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Quantum statistics of single-beam two-photon absorption

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Abstract. The effects of two-photon absorption from a single beam of light on the statistical properties of the beam are considered. Simple expressions are obtained for the initial rates of change and limiting steady-state values of the mean photon number and degree of second-order coherence of the beam. The general time dependences of these properties of the light and of the complete photon probability distribution are evaluated for several initial types of beam. The physical significance of the results is described, and their relationship to calculations which use a classical representation of the light is discussed.

1. Introduction

The linear absorption and stimulated emission of light take place with probabilities which are proportional to the number n of photons in the light. The time-averaged absorption and emission rates are proportional to the mean photon number $\bar{n}$ or mean intensity of the light; they are independent of its coherence properties (see for example Loudon 1973). The rates of nonlinear optical processes, on the other hand, usually depend on the degrees of coherence of the light beam and on correlations between light beams, when more than one beam participates in the process.

The present paper considers the statistical features of the nonlinear process of two-photon absorption from a single beam of light. The two-photon absorption probability is proportional to $n(n-1)$, and the time-averaged absorption rate is proportional to the square of the mean number of photons and to the degree of second-order coherence of the light, denoted by $g^{(2)}$. This result has been known for some time (Teich and Wolga 1966, Lambropoulos *et al* 1966, Shen 1967) and a derivation is given below (see equation (34)). One well known consequence is the prediction that for equal mean photon numbers $\bar{n}$, a beam of chaotic light is two-photon absorbed at twice the rate of a beam of coherent light, and this has received experimental support from the work of Shiga and Imamura (1967).

The proportionality of the time-averaged two-photon absorption rate to the degree of second-order coherence can be understood in qualitative terms. The size of $g^{(2)}$ provides a measure of the magnitude of the fluctuations in the photon number n. Since the probability of two-photon absorption is proportional to $n(n-1)$, enhanced absorption occurs for beams whose photon numbers fluctuate about the mean $\bar{n}$. The absorption from a fluctuating light beam occurs preferentially at the maxima of n. This property is clearly illustrated, for a classical model of the intensity fluctuations in chaotic light, by Weber (1971) in figure 2 of his paper. As the two-photon absorption proceeds the intensity peaks are rapidly eroded, while the intensity troughs are barely changed by the negligibly small absorption at low intensity.

Two-photon absorption therefore smooths intensity or photon-number fluctuations and causes changes in the statistical properties of the unabsorbed portion of the beam. In particular the magnitude of $g^{(2)}$ is changed by a significant amount of two-photon absorption, leading to associated changes in the absorption rate itself, which are often neglected in conventional treatments of the absorption process. The present paper is concerned with this interdependence of the two-photon absorption and the photon-number fluctuations of the beam.

The equations which describe the rate of change of the photon statistical distribution caused by two-photon absorption and emission are introduced in § 2. This is followed in § 3 by a brief review of the photon distributions and coherence properties of several types of light beam whose two-photon absorption is treated in later sections. The main calculations begin in § 4 with a determination of the changes in $\bar{n}$ and $g^{(2)}$ after a short period of two-photon absorption. The opposite extreme of the steady state achieved after a long period of absorption is treated in § 5. The short-time and steady-state solutions give physical insight into the mechanisms by which the changes in the beam statistical properties are brought about.

A generating function method introduced by Agarwal (1970) and developed by McNeil and Walls (1974) and Tornau and Bach (1974) is used in § 6 to obtain the complete time dependences of the mean photon number $\bar{n}$, the second factorial moment $\overline{n(n-1)}$ and the elements P_n of the photon probability distribution. The varieties of behaviour shown by a range of initial types of beam are discussed in physical terms in § 7, and the results are compared with those of calculations which ignore the time dependence of $g^{(2)}$ or use a classical model of the light beam.

2. Photon rate equations

Consider the photons in a single mode of the radiation field whose frequency allows two-photon absorption by a gas of N atoms. It is assumed that the atoms have no transitions of the required frequency for single-photon absorption. Suppose that N_1 atoms are in the lower state and a smaller number N_2 are in the upper state of the two-photon transition, with

$$N_1 + N_2 = N. \tag{1}$$

The numbers of atoms in the two states are assumed to be fixed by some external influence, but the number n of photons is regarded as a statistical quantity whose probability distribution P_n changes with time.

The probability per unit time that a two-photon absorption takes place, with a change in the photon number from n to $n-2$, can be written as

$$N_1 J n(n-1), \tag{2}$$

where an expression for J in terms of atomic energy levels and dipole matrix elements can be obtained for example from Loudon (1973). The corresponding probability per unit time of a two-photon emission, leading to an increase in the photon number from n to $n+2$, is

$$N_2 J(n+1)(n+2). \tag{3}$$

The two-photon absorption and emission cause changes in P_n, the probability that the radiation field contains n photons.

There are four types of transition which contribute to the rate of change of P_n. If n photons are indeed present in the radiation field, the absorption and emission described by (2) and (3) reduce P_n at a combined rate

$$-N_1Jn(n-1)P_n - N_2J(n+1)(n+2)P_n. \tag{4}$$

There are also two positive contributions to the rate of change of P_n. If $n-2$ photons are present, with probability P_{n-2}, emission of two photons increases P_n at a rate determined by (3) with n replaced by $n-2$:

$$N_2J(n-1)nP_{n-2}. \tag{5}$$

Similarly, if $n+2$ photons are present, two-photon absorption increases P_n at a rate determined by (2) with n replaced by $n+2$:

$$N_1J(n+2)(n+1)P_{n+2}. \tag{6}$$

The total rate of change of P_n obtained from (4), (5) and (6) is

$$\begin{aligned} \mathrm{d}P_n/\mathrm{d}t = & -N_1Jn(n-1)P_n - N_2J(n+1)(n+2)P_n \\ & + N_2J(n-1)nP_{n-2} + N_1J(n+2)(n+1)P_{n+2}. \end{aligned} \tag{7}$$

The four contributions to the rate of change are entered on the photon energy level diagram in figure 1. The first and third terms in (7) should be removed for $n = 0$ and $n = 1$, when the processes described by these terms cannot occur. An equation identical to (7) can be derived by density operator techniques using an explicit form for the photon–atom interaction Hamiltonian (Shen 1967, Lambropoulos 1967, Agarwal 1970, McNeil and Walls 1974).

