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Quantum statistics of the stimulated Raman effect

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Abstract. The nonlinear process of the stimulated Raman effect is treated theoretically. Simple expressions are derived for the initial rates of change and limiting steady state values of the first two moments, the degrees of second-order coherence and the correlation function of the pump and Stokes beams. An analytical solution for the time dependence of the joint photon probability distribution of the two light beams is obtained and then employed to calculate some numerical results for various special cases of interest, and the significance of these results is discussed in physical terms. Simpler approximate expressions are also derived for the case where the pump beam is much more intense than the Stokes beam.

1. Introduction

It has been known for some time (Shen 1967) that the nonlinear interaction between light and matter causes statistical changes in the properties of both, and therefore a complete description of any nonlinear effect requires the application of quantum statistics. The purpose of this paper is to consider in some detail the nonlinear process of the stimulated Raman effect by atoms from the quantum statistical point of view, and to answer the question of whether and how the statistical nature of the pump and Stokes fields is disturbed by such an effect. In a recent paper (Simaan and Loudon 1975b) the quantum statistical problem of the process of double-beam two-photon absorption was treated, and the time dependences of the joint photon probability distribution, its factorial moments, the degrees of second-order coherence and the correlation function of the two beams were determined. The stimulated Raman effect, as we shall see in this paper, also induces time-dependent changes in these properties.

The equations which describe the rate of change of the photon statistical joint distribution caused by the stimulated Raman effect and its inverse are presented in § 2, and equations for the rates of change of the moments and the correlation function are derived. These results are used in § 3 to calculate the initial short-time behaviours of the statistical properties of the pump and Stokes beams. The opposite extreme of the steady state achieved after a long period of time is treated in § 4. The short-time and steady state solutions give physical insight into the mechanisms by which the changes in the statistical properties of both the beams are brought about.

The general solution of the photon rate equations for the case where almost all the atoms are in their ground states is given in § 5. A Laplace transform method is used, similar to that employed by McNeil and Walls (1974) in their calculation of the photon probability distribution for the Stokes field alone in which both the pump and Stokes beams initially have definite numbers of photons. A more complete solution is derived for the time dependence of the joint photon probability distribution which enables the statistical properties of both beams to be calculated for any type of initial distribution. These results are compared with those of McNeil and Walls (1974) and some discrepancies are found.

Section 6 is devoted to an approximate solution which is valid when the initial mean number of photons in the pump beam is much larger than the initial mean photon number in the Stokes beam. In this section we shall assume that the depletion of power in the pump field is negligibly small, and hence the changes in its statistical properties can also be neglected. Within this limit the exact solution introduced in § 5 is approximated, and simple expressions for the probability distribution of the Stokes field and its first two moments are derived. These expressions are evaluated for various types of initial distribution and it is proved analytically that the growing Stokes field, in addition to the amplified version of its initial type, consists in general of a chaotic component which is generated by the amplification process. In § 7 all the statistical quantities obtained throughout the paper and in particular the first moments, the degrees of second-order coherence and the correlation function are discussed in physical terms and illustrated by some numerical results in the form of graphs for some special cases of interest.

2. Photon rate equations

Consider a single mode of a pump field whose frequency allows a photon to be absorbed by a gas of N two-level atoms and then scattered into another single mode of Stokes radiation field. It is assumed that the atoms only have transitions of the required frequency for the Stokes component of the stimulated Raman effect where its anti-Stokes component is ignored. The conditions needed for any other processes to occur are taken to be badly satisfied. Suppose that N_1 atoms are in the ground state and a smaller number N_2 in the excited state of the Raman transitions, with

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

The numbers of atoms in the two states are assumed to be kept constant by some external influence. The numbers n and m of photons respectively present in the pump and Stokes beams at time t are statistical quantities and governed by a joint probability distribution $P_{n,m}(t)$ which changes with time owing to the process of Raman scattering. At time $t = 0$, normally the two beams are considered to be statistically independent and therefore $P_{n,m}(0)$ can be written as a product of the photon distribution for the separate beams,

$$P_{n,m}(0) = Q_n(0)R_m(0). \tag{2}$$

The time-dependent changes in the $P_{n,m}$ are described by rate equations which shall be readily derived.

The probability per unit time that the Raman effect takes place with a change in the photon numbers from n and m to $n-1$ and $m+1$ can be written (Loudon 1973) as

$$N_1 Jn(m+1) \tag{3}$$

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

$$N_2 J(n+1)m. \tag{4}$$

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

$$-N_1 Jn(m+1)P_{n,m} - N_2 J(n+1)mP_{n,m}. \tag{5}$$

There are also two positive contributions to the rate of change of $P_{n,m}$. If $n-1$ and $m+1$ photons are present in the two beams, with probability $P_{n-1,m+1}$, the inverse Raman effect increases $P_{n,m}$ at a rate determined by (4) with n and m replaced by $n-1$ and $m+1$:

$$N_2 Jn(m+1)P_{n-1,m+1}. \tag{6}$$

Similarly if $n+1$ and $m-1$ photons are present, with probability $P_{n+1,m-1}$, the Raman effect increases $P_{n,m}$ at a rate given by (3) with n and m replaced by $n+1$ and $m-1$:

$$N_1 J(n+1)mP_{n+1,m-1}. \tag{7}$$

The total rate of change of $P_{n,m}$ from (5), (6) and (7) is

$$\begin{aligned} dP_{n,m}/dt = & -N_1 Jn(m+1)P_{n,m} - N_2 J(n+1)mP_{n,m} + N_2 Jn(m+1)P_{n-1,m+1} \\ & + N_1 J(n+1)mP_{n+1,m-1}. \end{aligned} \tag{8}$$

The four contributions to the rate of change are illustrated by the photon energy level diagram in figure 1. An equation identical to (8) can be derived by density operator

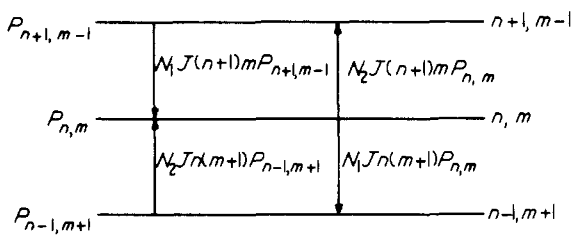

Figure 1. Energy level diagram for the photons. The level separation is equal to the difference of the photon energies of the pump and Stokes beams, and the transition rates indicated are the contributions to $dP_{n,m}/dt$.

techniques, using an explicit form for the photon–atom Hamiltonian (Shen 1967, McNeil and Walls 1974). For $N_2 = 0$, (8) reduces to an equation derived by Loudon (1973), using a method similar to that used above. The numbers of photons must of course be positive and the first and third terms in (8) should be removed for $n = 0$, when the processes described by these terms cannot occur. Similarly if $m = 0$, the second and fourth terms should be removed.

