PHOTON STATISTICS OF ATOMIC FLUORESCENCE, INDUCED BY A GAUSSIAN LASER FIELD

H.F. ARNOLDUS and G. NIENHUIS

Fysisch Laboratorium, Rijksuniversiteit Utrecht, Postbus 80 000, 3508 TA Utrecht, The Netherlands

Received 25 January 1985; revised manuscript received 11 March 1985

We investigate the photon correlations and statistics of the fluorescence radiation, emitted by a two-level atom in a strong phase-fluctuating laser field and a perturber bath. The laser lineshape is assumed to be gaussian, in contrast with the generally applied model of a lorentzian laser profile. It is shown that the stochastic process, which is responsible for the line broadening, can be dealt with exactly, just as in the lorentzian limit. The photon correlation functions do not factorize any more, which affects the photon statistics quite thoroughly. For long counting times, the variance in the number of detected photons increases with t^2, whereas in the lorentzian limit it was proportional to t. This implies that the variance will always exceed the average for t large, and hence the statistics is super-poissonian.

It has been recognized for quite a long time that the conditional probability for a photon detection from atomic fluorescence radiation at a time $t = \tau$ after a detection of a photon at $t = 0$, reveals the time evolution of the atom [1,2]. Hence the measurement of temporal photon correlations provides an important tool to investigate in detail the interaction of an atom with a strong laser field. A related issue which has attracted much attention in the last few years, pertains to the question whether fluorescence radiation can be represented by a stochastic classical electromagnetic field, or if a quantum description is inevitable. It was pointed out by Mandel [3] that this question is amenable to experimental investigation by means of a simple photon counting measurement. He showed that the variance $\sigma^2(t)$ in the number of detected photons in a time interval $[0, t]$ should always exceed (or equal) the average $\mu(t)$ for a classical stochastic field, so $\sigma^2(t) \geqslant \mu(t)$. A violation of this inequality would confirm the quantum nature of light, and this has been found recently [4]. The occurrence of sub-poissonian statistics, defined as a distribution with $\sigma^2(t) < \mu(t)$, is closely related to the photon correlations. For small counting times, we always have sub-poissonian statistics as a consequence of the anti-bunching of fluorescent photons. For long times, sub-poissonian statistics arises only if an effective, average anti-bunching survives for long delay times between two subsequently detected photons.

In contrast with the universal anti-bunching and the implied sub-poissonian statistics for small times, the long-time properties of the statistics will depend on the detailed mechanisms involved and the values of the various parameters. In a previous paper [5] we derived the condition for sub-poissonian statistics in the long-time limit, as a function of the detuning from resonance Δ, the collisional width γ and shift β, and the laser bandwidth λ'. The criterion turned out to be independent of the Rabi frequency Ω. It appeared that the statistics is sub-poissonian in general, except for small bandwidths λ', together with large detunings. In this paper we will show that not only the precise values of the parameters determine the long-time behaviour, but that the statistics is extremely sensitive to the underlying dynamics. In [5] we took the laser line to be a lorentzian with HWHM = λ', where the broadening was accomplished by stochastic process, which gives rise to a gaussian laser profile. It will turn out that this slight modification affects the results in a dramatic way.

The electric component of the laser field at the position of the atom is taken to be

$$E(t) = E_0 \, \mathrm{Re}\, \boldsymbol{\varepsilon}_{\mathrm{L}} \exp[-\mathrm{i}(\omega_{\mathrm{L}} t + \phi(t))], \tag{1}$$

with $\phi(t)$ an arbitrary stochastic process. Then the laser profile is given by

$$I(\omega) = \frac{1}{\pi} \mathrm{Re} \int_0^\infty \exp[\mathrm{i}(\omega - \omega_\mathrm{L})\tau] \times \langle \exp[-\mathrm{i} \textstyle\int_0^\tau \dot{\phi}(s)\,\mathrm{d}s] \rangle \,\mathrm{d}\tau, \tag{2}$$

where $\langle ... \rangle$ denotes a stochastic average. The equation of motion for the atomic density operator in the rotating frame attains the form [5]

$$\mathrm{i}\,\mathrm{d}\sigma/\mathrm{d}t = (L_\mathrm{d} + \dot{\phi}(t) L_\mathrm{g} - \mathrm{i}\Gamma - \mathrm{i}\Phi)\sigma,$$

$$\sigma^\dagger = \sigma, \quad \mathrm{Tr}\,\sigma = 1, \tag{3}$$

and the Liouville operators L_d, L_g, Γ and Φ can be expressed in the Hilbert space operators $P_\mathrm{g} = |\mathrm{g}\rangle\langle\mathrm{g}|$, $\mathrm{P_e} = |\mathrm{e}\rangle\langle\mathrm{e}|$, $d = |\mathrm{e}\rangle\langle\mathrm{g}|$. The dressed-atom dynamics is represented by

