Quantum statistics of double-beam two-photon absorption

1975 J. Phys. A: Math. Gen. 8 1140

(http://iopscience.iop.org/0305-4470/8/7/018)

J. Phys. A: Math. Gen., Vol. 8, No. 7, 1975. Printed in Great Britain. © 1975

Quantum statistics of double-beam two-photon absorption

H D Simaan and R Loudon

Physics Department, Essex University, Colchester CO4 3SQ, UK

Received 17 February 1975

Abstract. The process of two-photon absorption from two beams of light is treated theoretically. Expressions are derived for the time dependences of the joint photon probability distribution of the beams, their photon-number factorial moments and the photon-number correlation between the beams. These expressions are illustrated by numerical results for special cases and their significance is discussed in physical terms. Simpler approximate expressions are also derived for the case where one beam is much more intense than the other.

1. Introduction

We consider in this paper the process of two-photon absorption by atoms in which the photons are taken one each from two different beams of light. In a previous paper (Simaan and Loudon 1975) we have treated the analogous process in which both photons are removed from the same beam of light, and have determined the time dependence of the photon probability distribution, its factorial moments and the degree of second-order coherence. The double-beam process also causes time-dependent changes in these properties but there is the additional feature in this case of correlations—or more accurately anticorrelations—between the two beams which develop as the absorption proceeds.

The equations which describe the double-beam two-photon absorption are introduced in § 2 and the initial short-time behaviours of the statistical properties are derived. It is assumed throughout the paper that the parts of the joint photon probability distribution which refer to the individual beams can be factorized at time $t = 0$ before any absorption takes place. Apart from this condition the initial distributions are allowed to have quite general forms. Factorization of the joint distribution for $t > 0$ is prevented by the formation of correlations between the beams, as is discussed in later sections.

Section 3 is devoted to an approximate solution which is valid when the initial mean number of photons in one of the beams is much larger than the initial mean photon number in the other beam. This corresponds to a common experimental arrangement in two-photon spectroscopy where one beam is obtained from a laser and the other from a conventional source.

The general solution of the photon rate equations is given in § 4. A generating function method is used similar to that employed by McNeil and Walls (1974) in their calculation of the first two factorial moments for two-photon absorption in which both beams initially have definite numbers of photons. Expressions are derived for the time dependences of the joint probability distribution, the photon-number correlation function and the factorial moments of the two beams. Attention is also given to the degree of second-order coherence.

One of the aims of the paper is to provide physical insight into the mechanisms by which two-photon absorption changes the correlation and fluctuation properties of the beams. The general results are accordingly evaluated for various simple kinds of initial light beams.

2. Photon rate equations and short-time solutions

2.1. Derivation of the rate equations

Consider two modes of the radiation field whose frequencies are such that pairs of photons, one from each mode, can be simultaneously absorbed by a gas of N atoms. It is assumed that the atoms have no transition frequencies to cause single-photon absorption or absorption of two photons from the same mode. Suppose that N_1 atoms are in the ground state while a smaller number N_2 are in the excited state of the transition, with

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

The numbers of atoms in the two states are assumed to be maintained constant by some external influence.

The numbers n and m of photons in the two beams are statistical quantities governed by a joint probability distribution $P_{n,m}$ which changes with time owing to the two-photon absorption. The probability per unit time that the beam photon numbers n and m are reduced to $n-1$ and $m-1$ can be written (Loudon 1973) as

$$N_1Jnm \tag{2}$$

where J is shorthand for an expression which contains atomic dipole matrix elements and energy eigenvalues. The corresponding probability per unit time of a two-photon emission, leading to an increase in the photon numbers from n and m to $n+1$ and $m+1$, is

$$N_2J(n+1)(m+1). \tag{3}$$

These two processes reduce $P_{n,m}$ at a combined rate

$$-N_1JnmP_{n,m}-N_2J(n+1)(m+1)P_{n,m}. \tag{4}$$

There are also two positive contributions to the rate of change of $P_{n,m}$. If $n-1$ and $m-1$ photons are present, with probability $P_{n-1,m-1}$, emission of two photons increases $P_{n,m}$ at a rate given by (3) with n and m replaced by $n-1$ and $m-1$,

$$N_2JnmP_{n-1,m-1}. \tag{5}$$

Similarly if $n+1$ and $m+1$ photons are present in the two beams, two-photon absorption increases $P_{n,m}$ at a rate determined by (2) with n and m replaced by $n+1$ and $m+1$,

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

The total rate of change of $P_{n,m}$ from (4), (5) and (6) is

$$\begin{aligned} \mathrm{d}P_{n,m}/\mathrm{d}t = & -N_1JnmP_{n,m}-N_2J(n+1)(m+1)P_{n,m}+N_2JnmP_{n-1,m-1} \\ & +N_1J(n+1)(m+1)P_{n+1,m+1}. \end{aligned} \tag{7}$$

The four contributions to the rate of change are entered on the photon energy level diagram in figure 1. For $N_2 = 0$, (7) reduces to an equation derived by McNeil and Walls (1974) using density operator techniques. The numbers of photons must of course be positive and the first and third terms in (7) should be removed if either $n = 0$ or $m = 0$ when the corresponding processes cannot occur.

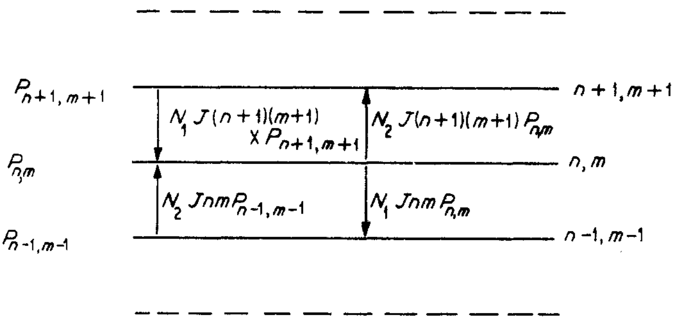

Figure 1. Energy level diagram for the photons. The level separation is equal to the sum of the photon energies of the two beams and the transition rates indicated are the contributions to $\mathrm{d}P_{n,m}/\mathrm{d}t$.

The probability distribution is assumed to be normalized,

$$\sum_{n,m} P_{n,m} = 1, \tag{8}$$

and it is seen by summation of (7) that a normalized distribution remains normalized as the two-photon absorption and emission proceed. The rth factorial moments of the two beams are defined by

$$m^{(r)} = \sum_{n,m} m(m-1)(m-2)\ldots(m-r+1)P_{n,m} \tag{9}$$

$$n^{(r)} = \sum_{n,m} n(n-1)(n-2)\ldots(n-r+1)P_{n,m}. \tag{10}$$

The mean numbers of photons in the two beams are thus denoted by $m^{(1)}$ and $n^{(1)}$ while their degrees of second-order coherence are defined to be

$$g_m^{(2)} = m^{(2)}/(m^{(1)})^2 \tag{11}$$

$$g_n^{(2)} = n^{(2)}/(n^{(1)})^2. \tag{12}$$

The photon-number correlation function of the two beams is

$$\overline{nm} = \sum_{n,m} nmP_{n,m}. \tag{13}$$

Equations for the rates of change of the factorial moments and the correlation function are obtained by differentiation of (9), (10) and (13) and by insertion of the rate of change of $P_{n,m}$ from (7). For example

$$\mathrm{d}m^{(1)}/\mathrm{d}t = -(N_1 - N_2)J\overline{nm} + N_2 J(m^{(1)} + n^{(1)} + 1) \tag{14}$$

$$\mathrm{d}m^{(2)}/\mathrm{d}t = -2(N_1 - N_2)J\sum_{n,m} nm^2 P_{n,m} + 2(N_1 + N_2)J\overline{nm} + 2N_2 J(m^{(2)} + 2m^{(1)}). \tag{15}$$

2.2. *Initial condition and short-time solutions*

Statistical independence of the two beams at the commencement of the two-photon absorption and emission implies that the initial probability distribution $P_{n,m}(0)$ can be written as a product of distributions for the individual modes

$$P_{n,m}(0) = Q_n(0)R_m(0). \tag{16}$$

In illustrating the general results the same types of initial beams will be used as in our previous paper (Simaan and Loudon 1975); the main properties of the simpler photon distributions are summarized in § 3 of this reference.

