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QUANTUM FLUCTUATIONS IN SECOND-HARMONIC GENERATION WITH PHOTON NUMBER-DEPENDENT COUPLING CONSTANT

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INTRODUCTION

The latter 1970-ies witnessed a revival of interest in the nature of light, and new arguments in favour of its quantum structure were put forward. A considerable amount of theoretical work was aimed at the production of so-called photon antibunching, first achieved in experiment by Kimble et al. (1977) who thus succeeded in proving the existence of this unique quantum effect for light of resonance fluorescence from single atoms (Walls, 1979).

Photon antibunching is related to a reduction in photon number fluctuations below their mean value. It entails a negative Hanbury Brown and Twiss (HBT) effect i.e. a deficit in photon count coincidences with respect to the random coincidences for coherent light; the deficit decreases with delay time. Roughly conceiving of the photons as point-shaped corpuscules, the above would correspond to their ordering, meaning an equalisation of the distances between the photons. Maximal antibunching would thus mean evenly spaced photons.

Light displaying photon antibunching has no classical counterpart. Classically speaking, the HBT effect can at the most be zero, thus corresponding to coherent light of constant intensity. The light sources generally used (including lasers) emit light that has classical analogs. The classical fluctuations of the number of emitters mask the quantum properties of the light emitted. From the theoretical viewpoint, the most direct proof of the correctness of quantum electrodynamics would consist in the construction of a unit source, e.g. a collisionally excited ion in a trap, and in performing a HBT experiment for the photons emitted in single, controlled events. The very closest to that is resonance fluorescence, already mentioned, from a two-level atom driven by a coherent field. The possibility of obtaining such non-classical fields is moreover offered by the now available wide variety of nonlinear processes of interaction between essentially coherent light and matter. In particular, we have in mind the processes of second-harmonic light generation, in which the role of the medium reduces to that of coupling between the fundamental mode (f) of frequency ω and the generated second harmonic (h) of frequency 2ω.

It should be stated that photon antibunching has already been the subject of a number of review articles (Walls, 1979; Peřina, 1980; Kozierowski, 1981; Kielich, 1981; Paul, 1982).

However, light with antibunched photons is by no means the only example of light with non-classical features. Recently, considerable interest is directed to electromagnetic fields in so-called squeezed states. In general, squeezing is meant to denote a reduction of the uncertainty in one of the non-commuting quantities below the minimal value of the root of the uncertainty product i.e. the value corresponding to the sign of equality in the uncertainty principle (obviously, both quantities are of the same dimension) . The fluctuations of the other non-commuting observable then increase, so as "not to violate the uncertainty principle". In concrete cases one takes as the observables in question the canonical variables P and Q. The minimum of the product of their uncertainties corresponds to coherent light as that kind of light the quantum description of which is the closest to classical. One is led to the conclusion that opportunities for the production of such fields (similarly to antibunching) should be sought in appropriate interaction processes involving laser light and matter. Although the two properties: squeezing and antibunching, are similar as to their quantum mechanical origin and become apparent under by nigh the same conditions, no general connection between them exists. For example, whereas maximal antibunching is expected to occur for the field in a pure number state, this kind of field exhibits no squeezing of any canonical variable (Walls and Zoller, 1981). Thus, in the case of nonlinear changes in light refractive index (anharmonic oscillator) the field emerges in a squeezed state but exhibiting no photon antibunching altogether (Tanaś, 1983). Nonetheless the two above properties of light are often considered jointly, because of their common origin in quantum fluctuations.

PHOTON ANTIBUNCHING

Second-harmonic generation has been discussed in the "short optical paths" approximation by Kozierowski and Tanaś (1977) from the viewpoint of photon antibunching and, quite recently, by Mandel (1982) from that of squeezing.

Generally, in the electric dipole approximation, at perfect phase matching, the process is described by the following effective interaction Hamiltonian:

$$H = \hbar c L_{2\omega} a_h^{+} a_f^{2} + \text{h.c.} \quad , \tag{1}$$

where $L_{2\omega}$ is the constant of coupling of the fundamental and harmonic beams, c is the light velocity in the medium, whereas a^{+} and a are photon creation and annihilation operators, respectively. Commonly, $L_{2\omega}$ is taken to be proportional to the susceptibility tensor of rank 3 and not dependent on the number of incident photons. In fact, the process is described as consisting in the vanishing of two photons ω and emission of one harmonic photon 2ω. In general, however, $L_{2\omega}$ can be dealt with as dependent on the number of photons incident on the medium; in a first approach, this dependence can be assumed to be linear (Kasprowicz et al., 1976):

