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A master equation approach to nonlinear optics

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Abstract. The nonlinear interaction of light with matter is described from a quantum-statistical point of view. The phenomena of two-photon emission and two-photon absorption including both the single- and two-mode cases and the Raman effect are discussed in detail. A master equation for the density operator of the light fields alone is derived. This operator equation is converted to a c number equation and analytic solutions are obtained for the diagonal matrix elements of the density operator in the Fock representation. No linearizing approximation is introduced. These solutions allow one to compute the moments of the photon distribution for the above nonlinear processes.

1. Introduction

A quantum-statistical description of the nonlinear interaction of light with matter is presented. This description is based on a hamiltonian formulation of the nonlinear interaction with both atoms and the electromagnetic field quantized (Shen 1967, Walls 1971). Loss mechanisms are neglected in this description. The central part of our analysis is based on an equation of motion derived for the density operator of the light field alone, the atomic variables having been removed by a tracing procedure. This equation was first derived by Shen (1967) in operator form. Analytic solutions to this equation have not however been presented except in special cases. We refer here to the work of Lambropolous (1967) on the degenerate case of two-photon emission into a single mode and Agarwal (1970) on the degenerate case of two-photon absorption from a single mode. Analytic solutions to a master equation describing the Raman effect where the parametric approximation has been made for the pump field have recently been presented by Walls (1973). This approximation however, linearizes the system and no allowance for pump depletion is included.

It is the purpose of the present paper to present analytic solutions to nonlinear master equations for a variety of nonlinear optical phenomena. Since the operator master equation is inaccessible to analytic solution we resort to finding solutions for the matrix elements of the density operator in the Fock representation. In order to determine the mean number and higher-order moments of the photon distribution it proves sufficient to solve for the diagonal matrix elements of the density operator.

The examples we consider are two-photon emission and absorption including both the single- and two-mode cases and the Raman effect. The nonlinear differential difference equations derived for the diagonal matrix elements of the density operator are solved either by Laplace transforms or a generating-function technique depending on which proves more readily applicable. Analytic solutions for the photon distributions are derived and the time evolutions of the mean number and variance of the distributions are presented.

2. Hamiltonian formulation of the nonlinear interaction

The electric-field operator for the free field may be expanded in terms of normal modes as (Glauber 1963a, b)

$$E(\boldsymbol{r}, t) = \mathrm{i} \sum_k \left(\frac{\hbar\omega_k}{2}\right)^{1/2} [a_k(t)u_k(\boldsymbol{r}) - a_k^\dagger(t)u_k^*(\boldsymbol{r})] \tag{2.1}$$

where $a_k(t)$ and $a_k^\dagger(t)$ are the boson annihilation and creation operators for the kth mode. The effects of the linear polarizability in a non-dissipative isotropic medium may be accounted for by including a dielectric constant $\epsilon = 1+4\pi\chi$ where χ is the linear susceptibility of the medium (Shen 1967). The mode functions are then taken to satisfy the wave equation

$$\left(\nabla^2 + \frac{\omega_k^2\epsilon_k}{c^2}\right) u_k(\boldsymbol{r}) = 0 \tag{2.2}$$

with the orthonormality condition

$$\int_V (\epsilon_k\epsilon_l)^{1/2} u_k^*(\boldsymbol{r}) u_l(\boldsymbol{r})\, \mathrm{d}^3\boldsymbol{r} = \delta_{kl}. \tag{2.3}$$

The total hamiltonian H describing the interaction of the electromagnetic field with a nonlinear medium may be expressed as the sum of a free hamiltonian H_0 and an interaction hamiltonian H_1

$$H = H_0 + H_1. \tag{2.4}$$

We consider the medium to consist of an ensemble of two-level atoms with transition frequency Ω. Hence the free hamiltonian may be written

$$H_0 = \sum_k \hbar\omega_k a_k^\dagger a_k + \tfrac{1}{2}\hbar\Omega \sum_i (c_{2i}^\dagger c_{2i} - c_{1i}^\dagger c_{1i}), \tag{2.5}$$

where the $c_{\alpha i}$ and $c_{\alpha i}^\dagger$ are the fermion annihilation and creation operators for the α level of the ith atom.

A general interaction hamiltonian describing an n photon process may be written as (Shen 1967, Walls 1971)

$$H_1 = \hbar\mathscr{E}^{(n)} \sum_i c_{2i}^\dagger c_{1i} \prod_{j=1}^{m} E_j^{(-)}(\boldsymbol{r}_i) \prod_{k=m+1}^{n} E_k^{(+)}(\boldsymbol{r}_i) + \text{adjoint} \tag{2.6}$$

where $\mathscr{E}^{(n)}$ is the matrix element for an n photon transition consisting of m emissions and $n-m$ absorptions. We take $E_k^{(+)}$ to represent a single mode of the electromagnetic field (this is appropriate for cavity modes)

$$E_k^{(+)}(\boldsymbol{r}_i) = \{E_k^{(-)}(\boldsymbol{r}_i)\}^\dagger = \mathrm{i}(\tfrac{1}{2}\hbar\omega_k)^{1/2} u_k(\boldsymbol{r}_i) a_k. \tag{2.7}$$

Making this substitution for $E_k^{(-)}$ and $E_k^{(+)}$ in equation (2.6) the interaction hamiltonian may be written in the form

$$H_i = \sum_i \mu_i^{(n)} c_{2i}^\dagger c_{1i} O^{(n)} + \text{adjoint} \tag{2.8}$$

where

$$O^{(n)} = \prod_{j=1}^{m} a_j^\dagger \prod_{k=m+1}^{n} a_k \tag{2.9}$$

and

$$\mu_i^{(n)} = \mathscr{E}^{(n)} \prod_{l=1}^{n} (\tfrac{1}{2}\hbar\omega_l)^{1/2} \prod_{j=1}^{m} u_j^*(\boldsymbol{r}_i) \prod_{k=m+1}^{n} u_k(\boldsymbol{r}_i). \tag{2.10}$$

No account of losses is included in the above description.