The derivation of the general time dependence of the photon statistical properties, given in § 4, is somewhat complicated. However, it is not difficult to evaluate the changes in these properties to first order in t. Consider the factorial moments. It is seen from (14) and (15) that, similar to the single-beam case, the rate of change of each moment depends on a moment or correlation of the next higher order and it is not possible to solve the equations straightforwardly. The time dependence correct to order t can however be obtained by substitution of the initial values of the various averages on the right-hand sides of (14) and (15), whence

$$m^{(1)} = m_0^{(1)} - Jt[N_1 n_0^{(1)} m_0^{(1)} - N_2(n_0^{(1)}+1)(m_0^{(1)}+1)] \tag{17}$$

$$m^{(2)} = m_0^{(2)} - 2Jt[N_1 n_0^{(1)} m_0^{(2)} - N_2(n_0^{(1)}+1)(m_0^{(2)}+2m_0^{(1)})] \tag{18}$$

where the zero subscripts denote the values of the moments at $t = 0$.

Substitution of (17) and (18) into (11) gives

$$g_m^{(2)} = g_{m0}^{(2)} + 2Jt(N_2/m_0^{(1)})(n_0^{(1)}+1)(2-g_{m0}^{(2)}). \tag{19}$$

The corresponding results for the other beam are obtained by a simple interchange of n and m, the basic equation (7) being symmetrical in these quantities.

The initial time dependence of the correlation function obtained in a similar way is

$$\begin{aligned}\overline{nm} = n_0^{(1)} m_0^{(1)} - Jt\{&N_1(n_0^{(2)} m_0^{(1)} + n_0^{(1)} m_0^{(2)} + n_0^{(1)} m_0^{(1)}) \\ &- N_2[n_0^{(2)}(m_0^{(1)}+1) + (n_0^{(1)}+1)m_0^{(2)} + 5n_0^{(1)} m_0^{(1)} + 3n_0^{(1)} + 3m_0^{(1)} + 1]\}.\end{aligned} \tag{20}$$

The initial values of the moments for the simpler photon distributions are summarized by Simaan and Loudon (1975).

These results show that the two-photon absorption associated with the N_1 ground state atoms causes a decrease in the factorial moments and the correlation function, while the emission associated with the N_2 excited atoms causes these quantities to increase. The degree of second-order coherence is not affected to order t by the two-photon absorptions; indeed it is not affected by the two-photon emissions either for a beam of initially chaotic statistics where $g_{m0}^{(2)}$ is equal to two. The time dependence is discussed in greater detail in subsequent sections when more complete solutions have been obtained.

3. Approximate solution

In this section we treat approximately the special case where one beam is initially much more intense than the other beam,

$$n_0^{(1)} \gg m_0^{(1)}. \tag{21}$$

It is clear that the ratio $n^{(1)}/m^{(1)}$ increases as the two-photon absorption proceeds. This case is an important one since it corresponds to the much used experimental arrangement in which one beam of fixed frequency is obtained from a laser while the other weaker beam is obtained by selection of a variable frequency from a conventional broad-band source (see for example Worlock 1972). The results could be obtained as special cases of the general solutions given in § 4. However, it is instructive to see the simple and direct way in which the strong-beam case can be treated.

It is assumed throughout the section that a negligible number of atoms is excited and we set $N_2 = 0$ and $N_1 = N$. We give greater attention to the weak beam since its properties are the more strikingly changed by the two-photon absorption.

3.1. *First moments*

The rate of change of $m^{(1)}$ given by (7) and (9) is

$$\mathrm{d}m^{(1)}/\mathrm{d}t = -NJ\sum_{n,m} nmP_{n,m}. \tag{22}$$

The summation generates the correlation function (13) whose value at a general time is as yet unknown. However, the value at $t = 0$ is very easily found because the photon distribution factorizes as in (16),

$$(\mathrm{d}m^{(1)}/\mathrm{d}t)_0 = -NJm_0^{(1)}n_0^{(1)}. \tag{23}$$

The higher time derivatives of $m^{(1)}$ can be found by successive differentiation of (22) and repeated use of (7). Thus

$$\mathrm{d}^2m^{(1)}/\mathrm{d}t^2 = (NJ)^2\sum_{n,m} nm(n+m-1)P_{n,m}. \tag{24}$$

The assumption that one beam is much more intense than the other is now used. In the parentheses of (24) the values of n and $m-1$ which make significant contributions to the summation are such that the latter can be neglected to a very good approximation, giving

$$(\mathrm{d}^2m^{(1)}/\mathrm{d}t^2)_0 = (NJ)^2m_0^{(1)}\sum_n n^2Q_n(0) \tag{25}$$

where (16) has been used. The approximation of retaining only the term of order n^r in the rth derivative leads to

$$(\mathrm{d}^rm^{(1)}/\mathrm{d}t^r)_0 = (-NJ)^rm_0^{(1)}\sum_n n^rQ_n(0). \tag{26}$$

Summation of the Taylor expansion of $m^{(1)}$ about $t = 0$ now gives

$$m^{(1)} = m_0^{(1)} + \sum_{r=1}^{\infty} (\mathrm{d}^rm^{(1)}/\mathrm{d}t^r)_0t^r/r! = m_0^{(1)}\sum_{n=0}^{\infty} \exp(-n\tau)Q_n(0) \tag{27}$$

where

$$\tau = NJt. \tag{28}$$

The time dependence of the first moment $n^{(1)}$ of the stronger beam is easily found. The time derivatives of $n^{(1)}$ and $m^{(1)}$ are indeed all equal and it follows that

$$n^{(1)} - m^{(1)} = n_0^{(1)} - m_0^{(1)}. \tag{29}$$

This equation is of the Manley–Rowe type; it expresses the equal rates of removal of photons from the two beams.

The above form of time dependence of $m^{(1)}$ shows that in the approximation used here the rate of absorption of the weaker beam depends on the statistical properties of the stronger beam but not on its own statistical properties. The summation in (27) can be performed without difficulty for the initial photon distributions given by Simaan and Loudon (1975) and we find

$$m^{(1)}/m_0^{(1)} = \begin{cases} \exp(-n_0^{(1)}\tau) & \text{(number)} \quad (30) \\ \exp\{n_0^{(1)}[\exp(-\tau)-1]\} & \text{(coherent)} \quad (31) \\ \{1+n_0^{(1)}-n_0^{(1)}\exp(-\tau)\}^{-1} & \text{(chaotic)} \quad (32) \\ 1-f+f\exp\{(n_0^{(1)}/f)[\exp(-\tau)-1]\} & \text{(pulsed)} \quad (33) \end{cases}$$

where (12), (15), (19) and (23) of this last reference have been used for the initial types of beam listed on the right.