The probability distribution is assumed to be normalized:

$$\sum_{n,m} P_{n,m} = 1, \tag{9}$$

and it is seen by summation of (8) that a normalized distribution remains normalized as the Raman effect and its inverse proceed. The rth moments of the two beams are defined by

$$\overline{n^r} = \sum_{n,m} n^r P_{n,m} \tag{10}$$

$$\overline{m^r} = \sum_{n,m} m^r P_{n,m} \tag{11}$$

while their degrees of second-order coherence are defined to be

$$g_n^{(2)} = (\overline{n^2} - \bar{n})/\bar{n}^2 \tag{12}$$

$$g_m^{(2)} = (\overline{m^2} - \bar{m})/\bar{m}^2. \tag{13}$$

Equations for the rates of change of the moments are obtained by time differentiation of (10) and (11) and insertion of the rate of change of $P_{n,m}$ from (8). For example,

$$\mathrm{d}\bar{n}/\mathrm{d}t = J \sum_{n,m} [-N_1 n(m+1) + N_2(n+1)m] P_{n,m} \tag{14}$$

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

In a similar way the rate of change of the correlation function

$$\overline{nm} = \sum_{n,m} nm P_{n,m} \tag{16}$$

can be written as

$$\mathrm{d}\overline{nm}/\mathrm{d}t = J \sum_{n,m} [N_1 n(m+1)(n-m-1) + N_2(n+1)m(m-n-1)] P_{n,m}. \tag{17}$$

The system of rate equations of which (8) is a representative only couples those elements $P_{n,m}$ for which

$$\nu = n+m = \text{constant} \tag{18}$$

where ν is a new variable and ranges from 0 to ∞ in integer steps. With the help of (18), it is convenient in deriving the steady state solutions, as we shall see in § 4, to write (8) as follows:

$$\begin{aligned} \mathrm{d}P_{n,\nu-n}/\mathrm{d}t = {} & -N_1 J n(\nu-n+1) P_{n,\nu-n} - N_2 J(n+1)(\nu-n) P_{n,\nu-n} \\ & + N_2 J n(\nu-n+1) P_{n-1,\nu-n+1} + N_1 J(n+1)(\nu-n) P_{n+1,\nu-n-1}. \end{aligned} \tag{19}$$

It is clear that for a given ν, this subset of equations is generated by taking the series of values of n which ranges from 0 to ν, rather than a pair of the variables n and m. The same procedure used in deriving (19) can be used to express the rate equations in terms of ν and m, but there is no difference in the final results obtained. A full account of the method of dividing the set of rate equations is given by Simaan and Loudon (1975b) for the double-beam two-photon absorption process, although this case is more complicated in that ν ranges from $-\infty$ to ∞, instead of the range 0 to ∞ for the stimulated Raman effect.

The sum of those elements of the probability distribution which have the same ν is seen from (19) to be a constant of the motion,

$$\sum_{n=0}^{\nu} P_{n,\nu-n} = \sum_{n=0}^{\nu} P_{n,\nu-n}(0) = \sum_{n=0}^{\nu} Q_n(0) R_{\nu-n}(0) \tag{20}$$

where (2) has been used. It follows that the average over the photon probability distribution of any function of ν is a constant of the motion which maintains its initial value.

For example the average of v itself generates the Manley–Rowe relation

$$\bar{n}+\bar{m} = \bar{n}_0+\bar{m}_0 \tag{21}$$

and similarly the average of v^2 generates a relation between the second moments and the correlation function of the two beams

$$\overline{n^2}+2\overline{nm}+\overline{m^2} = \overline{n_0^2}+2\bar{n}_0\bar{m}_0+\overline{m_0^2} \tag{22}$$

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

3. Short-time solutions

The fact that the rate of change of each moment, as is seen from (14) and (15), depends on a moment or correlation of the next higher order, renders the equations insoluble by simple techniques. Knowing however that (2) is valid at the commencement of the Raman transitions, the time dependences of the moments correct to order t may be obtained by substitution of the initial values of the various averages on the right-hand sides of (14) and (15), whence

$$\bar{n} = \bar{n}_0 - Jt[N_1\bar{n}_0(\bar{m}_0+1) - N_2(\bar{n}_0+1)\bar{m}_0] \tag{23}$$

$$\overline{n^2} = \overline{n_0^2} - Jt[N_1(2\overline{n_0^2}-\bar{n}_0)(\bar{m}_0+1) - N_2(2\overline{n_0^2}+3\bar{n}_0+1)\bar{m}_0]. \tag{24}$$

The corresponding expansion of $g_n^{(2)}$ can be obtained from these results and (12) with some algebra:

$$g_n^{(2)} = g_{n0}^{(2)} + 2N_2Jt(\bar{m}_0/\bar{n}_0)(2-g_{n0}^{(2)}). \tag{25}$$

The initial time dependence of the correlation function obtained from (17) in a similar way to that used in deriving (23) and (24) above is

$$\overline{nm} = \bar{n}_0\bar{m}_0 + Jt\{N_1[\overline{n_0^2}(\bar{m}_0+1) - \bar{n}_0(\overline{m_0^2}+2\bar{m}_0+1)] + N_2[\overline{m_0^2}(\bar{n}_0+1) - \bar{m}_0(\overline{n_0^2}+2\bar{n}_0+1)]\}. \tag{26}$$

If this equation together with (23) and (24) is employed to determine similar expressions for $\bar{m}$ and $\overline{m^2}$ respectively from (21) and (22), the expansion of $g_m^{(2)}$ correct to order t accordingly can be calculated from (13). For various initial light beams the linear time dependences of all the functions considered above can be obtained straightforwardly by substitution of the appropriate expressions for $\overline{n_0^2}$, $g_{n0}^{(2)}$ and $\overline{m_0^2}$ which can be found for the simpler photon distributions in § 3 of Simaan and Loudon (1975a).

The results obtained so far show that the Raman effect associated with the N_1 ground state atoms decreases the moments of the pump beam and increases those of the Stokes beam, while the inverse Raman effect associated with the N_2 excited state atoms causes the moments of the pump and Stokes beams respectively to increase and decrease. The short-time behaviour of the correlation function is more complicated and its variation in general is governed by the initial values of the moments. For example if we assume that $\bar{m}_0 = \overline{m_0^2} = 0$, the correlation function shows an initial increase by the Raman effect and no change by its inverse. The degree of second-order coherence $g_n^{(2)}$ on the other hand is not affected to order t by the Raman effect and it is not affected by its inverse either for a beam of initially chaotic distribution where $g_{n0}^{(2)} = 2$. Furthermore it is seen from (25) that $g_n^{(2)}$ tends to a chaotic value of 2 whether $g_{n0}^{(2)}$ is greater or less than 2.

