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TWO–PHOTON ABSORPTION IN UNPOLARIZED LIGHT[1]

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We study two-photon absorption in unpolarized light beams. The mean photon number and its variance for the total beam decreased as known from single-mode considerations. However, the time evolution of the variance for one of the polarization modes depends on the interaction process leading to sub- or super-Poissonian photon statistics.

1. Introduction

Several years ago two- and multi-photon absorption has found theoretical interest. Especially the change of photon statistics in a single-mode monochromatic light beam was studied [1]-[6]. An interesting feature of these processes is the reduction of the fluctuation, the photon statistics can become narrower than the Poisson distribution characterizing Glauber states. We want to extend these considerations to unpolarized light and thus to two polarization modes. Steps into this direction have already been made. In particular the absorption from two light modes - one photon from the first and second light beam, respectively, - has been studied intensively [7]-[12]. However, when we consider a two-mode light beam, as for instance unpolarized light, there are additional processes possible if, on average, balanced absorption from both modes is required. The question arises under which conditions a reduction of fluctuations and furthermore sub-Poissonian photon statistics occurs for the total beam and each polarization mode.

In this contribution we discuss two-photon absorption in unpolarized light including all possible two-photon absorption processes under the condition that the light beam leaves the absorber unpolarized. For simplicity reasons we restrict ourselves to monochromatic light beams. We consider a light beam as unpolarized of type II [13]

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when the corresponding density operator (or the quasidistribution) is (i) invariant when performing an arbitrary rotation on the axis of light propagation and (ii) symmetric with respect to left- and right-handed circular polarization. If there is (iii) an additional invariance with respect to phase retardation we call such light unpolarized of type I [13]. We study the time evolution of the total mean photon number as well as the mean photon number of one polarization mode and its fluctuations. In realistic experiments one will start with light beams with high mean photon numbers. However the numerical effort in calculating the effects exactly grows drastically when increasing the number of photons in the light beam. In order to demonstrate the principal characteristics we treat the process by means of a simple approximative procedure in analogy to considerations taking into account only a single mode [2]. We present a few characteristic examples to illustrate the influence of a specific absorption process on the fluctuations in each of the polarization modes.

2. The model

We consider only absorption processes conserving - at least - the type II properties of an unpolarized light beam, i.e. rotational invariance and symmetry with respect to left- and right-handed circular polarization. We are free to choose the quantization axis to be parallel to the direction of propagation. In addition, the atoms are supposed to be in the ground state $J = 0$. Thus the following processes are allowed: the medium can absorb (i) one left- and one right-handed circularly polarized photon where $\Delta m = 0$ as for instance in a transition $J = 0 \to J = 0$ or (ii) one left- and one right-handed or two left- or two right-handed circularly polarized photons where $\Delta m = 0, \pm 2$ in a transition $J = 0 \to J = 2$. The interaction Hamiltonian is given by

$$\hat{H}_{int} = \hbar \Big[\eta_2 \hat{c}^\dagger_{u2} \hat{c}_l \hat{a}_r \hat{a}_l + \eta_2^* \hat{c}_{u2} \hat{c}^\dagger_l \hat{a}^\dagger_r \hat{a}^\dagger_l + \eta_1 \hat{c}^\dagger_{u1} \hat{c}_l \hat{a}_r^2 + \eta_1^* \hat{c}_{u1} \hat{c}^\dagger_l \hat{a}^{\dagger 2}_r + \eta_3 \hat{c}^\dagger_{u3} \hat{c}_l \hat{a}_l^2 + \eta_3^* \hat{c}_{u3} \hat{c}^\dagger_l \hat{a}^{\dagger 2}_l \Big] , \quad (1)$$

where $\eta_1 = \eta_3 = 0$ for the $\Delta m = 0$ transition. In (1) the term $\hat{c}^\dagger_{u2} \hat{c}_l \hat{a}_r \hat{a}_l$ describes the population of the upper level 'u2' and the depletion of the lower level 'l' by absorbing a pair of left- and right-handed circularly polarized photons and so on. Standard techniques lead to the equation of motion

$$\begin{aligned} \frac{d\hat{\varrho}(t)}{dt} &= -\gamma_2 \left([\hat{a}^\dagger_r \hat{a}^\dagger_l, \hat{a}_r \hat{a}_l \hat{\varrho}] + [\hat{\varrho} \hat{a}^\dagger_r \hat{a}^\dagger_l, \hat{a}_r \hat{a}_l] \right) \\ &\quad -\gamma_1 \left([\hat{a}^{\dagger 2}_r, \hat{a}^2_r \hat{\varrho}] + [\hat{\varrho} \hat{a}^{\dagger 2}_r, \hat{a}^2_r] \right) - \gamma_3 \left([\hat{a}^{\dagger 2}_l, \hat{a}^2_l \hat{\varrho}] + [\hat{\varrho} \hat{a}^{\dagger 2}_l, \hat{a}^2_l] \right) . \end{aligned} \quad (2)$$

