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# Photon statistics of fluorescence radiation

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**Abstract.** We study the general properties of the photon statistics of fluorescence radiation, emitted by a two-level atom in a strong laser field and a perturber bath. We investigate the deviation of the factorial moments from a Poisson distribution, and we show that for long counting times the lowest-order correction can be expressed entirely by the quantity $\bar{Q}$, which represents the deviation of the variance from the average. We introduce and evaluate the quantities $\beta_n(t)$, which serve as a measure for the deviation of the higher-order statistics from Poissonian statistics. Subsequently we obtain explicit expressions for the average waiting time for the appearance of the $n$th photon, after an arbitrary initialization of the counting process. It turns out that the average time delay is again determined by $\bar{Q}$. Hence this parameter can be measured in a photon-counting experiment, which involves only the observation of a single photon, rather than an (in principle) infinite number of photons.

## 1. Introduction

We consider a two-level atom in a strong single-mode laser field, and surrounded by a perturber gas. The incident laser photons are scattered by the atom as dipole radiation [1–3]. The statistical distribution of the emitted photons is determined by the dynamics of the atom and its interaction with the laser field, and is modified by collisional effects. Temporal photon correlations and the photon statistics therefore carry information on the dynamics of the radiating system. In particular, it is well known that fluorescent photons tend to be emitted separated in time, this is termed antibunching. This characteristic feature of fluorescence radiation can be understood immediately from the mechanism of photon emission. After an emission, the atom is bound to be in its ground state, which prohibits a second photon being emitted immediately afterwards. Conversely, long after the first emission the memory of the system will be lost, and the probability for an emission will become independent of the initial one. Hence the conditional probability $f(t)\,\mathrm{d}t$ for an emission in the time interval $[t, t+\mathrm{d}t]$, after an emission in $[-\mathrm{d}t, 0]$, will have the properties

$$f(0)=0, \quad f(\infty)=I, \tag{1}$$

with $I$ the intensity, or $I\,\mathrm{d}t$ the unconditional probability for a photon emission in $[t, t+\mathrm{d}t]$. The function $f(t)$ has been evaluated by several methods [4–6], and has been measured in an atomic-beam experiment [7–9]. The notion of antibunching and its physical significance was reviewed by Walls [10] and Paul [11].

Besides the two-photon correlation function $f(t)$, we can also consider the statistics of the emitted photons. If the process of photon emission were completely random (a Poisson process), the average number of photons $\mu(t)$, emitted in the time interval $[0, t]$, would equal the variance $\sigma^2(t)$ of the photon-number distribution. Since fluorescent photons are correlated, the photon statistics will deviate from the Poisson distribution. In order to study this feature of fluorescence radiation, Mandel [12] introduced the $Q(t)$ factor, which is defined as

$$\sigma^2(t) = \mu(t)[1 + Q(t)]. \tag{2}$$

Since the variance is non-negative, we have the constraint $Q(t) \geqslant -1$ for any photon distribution. For a Poisson distribution we have $Q(t) = 0$, and if the radiation field would have a classical analogue, we would have the restriction $Q(t) \geqslant 0$ [12]. Therefore, Mandel stressed that any observation of $Q(t) < 0$ would indicate that the fluorescence radiation field is essentially a quantum field. A negative $Q$-factor reflects sub-Poisson statistics, which means that the variance is smaller than the average. We pointed out in a previous paper [13] that a negative $Q$-factor results from an average antibunching of the photons and we evaluated its limit for long counting times,

$$\bar{Q} = \lim_{t\to\infty} Q(t), \tag{3}$$

in terms of the atomic parameters. The behaviour of $Q(t)$ as a function of time was obtained by Singh [14]. Short and Mandel [15] measured $\bar{Q}$ in their beam experiment, and indeed it appeared to be negative.

