equations of motion for the operators a_+ and a_- describing the time evolution of these operators. Since we are considering propagation rather than a field in a cavity, we introduce the parameter $z=ct$ and obtain the equations (free evolution having been eliminated)

$$
\frac{\mathrm{d}}{\mathrm{d}z}a_\pm(z) = \mathrm{i}\{\phi_\pm + \varepsilon_\pm a_\pm^{+}(z)a_\pm(z) + \delta a_\mp^{+}(z)a_\mp(z)\}a_\pm(z). \tag{20}
$$

Since $a_+^{+}a_+$ and $a_-^{+}a_-$ are constants of motion, equations (20) have the simple exponential solutions

$$
a_\pm(z) = \exp\{\mathrm{i}z[\phi_\pm + \varepsilon_\pm a_\pm^{+}(0)a_\pm(0) + \delta a_\mp^{+}(0)a_\mp(0)]\}a_\pm(0), \tag{21}
$$

where we have introduced the notation

$$
\left.
\begin{aligned}
\phi_\pm &= (N/\hbar c)[\tilde{\alpha}(\omega) \pm \tilde{\rho}(\omega)], \\
\varepsilon_\pm &= (N/\hbar c)[\tilde{\gamma}_2(\omega) + \tilde{\gamma}_3(\omega) \pm (\tilde{\sigma}_2(\omega) + \tilde{\sigma}_3(\omega))], \\
\delta &= (N/\hbar c)[2\tilde{\gamma}_1(\omega) + \tilde{\gamma}_2(\omega) + \tilde{\gamma}_3(\omega)].
\end{aligned}
\right\} \tag{22}
$$

The formal solutions (21), which are strict operator solutions, allow one to calculate any characteristic of the field on traversal of the path z in the nonlinear medium. These solutions are in fact two-mode generalizations of the simple anharmonic solution (14). If there is only one mode of radiation propagating in the medium (either a_+ or a_-) we obtain the same equation as that of the anharmonic oscillator. Again, since $a_+^+ a_+$ and $a_-^+ a_-$ are constants of motion, there is no change in either the intensity or the photon statistics of these components. However, if a polarizer is used to select a particular cartesian component of the outgoing field, say the x component, the number of photons $a_x^+ a_x$ as well as the photon statistics of this component change as the field traverses the medium. As Tanaś and Kielich have shown [52] perturbatively, and as Ritze [53] has confirmed, giving exact solutions, photon antibunching can then occur. Tanaś and Kielich [50] have also predicted squeezed states of the electromagnetic field in such a system. Although there is no change in the photon statistics in the two circular components of the field, these components can be squeezed as a result of nonlinear interaction with the medium. For this squeezing effect, which is caused by the field itself, the present authors have introduced the term 'self-squeezing' [50].

If the incoming beam is in a coherent state, we obtain from the solutions (21) the equation

$$\begin{aligned}\langle:[\Delta Q_\pm(z)]^2:\rangle = 2\,\mathrm{Re}\,\{&\alpha_\pm^2 \exp[2iz\phi_\pm + iz\varepsilon_\pm + (\exp(2iz\varepsilon_\pm)-1)|\alpha_\pm|^2\\ &+(\exp(2iz\delta)-1)|\alpha_\mp|^2] - \alpha_\pm^2 \exp[2iz\phi_\pm + 2(\exp(iz\varepsilon_\pm)-1)|\alpha_\pm|^2\\ &+2(\exp(iz\delta)-1)|\alpha_\mp|^2]\} + 2|\alpha_\pm|^2\{1-\exp[2(\cos(\varepsilon_\pm z)-1)|\alpha_\pm|^2\\ &+2(\cos(\delta z)-1)|\alpha_\mp|^2]\}. \end{aligned} \tag{23}$$

A numerical analysis of this expression shows that it oscillates between positive and negative values. The negative values signify squeezing in the respective component of the field. (Corresponding curves for non-optically-active media are given elsewhere [50].) This self-squeezing effect is universal in the sense that it exists even for spherically symmetric molecules (atoms). The numerical values of this squeezing can be as high as 97 per cent of the maximum allowed by quantum mechanics. This means a reduction of quantum fluctuations by two orders of magnitude, which seems promising for the experimenter.