The coefficients γ_1, γ_2 and γ_3 contain all relevant parameters. In order to obtain a master equation conserving the type II properties we require

$$\gamma_1 = \gamma_3 . \quad (3)$$

In the special case

$$\gamma_1 = \gamma_3 = \gamma_2/2 \quad (4)$$

the additional retardation invariance of an unpolarized light beam of type I is conserved and the output light beam is again unpolarized light of type I.

3. Approximative solution

We are mainly interested in the time evolution of the mean photon number and higher photon number moments. Therefore we concentrate on the time evolution of the diagonal elements $\varrho_{n,m} = \langle m, n|\hat{\varrho}|n, m\rangle$ of the density operator given in the Fock-basis of left- and right-handed circular polarization $|n, m\rangle = |n\rangle_l |m\rangle_r$. ¿From (2) we find the equation of motion

$$\begin{aligned} \frac{d\varrho_{n,m}}{dt} &= -2\gamma_2 \left[nm\varrho_{n,m} - (n+1)(m+1)\varrho_{n+1,m+1} \right] \\ &\quad -2\gamma_1 \left[n(n-1)\varrho_{n,m} - (n+1)(n+2)\varrho_{n+2,m} \right] \\ &\quad -2\gamma_1 \left[m(m-1)\varrho_{n,m} - (m+1)(m+2)\varrho_{n,m+2} \right] , \end{aligned} \quad (5)$$

where we already have taken into account $\gamma_1 = \gamma_3$. The calculation is similar to the single-mode case [2]. In addition, the correlation between the two polarization modes must be taken into account. For the moments of one polarization mode we obtain

$$\frac{d\langle n^k \rangle}{dt} = -\gamma_2 \left\langle nm(n^k - (n-1)^k) \right\rangle - \gamma_1 \left\langle n(n-1)(n^k - (n-2)^k) \right\rangle . \quad (6)$$

Due to the left- and right-symmetry $\langle n^k \rangle = \langle m^k \rangle$ and the equation for the moments $\langle m^k \rangle$ of the orthogonal polarization mode is equivalent to (6). The correlations between the modes are governed by

$$\begin{aligned} \frac{d\langle n^k m^{k'} \rangle}{dt} &= -\gamma_2 \left\langle nm(n^k m^{k'} - (n-1)^k (m-1)^{k'}) \right\rangle \\ &\quad -\gamma_1 \left\langle n(n-1) m^{k'} (n^k - (n-2)^k) \right\rangle \\ &\quad -\gamma_1 \left\langle m(m-1) n^k (m^{k'} - (m-2)^{k'}) \right\rangle . \end{aligned} \quad (7)$$

From (6) and (7) we obtain equations for the mean photon number if $k = 1$, the second moment if $k = 2$ and the correlation of lowest order if $k = k' = 1$. These equations can be approximately solved in analogy to single-mode calculations [2]. We set $n(t) = \langle n(t) \rangle + \Delta n(t)$ where $\Delta n(t)$ is a small deviation from the mean photon number $\langle n(t) \rangle$ and define

$$h_n(t) \equiv \frac{\langle (\Delta n)^2 \rangle}{\langle n \rangle} \equiv \frac{\langle n^2(t) \rangle - \langle n(t) \rangle^2}{\langle n(t) \rangle} \ll \langle n(t) \rangle , \quad (8)$$

$$h_{nm}(t) \equiv \frac{\langle \Delta n \Delta m \rangle}{\langle n \rangle} \equiv \frac{\langle n(t) m(t) \rangle - \langle n(t) \rangle^2}{\langle n(t) \rangle} \ll \langle n(t) \rangle \quad (9)$$

and therefore

$$h_{tot}(t) \equiv \frac{\langle(\Delta n_{tot})^2\rangle}{\langle n_{tot}\rangle} \equiv \frac{\langle n_{tot}^2(t)\rangle - \langle n_{tot}(t)\rangle^2}{\langle n_{tot}(t)\rangle} = h_n(t) + h_{nm}(t)\,, \tag{10}$$