The $Q(t)$ factor from equation (2) measures the deviation of the variance from its Poissonian limit. In this paper we will extend this idea to the higher-order statistics. It will appear that the deviation from Poisson statistics, in the limit of long counting times, is again determined by $\bar{Q}$. Hence an accurate determination of the photon statistics of atomic fluorescence requires the measurement of $\bar{Q}$ as a function of the atomic parameters (dipole strength, laser intensity, laser linewidth, etc.). This experiment is rather difficult, because the large number of photons, which are detected in $[0, t]$ with $t \to \infty$, must all be emitted by the same atom. In this paper we propose a different experiment for the measurement of $\bar{Q}$, which involves only the observation of a single photon.

## 2. Counting statistics

The number $n(t)$ of detected photons in $[0, t]$ is a stochastic function of time. If we denote by $P_n(t)$, with $n = 0, 1, 2, \ldots$, the probability for the detection of $n$ photons in $[0, t]$, then we have the obvious properties

$$0 \leqslant P_n(t) \leqslant 1, \quad \sum_{n=0}^{\infty} P_n(t) = 1. \tag{4}$$

The average and the variance of the photon-number distribution can be written as

$$\mu(t) = \langle n\rangle(t), \quad \sigma^2(t) = \langle(n - \langle n\rangle)^2\rangle(t), \tag{5}$$

where the angle brackets indicate averaging with the set of probabilities $P_n(t)$. The factorial moments,

$$S_k(t) = \sum_{n=k}^{\infty} \frac{n!}{(n-k)!} P_n(t), \quad k = 0, 1, 2, \ldots \tag{6}$$

which gives in particular

$$S_0(t)=1, \quad S_1(t)=\mu(t), \quad S_2(t)-S_1(t)^2=\sigma^2(t)-\mu(t), \tag{7}$$

are quite convenient for the study of the deviation from Poisson statistics. According to (2), $Q(t)$ can be expressed in $S_1(t)$ and $S_2(t)$.

Next we introduce the generating function [16]

$$G(x;t)=\langle x^n\rangle(t)=\sum_{n=0}^{\infty} x^n P_n(t), \tag{8}$$

so that the $x$-dependence of $G(x;t)$ determines the set $P_n(t)$ as the Taylor coefficients in an expansion around $x=0$. On the other hand, if we differentiate $G(x;t)$ $k$ times with respect to $x$, and subsequently set $x$ equal to 1, we obtain precisely expression (6) for the factorial moments. So the $S_k(t)$s are the Taylor coefficients of $G(x;t)$ around $x=1$ and we can write

$$G(x;t)=\sum_{k=0}^{\infty}\frac{(x-1)^k}{k!}S_k(t). \tag{9}$$

A comparison of (8) and (9) yields

$$P_n(t)=\frac{1}{n!}\sum_{k=0}^{\infty}\frac{(-1)^k}{k!}S_{n+k}(t) \tag{10}$$

which is the inverse formula of equation (6). Both the sets $P_n(t)$ and $S_k(t)$ determine the statistics completely.

We introduce another quantity, defined as

$$F(x;t)=\exp\left[(1-x)\mu(t)\right]G(x;t). \tag{11}$$

The Taylor expansion around $x=1$

$$F(x;t)=\sum_{n=0}^{\infty}\frac{(x-1)^n}{n!}\beta_n(t), \tag{12}$$

defines the set $\beta_n(t)$ for $n=0,1,2,\ldots$ . The relation between $\beta_n$ and $S_k$ is obtained by substituting expansion (9) for $G(x;t)$ and taking the $n$th derivative. If we then set $x=1$ we find

$$\beta_n(t)=\sum_{k=0}^{n}\binom{n}{k}(-\mu(t))^{n-k}S_k(t). \tag{13}$$

In a similar way we obtain from the Taylor expansion of $G(x;t)=F(x;t)\times\exp\left[(x-1)\mu(t)\right]$ the inverse equation

$$S_n(t)=\sum_{k=0}^{n}\binom{n}{k}\mu(t)^{n-k}\beta_k(t), \tag{14}$$

which shows that a given set $\beta_n(t)$ also fixes the statistics. From (13) we find in particular

$$\beta_0(t)=1, \quad \beta_1(t)=0, \quad \beta_2(t)=\sigma^2(t)-\mu(t), \tag{15}$$

so that $\beta_2(t)$ is related to the $Q(t)$ factor.