3. Master equation for the light field

The equation of motion for the density operator χ of the light–atom system is given by

$$i\hbar\frac{\partial\chi}{\partial t} = [H_1(t), \chi(t)] \tag{3.1}$$

where $H_1(t)$ is the interaction hamiltonian in the interaction picture. We consider the atomic system to be in thermal equilibrium at temperature T. The density operator ρ of the field alone at time t is obtained by tracing out over the atomic variables:

$$\rho(t) = \mathrm{Tr}_A\{\chi(t)\}. \tag{3.2}$$

We assume that the thermal equilibrium of the atomic system is not disturbed by the photon fields thus we may write

$$\chi(t) = \rho(t) \otimes \rho_A(0) \tag{3.3}$$

$$\rho_A(0) = \prod_i \rho_i(0) \tag{3.4}$$

where $\rho_i(0)$ is the thermal equilibrium density operator for the ith atom.

The equation of motion for $\rho(t)$ may then be derived using the Born and Markoff approximations by standard techniques. (For a complete discussion of the derivation of quantum-mechanical master equations in the context of quantum optics see the excellent review articles of Haken 1970 and Agarwal 1973.) The result obtained is

$$\frac{\partial\rho}{\partial t} = k^{m,n-m}\beta_1([O^{(n)}\rho, O^{(n)\dagger}] + [O^{(n)}, \rho O^{(n)\dagger}]) - k^{m,n-m}\beta_2([O^{(n)}, O^{(n)\dagger}\rho] + [\rho O^{(n)}, O^{(n)\dagger}]) \tag{3.5}$$

where

$$k^{m,n-m} = 2\pi g(\Omega)|\mathscr{E}^{(n)}|^2 \prod_{l=1}^{n} (\tfrac{1}{2}\hbar\omega_l) \int_V \mathrm{d}^3\boldsymbol{r} N(\boldsymbol{r}) \prod_{j=1}^{n} |u_j(\boldsymbol{r})|^2 \tag{3.6}$$

and $g(\omega)$ is the lineshape function for the atoms. β_1 and β_2 are the thermal equilibrium occupation numbers for the atomic levels $|1\rangle$ and $|2\rangle$ and $N(\boldsymbol{r})$ is the atomic density in the medium. We have here only included resonant transitions where

$$\Omega = \sum_{i=1}^{m} \omega_i - \sum_{k=m+1}^{n} \omega_k.$$

The equation (3.5) is popularly known as the master equation for ρ.

In order to obtain the complete statistical description of the light field one must know ρ completely. However, the salient physical features of the light may be obtained from the moments of the number operator: $\mathrm{Tr}\{(a_j^\dagger a_j)^k\rho\}$ which do not require a complete knowledge of ρ. If we consider a single mode interacting with the medium the kth moment of the number operator is given by

$$\mathrm{Tr}\{(a^{\dagger}a)^k\rho\} = \sum_{n=0}^{\infty} n^k\langle n|\rho|n\rangle. \tag{3.7}$$

We see that only the diagonal matrix elements of ρ (in the number representation) are needed in order to obtain the mean photon number and the higher moments. It will therefore only be necessary for us to solve the above master equation for the diagonal matrix elements of ρ.

4. Two-photon emission

4.1. Single-mode case

We shall first consider the case of two-photon emission into a single mode. The population of the atomic ground state is considered to be completely depleted ($\beta_1 = 0$) and the population of the upper level of the atoms is held constant by external pumping. The master equation for two-photon emission follows then as a special case of equation (3.5) with $O^{(2)} = aa$

$$\frac{\partial\rho}{\partial t} = -k([aa, a^{\dagger}a^{\dagger}\rho]+[\rho aa, a^{\dagger}a^{\dagger}]) \tag{4.1}$$

where we have set $k^{(2,0)} = k$.

The equation of motion for the diagonal matrix elements of ρ in the Fock representation which follows immediately from equation (4.1) is

$$\frac{\partial\rho_n}{\partial\tau} = n(n-1)\rho_{n-2}-(n+2)(n+1)\rho_n \tag{4.2}$$

where $\tau = 2kt$ and $\rho_n = \langle n|\rho|n\rangle$.