Figure 2 shows graphs of these forms of time dependence of $m^{(1)}$. Note that the rather large steady-state values of $m^{(1)}$ for the pulsed stronger beams are a result of the absence of any absorption from those parts of the weaker beam which coincide with the dark sections of the pulsed beams.

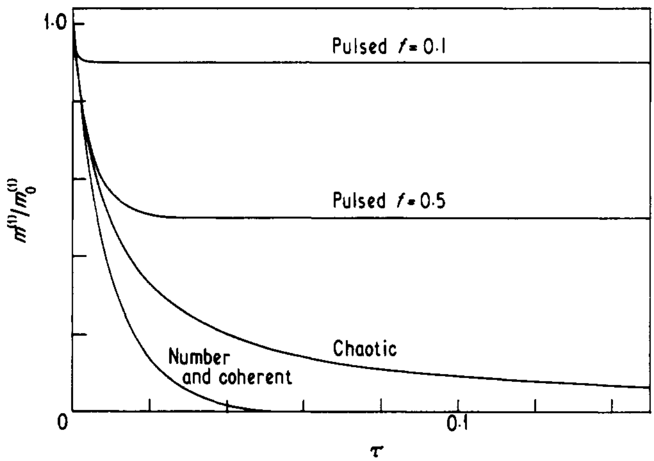

Figure 2. Time dependence of the mean photon numbers $m^{(1)}$ in the weaker beam for the initial types of stronger beam indicated. All the stronger beams have initial mean photon numbers $n_0^{(1)} = 100$. The pulsed beam result is shown for two different duty factors f.

Conventional treatments of two-photon absorption for the situation where one beam is much more intense than the other commonly solve (22) with the assumption that n can be set equal to $n_0^{(1)}$ and taken outside the summation. The resulting time dependence of $m^{(1)}$ is then identical to (30). However, the above analysis shows that this procedure is not generally valid; it holds when the stronger beam is a number state, and it is a good approximation for the coherent state, but it is a poor approximation for a chaotic stronger beam and it is completely wrong for the pulsed beam. The conventional treatment is in general only accurate to the term linear in t or τ where (17) shows that only the first moments of the beams are involved.

3.2. Second moment and second-order coherence

The second factorial moment of the weaker beam obtained from (7) and (9) by a similar approximation is

$$m^{(2)} = m_0^{(2)} \sum_{n=0}^{\infty} \exp(-2n\tau) Q_n(0). \tag{34}$$

It is seen that the summation in (34) is identical to that in the expression (27) for $m^{(1)}$ except that τ is replaced by 2τ. A corresponding replacement in the examples (30) to (33) for $m^{(1)}$ converts them into expressions for the time dependence of $m^{(2)}$ induced by various types of stronger beam.

The degree of second-order coherence of the weaker beam obtained from (11), (27) and (34) is given by

$$g_m^{(2)}/g_{m0}^{(2)} = \sum_n \exp(-2n\tau) Q_n(0) \left(\sum_n \exp(-n\tau) Q_n(0) \right)^{-2}. \tag{35}$$

In the limit of short times it is seen from (19) that $g_m^{(2)}$ has no term linear in the time for $N_2 = 0$ and this is verified by an expansion of (35) in powers of τ, where the first few terms give

$$g_m^{(2)}/g_{m0}^{(2)} = 1 + \tau^2 \sum_n (n - n_0^{(1)})^2 Q_n(0) - \tau^3 \sum_n (n - n_0^{(1)})^3 Q_n(0) + \dots . \tag{36}$$

The initial rate of increase of the degree of second-order coherence of the weaker beam is more rapid for strong beams whose mean square deviations in photon number are larger.

The summations in the numerator and denominator of (35) can be taken from (30) to (33) for the various kinds of initial strong beam and the resulting time dependences of $g_m^{(2)}$ are plotted in figure 3. For the strong number-state beam (35) gives

$$g_m^{(2)}/g_{m0}^{(2)} = 1 \tag{37}$$

but in general the degree of second-order coherence increases with time.

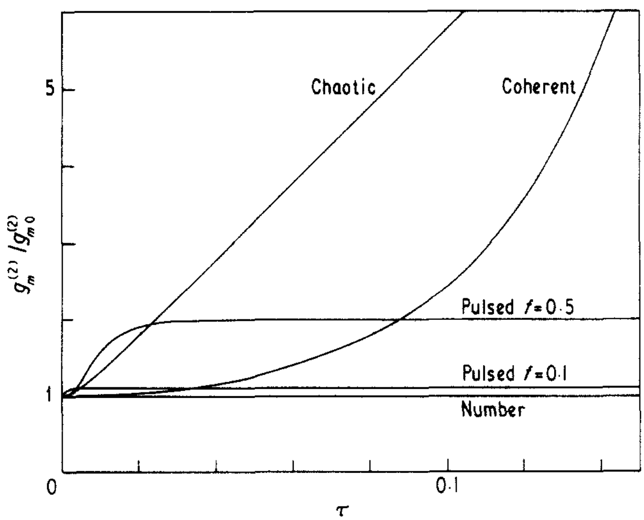

Figure 3. Time dependence of the degree of second-order coherence $g_m^{(2)}$ of the weaker beam for the same stronger beams as used in figure 2.

3.3. *Correlation function*

The Taylor expansion method can be applied finally to the correlation function $\overline{nm}$ defined in (13). The leading term in the rth derivative of $\overline{nm}$ with respect to time is of order n^{r+1} and the result obtained from these terms is

$$\overline{nm} = m_0^{(1)} \sum_{n=0}^{\infty} n \exp(-n\tau) Q_n(0). \tag{38}$$

It is not difficult to evaluate the summation for the various kinds of initial distribution in the intense beam, and the results are

$$\overline{nm}/n_0^{(1)}m_0^{(1)} = \begin{cases} \exp(-n_0^{(1)}\tau) & \text{(number)} \quad (39) \\ \exp\{n_0^{(1)}[\exp(-\tau)-1]-\tau\} & \text{(coherent)} \quad (40) \\ \exp(-\tau)\{1+n_0^{(1)}-n_0^{(1)}\exp(-\tau)\}^{-2} & \text{(chaotic)} \quad (41) \\ \exp\{(n_0^{(1)}/f)[\exp(-\tau)-1]-\tau\} & \text{(pulsed).} \quad (42) \end{cases}$$

Figure 4 shows graphs of the time dependence of $\overline{nm}/n^{(1)}m^{(1)}$ for these initial photon distributions, where the small changes in $n^{(1)}$ from the initial value have been ignored.

