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Conditional probability densities for photon emission in resonance fluorescence

Henk F. Arnoldus *, Robertsen A. Riehle

Department of Physics and Astronomy, Mississippi State University, P.O. Drawer 5167, Mississippi State, MS 39762-5167, USA

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ABSTRACT

A two-state atom in a laser beam, near resonance with the atomic transition, emits fluorescent photons. We have obtained the probability density for the emission time of the nth photon, after the emission of an initial photon at time zero. It is shown that the behavior of these probability densities depends strongly on the laser power. For low irradiation, the functions have a single peak with a long tail, but for higher power oscillations are present. The probability density for the first photon has strong oscillations, whereas for subsequent photons the probability density oscillates moderately on a smooth background.

1. Introduction

When a two-state atom is irradiated by a laser beam, near resonance with the atomic transition, the atom will absorb and emit laser photons in stimulated transitions, and emit fluorescent photons in spontaneous transitions from the excited state to the ground state. The fluorescence is electric dipole radiation, emitted in all directions, and is amenable to observation by a photon counter outside the laser beam. The detection of photons is a random process due to the quantum nature of the interaction between the light and the detector, and also due to possible randomness in the light being measured [1,2]. Usually, in such experiments alkali atoms like sodium or cesium are used, and the laser operates in the visible range of the spectrum. Only moderate laser power is needed to observe a sufficient number of photons for the determination of their statistical properties. In typical experiments, correlations between pairs of photons and photon probability distributions are measured. Also of interest is the variance of counting distributions, and their deviation from what would be expected for uncorrelated events (Poisson process). When all the fluorescent light emitted by an atom would be detected by a photon counter, then the counting statistics is the same as the statistics of the emitted photons, and this is the case we shall consider here.

Photons emitted in resonance fluorescence are correlated. After the emission of a fluorescent photon, the atom is in the ground state, and it takes a finite time for the atom to reach the excited state again (by means of stimulated absorption). Since fluorescent photons are emitted during spontaneous decay from the excited state to the ground state, the probability to detect a second photon immediately after the first is zero. This phenomenon is called antibunching, and has been observed experimentally [3–5]. As a direct consequence, the photon statistics for small counting times is sub-poissonian (variance smaller than the average) and the statistics for long counting times is sub-poissonian if photons are antibunched on average over time. Sub-Poisson statistics for long counting times has been observed experimentally for resonance fluorescence [6–10].

A complete account of all statistical properties of the emitted photons (or any random event process [11,12]) is given by the intensity correlation functions, defined as

$$\begin{aligned} &I_k(t_1,t_2,\ldots,t_k)\,\mathrm{d}t_1\,\mathrm{d}t_2\cdots\mathrm{d}t_k \\ &\quad = \text{probability for a photon emission in } [t_1,t_1+\mathrm{d}t_1],\ \text{and } \ldots \\ &\qquad \text{and a photon emission in } [t_k,t_k+\mathrm{d}t_k], \\ &\qquad \text{irrespective of emissions at other times,} \\ &\qquad \text{and with } t_1<t_2<\cdots<t_k. \end{aligned} \tag{1}$$

Antibunching of photons is expressed as $I_2(t_1,t_1)=0$ and the variance of the photon counts in a given time interval can be expressed in terms of $I_2(t_1,t_2)$. Therefore, antibunching and sub-Poisson statistics only involve the second order correlation function. We shall consider the conditional probability densities for the emission times of the photons, and these quantities involve the intensity correlation functions for all k.

* Corresponding author. Tel.: +1 662 325 2919; fax: +1 662 325 8898.
E-mail addresses: hfa1@msstate.edu (H.F. Arnoldus), rar102@ra.msstate.edu (R.A. Riehle).

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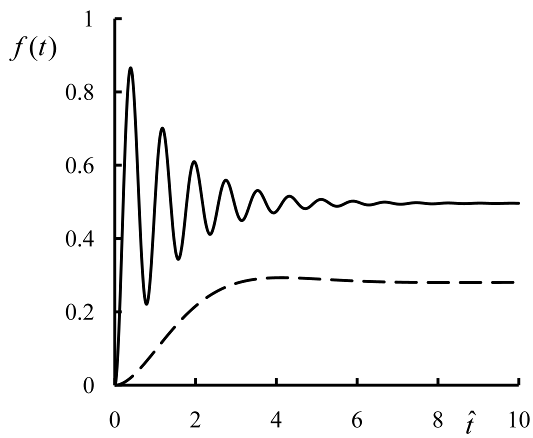

Fig. 1. Shown are graphs of the function $f(t)$, given by Eq. (4), for $\hat{\Omega}=0.8$ (dashed curve) and $\hat{\Omega}=8$ (solid curve).

