Laser-noise-induced intensity fluctuations in resonance fluorescence

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We present a simple method allowing computation of the intensity-intensity correlation function for the resonance fluorescence emitted by a two-level atom driven by a weak, noisy laser. The method is then applied to the two most commonly used models of the noisy laser light: phase diffusion and chaotic colored noise.

A study of the impact of laser noise on laser-atom interactions has been developed considerably over the past decade.[1] Continuing these investigations, in a recent paper,[2] its authors looked at the laser-noise-induced population fluctuations in a strongly and nearly resonantly driven two-level atom. By looking at the family of models with phase fluctuations they note that the precise value of the population's second moment depends on the fourth-order correlation function of the laser field and therefore may discriminate between different detailed models of the noise. This is of considerable theoretical importance, though any nonlinear-optical process is sensitive to higher-order correlation functions.

A possible experimental verification of the above results would involve a study of the intensity-intensity correlation function of the resonance fluorescence. In fact, such an experiment is underway by R. Jones.[3]

The purpose of this Brief Report is to present a simple method of computation of the intensity-intensity correlation function for arbitrary model of the noise, including an important case of chaotic light. The method, valid for weak field, expresses the intensity-intensity correlation function directly in terms of the fourth-order correlation function of the noise.

Our starting point is the following set of semiclassical Bloch equations for the quantum-mechanical expectation values of the dipole moment operators $\sigma_\pm$ and the inversion σ_z describing a two-level atom with the spontaneous-emission linewidth γ, subject to a time-dependent laser light, detuned by Δ from the atomic resonance, producing the instantaneous Rabi frequency $\Omega(t)$:

$$\begin{aligned}\dot{\sigma}_+ &= (i\Delta-\gamma/2)\sigma_+ - i\frac{\Omega(t)}{2}\sigma_z \,,\\ \dot{\sigma}_- &= (-i\Delta-\gamma/2)\sigma_- + i\frac{\Omega^*(t)}{2}\sigma_z \,,\\ \dot{\sigma}_z &= -\gamma(\sigma_z+1) - i\Omega^*(t)\sigma_+ + i\Omega(t)\sigma_- \,.\end{aligned} \tag{1}$$

They are solved perturbatively up to a second order with respect to the Rabi frequency $\Omega(t)$ with the natural initial conditions for the atom which is initially in its ground state:

$$\sigma_+(0)=\sigma_-(0)=0, \quad \sigma_z(0)=-1 \,. \tag{2}$$

The mean (in the sense of quantum mechanics) intensity of the fluorescence $I(t)$ is proportional to the population of the upper state:

$$I(t) \propto \frac{\sigma_z+1}{2} \,. \tag{3}$$

The perturbative expression for $I(t)$ reads

$$I(t)=\tfrac{1}{4}\int_0^t dt_1 \int_0^{t_1} dt_2 e^{-\gamma(t-t_1)}[e^{(i\Delta-\gamma/2)(t_1-t_2)}\Omega^*(t_1)\Omega(t_2)+\text{c.c.}] \,. \tag{4}$$

As explained in Ref. 2, the resonance fluorescence experiment done with a large number of independently radiating atoms will not exhibit intrinsically quantum fluctuations leading to the phenomenon of antibunching.[4] In fact, all the statistical properties of the time-dependent fluorescence intensity $I(t)$ are determined by the properties of the noisy driving signal $\Omega(t)$. In our Report, we shall study two most commonly used models of the noisy laser light: The phase diffusion model,[5] in which only the phase of the light changes stochastically with time, and the chaotic colored model,[5] in which both intensity and the phase undergo random changes. The phase diffusion model describes an idealized single mode, well-stabilized laser operating far above threshold, while the chaotic colored noise model is appropriate for multimode laser operation.

It is easy to understand that the two models may give different predictions for the intensity fluctuations of the fluorescence signal. Phase diffusing light has its instantaneous frequency changing in time. Hence, during interaction with the two-level atom, the light randomly goes on and off resonance with the atomic transition.

Since the fluorescence cross section exhibits the resonant behavior, the fluorescence must also fluctuate. Note, however, that the phase diffusing light does not have the intensity fluctuations.

