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Quantum theory of multiphoton lasers I. Systems in detailed balance

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Abstract. A quantum-mechanical model for a multiphoton laser is proposed by an obvious generalization of the usual one-photon laser model. In this first paper we restrict our considerations to a class of lasers which preserve detailed balance. This condition is ensured by assuming a multiphoton loss mechanism for the laser light of the same order as the multiphoton gain from the stimulated emission process. Solutions for the stationary density operator of the light field are obtained by a straightforward generalization of the techniques of Scully and Lamb. The multiphoton laser around threshold exhibits behaviour analogous to a second-order phase transition. A non-thermal distribution of multiphoton laser light is achieved above threshold.

1. Introduction

The phenomenon of laser action involving a one-photon emission in a single atomic decay has been satisfactorily explained by quantum theory. Review articles on this topic describing alternative but basically equivalent approaches are given by Haken (1970), Lax (1968) and Risken (1970). Multiphoton absorption experiments are now common and direct observation of two-photon spontaneous emission has been reported (Lipeles *et al* 1965). It is natural, therefore, to consider the possibility of achieving laser action involving the stimulated emission of two or more photons in a single atomic decay.

Analyses of the photon statistics of a two-photon emission process (Lambropolous 1967, McNeil and Walls 1974) have shown that the two-photon emission process is considerably more noisy than a single-photon emission process. This may tend to suggest that such an amplifier does not have the coherence properties of a laser amplifier since it tends to increase the statistical fluctuations of the field. However, these analyses did not include pumping and loss mechanisms and hence no conclusions about the action of a multiphoton laser may be drawn from them. It is clear, however, from general considerations of non-equilibrium thermodynamics (Glansdorff and Prigogine 1971) that for high enough values of the driving parameter a non-linear dissipative system will undergo a transition to a more highly ordered state. In the laser case the more highly ordered state is characterized by a narrower photon distribution than the thermal distribution.

It is our aim to present a fully quantum-mechanical model of a multiphoton laser starting from a microscopic Hamiltonian. In this paper we confine our attention to a restricted class of multiphoton lasers which preserve detailed balance.

2. Model and analytical approach

The model for the multiphoton laser is an obvious generalization of the usual one-photon laser model. We consider M electromagnetic field modes in a resonant cavity interacting with a system of N two-level atoms. These atoms are presumed to have an appreciable M-photon dipole matrix element.

The field modes are described by the boson creation and annihilation operators $b_j^\dagger$ and b_j which obey the commutation relations

$$[b_j, b_k^\dagger] = \delta_{jk}. \tag{2.1}$$

The atomic system is described by the operators S^-, S^+ and S_z which are analogous to the spin operators and obey in particular the commutation relations

$$[S^-, S^+] = 2S_z. \tag{2.2}$$

The free Hamiltonian is given by

$$H_0 = \hbar \sum_{j=1}^{M} \omega_j b_j^\dagger b_j + \tfrac{1}{2}\hbar\Omega S_z. \tag{2.3}$$

The sum frequency of the photons in the M field modes is assumed to be in resonance with the frequency separation of the atomic levels (ie $\Sigma_{j=1}^{M} \omega_j = \Omega$).

The atom–field interaction for an M-photon emission process may be described by the effective Hamiltonian (Shen 1967, Walls 1971):

$$H_1 = \hbar g^{(M)}\left(S^- \prod_{k=1}^{M} b_k^\dagger + S^+ \prod_{k=1}^{M} b_k\right). \tag{2.4}$$

The coupling constant $g^{(M)}$ is proportional to $x^{(M)}$, the dipole matrix element for an M-photon transition between the two atomic states.