Assuming that almost all the atoms are in their ground states, we can take $N_2 = 0$ and $N_1 = N$ for the remainder of the present section and define a new time variable

$$\tau = NJt. \tag{27}$$

At this stage it is instructive to extend our previous calculations and add an extra term or terms, in second order in t or even higher, to the expansions of the various functions. Further time differentiations of (14) and (15) and use of (8) respectively give expressions for the second derivatives of $\bar{n}$ and $\overline{n^2}$. With the help of (2), the magnitudes of these derivatives at $t = 0$ can be obtained and are the coefficients of t^2 in power series expansions, and therefore correct to second order in t, (23) and (24) can be written as

$$\bar{n} = \bar{n}_0 - \tau\bar{n}_0(\bar{m}_0+1) - \tfrac{1}{2}\tau^2[\overline{n_0^2}(\bar{m}_0+1) - \bar{n}_0(\overline{m_0^2}+3\bar{m}_0+2)] \tag{28}$$

$$\overline{n^2} = \overline{n_0^2} - \tau(2\overline{n_0^2} - \bar{n}_0)(\bar{m}_0+1) - \tfrac{1}{2}\tau^2[2\overline{n_0^3}(\bar{m}_0+1) - \overline{n_0^2}(4\overline{m_0^2}+13\bar{m}_0+9) + 3\bar{n}_0(\overline{m_0^2}+3\bar{m}_0+2)]. \tag{29}$$

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

$$\overline{nm} = \bar{n}_0\bar{m}_0 + \tau[\overline{n_0^2}(\bar{m}_0+1) - \bar{n}_0(\overline{m_0^2}+2\bar{m}_0+1)] + \tfrac{1}{2}\tau^2[\overline{n_0^3}(\bar{m}_0+1) - \overline{n_0^2}(4\overline{m_0^2}+11\bar{m}_0+7) + \bar{n}_0(\overline{m_0^3}+6\overline{m_0^2}+11\bar{m}_0+6)]. \tag{30}$$

The rather complicated expressions given above all simplify for the special case of having no Stokes photons present before the Raman effect takes place. This implies that $\bar{m}_0 = \overline{m_0^2} = \overline{m_0^3} = 0$, and therefore a direct substitution of (28) and (29) into (12) gives

$$g_n^{(2)} = g_{n0}^{(2)} + (\tau^2/\bar{n}_0^2)[(\overline{n_0^2}/\bar{n}_0)(\overline{n_0^2}+\bar{n}_0) - \overline{n_0^3} - \bar{n}_0], \tag{31}$$

while a combination of (30), (28) and (21) produces the relation

$$\overline{nm}/\bar{n}\bar{m} = g_{n0}^{(2)} + (\tau/2\bar{n}_0^2)(\overline{n_0^3} - 7\overline{n_0^2} + 6\bar{n}_0). \tag{32}$$

Now if the method which has been already developed throughout the section is again used to determine the expansions of $\bar{m}$ and $\overline{m^2}$ correct to third order in t, the corresponding expansion of $g_m^{(2)}$ for the present special case obtained from (13) is

$$g_m^{(2)} = 2g_{n0}^{(2)} + (\tau/\bar{n}_0^2)(12\overline{n_0^3} - 46\overline{n_0^2} + 40\bar{n}_0). \tag{33}$$

The initial and short-time variations of this function and those of (31) and (32) are shown at the short-time ends of figures 8 and 9 of § 7, where their behaviour is discussed in greater detail for various special cases of different types of initial pump beam. Note the odd behaviour of these functions for the case of an initially chaotic pump beam in comparison with other cases.

4. Steady state solutions

The photon system, after a sufficiently long period of time has elapsed, settles down into a steady state where the right-hand side of the rate equations (19) can be set equal to zero. If the steady state distribution is denoted $P_{n,\nu-n}(\infty)$, the rate equations give

$$N_1 P_{n,\nu-n}(\infty) = N_2 P_{n-1,\nu-n+1}(\infty). \tag{34}$$

This result is the condition for detailed balance between the photon levels n, m and $n-1$, $m+1$ as shown in figure 1.

By induction from (34),

$$P_{n,v-n}(\infty) = (N_2/N_1)^n P_{0,v}(\infty). \tag{35}$$

Hence for each value of v, a single element of the probability distribution remains unknown in the steady state, and its magnitude can be found making use of (20),

$$\sum_{n=0}^{v} P_{n,v-n}(\infty) = \frac{1-(N_2/N_1)^{v+1}}{1-(N_2/N_1)} P_{0,v}(\infty) = \sum_{n=0}^{v} Q_n(0)R_{v-n}(0). \tag{36}$$

Thus with the help of (35), this result enables the steady state distribution $P_{n,v-n}(\infty)$ to be determined for any initial distributions $Q_n(0)$ and $R_{v-n}(0)$.

It is seen that the subset of probability elements corresponding to a specified v and given by (35) has some similarity to a chaotic type of distribution (compare equation (10.17) of Loudon 1973). This photon distribution can now easily be used to determine the steady state values of the first two moments of the pump beam from (10),

$$\bar{n}_\infty = \frac{N_2/N_1}{[1-(N_2/N_1)]^2} \sum_{v=0}^{\infty} [1+v(N_2/N_1)^{v+1} - (v+1)(N_2/N_1)^v] P_{0,v}(\infty) \tag{37}$$

$$\overline{n_\infty^2} = \frac{(N_2/N_1)^2}{[1-(N_2/N_1)]^3} \sum_{v=0}^{\infty} [2+(v-v^2)(N_2/N_1)^{v+1} + 2(v^2-1)(N_2/N_1)^v - (v^2+v)(N_2/N_1)^{v-1}] P_{0,v}(\infty) + \bar{n}_\infty, \tag{38}$$

and of the correlation function of the two beams from (16),

$$\overline{nm} = \frac{N_2/N_1}{[1-(N_2/N_1)]^2} \sum_{v=0}^{\infty} [v+v^2(N_2/N_1)^{v+1} - (v^2+v)(N_2/N_1)^v] P_{0,v}(\infty) - \overline{n_\infty^2}. \tag{39}$$

The corresponding moments of the Stokes beam are obtained by a direct substitution of these results into (21) and (22).

The results again simplify when almost all the atoms are in their ground states and N_2 is negligibly small. In this case (35) gives

$$P_{n,v-n}(\infty) = 0 \qquad \text{for } n \geqslant 1, \tag{40}$$

and only those elements of the probability distribution with $n = 0$ and v ranging from 0 to ∞ can be nonzero. In other words this result suggests that the pump beam has no photons in the steady state and therefore its moments and the correlation function must vanish. This is verified by (37), (38) and (39) if we let $N_2 = 0$. Keeping this in mind, the first two moments of the Stokes field may be obtained directly from (21) and (22).

By setting $N_2 = 0$ in (36) and assuming that the Stokes beam has no photons at $t = 0$, we get

$$P_m(\infty) = Q_n(0), \tag{41}$$

in agreement with the large-time behaviours of the graphs shown in figures 4 and 6 of § 5, where the general-time dependence of $P_n(\tau)$ is discussed in greater detail. As a direct consequence of (41), we conclude immediately that the moments and degree of second-order coherence of the Stokes field at $\tau = \infty$ are identical to the corresponding quantities of the pump beam at $\tau = 0$. The first moments $\bar{m}$ and degrees of second-order coherence $g_m^{(2)}$ of the Stokes beams shown at the large-time ends of figures 7 and 8 are consistent with this conclusion.

5. General solutions

McNeil and Walls (1974) have pointed out that the rate equations for the photon probability distribution of the Stokes field alone can be solved by a Laplace transform method in the case where $N_2 = 0$. Here we follow the same general method as these authors but obtain a more complete solution which enables the time-dependent statistical properties of both the fields, pumped and Stokes, to be calculated for any kind of initial photon probability distribution.