2. Resonance fluorescence

For resonance fluorescence by an atom in the steady state, the intensity correlations are given by [13,14]

$$I_k(t_1,\ldots,t_k)=A^k f(t_k-t_{k-1})\cdots f(t_2-t_1)n_e,\quad k=2,3,\ldots. \tag{2}$$

Here, A is the Einstein coefficient for spontaneous decay, which equals the inverse lifetime of the excited state, and $f(t)$ is the population of the excited state at time t under the condition that the atom is in the ground state at time zero. The intensity of emission is $I_1(t_1)$, and for the steady state this is independent of time. We shall write $I_1(t_1)=I$, and we have $I=An_e$ with n_e the population of the excited state. We shall assume that the laser frequency is on resonance with the atomic transition, and indicate by Ω the Rabi frequency of the driven atom. We have

$$n_e=\frac{\hat{\Omega}^2}{1+2\hat{\Omega}^2}, \tag{3}$$

where we have set $\hat{\Omega}=\Omega/A$ for the Rabi frequency in units of A. The function $f(t)$ is [15]

$$f(t)=n_e\left\{1-e^{-\frac{3}{4}\hat{t}}\left[\frac{3}{4\rho}\sinh(\rho\hat{t})+\cosh(\rho\hat{t})\right]\right\}, \tag{4}$$

with $\hat{t}=At$ and $\rho=\sqrt{1/16-\hat{\Omega}^2}$. For $\hat{\Omega}>1/4$, the hyperbolic functions become trigonometric functions, and we get oscillations, as illustrated in Fig. 1.

3. Probability densities for photon emission

Let τ_n be the random variable representing the emission time for the nth photon, measured from an initial time $t=0$. The probability density for τ_n is defined as

$$w_n(t)\,\mathrm{d}t=\text{probability that }\tau_n\text{ lies in }[t,t+\mathrm{d}t], \tag{5}$$

and the conditional probability density for τ_n is

$$w_n(t|0)\,\mathrm{d}t=\text{probability that }\tau_n\text{ lies in }[t,t+\mathrm{d}t]\ \text{after an emission in }[-\mathrm{d}t,0]. \tag{6}$$

With $P_n(t)$ the probability for n emissions in $[0,t]$, we have [16]

$$w_n(t)=-\frac{\mathrm{d}}{\mathrm{d}t}\sum_{m=0}^{n-1}P_m(t), \tag{7}$$

and similarly

$$w_n(t|0)=-\frac{1}{I}\frac{\mathrm{d}}{\mathrm{d}t}\sum_{m=1}^{n}w_m(t). \tag{8}$$

We shall consider the conditional probability densities $w_n(t|0)$, since for these functions the initial time is well defined: At $t=0$ the first photon is emitted, and then we consider the arrival time of the nth photon after this initial photon. This function was obtained theoretically for $n=1$ in Ref. [17]. We shall generalize this result by obtaining $w_n(t|0)$ for all n.

The probabilities $P_n(t)$ are determined by the intensity correlation functions through Mandel's photon counting formula [18]. This yields $w_n(t)$ with Eq. (7) and then $w_n(t|0)$ with Eq. (8). The computation of $w_n(t|0)$ is greatly facilitated by adopting a Laplace transform. We set

$$\tilde{w}_n(s|0)=\int_0^\infty e^{-st}w_n(t|0)\,\mathrm{d}t, \tag{9}$$

and following the steps outlined above then yields

$$\tilde{w}_n(s|0)=\left(\frac{A\tilde{f}(s)}{1+A\tilde{f}(s)}\right)^n, \tag{10}$$

in terms of the Laplace transform $\tilde{f}(s)$ of $f(t)$.

4. Computation of $w_n(t|0)$

In order to obtain $w_n(t|0)$, we need to evaluate the Laplace inverse of the right-hand side of Eq. (10). To this end, we first note that the Laplace transform of $f(t)$ is

$$\tilde{f}(s)=\frac{\Omega^2}{2s}\frac{1}{(A+s)(\frac{1}{2}A+s)+\Omega^2}, \tag{11}$$

and this gives

$$\tilde{w}_n(s|0)=\left(\frac{\frac{1}{2}A\Omega^2}{(s+\frac{1}{2}A)[(s+\frac{1}{2}A)^2-(A\gamma)^2]}\right)^n, \tag{12}$$

with $\gamma=\sqrt{1/4-\hat{\Omega}^2}$. Therefore,

$$w_n(t|0)=\left(\frac{1}{2}A\Omega^2\right)^n e^{-\frac{1}{2}At}\mathcal{L}^{-1}\left(\frac{1}{s^n[s^2-(A\gamma)^2]^n}\right). \tag{13}$$

