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Semiclassical stochastic description of the two-photon laser

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We consider the problem of two-photon transitions between atomic levels having the same parity. The semiclassical theory is developed from Maxwell's equations and the resulting transcendental equations for the pulse envelope solved exactly. They lead to an unstable pulse envelope if the higher-order processes are included. We then treat the laser as a birth/death process, deriving a Markovian (macroscopic) master equation for the probability function, which may be solved exactly in the steady state. By a suitable truncation procedure, this equation predicts a stable steady-state envelope, and is consistent with the microscopic quantum theory when virtual processes are neglected. A comparison is made between the macroscopic (master equation) approach and the microscopic quantum theory at the level of the moment equations. We find that, in the high-gain limit, the fluctuations predicted by the two theories are the same.

I. INTRODUCTION

In recent years, considerable progress has been made in understanding the dynamics of coherent two-photon processes. For a two-photon transition to occur, it is necessary that the atomic levels between which it occurs have the same parity, so that electric dipole transitions are not dominant and may be excluded. It is seen, then, that virtual transitions between the other atomic levels occur, giving rise to higher-order terms in the dynamic equations so that the two-level picture is not a very good approximation in this case. Two-photon absorbtion has been treated theoretically[1] using a microscopic quantum-statistical approach via a master equation. The application of the well-known adiabatic following approximation to two-photon processes has also been considered[2] and used to describe self-induced transparency and pulse amplitude modulation. In a more recent publication,[3] the propagation of a pulse through a two-photon absorbing medium has been considered by Narducci *et al.* They derive the Bloch equations for this case and construct an equation for the pulse energy density, obtaining an unstable pulse envelope (because of the inclusion of virtual processes in the theory) with a Lorentzian line shape.

In this paper, the two-photon laser is treated as a birth-death process,[4] described by an appropriately defined macroscopic probability function P, which obeys a Markovian master equation. Such a theory is phenomenological in nature and has been applied[5] to a semiclassical treatment of the single-photon laser above threshold. It has been seen in this case, that, in the coherent signal regime, the fluctuations and statistics predicted by the master equation agree with the corresponding (microscopic) Scully-Lamb results. Similar techniques have been applied[6] to the optical bistability in resonance fluorescence, a system which is known to exhibit a first-order phase transition. In this case, the macroscopic master equation approach is found to predict a transition region which is slightly displaced from the corresponding microscopic predictions. The master equation also predicts a broader probability distribution. This is traced to the fact that the corresponding Fokker-Planck equation has a nonconstant diffusion term which differs from the corresponding term obtained through a microscopic approach. The laser Fokker-Planck equation, however, admits of a constant diffusion term, so that agreement between the two approaches is expected to be much closer, as has indeed been demonstrated in Ref. 5 on the one-photon case. The birth-death approach has also been extensively applied[4] in chemical kinetics, and, in particular, is a valuable tool in dealing with chemical systems exhibiting multiple stationary states, in which there is more than one time scale of interest.[7]

In this work, we outline first, the semiclassical theory of the two-photon laser as developed by Narducci *et al.*[3] By a simple extension of their results, we obtain an exact equation for the energy density which contains the analog of higher-order virtual processes in a second quantized theory. This equation is solved exactly for the pulse shape. Further, by expanding this equation and retaining only the lower-order processes, we arrive at a deterministic equation for the energy density. From this equation, we may derive a phenomenological (macroscopic) master equation which may be solved in the steady state yielding a coherent, stable pulse envelope, described by Poisson statistics above threshold. A comparison between the macroscopic approach and a simple microscopic (second-quantized) theory (derived as a simple extension of the one-photon Scully-Lamb theory) is further made in Sec. III, using the moment equations derived in the two theories.