Substituting the Laplace transform

$$\phi_{n,m}(s) = \int_0^\infty P_{n,m}(\tau) \exp(-s\tau)\, \mathrm{d}\tau, \tag{42}$$

where τ is defined by (27), into the rate equations (8) and solving for $\phi_{n,m}(s)$, one readily obtains

$$\begin{aligned}\phi_{n,m}(s) &= [P_{n,m}(0) + (n+1)m\phi_{n+1,m-1}(s)]/[s+n(m+1)] \\ &= \sum_{\alpha=0}^{m} \frac{\prod_{\beta=0}^{\alpha-1}(f_{\beta+1})}{\prod_{\beta=0}^{\alpha}(s+f_\beta)} Q_{n+\alpha}(0) R_{m-\alpha}(0)\end{aligned} \tag{43}$$

where

$$f_\beta = (n+\beta)(m-\beta+1), \tag{44}$$

the product in the numerator for $\alpha = 0$ is defined to be unity and (2) has been used. The inverse transform of (43) for $n > m$ (see Oberhettinger and Badii 1973, p 224) yields

$$P_{n,m}(\tau) = \sum_{\alpha=0}^{m} \sum_{\gamma=0}^{\alpha} \frac{\prod_{\beta=0}^{\alpha-1}(f_{\beta+1})}{\prod_{\beta=0, \beta\neq\gamma}^{\alpha}(f_\beta - f_\gamma)} \exp(-f_\gamma \tau) Q_{n+\alpha}(0) R_{m-\alpha}(0). \tag{45}$$

The denominator in (43) for $n \leqslant m$ contains repeated factors and therefore in order to determine the inverse transform of $\phi_{n,m}(s)$, it is convenient to rewrite (43) as follows:

$$\begin{aligned}\phi_{n,m}(s) = &\sum_{\alpha=0}^{\lambda} \frac{\prod_{\beta=0}^{\alpha-1}(f_{\beta+1})}{\prod_{\beta=0}^{\alpha}(s+f_\beta)} Q_{n+\alpha}(0) R_{m-\alpha}(0) \\ &+ \theta(m) \sum_{\alpha=\lambda+1}^{m} \frac{\prod_{\beta=0}^{\alpha-1}(f_{\beta+1})}{\prod_{\beta=0}^{\lambda}(s+f_\beta)\prod_{\beta'=\lambda+1}^{\alpha}(s+f_{\beta'})} Q_{n+\alpha}(0) R_{m-\alpha}(0)\end{aligned} \tag{46}$$

where

$$\theta(m) = \begin{cases} 0 & \text{for } m = 0 \\ 1 & \text{for } m > 0 \end{cases} \tag{47}$$

and

$$\lambda = \begin{cases} (m-n)/2 & \text{for } m-n \text{ even} \\ (m-n-1)/2 & \text{for } m-n \text{ odd.} \end{cases} \tag{48}$$

Making use of the convolution theorem, the inverse transform of (46) has the form

$$\begin{aligned} P_{n,m}(\tau) = & \sum_{\alpha=0}^{\lambda} \sum_{\gamma=0}^{\alpha} \frac{\Pi_{\beta=0}^{\alpha-1}(f_{\beta+1})}{\Pi_{\substack{\beta=0\\ \beta\neq\gamma}}^{\alpha}(f_\beta - f_\gamma)} \exp(-f_\gamma \tau) Q_{n+\alpha}(0) R_{m-\alpha}(0) \\ & + \theta(m) \sum_{\alpha=\lambda+1}^{m} \sum_{\gamma=0}^{\lambda} \sum_{\gamma'=\lambda+1}^{\alpha} \frac{\Pi_{\beta=0}^{\alpha-1}(f_{\beta+1})}{\Pi_{\substack{\beta=0\\ \beta\neq\gamma}}^{\lambda}(f_\beta - f_\gamma)\, \Pi_{\substack{\beta'=\lambda+1\\ \beta'\neq\gamma'}}^{\alpha}(f_{\beta'} - f_{\gamma'})} \\ & \times \{\delta_{f_{\gamma'} f_\gamma} \tau \exp(-f_{\gamma'}\tau) + (1-\delta_{f_{\gamma'} f_\gamma})[\exp(-f_\gamma \tau) - \exp(-f_{\gamma'}\tau)]/(f_{\gamma'} - f_\gamma)\} \\ & \times Q_{n+\alpha}(0) R_{m-\alpha}(0) \end{aligned} \tag{49}$$

where

$$\delta_{f_{\gamma'} f_\gamma} = \begin{cases} 1 & \text{for } f_{\gamma'} = f_\gamma \\ 0 & \text{otherwise.} \end{cases} \tag{50}$$

Equations (45) and (49) enable computation of the complete joint probability distribution $P_{n,m}$ and the separate distributions P_n and P_m respectively defined by

$$P_n(\tau) = \sum_{m=0}^{\infty} P_{n,m}(\tau) \tag{51}$$

$$P_m(\tau) = \sum_{n=0}^{\infty} P_{n,m}(\tau) \tag{52}$$

for arbitrary time τ and any arbitrary initial distributions $Q_{n+\alpha}(0)$ and $R_{m-\alpha}(0)$. For the sake of illustration the same types of initial distribution summarized by Simaan and Loudon (1975a) will be used to take account of $Q_{n+\alpha}(0)$ and $R_{m-\alpha}(0)$, but for the remainder of this section we assume that the Stokes field has no photons at $\tau = 0$, and therefore

$$R_{m-\alpha}(0) = \begin{cases} 1 & \text{for } \alpha = m \\ 0 & \text{for } \alpha \neq m. \end{cases} \tag{53}$$

If the incident beam is initially a number state with 10 photons in it, the time evolution of the joint distribution $P_{n,m}$ is shown in figure 2. The progressively slower decay of elements corresponding to the first four values of n and last four values of m of the photon numbers is apparent. At large values of τ, off the right-hand end of the figure, the joint distribution tends to its steady state form in which only $P_{0,10}$ is nonzero and has the value unity.

The time dependences of P_n and P_m for the same special case as that of figure 2 can be obtained from (51) and (52). We note here that McNeil and Walls (1974) have discussed the same special case and found expressions for P_m only, but their results are not in full agreement with the ones determined above. (The summation in (6.5) of McNeil and Walls has the incorrect lower limit of n_s^0 which should be replaced by zero so that the whole equation becomes correct for $n < N_0/2$ rather than $n \leqslant N_0/2$ as suggested; on the other hand (6.6) which is supposed to be valid for $n \geqslant N_0/2$ rather than just $n > N_0/2$ is a great deal different from our corresponding equation obtained from (52) and (49) for the special case we are dealing with.)

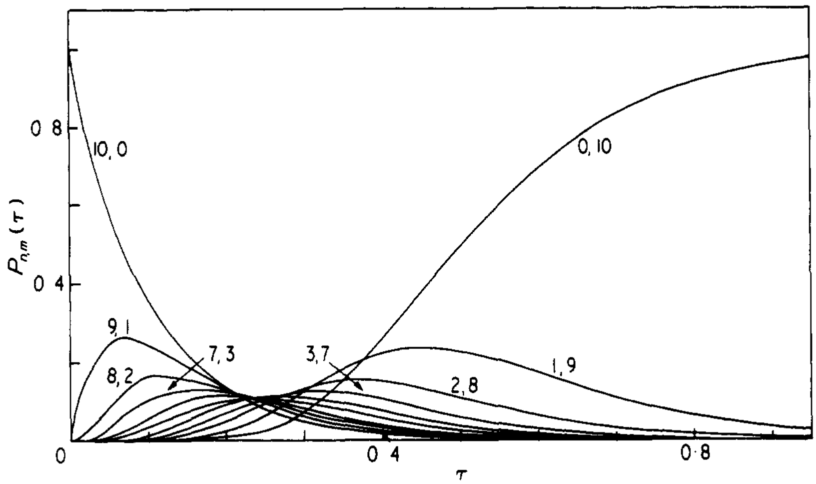

Figure 2. Time dependence of the joint photon probability distribution $P_{n,m}(\tau)$. The initial distribution is $P_{10,0} = 1$ and the only nonzero elements of the distribution at subsequent times are those for which the sum of the two subscripts of P is equal to 10. The numbers attached to the curves indicate the common values of n and m.