where $n_{tot}(t) = n(t) + m(t)$. These requirements are satisfied, for instance, by unpolarized light of type II (generated at a beam splitter [14])

$$\hat{\varrho} = \exp\left\{-|\alpha|^2\right\} \sum_{N=0}^{\infty} \left(\frac{|\alpha|^2}{2}\right)^N \frac{1}{N!} \sum_{n=0}^{N} \binom{N}{n} |N-n\rangle_l |n\rangle_r \, _r\langle n| \, _l\langle N-n| \tag{11}$$

and - in a restricted range - by unpolarized Glauber light of type I (see for instance [13])

$$\hat{\varrho} = \exp\left\{-|\alpha|^2\right\} \sum_{N=0}^{\infty} \frac{|\alpha|^{2N}}{(N+1)!} \sum_{n=0}^{N} |N-n\rangle_l |n\rangle_r \, _r\langle n| \, _l\langle N-n|\,. \tag{12}$$

On the assumption $\langle(\Delta n)^2\rangle \ll \langle n\rangle$ we obtain in the lowest order of approximation, i.e. when neglegting terms of the order $\langle n\rangle^{-1}$, the equations

$$\frac{d\langle n\rangle}{dt} = -(4\gamma_1 + 2\gamma_2)\langle n\rangle^2\,, \tag{13}$$

$$\begin{aligned}\frac{d\langle n^2\rangle}{dt} &= -(8\gamma_1 + 4\gamma_2)\langle n\rangle^3 + (16\gamma_1 + 2\gamma_2)\langle n\rangle^2 \\ &\quad -8\gamma_2\langle n\rangle\langle\Delta n\Delta_m\rangle - (24\gamma_1 + 4\gamma_2)\langle n\rangle\langle\Delta n\rangle^2\,,\end{aligned} \tag{14}$$

$$\begin{aligned}\frac{d\langle nm\rangle}{dt} &= -(8\gamma_1 + 4\gamma_2)[\langle n\rangle^3 + 2\langle n\rangle\langle\Delta n\Delta_m\rangle + \langle n\rangle\langle\Delta n\rangle^2] \\ &\quad +(8\gamma_1 + 2\gamma_2)\langle n\rangle^2.\end{aligned} \tag{15}$$

The approximate solution then can be easily obtained for $h_{tot} = h_n + h_{nm}$

$$h_{tot}(t) = \frac{2}{3} + \left\{\frac{\langle n(t)\rangle}{\langle n_0\rangle}\right\}^3 \left[h_{tot0} - \frac{2}{3}\right] \tag{16}$$

and $h_d = h_n - h_{nm}$

$$h_d(t) = \frac{4\gamma_1}{6\gamma_1 - \gamma_2} + \left\{\frac{\langle n(t)\rangle}{\langle n_0\rangle}\right\}^{\frac{6\gamma_1-\gamma_2}{2\gamma_1+\gamma_2}} \left[h_{d0} - \frac{4\gamma_1}{6\gamma_1 - \gamma_2}\right]\,, \tag{17}$$

where $\langle n_0\rangle = \langle n(0)\rangle$, $h_{tot0} = h_{tot}(0)$ and $h_{d0} = h_d(0)$. In case of unpolarized Glauber-light of type II as given in (11) $h_{tot0} = 1$ and $h_{d0} = 1$. For unpolarized Glauber–light of type I given in (12) we find $h_{tot0} = 1$ and $h_{d0} = 1 + 2\langle n_0\rangle/3$. When we disregard the polarization and consider the relative fluctuations of the total beam (10) we find the same expression as for single–mode light presented in [2] and therefore a reduction of the fluctuations in the total photon number leading to sub-Poissonian statistics. The time evolution of $h_{tot}(t)$ depends on the time evolution of the mean photon number and

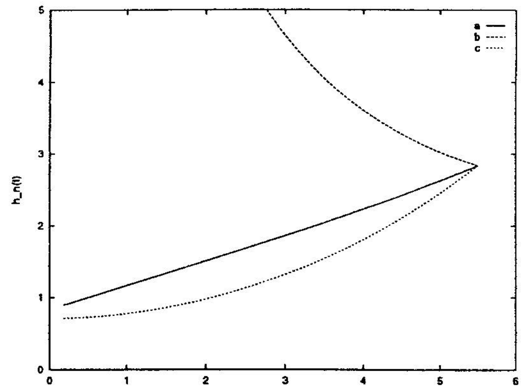

Fig. 1. Relative fluctuation of one polarization mode $h_n(t)$ depending on the mean photon number $\langle n(t)\rangle$ of this mode for specific two-photon absorption processes: (a) $\gamma_1 = \gamma_3 = \gamma_2/2$, (b) $\gamma_1 = \gamma_3 = 0$ and $\gamma_2 = 1$ and (c) $\gamma_1 = \gamma_3 = 3\gamma_2/2$. Depending on the kind of absorption process the relative fluctuation are increased or decreased. The initial state is an unpolarized Glauber light beam of type I (12) and $h_n(0) = 1 + \langle n_0\rangle/6$.