II. SEMICLASSICAL THEORY OF THE TWO-PHOTON LASER AND THE MASTER EQUATION

A. Semiclassical theory

In this subsection, we briefly outline the results of Narducci *et al.*[3] (adhering to their notation) and then find the exact form of the steady-state envelope. We consider the total Hamiltonian of the form

$$H=E_a|a\rangle\langle a|+E_b|b\rangle\langle b| + \sum_{j\neq a,b} E_j|j\rangle\langle j| - \vec{p}\cdot\vec{\mathcal{E}}(\vec{x},t), \qquad (1)$$

where the two-photon transition is assumed to occur between the atomic levels $|a\rangle$ and $|b\rangle$ (of the same parity) having a frequency separation $\omega_{ab} = E_a - E_b$ (in units of $\hbar$). The subscript j denotes the remaining levels which, as we shall see, contribute to the classical analog of higher-order virtual transitions. The last term in (1) represents the interaction of the atoms with the electric field, $\vec{p}$ being the atomic-dipole moment. We describe the electric field by a plane wave,

$$\vec{\mathcal{E}}(\vec{x},t)=\vec{\mathcal{E}}_0(\vec{x},t)\cos\{\omega t-kx+\varphi(x,t)\}\ , \qquad (2)$$

which obeys the well-known wave equation

$$\frac{\partial^2\mathcal{E}}{\partial x^2}+\frac{1}{c^2}\frac{\partial^2\mathcal{E}}{\partial t^2}+\mu_0\sigma\frac{\partial\mathcal{E}}{\partial t}=-\frac{1}{c^2\epsilon_0}\frac{\partial^2 P}{\partial t^2}\ . \qquad (3)$$

In this equation, P is the macroscopic polarization in the laser cavity, obtained in a self-consistent way be summing the individual polarizations of each atomic dipole (induced by the cavity field) and incorporating the result into Maxwell's equations as a source term. The polarization P may be broken up into an in-phase (with $\vec{\mathcal{E}}$) component P_c and a quadrature component P_s according to

$$P=P_c\cos(\omega t-kx+\varphi)+P_s\sin(\omega t-kx+\varphi)\ . \qquad (4)$$

Substituting (2) and (4) into (3) and equating the coefficients of the sine terms, we find in the slowly varying envelope approximation[8]

$$c\frac{\partial\mathcal{E}_0}{\partial x}-\frac{\partial\mathcal{E}_0}{\partial t}-\tfrac{1}{2}c^2\mu_0\sigma\mathcal{E}_0=\frac{\omega}{2\epsilon_0}P_s\ . \qquad (5)$$

The atom may be described by a state vector,

$$|\psi(t)\rangle=\sum_j C_j(t)e^{-iE_jt}|j\rangle+C_a(t)e^{-iE_at}|a\rangle + C_b(t)e^{-iE_bt}|b\rangle\ , \qquad (6)$$

where the atomic amplitudes C_a and C_b, have been shown[3] to satisfy Bloch equations which depend on the square of the electric field vector, rather than on its first power as in the one-photon case.[9] In terms of these amplitudes, the polarization P is found to have the form

$$P=N\left(\sum_j \mu_{ja}C_aC_j^*e^{i\omega_{ja}t}+\sum_j\mu_{jb}C_bC_j^*e^{i\omega_{jb}t}\right)\ . \qquad (7)$$

This contains terms which depend on the coupling of all the remaining atomic levels $|j\rangle$ $(j\neq a,b)$ to $|a\rangle$ and $|b\rangle$. However, if we consider only the componets of P which oscillate at the frequency of the injected signal we obtain[3]

$$P_s=-Nk_{ab}R_1\mathcal{E}_0\ , \qquad (8)$$

where N is the number of atoms and k is an atomic linewidth. We have introduced here the Bloch vector component,

$$R_1=i(C_a^*C_be^{i\alpha}-\text{c.c.})\ , \qquad (9)$$

with

$$\alpha=(2\omega-\omega_{ab})t-2kx+2\varphi\ .$$

We also introduce the Rabi frequency ω_R through the relation

$$\omega_R(2\hbar/k_{ab})(1+\gamma^2)^{-1/2}=\mathcal{E}_0^2\ , \qquad (10)$$

where $\gamma=(k_{bb}-k_{aa})/2k_{ab}$. Equation (5) then reduces to

$$\frac{\partial\omega_R}{\partial t}=-c^2\mu_0\sigma\,\omega_R+gR_1\omega_R\ , \qquad (11)$$

where $g=\omega Nk_{ab}/\epsilon_0$. We have ignored the spatial dependence of the pulse, considering only its time evolution. Equation (11) is one of the principal results of Narducci *et al.* The first term on the right-hand side is a loss term and the second term represents the gain. This equation is our starting point for the rest of this work. Throughout the rest of this section, we shall concentrate on the gain term, which is responsible for the nonlinear effects of interest.