The time dependences of the separate distributions of both the pump and Stokes beams for an initially coherent incident beam with a mean number of photons $\bar{n}_0 = 2$, are respectively shown in figures 3 and 4. It is seen from these figures that at large values of τ, the distributions tend to their steady state forms in which the zeroth element of P_n and all the elements of P_m are nonzero. The behaviour of the elements of the distribution illustrated in figure 4 suggests that the growing Stokes field has a chaotic type of distribution for the values of time τ smaller than 1·1, while as we move away towards greater values of τ, this distribution continuously changes from the chaotic form to the coherent

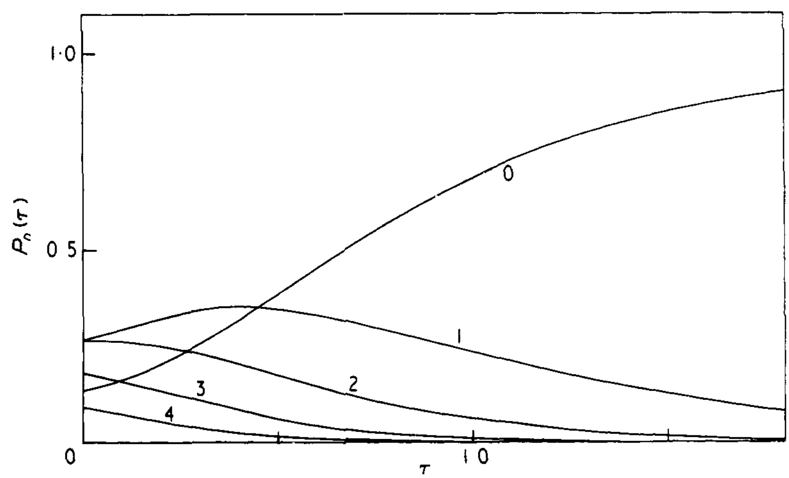

Figure 3. Time dependence of $P_n(\tau)$ for initial distributions in which beam n is coherent with a mean photon number $\bar{n}_0 = 2$ and beam m has no photons in it. The numbers attached to the curves indicate the corresponding values of n. Only the first five elements of the distribution are shown.

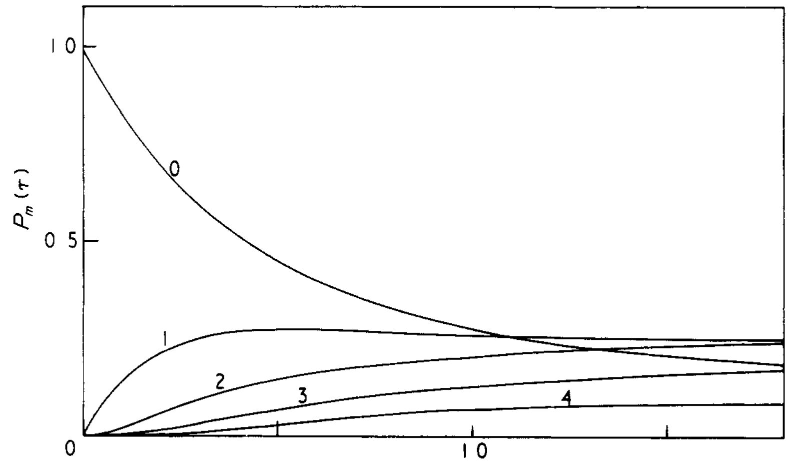

Figure 4. Time dependence of $P_m(\tau)$ for the same initial distributions as figure 3. The numbers attached to the curves indicate the values of m.

form, and as the steady state condition is achieved, the result predicted by (41) is verified and the distribution of the Stokes beam becomes purely coherent.

The time evolution of P_n and P_m for another simple example of an initially chaotic pump beam this time, with $\bar{n}_0 = 2$ again, are respectively shown in figures 5 and 6. Clearly the behaviours of the graphs in figure 5 are similar to those of figure 3, and the same thing can be said about figures 6 and 4 in the sense that the Stokes field grows chaotically and (41) is verified when the steady state is achieved. This means that since for the present special case the pump beam is initially chaotic, the steady state distribution of the Stokes beam is chaotic too, and therefore in full agreement with the result

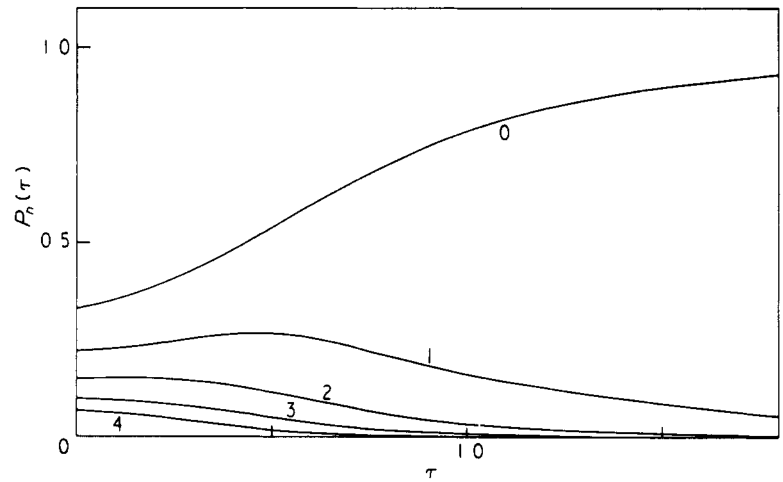

Figure 5. Time dependence of $P_n(\tau)$ for initial distributions in which beam n is chaotic with $\bar{n}_0 = 2$ as the mean photon number, and beam m has no Stokes photons. Again only the first five elements of the distribution are shown.

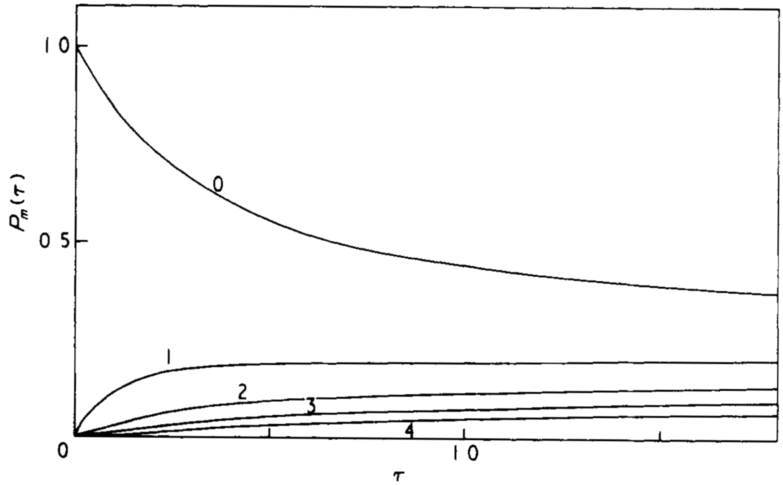

Figure 6. Time dependence of $P_m(\tau)$ for the same initial distributions as used in figure 5.

shown at the large-time end of figure 6 as the elements of P_m are tending to saturate at the initial values of the corresponding elements of P_n given at the vertical axis of figure 5 where $\tau = 0$.