The remaining inverse can be evaluated with the Bromwich integral [19], with result

$$w_n(t|0)=A\frac{2\hat{\Omega}^{2n}}{(2\gamma)^{3n-1}}e^{-\frac{1}{2}\hat{t}}\sum_{k=0}^{n-1}\frac{(\gamma\hat{t})^{n-1-k}}{(n-1-k)!}\times\left\{\frac{1}{2}\left[(-1)^k e^{\gamma\hat{t}}+e^{-\gamma\hat{t}}\right]A_{n-1,k}-B_{n-1,k}\right\}. \tag{14}$$

The universal functions $A_{n,k}$ and $B_{n,k}$ are defined as

$$A_{n,k}=\sum_{m=0}^{k}\binom{n+m}{n}\binom{n+k-m}{n}2^{n-m}, \tag{15}$$

$$B_{n,k}=2^{2n}\sum_{m=0}^{k}\binom{n+m}{n}\binom{n+k-m}{n}(-1)^{n-m}, \tag{16}$$

for $n,k=0,1,2,\ldots$. Expression (14) for $w_n(t|0)$ is the main result of this Letter. It gives the conditional probability density for the emission of the nth photon, after an emission at $t=0$.

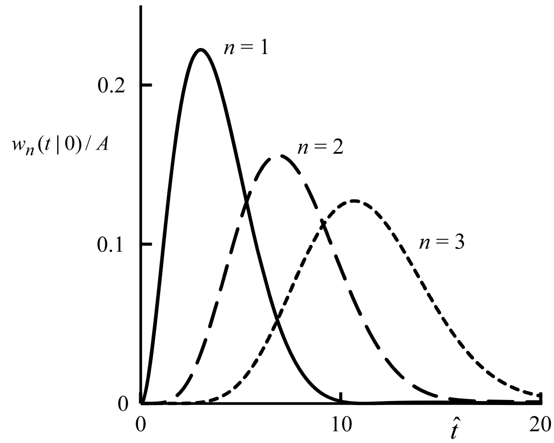

Fig. 2. The figure shows $w_n(t|0)$ for $n = 1, 2$ and 3, and for $\hat{\Omega} = 0.75$.

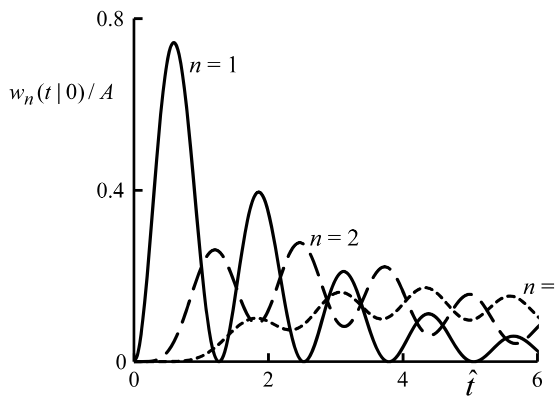

Fig. 3. Shown are graphs of $w_n(t|0)$ for $\hat{\Omega} = 5$.

5. Results

The conditional probability for the first photon follows from Eq. (14) with $n = 1$. With Eqs. (15) and (16) we have $A_{0,0} = B_{0,0} = 1$, so that

$$w_1(t|0) = A\frac{\hat{\Omega}^2}{2\gamma^2}e^{-\frac{1}{2}\hat{t}}\big[\cosh(\gamma\hat{t}) - 1\big], \tag{17}$$

in agreement with Ref. [17]. For $n = 2$ we have $A_{1,0} = 2$, $B_{1,0} = -4$, $A_{1,1} = 6$ and $B_{1,1} = 0$. This gives

$$w_2(t|0) = A\left(\frac{\hat{\Omega}^2}{2\gamma^2}\right)^2 e^{-\frac{1}{2}\hat{t}}\left\{\hat{t}\left[1 + \frac{1}{2}\cosh(\gamma\hat{t})\right] - \frac{3}{2\gamma}\sinh(\gamma\hat{t})\right\}, \tag{18}$$

and so on. Fig. 2 shows $w_n(t|0)$ for $n = 1, 2$ and 3, and for $\hat{\Omega} = 0.75$. Each distribution has a peak and the positions of the peaks increase with n, as it should be. We also see that $w_n(0|0) = 0$, which reflects the antibunching between the first photon and the conditional photon at $t = 0$. Fig. 3 shows the same functions, but for $\hat{\Omega} = 5$, corresponding to a much higher intensity of the driving laser than in Fig. 2. The functions are now oscillatory, with no clear peak anymore. We also notice that the functions have a long tail at the high t end. Fig. 4 shows $w_n(t|0)$ for $n = 1$ and 3, with $\hat{\Omega} = 15$. The function $w_1(t|0)$ has very fast oscillations with a large amplitude, and it has numerous zeros (where $\cosh(\gamma\hat{t}) = 1$, and here γ is positive imaginary). The oscillations in $w_3(t|0)$ are also fast, but very small in amplitude.

