CORRELATED STATISTICS OF PHOTONS IN THE COMPONENTS OF THE FLUORESCENCE TRIPLET

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We derive the statistics of the number of photons emitted in the three lines of the fluorescence spectrum of a two-level atom from an explicit expression for the generating function. Photons from opposite sidebands alternate when the excitation is purely monochromatic and in the absence of collisions. For a driving field with a finite bandwidth or when collisions are present, this is no longer true. Photons in the Rayleigh line are emitted independently of emissions in other lines and have a Poisson probability distribution. We derive explicit expressions for the deviation from poissonian statistics for emissions of photons in the sidebands. In the limit of large counting times, the factorial moments of the distribution tend to their poissonian value. Finally we show that collisions or a finite bandwidth can narrow the probability distribution of photon emissions.

Photon statistics of a radiation field is determined by the hierarchy of joint probability densities $I_n(t_1, t_2, ..t_n)$ for the detection of a photon at time t_1, a photon at time t_2, ..., and a photon at time t_n. In the case of fluorescence radiation from a two-level atom in the steady state, these probability densities take the form [1–3]

$$I_n(t_1, t_2, ...t_n) = f(t_n - t_{n-1})...f(t_3 - t_2) f(t_2 - t_1) S \tag{1}$$

for $t_n > ... > t_2 > t_1$. Here S is the integrated strength of the fluorescence spectrum and $f(\tau)$ is the conditional probability for detecting a photon at time $t = \tau$, when another photon was detected at time $t = 0$. In the limit $\tau \downarrow 0$, $f(\tau)$ approaches zero, indicating that the atom needs a finite recovery time before a subsequent photon emission can occur. This phenomenon of antibunching has been observed experimentallly [4]. In the limit $\tau \to \infty$, the memory of the initial emission is lost and $f(\tau)$ approaches the steady-state strength S.

These results apply to the situation that the photons are detected without spectral resolution. On the other hand, it is known that the fluorescence spectrum of a two-level atom contains three separated lines in the limit of large Raby frequency Ω or large detuning Δ from resonance [5]. The central line, positioned at the frequency ω_L of the driving field is termed the Rayleigh line (R) and the two sidebands at $\omega_L - \Omega'$ and $\omega_L + \Omega'$ are the fluorescence line (F) and the three photon line (T), with

$$\Omega' = \Delta(1 + \Omega^2/\Delta^2)^{1/2} . \tag{2}$$

It has been demonstrated experimentally [6,7] that photons from the two sidebands tend to be emitted in a well-defined order (T before F). The intensity correlation functions for each pair of lines in the triplet have been studied by several authors [8–10]. The frequency resolution, needed for the spectral separation of the lines, necessarily leads to a restricted time resolution so that rapid oscillations with frequency Ω' are washed out. These oscillations correspond to off-diagonal terms between the eigenstates of the dressed atom [9]. When the exciting radiation has a finite bandwidth λ, resulting from phase fluctuations, and when the atom suffers line-broadening collisions with foreign gas particles, the steady-state populations n_1 and n_2 of the eigenstates of the dressed atom are parametrised as [11]

$$n_1 = p/(p+q), \quad n_2 = q/(p+q). \tag{3}$$

Here $p = Ag_F^2 + 2\lambda g_R^2 + k(\Omega, \Delta)$ and $q = Ag_T^2 + 2\lambda g_R^2 + k(\Omega, \Delta)$, with $k(\Omega, \Delta)$ the rate of optical collisions between the dressed states, A the Einstein coefficient for spontaneous emission and

$$g_F = (\Omega' + \Delta)/2\Omega', \quad g_T = (\Omega' - \Delta)/2\Omega', \quad g_R = \Omega/2\Omega'. \tag{4}$$