The system of rate equations, of which (7) is a representative, can be divided into two sets, one of which couples all the P_n for positive odd integers n, and the other of which couples all the P_n for positive even integers n and $n = 0$. It is seen by addition of all the equations in each of the sets that the sums of P_n over all even or all odd integers n are constants of the motion. Thus if $P_n(0)$ is the photon probability distribution at time $t = 0$,

$$\sum_n^{\text{even}} P_n = \sum_n^{\text{even}} P_n(0) \tag{8}$$

$$\sum_n^{\text{odd}} P_n = \sum_n^{\text{odd}} P_n(0). \tag{9}$$

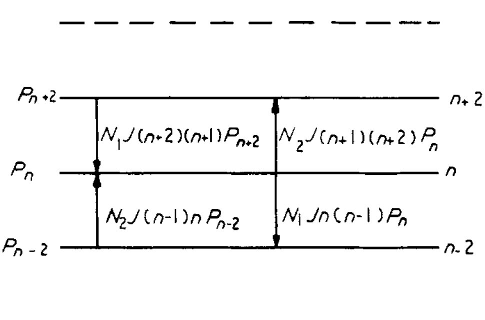

Figure 1. Energy level diagram for the photons. The level separation is twice the photon energy and the transition rates indicated are the contributions to $\mathrm{d}P_n/\mathrm{d}t$.

The rate equations used here are independent of position, since propagation effects have not been included. The analysis which follows is valid for the interaction of radiation with atoms in a cavity whose length is sufficiently short for the transit time of light to be small compared with the time scale of changes in P_n.

3. Photon probability distributions

The two-photon absorption of several types of initial light beam is treated in later sections. The main properties of these beams are summarized without proof in the present section. Most of the results can be taken directly from the literature (see for example Loudon 1973), while the remainder require only a little derivation. In each case expressions are given for the normalized photon probability distribution, for its second and third moments, where the rth moment is defined by

$$\overline{n^r} = \sum_n n^r P_n, \tag{10}$$

and for the degree of second-order coherence $g^{(2)}$. This latter quantity is determined for any single-mode light beam by the second factorial moment of the probability distribution,

$$g^{(2)} = (\overline{n^2} - \bar{n})/\bar{n}^2. \tag{11}$$

3.1. The number-state beam

A beam with a definite number $\bar{n}_0$ of photons at time $t = 0$ has the initial probability distribution

$$P_n(0) = \delta_{n\bar{n}_0} \qquad \delta_{n\bar{n}_0} = 1 \text{ for } n = \bar{n}_0 \tag{12}$$
$$= 0 \text{ otherwise.}$$

The second and third moments are

$$\overline{n_0^2} = \bar{n}_0^2, \qquad \overline{n_0^3} = \bar{n}_0^3, \tag{13}$$

and the degree of second-order coherence is

$$g_0^{(2)} = (\bar{n}_0 - 1)/\bar{n}_0 \qquad \text{for} \qquad \bar{n}_0 \geqslant 1 \tag{14}$$

and is indeterminate for $\bar{n}_0 = 0$. The number state is not of great practical interest but it is a useful limit in which to evaluate the theory.

3.2. The coherent beam

The initial probability $P_n(0)$ for a mean photon number $\bar{n}_0$ is given by a Poisson distribution

$$P_n(0) = (\bar{n}_0^n/n!)\exp(-\bar{n}_0), \tag{15}$$

leading to

$$\overline{n_0^2} = \bar{n}_0^2 + \bar{n}_0 \tag{16}$$

$$\overline{n_0^3} = \bar{n}_0^3 + 3\bar{n}_0^2 + \bar{n}_0 \tag{17}$$

$$g_0^{(2)} = 1. \tag{18}$$

The photon state of a single-mode laser well above threshold is a close approximation to a coherent beam.

3.3. The chaotic or thermal beam

The photon statistics in this case are given by a Bose–Einstein distribution

$$P_n(0) = \bar{n}_0^n/(1+\bar{n}_0)^{1+n}, \tag{19}$$

leading to

$$\overline{n_0^2} = 2\bar{n}_0^2 + \bar{n}_0 \tag{20}$$

$$\overline{n_0^3} = 6\bar{n}_0^3 + 6\bar{n}_0^2 + \bar{n}_0 \tag{21}$$

$$g_0^{(2)} = 2. \tag{22}$$

Lasers below threshold and conventional light sources provide chaotic or thermal beams.

3.4. The pulsed coherent beam

This kind of beam is obtained by repeatedly switching a beam of coherent light on and off. If $\bar{n}_0/f$ is the mean photon number of the coherent beam and the duty factor f is the fraction of the time for which the beam is turned on, then the mean photon number of the pulsed beam is $\bar{n}_0$, independent of f. The probability distribution is

$$P_n(0) = f[(\bar{n}_0/f)^n/n!]\exp(-\bar{n}_0/f) \qquad \text{for } n \neq 0 \tag{23}$$

$$P_0(0) = 1 - f + f\exp(-\bar{n}_0/f),$$

leading to

$$\overline{n_0^2} = (\bar{n}_0^2/f) + \bar{n}_0 \tag{24}$$

$$\overline{n_0^3} = (\bar{n}_0^3/f^2) + (3\bar{n}_0^2/f) + \bar{n}_0 \tag{25}$$

$$g_0^{(2)} = 1/f. \tag{26}$$

The degree of second-order coherence of the pulsed beam can have any value greater than unity by suitable choice of f.