The solution to equation (4.2) has been given by Lambropolous (1967). We shall include the results here for completeness. The equation may be solved by Laplace transforms yielding

$$\rho_{2m}(\tau) = (2m)!\sum_{n=0}^{m}\frac{\rho_{2n}(0)}{(2n)!}\sum_{j=n}^{m} A_{jn}\exp[-(2j+1)(2j+2)\tau]$$

$$\rho_{2m+1}(\tau) = (2m+1)!\sum_{n=0}^{m}\frac{\rho_{2n+1}(0)}{(2n+1)!}\sum_{j=n}^{m} B_{jn}\exp[-(2j+2)(2j+3)\tau] \tag{4.3}$$

where

$$A_{jn} = \left(\prod_{\substack{q=n\\ q\neq j}}^{m}(q-j)[2(q+j)+3]\right)^{-1}$$

$$B_{jn} = \left(\prod_{\substack{q=n\\ q\neq j}}^{m}(q-j)[2(q+j)+5]\right)^{-1}.$$

An initial coherent or chaotic distribution of photons may be accounted for by putting the appropriate initial distribution $\rho_k(0)$ directly in equation (4.3).

The time-dependent behaviour of the mean number and variance of the photon distribution for an initial vacuum state are shown in figures 1 and 2 respectively.

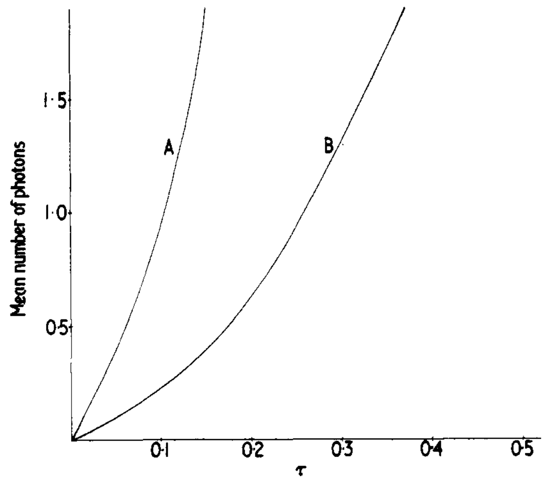

Figure 1. Two-photon emission—initial vacuum state. Graph of the mean number of photons $\bar{n}$ against τ for single-mode and two-mode emission: A, single mode; B, two mode.

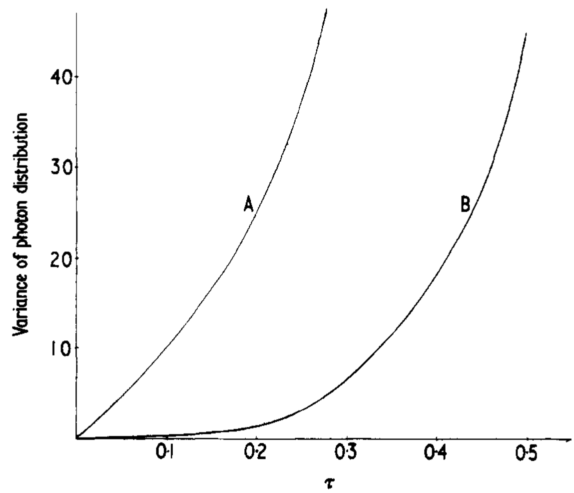

Figure 2. Two-photon emission—initial vacuum state. Graph of the variance of the photon distribution against τ for single-mode and two-mode emission: A, single mode; B, two mode.

4.2. Two-mode case

We now consider the case of two-photon emission into two modes with frequencies ω_1 and ω_2. The master equation follows from equation (3.5) with $O^{(2)} = a_1 a_2$:

$$\frac{\partial \rho}{\partial t} = -k([a_1 a_2, a_1^\dagger a_2^\dagger \rho] + [\rho a_1 a_2, a_1^\dagger a_2^\dagger]). \tag{4.4}$$

The equation of motion for the diagonal matrix elements of ρ follows immediately

$$\frac{\partial \rho_{n_1,n_2}}{\partial t} = n_1 n_2 \rho_{n_1-1,n_2-1} - (n_1+1)(n_2+1)\rho_{n_1,n_2} \tag{4.5}$$

where $\tau = 2kt$ and we have used the notation $\rho_{n_1,n_2} = \langle n_1, n_2|\rho|n_1, n_2\rangle$. Thus the operator equation (4.1) has been reduced to a c number equation and is thus accessible to analytic solution.

To facilitate the solution of equation (4.5) we assume that there is a definite number of photons present at $t = 0$,

$$N_0 = n_2(0) - n_1(0) = n_2^0 - n_1^0. \tag{4.6}$$

Then since $[H, a_2^\dagger a_2 - a_1^\dagger a_1] = 0$, $n_2(t) - n_1(t)$ is a conserved quantity so that

$$N_0 = n_2(t) - n_1(t), \qquad \text{for all } t. \tag{4.7}$$

Using this constant of the motion in equation (4.5) to eliminate n_2 yields

$$\frac{\partial \rho_n}{\partial t} = n(N_0 + n)\rho_{n-1} - (n+1)(N_0 + n + 1)\rho_n \tag{4.8}$$

where we have set $n_1 = n$ and summed over n_2 to obtain the reduced density operator for mode 1. We proceed to solve equation (4.8) by the method of Laplace transforms. The Laplace transform of equation (4.8) is

$$\bar{\rho}_n(s) - \frac{n(N_0 + n)}{[s + (n+1)(N_0 + n + 1)]}\bar{\rho}_{n-1}(s) = \frac{\rho_n(0)}{[s + (n+1)(N_0 + n + 1)]} \tag{4.9}$$

where

$$\bar{\rho}_n(s) = \int_0^\infty e^{-s\tau}\rho_n(\tau)\,d\tau. \tag{4.10}$$

The solution of equation (4.9) is readily determined to be

$$\bar{\rho}_n(s) = n!(N_0 + n)! \sum_{m=0}^{n} \rho_n(0)\left(m!(N_0 + m)! \prod_{q=m}^{n} [s + (q+1)(N_0 + q + 1)]\right)^{-1}. \tag{4.11}$$