At resonance we obtain[10]

$$R_1=\frac{R_3^e}{(1+\gamma^2)^{1/2}}\sin\int_{-\infty}^t\omega_R(\tau)\,d\tau, \qquad (12)$$

R_3^e being the equilibrium population inversion between $|a\rangle$ and $|b\rangle$. Setting $\mathcal{E}_0^2=n$ in analogy to the "photon" energy density, we obtain from (11),

$$\dot n=bn\sin\zeta\sigma'\ , \qquad (13)$$

where

$$b=gR_3^e(1+\gamma^2)^{-1/2};\quad \zeta=(k_{ab}/2\hbar)(1+\gamma^2)^{1/2}\ , \qquad (14)$$

and

$$\sigma'(t)=\int_{-\infty}^t n(\tau)\,d\tau. \qquad (15)$$

Differentiating (13) with respect to time and using (15) we obtain

$$\frac{d^2\sigma'}{dt^2}=b\,\frac{d\sigma'}{dt}\sin\zeta\sigma', \tag{16}$$

which may be integrated once to give

$$\frac{d\sigma'}{dt}=b'(1-\cos\zeta\sigma'), \tag{17}$$

where $b'=b/\zeta$. Since $\dot{\sigma}'=n$, we find from (17)

$$\zeta\sigma'=\cos^{-1}(1-n/b'), \tag{18}$$

so that Eq. (13) reduces to

$$\dot{n}=bn\sin[\cos^{-1}(1-n/b')]\,. \tag{19}$$

We shall return to this later. Let us now combine Eqs. (18) and (14). This gives

$$\dot{n}=bn(2n/b'-n^2/b'^2)^{1/2}\,. \tag{20}$$

This equation is exact and has steady states at $n=0$ and $n=2b'$. It may be solved to give

$$n=2b'(1+b^2t^2)^{-1}, \tag{21}$$

which is Lorentzian. We have thus obtained a time-dependent solution for the photon energy density n. This solution is shown in Fig. 1. The pulse is seen to be unstable, a fact that has been pointed out in Ref. 3. The instability arises because of the retention of all the higher-order processes in (11). If these processes are ignored, as is done in the next subsection, the pulse envelope will be stable.

For the purpose of comparison with a microscopic (second-quantized) theory and also in order to derive a macroscopic master equation, we reconsider Eq. (17). This may be readily integrated to give

$$\zeta\sigma'=2\cot^{-1}bt, \tag{22}$$

so that we obtain from (13)

$$\dot{n}=bn\sin(2\cot^{-1}bt)\,. \tag{23}$$

Upon making the simple change of variables $z=\cot^{-1}bt$, we may integrate (23) to obtain the particular solution,

$$n=2b'\sin^2(\cot^{-1}bt)\,. \tag{24}$$

It may be checked that this solution is equivalent to (21). Alternatively, we may consider Eq. (19). Setting $z'=\cos^{-1}(1-n/b')$, we may reduce (19) to the simple form

$$\dot{z}'=-b(1-\cos z'), \tag{25}$$

which also yields (24) as a solution.

We now return to Eq. (19) which we write as

$$\dot{n}=bn\sin(\cos^{-1}x), \tag{26}$$

where we have set $x=1-n/b'$. The arcosine may be expanded in a Taylor series to give

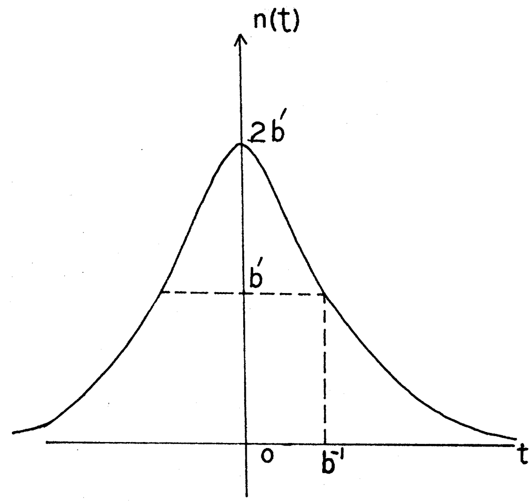

FIG. 1. Form of the steady-state pulse envelope as predicted by the semiclassical theory.