6. Approximate solutions

Consider the special case where beam n is initially much more intense than beam m,

$$\bar{n}_0 \gg \bar{m}_0. \tag{54}$$

It is clear that as the Raman effect proceeds, the ratio $\bar{n}/\bar{m}$ decreases, the poorer the approximate solutions presented in this section become, and therefore should not be evaluated for large values of τ. In the limit of (54), only those terms in the joint distribution with $n \gg m$ have significant magnitude, and hence in deriving an approximate expression for $P_{n,m}$, (45) alone need be considered. Within these remarks, α and β which respectively range from 0 to m and 0 to α, can be neglected in comparison to n, and accordingly (45) gives

$$P_{n,m}(\tau) = \sum_{\alpha=0}^{m} \sum_{\gamma=0}^{\alpha} \frac{\prod_{\beta=0}^{\alpha-1} (m-\beta)}{\prod_{\substack{\beta=0 \\ \beta \neq \gamma}}^{\alpha} (\gamma-\beta)} \exp[-n(m-\gamma+1)\tau] Q_n(0) R_{m-\alpha}(0). \tag{55}$$

This equation with the help of the relations

$$\prod_{\beta=0}^{\alpha-1} (m-\beta) = m!/(m-\alpha)! \tag{56}$$

$$\prod_{\substack{\beta=0 \\ \beta \neq \gamma}}^{\alpha} (\gamma-\beta) = (-1)^{\alpha-\gamma} \gamma!(\alpha-\gamma)! \tag{57}$$

$$[\exp(n\tau)-1]^{\alpha} = \sum_{\gamma=0}^{\alpha} (-1)^{\alpha-\gamma} \binom{\alpha}{\gamma} \exp(n\gamma\tau), \tag{58}$$

can be written as follows:

$$P_{n,m}(\tau) = \sum_{\alpha=0}^{m} \binom{m}{\alpha} [\exp(n\tau)-1]^{\alpha} \exp[-n(m+1)\tau] Q_n(0) R_{m-\alpha}(0). \tag{59}$$

With some lengthy algebra, and making use of (59) in (11), the first two moments of the Stokes field are

$$\bar{m} = (\bar{m}_0+1) \sum_{n=0}^{\infty} \exp(n\tau) Q_n(0) - 1 \tag{60}$$

$$\overline{m^2} = (\overline{m_0^2}+3\bar{m}_0+2) \sum_{n=0}^{\infty} \exp(2n\tau) Q_n(0) - 3(m_0+1) \sum_{n=0}^{\infty} \exp(n\tau) Q_n(0) + 1. \tag{61}$$

In a similar way the first two moments of the pump beam can be obtained without difficulty, but since the variations in these moments are assumed to be negligible in comparison to their initial values, we have decided to devote this section to the study of the changes in the statistical properties of the Stokes field alone. Equation (60) is in agreement with the corresponding equations (12.103) of Loudon (1973) and (25) of Shen (1967). The method followed by Loudon can easily be used to derive (61) as well.

The summations in (60) and (61) may be performed without difficulty for the initial distributions given by Simaan and Loudon (1975a), and for an initial number-state pump beam containing n_0 photons we find

$$\bar{m} = (\bar{m}_0+1)\exp(n_0\tau) - 1 \tag{62}$$

$$\overline{m^2} = (\overline{m_0^2}+3\bar{m}_0+2)\exp(2n_0\tau) - 3(\bar{m}_0+1)\exp(n_0\tau) + 1, \tag{63}$$

while the corresponding results for an initially coherent beam of a mean photon number $\bar{n}_0$ are

$$\bar{m} = (\bar{m}_0+1)\exp\{\bar{n}_0[\exp(\tau)-1]\} - 1 \tag{64}$$

$$\overline{m^2} = (\overline{m_0^2}+3\bar{m}_0+2)\exp\{\bar{n}_0[\exp(2\tau)-1]\} - 3(\bar{m}_0+1)\exp\{\bar{n}_0[\exp(\tau)-1]\} + 1. \tag{65}$$

In the limit where $\bar{n}_0 \exp(\tau)$ and $\bar{n}_0 \exp(2\tau)$ are less than $(1+\bar{n}_0)$, and for the case of an initially chaotic pump beam, (60) and (61) can also be performed to become

$$\bar{m} = (\bar{m}_0+1)/[1+\bar{n}_0-\bar{n}_0\exp(\tau)] - 1 \tag{66}$$

$$\overline{m^2} = (\overline{m_0^2}+3\bar{m}_0+2)/[1+\bar{n}_0-\bar{n}_0\exp(2\tau)] - 3(\bar{m}_0+1)/[1+\bar{n}_0-\bar{n}_0\exp(\tau)] + 1. \tag{67}$$

A direct substitution of (59) into (52) gives an expression for $P_m(\tau)$, and after setting $n = n_0$ it can be written as

$$P_m(\tau) = \sum_{n_0=0}^{\infty} Q_{n_0}(0) P_m(n_0, \tau) \tag{68}$$

where

$$P_m(n_0, \tau) = \left(\frac{\bar{m}_t^m}{(1+\bar{m}_t)^{1+m}}\right) \sum_{\alpha=0}^{m} \binom{m}{\alpha} \left(\frac{1}{\bar{m}_t}\right)^{m-\alpha} R_{m-\alpha} \tag{69}$$

represents the probability distribution of the growing Stokes field for an initially number-state pump beam containing n_0 photons, and

$$\bar{m}_t = \exp(n_0\tau) - 1. \tag{70}$$

Although carrying on discussing and evaluating all the above results for any type of initial distribution of both the pump and Stokes beam is possible, in the rest of this section we shall only consider the case of an initially number-state pump field and evaluate (69) for the various kinds of initial distributions given in § 3 of Simaan and Loudon (1975a).

Equation (69) enables the study of the sort of amplification occurring for any type of initial Stokes field by the Raman effect. In what follows four examples of these fields are considered to evaluate $P_m(n_0, \tau)$. As a trivial but interesting example at the same time, let us assume the case described by (53) where the Stokes field initially has no photons in it. In this case (69) takes the form

$$P_m(n_0, \tau) = \bar{m}_t^m/(1+\bar{m}_t)^{1+m}. \tag{71}$$

Clearly the right-hand side of this equation is the Bose–Einstein distribution, appropriate to a thermal beam which has a mean photon number $\bar{m}_t$ given by (70). Therefore we conclude that for the very special case of no initial Stokes photons, the growing field is thermal or what is sometimes called chaotic, and its first two moments are well known to be

$$\bar{m} = \bar{m}_t \tag{72}$$

$$\overline{m^2} = 2\bar{m}_t^2 + \bar{m}_t. \tag{73}$$

Respectively these relations can be obtained directly from (62) and (63) by taking $\bar{m}_0 = \overline{m_0^2} = 0$.