The strengths of the line lines are

$$S_R = Ag_R^2, \quad S_F = Ag_F^2 n_2, \quad S_T = Ag_T^2 n_1. \tag{5}$$

The intensity correlations between the lines α and β in the spectrum are found to be [8,10]

$$I_2(\alpha t_1, \beta t_2) = S_\alpha S_\beta \qquad \text{for } \alpha = \mathrm{R}, \text{ or } \beta = \mathrm{R} \tag{6}$$

indicating that an emission of a photon in the Rayleigh line is uncorrelated to previous or later emissions. The other intensity correlation functions are

$$I_2(\mathrm{F}t_1, \mathrm{F}t_2) = S_F f(\mathrm{FF}, t_2 - t_1), \qquad I_2(\mathrm{F}t_1, \mathrm{T}t_2) = S_F f(\mathrm{FT}, t_2 - t_1)$$

$$I_2(\mathrm{T}t_1, \mathrm{T}t_2) = S_T f(\mathrm{TT}, t_2 - t_1), \qquad I_2(\mathrm{T}t_1, \mathrm{F}t_2) = S_T f(\mathrm{TF}, t_2 - t_1) \tag{7}$$

for $t_2 > t_1$. The functions $f(\alpha\beta, \tau)$ have the physical significance of the conditional probability of a photon emission at time $t = \tau$ in the line β, when a photon in the line α was detected at time $t = 0$. Their explicit expressions are

$$f(\mathrm{FF}, \tau) = S_F[1 - \exp(-(p+q)\tau)], \qquad f(\mathrm{TT}, \tau) = S_T[1 - \exp(-(p+q)\tau)],$$

$$f(\mathrm{FT}, \tau) = S_T[1 + (q/p)\exp(-(p+q)\tau)], \qquad f(\mathrm{TF}, \tau) = S_F[1 + (p/q)\exp(-(p+q)\tau)]. \tag{8}$$

The properties $f(\mathrm{FF}, 0) = f(\mathrm{TT}, 0) = 0$ indicate antibunching for F photons and for T photons, whereas successively emitted photons from opposite sidebands tend to bunch. This is understandable in a dressed atom picture. In eq. (5) we see that the probability of an F emission is proportional to the population of the dressed state $|2\rangle$, but an emission in the F line corresponds to a $|2\rangle \to |1\rangle$ transition [9] and the other way around for a T emission. This explains the bunching and antibunching properties of F and T photons. Emission of an R photon leaves the state of the atom unchanged, which explains the factorisation (6).

In this paper we intend to study the correlated statistics of the photons emitted in the three lines. This statistics is fully contained in the joint probability densities $I_n(\alpha_1, t_1; \alpha_2, t_2; ...\alpha_n, t_n)$ for detection of a photon in line α_1 at time t_1, ..., and a photon in line α_n at time t_n [10]. These probability densities are found as direct generalisations of eqs. (6) and (7). Each photon $\alpha_i = \mathrm{R}$ contributes a simple factor S_R to I_n, so it is sufficient to restrict ourselves to the case that $\alpha_i = \mathrm{F}$ or T for all i. Then the probability density I_n is given by

$$I_n(\alpha_1, t_1; ...; \alpha_n, t_n) = f(\alpha_{n-1}\alpha_n, t_n - t_{n-1}) ... f(\alpha_1\alpha_2, t_2 - t_1) S_{\alpha_1} \tag{9}$$

for $\alpha_i = \mathrm{F}$ or T and $t_n > t_{n-1} > ... > t_1$. This determines the probability density I_n for arbitrary time ordering, since by definition I_n must be invariant for an exchange of (α_i, t_i) and (α_j, t_j).

We denote as $P(K, L, M; t)$ the probability of detecting precisely K photons in the R line, L photons in the F line and M photons in the T line during the counting interval $[0, t]$. It is convenient to introduce the generating function [12], which in the present case of three types of photons is a function of three variables. We define

$$G(x, y, z; t) \equiv \langle x^K y^L z^M \rangle(t) = \sum_{KLM} P(K, L, M; t)\, x^K y^L z^M. \tag{10}$$