The pumping and loss mechanisms for the atoms are included by coupling the atoms individually to thermal reservoirs. The loss mechanism for the laser light is principally by transmission through the end mirror. Here we restrict our attention to a laser cavity where the end mirror is partially transmitting at the sum frequency $\Sigma_{i=1}^{M} \omega_i$ but totally reflecting at the individual photon frequencies. This M-photon loss mechanism is described by the Hamiltonian

$$H_{\mathrm{FR}} = \Gamma_{\mathrm{R}}^\dagger \prod_{i=1}^{M} b_i + \Gamma_{\mathrm{R}} \prod_{i=1}^{M} b_i^\dagger \tag{2.5}$$

where the Γ_{R} represent the reservoir operators.

The restriction to an M-photon loss mechanism is necessary to retain the property of detailed balance. (This is lifted in the following paper.)

It is possible, using standard methods developed for the one-photon laser, to derive an equation of motion for the density operator or distribution function of the light field. Here we adopt the method of Scully and Lamb (1967) and derive an equation of motion for $\rho_{n,n}$ the matrix elements of the reduced density operator for the field in Fock space. We consider separately the cases of M-photon emission into a single mode and M-photon emission into M modes.

3. Single-mode M-photon laser

A straightforward application of the method of Scully and Lamb to the model described in § 2 yields the following master equation for the M-photon single-mode laser:

$$\frac{\mathrm{d}\rho_{n,n}}{\mathrm{d}t} = -A_M \prod_{k=1}^{M}(n+k)\left(1+\frac{B_M}{A_M}\prod_{k=1}^{M}(n+k)\right)^{-1}\rho_{n,n} + A_M\prod_{j=0}^{M-1}(n-j)\left(1+\frac{B_M}{A_M}\prod_{k=0}^{M-1}(n-k)\right)^{-1}\rho_{n-M,n-M} + C_M\prod_{k=1}^{M}(n+k)\rho_{n+M,n+M} - C_M\prod_{j=0}^{M-1}(n-j)\rho_{n,n} \tag{3.1}$$

where A_M, B_M and C_M are the usual gain, non-linear and loss parameters defined by Scully and Lamb, except that A_M and B_M are proportional to $(g^{(M)})^2$ that is, to the square of the coupling constant for an M-photon dipole transition.

The steady-state solution of equation (3.1) may be obtained by invoking detailed balance considerations. This reduces the second-order difference equations to two equivalent systems of first-order difference equation of the form

$$A_M\prod_{k=1}^{M}(n+k)\left(1+\frac{B_M}{A_M}\prod_{k=1}^{M}(n+k)\right)^{-1}\rho_{n,n} - C_M\prod_{k=1}^{M}(n+k)\rho_{n+M,n+M} = 0 \tag{3.2}$$

Assuming that the initial photon distribution depends only on $\rho_{mM,mM}$ (where m is an integer), the solution to equation (3.2) is clearly

$$\left.\begin{aligned}\rho_{nM,nM} &= \rho_{0,0}\prod_{k=1}^{n}\frac{A_M/C_M}{1+(B_M/A_M)\prod_{j=0}^{N-1}(kM-j)} \\ \rho_{nM+j,nM+j} &= 0 \qquad j = 1,2,\ldots,M-1\end{aligned}\right\} n = 1,2,\ldots. \tag{3.3}$$

This distribution exhibits a similar behaviour to that obtained for the one-photon laser with the difference that only the multiples of M photon numbers are present. This is a consequence of the model chosen where the photons are produced and lost only in multiples of M.

There is a threshold $A_M \simeq C_M$ below which the distribution rapidly falls away from zero and above which the distribution is peaked away from zero.

Below threshold we find

$$\rho_{nM,nM} \simeq \rho_{0,0}\left[\left(\frac{A_M}{C_M}\right)^{1/M}\right]^{nM} \tag{3.4}$$

which is a geometric-like distribution (in the variable nM) with parameter $(A_M/C_M)^{1/M}$.

Well above threshold ($A_M/C_M \gg 1$), we find

$$\rho_{nM,nM} \simeq \rho_{0,0}\frac{[(A_M^2/B_MC_M)^{1/M}]^{nM}}{(nM)!} \tag{3.5}$$

which is a Poisson-like distribution (in the variable nM) with parameter $(A_M^2/B_MC_M)^{1/M}$.