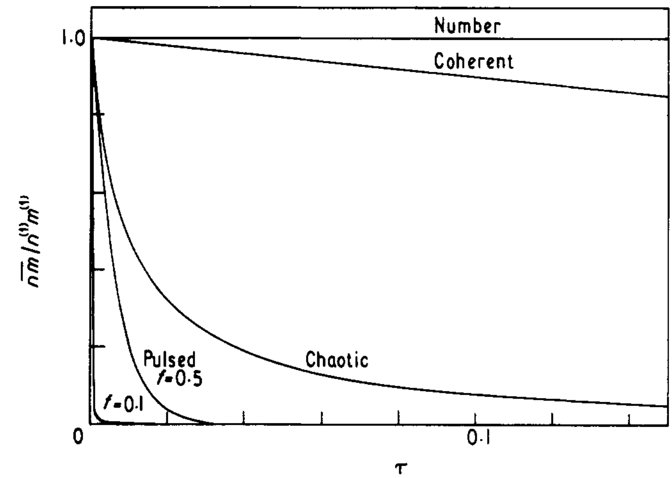

Figure 4. Time dependence of the normalized correlation function $\overline{nm}/n^{(1)}m^{(1)}$ for the same stronger beams as used in figures 2 and 3.

In all cases illustrated $\overline{nm}$ falls from its value $n_0^{(1)}m_0^{(1)}$ at $\tau = 0$, and at later times the inequality

$$\overline{nm} < n^{(1)}m^{(1)} \tag{43}$$

is satisfied, except for the number-state strong beam where the two quantities remain equal. There is thus a tendency for anticorrelations between the beams to develop as the two-photon absorption proceeds.

This aspect of the two-photon absorption can be understood by considering the simpler case of a classical description in which the strong beam is pulsed and the weak beam initially has constant intensity. The photon numbers n and m are replaced by classical beam intensities in this description. Figure 5 shows the time or space dependences of the intensities of short sections of the two beams before any absorption takes place and after a long period of absorption. Two-photon absorption can occur only during the pulses of the stronger beam and the absorption ceases when those sections of the weaker beam coincident with the pulses have been exhausted, leaving a fraction $1-f$ of the initial intensity of the weaker beam unabsorbed. The anticorrelation effect is clearly shown in figure 5; at the cessation of the two-photon absorption both beams have nonzero mean intensities but the intensity correlation function of the two beams is zero. At intermediate times the intensity correlation function satisfies an inequality analogous to (43). It is seen that the strong beam imposes a negative image of its intensity fluctuations on the weak beam in this classical analogue.

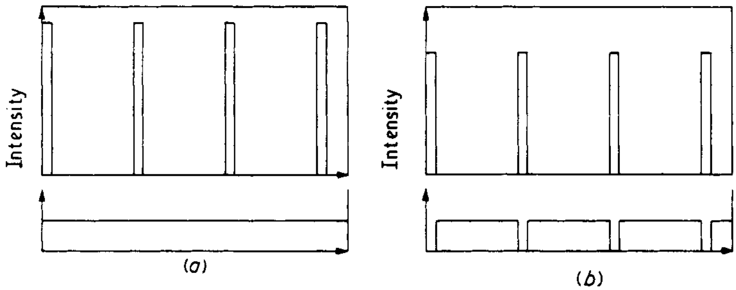

Figure 5. Beam intensities (*a*) before absorption and (*b*) after the cessation of absorption for a classical model in which the strong beam is pulsed with duty factor $f = 0{\cdot}1$ and the weak beam initially has constant intensity. The upper part of each diagram shows the strong beam and the lower part shows the weak beam. The horizontal axes can represent position for a view of the beams at a single time, or vice versa.

The differing behaviours of $\overline{nm}$ predicted by the quantum-mechanical calculations and illustrated in figure 4 arise for the same qualitative reasons. Thus the pulsed beams produce a rapid decrease in $\overline{nm}/n^{(1)}m^{(1)}$ because of the decay to zero of $\overline{nm}$ with only a modest decrease in $m^{(1)}$. A fast decrease in the correlation function also occurs for the chaotic strong beam while the much smaller photon-number fluctuations in the coherent beam lead to a slow development of anticorrelations in this case. The absence of fluctuations in the number-state beam prevents the formation of anticorrelations altogether.

The absorption rate (22) is proportional to the correlation function $\overline{nm}$, and the development of photon-number troughs in the weak beam coincident with the photon-number peaks in the strong beam inhibits the two-photon absorption. This effect can be seen by comparison of figure 4 with figure 2 where it is seen that the strong beams for which $\overline{nm}/n^{(1)}m^{(1)}$ decreases more rapidly are those for which $m^{(1)}$ decreases more slowly after the initial common rate of decay. Conventional treatments of double-beam two-photon absorption (see for example chapter 12 of Loudon 1973) omit beam correlation effects which begin to influence $m^{(1)}$ in order τ^2.

The time dependence of the degree of second-order coherence shown in figure 3 is also consistent with this description of the imposition on the weak beam of a negative image of the fluctuations in the strong beam. This is apparent in the τ^2 term in the short-time behaviour given by (36) and in the longer-time behaviours shown in the figure for the different types of strong beam. Thus the lack of fluctuations in the number-state strong beam leads to absorption of the weaker beam without any increase in its degree of second-order coherence. At the other extreme the large fluctuations in the strong chaotic and pulsed beams produce rapid initial increases in $g_m^{(2)}$, which are followed for the pulsed beams by saturation at a value close to $g_{m0}^{(2)}/(1-f)$. This is again in agreement with the behaviour shown in figure 5 where the initially coherent classical weak beam having $g_{m0}^{(2)} = 1$ is converted into a pulsed beam of duty factor $1-f$ and hence a degree of second-order coherence of $1/(1-f)$ at large times (see equation (5.109) of Loudon 1973).

It should be mentioned finally that the approximations made in the present section are such that the results agree with the short-time behaviours considered in § 2 in the case $N_2 = 0$, but only approximate to the exact steady-state results derived in § 4.

4. General solution

4.1. Separation of rate equations

The system of rate equations of which (7) is a representative only couples together series of elements of the joint probability distribution for which the difference between the two subscripts of P is constant. We define a new variable

$$\nu = n - m \tag{44}$$

whose value ranges from $-\infty$ to $+\infty$ in integer steps. Then the rate equations can be divided into subsets, each of which involves only those $P_{n,m}$ with the same value of ν. This division is illustrated schematically for small values of n and m by the oblique lines in figure 6.

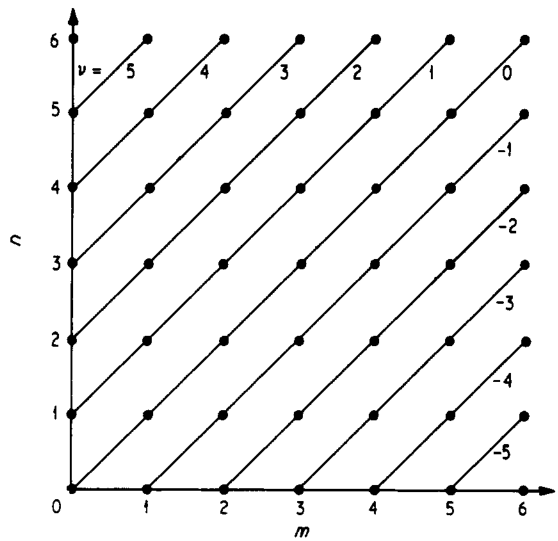

Figure 6. Schematic representation of the separation of the photon rate equations into subsets. Each blob represents a pair of values of n and m. Only those blobs connected by one of the oblique lines have $P_{n,m}$ which are coupled by the rate equations. The numbers attached to these lines are the values of $\nu = n-m$. The probability flows downwards towards the axes for increasing time when $N_2 < N_1$.