$$\cos^{-1}x=\frac{\pi}{2}-\frac{7}{6}+\frac{3}{2}\,\frac{n}{b'}-\frac{n^2}{2b'^2}+\frac{n^3}{2b'^3}\cdots\,. \tag{27}$$

Hence (26) becomes

$$\dot{n}=\frac{3}{2}\,\frac{b}{b'}\,n^2-\frac{19}{48}\,\frac{b}{b'^3}\,n^4, \tag{28}$$

where we have retained only the terms in n^2 and n^4. For small energy densities, the neglect of higher-order terms corresponds to the neglect of virtual processes in a quantum theory.[1] We shall write (28) in the form

$$\dot{n}=k_1n^2-k_2n^4\,. \tag{29}$$

In the steady state, this deterministic equation yields the solutions for the average energy density $\langle n\rangle$:

$$\langle n\rangle=0;\quad (k_1/k_2)^{1/2}, \tag{30}$$

where we find from (28),

$$\alpha=(k_1/k_2)^{1/2}=1.95b'\,. \tag{31}$$

α is thus the steady-state value of the energy density. By retaining higher-order terms in the expansion (27), we obtain a better approximation to the exact value $\alpha=2b'$ as obtained from the solutions (21) or (24). The laser exhibits a second-order phase transition at threshold[11] with order parameter $\langle n\rangle^2$. This deterministic equation (29) will be our starting point in deriving a phenomenological Markovian master equation for the two-photon laser in the absence of virtual processes. This is done in Sec. II B.

B. Master equation

The deterministic equation (29) has a gain-loss structure. The first term on the right-hand side is a nonlinear gain and the second term represents a loss. We now introduce a macroscopic probability distribution $P(n,t)$. By exploiting the analogy of the laser with an autocatalytic chemical reaction as done in the single-photon case,[5] we find the following macroscopic Markovian master equation for P:

$$\frac{dP}{dt}=\lambda_{n-2}P(n-2,t)+\mu_{n+2}P(n+2,t)-(\lambda_n+\mu_n)P(n,t). \qquad (32)$$

This equation has the familiar gain-loss structure associated with stochastic master equations.[4] The "birth" and "death" amplitudes λ_n and μ_n are given by

$$\lambda_n=k_1n(n+1),\quad \mu_n=k_2n(n-1)(n-2)(n-3), \qquad (33)$$

so that the right-hand side of (32) is the sum of a probability flow from the level $n+2$ to n and a flow from $n-2$ to n.

Let us now introduce a generating function[12] corresponding to the probability P:

$$F(s,t)=\sum_{n=0}^{\infty}s^nP(n,t), \qquad (34)$$

subject to the condition

$$F(1,t)=1, \qquad (35)$$

corresponding to probability conservation. In terms of F, the differential-difference equation (32) becomes a partial differential equation,

$$\frac{\partial F}{\partial t}=s(s^2-1)\left(2k_1\frac{\partial F}{\partial s}+k_1s\frac{\partial^2F}{\partial s^2}-k_2s\frac{\partial^4F}{\partial s^4}\right), \qquad (36)$$

which may be solved in the steady state using Laplace transforms to give

$$F(s)=\frac{\alpha s\cosh\alpha s-\sinh\alpha s}{\alpha\cosh\alpha-\sinh\alpha}, \qquad (37)$$

where we have used the condition (35) to determine the integration constant. We may now determine the mean and variance of the photon number distribution. From (34) it may be seen that[12]

$$\langle n\rangle=\left.\frac{\partial F}{\partial s}\right|_{s=1}=\frac{\alpha^2}{\alpha\coth\alpha-1}, \qquad (38)$$

where we have used (37). In the high-gain limit we find for large α

$$\langle n\rangle\to\alpha=(k_1/k_2)^{1/2}, \qquad (39)$$

in accordance with (30). The variance is given by[12]

$$\langle(\delta n)^2\rangle=\left.\frac{\partial^2F}{\partial s^2}\right|_{s=1}-\left(\frac{\partial F}{\partial s}\right)^2_{s=1}+\left.\frac{\partial F}{\partial s}\right|_{s=1}=\alpha, \qquad (40)$$

where we have, once again, used the strong-signal limit. The variance is thus Poisson and since we know $F(s)$ from (37), all the higher-order moments of the photon number may be uniquely determined in terms of $\langle n\rangle$, corresponding to a truncation of the hierarchy of moment equations at arbitrary order. This has already been done for the single-photon case[5] even near threshold.