In the chaotic, colored light, not only the frequency of the light undergoes random changes but also the intensity fluctuates. In fact, such a light exhibits 100% intensity fluctuations. Therefore on top of the mechanism mentioned above there is also the simple (in a perturbative regime, far from saturation) proportionality of the fluorescence to the changing intensity.

Both above-mentioned models have the same second-order correlation function of the Rabi frequency:

$$\langle \Omega(t)\Omega^*(t')\rangle = \Omega_0^2 e^{-\Gamma|t-t'|} , \tag{5}$$

where Γ is the width of the Lorentzian laser spectral line. Therefore they lead to exactly the same mean intensity of the fluorescence. Its long-time steady-state value $\langle I\rangle$ is given by

$$\langle I\rangle = \lim_{t\to\infty} \langle I(t)\rangle = \frac{\Omega_0^2(\gamma/2+\Gamma)}{2\gamma[(\gamma/2+\Gamma)^2+\Delta^2]} . \tag{6}$$

Our next task is to compute the mean value of the long-time normalized intensity-intensity correlation function $S(T)$ defined as

$$S(T) = \lim_{t\to\infty} \frac{\langle I(t)I(t+T)\rangle - \langle I\rangle^2}{\langle I\rangle^2} . \tag{7}$$

It is obvious that it depends on the fourth-order correlation functions of the field. The two models considered differ here. In the phase diffusion model we have

$$\langle \Omega(t_1)\Omega(t_2)\Omega^*(t_3)\Omega^*(t_4)\rangle = \Omega_0^4 e^{-\Gamma|t_1-t_3|} e^{-\Gamma|t_2-t_4|} (e^{2\Gamma d(t_1,t_3;t_2,t_4)} + e^{-2\Gamma d(t_1,t_3;t_2,t_4)}) , \tag{8}$$

where the function $d(t_1,t_3;t_2,t_4)$ is defined as

$$d(t_1,t_3;t_2,t_4) = \int d\tau\, \chi[t_1,t_3]\chi[t_2,t_4] \tag{9}$$

and

$$\chi[t_1,t_3] = \begin{cases} 1 & \text{if } \tau\in[t_1,t_3] \\ 0 & \text{if } \tau\notin[t_1,t_3] \end{cases} . \tag{10}$$

The function d is simply a length of the overlap of intervals $[t_1,t_3]$ and $[t_2,t_4]$.

For the chaotic, colored light, for which the complex field amplitude is a Gaussian process, we have

$$\langle \Omega(t_1)\Omega(t_2)\Omega^*(t_3)\Omega^*(t_4)\rangle = \Omega_0^4 (e^{-\Gamma|t_1-t_3|} e^{-\Gamma|t_2-t_4|} + e^{-\Gamma|t_1-t_4|} e^{-\Gamma|t_2-t_3|}) . \tag{11}$$

The fourfold integration that is necessary to compute correlation function $S(T)$ is straightforward but tedious. We carried out the integrals using the algebraic computation package MATHEMATICA on Apple Macintosh II.

The intensity-intensity correlation function $S(T)$ for the phase diffusion model is

$$S(T) = \frac{\gamma^2[(\gamma/2+\Gamma)^2+\Delta^2]^2}{(\gamma/2+\Gamma)^2} \mathrm{Re}(A+B) , \tag{12}$$

where

$$A = c\left[\frac{e^{-(\gamma/2+\Gamma-i\Delta)T}}{\gamma/2+\Gamma-i\Delta} - \frac{e^{-\gamma T}}{\gamma}\right] ,$$
$$c = \frac{\Gamma}{2(\gamma/2-i\Delta)(\gamma/2-\Gamma+i\Delta)(\gamma/2+\Gamma-i\Delta)(3\gamma/2+\Gamma-i\Delta)} , \tag{13}$$

and

$$B = d\left[\frac{(\gamma/2+\Gamma)e^{-\gamma T}}{\gamma(3\gamma/2+\Gamma-i\Delta)} - \frac{e^{-(\gamma/2+\Gamma+i\Delta)T}}{3\gamma/2+\Gamma+i\Delta}\right] ,$$
$$d = \frac{\Gamma}{[(\gamma/2+\Gamma)^2+\Delta^2](\gamma+4\Gamma)(\gamma/2-\Gamma-i\Delta)} . \tag{14}$$

Note that our result reproduces the value $S(0)$ computed in Ref. 1.

