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LETTER TO THE EDITOR

Two photon absorption with coherent and partially coherent driving fields

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Abstract. A model of two photon absorption in a cavity with an external driving field is presented. Phase and amplitude fluctuations in the driving field are taken into account to model a partially coherent multimode source. The photon statistics of the light in the cavity are investigated through a calculation of the second-order correlation function of the light field in the steady state.

For a coherent driving field the field inside the cavity will exhibit photon antibunching for low intensities. As the fluctuations in the driving field are increased the photon antibunching is lost.

For a partially coherent field, $g^{(2)}(0)$ is reduced with increasing laser intensity. This reduction with laser intensity is slower with increasing laser amplitude fluctuations. This could provide a possible explanation for the recent experimental observations of Kransinski and Dinev.

We consider a model of two photon absorption from a single-mode field inside a cavity. The single-mode field is pumped by an external driving field. We include phase and amplitude fluctuations in the driving field to simulate a partially coherent multimode laser. A steady state field of finite intensity may be achieved inside the cavity.

The model differs in this respect from previous classical (Weber 1971) and quantum (Chandra and Prakash 1970, McNeil and Walls 1974, Simaan and Loudon 1975, Every 1975, Paul *et al* 1976) treatments of two photon absorption which have no driving field and hence only consider the transient situation. (A steady state is achieved in a related model where the two photon absorber is placed inside the laser cavity (Bandilla and Ritze 1976).) The above quantum treatments do not correctly model a coherent or partially coherent field since they represent the field as a statistical mixture of number states. In our analysis these fields are correctly characterised using the coherent state representation.

We follow the treatment of McNeil and Walls (1974) for the two photon absorption process. The two photon absorbing medium is characterised by the reservoir operators Γ and $\Gamma^\dagger$. The Hamiltonian for the two photon absorber in a cavity with an external driving field is

$$\begin{aligned} H &= H_0 + H_1 \\ H_0 &= \hbar\omega a^\dagger a + H_{\mathrm{R}} \qquad (1) \\ H_1 &= \hbar\kappa^{(2)}(a^{\dagger 2}\Gamma + a^2\Gamma^\dagger) - \mathrm{i}\hbar(\epsilon^*(t)\,\mathrm{e}^{\mathrm{i}\omega t}a - \epsilon(t)\,\mathrm{e}^{-\mathrm{i}\omega t}a^\dagger \end{aligned}$$

where H_{R} is the reservoir Hamiltonian, a and $a^\dagger$ are the creation and annihilation operators for the light field; $\kappa^{(2)}$ is the dipole matrix element for the two photon absorption process and $\epsilon(t)$ is the complex amplitude of the driving field which is assumed on resonance with the cavity mode of frequency ω.

The master equation for the density operator ρ of the field mode in the interaction picture is

$$\frac{\partial\rho}{\partial t}=\tfrac{1}{2}d^2(2a^2\rho a^{\dagger 2}-a^{\dagger 2}a^2\rho-\rho a^{\dagger 2}a^2)-[a\epsilon^*(t)-a^\dagger\epsilon(t),\rho] \tag{2}$$

where d^2 is the two photon absorption rate and is proportional to $|\kappa^{(2)}|^2N$ where N is the density of absorbing atoms.

This may be written in equivalent c-number form using the P-representation (Glauber 1963)

$$\rho=\int P(\alpha,t)|\alpha\rangle\langle\alpha|\,\mathrm{d}^2\alpha. \tag{3}$$

Following the techniques described in Louisell (1973) this yields the following Fokker–Planck equation for $P(\alpha, t)$:

$$\frac{\partial P(\alpha,t)}{\partial t}=-\frac{\partial}{\partial\alpha}(\epsilon^*(t)-d^2\alpha^*\alpha^2)P(\alpha,t)-\frac{1}{2}d^2\frac{\partial^2}{\partial\alpha^2}\alpha^2P(\alpha,t)+\text{cc}. \tag{4}$$