Finally, let us return to Eqs. (35) and (37) and calculate the distribution function $P(n)$ in the steady state. Setting $s=e^{-\phi}$ we may invert (34) to write[12]

$$P(n)=\frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty}e^{n\phi}F(\phi)\,d\phi \qquad (41)$$

$$=\frac{1}{2\pi i}\left(\oint_C-\oint_\Gamma\right)e^{n\phi}F(\phi)\,d\phi, \qquad (42)$$

where C is the contour shown in Fig. 2, Γ being the arc bounded by the straight line $\phi=\gamma$ in the right half-plane. The integral over Γ vanishes if we have $F(\phi)\equiv P'(\phi)/Q(\phi)$ and P' is of lower degree in ϕ than Q. Then we have

$$P(n)=\frac{1}{2\pi i}\frac{1}{\alpha\cosh\alpha-\sinh\alpha}\times\oint_{C'}s^{-n-1}(\alpha s\cosh\alpha s-\sinh\alpha s)\,ds, \qquad (43)$$

where C' now encloses all the singularities of s. We then obtain from the residue theorem,

$$P(n)=\frac{(n!)^{-1}}{\alpha\cosh\alpha-\sinh\alpha}\frac{d^n}{ds^n}(\alpha\cosh\alpha s-\sinh\alpha s)_{s=0}, \qquad (44)$$

which may be easily evaluated to give,

$$P(n)=\frac{1}{\alpha\cosh\alpha-\sinh\alpha}\frac{\alpha^n}{n!}(n-1). \qquad (45)$$

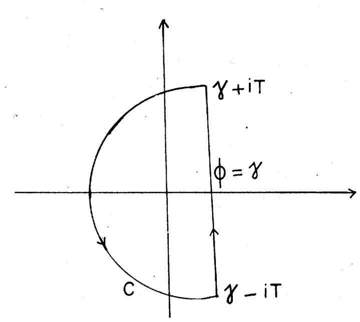

FIG. 2. Integration contour for Eq. (42). Γ is the arc of the semicircle bounded by the line $\phi=\gamma$. The entire contour is denoted by C.

For strong signals we obtain

$$\alpha\cosh\alpha - \sinh\alpha \approx \alpha e^{\alpha}, \quad (46)$$

so that

$$P(n) \to e^{-\alpha}\alpha^{n-1}/(n-1)!, \quad (47)$$

a Poisson distribution, with mean value α. It may be noted that the hyperbolic functions in (46) reach their high-gain values very rapidly above threshold, so that the laser statistics may be assumed to be coherent as soon as we are slightly above threshold.

We may now compare the results of this section with the microscopic theory given in Sec. II A. The most striking feature is that the pulse envelope is stable in the absence of higher-order terms. To see this, let us return to our original deterministic equation (29) whose time-dependent solution may be written down immediately:

$$\frac{\alpha + \langle n\rangle}{\alpha - \langle n\rangle} = K_2 \exp(2k_2\alpha^3 t), \quad (48)$$

where K_2 is an integration constant which we set equal to unity by requiring $\langle n\rangle$ to vanish for $t=0$. Equation (48) admits of a steady state $\langle n\rangle = \alpha = 2b'$ for long times which is stable. In Sec. III, we shall briefly outline a quantum microscopic approach to the same problem and show that our macroscopic results are consistent with this approach by comparing the moment equations given by the two theories, in the absence of virtual transitions.

In concluding this subsection, we note that our results may be extended to the M-photon case as long as no single-photon loss mechanism is allowed. This result has been formally written down by McNeil and Walls[1] using the M-photon extension of the microscopic quantum theory which we shall outline briefly below.