Expanding this function in a Taylor series around $x = y = z = 1$ gives

$$G(x,y,z;t) = \sum_{klm} \frac{(x-1)^k}{k!} \frac{(y-1)^l}{l!} \frac{(z-1)^m}{m!} s(k,l,m;t) , \tag{11}$$

with

$$s(k,l,m;t) = \left\langle \frac{K!}{(K-k)!} \frac{L!}{(L-l)!} \frac{M}{(M-m)!} \right\rangle (t) \tag{12}$$

the factorial moments. Knowledge of the generating function or of the complete set of all factorial moments is equivalent to the knowledge of the photon number probabilities P. On the other hand, the factorial moments $s(k, l, m; t)$ can be expressed as multiple integrals over a joint probability density I_n with $n = k + l + m$ [12]

$$s(k,l,m;t) = \int_0^t \mathrm{d}t_1 \ldots \mathrm{d}t_n \, I_n(\alpha_1, t_1; \alpha_2, t_2; \ldots; \alpha_n, t_n) , \tag{13}$$

where the set of labels α_i contains k times R, l times F and m times T. The R photons contribute a factor $(S_\mathrm{R} t)^k$ and we have the explicit expression (9) for the remaining probability density of F and T photons. If we substitute (13) into (11), transform the multiple integral in a time-ordered integral and perform the summation over k, we obtain

$$G(x,y,z;t) = \exp[S_\mathrm{R} t(x-1)]$$
$$\times \left[1 + \sum_{n=1}^{\infty} \sum_{\{\alpha_i\}} (y-1)^l (z-1)^m \int_0^t \mathrm{d}t_n \int_0^{t_n} \mathrm{d}t_{n-1} \ldots \int_0^{t_2} \mathrm{d}t_1 \, I_n(\alpha_1, t_1; \alpha_2, t_2; \ldots; \alpha_n, t_n) \right] . \tag{14}$$

The summation over $\{\alpha_i\}$ runs over all ordered series $\alpha_1, \alpha_2, \ldots \alpha_n$ with α_i = F or T, and l is the number of F's and m the number of T's in the series. After substituting (9) into (14), we directly obtain an expression for the Laplace transform of G,

$$\tilde{G}(x,y,z;v) = \int_0^\infty \mathrm{e}^{-vt} G(x,y,z;t) \, \mathrm{d}t \tag{15}$$

with the result (for $x = 1$)

$$\tilde{G}(1,y,z;v) = \frac{1}{v} + \frac{1}{v^2} \sum_{n=1}^{\infty} \sum_{\{\alpha_i\}} (y-1)^l (z-1)^m \tilde{f}(\alpha_{n-1}\alpha_n, v) \ldots \tilde{f}(\alpha_1 \alpha_2, v) S_{\alpha_1} , \tag{16}$$

where $\tilde{f}(v)$ is the Laplace transform of $f(t)$. The x dependence of $\tilde{G}$ is given by the property

$$\tilde{G}(x,y,z;v) = \tilde{G}(1,y,z;v - S_\mathrm{R}(x-1)) . \tag{17}$$

The summation in (16) can be performed and the resulting expression reads

$$\tilde{G}(1,y,z;v) = \frac{p + q + v + S_\mathrm{F}(y-1) + S_\mathrm{T}(z-1)}{v(p+q+v) - (p+q)[S_\mathrm{F}(y-1) + S_\mathrm{T}(z-1)] - (p+q)^2 S_\mathrm{F} S_\mathrm{T}(y-1)(z-1)/pq} \tag{18}$$

and can be simplified by noting that $(p + q)^2 S_\mathrm{F} S_\mathrm{T}/pq = S_\mathrm{R}^2$.

Eq. (18) is the main result of this paper. Together with (17) it contains the full information on the statistics of the photons in the three lines, including all correlations between them. The derivatives with respect to x, y and z at the arguments $x = y = z = 0$ determine the probabilities $\tilde{P}(K, L, M; v)$ whereas the derivatives at the arguments $x = y = z = 1$ determine the factorial moments $\tilde{s}(k, l, m; v)$. In the remaining part of this paper we will give some explicit results, which are all extracted from the general result (17), (18).

