# FINITE LASER BANDWIDTH EFFECT ON *n*-PHOTON RESONANCE PHENOMENA

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Applying the theory of multiplicative stochastic processes, the effect of finite laser bandwidth on $n$-photon resonance phenomena is discussed and shown to be stronger ($\sim n^2$) in the $n$-photon case than in the one-photon case.

## 1. Introduction

Resonance interaction of optical fields and atoms leads to many interesting effects [1]. With regard to resonance interaction one is justified in applying the two-level approximation, which takes into account only two levels of the atom. The evolution of such a system is well described by the optical equations of Bloch. Agarwal [2] and, quite recently, Wódkiewicz [3], applying the theory of multiplicative stochastic processes, have shown how the stochastic nature of the exciting field affects the resonance phenomena. The stochastic approach enables to take into account the finite linewidth of real lasers by considering the phase of the laser field as a stochastic variable [4]. Usually, "resonance" is intended to mean one-photon resonance, which implies matching of the photon and atomic transition energies. In this paper, we propose results concerning the influence of the finiteness of the laser linewidth on $n$-photon resonance processes i.e. ones in which the energy of $n$-photons is matched to the energy of the atomic transition ($n\omega \approx \omega_0$). In our description of the finite laser linewidth we apply a phase diffusion model, based on an analogy with Brown's motion [4].

## 2. The hamiltonian

We assume the hamiltonian of the system in the form:

$$H = H_F + H_A + H_1 + H_2, \tag{1}$$

where

$$H_F = \sum_k \hbar \omega_k a_k^\dagger a_k \tag{2}$$

desribes the free radiation field and

$$H_A = \hbar \omega_0 S^z \tag{3}$$

the material system; we assume that, at interaction with the laser field, only the populations of two of its levels, differing by $\hbar\omega_0$, change significantly; $S^z$ is the energetic spin operator component. In fact, we deal with the atom as a two-level one.

The hamiltonian of interaction of the atom and radiation field consists of two parts:

$$H_1 = \hbar (\eta^{(n)} a^n S^+ + \eta^{(n)} a^{\dagger n} S^-), \tag{4}$$

$$H_2 = {\sum_k}' \hbar (\eta_k^{(1)} a_k S^+ + \eta_k^{(1)*} a_k^\dagger S^-). \tag{5}$$

Eq. (4) accounts for interaction between the atom and the strong laser beam, at $n$-photon resonance with the atom ($n\omega \approx \omega_0$). Eq. (5) describes weak interaction with the field leading to spontaneous emission in the system (the prime at $\Sigma$ denotes that the incident beam is not comprised in the sum). $S^+$ and $S^- = [S^+]^+$ are operators raising and lowering the energy of the atom which fulfil the commutation rules:

$$[S^+, S^-] = 2S^z, \qquad [S^z, S^\pm] = \pm S^\pm, \tag{6}$$

whereas $a_k^\dagger$ and $a_k$ are creation and annihilation operators of a photon in the $k$th mode, and $\eta^{(n)}$, $\eta_k^{(1)}$ are the respective atom–field coupling constants. In (4) and (5) the rotating wave approximation is made and rapidly oscillating terms omitted.

On the assumption that the laser beam is in coherent state $a|\alpha\rangle = \alpha|\alpha\rangle$, the hamiltonian $H_1$ reduces to:

$$H_1 = \hbar(\eta^{(n)}\alpha^n \exp(-\mathrm{i}n\omega t)S^+ + \text{h.c.}), \tag{7}$$

where $\alpha$ is the complex amplitude of the field, to be written as:

$$\alpha = r\exp(-\mathrm{i}\varphi). \tag{8}$$

The form (7) of $H_1$ together with (8) permit the modelling of the finite width of the laser line by introducing fluctuations in the phase $\varphi$ (at constant $r$) and the application of the so-called phase diffusion model, based on an analogy with Brown motion.

## 3. Phase-diffusion model

In this model, the phase $\varphi(t)$ is the stochastic variable of a distribution function $P$, obeying the diffusion equation [4]:

$$\partial P/\partial t = \Gamma_L\, \partial^2 P/\partial\varphi^2, \tag{9}$$

$\Gamma_L$ is the laser linewidth.

The Langevin equation corresponding to (9) has the form:

$$\mathrm{d}\varphi(t)/\mathrm{d}t = f(t), \tag{10}$$

where $f(t)$ is a $\delta$-correlated gaussian random process with

$$\langle f(t)\rangle = 0, \qquad \langle f(t)\, f(s)\rangle = 2\Gamma_L(t-s). \tag{11}$$

Single brackets denote here the classical ensemble average with respect to the distribution of the random process $f(t)$.

