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LETTER TO THE EDITOR

Attenuation of a multimode field due to two-photon absorption

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Abstract. Starting from an effective Hamiltonian describing the interaction of a multimode field with a two-photon absorber, an equation of motion for the quantised slowly varying field amplitude is derived which exhibits a memory effect due to the finite atomic linewidth. Averaging this equation, as a whole, over the memory time leads to a single-mode formalism, thus providing a basis for the physical interpretation of the conventional one-mode treatment of two-photon absorption, especially with respect to the photon antibunching phenomenon. In particular, a precise physical meaning can be attributed to the mode volume.

In recent years, the change in photon statistics due to multiphoton absorption has become an object of several theoretical studies (see e.g. Tornau and Bach 1974, McNeil and Walls 1974, Simaan and Loudon 1975, Bandilla and Ritze 1975, Paul *et al* 1976). The most interesting result is that such a process may produce, at least in principle, photon antibunching, as expressed by the relation

$$\Delta n^2/\bar{n}^2 = \alpha/\bar{n} \qquad (\tfrac{1}{2} \leqslant \alpha < 1). \tag{1}$$

Here $\bar{n}$ is the mean value and Δn^2 the mean-square deviation for the photon number. The definite value of the constant α depends on the type of process. For two-photon absorption we have $\alpha = \frac{2}{3}$ for the asymptotic state of the field which is attained after sufficiently strong absorption (Paul *et al* 1976), and $\alpha = \frac{1}{2}$ for the final steady state when $\bar{n} = \frac{1}{2}$ (cf. Simaan and Loudon 1975).

From equation (1) it becomes obvious that the antibunching effect can be observed only at low values of $\bar{n}$ (say $\bar{n} \leqslant 100$). Now equation (1) has been deduced in the single-mode formalism, i.e. $\bar{n}$ refers to the mode volume, and the question, left open in the literature, arises as to how this volume is defined physically. In the following we shall try to give an answer.

Our procedure consists of deriving the commonly used single-mode formalism, as an approximate scheme, from general equations describing the interaction between a two-photon absorber and a multimode field. (The generalisation to k-photon absorption, where $k = 3, 4, \ldots$, is straightforward.) We consider a light beam travelling in the x direction and coherent over its cross section. The absorption cell is homogeneously filled with atoms capable of two-photon absorption. Then the problem is essentially

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one-dimensional (with respect to space). We start from the following effective interaction Hamiltonian (cf. Shen 1967):

$$H_{\text{int}} = \hbar\eta \sum_{\mu} \hat{E}^{(-)2}(x_\mu) a_\mu + \text{h.c.} \qquad (\hat{E}^{(-)}(x) \equiv \exp(\mathrm{i}k_0 x) E^{(-)}(x)). \tag{2}$$

Here a_μ^+ and a_μ denote the raising and lowering operators, respectively, for the μth atom located at x_μ, $E^{(-)}$ is the negative-frequency part of the operator for the electric field strength, k_0 is the wavenumber at the centre frequency ω_0, and η is the coupling constant. We take into account inhomogeneous as well as homogeneous line broadening, i.e. we ascribe to an individual atom both a resonance frequency Ω_μ (with respect to two-photon absorption) and a homogeneous linewidth 2γ.

From the formal point of view it is desirable to consider the atomic operators—like the field operators—as *continuous* functions in space. Doing so, we rewrite the interaction Hamiltonian (2) as

$$H_{\text{int}} = \hbar \sum_j \zeta_j \int_0^L \mathrm{d}x\, \hat{E}^{(-)2}(x) a_j(x) + \text{h.c.} \tag{3}$$

where the subscript j has been used to distinguish between groups of atoms which differ by their resonance frequencies Ω_j. The dependence of the new coupling constant ζ_j on j reflects the decrease in the number of atoms in the jth group with the growing deviation of Ω_j from the line centre. Taking into account only those modes of the field which propagate strictly in the (positive) x direction, we find the commutator for the field operators to be

$$[\hat{E}^{(+)}(x_1), \hat{E}^{(-)}(x_2)] = -2\pi\mathrm{i}\hbar c\delta'(x_1 - x_2) + 2\pi\hbar\omega_0\delta(x_1 - x_2) \tag{4}$$

where δ' is the derivative of Dirac's delta function, and c is the velocity of light. The commutator for the atomic variables, on the other hand, reads

$$[a_j^+(x_1), a_j(x_2)] = \sigma_j(x_1)\delta(x_1 - x_2) \tag{5}$$

where $\sigma_j(x)$ is the inversion density associated with the group j. In the following, we shall approximate $\sigma_j(x)$ by -1, thus neglecting saturation effects.