$$L_{2\omega} = L_3 + L_5 \hat{n}_f \quad . \tag{2}$$

Above, $\hat{n}$ is the photon number operator. The coupling constant L_3 is proportional to the third-rank susceptibility tensor, whereas L_5 —— to the fifth rank tensor. The process governed by L_5 takes place with the vanishing of three photons ω and the emission of a photon 2ω and a new photon ω. The two contributions L_3 and L_5 are practically unconnected. However, an event can occur in which e.g. four photons interact with the medium; then if, say, L_3 occurs first, L_5 is ruled out for the lack of one photon. But both processes can appear in the reverse order.

We now make use of the perturbative solution. With the Hamiltonian

$$H = \hbar c L_3 a_h^+ a_f^2 + \hbar c L_5 a_h^+ \hat{n}_f a_f^2 + h.c. \quad , \tag{3}$$

we calculate the quantities of interest to us (firstly, the slowly variable parts of the creation and annihilation operators of photons of the two beams in the rotating wave approximation) in the form of a Taylor series in the path z traversed by the beams in the medium. Whatever the z-approximation applied, we retain only the lowest, linear power of L_5.

In this way, for the fundamental beam, we obtain the normally ordered correlation functions of the first and second orders in the form

$$\begin{aligned} g_f^{(1)}(z) &= g_f^{(1)}(0) - 2L_3^2 z^2\left\{ g_f^{(2)}(0) + 2\frac{L_5}{L_3} g_f^{(3)}(0) + \cdots\right\} + \cdots \; , \\ g_f^{(2)}(z) &= g_f^{(2)}(0) - 2L_3^2 z^2\Big\{ 2g_f^{(3)}(0) + g_f^{(2)}(0) + \\ &\quad + 2\frac{L_5}{L_3}\left[2g_f^{(4)}(0) + 3g_f^{(3)}(0)\right] + \cdots \Big\} + \cdots \; , \end{aligned} \tag{4}$$

where $g_f^{(k)}(0)$ are the field correlation functions at the input to the medium i.e. in the plane z=0.

The response in the HBT experiment is proportional to the difference $g^{(2)}(z) - [g^{(1)}(z)]^2$ i.e. to the departure from Poisson statistics. In particular, calculating the scaled HBT parameter (divided by $g^{(1)}(z)$), we find:

$$\begin{aligned} HBT_f = \frac{g_f^{(2)}(0) - [g_f^{(1)}(0)]^2}{g_f^{(1)}(0)} &- 2L_3^2 z^2\Big\{ 2\left[g_f^{(3)}(0) - g_f^{(2)}(0) g_f^{(1)}(0)\right] \\ + g_f^{(2)}(0) + 2\frac{L_5}{L_3}\Big[2\Big(g_f^{(4)}(0) &- g_f^{(3)}(0) g_f^{(1)}(0)\Big) + 3g_f^{(3)}(0)\Big] + \cdots \Big\} : g_f^{(1)}(0) \; . \end{aligned} \tag{5}$$

At coherent incident light, the correlation functions $g_f^{(k)}(0) = \langle n_{f0}\rangle^k$ are given by the respective power of the mean number of photons incident on the nonlinear medium. Accordingly, in Eq.(5), all "classical" terms with the highest powers of $\langle n_{f0}\rangle$ (square brackets) vanish and only strictly quantum terms remain:

$$HBT_f = -2k(z)\left\{ 1 + 6\frac{L_5}{L_3}\langle n_{f0}\rangle \right\} \; , \tag{6}$$

where $k(z) = \langle n_h(z)\rangle / \langle n_{f0}\rangle = L_3^2 \langle n_{f0}\rangle z^2$ is the conversion ratio.

The term proportional to L_5 tends to enhance photon antibunching, signified by "-" in Eq. (6). From the classical viewpoint, a beam of constant intensity should generate a harmonic with non-fluctuating intensity, thus itself remaining unfluctuating albeit experiencing a loss in its initial intensity. The whole picture, however, changes completely even in our rough, point-shaped photon approach (Kozierowski, 1981). In the absence of the medium, the photons of coherent light would be incident randomly onto the photodetector, like drops of rain. The phenomenon of generation, involving "extraction" of at least two phonons simultaneously in a single elementary event, occurs more strongly in intervals in which the number of incident photons is greater. Consequently, beside a decrease in the mean number of photons, we deal with their "rarefaction", meaning that the distances between successive photons tend to become equal. This now is the essence of photon antibunching. From the viewpoint of the fundamental beam the L_5-process is an additional factor in lowering the number of photons and their disorder — hence its synergetic role in Eq.(6). The scaled HBT_f parameter increases with the number of photons by way of the increase in both components, whereas the normalized parameter (divided by $[g^{(\omega)}(0)]^2$) has its first term constant and its second term increasing to a limit for $\langle n_{f0}\rangle$ tending to values corresponding to optical breakdown.