The Green's function solution of eq. (9) can be written as follows [4]:

$$P(\varphi, t|\varphi_0, 0) = \frac{1}{2\pi}\sum_{-\infty}^{\infty} \exp\{\mathrm{i}m(\varphi-\varphi_0)\}\exp(-m^2\Gamma_L t). \tag{12}$$

Together with the uniform distribution over $2\pi$ of the initial phase $\varphi_0 = \varphi(0)$, eq. (12) permits the calculation of the average over the ensemble of phases. With (12), one easily calculates the following average:

$$\langle\exp\{\mathrm{i}n(\varphi-\varphi_0)\}\rangle = \frac{1}{2\pi}\int_0^{2\pi}\mathrm{d}\varphi_0\int_0^{2\pi}\frac{1}{2\pi} \times \sum_{m=-\infty}^{\infty}\exp\{\mathrm{i}(n+m)(\varphi-\varphi_0) - m^2\Gamma_L t\}\,\mathrm{d}\varphi$$

$$= \exp(-n^2\Gamma_L|t|), \tag{13}$$

occurring in $n$-photon processes, as seen from the form of the hamiltonian (7). The preceding simple calculation suffices to convince us that, in the $n$-photon case, the influence of the finite width of the laser line is stronger than in the one-photon case. The effective linewidth is, then, equal to $n^2\Gamma_L$.

## 4. Optical Bloch equations with fluctuating phase

Using eqs. (1)–(8), one can derive the quantum equations of motion for the spin operators describing the atom, i.e. the optical Bloch equations [1]. For the slowly varying parts of these operators, in our case, these equations take the form:

$$\dot{S}_s^+(t) = (\mathrm{i}\Delta - \tfrac{1}{2}\Gamma)S_s^+(t) + 2\lambda^*\exp\{\mathrm{i}n\varphi(t)S_s^z(t),$$

$$\dot{S}_s^-(t) = (-\mathrm{i}\Delta - \tfrac{1}{2}\Gamma)S_s^-(t) + 2\lambda\exp\{-\mathrm{i}n\varphi(t)\}S_s^z(t),$$

$$\dot{S}_s^z(t) = -\Gamma(S_s^z(t) + \tfrac{1}{2}) \tag{14}$$

$$-\lambda\exp\{-\mathrm{i}n\varphi(t)\}S_s^+(t) - \lambda^*\exp\{\mathrm{i}n\varphi(t)\}S_s^-(t),$$

where $\Delta = \omega_0 - n\omega$ is the detuning parameter and $\lambda = \mathrm{i}\eta^{(n)}r^n$ provides a measure of the magnitude of the coupling between the atom and laser beam. Here, $r$ is assumed as constant i.e. we neglect fluctuations of the beam intensity.

In eqs. (14), by writing the damping constant $\Gamma$, we have taken into account the interaction $H_2$, given by the expression (5). We thus assume that the interaction (5), leading to spontaneous emission in the system, causes radiative damping of the atom with the decay constant $\Gamma$ which, in our case, is equal to the Einstein coefficient $A$.

The slowly varying parts of the spin operators of the atom occurring in (14) are obtained going over to a system of reference, rotating with the frequency $n\omega$:

$$S_s^+(t) = S^+(t)\exp(-in\omega t),$$

$$S_s^-(t) = S^-(t)\exp(in\omega t), \qquad S_s^z(t) = S^z(t). \tag{15}$$

With regard to the phase $\varphi(t)$ occurring in (14) which we consider as a stochastic variable, eqs. (14) are stochastic equations, of the type recently discussed by Agarwal [2] and Wódkiewicz [3] for one-photon resonance on the basis of the theory of multiplicative stochastic processes. We here extend those results to $n$-photon resonance, where the influence of the finite laser linewidth is found to be much stronger.

On introducing the new stochastic processes

$$A^+(t) = S_s^+(t)\exp\{-in\varphi(t)\},$$

$$A^-(t) = S_s^-(t)\exp\{in\varphi(t)\}, \tag{16}$$

into eqs. (14) and applying (10) we arrive at the following equations:

$$\dot{A}^+(t) = (i\Delta - \tfrac{1}{2}\Gamma)A^+(t) + 2\lambda^* S_s^z(t) - in\, f(t) A^+(t),$$

$$\dot{A}^-(t) = (-i\Delta - \tfrac{1}{2}\Gamma)A^-(t)$$

$$+ 2\lambda S_s^z(t) + in\, f(t) A^-(t), \tag{17}$$

$$\dot{S}_s^z(t) = -\Gamma(S_s^z(t) + \tfrac{1}{2}) - \lambda A^+(t) - \lambda^* A^-(t).$$

Applying (11) and (17), one readily obtains exact equations describing the time-evolution of the averages over the ensemble of random phases of the stochastic processes considered here. These equations are of the form:

$$(\mathrm{d}/\mathrm{d}t)\langle A^+(t)\rangle = (i\Delta - \tfrac{1}{2}\Gamma - n^2\Gamma_L)\langle A^+(t)\rangle + 2\lambda^*\langle S_s^z(t)\rangle,$$

$$(\mathrm{d}/\mathrm{d}t)\langle A^-(t)\rangle = (-i\Delta - \tfrac{1}{2}\Gamma - n^2\Gamma_L)\langle A^-(t)\rangle + 2\lambda\langle S_s^z(t)\rangle,$$

$$(\mathrm{d}/\mathrm{d}t)\langle S_s^z(t)\rangle$$

$$= -\Gamma(\langle S_s^z(t)\rangle + \tfrac{1}{2}) - \lambda\langle A^+(t)\rangle - \lambda^*\langle A^-(t)\rangle, \tag{18}$$

at the initial conditions $\langle A^+(0)\rangle = \langle A^-(0)\rangle = 0$, $\langle S_s^z(0)\rangle = -\frac{1}{2}$.