4. Short-time solutions

It is instructive to consider the changes in the initial statistical properties of the photons to first order in the time t. We derive the initial rates of change of the first and second moments of the photon distribution, and combine these to obtain the rate of change of $g^{(2)}$.

Equations for the rates of change of the moments are obtained by time-differentiation of (10) and insertion of the rate of change of P_n from (7):

$$\mathrm{d}\bar{n}/\mathrm{d}t = 2(N_2-N_1)J\overline{n^2}+2(3N_2+N_1)J\bar{n}+4N_2J \tag{27}$$

$$\mathrm{d}\overline{n^2}/\mathrm{d}t = 4(N_2-N_1)J\overline{n^3}+8(2N_2+N_1)J\overline{n^2}+4(5N_2-N_1)J\bar{n}+8N_2J. \tag{28}$$

These expressions differ by numerical factors from the corresponding equations (16) of Shen (1967) and (7.2) of McNeil and Walls (1974), but agree with Lambropoulos (1967). It is seen from (27) and (28) that the change with time of each moment of the distribution depends on the next higher moment, making a simple solution of the equations impossible.

It is however easily possible to obtain the initial behaviours of the moments. If the values of the moments at $t = 0$ are substituted on the right-hand sides of (27) and (28), then these expressions are the coefficients of t in power-series expansions, and correct to order t,

$$\bar{n} = \bar{n}_0+2Jt[N_2(\overline{n_0^2}+3\bar{n}_0+2)-N_1(\overline{n_0^2}-\bar{n}_0)] \tag{29}$$

$$\overline{n^2} = \overline{n_0^2}+4Jt[N_2(\overline{n_0^3}+4\overline{n_0^2}+5\bar{n}_0+2)-N_1(\overline{n_0^3}-2\overline{n_0^2}+\bar{n}_0)]. \tag{30}$$

The corresponding expansion of $g^{(2)}$ can be obtained from these results and (11) with some algebra:

$$g^{(2)} = g_0^{(2)}+(2Jt/\bar{n}_0^2)\{N_2[2\overline{n_0^3}+3\overline{n_0^2}+13\bar{n}_0+6-(\overline{n_0^2}/\bar{n}_0)(2\overline{n_0^2}+4)] \\ -N_1[2\overline{n_0^3}-\overline{n_0^2}+\bar{n}_0-2(\overline{n_0^2}/\bar{n}_0)\overline{n_0^2}]\}. \tag{31}$$

The linear time dependences of $\bar{n}$, $\overline{n^2}$ and $g^{(2)}$ for various types of initial light beam can now be obtained straightforwardly by substitution of the appropriate expressions for $\overline{n_0^3}$, $\overline{n_0^2}$ and $g_0^{(2)}$ from § 3. For example, in the case of initially coherent light (31) gives

$$g^{(2)} = 1+(2Jt/\bar{n}_0^2)[(5N_2-N_1)\bar{n}_0^2+12N_2\bar{n}_0+2N_2], \tag{32}$$

in agreement with the finding of Chandra and Prakash (1970) that $g^{(2)}$ decreases below the value unity if $\bar{n}_0 \gg 1$ and $N_2 < N/6$.

The rather complicated expressions given above all simplify for the special case where almost all the atoms are in their lower levels. This also corresponds closely to the normal experimental situation where there is a negligibly small excited state population. We take $N_2 = 0$ and $N_1 = N$ for the remainder of the present section and define a new time variable

$$\tau = NJt. \tag{33}$$

Then (27) becomes

$$\mathrm{d}\bar{n}/\mathrm{d}\tau = -2g^{(2)}\bar{n}^2 \tag{34}$$

where (11) has been used, and (29) can be written as

$$\bar{n} = \bar{n}_0(1-2g_0^{(2)}\bar{n}_0\tau). \tag{35}$$

Thus for small t, where this equation is valid, and taking $g_0^{(2)}$ from § 3, it is seen that chaotic light is two-photon absorbed at twice the rate for coherent light, as mentioned in § 1, the rate for the number-state beam is close to that for coherent light, and the rate for the pulsed beam increases with decreasing pulse duration.

The results for the degree of second-order coherence, obtained by substitution into (31), are

$$g^{(2)} = [(\bar{n}_0-1)/\bar{n}_0](1+2\tau) \quad \text{(number)} \tag{36}$$

$$g^{(2)} = 1-2\tau \quad \text{(coherent)} \tag{37}$$

$$g^{(2)} = 2-4\tau(2\bar{n}_0+1) \quad \text{(chaotic)} \tag{38}$$

$$g^{(2)} = (1/f)(1-2\tau) \quad \text{(pulsed).} \tag{39}$$

It is seen that the rate of decrease in $g^{(2)}$ for chaotic light exceeds that for coherent light by a factor $2(2\bar{n}_0+1)$. The number-state beam differs from the other three in showing an initial increase in $g^{(2)}$.

The initial variations in $\bar{n}$ and $g^{(2)}$ are shown at the short-time ends of the figures given in § 7, where their behaviour is discussed in greater detail.