In the case of the harmonic beam the L_5-process counteracts the photon antibunching arising from the main L_3-process. Moreover, the influence of the L_5-term derived by us is apparent already in the z^2-approximation, whereas the essential L_3-component does not appear before z^4:

$$HBT_h = 4L_3L_5\langle n_{f0}\rangle^2 z^2 - \frac{8}{3}L_3^4\langle n_{f0}\rangle^2 z^4 \simeq \simeq -\frac{8}{3}k^2(z)\left\{1 - \frac{3}{2}\frac{L_5}{L_3}\frac{\langle n_{f0}\rangle}{k(z)}\right\}. \tag{7}$$

The conversion ratio is significantly less than unity. In this case only, our perturbation expansion is physically meaningful. The influence of the L_5-process, now counteractive, is additionally enhanced by the inverse of the conversion ratio. Photon antibunching given by the term at z^4, already less than antibunching in the fundamental mode (fraction to power 2), is further reduced. The term in L_5 now has the following effects:

firstly, it raises the number of photons in the second harmonic; secondly, representing a process practically independent of the principal L_3-process, it favours greater randomness in the distances between the newly created photons. All this counteracts photon antibunching.

SQUEEZING

Squeezed states involve the quantum phase properties of the field. Such states arise if the variance of whichever of the observables

$$Q = a^{+} + a \ , \ P = i(a^{+} - a) \tag{8}$$

fulfils the conditions: $\langle(\Delta Q)^2\rangle < 1$ or $\langle(\Delta P)^2\rangle < 1$. We restrict ourself to the determination of changes in the slowly variable parts of the variables.

For the fundamental beam we find, at coherent incident light,

$$\left.\begin{array}{l}\langle[\Delta Q_f(z)]^2\rangle \\ \langle[\Delta P_f(z)]^2\rangle\end{array}\right\} = 1 \mp 2k(z)\left\{1 + 6\frac{L_5}{L_3}\langle n_{f0}\rangle\right\}\cos 2\theta , \tag{9}$$

where θ is the initial phase of the complex amplitude α_f, and $|\alpha_f|^2 = \langle n_{f0}\rangle$. It is seen that, dependent on the phase, Q or P is squeezed. The maximal deviation from unity is equal to the photon antibunching, so that the influence of the higher-order susceptibility inherent in L_5 is here quite similar. Usually, the quantities (8) are defined with a coefficient of 1/2; to take account of this, the parameters (9) would have to be divided by 4. However, the normalized ones would remain unchanged.

With regard to the harmonic beam, the L_5-process is counteractive, lowering squeezing effect /as it did in photon antibunching/:

$$\left.\begin{array}{l}\langle[\Delta Q_h(z)]^2\rangle \\ \langle[\Delta P_h(z)]^2\rangle\end{array}\right\} = 1 \pm \frac{\overset{4}{\cancel{8}}}{3}k^2(z)\left\{1 - \frac{3}{2}\frac{L_5}{L_3}\frac{\langle n_{f0}\rangle}{k(z)}\right\}\cos 4\theta . \tag{10}$$

In the general Eqs (9) and (10) the distribution of signs at the variances of Q and P is opposite. But of course the final signs of the variances are phase dependent, and the factors at the phase are different in (9) and (10).

All in all, photon antibunching and squeezing should more easily be observed in the fundamental mode. In the latter, not only the contributions from the main L_3- process are greater than in the harmonic mode, but moreover there is a synergetic effect from all the other higher-order nonlinearities in general.

As in the case of photon antibunching in arbitrary harmonics (Kielich et al., 1978; Kozierowski, 1981), one can consider squeezed states of higher harmonics (Kozierowski and Kielich, 1983) of the laser beam.

Photon antibunching and squeezing are intimately related to the quantum nature of light. The former is primarily a manifestation of corpuscularity, whereas the latter reveals the wave nature of light. Both effects offer the opportunity of performing experiments being very much in the spirit of "experimenta crucis" testing Q.E.D. . Higher-harmonic generation processes offer one of the possibilities in this direction.

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