thus indirectly on the respective absorption process. The variance in one polarization mode is given by

$$\begin{aligned}h_n(t) = \frac{12\gamma_1 - \gamma_2}{18\gamma_1 - 3\gamma_2} &+ \left\{\frac{\langle n(t)\rangle}{\langle n_0\rangle}\right\}^{\frac{6\gamma_1-\gamma_2}{2\gamma_1+\gamma_2}} \left[\frac{h_{d0}}{2} - \frac{2\gamma_1}{6\gamma_1 - \gamma_2}\right] \\ &+ \left\{\frac{\langle n(t)\rangle}{\langle n_0\rangle}\right\}^3 \left[\frac{h_{tot0}}{2} - \frac{1}{3}\right].\end{aligned} \tag{18}$$

For the corresponding equation for the correlations we obtain

$$\begin{aligned}h_{nm}(t) = \frac{-\gamma_2}{18\gamma_1 - 3\gamma_2} &- \left\{\frac{\langle n(t)\rangle}{\langle n_0\rangle}\right\}^{\frac{6\gamma_1-\gamma_2}{2\gamma_1+\gamma_2}} \left[\frac{h_{d0}}{2} - \frac{2\gamma_1}{6\gamma_1 - \gamma_2}\right] \\ &+ \left\{\frac{\langle n(t)\rangle}{\langle n_0\rangle}\right\}^3 \left[\frac{h_{tot0}}{2} - \frac{1}{3}\right].\end{aligned} \tag{19}$$

Depending on the coefficients γ_1 and γ_2 two-photon absorption leads to super- or sub-Poissonian photon statistics for one polarization mode of the light beam.

Examples

Figs. 1 and 2 illustrate Eq. (18) for different absorption processes distinguished by their coefficients $\gamma_1 = \gamma_3$ and γ_2. As initial state we chose the two types of unpolarized Glauber-light beams given in (11) and (12). The influence of the kind of the absorption process is clearly visible. If $\gamma_1 = \gamma_3 = 0$ then $h_n(t)$ increases, while $h_{tot}(t)$ for the total mean photon number decreases. (Note that the approximation breaks down if $h_n(t)$ or $h_{nm}(t)$ is in the order of $\langle n(t)\rangle$.) This absorption process does not reduce relative fluctuations h_n of each polarization mode but only fluctuations h_{tot} of the total light

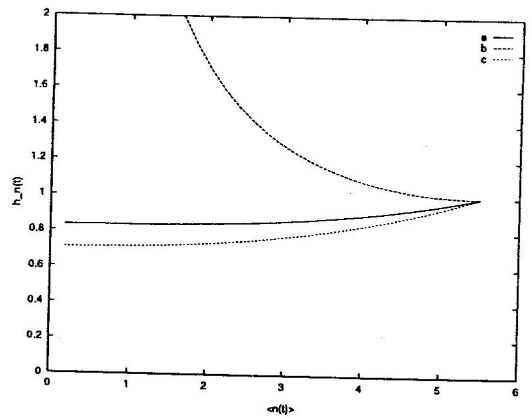

Fig. 2. The same as in Fig. 1. The initial state is an unpolarized Glauber light beam of type II (11) and $h_n(0) = 1$.

beam. Due to the interaction only pairs consisting of one left- and one right-handed circularly polarized photon are absorbed. Thus the contribution of fluctuations of 'pure polarization' increases relatively. This means that super-Poissonian photon statistics is present in each polarization mode. If $3\gamma_1 > \gamma_2$ we find that $h_n(\infty) < 1$. In such processes not only the relative fluctuations h_{tot} of the total mean photon number but also relative fluctuations h_n in each of the polarization modes are reduced. Accordingly, the correlation between the two polarization modes increases drastically. In order to obtain sub-Poissonian photon statistics also for high mean photon numbers it is required to start with suitable initial conditions as satisfied for unpolarized Glauber light of type II.

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