Inversion of equation (4.11) yields

$$\rho_n(\tau) = n!(N_0 + n)! \sum_{m=0}^{n} \frac{\rho_m(0)}{m!(N_0 + m)!} \sum_{j=m}^{n} A_{jm}^n \exp[-(j+1)(N_0 + j + 1)\tau]. \tag{4.12}$$

where

$$A_{jm}^n = \left(\prod_{\substack{i=m \\ i \neq j}}^{n} (i - j)(N_0 + 2 + j + i)\right)^{-1}.$$

Recalling the initial condition $\rho_m(0) = \delta_{m,n_1^0}$ we obtain the following expression for the distribution of photons in mode 1:

$$\rho_n(\tau) = \begin{cases} \dfrac{n!(N_0 + n)!}{n_1^0!\, n_2^0!} \displaystyle\sum_{j=n_1^0}^{n} A_{jn_1^0}^n \exp[-(j+1)(N_0 + j + 1)\tau], & n \geqslant n_1^0 \\ 0, \qquad n < n_1^0. \end{cases} \tag{4.13}$$

An expression for the distribution of photons in mode 2 may be derived in a like manner. An initial number state for the photon field is not however a realistic description for the light fields encountered in optics which are more realistically represented by a distribution in photon number.

If initially the photon distributions in modes 1 and 2 are given by $P_1(n_1^0)$ and $P_2(n_2^0)$ respectively then the photon distribution in mode 1 at time t is given by

$$\rho_n(\tau) = \sum_{n_1^0=0}^{\infty} \sum_{n_2^0=0}^{\infty} P_1(n_1^0)P_2(n_2^0)\rho_n(n_1^0, n_2^0, \tau) \tag{4.14}$$

where we have denoted the expression for $\rho_n(\tau)$ in equation (4.13) as $\rho_n(n_1^0, n_2^0, \tau)$.

For a field initially in a coherent state such as an ideal laser field $P(n^0)$ is given by a Poisson distribution whereas for a field initially in a chaotic state $P(n^0)$ is given by a Bose–Einstein distribution (Glauber 1963a, b).

The time evolutions of the mean number and variance of the photon distribution for an initial vacuum state are shown in figures 1 and 2 respectively. On comparison of the single- and two-mode cases one sees a much more rapid increase in both mean number and variance of the photon distribution for two-photon emission into a single mode. The noise of course arises from spontaneous emission. This is amplified by stimulated emission, the amplification being greater if all the photons are concentrated in a single mode.

5. Two-photon absorption

5.1. Single-mode case

We consider the case of two-photon absorption from a single mode. The atomic system is considered to be at a low temperature where the atoms are all principally in their ground state, thus $\beta_1 = 1, \beta_2 = 0$. The master equation may be deduced from the general equation (3.5) yielding

$$\frac{\partial\rho}{\partial t} = k([aa\rho, a^\dagger a^\dagger] + [aa, \rho a^\dagger a^\dagger]) \tag{5.1}$$

which leads to the following equation for the diagonal matrix elements of ρ:

$$\frac{\partial\rho_n}{\partial\tau} = (n+1)(n+2)\rho_{n+2} - (n-1)n\rho_n \tag{5.2}$$

where $\tau = 2kt$. To solve this equation we introduce the generating function $F(x, t)$ defined by

$$F(x, \tau) = \sum_{n=0}^{\infty} \rho_n(\tau)x^n, \qquad \tau \geqslant 0 \text{ and } |x| \leqslant 1. \tag{5.3}$$

In terms of this generating function equation (5.2) may be written

$$\frac{\partial F}{\partial\tau} = (1-x^2)\frac{\partial^2 F}{\partial x^2}. \tag{5.4}$$

A general solution to this equation may be obtained using a separation of variables technique (McQuarrie 1967) yielding

$$F(x, \tau) = \sum_{n=0}^{\infty} A_n C_n^{-1/2}(x) \exp[-n(n-1)\tau], \tag{5.5}$$

where $C_n^{-1/2}(x)$ is a Gegenbauer polynomial.

Agarwal (1970) has obtained such a series solution for this problem but did not explicitly evaluate the coefficients A_n. We proceed here to evaluate the coefficients A_n from the initial condition $F(x, 0)$. By differentiating the initial condition with respect to x and using the following properties of the Gegenbauer and Legendre polynomials:

$$\frac{dC_n^{-1/2}(x)}{dx} = -C_{n-1}^{1/2}(x) = -P_{n-1}(x) \qquad (n \geqslant 1) \tag{5.6}$$

$$\int_{-1}^{1} x^\lambda P_n(x)\, dx = 0 \qquad (\lambda < n) \tag{5.7}$$

$$\int_0^1 x^\lambda P_n(x)\, dx = \frac{\sqrt{\pi}\Gamma(\lambda+1)}{2^{\lambda+1}\Gamma(1+\frac{1}{2}(\lambda-n))\Gamma(\frac{1}{2}(\lambda+n+3))} \tag{5.8}$$

together with the orthogonality of the Legendre polynomials we obtain

$$A_{2n+1} = -\sqrt{\pi}(4n+1)\sum_{l=n}^{\infty} \rho_{2l+1}(0)\frac{(2l+1)!}{2^{2l+1}(l-n)!\Gamma(n+l+\frac{3}{2})}, \qquad n = 0, 1, \ldots \tag{5.9}$$

$$A_{2n} = -\sqrt{\pi}(4n-1)\sum_{l=n}^{\infty} \rho_{2l}(0)\frac{(2l)!}{2^{2l}(l-n)!\Gamma(n+l-\frac{1}{2})}, \qquad n = 1, 2, \ldots . \tag{5.10}$$