The next simplest example to consider is an initially number-state Stokes field containing $\bar{m}_0$ photons, for which (69) can be reduced to

$$P_m(n_0, \tau) = \left(\frac{\bar{m}_t^m}{(1+\bar{m}_t)^{1+m}}\right)\binom{m}{\bar{m}_0}\left(\frac{1}{\bar{m}_t}\right)^{\bar{m}_0}. \tag{74}$$

We note that this equation is consistent with the form taken by (5.10) of Schell and Barakat (1973) in the limit where only photon emission is considered. The distribution given by their equation is obtained by starting with a number state which then interacts with the atomic transitions to produce thermal photons. The first two moments of (74) are

$$\bar{m} = \bar{m}_N + \bar{m}_t \tag{75}$$

$$\overline{m^2} = \bar{m}_N^2 + 3\bar{m}_N\bar{m}_t + 2\bar{m}_t^2 + \bar{m}_t \tag{76}$$

where

$$\bar{m}_N = \bar{m}_0 \exp(n_0\tau), \tag{77}$$

and $\bar{m}_t$ given by (70), are the two time-dependent parts of $\bar{m}$, respectively contributed by the number-state and thermal fields. Note that (75) is in agreement with (62), and since for a number-state distribution, $\overline{m_0^2} = \bar{m}_0^2$, (63) and (76) are also identical. The conclusion to be drawn from the above results hence is that the initially number-state field is amplified in accordance with (77), and the amplification process generates a thermal field of a mean photon number $\bar{m}_t$ to be determined from (70).

The third in our series of examples is an initially coherent Stokes field with a mean number of photons $\bar{m}_0$. This case reduces (69) to the following well known form of distribution which describes a mixture of the coherent and thermal radiation (Jakeman and

Pike 1969, Lachs 1965, Schell and Barakat 1973):

$$P_m(n_0, \tau) = [\bar{m}_t^m/(1+\bar{m}_t)^{1+m}]\exp[-\bar{m}_c/(1+\bar{m}_t)]L_m[-\bar{m}_c/\bar{m}_t(1+\bar{m}_t)] \qquad (78)$$

where

$$\bar{m}_c = \bar{m}_0 \exp(n_0\tau) \qquad (79)$$

is the mean photon number of the amplified coherent beam and L_m is a Laguerre polynomial. The first and second moments of the distribution shown in (78) are

$$\bar{m} = \bar{m}_c + \bar{m}_t \qquad (80)$$

$$\overline{m^2} = \bar{m}_c^2 + 4\bar{m}_c\bar{m}_t + \bar{m}_c + 2\bar{m}_t^2 + \bar{m}_t. \qquad (81)$$

Again these relations can directly be obtained from (62) and (63) by setting $\overline{m_0^2} = \bar{m}_0^2 + \bar{m}_0$ which is valid if the Stokes beam is initially coherent. The conclusion to be drawn from the results obtained above for the present example is that an initially coherent Stokes beam is amplified in accordance with (79), the amplified version of the beam maintains its initial type and the amplification process generates a thermal field of a mean photon number $\bar{m}_t$ given by (70). For more discussion and physical interpretation of this point a paper by Loudon (1970) is recommended. It remains to mention here that the problem of Raman scattering from phonons, in the limit of parametric approximation of the pump beam, has been considered by Walls (1973), and we note the closely related results of his § 3, obtained for the case of an initially coherent Stokes field, to one presented above.

Finally we end the series of our examples by assuming that the Stokes field is initially thermal with $\bar{m}_0$ as the mean photon number; this changes (69) to another thermal distribution of the Bose–Einstein type, similar to that of (71) but with every $\bar{m}_t$ replaced by the $\bar{m}$ given in (62). Therefore an initially thermal field is amplified, but its statistical properties remain unchanged.

All the above four examples apply to the case of an initially number state (any other kind of pump field with $Q_{n0}(0)$ as the initial distribution can be taken into account by using (68)), and show that in general the growing Stokes beam consists of an amplified version of its initial type in addition to a thermal component which is generated by the process of amplification. We note here the close similarity of this result to the general conclusion drawn from a paper by Gordon *et al* (1963).

7. Discussion

The general results derived in § 5 have been used to construct graphs for the time dependences of the joint probability distribution and the distributions of the individual beams for a variety of initial types of photon distribution, as shown in figures 2 to 6. The moments of these distributions and the correlation function of the two beams are often of more immediate physical interest than the distributions themselves, and their time dependences, obtained from (10), (11) and (16) with the direct use of (45) and (49), give the most compact description of the effects of the Raman scattering process on the pump and Stokes light beams. In this section we present graphs for the time dependences of the mean photon numbers, the degrees of second-order coherence and the correlation function of the two beams for initial states in which the Stokes beam distribution has the form of (53) for the case of no Stokes photons at $t = 0$, and the pump beam initially has various kinds of distribution, namely number-state, coherent and thermal with mean number of photons $\bar{n}_0 = 2$.

The decaying and growing curves in figure 7 respectively show the exact time variations of $\bar{n}$ and $\bar{m}$ for the various special cases mentioned above. The behaviours of these moments at short times are consistent with the results of § 3. It is clear that an initially thermal pump beam has caused faster decaying of $\bar{n}$ and growing of $\bar{m}$ than that induced by initial pump fields of the coherent type or even number state. As long as $\bar{n}_0 \gg \bar{m}_0$,

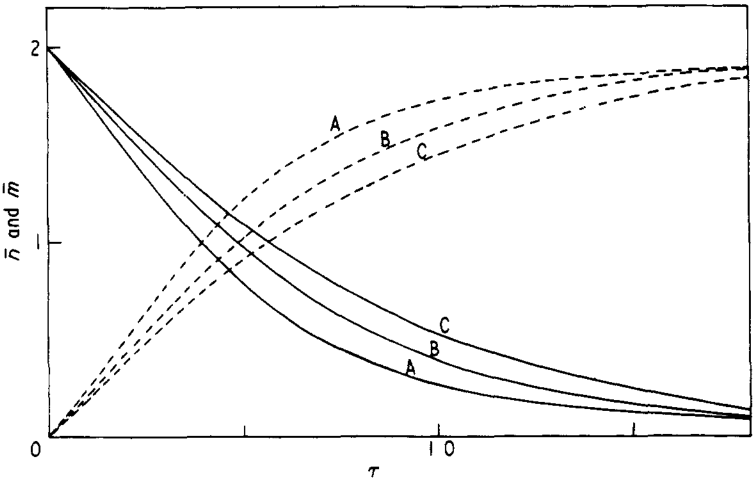

Figure 7. Time dependence of the mean photon numbers $\bar{n}$ (———) and $\bar{m}$ (- - - - -) for initial distributions in which beam n has the character indicated on the curves and beam m initially has no photons in it (A: chaotic; B: coherent; C: number). The initial mean number $\bar{n}_0$ is equal to 2 for all three types of pump beam.

that is for $\tau < 0{\cdot}2$ for the cases shown in figure 7, the Stokes beam is chaotic irrespective of the nature of the pump field. Examples are shown for $\tau < 0{\cdot}2$ in figures 4 and 6. At longer times the nature of the Stokes beam becomes more complicated as the nature of the pump field begins to influence the statistical properties of the Stokes beam. However in the $\tau \to \infty$ steady state limit, where all the photons have been transferred to the Stokes beam, the situation is again simple because the $\tau \to \infty$ probability distribution of the Stokes beam is the same as the $\tau = 0$ probability distribution of the pump beam.