5. Steady-state solutions

The photon system settles down into a steady state after a sufficiently long period of time has elapsed. The rates of change (7) are equal to zero in the steady state, leading to two chains of equations for the P_n, one for even n and the other for odd n. If the steady-state probability distribution is denoted $P_n(\infty)$, the rate equations give

$$N_1 P_n(\infty) = N_2 P_{n-2}(\infty). \tag{40}$$

It is seen by reference to figure 1 that this is just the condition for detailed balance between the photon levels n and $n-2$.

By iteration of (40)

$$P_n(\infty) = (N_2/N_1)^{n/2} P_0(\infty) \quad \text{for } n \text{ even} \tag{41}$$

$$P_n(\infty) = (N_2/N_1)^{(n-1)/2} P_1(\infty) \quad \text{for } n \text{ odd.} \tag{42}$$

Hence

$$\sum_n^{\text{even}} P_n(\infty) = [1-(N_2/N_1)]^{-1} P_0(\infty) = \sum_n^{\text{even}} P_n(0) \tag{43}$$

$$\sum_n^{\text{odd}} P_n(\infty) = [1-(N_2/N_1)]^{-1} P_1(\infty) = \sum_n^{\text{odd}} P_n(0), \tag{44}$$

where (8) and (9) have been used. Equations (41) to (44) enable $P_n(\infty)$ to be determined for any given initial distribution $P_n(0)$. Addition of (43) and (44) gives the simple result

$$P_0(\infty)+P_1(\infty) = 1-(N_2/N_1). \tag{45}$$

It is seen that the steady-state photon distribution given by (41) and (42) has some resemblance to a chaotic distribution (compare equation (10.17) of Loudon 1973), but it differs by the separation into two parts which are not coupled by the two-photon absorption and emission. The moments of the distribution can be determined without difficulty; the mean number of photons in the steady state is

$$\bar{n}_\infty = \frac{2(N_2/N_1)+P_1(\infty)}{1-(N_2/N_1)}, \tag{46}$$

and the second factorial moment is

$$\overline{n^2_\infty} - \bar{n}_\infty = \frac{N_2}{N_1}\,\frac{6(N_2/N_1)+4P_1(\infty)+2}{[1-(N_2/N_1)]^2}. \tag{47}$$

The results again simplify when N_2 can be set equal to zero. In this case (41) and (42) show that

$$P_n(\infty) = 0 \qquad \text{for } n \geqslant 2, \tag{48}$$

and the steady state is achieved when there are no pairs of photons left to be absorbed. From (46) and (47)

$$\bar{n}_\infty = P_1(\infty) \tag{49}$$

and

$$\overline{n^2_\infty} - \bar{n}_\infty = 0. \tag{50}$$

It follows from (11) that

$$g^{(2)}_\infty = 0, \tag{51}$$

except in the special case where $\bar{n}_\infty = 0$, considered in § 7. Table 1 shows the values of $P_0(\infty)$ and $P_1(\infty)$ for several examples.

The steady-state values of $\bar{n}$, $g^{(2)}$ and P_n are shown at the large-time ends of the figures of § 7. We note that figure 1 of Agarwal (1970), showing the time dependence of $\bar{n}$ for an initially coherent beam with $\bar{n}_0 = 10$, has the incorrect steady-state value $\bar{n}_\infty = 1$. In addition, the variance of the distribution shown in figure 3 of this reference should have the steady-state value of 0·25.

6. General solutions

Agarwal (1970) has pointed out that the rate equations (7) can be solved for the time-dependent photon probability distribution $P_n(\tau)$ by a generating function method in the case where $N_2 = 0$. We define

$$F(y, \tau) = \sum_{n=0}^{\infty} y^n P_n(\tau), \tag{52}$$

where τ is defined by (33). On multiplication of both sides of (7) by y^n and summation over n we obtain

$$\partial F/\partial \tau = (1-y^2)\partial^2 F/\partial y^2. \tag{53}$$

The generating function provides a simple means of obtaining the factorial moments of the distribution, and it is seen from (10) and (52) that

$$\overline{n(n-1)(n-2)\ldots(n-r+1)} = (\partial^r F/\partial y^r)_{y=1}, \tag{54}$$

while the distribution itself is obtained from

$$P_n(\tau) = (n!)^{-1}(\partial^n F/\partial y^n)_{y=0}. \tag{55}$$

The solution of (53) by the method of separation of the variables has been considered by Agarwal (1970), McNeil and Walls (1974) and Tornau and Bach (1974).

The generating function has the form

$$F(y,\tau) = \sum_{k=0}^{\infty} A_k C_k^{-\frac{1}{2}}(y)\exp[-k(k-1)\tau], \tag{56}$$

where $C_k^{-\frac{1}{2}}(y)$ is a Gegenbauer polynomial. The coefficients A_k can be obtained by a method described by McNeil and Walls, and we find

$$A_k = -(2k-1) \sum_{m=k}^{\infty}{}^{(m-k\,\text{even})} \frac{m!\Gamma(\frac{1}{2}m-\frac{1}{2}k+\frac{1}{2})}{2^k(m-k)!\Gamma(\frac{1}{2}m+\frac{1}{2}k+\frac{1}{2})}P_m(0), \tag{57}$$

where $P_m(0)$ is the initial photon distribution. The restriction of the summation to values of m which differ from k by even integers is a consequence of the separation of the rate equations into two chains for even and odd photon numbers. We note that (57) differs by a sign in the gamma function in the denominator from a result for even k given by McNeil and Walls (Walls, private communication, has confirmed the presence of a misprint in equation (5.10) of this paper).