This determines $F(x, 0)$ apart from a constant which is defined to be zero by setting $A_0 = 1$. The equations (5.5), (5.9) and (5.10) then completely define $F(x, \tau)$. Initial coherent or chaotic states of the photon field are accounted for by substituting the appropriate distribution directly in equations (5.9) and (5.10).

The mean number and second factorial moment of the photon distribution may be obtained directly from $F(x, \tau)$ using the relations

$$\langle n \rangle = \left(\frac{\partial F}{\partial x}\right)_{x=1} \tag{5.11}$$

$$\langle n(n-1) \rangle = \left(\frac{\partial^2 F}{\partial x^2}\right)_{x=1}. \tag{5.12}$$

This yields

$$\langle n \rangle = -\sum_{n=1}^{\infty} A_n \exp[-n(n-1)\tau] \tag{5.13}$$

$$\langle n(n-1) \rangle = -\tfrac{1}{2}\sum_{n=2}^{\infty} n(n-1)A_n \exp[-n(n-1)\tau]. \tag{5.14}$$

The time evolution of the mean number of photons for an initial number state is shown in figure 3. It is well known that the two-photon absorption rate from chaotic light exceeds (by a factor of two for short times) that from coherent light (Teich and Wolga 1966).

5.2. Two-mode case

For two-photon absorption from two modes with frequencies ω_1 and ω_2 the master equation which follows from equation (3.5) is

$$\frac{\partial \rho}{\partial t} = k([a_1 a_2 \rho, a_1^\dagger a_2^\dagger] + [a_1 a_2, \rho a_1^\dagger a_2^\dagger]) \tag{5.15}$$

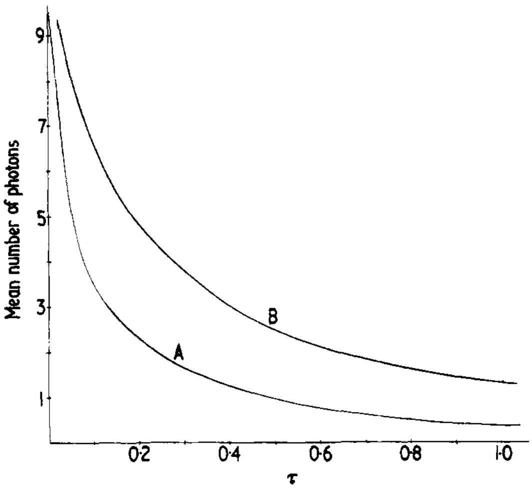

Figure 3. Two-photon absorption—initial number state. Graph of the mean number of phonons $\bar{n}$ against τ for absorption from a single mode and two modes for $\langle n(0)\rangle = 10$: A, single mode; B, two modes.

which leads to the following equation for the diagonal matrix elements of ρ:

$$\frac{\partial \rho_{n_1,n_2}}{\partial \tau} = (n_1+1)(n_2+1)\rho_{n_1+1,n_2+1} - n_1 n_2 \rho_{n_1,n_2}. \tag{5.16}$$

To facilitate the solution of equation (5.16) we assume that at $t = 0$ there is a definite number of photons n_1^0, n_2^0 present in each mode. The difference in photon number $N_0 = n_2^0 - n_1^0$ is a constant of the motion. Equation (5.2) may then readily be transformed into an equation for the reduced density operator for mode 1:

$$\frac{\partial \rho_n}{\partial \tau} = (n+1)(N_0+n+1)\rho_{n+1} - n(N_0+n)\rho_n \tag{5.17}$$

where we have set $n_1 = n$. Again this equation proves tractable to solution by the generating function technique. Multiplying equation (5.17) by x^n and summing over n gives the following partial differential equation for the generating function $F(x, \tau)$:

$$\frac{\partial F}{\partial \tau} = x(1-x)\frac{\partial^2 F}{\partial x^2} + (N_0+1)(1-x)\frac{\partial F}{\partial x}. \tag{5.18}$$

A general solution of this equation may be obtained using a separation of variables technique (McQuarrie 1967) with the result

$$F(x, \tau) = \sum_{n=0}^{\infty} A_n J_n(N_0, N_0+1, x)\exp[-n(N_0+n)\tau] \tag{5.19}$$

where J_n is a Jacobi polynomial.

