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Exact Solution for the Influence of Laser Temporal Fluctuations on Resonance Fluorescence

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The influence of the laser fluctuations on resonance fluorescence is treated exactly within the framework of multiplicative stochastic processes. For the phase diffusion model of the laser, I find that, in the limit of strong fields, the ratios of the width and height of the central peak to the side peak are, respectively, $2/3x$ and $3x$, where $x = (\gamma + \gamma_c)/(\gamma + 2\gamma_c)$. The envelope of the intensity correlations is found to decay at the rate $(3\gamma + \gamma_c)/2$.

The theory of resonance fluorescence in single-mode laser fields with definite amplitude and phase is by now fairly standard[1-3] and has recently been investigated by several experimental workers.[4,5] An important problem here having direct bearing on experiments is how the temporal fluctuations of the laser beam affect the characteristics of the resonance fluorescence. In a recent paper,[6] Eberly has developed a theory to take into account the effect of the amplitude fluctuations of the laser beam on the spectrum of resonance fluorescence. In this Letter, I report an exact theory which takes into account the effect of temporal fluctuations of the laser beam on the spectrum of resonance fluorescence, the antibunching effects, and the evolution of the atomic populations, dipole moment, etc. The theory which I present here is for a single two-level atomic system and is easily generalized to multilevel systems,[7] to cases when other relaxation mechanisms, such as collisional relaxation,[8] affect strongly the scattering. The approach in the present work is also applicable to experiments on level crossing with paritally coherent light. The theory should also have applications to laser-induced chemical reactions, since the underlying dynamical equations have very similar structure. From the viewpoint of statistical mechanics, my work provides one example of the very few exactly soluble models.

A fully quantum-electrodynamic theory of resonance fluorescence in nonfluctuating fields is presented in Ref. 2 (see also Refs. 1b, 3). The spectrum calculated there is in agreement with the one calculated by Mollow[1a] earlier. In this approach, the atomic dynamics of a two-level atom (with energy separation ω) interacting with the zero-point fluctuations and an external laser beam is described by

$$\frac{\partial\rho}{\partial t} = -i(\omega-\omega_0)[s^z,\rho] - \gamma(s^+s^-\rho - 2s^-\rho s^+ + \rho s^+ s^-) + \tfrac{1}{2}id[s^+\mathcal{E}(t) + s^-\mathcal{E}^*(t),\rho], \tag{1}$$

where $\mathcal{E}(t)$ is the slowly varying part of the electric field of the laser beam of frequency ω_0, 2γ is equal to Einstein A coefficient. The density operator ρ is in a frame rotating with the frequency of the external field. For fluctuating laser beams, the field $\mathcal{E}(t)$ is a stochasic variable and, thus, I have a case of stochasic Liouvillian; and such cases have been treated in a number of approximate ways in the literature.[9]

Here I treat the phase diffusion model of the laser and obtain exact results using the techniques of multiplicative stochastic processes.[10] In this model the temporal phase fluctuations of the laser beam are described by

$$\mathcal{E}(t) = \mathcal{E}_0 e^{-i\varphi(t)}, \quad \varphi(0) = \varphi_0, \tag{2}$$

where $\mathcal{E}_0$ is nonstochastic, φ_0 is the phase variable which is uniformly distributed, and $\varphi(t)$ is the stochastic phase variable; the dynamics for $\varphi(t)$ is Markovian:

$$\dot{\varphi}(t) = \mu(t), \quad \overline{\mu}(t) = 0, \; \overline{\mu(t_1)\mu(t_2)} = 2\gamma_c \delta(t_1 - t_2), \tag{3}$$

and $\mu(t)$ is a Gaussian random force which is δ correlated. For this model all the correlation functions of $\mathcal{E}(t)$ can be calculated in analytic form. I now introduce the following column matrices

$$\psi_1 = \psi_2{}^* = e^{-i\varphi(t)}\langle s^+\rangle, \quad \psi_3 = \langle s^z\rangle, \quad \psi_4 = 1, \quad \chi(t) = \psi(t) e^{i\varphi(t)}. \tag{4}$$