Self-generation of quantum fields can occur in arbitrary isotropic media as well as in crystals (both optically active and inactive) belonging to certain symmetry classes [54].

5. Harmonic generation

Harmonic generation processes also produce light which exhibits photon antibunching and squeezing together. Second-harmonic generation has been discussed by Kozierowski and Tanaś [55] in the 'short optical paths' approximation from the viewpoint of photon antibunching, and by Mandel [56] from the viewpoint of squeezing. The present authors have generalized these results to higher-harmonic generation for photon antibunching [57] and squeezing [58], and extended this perturbative approach to second-harmonic generation with a photon-number-dependent coupling constant [59].

In the electric dipole approximation and for perfect phase matching, the generation of the kth harmonic is described by the effective interaction hamiltonian

$$H_I = \hbar c L_{k\omega} a_{k\omega}^+ a_f^k + \text{h.c.}, \tag{24}$$

where $L_{k\omega}$ is the coupling constant, the subscripts $k\omega$ and f denote the kth harmonic and fundamental mode, respectively and h.c. represents the hermitian conjugate.

If the incident light is coherent and both the fundamental and harmonic beams traverse the 'short' path z in the medium, their photons are antibunched and the scaled HBT parameter $\langle:[\Delta n(z)]^2:\rangle/\langle n(z)\rangle$ takes the forms:

$$\mathrm{HBT}_{\mathrm{f}} \simeq -k(k-1)C(z), \tag{25}$$

$$\mathrm{HBT}_{k\omega} \simeq -\tfrac{1}{3}kC^2(z) \sum_{m=0}^{k-1} \sum_{s=0}^{m} \sum_{t=1}^{k-1-s} s!t! \binom{m}{s}\binom{k-1}{s}\binom{k}{t}\binom{k-1-s}{t} \langle n_{\mathrm{f}0}\rangle^{1-s-t}, \tag{26}$$

where

$$C(z) = \langle n_{k\omega}(z)\rangle / \langle n_{\mathrm{f}0}\rangle \simeq L_{k\omega}^2 \langle n_{\mathrm{f}0}\rangle^{k-1} z^2$$

is the conversion ratio. The parameter (26) though expressed here in shortened form, is strictly equivalent to the form given previously [59].

In the classical approach a wave of constant amplitude generates a harmonic with non-fluctuating intensity, thus itself remaining unfluctuating, albeit with diminished intensity. This picture changes drastically when quantum effects are considered, even under the naive assumption that photons are dimensionless points [7, 59]; generation proceeds with the simultaneous 'extraction' of k photons from the fundamental beam in a single event. Hence, the phenomenon occurs more strongly in intervals of time during which the number of incident photons is greater. In consequence, as well as a decrease in the mean number of fundamental photons, we have to consider their 'rarefaction', meaning that the distances between successive photons tend to become equal. This is precisely the essence of photon antibunching. As for the generated harmonic, of the k fundamental photons only one photon of k-fold frequency is generated in each elementary event, so the disorder in the harmonic will be less than in the incident coherent beam at the input to the medium. This is of course characteristic of photon antibunching.

The same rough explanation of the origin of photon antibunching in a beam of fundamental frequency ω can easily be applied to other nonlinear phenomena. For one-mode nonlinear processes, for instance in the anharmonic oscillator or in the nonlinear propagation of circularly polarized light, the number of photons—and hence their disorder—is not affected. The annihilated photons are re-emitted into the same mode, and so photon antibunching does not occur.