The Gegenbauer polynomials have the following properties (see for example § 10.9 of Erdélyi *et al* 1953):

$$\begin{aligned}(\partial/\partial y)^l C_k^{-\frac{1}{2}}(y) &= [2^l\Gamma(l-\tfrac{1}{2})/\Gamma(-\tfrac{1}{2})]C_{k-l}^{l-\frac{1}{2}}(y) \qquad &\text{for } k \geqslant l\\ &= 0 &\text{for } k < l\end{aligned} \tag{58}$$

and

$$C_{k-r}^{r-\frac{1}{2}}(1) = (k+r-2)!/(k-r)!(2r-2)! \tag{59}$$

$$\begin{aligned}C_{k-n}^{n-\frac{1}{2}}(0) &= (-1)^{\frac{1}{2}k-\frac{1}{2}n}\Gamma(\tfrac{1}{2}k+\tfrac{1}{2}n-\tfrac{1}{2})/\Gamma(n-\tfrac{1}{2})\Gamma(\tfrac{1}{2}k-\tfrac{1}{2}n+1) \qquad &\text{for } k-n \text{ even}\\ &= 0 &\text{for } k-n \text{ odd.}\end{aligned} \tag{60}$$

It follows from (54), (56), (58) and (59) that the first two factorial moments are given by

$$\bar{n} = -\sum_{k=1}^{\infty} A_k \exp[-k(k-1)\tau] \tag{61}$$

$$\overline{n(n-1)} = -\tfrac{1}{2}\sum_{k=2}^{\infty} k(k-1)A_k\exp[-k(k-1)\tau], \tag{62}$$

while the photon probability distribution obtained from (55), (56), (58) and (60) is

$$P_n(\tau) = \sum_{k=n}^{\infty}{}^{(k-n\,\text{even})} \frac{(-1)^{\frac{1}{2}k-\frac{1}{2}n}2^n\Gamma(\frac{1}{2}k+\frac{1}{2}n-\frac{1}{2})}{n!\Gamma(-\frac{1}{2})\Gamma(\frac{1}{2}k-\frac{1}{2}n+1)}A_k\exp[-k(k-1)\tau]. \tag{63}$$

For any given initial photon distribution, A_k is determined by (57) and these equations provide explicit expressions for the photon distribution and its moments as functions of the time.

We conclude this section by relating the general solution for the photon distribution to its steady-state properties derived in the previous section. At infinitely long times τ, (63) gives

$$P_0(\infty) = A_0 \tag{64}$$

$$P_1(\infty) = -A_1, \tag{65}$$

and the probability for two or more photons is zero, in agreement with (48). From (57),

$$A_0 = \sum_m^{\text{even}} P_m(0) \tag{66}$$

$$A_1 = -\sum_m^{\text{odd}} P_m(0) \tag{67}$$

and the steady-state values of P_0 and P_1 given by (64) and (65) thus agree with our previous results (43) and (44) in the case $N_2 = 0$. It is seen from (66) that A_0 does not in general have the value unity assumed by McNeil and Walls (1974).

7. Discussion

The main features of the changes in photon statistical properties brought about by two-photon absorption are best appreciated by consideration of some special cases. Graphs are presented in this section for the time dependences of the mean photon number, the degree of second-order coherence, and the photon probability distribution for the case $N_2 = 0$ and for a variety of initial photon distributions. The graphs have been constructed by evaluation of the summations in (61), (62) and (63) with the help of a computer.

The five curves in figures 2 and 3 show the time variations of $\bar{n}$ for the five types of initial photon state listed in table 1. The behaviours of $\bar{n}$ at short times are consistent with equation (35), and in particular the factor of two difference between the initial absorption rates of chaotic and coherent light is clearly shown in figure 3. The steady-state limits of $\bar{n}$ are as indicated in table 1; the differences in steady-state behaviour are particularly marked for the odd and even number-state beams shown in figure 2.

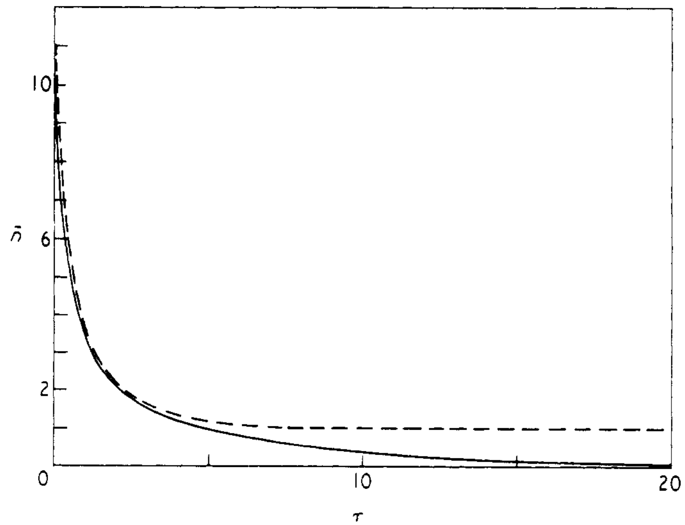

Figure 2. Time dependences of the mean photon numbers $\bar{n}$ for beams which are initially number states containing 10 photons (———) and 11 photons (– – –).