III. DISCUSSION

We have demonstrated above, that, in the absence of higher-order terms in the expansion of $\cos^{-1}(1-n/b')$, the semiclassical microscopic theory and the macroscopic approach based on a Markovian master equation agree. One may also construct a quantum (microscopic) theory in which the atoms *and* field are treated quantum mechanically, the atoms being adiabatically eliminated. Such a second-quantized theory is an extension of the one-photon Scully-Lamb theory[9] and has been formally given by McNeil and Walls.[1] Without going into the details, we give here, for the purpose of discussion, the microscopic master equation for the field density matrix ρ_{nn} in Fock space:

$$\begin{aligned}\dot\rho_{nn} = &-A_2(n+1)(n+2)\left(1-\frac{B_2}{A_2}(n+1)(n+2)\right)\rho_{nn}\\ &+A_2 n(n-1)\left(1-\frac{B_2}{A_2}n(n-1)\right)\rho_{n-2,n-2}\\ &-C_2 n(n-1)\rho_{nn} + C_2(n+1)(n+2)\rho_{n+2,n+2}. \end{aligned} \quad (49)$$

Here, A_2, C_2, B_2, are the two-photon analogs of the gain, loss, and saturation parameters defined for the one-photon case.[9] A_2 and B_2 are proportional to g^4 (g being the coupling constant), in contrast to the one-photon case where they vary as g^2. It is important to note here that, in Eq. (49), the higher-order virtual processes have been omitted. A similar procedure is followed by Lambropoulos.[1] Let us now consider the equation for the mean photon number. This is found by multiplying both sides of (49) by n and summing over n. In the large-photon-number limit the leading contributions give

$$\frac{\partial\langle n\rangle}{\partial t} = 2(A_2 - C_2)\langle n^2\rangle - 2B_2\langle n^4\rangle. \quad (50)$$

In the large-photon-number limit, we may assume factorization of the form[5,9] $\langle n^k\rangle \approx \langle n\rangle^k$. In this case, the equation has exactly the same form as the truncated semiclassical equation (29), and admits of the steady-state solution,

$$\langle n\rangle = 0; \quad [(A_2 - C_2)/B_2]^{1/2}. \quad (51)$$

Equation (50) and its solution (51) are to be compared with (29) and (30). We readily see that

$$\langle n\rangle = 2b' = [(A_2 - C_2)/B_2]^{1/2}, \quad (52)$$

when the two theories are consistent. One may readily verify that use of the above procedure in the master equation (32), yields the deterministic equation (29) for $\langle n\rangle$, in the absence of lower-order contributions. Let us write down the equation for the second moment derived analogous to (50):

$$\frac{\partial}{\partial t}\langle n^2\rangle = 2(A_2 - C_2)\langle n^3\rangle - 2B_2\langle n^5\rangle, \quad (53)$$

where we have once again, retained only the leading terms in the semiclassical limit. Once again, it may be shown that the corresponding equation for $\langle n^2\rangle$ derived from the master equation (32) is identical to (53) in the high-gain limit (it has been seen[5] that this is the only region where the semiclassical and quantum results are expected to agree). The above correspondence may be made for all the moment equations of the hierarchy obtained from each of the two theories. In the strong-signal limit, the fluctuations predicted by the macroscopic birth-death description and the fluctuations predicted by the microscopic quantum theory are the same. This has already been observed in the single-photon laser, where numerical comparisons of the moments predicted by the two theories have been made.[5] Such a comparison is not possible in this case.

The results of this paper have thus demonstrated another instance in which the stochastic master equation approach yields the microscopic results (this has already been demonstrated on the single-photon laser[5]). The success of this approach may be attributed to the fact that the pulse envelope in this case, has a constant diffusion coefficient (as in the one-photon case[5]) associated with it [this may be verified by deriving a Fokker-Planck equation from (32) via a Poisson transform].[5] Indeed, it has been seen[6] that for a system with a nonconstant diffusion coefficient, the results obtained via a Markovian master equation do not agree so closely with the results of a microscopic theory. In this case, it has been seen that the master equation predicts a broader probability distribution than the distribution obtained from the corresponding microscopic theory even though both theories predict the same mean values for the quantities of physical interest.

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