## 3. The Poisson distribution

The Poisson distribution is defined as

$$P_n(t) = \frac{\mu(t)^n}{n!} \exp(-\mu(t)) \tag{16}$$

in terms of an arbitrary function $\mu(t) \geqq 0$. From (6) we then find the factorial moments to be

$$S_k(t) = \mu(t)^k \tag{17}$$

and with (8) we obtain the generating function

$$G(x; t) = \exp[(x-1)\mu(t)]. \tag{18}$$

The function $F(x; t)$ and the $\beta_n(t)$s are then

$$F(x; t) = 1 \tag{19}$$

$$\beta_n(t) = \delta_{n,0}. \tag{20}$$

This clearly reveals the significance of the set $\beta_n(t)$: a non-zero value of $\beta_n(t) (n > 1)$ reflects a deviation from Poissonian statistics. Especially a non-zero $\beta_2(t)$ is equivalent to a non-zero $Q(t)$, so the definition of the $\beta_n(t)$s can be viewed as a generalization of the $Q$-factor (apart from a normalization). Furthermore $\beta_n(t)$ is determined by $S_1(t), \ldots, S_n(t)$ and does not depend on higher-order moments. This implies that the series $\beta_n(t)$ describes successively the deviation of $S_n(t)$ from their Poissonian value (17). For $n = 1$ we have $S_1(t) = \mu(t)$, so that $\beta_1(t) = 0$ by definition. For $n = 2$ we have

$$\beta_2(t) = \mu(t)Q(t) \tag{21}$$

$$S_2(t) = S_1(t)^2 + \beta_2(t). \tag{22}$$

## 4. Random events

The process of photon counting can be regarded as the observation of dots or random events on the time axis. The statistics of these events is determined by the Stratonovich distribution functions $I_k(t_1, \ldots, t_k) (k \geqslant 1)$, where $I_k(t_1, \ldots, t_k)$ $\mathrm{d}t_1 \ldots \mathrm{d}t_k$ is the probability for the occurrence of an event in $[t_1, t_1 + \mathrm{d}t_1], \ldots$ and an event in $[t_k, t_k + \mathrm{d}t_k]$, irrespective of possible events at other times [17]. The set of distribution functions determines the factorial moments according to

$$S_k(t) = k! \int_0^t \mathrm{d}t_k \int_0^{t_k} \mathrm{d}t_{k-1} \ldots \int_0^{t_2} \mathrm{d}t_1 I_k(t_1, \ldots, t_k) \tag{23}$$

and thereby the complete statistics of the number of events for each value of the counting time $t$. From (23) we find the initial values

$$S_k(0) = \delta_{k,0}, \quad P_n(0) = \delta_{n,0}, \quad \beta_n(0) = \delta_{n,0}. \tag{24}$$

Suppose we have a classical stochastic electromagnetic field with intensity $\hat{I}(t)$ incident upon a photomultiplier tube. Then the distribution functions are given by the intensity correlation functions [18, 19]

$$I_k(t_1, \ldots, t_k) = \{\hat{I}(t_1) \ldots \hat{I}(t_k)\}, \tag{25}$$

where $\{\ldots\}$ denotes an average over the stochastic process $\hat{I}(t)$. The average number of events for a given realization of $\hat{I}(t)$ is

$$\hat{\mu}(t) = \int_0^t \hat{I}(s)\,\mathrm{d}s \tag{26}$$

which itself is a stochastic process. With (23) we then find the factorial moments to be

$$S_k(t) = \langle\!\langle \hat{\mu}(t)^k \rangle\!\rangle, \tag{27}$$

and in particular

$$S_1(t) = \mu(t) = \langle\!\langle \hat{\mu}(t) \rangle\!\rangle. \tag{28}$$

If the incident field $\hat{I}(t)$ is not stochastic, we have $\mu(t) = \hat{\mu}(t)$, so $S_k(t) = \mu(t)^k$, and the number of events has a Poisson distribution.