It is convenient in deriving the general solution to alter the notation by replacing one of the beam photon numbers as follows:

$$P_{n,m} = \begin{cases} P_{m+\nu,m} & \text{for } \nu \geqslant 0 \\ P_{n,n-\nu} & \text{for } \nu \leqslant 0. \end{cases} \tag{45}$$

The notation now shows explicitly the dependence on ν, and (7) takes the forms

$$\begin{aligned} \mathrm{d}P_{m+\nu,m}/\mathrm{d}t = & -N_1 J(m+\nu)mP_{m+\nu,m} - N_2 J(m+\nu+1)(m+1)P_{m+\nu,m} \\ & + N_2 J(m+\nu)mP_{m+\nu-1,m-1} + N_1 J(m+\nu+1)(m+1)P_{m+\nu+1,m+1} \end{aligned} \tag{46}$$

$$\begin{aligned} \mathrm{d}P_{n,n-\nu}/\mathrm{d}t = & -N_1 Jn(n-\nu)P_{n,n-\nu} - N_2 J(n+1)(n-\nu+1)P_{n,n-\nu} \\ & + N_2 Jn(n-\nu)P_{n-1,n-\nu-1} + N_1 J(n+1)(n-\nu+1)P_{n+1,n-\nu+1}. \end{aligned} \tag{47}$$

It is clear that for each value of ν the set of rate equations has only a single variable, n *or* m, instead of the pair of variables, n *and* m, which are apparently coupled in the original form (7). In the two-photon absorption of two beams both initially represented by number states, as treated by McNeil and Walls (1974), only those elements of the joint probability distribution lying on a single line in figure 6 need be considered.

The sums of those elements of the probability distribution which have the same ν are seen from (46) and (47) to be constants of the motion. Thus denoting these zeroth moments $m^{(0)}(\nu)$ and $n^{(0)}(\nu)$, we have

$$m^{(0)}(\nu) = \sum_{m=0}^{\infty} P_{m+\nu,m} = \sum_{m=0}^{\infty} P_{m+\nu,m}(0) = \sum_{m=0}^{\infty} Q_{m+\nu}(0)R_m(0) \qquad (\nu \geqslant 0) \tag{48}$$

$$n^{(0)}(\nu) = \sum_{n=0}^{\infty} P_{n,n-\nu} = \sum_{n=0}^{\infty} P_{n,n-\nu}(0) = \sum_{n=0}^{\infty} Q_n(0)R_{n-\nu}(0) \qquad (\nu \leqslant 0) \tag{49}$$

where (16) has been used. It follows that the average over the photon distribution of any function of ν is a constant of the motion which maintains its initial value. Application of this principle to the average of ν itself generates the Manley–Rowe relation (29) which is a general result, not limited to the case where one beam is much more intense than the other. Similarly the average of ν^2 generates a relation between the second moments and the correlation function,

$$n^{(2)} - 2\overline{nm} + m^{(2)} = n_0^{(2)} - 2n_0^{(1)}m_0^{(1)} + m_0^{(2)}. \tag{50}$$

4.2. *Steady-state limit*

After a sufficiently long period of time has elapsed, the photon system arrives at a steady state in which the rates of change (46) and (47) are zero. Solution of the resulting chains of simultaneous equations for the steady-state probabilities, denoted by $P_{m+\nu,m}(\infty)$ or $P_{n,n-\nu}(\infty)$, gives

$$P_{m+\nu,m}(\infty) = (N_2/N_1)^m P_{\nu,0}(\infty) \qquad (\nu \geqslant 0) \tag{51}$$

$$P_{n,n-\nu}(\infty) = (N_2/N_1)^n P_{0,-\nu}(\infty) \qquad (\nu \leqslant 0). \tag{52}$$

Hence

$$\sum_{m=0}^{\infty} P_{m+\nu,m}(\infty) = [1-(N_2/N_1)]^{-1} P_{\nu,0}(\infty) = m^{(0)}(\nu) \tag{53}$$

$$\sum_{n=0}^{\infty} P_{n,n-\nu}(\infty) = [1-(N_2/N_1)]^{-1} P_{0,-\nu}(\infty) = n^{(0)}(\nu). \tag{54}$$

Thus with the help of (48) and (49) the last four equations enable the steady-state photon distribution to be determined for any initial distributions $Q_n(0)$ and $R_m(0)$.

It is seen from (51) and (52) that the subset of probability elements corresponding to an oblique line in figure 6 has a chaotic type of distribution in the steady state (compare equation (10.17) of Loudon 1973). However, the distribution as a whole is not in general chaotic since the zeroth moments $m^{(0)}(\nu)$ and $n^{(0)}(\nu)$ for different ν are not appropriately related.

The results simplify when almost all the atoms are in their ground states and N_2 is negligibly small. In this case (51) and (52) give

$$P_{m+\nu,m}(\infty) = 0 \qquad \text{for } m \geqslant 1 \qquad (\nu \geqslant 0) \tag{55}$$

$$P_{n,n-\nu}(\infty) = 0 \qquad \text{for } n \geqslant 1 \qquad (\nu \leqslant 0) \tag{56}$$

and only those elements of the probability distribution which lie on the two axes of figure 6 can be nonzero. The steady-state values of the first two factorial moments of the two beams are then seen by inspection of figure 6 to be given by

$$m_{\infty}^{(1)} = -\sum_{\nu=-\infty}^{-1} \nu P_{0,-\nu}(\infty) \tag{57}$$

$$n_{\infty}^{(1)} = \sum_{\nu=1}^{\infty} \nu P_{\nu,0}(\infty) \tag{58}$$

$$m_{\infty}^{(2)} = \sum_{\nu=-\infty}^{-2} \nu(\nu+1) P_{0,-\nu}(\infty) \tag{59}$$

$$n_{\infty}^{(2)} = \sum_{\nu=2}^{\infty} \nu(\nu-1) P_{\nu,0}(\infty). \tag{60}$$

The correlation function must vanish in the steady state for $N_2 = 0$,

$$\overline{nm}_{\infty} = 0, \tag{61}$$

since by (55) and (56) the only nonzero elements of the distribution are those for which there are no photons in one of the beams.

4.3. Generating function method

McNeil and Walls (1974) have pointed out that the rate equations for the photon probability distributions can be solved by a generating function method in the special case $N_2 = 0$. We follow the same general method as these authors but obtain a more complete solution to the problem.

Consider the rate equation (46) for the elements $P_{n,m}$ which have $n \geqslant m$, that is $\nu \geqslant 0$. We define a generating function

$$F_{\nu}(y, \tau) = \sum_{m=0}^{\infty} y^m P_{m+\nu,m}(\tau) \tag{62}$$

which involves only those elements corresponding to a single oblique line in figure 6.

On setting $N_2 = 0$, multiplying both sides of (46) by y^m and summing over m, we obtain

$$\frac{\partial F_\nu}{\partial \tau} = y(1-y)\frac{\partial^2 F_\nu}{\partial y^2} + (1+\nu)(1-y)\frac{\partial F_\nu}{\partial y}. \tag{63}$$

The solution obtained by separation of the variables in the manner of McNeil and Walls is

$$F_\nu(y,\tau) = \sum_{k=0}^{\infty} A_k J_k(\nu, 1+\nu, y)\exp[-k(k+\nu)\tau] \tag{64}$$

where the Jacobi polynomial J_k defined for $\nu \geqslant 0$ is equal to the hypergeometric function ${}_2F_1(-k, k+\nu; 1+\nu; y)$ (Morse and Feshbach 1953).