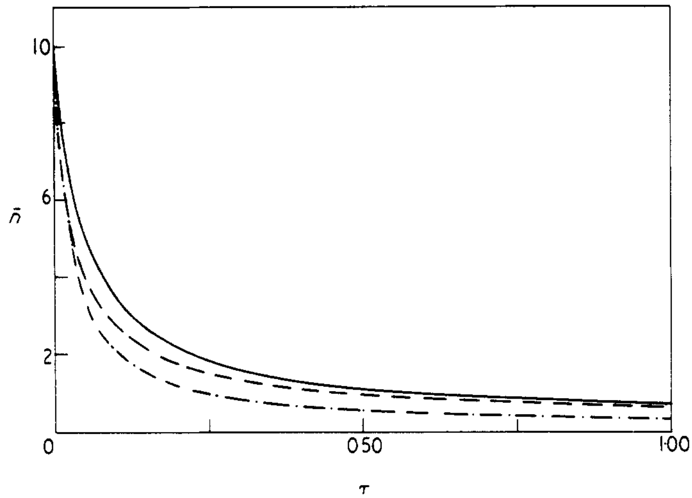

Figure 3. Time dependences of the mean photon numbers $\bar{n}$ for beams which are initially coherent (———), chaotic (- - -) and pulsed (-·-·-·-). All the beams have an initial mean photon number $\bar{n}_0 = 10$, and the pulsed beam has a duty factor $f = \frac{1}{2}$.

Table 1. The steady-state photon probability distributions for $N_2 = 0$ and various types of initial photon distribution

	number state $\bar{n}_0$ even	number state $\bar{n}_0$ odd	coherent $\bar{n}_0 = 10$	chaotic $\bar{n}_0 = 10$	pulsed $\bar{n}_0 = 10, f = \frac{1}{2}$
$P_0(\infty)$	1	0	0·50	0·53	0·75
$P_1(\infty) = \bar{n}_\infty$	0	1	0·50	0·47	0·25

In conventional treatments of two-photon absorption it is customary to ignore the time dependence of $g^{(2)}$, which is set equal to the initial value $g_0^{(2)}$. With this substitution the solution of (34) is

$$\bar{n} = \frac{\bar{n}_0}{1+2g_0^{(2)}\bar{n}_0\tau}. \tag{68}$$

This approximate solution for $\bar{n}$ can be compared with the exact solutions shown in figures 2 and 3. It is seen that (68) agrees with the exact solution at short times given in (35), but at longer times the approximate solution gives smaller $\bar{n}$ than the exact results. The discrepancy is particularly serious for initially chaotic light.

The approximate solution fails at longer times because the degree of second-order coherence falls below its initial value and thus causes a decrease in the absorption rate. Exact results for the time variations of $g^{(2)}$ for the five light beams are shown in figures 4 and 5. The short-time behaviours of $g^{(2)}$ are in accordance with equations (36) to (39), while $g^{(2)}$ generally tends to zero in the steady state. An exception to this behaviour occurs for any initial number state in which an even number of photons is present. In this case the denominator of $g^{(2)}$ defined in (11) tends to zero at a faster rate than the

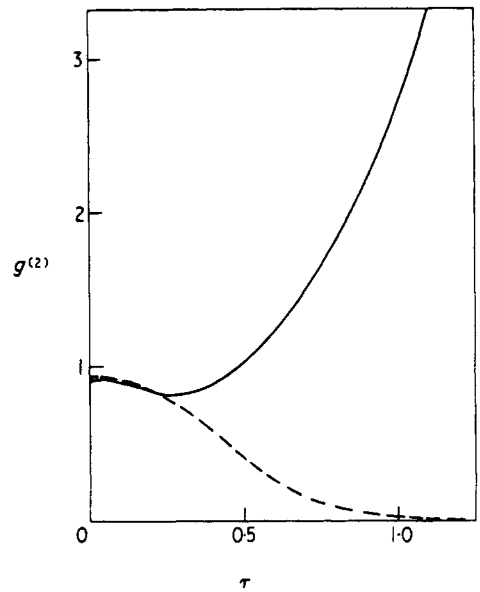

Figure 4. Time dependences of the degrees of second-order coherence $g^{(2)}$ for beams which are initially number states containing 10 photons (———) and 11 photons (– – –).

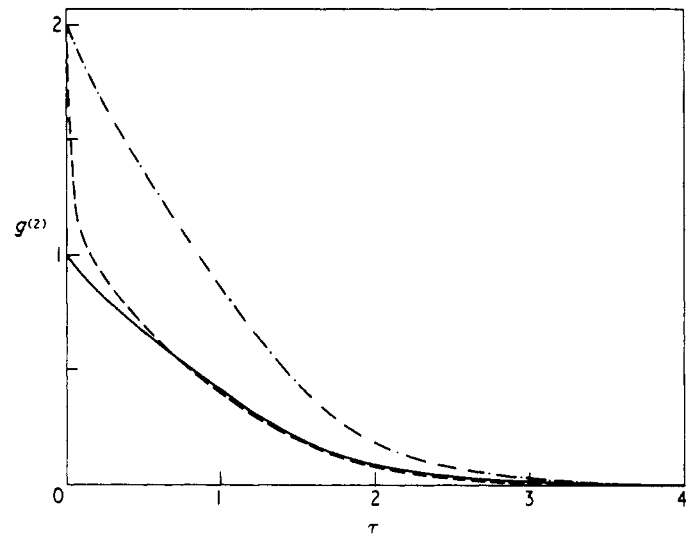

Figure 5. Time dependences of the degrees of second-order coherence $g^{(2)}$ for beams which are initially coherent (———), chaotic (– – –) and pulsed (– · – · –), with the same parameters as in figure 3.

numerator as τ tends to infinity, leading to an anomalous divergence in $g^{(2)}$. Experimental light beams have $\bar{n}_\infty$ different from zero and this anomalous behaviour of $g^{(2)}$ is not of practical importance.

Apart from this exception the smoothing of fluctuations by the two-photon absorption produces degrees of second-order coherence which reduce with time. Of the beams considered, the fall-off is particularly noteworthy for the chaotic or thermal case. It is seen in figure 5 that $g^{(2)}$ for chaotic light falls rapidly from its initial value of two and that for τ greater than about 0·5 the initially chaotic and coherent beams have very similar values of their degrees of second-order coherence.