It is clear that the Fourier transform of this function, which gives the spectrum of intensity fluctuations, consists of three peaks: The central one of width γ and two satellites of width $\Gamma+\gamma/2$ symmetrically displaced by the detuning Δ. On resonance the formula simplifies:

$$S(T) = \frac{4\Gamma^2}{(\gamma/2-\Gamma)(3\gamma+\Gamma)(\gamma+4\Gamma)} \times[\gamma e^{-(\gamma/2+\Gamma)T} - (\gamma/2+\Gamma)e^{-\gamma T}] . \tag{15}$$

The resulting spectrum $W(\omega)$, defined as

$$W(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} S(T) e^{i\omega T} dT \tag{16}$$

at resonance is given by

$$W(\omega)=\left[\frac{2}{\pi}\right]^{1/2}\frac{4\gamma\Gamma^2(\gamma/2+\Gamma)}{\gamma+4\Gamma}\times\frac{1}{[\omega^2+(\gamma/2+\Gamma)^2](\omega^2+\gamma^2)} . \tag{17}$$

Note that the spectrum falls off in the wings as the square of a Lorentzian. This last property is a consequence of the vanishing of the linear term in the power-series expansion of $S(T)$ given by (15) near $T=0$.

For the chaotic colored model, the correlation function $S(T)$ is given by

$$S(T)=\frac{(\gamma/2+\Gamma)^2+\Delta^2}{(\gamma/2+\Gamma)^2[(\gamma/2-\Gamma)^2+\Delta^2]}[(\gamma/2)^2e^{-2\Gamma T}+\Gamma^2e^{-\gamma T}-\gamma\Gamma e^{-(\gamma/2+\Gamma)T}(a\cos\Delta T-b\sin\Delta T)] , \tag{18}$$

where

$$a=\frac{(\gamma/2+\Gamma)^2-\Delta^2}{(\gamma/2+\Gamma)^2+\Delta^2} ,$$

and

$$b=\frac{2\Delta(\gamma/2+\Gamma)}{(\gamma/2+\Gamma)^2+\Delta^2} .$$

We obviously have another three-peak spectrum in this case. The main difference is that the central peak now has two components: one of width γ, and another of width 2Γ. Note that always $S(0)=1$. This fact reflects the Gaussian character of our fluctuating signal and should be contrasted with the size of the fluctuations in the phase diffusion model, where $S(0)$ has a nontrivial dependence on the detuning and goes to zero as the laser bandwidth Γ goes to zero.

Again, at resonance, the formula (15) simplifies and we get

$$S(T)=\frac{1}{(\gamma/2-\Gamma)^2}[(\gamma/2)^2e^{-2\Gamma T}+\Gamma^2e^{-\gamma T}-\gamma\Gamma e^{-(\gamma/2+\Gamma)T}] . \tag{19}$$

The Fourier transform gives the spectrum of intensity fluctuations at resonance:

$$W(\omega)=\left[\frac{2}{\pi}\right]^{1/2}\gamma\Gamma(\gamma/2+\Gamma)\times\frac{\omega^2+\gamma^2+6\gamma\Gamma+4\Gamma^2}{(\omega^2+4\Gamma^2)(\omega^2+\gamma^2)[\omega^2+(\gamma/2+\Gamma)^2]} . \tag{20}$$

We see that again the spectrum falls off in the wings as the square of a Lorentzian. As in the previous case, the correlation function $S(T)$ has a parabolic maximum at $T=0$.

We compare the spectra of intensity fluctuations of the phase diffusing versus chaotic light in Fig. 1.