The photon distribution at $\tau = 0$ is assumed known and the initial condition given by (62) and (64) is

$$F_\nu(y,0) = \sum_{m_0=0}^{\infty} y^{m_0} P_{m_0+\nu, m_0}(0) = \sum_{k=0}^{\infty} A_k J_k(\nu, 1+\nu, y) \tag{65}$$

where the running index for the initial distribution has been changed to m_0 to avoid confusion with m in later equations. The coefficients A_k can be determined by multiplication of the second and third parts of this relation by

$$y^\nu(1-y)^{-1}J_{k'}(\nu, 1+\nu, y)$$

followed by integration with respect to y over the range 0 to 1. Then with the help of an orthogonality relation satisfied by the Jacobi polynomials and a standard integral (see for example p 1755 of Morse and Feshbach 1953, p 398 of Erdélyi *et al* 1954, p 849 of Gradshteyn and Ryzhik 1965)

$$A_k = \frac{(-1)^k(2k+\nu)(k+\nu-1)!}{k!\,\nu!}\sum_{m_0=k}^{\infty}\frac{m_0!(m_0+\nu)!}{(m_0-k)!(m_0+k+\nu)!}P_{m_0+\nu, m_0}(0). \tag{66}$$

It is seen from (62) that the probability distribution is obtained from the generating function according to

$$P_{m+\nu, m}(\tau) = (m!)^{-1}(\partial^m F_\nu(y,\tau)/\partial y^m)_{y=0}. \tag{67}$$

Similarly the rth factorial moment of the distribution defined as in (9), but with the summation extending only over those elements which have the same value of ν, similar to the definition of the zeroth moment in (48), is given by

$$m^{(r)}(\nu) = (\partial^r F_\nu(y,\tau)/\partial y^r)_{y=1}. \tag{68}$$

The Jacobi polynomials have the properties

$$\left(\frac{\partial}{\partial y}\right)^l J_k(\nu, 1+\nu, y) = \begin{cases} \dfrac{(-1)^l k!(k+l+\nu-1)!\,\nu!}{(k-l)!(k+\nu-1)!(l+\nu)!}J_{k-l}(\nu+2l, 1+\nu+l, y) & \text{for } l \leqslant k \\ 0 & \text{for } l > k \end{cases} \tag{69}$$

and

$$J_{k-l}(\nu+2l, 1+\nu+l, 0) = 1 \tag{70}$$

$$J_{k-l}(\nu+2l, 1+\nu+l, 1) = (-1)^{k-1}(k-1)!(l+\nu)!/(l-1)!(k+\nu)! \qquad (l \geqslant 1). \tag{71}$$

It therefore follows from (64), (66), (67), (69) and (70) that

$$P_{m+\nu,m}(\tau) = \frac{(-1)^m}{m!(m+\nu)!}\sum_{k=m}^{\infty}\frac{(-1)^k(2k+\nu)(k+m+\nu-1)!}{(k-m)!}\exp[-k(k+\nu)\tau]$$
$$\times \sum_{m_0=k}^{\infty}\frac{m_0!(m_0+\nu)!}{(m_0-k)!(m_0+k+\nu)!}P_{m_0+\nu,m_0}(0) \tag{72}$$

and from (64), (66), (68), (69) and (71) that for $r \geqslant 1$

$$m^{(r)}(\nu) = \frac{1}{(r-1)!}\sum_{k=r}^{\infty}\frac{(2k+\nu)(k+r+\nu-1)!(k-1)!}{(k-r)!(k+\nu)!}\exp[-k(k+\nu)\tau]$$
$$\times \sum_{m_0=k}^{\infty}\frac{m_0!(m_0+\nu)!}{(m_0-k)!(m_0+k+\nu)!}P_{m_0+\nu,m_0}(0) \qquad (\nu \geqslant 0). \tag{73}$$

All the above analysis has been concerned with the rate equation (46). The corresponding equation (47) for the elements of the probability distribution which have $n \leqslant m$ can be solved by the same technique and the results are

$$P_{n,n-\nu}(\tau) = \frac{(-1)^n}{n!(n-\nu)!}\sum_{k=n}^{\infty}\frac{(-1)^k(2k-\nu)(k+n-\nu-1)!}{(k-n)!}\exp[-k(k-\nu)\tau]$$
$$\times \sum_{n_0=k}^{\infty}\frac{n_0!(n_0-\nu)!}{(n_0-k)!(n_0+k-\nu)!}P_{n_0,n_0-\nu}(0) \tag{74}$$

$$n^{(r)}(\nu) = \frac{1}{(r-1)!}\sum_{k=r}^{\infty}\frac{(2k-\nu)(k+r-\nu-1)!(k-1)!}{(k-r)!(k-\nu)!}\exp[-k(k-\nu)\tau]$$
$$\times \sum_{n_0=k}^{\infty}\frac{n_0!(n_0-\nu)!}{(n_0-k)!(n_0+k-\nu)!}P_{n_0,n_0-\nu}(0) \qquad (\nu \leqslant 0). \tag{75}$$

The latter quantity is the rth factorial moment (valid for $r \geqslant 1$) defined as in (10) but with the sum restricted to elements which have the same negative value of ν.

Equations (72) and (74) enable computation of the complete joint probability distribution for arbitrary time τ and arbitrary initial distribution. Figure 7 shows some results for a simple example in which both beams are initially number states with ten photons in each so that only the $\nu = 0$ elements can have values different from zero. The progressively slower decay of elements corresponding to smaller photon numbers is apparent, as is the approach at the right-hand side of the figure to the steady-state condition in which all the initial photons have been absorbed.

In the steady-state limit where τ tends to infinity all the terms on the right-hand sides of (72) and (74) are zero except those with $k = 0$. The only nonzero probability elements are therefore

$$P_{\nu,0}(\infty) = \sum_{m_0=0}^{\infty} P_{m_0+\nu,m_0}(0) = m^{(0)}(\nu) \tag{76}$$

$$P_{0,-\nu}(\infty) = \sum_{n_0=0}^{\infty} P_{n_0,n_0-\nu}(0) = n^{(0)}(\nu) \tag{77}$$

in agreement with (53) to (56).

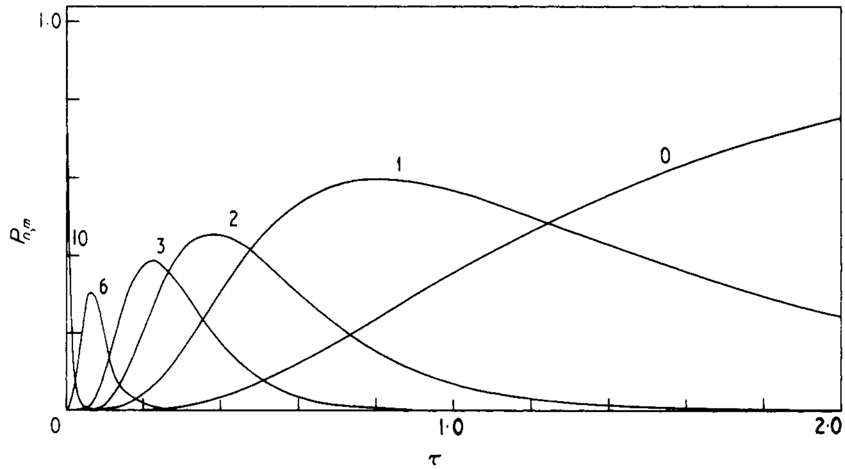

Figure 7. Time dependence of selected elements of the photon probability distribution. The initial distribution is $P_{10,10}(0) = 1$ and only diagonal elements of the distribution have nonzero values at subsequent times. The numbers attached to the curves indicate the common values of n and m.