With equation (13) we now find

$$\beta_n(t) = \langle\!\langle (\hat{\mu}(t) - \mu(t))^n \rangle\!\rangle, \tag{29}$$

which reduces to $\beta_n(t) = \delta_{n,0}$ for a non-stochastic field $\hat{I}(t)$. From equation (29) we immediately find the restriction

$$\beta_n(t) \geqslant 0 \quad \text{for } n = 0, 2, 4, \dots \tag{30}$$

for photon detection from a classical field. This generalizes $Q(t) \geqslant 0$, which is (30) for $n = 2$. Notice that equation (29) sets no lower bound for $n$ odd.

## 5. Fluorescence radiation

The photon statistics of fluorescence radiation, emitted by a two-level atom, is governed by the conditional probability density $f(t)$ for a photon emission at time $t$, after an emission at time zero. This $f(t)$ equals the population of the excited state at time $t$, with the condition that the atom is in the ground state at time zero. We suppose that the atom has spent a sufficiently long time in the laser field, so that it has reached a steady state. Then the distribution functions for the photon emissions are given by [5, 20]

$$I_k(t_1, \dots, t_k) = f(t_k - t_{k-1}) \dots f(t_3 - t_2) f(t_2 - t_1) I, \quad k > 1 \tag{31}$$

for $t_k \geqslant \dots \geqslant t_1$, and $I_1(t) = I$. From (23) we find the average number of detected photons in $[0, t]$ to be

$$\mu(t) = S_1(t) = It. \tag{30}$$

The second factorial moment is

$$S_2(t) = 2I \int_0^t (t - \tau) f(\tau)\,\mathrm{d}\tau \tag{33}$$

and combination of (32) and (33) determines the $Q(t)$-factor [13].

The higher-order statistics is most easily found in the Laplace domain. If we make the transformation

$$\tilde{P}_n(s) = \int_0^\infty \exp(-st) P_n(t)\,\mathrm{d}t, \tag{34}$$

and similarly for other time-dependent quantities, then we find from (31) and (23) the factorial moments [21]

$$\tilde{S}_0(s) = \frac{1}{s}, \quad \tilde{S}_k(s) = \frac{I^k k!}{s^{k+1}} \left( \frac{s\tilde{f}(s)}{I} \right)^{k-1}, \quad k \geq 1. \tag{35}$$

With (10) we obtain the probabilities

$$\tilde{P}_0(s) = \frac{1}{s} - \frac{1}{s^2} \frac{I}{1+\tilde{f}(s)}$$
$$\tilde{P}_n(s) = \frac{I}{s^2} \frac{\tilde{f}(s)^{n-1}}{(1+\tilde{f}(s))^{n+1}}, \quad n \geq 1, \tag{36}$$

and the generating function

$$G(x; s) = \frac{1}{s} + \frac{1}{s^2} \frac{(x-1)I}{1-(x-1)\tilde{f}(s)} \tag{37}$$

follows from (9). The relation (11) between $F(x;t)$ and $G(x;t)$ is in the Laplace domain

$$\tilde{F}(x; s) = \tilde{G}(x; s + (x-1)I). \tag{38}$$

These results determine the photon statistics of fluorescence radiation.

The property $f(\infty) = I$ from equation (1) becomes in the Laplace domain

$$\lim_{s \to 0} s\tilde{f}(s) = I. \tag{39}$$

This implies that we can write

$$\frac{s\tilde{f}(s)}{I} = 1 + \tilde{g}(s) \quad \text{with} \quad \tilde{g}(0) = 0, \tag{40}$$

which defines the function $\tilde{g}(s)$. With (40) we can expand $\tilde{S}_k(s)$ from (35) around $s = 0$, and the Laplace inverse then corresponds to an expansion of $S_k(t)$ around $t = \infty$. This yields the factorial moments for long counting times

$$S_k(t) = (It)^k (1 + \mathcal{O}(t^{-1})), \quad t \to \infty. \tag{41}$$

The leading term $(It)^k$ is the Poisson limit, which will be reached for $t$ sufficiently large. The vanishing component $\mathcal{O}(t^{-1})$ accounts for the deviation from Poisson statistics. Expansion (41) indicates that a deviation from Poisson statistics will be difficult to observe in a photon-counting experiment, in which simply the number of photons in $[0, t]$ is registered.