This Fokker–Planck equation may equivalently be written as the following Langevin equations (Gikhman and Skorokhod 1971, Gardiner and Chaturvedi 1977):

$$\frac{\mathrm{d}\alpha}{\mathrm{d}t}=\epsilon(t)-d^2\alpha^*\alpha^2+\mathrm{i}\,d\alpha\eta_1(t) \tag{5a}$$

$$\frac{\mathrm{d}\alpha^*}{\mathrm{d}t}=\epsilon^*(t)-d\alpha\alpha^{*2}-\mathrm{i}d\alpha^*\eta_2(t). \tag{5b}$$

(These are Itô stochastic differential equations in that the equations for the moments of $P(\alpha)$ as given by equation (4) are reproduced by equation (5).) η_1 and η_2 are independent noise sources with

$$\begin{aligned}&\langle\eta_1(t)\eta_1(t')\rangle=\langle\eta_2(t)\eta_2(t')\rangle=\delta(t-t')\\&\langle\eta_1(t)\eta_2(t')\rangle=0.\end{aligned} \tag{6}$$

In the case of a completely coherent driving field $\epsilon(t)$ is a constant. However we wish to allow for the possibility of phase and amplitude fluctuations in the driving field. To this end we write $\epsilon(t)$ as

$$\epsilon(t)=\nu(t)\,\mathrm{e}^{\mathrm{i}\phi(t)}. \tag{7}$$

Defining

$$\beta(t)=\alpha(t)\,\mathrm{e}^{-\mathrm{i}\phi(t)} \tag{8}$$

equations (5) may be written as

$$\frac{\mathrm{d}\beta}{\mathrm{d}t}=\nu(t)-d^2(\beta^*\beta)\beta+\mathrm{i}\,d\beta\eta_1(t)-\mathrm{i}\dot\phi(t)\beta \tag{9a}$$

$$\frac{\mathrm{d}\beta^*}{\mathrm{d}t}=\nu(t)-d^2(\beta^*\beta)\beta^*-\mathrm{i}d\beta^*\eta_2(t)+\mathrm{i}\dot\phi(t)\beta^*. \tag{9b}$$

Elementary laser theory suggests the following stochastic properties for the phase:

$$\dot{\phi}(t) \equiv \gamma(t) \qquad \langle\gamma(t)\gamma(t')\rangle = \Gamma\,\delta(t-t') \tag{10}$$

and amplitude:

$$\begin{aligned} &\nu(t) = \nu_0 + \nu_1(t) \\ &\langle\nu_1(t)\rangle = 0 \\ &\langle\nu_1(t)\nu_1(t')\rangle = a\,\mathrm{e}^{-b|t-t'|} \end{aligned} \tag{11}$$

(see for example Haken 1970).

Equations (9) may be written as

$$\frac{\mathrm{d}\beta}{\mathrm{d}t} = \nu_0 - d^2(\beta^*\beta)\beta + \mathrm{i}d\beta\eta_1(t) - \mathrm{i}\gamma(t)\beta + \nu_1(t) \tag{12a}$$

$$\frac{\mathrm{d}\beta^*}{\mathrm{d}t} = \nu_0 - d^2(\beta^*\beta)\beta^* - \mathrm{i}d\beta^*\eta_2(t) + \mathrm{i}\gamma(t)\beta^* + \nu_1(t). \tag{12b}$$

We wish to calculate the second-order correlation of the light field in the steady state. Neglecting fluctuations the steady state solution is

$$\beta_0 = \beta_0^* = (\nu_0/d^2)^{1/3}. \tag{13}$$

We shall solve equations (12) by a perturbation expansion about this steady state solution valid for $\nu_0 \gg d\langle\beta\rangle$, $\gamma\langle\beta\rangle$, ν_1 by substituting

$$\beta = \beta_0 + \beta_1 \tag{14}$$

we have

$$\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix}\beta_1\\ \beta_1^*\end{pmatrix} = -A\begin{pmatrix}\beta_1\\ \beta_1^*\end{pmatrix} + \begin{pmatrix}\mathrm{i}d\beta_0\eta_1(t) - \mathrm{i}\beta_0\gamma(t) + \nu_1(t)\\ -\mathrm{i}d\beta_0\eta_2(t) + \mathrm{i}\beta_0\gamma(t) + \nu_1(t)\end{pmatrix} \tag{15}$$