If we set our spectrometer on a line of the fluorescence triplet, only one kind of photons is detected. In view of (10), the generating function for photon detections in one line can be found from the general expression by setting two of the variables x, y, z equal to one. In this way, the statistics of Rayleigh photons is described by

$G(x, 1, 1; t) = \exp[S_R t(x-1)]$ which corresponds to a Poisson distribution. From (14) we see that this x dependence of $G(x, y, z; t)$ factorizes from the y, z dependence, so R photons are emitted randomly and independent of emissions in other lines. This can be understood from the fact that an emission of an R photon does not affect the state of the atom. The average number of emitted photons in $[0, t]$ equals $S_R t$ and the factorial moments of this distribution are simply $S(k, 0, 0; t) = (S_R t)^k$.

Since we know that photon emissions in a sideband are correlated, we expect a deviation from poissonian statistics. If we write the factorial moments for emissions in the F line as

$$s(0, l, 0; t) = \langle L!/(L-l)!\rangle = (S_F t)^l \{1 + C^{(l)}((p+q)t)\} \tag{19}$$

then we find from (18) with $x = z = 1$

$$C^{(l)}(\tau) = \frac{-l(l-1)}{\tau} + \sum_{k=2}^{l} \frac{(l+k-2)!}{(l-k)!(k-2)!(-\tau)^k}\left(\frac{l(l-1)}{k(k-1)} - (-1)^{l+k}\mathrm{e}^{-\tau}\right), \quad l = 2, 3, \ldots \tag{20}$$

and $C^{(l)}(\tau) = 0$ for $l = 0, 1$. The same result holds for T photons if we replace S_F by S_T. The function $C^{(l)}(\tau)$ is the same in both cases and is approximately $-l(l-1)/\tau$ for τ large. Hence for long counting times the factorial moments will approach their poissonian limit $(S_\alpha t)^l$ with α = F or T. The behaviour of $C^{(l)}(\tau)$ for small times is found by an expansion in τ, which is obtained directly after expanding $s(0, l, 0; v)$ in v^{-1}. The result is

$$C^{(l)}(\tau) = -1 + l(l-1)\sum_{k=0}^{\infty}(-1)^k \frac{(k+l-2)!}{k!(k+2l-1)!}\tau^{l+k-1} \tag{21}$$

for $l \geqslant 2$. Hence $C^{(l)}(0) = -1$ and the approach to zero for increasing τ is very smooth.

As an application of these results we evaluate Mandel's Q-factor [2], which is a measure for the deviation from poissonian statistics. With $\mu(t) = S_\alpha t$ the mean and $\sigma^2(t)$ the variance of the number of photons emitted in the line α = F or T, we find

$$Q_\alpha(t) \equiv [\sigma^2(t) - \mu(t)]/\mu(t) = \mu(t)C^{(2)}((p+q)t)$$

$$= -2S_\alpha\{1 - [1 - \exp(-(p+q)t)]/(p+q)t\}/(p+q), \quad \alpha = \mathrm{F}, \mathrm{T}. \tag{22}$$

Hence Q_α is negative for α = F or T and $t > 0$, and the probability distribution is narrower than the corresponding Poisson distribution with the same mean. This reflects the net anti-bunching property of the photons from either one of the sidebands.

Another instructive example is the correlated statistics of F and T photons. If we consider the case of a free atom in a monochromatic driving field ($k(\Omega, \Delta) = 0$, $\lambda = 0$), we can simplify the generating function (18) by using the explicit expressions (5) for S_F and S_T (which are equal in this case). The result is

$$G(1, y, z; v) = [p + q + v + S_F(y + z - 2)]/[v(p + q + v) - pq(yz - 1)], \tag{23}$$