For later purposes we recall that $\bar{Q}$ can be expressed in $\tilde{g}(s)$ as [13]

$$\bar{Q} = 2I \lim_{s \to 0} \frac{\mathrm{d}}{\mathrm{d}s} \tilde{g}(s). \tag{42}$$

## 6. Deviation from Poisson statistics

It was pointed out in § 2 that non-zero values of $\beta_n(t)$ for $n \geq 2$ indicate a deviation from a Poisson distribution. In this section we will evaluate $\tilde{\beta}_n(s)$ explicitly. The Laplace transform of (13) reads

$$\tilde{\beta}_n(s) = \sum_{k=0}^{n} \binom{n}{k} \left( I \frac{\mathrm{d}}{\mathrm{d}s} \right)^{n-k} \tilde{S}_k(s). \tag{43}$$

If we now substitute $\tilde{S}_k(s)$ from equation (35), apply equation (40) and perform the summations, we find

$$\tilde{\beta}_n(s) = \frac{\delta_{n,0}}{s} + \frac{n!\,I^n}{s^{n+1}} \sum_{k=0}^{n-2} \frac{s^k}{k!} \frac{\mathrm{d}^k}{\mathrm{d}s^k} \frac{\tilde{g}(s)^{n-k} + \tilde{g}(s)(-1)^{n-k}}{1+\tilde{g}(s)}, \tag{44}$$

which can also be written as

$$\tilde{\beta}_n(s) = \frac{\delta_{n,0}}{s} + \frac{n!\,I^n}{s^{n+1}} \sum_{k=0}^{n-2} \sum_{l=1}^{n-k-1} (-1)^{n+k+l+1} \frac{s^k}{k!} \frac{\mathrm{d}^k}{\mathrm{d}s^k} \tilde{g}(s)^l. \tag{45}$$

Here the summations account for the deviation from Poisson statistics.

The behaviour of $\beta_n(t)$ for $t \to \infty$ is governed by the expansion of (45) around $s=0$. From

$$s^k \frac{\mathrm{d}^k}{\mathrm{d}s^k} \tilde{g}(s)^l = \mathcal{O}(s^{\max(k,l)}) \tag{46}$$

we find $\tilde{\beta}_n(s) = \delta_{n,0}/s + \mathcal{O}(s^{-n})$, which corresponds to $\beta_n(t) = \delta_{n,0} + \mathcal{O}(t^{n-1})$. This also follows from a substitution of (41) into (13), since the Poisson terms cancel. We will now show that the $\beta_n(t)$s do not diverge as $\mathcal{O}(t^{n-1})$ for $t \to \infty$, but much slower, due to the exact cancelation of many low-order terms in equation (45).

To this end we notice that

$$\sum_{k=0}^{\infty} (-1)^{k+l} \frac{s^k}{k!} \frac{\mathrm{d}^k}{\mathrm{d}s^k} \tilde{g}(s)^l = (-\tilde{g}(0))^l = \delta_{l,0}. \tag{47}$$

Then we can write equation (45) as

$$\tilde{\beta}_n(s) = \frac{\delta_{n,0}}{s} + \frac{(-1)^n n!\,I^n}{s^{n+1}} \sum_{l=1}^{n-1} \sum_{k=n-1}^{\infty} (-1)^{k+l} \frac{s^k}{k!} \frac{\mathrm{d}^k}{\mathrm{d}s^k} \tilde{g}(s)^l, \tag{48}$$

and with (46) we find that the double sum behaves as $\mathcal{O}(s^{n'})$ with $n' = \frac{1}{2}n$ for $n$ even and $n' = \frac{1}{2}n + \frac{1}{2}$ for $n$ odd. The lowest order contribution in $s$ then follows from $k = l = n'$ for $n$ even. For $n$ odd however, the three terms with $k = l = n'$, $k = l + 1 = n'$ contribute. If we define the Taylor coefficients of $\tilde{g}(s)$ as

$$\zeta_n = \lim_{s \to 0} \frac{\mathrm{d}^n}{\mathrm{d}s^n} \tilde{g}(s), \tag{49}$$