From equations (3)–(5) we obtain the equations of motion in the Heisenberg picture which, after separation of the high-frequency time dependence of the operators, take the following form:

$$\dot{\tilde{a}}_j^+(x, t) = \mathrm{i}(\Omega_j - 2\omega_0)\tilde{a}_j^+(x, t) - \gamma\tilde{a}_j^+(x, t) + \mathrm{i}\zeta_j\tilde{E}^{(-)2}(x, t) + \tilde{f}_j^+(x, t) \tag{6}$$

$$(\partial/\partial t + c\,\partial/\partial x)\tilde{E}^{(-)}(x, t) = 4\pi\mathrm{i}\hbar\omega_0 \sum_j \zeta_j^* \tilde{a}_j^+(x, t)\tilde{E}^{(+)}(x, t) \tag{7}$$

where

$$\tilde{E}^{(-)}(x, t) = \exp(-\mathrm{i}\omega_0 t)\hat{E}^{(-)}(x, t) = \exp[\mathrm{i}(k_0 x - \omega_0 t)]E^{(-)}(x, t) \tag{8}$$

$$\tilde{a}(x, t) = \exp(2\mathrm{i}\omega_0 t)a(x, t) \tag{9}$$

and ω_0 is the centre frequency for the field whose doubled value is assumed to coincide with the atomic line centre.

In equation (6) a damping term characteristic of homogeneous line broadening (linewidth 2γ) together with a fluctuating (Langevin) force $\tilde{f}_j^+(x, t)$ ensuring quantum-mechanical consistency have been introduced in a familiar manner (see e.g. Lax 1966). In the derivation of the interaction term of equation (7) only the dominant second term on the right-hand side of equation (4) has been taken into account.

Integrating equation (6) and substituting the result into equation (7) gives us

$$(\partial/\partial t + c\,\partial/\partial x)\tilde{E}^{(-)}(x,t)$$
$$= -4\pi\hbar\omega_0\rho\int_{-\infty}^{t}\exp[-(\Gamma+\gamma)(t-t')]\tilde{E}^{(-)2}(x,t')\,\mathrm{d}t'\,\tilde{E}^{(+)}(x,t)$$
$$+\tilde{F}^{+}(x,t)\tilde{E}^{(+)}(x,t) \tag{10}$$

where

$$\tilde{F}^{+}(x,t) = 4\pi\mathrm{i}\hbar\omega_0\sum_j \zeta_j^{*}\int_{-\infty}^{t}\exp\{[\mathrm{i}(\Omega_j - 2\omega_0)-\gamma](t-t')\}\tilde{f}_j^{+}(x,t')\,\mathrm{d}t'. \tag{11}$$

Here we have assumed the inhomogeneous line to be of Lorentzian shape (linewidth 2Γ), i.e. we have replaced

$$\sum_j |\zeta_j|^2 \ldots \qquad \text{by} \qquad \rho\Gamma\pi^{-1}\int \mathrm{d}\Omega[(\Omega-2\omega_0)^2+\Gamma^2]^{-1}\ldots$$

in equation (10). Since the right-hand side of equation (10) contains temporal averages (the weighting function being $(\Gamma+\gamma)\exp[-(\Gamma+\gamma)(t-t')]$), it appears natural to subject equation (10), as a whole, to the same averaging procedure. The result can be written in the approximate form†

$$(\partial/\partial t + c\,\partial/\partial x)\mathscr{E}^{(-)}(x,t) = -4\pi\hbar\omega_0\rho(\Gamma+\gamma)^{-1}\mathscr{E}^{(-)2}(x,t)\mathscr{E}^{(+)}(x,t) + \tilde{F}^{+}(x,t)\mathscr{E}^{(+)}(x,t) \tag{12}$$

where

$$\mathscr{E}^{(\pm)}(x,t) = (\Gamma+\gamma)\int_{-\infty}^{t}\exp[-(\Gamma+\gamma)(t-t')]\tilde{E}^{(\pm)}(x,t')\,\mathrm{d}t'. \tag{13}$$

The commutator relation for the averaged electric field strength is easily calculated to be

$$[\mathscr{E}^{(+)}(x,t),\,\mathscr{E}^{(-)}(x,t)] = \pi\hbar k_0(\Gamma+\gamma). \tag{14}$$