where

$$A = \beta_0^2 d^2\begin{pmatrix}2 & 1\\ 1 & 2\end{pmatrix}.$$

Equation (14) has the solution

$$\begin{pmatrix}\beta_1(t)\\ \beta_1^*(t)\end{pmatrix} = \int_0^t \mathrm{d}t'\,\mathrm{e}^{-A(t-t')}\begin{pmatrix}\mathrm{i}d\beta_0\eta_1(t') - \mathrm{i}\beta_0\gamma(t') + \nu_1(t')\\ -\mathrm{i}d\beta_0\eta_2(t') + \mathrm{i}\beta_0\gamma(t') + \nu_1(t')\end{pmatrix}. \tag{16}$$

The correlation functions may be calculated using the correlation properties (10) and (11) of the noise sources. The correlation matrix in the steady state is

$$C_{\mathrm{ss}} = \left\langle\begin{pmatrix}\beta_1(t)\beta_1(t) & \beta_1^*(t)\beta_1(t)\\ \beta_1(t)\beta_1^*(t) & \beta_1^*(t)\beta_1^*(t)\end{pmatrix}\right\rangle_{\mathrm{ss}} = \int_0^\infty \mathrm{e}^{-A(t-t')}B\,\mathrm{e}^{-A(t-t')} \tag{17}$$

where

$$B = \begin{pmatrix} -\beta_0^2 d^2 - \beta_0^2\Gamma + \dfrac{2a}{b+3\beta_0^2 d^2}, & \beta_0^2\Gamma + \dfrac{2a}{b+3\beta_0^2 d^2} \\[2ex] \beta_0^2\Gamma + \dfrac{2a}{b+3\beta_0^2 d^2}, & -\beta_0^2 d^2 - \beta_0^2\Gamma + \dfrac{2a}{b+3\beta_0^2 d^2} \end{pmatrix}.$$

Using the result derived in Chaturvedi *et al* (1977) we obtain from equation (17) the following result:

$$C_{ss}=\frac{(\det A)B-(A-\operatorname{Tr} A)B(A-\operatorname{Tr} A)}{2(\operatorname{Tr} A)(\det A)}$$
$$=\begin{pmatrix} -\frac{1}{3}-\frac{\Gamma}{2d^2}+\frac{a}{3\beta_0^2d^2}\left(\frac{1}{b+3\beta_0^2d^2}\right), & \frac{1}{6}+\frac{\Gamma}{2d^2}+\frac{a}{3\beta_0^2d^2}\left(\frac{1}{b+3\beta_0^2d^2}\right) \\ \frac{1}{6}+\frac{\Gamma}{2d^2}+\frac{a}{3\beta_0^2d^2}\left(\frac{1}{b+3\beta_0^2d^2}\right), & -\frac{1}{3}-\frac{\Gamma}{2d^2}+\frac{a}{3\beta_0^2d^2}\left(\frac{1}{b+3\beta_0^2d^2}\right) \end{pmatrix}. \tag{18}$$

With this result we may determine the steady state correlation functions of the cavity mode.

The mean photon number is:

$$\bar{n}=\langle\alpha^*\alpha\rangle_{ss}=\langle\beta^*\beta\rangle_{ss}=\beta_0^2+\langle\beta_1\beta_1^*\rangle=\beta_0^2+\frac{1}{6}+\frac{\Gamma}{2d^2}+\frac{a}{3\beta_0^2d^2}\left(\frac{1}{b+3\beta_0^2d^2}\right). \tag{19}$$