Finally, for completeness, let us consider light which has two independent components: phase diffusing component $\Omega_p(t)$ and chaotic one $\Omega_c(t)$:

$$\Omega(t)=\Omega_c(t)+\Omega_p(t) , \tag{21}$$

where each of the components is characterized by its strength and its width:

$$\langle\Omega_p^*(t)\Omega_p(t')\rangle=\Omega_0^2\exp(-\Gamma_p|t-t'|) ,$$
$$\langle\Omega_c^*(t)\Omega_c(t')\rangle=x\Omega_0^2\exp(-\Gamma_c|t-t'|) , \tag{22}$$

where the parameter x denotes the relative strength of the components.

We quote only the result for the correlation function at resonance:

$$S(t)=S_p(t)+S_c(t)+S_{pc}(t) , \tag{23}$$

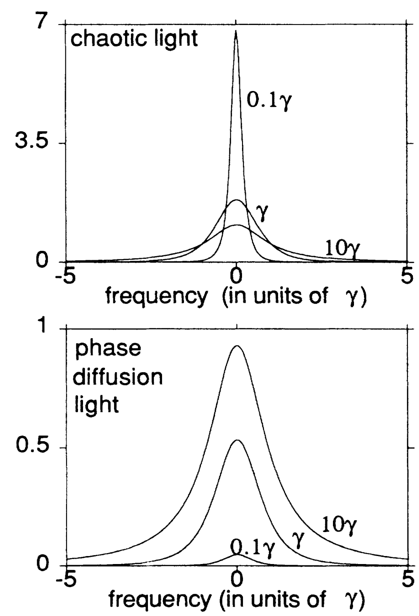

FIG. 1. Spectrum of intensity fluctuations of the resonance fluorescence of a two-level atom driven by the noisy resonant laser light. Comparison of the chaotic light (upper figure) with the phase diffusing light (lower figure) for three different widths of the light measured in the spontaneous width γ. Note the constant amount of fluctuations (area under the spectrum) in the chaotic case and the reduced fluctuations with the decreasing width for the phase diffusing light. Note different vertical scale in the two cases.

where the three components are given by

$$S_p(t)=A[\gamma e^{-(\gamma/2+\Gamma_p)T}-(\gamma/2+\Gamma_p)e^{-\gamma T}]\ , \tag{24}$$

$$S_c(T)=B[(\gamma/2)^2 e^{-2\Gamma_c T}-\gamma\Gamma_c e^{-(\gamma/2+\Gamma_c)T}+\Gamma_c^2 e^{-\gamma T}]\ , \tag{25}$$

$$\begin{aligned} S_{pc}(T)=C[&(\gamma/2)^2 e^{-(\Gamma_c+\Gamma_p)T} \\ &-(\gamma/2)e^{-\gamma T/2}(\Gamma_c e^{-\Gamma_p T}+\Gamma_p e^{-\Gamma_c T}) \\ &+\Gamma_c\Gamma_p e^{-\gamma T}]\ , \end{aligned} \tag{26}$$

with constants A, B, and C:

$$A=\frac{4\Gamma_p(\gamma/2+\Gamma_c)^2}{D(\gamma/2-\Gamma_p)(3\gamma/2+\Gamma_p)(\gamma+4\Gamma_p)}\ , \tag{27}$$

$$B=\frac{x^2(\gamma/2+\Gamma_p)^2}{D(\gamma/2+\Gamma_c)^2}\ , \tag{28}$$

$$C=\frac{2x(\gamma/2+\Gamma_p)(\gamma/2+\Gamma_c)}{D(\gamma/2-\Gamma_p)(\gamma/2-\Gamma_c)}\ , \tag{29}$$

$$D=[\gamma/2+\Gamma_c+x(\gamma/2+\Gamma_p)]^2\ . \tag{30}$$

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[1]For a review of the laser bandwidth problems, see P. Zoller, in *Multiphoton Processes,* edited by P. Lambropoulos and S. J. Smith (Springer-Verlag, Berlin, 1984), Vol. 2, p. 68.

[2]Th. Haslwanter, H. Ritsch, J. Cooper, and P. Zoller, Phys. Rev. A **38**, 5652 (1988).

[3]R. Jones (private communication).

[4]See, for example, L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).

[5]See, for examples, R. Loudon, *Quantum Theory of Light* (Clarendon Press, Oxford, 1983).