Obviously the operators

$$\tilde{q} = [\pi\hbar k_0(\Gamma+\gamma)]^{-1/2}\mathscr{E}^{(+)}, \qquad \tilde{q}^{+} = [\pi\hbar k_0(\Gamma+\gamma)]^{-1/2}\mathscr{E}^{(-)} \tag{15}$$

obey the correct commutation relation for the familiar photon creation and annihilation operators. Hence rewriting equation (12) as

$$(\partial/\partial\tau)\tilde{q}^{+}(\xi,\tau) = -\beta\tilde{q}^{+2}(\xi,\tau)\tilde{q}(\xi,\tau) + \tilde{F}^{+}(\xi,\tau)\tilde{q}(\xi,\tau) \tag{16}$$
$$(\xi = x - ct, \quad \tau = t, \quad \beta = (2\pi\hbar)^2\omega_0 k_0\rho)$$

we finally arrive at an equation of motion for a single 'mode'. In contrast to the

† Strictly speaking, the approximate replacement of $(\Gamma+\gamma)\int_{-\infty}^{t}\exp[-(\Gamma+\gamma)(t-t')]\tilde{E}^{(-)2}(x,t')\,\mathrm{d}t'$ by $\mathscr{E}^{(-)2}(x,t)$ is not correct when the contributions from all modes (including those which remain in the vacuum state during the interaction) are considered. Physically, it seems reasonable, however, to take into account only those modes which are actually affected by the atoms (either modes present in the incident field and experiencing attenuation due to the atoms, or modes originally in the vacuum state and growing up to some extent in the course of interaction). If we assume the linewidth of the incident field $\Delta\omega$ to be smaller than $\gamma+\Gamma$, we may restrict ourselves to those modes whose frequencies ω obey the inequality $|\omega-\omega_0| \leqslant \gamma+\Gamma$. In this way the above mentioned replacement can be justified.

conventional *ab initio* single-mode formalism, however, our analysis yields a physical definition for the mode volume. In fact, comparing the relation

$$\langle \tilde{q}^+ \tilde{q} \rangle = \langle \mathscr{E}^{(-)} \mathscr{E}^{(+)} \rangle / \pi \hbar k_0 (\Gamma + \gamma) \tag{17}$$

following from equations (15) with the corresponding equation in the single-mode treatment, we find the dimension of the mode volume, in the direction of beam propagation, to be $2c(\Gamma+\gamma)^{-1}$.

To show the definite correlation with the conventional treatment of two-photon absorption we mention that equation (16) yields precisely the same equation of motion for the expectation value of any product of photon creation and annihilation operators as does the density matrix formalism commonly used. (Note that in the case of normally ordered products the term $\tilde{F}^+ \tilde{q}$ gives no contribution, owing to the fact that the expectation value for any normally ordered product of the fluctuating forces $\tilde{f}_i^+, f_i$ vanishes.)

Finally, let us discuss two physically important cases.

(i) *Entirely homogeneous line broadening* ($\Gamma = 0$). Assuming the linewidth of the incident field $\Delta\omega$ to be smaller than γ, we arrive at the conclusion that it is not the coherence time $(\Delta\omega)^{-1}$ but the smaller dephasing time $(2\gamma)^{-1}$ which determines what has to be taken as the mode volume. Similarly, considering a field travelling in a ring resonator, it is legitimate to identify the mode volume appearing in the single-mode treatment of two-photon absorption with the resonator volume only when the round-trip time does not exceed the dephasing time.

Our result is in accordance with the following simple, intuitive picture based on Einstein's original concept of localised light quanta. An individual atom can absorb a pair of photons only during a time interval in which the field interacts coherently with the atom. Hence, in an elementary absorption process, the temporal 'distance' between the two photons involved cannot exceed the dephasing time. Therefore intensity correlations characteristic for antibunching, as they are produced by the absorber in favourable circumstances, can exist between two times t_1 and t_2 (at fixed x) only if $|t_1 - t_2| \leqslant (2\gamma)^{-1}$.

(ii) *Strong inhomogeneous line broadening* ($\Gamma \gg \Delta\omega \gg \gamma$). The linewidth of the field $\Delta\omega$ is larger than γ. Since only those atoms whose resonance frequencies obey the inequality $|\Omega_j - 2\omega_0| \leqslant \Delta\omega$ can experience absorption, it appears reasonable to take into consideration only the effective part of the inhomogeneous linewidth $\Gamma_{\text{eff}} \approx \Delta\omega$. Then the mode volume, apart from a factor 2, is equal to the coherence volume. In this case the coherent interaction of an individual atom with the field terminates because the phase of the field changes in a random manner when a time of the order of the coherence time has elapsed.

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