The correlation function is:

$$\begin{aligned}\langle\alpha^*\alpha\alpha^*\alpha\rangle_{ss}&=\langle(\beta^*\beta)^2\rangle_{ss}\\ &=\beta_0^4+4\beta_0^2\langle\beta_1\beta_1^*\rangle_{ss}+\beta_0^2(\langle\beta_1^2\rangle_{ss}+\langle\beta_1^{*2}\rangle_{ss})\\ &=\beta_0^4+\beta_0^2\left[\frac{\Gamma}{2d^2}+\frac{2a}{\beta_0^2d^2}\left(\frac{1}{b+3\beta_0^2d^2}\right)\right].\end{aligned} \tag{20}$$

The normalised second-order correlation function for $b=0$, i.e. a long correlation time of the amplitude fluctuations, may be written as

$$g_{ss}^{(2)}(0)=\frac{\langle(\alpha^*\alpha)^2\rangle_{ss}}{\langle\alpha^*\alpha\rangle_{ss}^2}=1-\frac{1}{3\bar{n}}+\frac{4a}{9\nu_0^2}+\left(\text{terms of order }\frac{\Gamma}{d^2},\frac{a^2}{\nu_0^4},\frac{1}{\bar{n}^2}\right). \tag{21}$$

We note that to this order in Γ the phase fluctuations do not show up in $g_{ss}^{(2)}(0)$. We discuss this result for the cases of a fully coherent field and a partially coherent field.

(*a*) *Coherent field*

For a perfectly coherent driving field $a=b=0$ and

$$g^{(2)}(0)=1-(1/3\bar{n}). \tag{22}$$

Hence photon antibunching $g^{(2)}(0)<1$ may be expected to be observable for low light intensities, i.e. $\bar{n}\sim 1$.

A calculation of the two-time correlation function using the quantum regression theorem (Lax 1968) yields

$$g^{(2)}(\tau)=1+(g^{(2)}(0)-1)\,e^{-\tau/\tau_c} \tag{23}$$

where

$$\tau_c=\frac{1}{3\bar{n}\,d^2}.$$

For $\bar{n}=1$ this gives a correlation time of the order of the inverse of the two photon absorption rate.

The photon count rate may be estimated as follows. The extraction rate of the photons from the cavity must be less than the rate required for the light field to reach a steady state. This relaxation time is given by τ_c, hence the photon count rate must be less than $\bar{n}d^2$. Hence in order to observe photon antibunching which from equation (22) requires $\bar{n}\sim 1$, a two photon absorber with a large d^2 is required.

Photon antibunching as a result of two photon absorption from a coherent light beam has been predicted by several authors (Chandra and Prakash 1970, Tornau and Bach 1974, Simaan and Loudon 1975, Every 1975, Paul *et al* 1976). However all these analyses are for the transient situation where an initially coherent field is approximated by a Poissonian ensemble of number states.

(*b*) *Partially coherent fields*

For a partially coherent driving field one sees from equation (21) that the photon antibunching may be lost owing to the amplitude fluctuations in the driving field. It will therefore be necessary to achieve sufficient two photon absorption with stabilised coherent lasers if photon antibunching is to be observed.

In the high intensity limit we may neglect the term $-1/3\bar{n}$ and write $g^{(2)}(0)$ as

$$g^{(2)}(0)=1+\frac{4}{9}\frac{a(\nu_0)}{\nu_0^2}, \tag{24}$$

where $a(\nu_0)$ gives the intensity dependence of the amplitude correlation function in equation (11). Thus $g^{(2)}(0)$ decreases as the ratio of the variance of the amplitude fluctuations over the square of the amplitude. This result holds for partially coherent fields but not fully chaotic fields where $a(\nu_0)\sim\nu_0^2$ and the perturbation expansion breaks down. The result given by equation (24) is in qualitative agreement with the recent experiments of Kransinski and Dinev (1976) on two photon absorption from a pulsed dye laser. They observed a slower decrease of $g^{(2)}(0)$ with the laser intensity than predicted by the analysis of Weber (1971).

Equation (24) predicts a slower decrease of $g^{(2)}(0)$ with laser intensity as the laser amplitude fluctuations are increased. As such this could provide an explanation for the observations of Kransinski and Dinev (1976).

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