Using Eqs. (1) and (3), one finds that these column matrices satisfy

$$\dot{\psi} = [A_0 + i\mu(t) A_1]\psi, \quad \dot{\chi} = [A_0 + i\mu(t)(A_1 + 1)]\chi, \tag{5}$$

$$A_0 = \begin{pmatrix} -\gamma & 0 & 2i\alpha & 0 \\ 0 & -\gamma & -2i\alpha & 0 \\ i\alpha & -i\alpha & -2\gamma & -\gamma \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad (A_1)_{ij} = \delta_{ij}(\delta_{i2} - \delta_{i1}), \quad \alpha = \tfrac{1}{2} d\mathcal{E}_0. \tag{6}$$

Since (5) has the form of the equations of multiplicative stochastic processes,[10] the *exact* equations for ψ and χ averaged over the ensemble of $\mu(t)$ are found to be

$$\dot{\overline{\psi}} = -\begin{pmatrix} \gamma+\gamma_c & 0 & -2i\alpha & 0 \\ 0 & \gamma+\gamma_c & 2i\alpha & 0 \\ -i\alpha & i\alpha & 2\gamma & \gamma \\ 0 & 0 & 0 & 0 \end{pmatrix}\overline{\psi}, \quad \dot{\overline{\chi}} = -\begin{pmatrix} \gamma & 0 & -2i\alpha & 0 \\ 0 & \gamma+4\gamma_c & 2i\alpha & 0 \\ -i\alpha & i\alpha & 2\gamma+\gamma_c & \gamma \\ 0 & 0 & 0 & \gamma_c \end{pmatrix}\overline{\chi} \equiv B\overline{\chi}. \tag{7}$$

Equation (7) gives the ensemble-averaged mean values of $\langle s^\pm\rangle$ and $\langle s^z\rangle$. Explicitly, I have for the Laplace transforms of $\overline{\langle s^+\rangle}$ and $\overline{\langle s^z\rangle}$,

$$\hat{\overline{\langle s^z\rangle}} = f^{-1}(z)(z+\gamma+\gamma_c)^2\{\langle s^z(0)\rangle - \gamma/z\}, \tag{8}$$

$$\hat{\overline{\langle s^+\rangle}} = g^{-1}(z)\{(z+\gamma+4\gamma_c)(z+2\gamma+\gamma_c) + 2\alpha^2\}\langle s^+(0)\rangle, \tag{9}$$

where the polynomials f and g are given by

$$f(z) = (z+\gamma+\gamma_c)[(z+2\gamma)(z+\gamma+\gamma_c) + 4\alpha^2], \tag{10}$$

$$g(z) = (z+\gamma)(z+2\gamma+\gamma_c)(z+\gamma+4\gamma_c) + 4\alpha^2(z+\gamma+2\gamma_c). \tag{11}$$

In deriving (8) and (9) one has assumed that the initial values of $\langle s^\pm\rangle$ and $\langle s^z\rangle$ are independent of the distribution of μ. Note that in the limit of infinite correlation time $\gamma_c \to 0$, $f(z) = g(z)$. Thus in presence of laser correlations, the time dependences of $\langle s^z\rangle$ and $\langle s^\pm\rangle$ are governed by two different polynomials. I shall show later that the roots of $f(z)$ are important for the antibunching effects, whereas the roots of $g(z)$ are important for the spectrum of the resonance fluorescence.

It should be noted that the stochastic behavior of the extended system is Markovian and hence I can still use the regression theorem to calculate the two-time correlations. Using (7) and the operator algebra, I get the two-time atomic correlation function

$$\overline{\langle s^+(t+\tau)s^-(t)\rangle} = (e^{B\tau})_{11}\{\tfrac{1}{2} + \overline{\psi_3(t)}\} + \overline{\psi}_2(t)\{(e^{B\tau})_{14} - \tfrac{1}{2}(e^{B\tau})_{13}\}, \tag{12}$$

where the solution for $\overline{\psi}$ is obtained from (7). From (12), the steady-state spectrum $\mathcal{S}(\omega)$ follows:

$$\mathcal{S}(\omega) = \operatorname*{Re}_{z\to i\omega}\Big(g^{-1}(z)(z+\gamma_c)^{-1}\alpha^2[\alpha^2 + \tfrac{1}{2}\gamma(\gamma+\gamma_c)]^{-1}[2\gamma^2(z+\gamma+4\gamma_c) + (z+\gamma_c)\{2\alpha^2 + (z+\gamma+4\gamma_c)(z+3\gamma+\gamma_c)\}]\Big). \tag{13}$$