The A_n may be calculated by differentiating the initial condition $F(x, 0)$ with respect to x. With the help of the relation (Abramowitz and Stegun 1965)

$$\frac{d}{dx}J_n(p, q, x) = \frac{-n(n+p)}{q}J_{n-1}(p+2, q+1, x) \tag{5.20}$$

and the orthogonality relation

$$\int_0^1 x^{q-1}(1-x)^{p-q}J_n(p,q,x)J_m(p,q,x)\,\mathrm{d}x = \delta_{mn}\frac{n!(\Gamma(q))^2\Gamma(n+p-q+1)}{(2n+p)\Gamma(n+p)\Gamma(n+q)} \tag{5.21}$$

where $\Gamma(x)$ is the gamma function, together with the initial condition $\rho_l(0) = \delta_{l,n_1^0}$ we find

$$A_n = \begin{cases} \dfrac{(-1)^n(2n+N_0)(n+N_0-1)!n_1^0!(n_1^0+N_0)!}{n!N_0!(n_1^0-n)!(n_1^0+N_0+n)!}, & n \leqslant n_1^0 \\ 0, & n > n_1^0. \end{cases} \tag{5.22}$$

Setting $A_0 = 1$, the equations (5.19) and (5.22) then completely determine $F(x, \tau)$ and hence the ρ_n. The mean number and higher factorial moments may be obtained directly from $F(x, \tau)$ using the relations (5.11) and (5.12). This yields

$$\langle n\rangle = \sum_{n=1}^{n_1^0} \frac{(2n+N_0)n_1^0!(n_1^0+N_0)!}{(n_1^0-n)!(n_1^0+N_0+n)!}\exp[-n(n+N_0)\tau]. \tag{5.23}$$

$$\langle n(n-1)\rangle = \sum_{n=2}^{n_1^0} \frac{(n-1)(n+N_0+1)(2n+N_0)(n_1^0)!(n_1^0+N_0)!}{(n_1^0-n)!(n_1^0-N_0+n)!}\exp[-n(n+N_0)\tau]. \tag{5.24}$$

The time evolution of the mean number of photons for both modes initially in number states is shown in figure 3. On comparison of the one- and two-mode results it is seen that a higher rate of absorption occurs from a single-mode source. It is clear that greater efficiency may be achieved in multi-photon absorption experiments using a single-mode rather than a multi-mode light source. This effect becomes increasingly marked the higher the order of the multi-photon process.

When the field mode at one frequency is considerably more intense than the other mode the two-photon transition changes to a 'pseudo-one-photon' transition. That is when $N_0 = n_2^0 - n_1^0$ is very large only the first term in the expansions (equations (5.23) and (5.24)) is necessary and the expressions for the mean and second factorial moment reduce to

$$\langle n\rangle = n_1^0\,\mathrm{e}^{-n_2^0\tau} \tag{5.25}$$

$$\langle n(n-1)\rangle = n_1^0(n_1^0-1)\,\mathrm{e}^{-2n_2^0\tau} \tag{5.26}$$

which correspond to the expressions obtained for a one-photon absorption process.

For initial photon distributions in modes 1 and 2 given by $P_1(n_1^0)$ and $P_2(n_2^0)$ the photon distribution at time t is obtained by the averaging procedure described by equation (4.14). For example a tunable two-photon absorption experiment using a laser mode with frequency ω_1 and a tunable thermal source in mode 2 would require a Poisson distribution for $P_1(n_1^0)$ and a Bose–Einstein distribution for $P_2(n_2^0)$.

6. The Raman effect

The Raman effect is also described by the general master equation (3.5). In the Raman effect an incident laser photon at frequency ω_L is annihilated and either a Stokes photon with frequency $\omega_S = \omega_L - \Omega$ or an anti-Stokes photon with frequency $\omega_{aS} = \omega_L + \Omega$ is emitted. The Stokes process is accompanied by an atomic excitation with energy $\hbar\Omega$ and the anti-Stokes process is accompanied by a corresponding de-excitation of the atoms. A similar master equation involving the Raman scattering from phonons has been considered by Walls (1973). Walls gives solutions for both the density operator of the Stokes field alone and the coupled Stokes, anti-Stokes density operator. However, in his analysis the parametric approximation is made for the laser pump field and hence his results do not include pump depletion.

Here we wish to present a solution to the nonlinear problem, thus including laser depletion. We shall, however, limit our considerations to the Stokes effect alone, neglecting anti-Stokes and higher-order Stokes generation.

The master equation for the density operator of the coupled Stokes laser field may be obtained from the general master equation (3.5) with the substitution $O^{(2)} = a_L a_S^\dagger$ where a_L, a_S are the annihilation operators for the laser pump and Stokes modes respectively. This substitution yields

$$\frac{\partial \rho}{\partial t} = k\beta_1([a_L a_S^\dagger \rho, a_L^\dagger a_S] + [a_L a_S^\dagger, \rho a_L^\dagger a_S]) - k\beta_2([a_L a_S^\dagger, a_L^\dagger a_S \rho] + [\rho a_L a_S^\dagger, a_L^\dagger a_S]) \tag{6.1}$$

where we have set $k^{(1,1)}$ defined by equation (3.6) equal to k. We further consider the temperature of the medium to be low so that $\beta_1 \simeq 1$, $\beta_2 \simeq 0$. This justifies our neglect of the anti-Stokes production. The equation of motion for the diagonal matrix elements of ρ is

$$\frac{\partial \rho_{n_L, n_S}}{\partial \tau} = n_S(n_L+1)\rho_{n_L+1, n_S-1} - n_L(n_S+1)\rho_{n_L, n_S} \tag{6.2}$$

where $\tau = kt$. We note that $n_L(t) + n_S(t)$ is a constant of the motion, hence if initially n_L and n_S are well defined with $n_L^0 + n_S^0 = N_0$ say at $t = 0$, then $n_L(t) + n_S(t) = N_0$ for all time. Using this property we may reduce equation (6.2) to an equation of motion for the density operator of the Stokes field alone