The dependences of the degrees of second-order coherence of the two beams on time τ for the same initial distributions as used in figure 7 are illustrated in figure 8. The initial and short-time behaviours of $g_n^{(2)}$ and $g_m^{(2)}$ are respectively in accordance with (31) and (33), and the direct substitution of the appropriate expressions for $\overline{n_0^3}$, $\overline{n_0^2}$ and $g_{n0}^{(2)}$ of the various types of initial beam in these equations explains the rather peculiar nature of the curves at the short-time end of figure 8, and especially the oddity of the curves associated with the special case of an initially thermal pump field. Apart from this case $g_n^{(2)}$ generally tends to increase, while $g_m^{(2)}$ tends to decrease until it reaches the steady state limit of saturation which is equal to $g_{n0}^{(2)}$ as shown in § 4.

According to (33) the initial value of $g_m^{(2)}$ is $2g_{n0}^{(2)}$. Thus the pump beams which have larger fluctuations generate initial Stokes beams which also have correspondingly larger fluctuations. The $\tau \to \infty$ value of $g_m^{(2)}$ is $g_{n0}^{(2)}$ and the magnitude of $g_m^{(2)}$ is thus reduced by a factor of 2 between $\tau = 0$ and $\tau = \infty$. The large fluctuations in the chaotic pump beam produce a positive term linear in τ in (33) so that $g_m^{(2)}$ rises from its initial value of 4 in this case before conforming to the requirement that its $\tau = \infty$ value must be

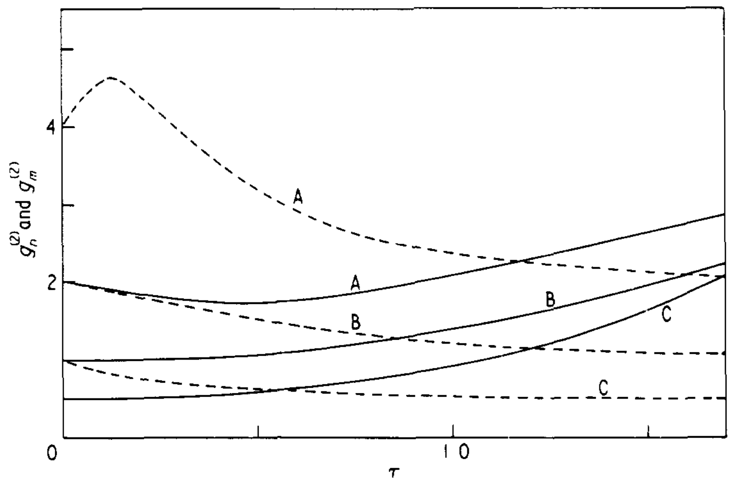

Figure 8. Time dependence of the degrees of second-order coherence $g_n^{(2)}$ (———) and $g_m^{(2)}$ (- - - - -) for the same initial distributions as figure 7 (A, B, C as for figure 7).

2. Nothing has so far been said about the steady state value of $g_n^{(2)}$, and the reason is because physically it has no meaning to discuss the statistical properties of a beam which contains no photons and obviously is the case of the pump beam in the steady state.

Figure 9 shows graphs of the time dependence of the normalized correlation function $\overline{nm}/\bar{n}\bar{m}$ for the same initial states used in the two previous figures. The initial and short-time behaviours of these graphs are consistent with (32). According to this equation $\overline{nm}/\bar{n}\bar{m}$ has an initial value of $g_{n0}^{(2)}$, which is larger for initial pump beams which have larger fluctuations and again the term linear in τ in (32) is positive for a chaotic pump, producing a hump in the corresponding curve in figure 9. The general behaviour of the curves is qualitatively explained as follows. At short times, the Stokes beam is preferentially produced at the peak fluctuations in the pump beam, leading to a positive correlation

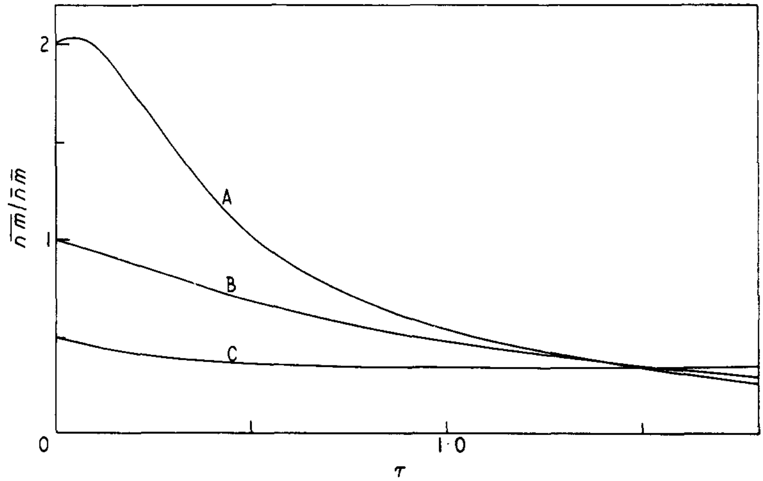

Figure 9. Time dependence of the normalized correlation function $\overline{nm}/\bar{n}\bar{m}$ for the same initial distributions as figure 7 (A, B, C as for figure 7).

between the beams, ie $\overline{nm} > \bar{n}\bar{m}$ in the case of the chaotic pump. At longer times the peaks in the time-dependent fluctuations of the pump beam have been transferred to the Stokes beam, leaving gaps in the pump coincident with the peaks in the Stokes beam. Thus an anticorrelation effect, ie $\overline{nm} < \bar{n}\bar{m}$, sets in at longer times, accounting for the downward trend of the curves in figure 9.

All the above remarks apply to the case of no Stokes photons at $t = 0$. Any other cases different from this are much more complicated, and for most times simple conclusions are not available—even the $\tau \to \infty$ distribution obtained from (36) is fairly complicated. The case of an initially number-state pump, containing $\bar{n}_0$ photons, however is not difficult to discuss as follows. For $\bar{n}_0 \gg \bar{m}_0$ the Stokes beam has the thermal component it would have had in the absence of any Stokes photons initially, and the initial Stokes beam is amplified while maintaining its initial statistical properties. The Stokes beam thus contains two identifiable components. At longer times the Stokes distribution becomes complicated. In the $\tau \to \infty$ limit, the Stokes beam has the same distribution as it had at $\tau = 0$, but with the origin of the distribution shifted from $m = 0$ to $m = \bar{n}_0$.

There have not to the author's knowledge been any observations of coherence or correlation effects in experiments on the stimulated Raman effect. There is a similar lack of experimental data on coherence changes in the two-photon absorption process. However, the stimulated Raman effect should be the more favourable for such observations since the growing Stokes beam is expected to show interesting coherence effects beginning with the initiation of the beam. By contrast, in two-photon absorption, an experiment would need to detect changes from the initial value of the coherence of a beam which is attenuated in the absorption process. It appears that the coherence properties of the Stokes beam predicted in this paper should be observable with currently available materials and techniques.

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