The calculations shown in figures 4 and 5 are quantum mechanical, since they make use of the degree of second-order coherence $g^{(2)}$ defined by (11) in terms of the mean and mean-square photon numbers. A previous study of the two-photon absorption of chaotic and coherent light has been made by Weber (1971) using a classical description of the light. The classical degree of second-order coherence is defined by

$$g^{(2)} = \overline{I^2}/\bar{I}^2, \tag{69}$$

where I is the fluctuating intensity of the light. The classical $g^{(2)}$ does not take values smaller than unity, in contrast to the behaviour of the quantum-mechanical $g^{(2)}$ illustrated in figures 4 and 5. There is thus a major difference between the effect of two-photon absorption on the magnitudes of the degrees of second-order coherence defined quantum mechanically and classically.

The source of the discrepancy lies in the nature of the photon state to which the light tends in the steady state. When $N_2 = 0$ it is seen from (41) and (42) that the only nonzero elements of the photon distribution in the steady state are $P_0(\infty)$ and $P_1(\infty)$, their sum being unity in accordance with (45). The steady state is thus a mixture of the photon number states $|0\rangle$ and $|1\rangle$, the quantum-mechanical $g^{(2)}$ being zero for both states. Now any light beam which has a definite number of photons (other than zero) or is a mixture of a few restricted number states can only be treated quantum mechanically since it cannot be represented by any probability distribution of classical intensity (see for example Perina 1972). The classical $g^{(2)}$ is therefore incapable of describing the second-order coherence either of the number-state beams of figure 4 or of the steady states of the beams represented in figure 5 (see Klauder and Sudarshan (1968) for discussion of this point).

It is seen from comparison of figures 3 and 5 that the quantum-mechanical $g^{(2)}$ falls significantly below the classical lower bound of unity only when the mean photon numbers have been reduced to the order of one or two. The quantum-mechanical effects thus occur at very low light intensities where experimental observation is difficult. No similar effects occur in linear or single-photon absorption, where for $N_2 = 0$ the photon distribution retains its initial nature as the mean photon number is reduced.

Because of the difference between the effects of two-photon absorption in quantum mechanics and classical theory it is not possible to make a detailed comparison of our results with those of Weber (1971). However, the classical calculations show the same qualitative behaviour as figure 5 in that the $g^{(2)}$ of initially chaotic light approaches that of initially coherent light after a period of two-photon absorption corresponding to τ of the order of 0·5.

The mean photon number and the degree of second-order coherence have been discussed in some detail because their time dependences give the most compact description of the effects of the two-photon absorption on the light. However, the time dependence of the complete photon probability distribution is readily obtainable from (63), and the remaining figures show some results for the first four initial distributions listed in table 1. Figures 6 and 7 show the time dependence of $P_n(\tau)$ for initial number-state beams with even and odd numbers of photons. All nonzero elements of the distributions are shown. At large values of τ, off the right-hand ends of the figures, the distributions tend to their steady-state forms in which only $P_0(\infty)$ and $P_1(\infty)$ are nonzero. For the special case of the initial number states, in which only even or odd elements

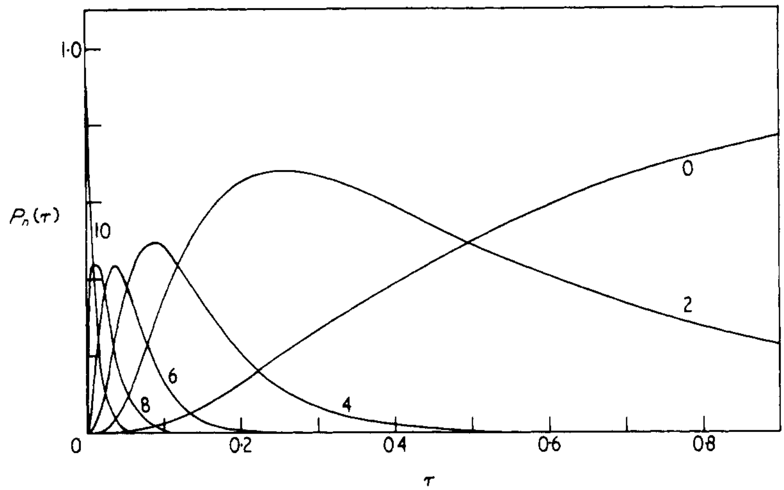

Figure 6. Time dependence of the photon probability distribution $P_n(\tau)$ for an initial number-state beam which contains 10 photons. The numbers attached to the curves indicate the corresponding values of n.

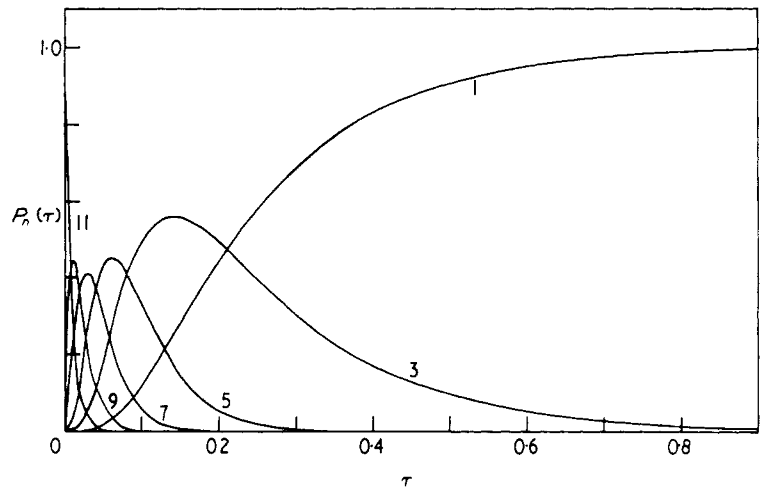

Figure 7. Time dependence of $P_n(\tau)$ for an initial number-state beam which contains 11 photons.

of the distribution occur, only $P_0(\infty)$ *or* $P_1(\infty)$ is nonzero, as shown in table 1. Figures 8 and 9 show corresponding results for initially coherent and chaotic beams respectively. In these cases the initial distribution extends over a wide range of values of n and the figures show the time dependences only of selected elements of the distribution. At large τ the distributions again tend to the appropriate steady-state limits given in table 1.