According to the definitions (8), the results for the variances of the canonical variables show squeezing. For the fundamental beam we have that [58]

$$\left.\begin{matrix}\langle:[\Delta Q_{\mathrm{f}}(z)]^2:\rangle \\ \langle:[\Delta P_{\mathrm{f}}(z)]^2:\rangle\end{matrix}\right\} = \mp k(k-1)C(z)\cos(2\phi_0), \tag{27}$$

where ϕ_0 is the initial phase of the complex amplitude of the incident light; it is clear that $\langle n_{\mathrm{f}0}\rangle = |\alpha_{\mathrm{f}}|^2$. Depending on the phase ϕ_0, either Q_{f} or P_{f} is squeezed. In particular, by setting $k=2$ in equation (27) Mandel's result [56] is recovered. The amount of maximal squeezing in the fundamental beam is equal to the degree of photon antibunching, as given by equation (25).

For the generated harmonic we obtain

$$\left.\begin{matrix}\langle:[\Delta Q_{k\omega}(z)]^2:\rangle\\ \langle:[\Delta P_{k\omega}(z)]^2:\rangle\end{matrix}\right\} = \pm\tfrac{1}{3}kC^2(z)\sum_{m=0}^{k-1}\sum_{s=0}^{m}\sum_{t=1}^{k-1-s} s!t!$$

$$\times\binom{m}{s}\binom{k-1}{s}\binom{k}{t}\binom{k-1-s}{t}\langle n_{f0}\rangle^{1-s-t}\cos(2k\phi_0)$$

$$\mp\tfrac{1}{6}k^2C^2(z)\sum_{m=0}^{k-1}\sum_{s=0}^{k-2}\sum_{t=0}^{s}\sum_{r=0}^{\min(m,k-2-t)} r!t!$$

$$\times\binom{s}{t}\binom{k-1}{t}\binom{m}{r}\binom{k-2-t}{r}\langle n_{f0}\rangle^{-r-t}\cos(2k\phi_0). \quad (28)$$

In a previous analysis [58] the second term in equation (28) was omitted.

In the generated k-harmonic beam, the maximal squeezing is no longer equal to the degree of photon antibunching. In particular, for the second harmonic the maximal squeezing amounts to $4C^2(z)/3$, in agreement with the result obtained by Peřina *et al.* [60], which is half the photon antibunching.

Chmela [61] has shown that, by generating second harmonics in cascade in thin plates, filtering out the generated harmonic behind each plate, photon antibunching in the fundamental beam increases considerably. The same is expected for squeezing.

As well as generating harmonics, the processes of spontaneous Rayleigh and Raman scattering of harmonics [62] provide information on the statistics of light. Simaan [63] and, independently, Szlachetka and Kielich [64], have drawn attention to the possibility of photon antibunching occurring in hyper-Raman scattering. Effects of correlation and anticorrelation of incident and scattered photons in the presence of phonon fluctuations have been analysed closely by Peřinova *et al.* [65] and Szlachetka *et al.* [66] for various initial statistical properties of laser and Stokes or anti-Stokes modes—whether coherent, chaotic or in the vacuum state (see also [67–69]). A similar analysis has also been performed for Brillouin scattering [70, 71]. Sub-poissonian statistics and squeezing phenomena in hyper-Raman scattering have been discussed recently by Peřina *et al.* [60], and by Peřinova and Tiebel [72].

6. Conclusions and outlook

Some phenomena of nonlinear optics which can give rise to fields displaying such nonclassical features as photon antibunching and squeezing have been described. Although such fields are important in their own right, research in this area is also motivated by their possible applications in optical communications [15, 73, 74] and in the detection of gravity waves [75]. Usually both the photon number and the phase of the field are affected by nonlinear interaction, and the two non-classical effects can occur simultaneously. However, fields exhibiting photon antibunching but not squeezing (and vice versa) can be produced as well. Squeezing has yet to be observed experimentally. Mandel [12] and Shapiro *et al.* [76] have proposed some schemes for its observation (see also [77, 78]). Given current rates of progress, it seems safe to predict that squeezing will soon be verified experimentally, and that photon antibunching will be found to be associated with phenomena other than resonance fluorescence.

There have been numerous papers [79–101] analysing various aspects of quantum field generation, but lack of space prevents them from being discussed here.

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