Figure 1 compares the photon distribution given by equation (3.3) for a two-photon laser with a one-photon laser distribution, both at 20% above threshold, with the non-linear parameters chosen to give the same mean. It is interesting to observe that the

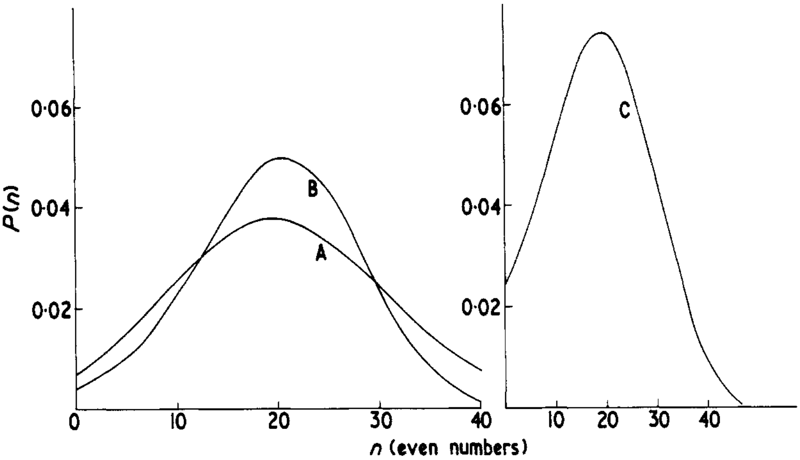

Figure 1. Photon number distributions 20% above threshold (mean number 20) for: A, one-photon laser; B, two-photon two-mode laser; C, two-photon single-mode laser.

two-photon laser number distribution has approximately the same variance as the one-photon laser number distribution. Why this should be is not immediately obvious, since the two-photon emission process is noisier than the one-photon emission process. These results show that the two-photon damping mechanism is sufficient to inhibit the noise, and allow the onset of lasing.

4. Multimode M-photon laser

We now consider the case where the M photons are emitted into M different modes. This may be achieved in a continuous-wave laser by suitable tuning of the cavity or in a pulsed laser by choosing the inverse frequency of the triggering pulse to be other than an integral multiple of the frequency of the atomic transition.

The master equation for the matrix elements $\rho_{\{n_i\}} = \langle\{n_i\}|\rho|\{n_i\}\rangle$ of the field density operator is readily shown to be

$$\begin{aligned}\frac{\mathrm{d}\rho_{(\{n_i\})}}{\mathrm{d}t} = & -A_M \prod_{i=1}^{M} (n_i+1)\left(1+\frac{B_M}{A_M}\prod_{i=1}^{M}(n_i+1)\right)^{-1}\rho_{\{n_i\}} \\ & +A_M \prod_{i=1}^{M} n_i\left(1+\frac{B_M}{A_M}\prod_{i=1}^{M} n_i\right)^{-1}\rho_{\{n_i-1\}} \\ & +C_M \prod_{i=1}^{M}(n_i+1)\rho_{\{n_i+1\}} - C_M \prod_{i=1}^{M} n_i\rho_{\{n_i\}}. \end{aligned} \tag{4.1}$$

The stationary solution to this equation may again be obtained employing considerations of detailed balance. Since the photons are produced and lost in multiples of M, we have the independent constants of the motion $n_1 - n_k = 0$. The stationary solution of equation (4.1) is found to be

$$\rho_{\{n_i\}} = \prod_{k=1}^{M} \delta_{n_1,n_k}\rho_{n,n} \tag{4.2}$$

where

$$\rho_{n,n} = \rho_{0,0} \prod_{k=1}^{n} \frac{A_M/C_M}{1+(B_M/A_M)k^M}. \tag{4.3}$$

This model of a multimode M-photon laser again has a threshold at $A_M \simeq C_M$. Below threshold

$$\rho_{n,n} \simeq \rho_{0,0}\left(\frac{A_M}{C_M}\right)^n \tag{4.4}$$

that is, each mode has a geometric distribution with parameter A_M/C_M.