All the above remarks apply to the case where $N_2 = 0$. The behaviour of the light is more complicated if some of the atoms are maintained in the excited state and we make only a brief comment on this case. As described in § 5, the steady state for

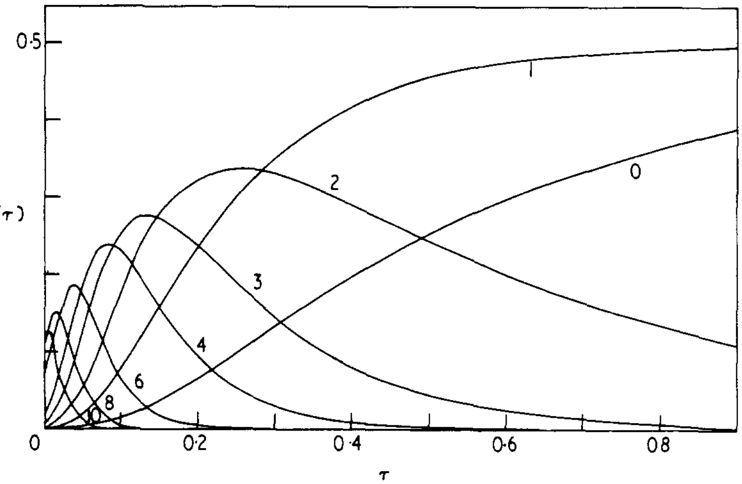

Figure 8. Time dependence of $P_n(\tau)$ for an initially coherent beam with mean photon number $\bar{n}_0 = 10$. Only selected elements of the distribution are shown.

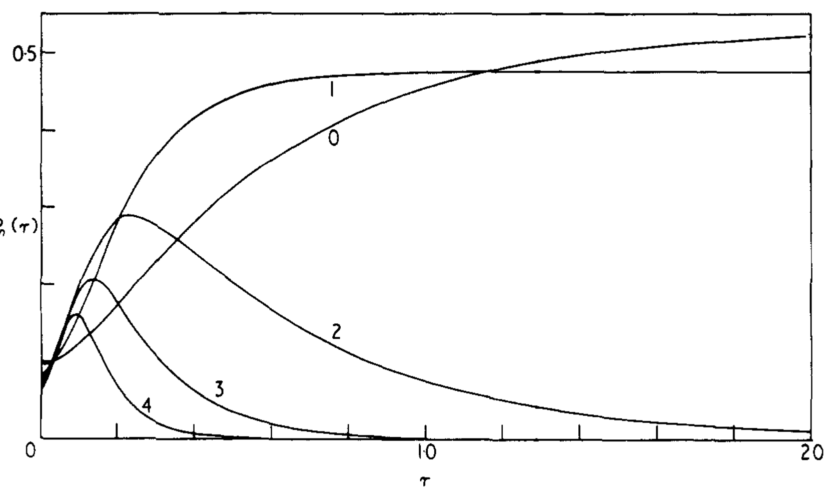

Figure 9. Time dependences of the low-n elements of $P_n(\tau)$ for an initially chaotic beam with mean photon number $\bar{n}_0 = 10$.

$N_2 \neq 0$ is one in which the even and odd photon-number states have separate probability distributions of a chaotic nature. The weights of the two parts of the distribution are determined by the initial photon distribution of the light in accordance with (43) and (44). In the very special case where these equations give

$$P_1(\infty) = (N_2/N_1)^{\frac{1}{2}} P_0(\infty) \tag{70}$$

(41) and (42) show that the two separate distributions combine to form a single chaotic distribution $P_n(\infty)$ which has the same form as (19) with a mean photon number given by (45), (46) and (70) as

$$\bar{n}_\infty = (N_2/N_1)^{\frac{1}{2}}[1-(N_2/N_1)^{\frac{1}{2}}]^{-1}. \tag{71}$$

However, the two parts of the distribution cannot in general be represented by a single expression $P_n(\infty)$ which is valid for both even and odd n. The degree of second-order coherence $g^{(2)}_\infty$ of the steady state is determined by (46) and (47); it is not difficult to show that $g^{(2)}_\infty$ can take any positive value by appropriate choices of the magnitudes of N_2/N_1 and $P_1(\infty)$. A wide variety of behaviours is therefore possible when $N_2 \neq 0$.

In summary, the calculations described above illustrate the effects of two-photon absorption on the fluctuations of light. Calculations which ignore changes in the degree of second-order coherence are correct only to first order in the time, and they are in error for the higher orders in t or τ, where the reaction of the two-photon absorption on the beam begins to influence the absorption rate. The analysis presented in this paper enables the time-dependent statistical properties of the light to be evaluated for any kind of initial photon probability distribution.

Acknowledgments

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Note added in proof. Stoler (1974) has shown that light having $g^{(2)} < 1$, which he calls the photon antibunching or anticorrelation effect, can be generated by a nonlinear optical experiment involving degenerate parametric amplification. The present paper shows that antibunched photons can also be obtained by two-photon absorption, as in the beams which have $g^{(2)} < 1$ in figures 4 and 5.

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