then we can expand (48) around $s=0$ in terms of the $\zeta_n$s. Keeping only the lowest-order terms and transforming back to the time domain then yields the long-time behaviour of $\beta_n(t)$. We find

$$\begin{aligned} \beta_{2k}(t) &= \frac{(2k)!}{k!} (I^2\zeta_1 t)^k (1 + \mathcal{O}(t^{-1})) \\ \beta_{2k+1}(t) &= \frac{(2k+1)!}{(k-1)!} (I^2\zeta_1 t)^k I\zeta_1 \left(1 + \frac{\zeta_2}{2\zeta_1{}^2}\right)(1 + \mathcal{O}(t^{-1})) \end{aligned} \tag{50}$$

for $k = 1, 2, 3, \ldots$. Hence the $\beta_n(t)$s grow as $\mathcal{O}(t^{n/2})$ for $n$ even and as $\mathcal{O}(t^{n/2-1/2})$ for $n$ odd.

With (39) we notice that

$$\bar{Q} = 2I\zeta_1, \tag{51}$$

so the leading term in $\beta_{2k}(t)$ is completely determined by $\bar{Q}$. The long-time behaviour of $\beta_n(t)$ for $n$ odd however, involves also the parameter $\zeta_2$. For photon detection from a classical field, we have the constraint (30) for $\beta_n(t)$ with $n$ even. Sub-Poisson statistics is usually defined as a violation of the inequality (30) for $n=2$. From (50) we then find that (30) is also violated for $n=6, 10, 14, \ldots$, but not for $n=8, 12, 16, \ldots$ .

Substitution of (50) into (14) gives the long-time behaviour of the factorial moments. We find

$$S_k(t)=\mu(t)^k(1+k(k-1)\bar{Q}/2It+\mathcal{O}(t^{-2})), \quad t\to\infty \tag{52}$$

for $k=0,1,2,\ldots$ . This result expresses that the asymptotic deviation from Poisson statistics of the factorial moments is completely determined by $\bar{Q}$.

## 7. Waiting times

The set $P_n(t)$ for a given time $t$ gives the probability for the detection of $n$ photons in the time interval $[0,t]$. From the time dependence of $P_n(t)$, we can deduce the probability distribution for the detection time of the $n$th photon. To this end, we introduce the probability densities $w_n(t)$ as [16]

$$w_n(t)\,\mathrm{d}t = \text{probability that the } n\text{th photon is detected in } [t, t+\mathrm{d}t], \quad n\geqslant 1, \tag{53}$$

which is identical to the probability for the detection of $n-1$ photons in $[0,t]$ and one photon in $[t,t+\mathrm{d}t]$. From the fact that we can only detect one photon or no photon in an infinitesimal time interval, we derive immediately the relations

$$\frac{\mathrm{d}}{\mathrm{d}t}P_0(t)=-w_1(t),$$
$$\frac{\mathrm{d}}{\mathrm{d}t}P_n(t)=w_n(t)-w_{n+1}(t), \qquad n\geqslant 1. \tag{54}$$

The solution of these equations is

$$w_n(t)=-\frac{\mathrm{d}}{\mathrm{d}t}\sum_{k=0}^{n-1}P_k(t), \tag{55}$$

which expresses the $w_n(t)$s with $k<n$. The inverse relation follows from integration of (54)

$$P_0(t)=1-\int_0^t w_1(\tau)\,\mathrm{d}\tau,$$
$$P_n(t)=\int_0^t w_n(\tau)\,\mathrm{d}\tau-\int_0^t w_{n+1}(\tau)\,\mathrm{d}\tau, \qquad n\geqslant 1 \tag{56}$$

so the set $w_n(t)$ also fixes the statistics.

From (53) we have the obvious interpretation

$$\int_0^t w_n(\tau)\,\mathrm{d}\tau = \text{probability that the } n\text{th photon is detected in } [0,t], \tag{57}$$

and with (55) we find

$$\int_0^t w_n(\tau)\,\mathrm{d}\tau=1-\sum_{k=0}^{n-1}P_k(t)=\sum_{k=n}^{\infty}P_k(t), \tag{58}$$

which expresses that the probability to detect the $n$th photon in $[0, t]$ equals the probability to detect $n$ or more photons in $[0, t]$.