4.4. *First moments*

The moments of the distribution are often of more immediate physical interest than the distribution itself, and these can be obtained from the general results (73) and (75) for the factorial moments taken over the part of the distribution having a given value of v. The summation in the definition (9) of the first moment $m^{(1)}$ can be rearranged with the help of (44) and (45) to give

$$m^{(1)} = \sum_{n,m} mP_{n,m} = \sum_{v=0}^{\infty}\sum_{m=0}^{\infty} mP_{m+v,m} + \sum_{v=-\infty}^{-1}\sum_{n=0}^{\infty}(n-v)P_{n,n-v}$$

$$= \sum_{v=0}^{\infty} m^{(1)}(v) + \sum_{v=-\infty}^{-1}(n^{(1)}(v) - vn^{(0)}(v)) \tag{78}$$

where (49) has been used. The rearrangement of the summation can readily be visualized by reference to figure 6. A similar result can be derived for the first moment $n^{(1)}$ of the other beam but the Manley–Rowe relation (29) makes independent numerical calculations of $n^{(1)}$ unnecessary. We note that the expression for $m^{(1)}(v)$ obtained from (73) for the case of an initial number-state distribution agrees with (5.23) of McNeil and Walls (1974).

Figure 8 shows the results of some calculations of the time dependence of the first moments for initial states in which beam n is a number state while beam m has various kinds of distribution. Both beams have initial first moments equal to ten, and $m^{(1)}$ and $n^{(1)}$ therefore remain equal as the two-photon absorption proceeds. Their steady-state values are zero for the case where both beams are initially number states and only $v = 0$ probability elements occur, but the steady-state values differ from zero for the other cases and they can be obtained by numerical evaluation of (57) and (58).

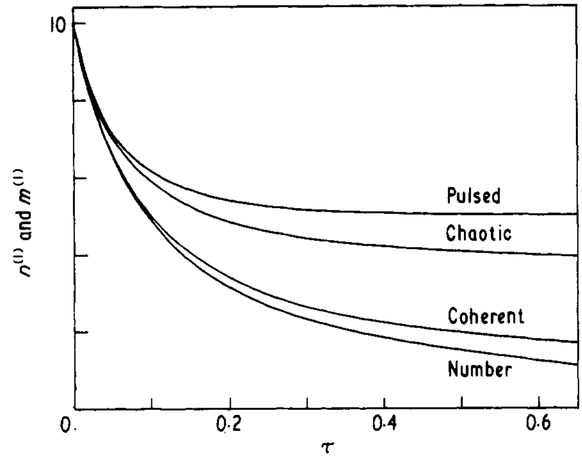

Figure 8. Time dependence of the mean photon numbers $m^{(1)}$ and $n^{(1)}$ for initial distributions in which beam n is a number state with ten photons and beam m has the character indicated on the curves. The initial mean photon number $m_0^{(1)}$ is equal to ten for all four types of beam and the duty factor in the pulsed case is 0·5.

4.5. Correlation function

The correlation function defined in (13) can be obtained from (73) and (75) by a rearrangement of the summation similar to that used in (78), and we find

$$\overline{nm} = \sum_{v=0}^{\infty} [m^{(2)}(v) + (1+v)m^{(1)}(v)] + \sum_{v=-\infty}^{-1} [n^{(2)}(v) + (1-v)n^{(1)}(v)]. \qquad (79)$$

Figure 9 shows the results of calculations of the time dependence of the correlation function for the same initial distributions as used in figure 8.

The qualitative aspects of figures 8 and 9 can be accounted for by explanations very similar to those given in § 3.3 for the behaviour of the weaker beam in the presence of another beam which is much more intense. Thus the anticorrelations between the beams occur for the same reasons as before and lead to a zero correlation function in the steady state even though the mean numbers of photons in the two beams do not vanish, in accordance with (57), (58) and (61). This behaviour is shown by three of the examples plotted in figure 9. The fourth example, that of two initial number-state beams with equal numbers of photons, is an exception in that $n^{(1)}$, $m^{(1)}$ and $\overline{nm}$ all tend to zero at long times with $\exp(-\tau)$ time dependences, leading to an exponential *divergence* of the normalized correlation function.

The behaviours of $n^{(1)}$, $m^{(1)}$ and $\overline{nm}$ are closely related for all types of initial beams, the rates of change of the first two quantities being proportional to the third as in (22). Thus, similar to the intense beam case, the curves for the four examples appear in reverse order in figures 8 and 9 since a larger value of $\overline{nm}$ corresponds to a more rapid decrease of $n^{(1)}$ and $m^{(1)}$.

4.6. Second moments

The second factorial moments of the two beams and their correlation function are

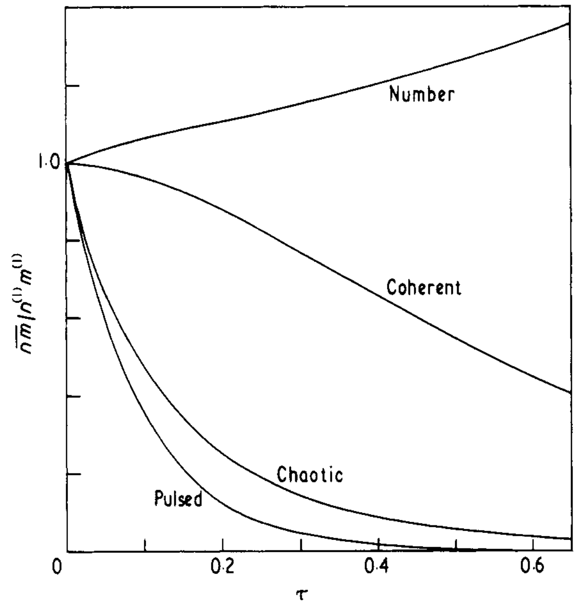

Figure 9. Time dependence of the normalized correlation function $\overline{nm}/n^{(1)}m^{(1)}$ for the same initial distributions as figure 8.

related by (50), so that in addition to (79) it is only necessary to make numerical calculations of one of the factorial moments. A rearrangement of the summation in (9) for $r = 2$ similar to that made in (78) gives

$$m^{(2)} = \sum_{v=0}^{\infty} m^{(2)}(v) + \sum_{v=-\infty}^{-1} [n^{(2)}(v) - 2vn^{(1)}(v) + v(v+1)n^{(0)}(v)]. \tag{80}$$

The degrees of second-order coherence of the two beams, given in terms of the first two factorial moments by (11) and (12), are plotted in figures 10 and 11 for the same examples of initial beams as used in the two previous figures.

The time dependences shown in figures 10 and 11 also have some similarities with those found in the intense beam case. Thus the initial number state shown in figure 11 acquires fluctuations as a result of its absorption, which occurs preferentially at the peaks of the other beam, and the order of the curves in figure 11 is the same as that at the short-time end of figure 3. The case where both beams are initially number states is again exceptional in that the first two factorial moments of both beams tend to zero in the steady state; a more careful inspection shows that the numerators of (11) and (12) fall off more rapidly than the denominators to give an $\exp(-2\tau)$ dependence of the degree of second-order coherence at large τ. The degrees of second-order coherence increase with time in all the other examples shown and their steady-state limits can be determined by evaluation of (57) to (60). Thus for example, the beam which initially has a chaotic distribution arrives at the steady state with a second-order coherence close to 5·2, and the initially pulsed beam remains pulsed with a degree of second-order coherence in the steady state a little larger than two while that of its partner increases to become a little smaller than two.