Well above threshold ($A_M/C_M \gg 1$)

$$\rho_{n,n} \simeq \frac{(A_M^2/B_M C_M)^n}{(n!)^M} \tag{4.5}$$

which is a sharply peaked distribution.

In figure 1, the photon distribution for two-photon emission into two modes is compared with that of two-photon emission into a single mode. A broader photon number distribution results from the two-photon emission into a single mode since the noise from the spontaneous emission of the first photon is effectively amplified by the second photon. Again we find that this particular two-photon laser has a photon number distribution which is narrower than the corresponding one-photon laser distribution.

5. Macroscopic behaviour

In conclusion we shall examine the macroscopic behaviour of the system around threshold. Close to threshold we may expand the denominator in the master equations and take terms up to B_M/A_M only. We may then obtain equations of motion for the mean number of photons as follows.

(i) Single-mode case

$$\frac{d\langle n\rangle}{dt} \simeq M[\langle n^M\rangle(A_M - C_M) - \langle n^{2M}\rangle B_M] \tag{5.1}$$

(ii) M-mode case

$$\frac{d\langle n_i\rangle}{dt} \simeq \left[\left\langle \prod_{i=1}^{M} n_i \right\rangle(A_M - C_M) - \left\langle\left(\prod_{i=1}^{M} n_i\right)^2\right\rangle B_M\right]. \tag{5.2}$$

If we ignore fluctuations, that is set $\langle n^M\rangle \simeq \langle n\rangle^M$ we obtain the macroscopic equations. It is readily seen that the multiphoton laser exhibits behaviour analogous to a second-order phase transition about threshold in a like fashion to the one-photon laser (Graham and Haken 1970, De Giorgio and Scully 1970, Grossman and Ritcher 1971). The order parameters for this particular type of multiphoton laser are (i) n^M, (ii) $\Pi_{i=1}^M n_i$. The behaviour of these quantities as a function of the pumping parameter is shown in figure 2.

In summary, it has been shown that it is possible in principle to achieve non-thermal photon distributions from stimulated multiphoton emission. The distributions obtained resemble the Poisson distributions characteristic of coherent laser light; however, only

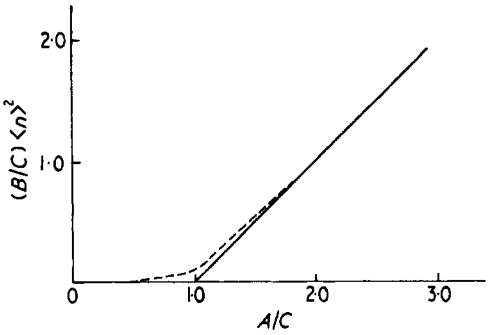

Figure 2. Mean photon number squared as a function of the pumping parameter for a two-photon single-mode laser: full curve, macroscopic; broken curve, as given by the photon number distribution.

photon numbers in multiples of M appear in the distribution. This is a consequence of the particular multiphoton laser model chosen to preserve detailed balance. A less restricted class of possible multiphoton lasers is considered in the following paper.

6. Discussion of experimental possibilities

Though at present no-one has reported achieving two-photon laser amplification, there is substantial development work proceeding to this end. One potential scheme under development at Livermore was recently reported by Carman *et al* (1974). Other potential two-photon lasers are being developed by R Byer (1974, private communication) at Stanford and by P Sorokin (1974, private communication) at IBM. These systems will be pulsed single-pass amplifiers and hence it will be necessary to generalize the foregoing calculations to a multimode analysis in order to derive information on quantities such as the pulse width.

Interest in developing such two-photon lasers is considerable because of the possibility of achieving high light intensities with these systems. This possibility arises since the strength of the coupling between the laser-active atoms and the light field is now proportional to the light intensity as against the square root of the light intensity in the one-photon laser. Thus, once one has achieved sufficient energy storage to overcome the relative weakness of the two-photon transition, one has the potential to achieve higher laser power than with the usual one-photon laser.

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