The set $P_n(t)$ is uniquely related to the set $S_k(t)$, so the set $w_n(t)$ must also be uniquely related to the set $S_k(t)$. With (6) we find

$$s_0(t) = 1,$$
$$S_k(t) = k \sum_{m=0}^{\infty} \frac{(m+k-1)!}{m!} \int_0^t w_{m+k}(\tau)\,\mathrm{d}\tau, \qquad k \geqq 1, \tag{59}$$

which has the inverse relation

$$\int_0^t w_k(\tau)\,\mathrm{d}\tau = \frac{1}{(k-1)!} \sum_{m=0}^{\infty} (-1)^m \frac{(m+k-1)!}{m!} S_{m+k}(t). \tag{60}$$

The generating function $G(x; t)$ becomes in terms of $w_n(t)$

$$G(x; t) = 1 + \frac{x-1}{x} \sum_{m=1}^{\infty} x^m \int_0^t w_m(\tau)\,\mathrm{d}\tau. \tag{61}$$

Let us consider the Poisson distribution (16). It is easy to check that the $P_n(t)$s obey the relations

$$\frac{\mathrm{d}}{\mathrm{d}t} P_0(t) = -I(t)\,P_0(t),$$
$$\frac{\mathrm{d}}{\mathrm{d}t} P_n(t) = -I(t)\,(P_n(t) - P_{n-1}(t)), \qquad n \geqslant 1, \tag{62}$$

with $I(t) = \mathrm{d}\mu(t)/\mathrm{d}t$. If we substitute this into (55) we obtain

$$w_n(t) = P_{n-1}(t)\,I(t). \tag{63}$$

This expresses that the probability for the detection of the $n$th photon in $[t, t + \mathrm{d}t]$ is proportional to $I(t)$ and independent of the previous $n - 1$ detections. In other words, the photon detections are uncorrelated.

An interesting quantity will appear to be the average elapsed time before the $n$th photon is detected. If we define the waiting time as

$$T_n = \int_0^{\infty} t\, w_n(t)\,\mathrm{d}t \tag{64}$$

then we find with equation (55)

$$T_n = \sum_{k=0}^{n-1} \int_0^{\infty} P_k(t)\,\mathrm{d}t, \tag{65}$$

or in the Laplace domain

$$T_n = -\lim_{s \to 0} \frac{\mathrm{d}}{\mathrm{d}s} \tilde{w}_n(s) = \sum_{k=0}^{n-1} \tilde{P}_k(0). \tag{66}$$

The variance of the waiting time for the $n$th photon is

$$\Delta T_n^2 = \int_0^{\infty} (t - T_n)^2 w_n(t)\,\mathrm{d}t, \tag{67}$$

and with (55) this can be expressed in $\tilde{w}_n(s)$ or $\tilde{P}_k(s)$ as

$$\Delta T_n^2 = \lim_{s\to 0}\frac{d^2}{ds^2}\tilde{w}_n(s) - T_n^2 = -2\sum_{k=0}^{n-1}\lim_{s\to 0}\frac{d}{ds}\tilde{P}_k(s) - T_n^2. \quad (68)$$

## 8. Fluorescent photons

In this section we evaluate the $\tilde{w}_n(s)$ and the waiting times for photon detection from fluorescence radiation. If we take the Laplace transform of equation (55) and substitute the result (36), we obtain

$$\tilde{w}_n(s) = \frac{I}{s(1+\tilde{f}(s))}\left(\frac{\tilde{f}(s)}{1+\tilde{f}(s)}\right)^{n-1}, \quad n \geqslant 1 \quad (69)$$

which can be written as

$$\tilde{w}_n(s) = s(1+\tilde{f}(s))\tilde{P}_n(s), \quad n \geqslant 1. \quad (70)$$

For $n=1$ we have the alternative form

$$\tilde{w}_1(s) = 1 - s\tilde{P}_0(s) \quad (71)$$

as follows from (54).