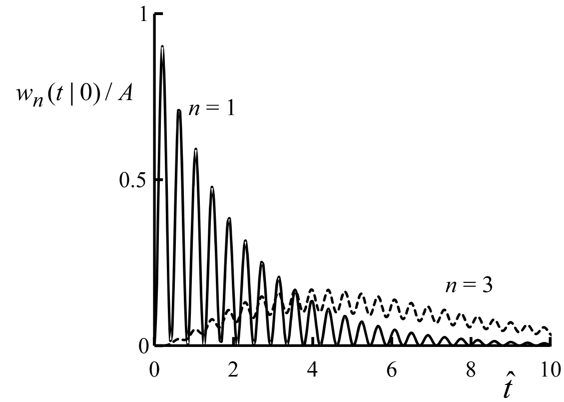

Fig. 4. The figure shows $w_1(t|0)$ and $w_3(t|0)$ for $\hat{\Omega} = 15$, corresponding to very strong illumination of the atom.

6. The functions $A_{n,k}$ and $B_{n,k}$

Eqs. (15) and (16) define the functions $A_{n,k}$ and $B_{n,k}$ that appear in the solution for $w_n(t|0)$. In this section we shall derive some properties of these functions. First, for $k = 0$ there is only one term, and we get immediately

$$A_{n,0} = 2^n, \qquad B_{n,0} = (-1)^n 2^{2n}. \tag{19}$$

If we make the substitution $m' = k - m$ in Eq. (16) we get $B_{n,k} = (-1)^k B_{n,k}$ and therefore

$$B_{n,k} = 0, \quad k \text{ odd}. \tag{20}$$

We now construct generating functions in k, for fixed n. From Eqs. (15) and (16) we derive

$$\sum_{k=0}^{\infty} A_{n,k}y^k = \frac{2^{2n+1}}{[(1-y)(2-y)]^{n+1}}, \tag{21}$$

$$\sum_{k=0}^{\infty} B_{n,k}y^k = \frac{(-1)^n 2^{2n}}{(1-y^2)^{n+1}}, \tag{22}$$

both convergent around $y = 0$. Then we differentiate Eq. (21) with respect to y, regroup, and then equate equal powers of y. This yields a three-term recursion relation between $A_{n,k}$'s with different k:

$$2kA_{n,k} = 3(n+k)A_{n,k-1} - (2n+k)A_{n,k-2}, \quad k = 2, 3, \ldots. \tag{23}$$

The initial values are $A_{n,0} = 2^n$ and

$$A_{n,1} = 3(n+1)2^{n-1}. \tag{24}$$

In the same way we find from Eq. (22)

$$B_{n,k} = \frac{2n+k}{k}B_{n,k-2}, \quad k = 2, 3, \ldots. \tag{25}$$

For k odd we have $B_{n,k} = 0$, and for k even, the solution of Eq. (25) is

$$B_{n,2m} = (-1)^n 2^{2n}\binom{n+m}{n}, \quad m = 0, 1, \ldots. \tag{26}$$

Many other relations for the functions $A_{n,k}$ and $B_{n,k}$ can be found. For instance, $A_{0,k} = 2 - 2^{-k}$. With considerable more effort, various sum rules for the function $A_{n,k}$ can be derived. We mention without proof:

$$\frac{1}{2}\left[(-1)^n+(-1)^p\right]\sum_{\ell=0}^{p}(-1)^\ell\binom{p}{\ell}A_{n,n-\ell}=B_{n,n-p},\quad p=0,1,\ldots,n, \tag{27}$$

$$\left[(-1)^n+(-1)^p\right]\sum_{\ell=0}^{n}(-1)^\ell\binom{p}{\ell}A_{n,n-\ell}=0,\quad p=n+1,n+2,\ldots,3n+1, \tag{28}$$

$$\sum_{\ell=0}^{n}(-1)^{n-\ell}\binom{3n+2}{\ell}A_{n,n-\ell}=2^{2n}. \tag{29}$$

Setting $p=0$ in Eq. (27) yields

$$A_{n,n}=B_{n,n},\quad n\text{ even}. \tag{30}$$

7. Conclusions

The dynamics of a two-state atom in a laser beam is arguably the most important problem in quantum optics. We have studied the conditional probability densities $w_n(t|0)$ for emissions of fluorescent photons by this atom. The initial observation time $t=0$ is set by the emission of the first photon, and then $w_n(t|0)$ gives the probability density for the emission of the nth photon after the initial one. Eq. (14) gives the explicit result for $w_n(t|0)$, and Figs. 2–4 illustrate the behavior of these functions. For small to moderate laser power, these functions simply have a peak, as shown in Fig. 2. For higher power, oscillations set in, and $w_1(t|0)$ has persisting oscillations with a large amplitude. For larger n there only appear small oscillations on a smooth background, as seen in Fig. 4.

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