$$\frac{\partial \rho_n}{\partial \tau} = n(N_0 - n + 1)\rho_{n-1} - (n+1)(N_0 - n)\rho_n \tag{6.3}$$

where we have set $n_S = n$. Taking the Laplace transform of equation (6.3) we obtain the following solution for $\bar{\rho}_n(s)$, the Laplace transform of $\rho_n(\tau)$:

$$\bar{\rho}_n(s) = \begin{cases} \dfrac{n!}{(N_0-n)!} \displaystyle\sum_{m=0}^{n} \dfrac{(N_0-m)!}{m!} \dfrac{\rho_m(0)}{\prod_{j=m}^{n} [s + (j+1)(N_0 - j)]}, & n \leqslant N_0 \\ 0, \quad n > N_0. \end{cases} \tag{6.4}$$

The inverse transform for $n \leqslant N_0/2$ is

$$\rho_n(\tau) = \frac{n! N_0!}{(N_0 - n)!} \sum_{j=n_S^0}^{n} A_{nj} \exp[-(j+1)(N_0 - j)\tau] \tag{6.5}$$

where

$$A_{nj} = \left(\prod_{\substack{l=0 \\ l \neq j}}^{n} (j-l)[(j+l+1) - N_0] \right)^{-1}.$$

For $n > N_0/2$ the denominator in equation (6.5) contains repeated factors so that the inverse transforms will involve convolutions. For example for N_0 even

$$\rho_n(\tau) = 2\sum_{j=i=N_0-n}^{N_0/2}\sum^{j-1} B_{ni}B_{nj}\frac{\exp[-(i+1)(N_0-i)\tau]-\exp[-(j+1)(N_0-j)\tau]}{(j+1)(N_0-j)-(i+1)(N_0-i)} - \sum_{j=N_0-n}^{N_0/2} B_{nj}^2\exp[-(j+1)(N_0-j)\tau] \tag{6.6}$$

where

$$B_{nj} = \left(\prod_{\substack{l=N_0-n\\ l\neq j}}^{N_0/2} (j-l)[(j+l+1)-N_0]\right)^{-1}.$$

The initial photon distribution of the pumping field may be taken into account by a similar averaging procedure to that described by equation (4.14).

The time evolution of the mean number of Stokes photons for an initially coherent laser pump is shown in figure 4. Comparison is made with the predictions of the approximation solution (see equation (7.10)) and the solution deduced using the parametric approximation.

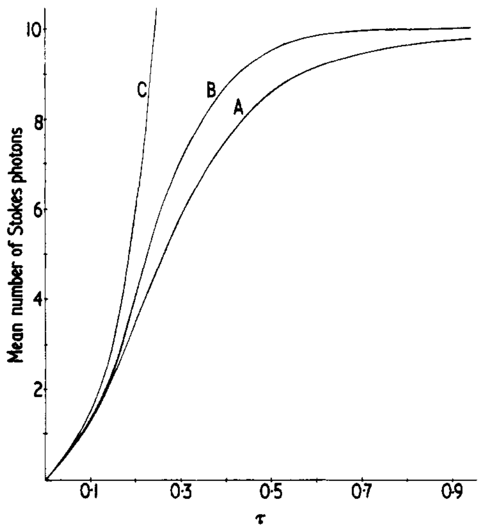

Figure 4. Raman effect—initial coherent laser pump. Graph of the mean number of Stokes photons against τ for $\langle n_L(0)\rangle = 10$, $\langle n_S(0)\rangle = 0$: A, exact solution; B, approximate solution; C, parametric approximation.

7. Approximate methods of solution

Instead of attempting to solve the master equation directly one may first derive the equations of motion for the mean number and higher-order moments of the photon distribution. These differential equations are then amenable to certain approximate methods of solution as we shall illustrate in the examples chosen below.

7.1. Two-photon absorption—single mode

The equations of motion for the mean and second-order moment of the photon distribution in the case of two-photon absorption from a single mode derived directly from equation (5.2) are

$$\frac{d\langle n\rangle}{d\tau} = -2\langle n^2\rangle + 2\langle n\rangle, \tag{7.1}$$

$$\frac{d\langle n^2\rangle}{d\tau} = -2\langle n^3\rangle + 4\langle n^2\rangle - 2\langle n\rangle. \tag{7.2}$$

These equations form part of an infinitely coupled set. They may, however, be solved approximately in the limit where the variance of the photon distribution is very small in comparison with the mean. That is, we ignore fluctuations and set $\langle n^2\rangle \simeq \langle n\rangle^2$ in equation (7.1), and $\langle n^3\rangle \simeq \langle n\rangle\langle n^2\rangle$ in equation (7.2). The approximate solutions to equations (7.1) and (7.2) are then

$$\langle n(\tau)\rangle \simeq [\langle n(0)\rangle + \langle n(0)\rangle(1-\langle n(0)\rangle)\,e^{\tau}]^{-1}\langle n(0)\rangle \tag{7.3}$$

$$\langle n^2(\tau)\rangle \simeq \langle n(\tau)\rangle^2\left(1+\frac{2}{3}\frac{1-\langle n(0)\rangle}{\langle n(0)\rangle}e^{-\tau}+\frac{\langle n^2(0)\rangle-\frac{1}{3}\langle n(0)\rangle(\langle n(0)\rangle+2)}{\langle n(0)\rangle^2}e^{\tau}\right). \tag{7.4}$$

From the above results we may estimate the variance in the photon distribution for various initial states.