From (36) and (39) we find

$$\tilde{P}_0(0) = \frac{1}{I} + \frac{\bar{Q}}{2I},$$

$$\tilde{P}_n(0) = \frac{1}{I}, \qquad n \geqslant 1 \quad (72)$$

and with (66) this gives

$$T_n = \frac{n}{I} + \frac{\bar{Q}}{2I}. \quad (73)$$

For $n=1$ we have the inverse relation

$$\bar{Q} = 2(IT_1 - 1). \quad (74)$$

Hence $\bar{Q}$ can be determined from a measurement of the average waiting time $T_1$ for the appearance of the first photon. This should be much easier than the measurement of $\mu(t)$ and $\sigma^2(t)$ for $t\to\infty$, as it is commonly done. Notice that the constraint $\bar{Q} \geqslant -1$ sets the lower bound to the waiting time

$$T_1 \geqslant 1/2I \quad (75)$$

but for classical fields ($\bar{Q} \geqslant 0$) we have

$$T_1 \geqslant 1/I. \quad (76)$$

Hence a waiting time which is smaller than $1/I$ reflects sub-Poissonian statistics, and thereby the non-classical behaviour of fluorescent photons.

The measurement of $T_1$ determines $\bar{Q}$, and thereby the asymptotic behaviour for $t\to\infty$ of $S_k(t)$, according to equation (52). Combination of (51) and (74) gives

$$\zeta_1 = T_1 - \frac{1}{I}, \quad (77)$$

and this parameter governs the asymptotic behaviour of the normalized quantity

$\beta_{2k}(t)$. The behaviour of the $\beta_n(t)$s with $n$ odd involves the second independent parameter $\zeta_2$, which is not determined by $T_1$. It can however also be found from the waiting-time distribution. To this end, we consider the variance $\Delta T_n^2$. From (68) and (69) we find immediately

$$\Delta T_n^2 = -\zeta_2 + \zeta_1 T_n + (n/I) T_1 \tag{78}$$

for the variance of the waiting time for the $n$th photon. For $n = 1$ this equation reads

$$\zeta_2 = T_1^2 - \Delta T_1^2. \tag{79}$$

This shows that $T_1$ and $\Delta T_1^2$ determine $\zeta_1$ and $\zeta_2$, and thereby the asymptotic behaviour for $t \to \infty$ of $\beta_n(t)$ for all $n$. It will be obvious that we can generalize this procedure to obtain the $\zeta_n$ for $n = 1, 2, 3, \ldots$ from the waiting-time distribution $w_n(t)$. Hence the complete photon statistics for $t \to \infty$ can be found from the measurement of the time delay of the first photon, after a random initialization of the counting process.

## 9. Conclusions

In this paper we elaborated the theory of photon statistics of fluorescence radiation, with particular emphasis on the deviation from Poisson statistics. This was accomplished by the introduction of normalized quantities $\beta_n(t)$, which vanish identically for a Poisson distribution ($n \geqslant 2$). We evaluated $\tilde{\beta}_n(s)$ explicitly and we studied its behaviour in the limit of long counting times. It appeared that the first term in the asymptotic expansion of $\beta_2(t), \beta_3(t), \ldots$ around $t = \infty$, involves only the two independent atomic parameters $\zeta_1$ and $\zeta_2$. Here $\zeta_1$ is proportional to the familiar $\bar{Q}$-factor. Furthermore it is shown that the deviation of the factorial moments from their Poisson value $S_k(t) = \mu(t)^k$, is determined by $\bar{Q}$ for all $k$ in the limit $t \to \infty$.

Subsequently we considered the distribution $w_n(t)$ of the waiting time for the appearance of the $n$th photon. We evaluated the average waiting time $T_n$ and the variance $\Delta T_n^2$. It was pointed out that $T_1$ and $\Delta T_1^2$ determine the parameters $\zeta_1$ and $\zeta_2$. Thus the asymptotic limit $t \to \infty$ of the photon statistics, which corresponds to the detection of an infinite number of photons, can be found from the measurement of the distribution of the detection times of the first photon.

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