As before, the spectrum is found to be symmetric. The contribution from the pole at $z = -\gamma_c$ should be subtracted—this is the analog of the coherent contribution which is now broadened because of the finite correlation time of the laser beam. I will give the numerical evaluation of the spectrum (13) [including the transient spectrum (12)] elsewhere. In order to bring out some salient features of the influence of the laser phase fluctuations, I consider the limiting case when the Rabi frequency is much larger than γ (i.e., $2\alpha \gg \gamma$). In this case, the roots of $g(z)$ are simply

$$z = -(\gamma + 2\gamma_c), \quad -\tfrac{3}{2}(\gamma + \gamma_c) \pm 2i\alpha. \tag{14}$$

It is then seen that the ratio of the widths of the central peak to side peak is $\frac{2}{3}(\gamma + 2\gamma_c)/(\gamma + \gamma_c)$, which should be compared with the case of no correlations (i.e., when the widths are in the ratio $1:\frac{3}{2}$). An examination of peak heights shows that the ratio between the central- and the side-peak heights is $3(\gamma + \gamma_c)/(\gamma + 2\gamma_c)$ which should be compared with the factor of 3 in the absence of laser correlations. It is thus clear that in order to see the effects of laser fluctuations, the bandwidth of the laser should be comparable to the natural width; and hence the experiments to look for the effect of laser fluctuations should be done with partially coherent light. In this connection it may be noted here that many experiments on laser spectroscopy are performed with dye lasers, which may have large bandwidths.[11-13]

I now examine the effect of the laser phase fluctuations on the antibunching effects[14] in resonance fluorescence. Since the behavior of the augmented system is Markovian, the result for the intensity correlations follows from a recently proved theorem.[15]

$$\overline{\langle : I(t) I(t+\tau) : \rangle} \propto f(t) g(\tau), \tag{15}$$

where $f(t)$ $[g(t)]$ is the probability of finding the atom in the excited state at time t, if at $t=0$ it was in the state $\rho(0)$ (ground state). Using (7), I have again checked the validity of (15). The explicit result for the second-order intensity correlations is

$$\lim_{t\to\infty} \overline{\langle : I(t) I(t+\tau) : \rangle} / (\lim_{t\to\infty} \overline{\langle I(t) \rangle})^2 - 1 = -e^{-x\tau}[(x/y)\sin y\tau + \cos y\tau],$$

$$x = (3\gamma + \gamma_c)/2, \quad y = 2\alpha\{1 - [(\gamma - \gamma_c)^2/16\alpha^2]\}^{1/2}. \tag{16}$$

One again has antibunching effects. The envelope of oscillations (in the limit $2\alpha \gg \gamma$) now decays faster at the rate $(3\gamma + \gamma_c)/2$.

I would like to emphasize here that I was able to obtain exact results because the model for the laser beam was a rather simple one. For more complicated models, one should resort to other techniques. I have, for example, used the projection operator techniques[16] to derive exact equations of motion for the laser field ensemble-averaged, one-time, and multitime correlation functions. I have further shown that these equations lead, for the phase diffusion model, to the results discussed above. Both amplitude and phase fluctuations of the laser beam can be taken into account by combining the techniques of projection operators and those of this work. A result of this analysis is that the ratio of the widths of the side peak to the center peak is $\frac{3}{2}(\gamma + \gamma_c + \lambda_c/3)/(\gamma + 2\gamma_c)$, where λ_c^{-1} is the correlation time of the amplitude fluctuations and $2\alpha \gg \gamma$.

Finally, I would like to mention that in the above treatment, the laser field has been treated classically—this restriction, however, is easily removed. I hope to discuss this and the above-mentioned generalizations elsewhere.

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