with $S_F = S_T = pq/(p+q)$. One notices that the numerator in (23) is linear in y and z, whereas the denominator depends only on the product yz. If we make the expansion (10) to find the probabilities $\widetilde{P}(K, L, M; v)$, we only obtain terms with $|L - M| = 0$ or 1. Hence the probability for L photons in the F line and M photons in the T line vanishes whenever L and M differ by more than one. Since this is true for an arbitrary time interval, we may conclude that F and T photons are emitted in an alternating fashion: after emission of an F photon, a subsequent emission of an F photon is only possible when it is preceded by emission of a T photon. This result is directly understood in a dressed-atom picture, as was pointed out before by Cohen–Tannoudji [13]. Emission of an F photon corresponds to a spontaneous emission from the state $|2\rangle$ to the state $|1\rangle$ of the dressed atom, whereas a T photon is emitted during a spontaneous transition from $|1\rangle$ to $|2\rangle$. For a free atom in a monochromatic field these sideband emissions are the only mechanism for transitions between the dressed states, since emission of a Rayleigh photon leaves the state of the atom unchanged. On the other hand, when collisions are present, transitions between the dressed states are possible without emission of a photon. In a non-monochromatic field, the dressed states are possible without emission of a photon. In a non-monochromatic field, the dressed states are effectively coupled by the field fluctuations. As is obvious from eq. (18), a non-zero rate of optical collisions k or a non-zero bandwidth λ introduces linear terms in y and z in the denominator. This causes the probabilities $P(K, L, M; t)$ to be non-zero, even if L and M differ by more than one, indicating that the purely alternating character of sideband photon emissions is destroyed.

When we set x, y and z equal in the generating function, we obtain a generating function of only one variable. This function signifies the average $\langle x^N \rangle$ with $N = K + L + M$, which generates the statistics of all photon emissions, irrespective of their frequency. In the present limit of separated lines, we have assumed Ω' to be large, and an average over rapid oscillations with this frequency is made implicitly by the detection method of resolving the lines. If we calculate the Q-factor from $G(x, x, x; t)$, we obtain in the limit of a large detection interval

$$\bar{Q} = Q(\infty) = -\frac{2A\Delta^2}{\Omega'^2} \frac{2(\lambda - \frac{1}{2}A)g_R^2 + k(\Omega, \Delta)}{[A + 4(\lambda - \frac{1}{2}A)g_R^2 + 2k(\Omega, \Delta)]^2} . \tag{24}$$

For a free atom in a monochromatic field ($\lambda = k = 0$), this $\bar{Q}$-factor is positive, indicating that the statistics of the total number of emitted photons is super-poissonian. However, due to collisions or a finite bandwidth, $\bar{Q}$ can become negative, which reflects sub-poissonian statistics. Hence collisions or field fluctuations can make the probability distribution narrower. The same results can also be obtained from a previous paper [14] in the appropriate limit of separated lines.

References

[1] G.S. Agarwal, Phys. Rev. A15 (1977) 814.
[2] L. Mandel, Optics Lett. 4 (1979) 205.
[3] A.T. Georges, Optics Comm. 38 (1981) 274.
[4] H.J. Kimble, M. Dagenais and L. Mandel, Phys. Rev. A18 (1978) 201.
[5] B.R. Mollow, Phys. Rev. 188 (1969) 1969.
[6] J. Dalibard, thesis (Laboratoire de Physique de l'Ecole Normale Supérieure, Paris, 1981).
[7] A. Aspect, G. Roger, S. Reynaud, J. Dalibard and C. Cohen-Tannoudji, Phys. Rev. Lett. 45 (1980) 617.
[8] P.A. Apanasevich and S.Ja. Kilin, J. Phys. B12 (1979) L83.
[9] C. Cohen-Tannoudji and S. Reynaud, Phil. Trasn. Roy. Soc. London A293 (1979) 223.
[10] H.F. Arnoldus and G. Nienhuis, J. Phys. B, submitted.
[11] H.F. Arnoldus and G. Nienhuis, J. Phys. B16 (1983) 2325.
[12] N.G. van Kampen, Stochastic processes in physics and chemistry (North-Holland, Amsterdam, 1981).
[13] C. Cohen-Tannoudji, in: Laser spectroscopy VI, eds. H.P. Weber and W. Lüthy (Springer, Berlin, 1983) p. 2.
[14] H.F. Arnoldus and G. Nienhuis, Optica Acta, in press.