The set of equations (18) can be solved using the Laplace transform method, similarly as done by Torrey [5]. The evolution in time of the quantities searched for is defined by the roots of the following equation:

$$(z+\Gamma)[(z + \tfrac{1}{2}\Gamma + n^2\Gamma_L)^2 + \Delta^2]$$

$$+ 4|\lambda|^2 (z + \tfrac{1}{2}\Gamma + n^2\Gamma_L) = 0. \tag{19}$$

On inspection of the form of (19) one notes that these roots can be obtained from the Torrey equation for a coherent beam [1] on replacing the longitudinal and transversal relaxation times respectively by:

$$1/T_1 = \Gamma, \text{ and } \quad 1/T_2 = \tfrac{1}{2}\Gamma + n^2\Gamma_L. \tag{20}$$

Hence the finiteness of the laser linewidth $\Gamma_L$ affects the transversal relaxation time only; in $n$-photon resonance, the variation thus caused is proportional to $n^2$. This result is in agreement with the result (13), obtained along other lines above.

## 5. Resonance fluorescence spectrum in the $n$-photon resonance case

As an example of the finite laser linewidth effect on resonance processes involving more than one photon, so as to fulfil the condition for resonance, we adduce here an expression for the resonance fluorescence from such a system which we express by the formula [6]:

$$P(\omega, T) \sim 2\,\mathrm{Re}\int_0^T \mathrm{d}t \int_0^t \mathrm{d}\tau$$

$$\times \exp\{i(\omega - \omega_0)\tau\}\, \langle\langle S_s^+(t) S_s^-(t+\tau)\rangle\rangle. \tag{21}$$

Above, the symbol $\langle\langle\ \ \rangle\rangle$ denotes the quantum mechanical mean value over the atomic states and the average over the ensemble of random phases. Applying the Bloch eqs. (14) one can calculate the atomic correlation function occurring in (21) and, thus, the resonance fluorescence spectrum. In general, the latter is asymmetrical; its form, with the finite linewidth taken into account, is to be found in ref. [7] for the case of one-photon resonance. In that of $n$-photon resonance, the spectrum is modified only in that $\Gamma_L$ is everywhere replaced by $n^2\Gamma_L$, as it results from eqs. (19) and (20). The spectrum takes a particularly simple, symmetrical form in the case of exact resonance ($\Delta$=0) and an intense laser beam, meaning that $2|\lambda| \gg \Gamma$ and $2|\lambda| \gg n^2\Gamma_L$. In the stationary case i.e. at $T \to \infty$, eq. (21) takes the three-peak form:

$$P(\omega,T\to\infty)\sim\left\{\frac{1}{2}\frac{\Gamma^2(\Gamma+3n^2\Gamma_L)}{4|\lambda|^2(\Gamma+n^2\Gamma_L)}\frac{1}{(\frac{1}{2}n^2\Gamma_L)^2+(\omega-\omega_0)^2}\right.$$
$$+\frac{\frac{1}{2}(\frac{1}{2}\Gamma+n^2\Gamma_L)}{(\frac{1}{2}\Gamma+n^2\Gamma_L)^2+(\omega-\omega_0)^2}$$
$$+\frac{\frac{3}{16}(\Gamma+n^2\Gamma_L)}{\frac{9}{16}(\Gamma+n^2\Gamma_L)^2+(\omega-\omega_0+2|\lambda|)^2}$$
$$\left.+\frac{\frac{3}{16}\Gamma+n^2\Gamma_L}{\frac{9}{16}(\Gamma+n^2\Gamma_L)^2+(\omega-\omega_0-2|\lambda|)^2}\right\}. \quad (22)$$

For $n=1$, eq. (22) goes over into the expression of Kimble and Mandel [7]. For $n>1$, the change in width of the various lines of the spectrum, which amounts to $n^2\Gamma_L$, is greater than in the case of one-photon resonance. One thus can expect a considerable broadening of the line in processes involving resonances of higher order. With regard to such processes, however, it is less easy to fulfil the strong field condition $2|\lambda|\gg n^2\Gamma_L$. We nonetheless believe that such spectra should be accessible to observation, at least in the two-photon resonance case [8]. A similar finite linewidth effect could also be apparent in the fluorescence spectrum of a three-level atom, albeit with two-photon resonance [9]. Quite recently, the laser bandwidth effect on two-photon ionisation in caesium has also been discussed [10].

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