(i) For an initial number state n_0 the coefficient of variation is

$$C_v = \frac{\sqrt{\text{var}}}{\langle n\rangle} = \left(\frac{2}{3}\frac{(n_0-1)}{n_0}(e^{2\tau}-e^{-\tau})\right)^{1/2}. \tag{7.5}$$

(ii) For an initially coherent state with a mean photon number n_0 the coefficient of variation is

$$C_v = \left(\frac{2}{3}\frac{1}{n_0}[(1-n_0)e^{-\tau}+\tfrac{1}{2}(2n_0+1)e^{2\tau}]\right)^{1/2}. \tag{7.6}$$

7.2. Two-photon absorption—two modes

In the case of two-photon absorption from two different modes the equation of motion for the mean number of photons in mode one may be derived directly from equation (5.17) with the result

$$\frac{d\langle n_1\rangle}{d\tau} = -(n_2^0-n_1^0)\langle n_1\rangle - \langle n_1^2\rangle. \tag{7.7}$$

Again we ignore the variance in the photon number distribution and approximate $\langle n_1^2\rangle \simeq \langle n_1\rangle^2$. The solution to equation (7.7) may then readily be shown to be

$$\langle n_1(\tau)\rangle = \begin{cases} \dfrac{n_1^0(n_2^0-n_1^0)}{n_2^0\exp[-(n_2^0-n_1^0)\tau]-n_1^0} & n_1^0 \neq n_2^0 \\ \dfrac{n_1^0}{n_1^0\tau+1} & n_1^0 = n_2^0 \end{cases} \tag{7.8}$$

which is in exact agreement with the classical result obtained by T P McLean (1963, unpublished lecture notes). This is not unexpected since spontaneous emission which is not included in a classical analysis plays no role in a pure absorption process.

7.3. Two-photon emission—single mode

The time evolution of the mean photon number in two-photon emission into a single mode may be derived from equation (4.13), and is:

$$\frac{\mathrm{d}\langle n\rangle}{\mathrm{d}\tau} = 2\langle n^2\rangle + 6\langle n\rangle + 4. \tag{7.9}$$

Employing the approximation $\langle n^2\rangle \simeq \langle n\rangle^2$, the solution to equation (7.9) is found to be

$$\langle n(\tau)\rangle = \frac{2(n_0+1)\,\mathrm{e}^{2\tau} - (n_0+2)}{(n_0+2) - (n_0+1)\,\mathrm{e}^{2\tau}} \tag{7.10}$$

where n_0 is the initial photon number. This expression differs from what one would obtain by a purely classical calculation since the onset of radiation by spontaneous emission is included explicitly.

7.4. The Raman effect

The approximate equation of motion for the mean number of Stokes photons derived from equation (6.2) by neglecting the variance in the photon number is

$$\frac{\mathrm{d}n_{\mathrm{S}}}{\mathrm{d}\tau} = n_{\mathrm{L}}(n_{\mathrm{S}}+1) \tag{7.11}$$

where n_{L} and n_{S} are the mean number of laser and Stokes photons respectively. The solution to this equation is

$$n_{\mathrm{S}}(\tau) = \frac{(n_{\mathrm{S}}^0+1)(n_{\mathrm{S}}^0+n_{\mathrm{L}}^0) - n_{\mathrm{L}}^0 \exp[-(n_{\mathrm{L}}^0+n_{\mathrm{S}}^0+1)\tau]}{(n_{\mathrm{S}}^0+1) + n_{\mathrm{L}}^0 \exp[-(n_{\mathrm{L}}^0+n_{\mathrm{S}}^0+1)\tau]}. \tag{7.12}$$

We note that this solution differs significantly from the classical results obtained by McLean (1963, unpublished) and Bloembergen (1965) since it allows a build-up of Stokes radiation directly from the vacuum by the amplification of spontaneous emission.

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Note added in proof. A recent discussion of photon statistical properties in nonlinear optics has been given by Loudon (1973) who also obtains some of the approximate solutions reported in § 7.

References

Abramowitz M and Stegun I A 1965 *Handbook of Mathematical Functions* (New York: Dover)
Agarwal G S 1970 *Phys. Rev.* A **1** 1445–59
—— 1973 *Progress in Optics* vol 11, ed E Wolf (Amsterdam: North Holland)
Bloembergen N 1965 *Non Linear Optics* (New York: Benjamin)
Glauber R J 1963a *Phys. Rev.* **130** 2529–39
—— *Phys. Rev.* **131** 2766–88
Haken H 1970 *Handb. Phys.* vol XXV/2C (Heidelberg: Springer Verlag)
Lambropolous P 1967 *Phys. Rev.* **156** 286–97
Loudon R 1973 *The Quantum Theory of Light* (Oxford: Clarendon)
McQuarrie D A 1967 *J. appl. Prob.* **4** 413–78
Shen Y R 1967 *Phys. Rev.* **155** 921–31
Teich M C and Wolga G J 1966 *Phys. Rev. Lett.* **16** 625–7
Walls D F 1971 *J. Phys.* A: *Gen. Phys.* **4** 813–26
—— 1973 *J. Phys.* A: *Math., Nucl. Gen.* **6** 496–505