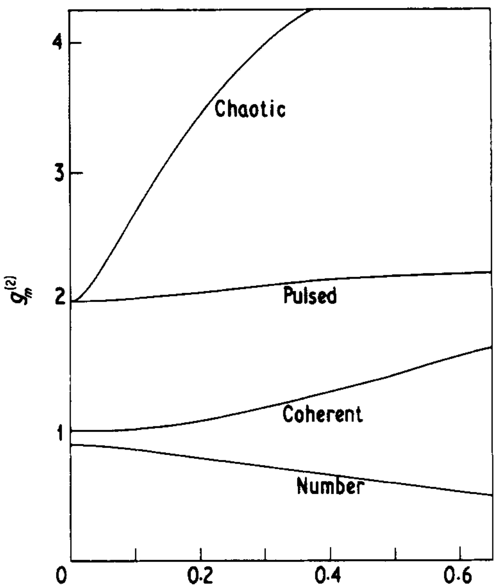

Figure 10. Time dependence of the degree of second-order coherence $g_m^{(2)}$ for the same initial distributions as figure 8.

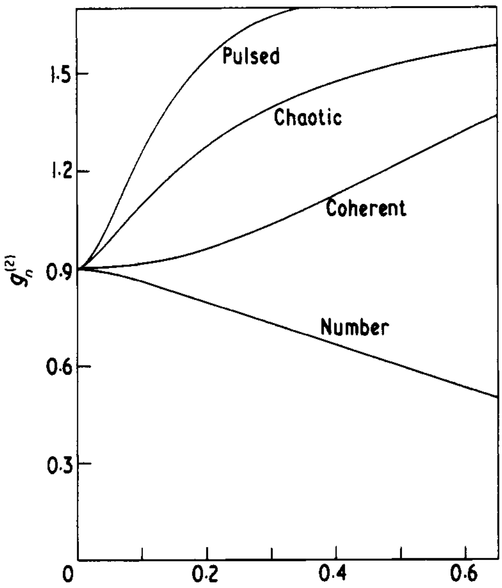

Figure 11. Time dependence of the degree of second-order coherence $g_n^{(2)}$ for the same initial distributions as figure 8.

4.7. Intense beam limit

We conclude the section by relating the general solutions to the approximate solutions found in § 3 for the case where beam n is much more intense than beam m. In this limit only those terms in the photon probability distribution with $\nu \gg 1$ have significant magnitude. The exponential factors in (72) and (73) then fall off extremely rapidly with increasing τ and it is a permissible approximation to retain only that term in the summation which falls off least rapidly. The approximate form of the probability distribution (72) obtained by retaining only the $k = m$ term on the right-hand side is

$$P_{m+\nu,m}(\tau) \simeq \frac{(2m+\nu)!}{m!(m+\nu)!}\exp[-m(m+\nu)\tau]\sum_{m_0=m}^{\infty}\frac{m_0!(m_0+\nu)!}{(m_0-m)!(m_0+m+\nu)!}P_{m_0+\nu,m_0}(0)$$

$$\simeq (m!)^{-1}\exp(-m\nu\tau)\sum_{m_0=m}^{\infty}[m_0!/(m_0-m)!]P_{m_0+\nu,m_0}(0). \tag{81}$$

The moments can be approximated in a similar way. Thus from (73)

$$m^{(1)}(\nu) \simeq \exp(-\nu\tau)\sum_{m_0=1}^{\infty} m_0 P_{m_0+\nu,m_0}(0) \tag{82}$$

where only the $k = 1$ term has been retained in the first summation on the right of (73). Only the first term on the right of (78) contributes for $\nu \gg 1$, and the approximate expression for the first moment of the complete beam is accordingly

$$m^{(1)} \simeq \sum_{\nu=0}^{\infty}\exp(-\nu\tau)\sum_{m_0=1}^{\infty} m_0 Q_{m_0+\nu}(0)R_{m_0}(0) \simeq m_0^{(1)}\sum_{\nu=0}^{\infty}\exp(-\nu\tau)Q_\nu(0) \tag{83}$$

where (16) has been used, and the second step is valid only for the large-ν terms in the summation. We have thus arrived by a more difficult and less direct route at the same expression (27) as previously derived for the intense beam case. The other approximate results of § 3 can be rederived in a similar manner by making use of large-ν approximations to (79) and (80).

5. Discussion

The various statistical properties of the double-beam two-photon absorption process derived in the paper have been extensively illustrated and discussed in the earlier sections, and it remains only to summarize the main conclusions.

The rate of two-photon absorption of each beam depends upon the statistical properties of the other beam via the correlation function $\overline{nm}$. The correlation function is equal to the product of the mean photon numbers $n^{(1)}m^{(1)}$ of the individual beams when the beams are statistically independent, as assumed at the beginning of the two-photon absorption. However, the absorption process itself generates anticorrelations between the beams as the enhanced absorption associated with peaks in the photon-number fluctuations of one beam cuts out troughs in the time-dependent fluctuations of the other beam. The result is a reduction of $\overline{nm}$ below the value of $n^{(1)}m^{(1)}$ and a fall-off in the rate of absorption, the effect being more marked for beams with larger fluctuations in photon number. Another aspect of the interaction between the beams is the transfer of fluctuations from one beam to the other, leading to a tendency for their degrees of second-order coherence to increase with time. This contrasts with the opposite tendency in single-beam two-photon absorption (Simaan and Loudon 1975) where the beam fluctuations are smoothed by the absorption process. It should also be noted that there are exceptions to the generality of the above remarks, as in the case of two initial number-state beams, discussed in detail in § 4.

The conventional theory of double-beam two-photon absorption (see for example chapter 12 of Loudon 1973) ignores the development of anticorrelations between the beams and thus gives results which are generally correct only as far as the terms linear in the time. It is necessary to use the exact results given here if the terms of higher order in the time are required. The examples treated in the paper have used rather simple special cases of initial photon distributions in the two beams in order to illustrate the physical processes which control the rate of absorption. However, the results presented in § 4 enable the time-dependent statistical properties of the light to be evaluated for any initial photon probability distributions in the two beams.

Acknowledgment

H D Simaan thanks the Iraqi Government for financial support.

References

Erdélyi A, Magnus W, Oberhettinger F and Tricomi F G 1954 *Tables of Integral Transforms* vol 2 (New York: McGraw-Hill)
Gradshteyn I S and Ryzhik I M 1965 *Tables of Integrals, Series and Products* (New York: Academic Press)
Loudon R 1973 *The Quantum Theory of Light* (Oxford: Clarendon)
McNeil K J and Walls D F 1974 *J. Phys. A: Math., Nucl. Gen.* **7** 617–31
Morse P M and Feshbach H 1953 *Methods of Theoretical Physics* (New York: McGraw-Hill)
Simaan H D and Loudon R 1975 *J. Phys. A: Math. Gen.* **8** 539–54
Worlock J M 1972 *Laser Handbook* eds E O Schulz-Dubois and F T Arecchi (Amsterdam: North-Holland)