$$L_\mathrm{d}\sigma = -\tfrac{1}{2}[\Delta(P_\mathrm{e} - P_\mathrm{g}) + \Omega(d + d^\dagger), \sigma], \tag{4}$$

with $\Delta = \omega_\mathrm{L} - \omega_0$, $\hbar\Omega = E_0 \boldsymbol{\mu}_\mathrm{eg} \cdot \boldsymbol{\varepsilon}_\mathrm{L}$ as usual, and the phase fluctuations, spontaneous decay and collisions with perturbers are included as

$$L_\mathrm{g}\sigma = [P_\mathrm{g}, \sigma], \tag{5}$$

$$\Gamma\sigma = \tfrac{1}{2}A(P_\mathrm{e}\sigma + \sigma P_\mathrm{e} - 2d^\dagger \sigma d), \tag{6}$$

$$\Phi\sigma = L_\mathrm{g}(\gamma L_\mathrm{g} - \mathrm{i}\beta)\,\sigma, \tag{7}$$

with A the Einstein coefficient. The solution of eq. (3) can be written as

$$\sigma(t_2) = \theta \exp\left(-\mathrm{i} \int_{t_1}^{t_2} (L_\mathrm{d} + \dot{\phi}(s) L_\mathrm{g} - \mathrm{i}\Gamma - \mathrm{i}\Phi)\,\mathrm{d}s\right) \sigma(t_1), \tag{8}$$

and the time-ordered exponential will be abbreviated as $Y(t_2, t_1)$.

The photon correlation functions, which are proportional to the intensity correlations of the fluorescence radiation, can be expressed in the stochastic evolution operator $Y(t_{i+1}, t_i)$ as [5]

$$I_k(t_1, ..., t_k) = (\alpha A)^k \,\mathrm{Tr}\,\mathcal{R}\, Y(t_k, t_{k-1}) \times \mathcal{R} ... \mathcal{R}\, Y(t_2, t_1) \mathcal{R}\, \sigma(t_1), \qquad t_k \geqslant ... \geqslant t_1 \tag{9}$$

where α is the probability that an emitted photon is detected. The photon emission operator $\mathcal{R}$ is defined by

$$\mathcal{R}\sigma = d^\dagger \sigma d = P_\mathrm{g}\langle \mathrm{e}|\sigma|\mathrm{e}\rangle, \tag{10}$$

and the property $\mathcal{R}^2 = 0$ reflects anti-bunching.

Eq. (9) holds for an arbitrary stochastic phase $\phi(t)$. Let us now suppose that $\dot{\phi}(t)$ is a gaussian process. Then all moments can be expressed in the second moment [6]

$$\langle \dot{\phi}(t_1) ... \dot{\phi}(t_n) \rangle = (2^m m!)^{-1} \sum_\mathrm{P} \prod_{(\alpha,\beta)} \langle \dot{\phi}(t_\alpha)\dot{\phi}(t_\beta) \rangle \tag{11}$$

for $n = 2m = 2, 4, ...$ and the odd momenta vanish. Commonly [7–9], $\dot{\phi}(t)$ is taken to be the Ornstein–Uhlenbeck process, with

$$\langle \dot{\phi}(t_\alpha)\dot{\phi}(t_\beta) \rangle = \lambda^2 \exp[-|t_\alpha - t_\beta|\lambda^2/\lambda'],$$

$$\lambda > 0, \quad \lambda' > 0, \tag{12}$$

where λ and λ' are two independent parameters. If we expand the exponential in brackets in (2) and apply (11), (12), we find for the laser profile

$$I(\omega) = \frac{1}{\pi} \mathrm{Re} \frac{1}{\lambda' - \mathrm{i}(\omega - \omega_\mathrm{L})} \times M\left(1, \frac{\lambda' - \mathrm{i}(\omega - \omega_\mathrm{L})}{\lambda^2/\lambda'} + 1, (\lambda'/\lambda)^2\right), \tag{13}$$

with $M(a, b, z)$ the regular confluent hypergeometric function, which is related to the incomplete gamma function by $M(1, a+1, z) = a\mathrm{e}^{-z} z^{-a} \gamma(a, z)$ [10]. If we would apply (12) to obtain the stochastic average of $I_k(t_1, ..., t_k)$, this would give rise to very awkward and untransparent expressions. Therefore we consider two limits of special importance, defined as

$$\lambda' \to \infty, \lambda \text{ fixed} \quad \text{gives } \langle \dot{\phi}(t_\alpha)\dot{\phi}(t_\beta) \rangle = \lambda^2, \tag{14}$$

$$\lambda \to \infty, \lambda' \text{ fixed} \quad \text{gives } \langle \dot{\phi}(t_\alpha)\dot{\phi}(t_\beta) \rangle = 2\lambda'\delta(t_\alpha - t_\beta), \tag{15}$$

and the corresponding laser profiles reduce to

$$I_\mathrm{G}(\omega) = (1/\lambda\sqrt{2\pi}) \exp[-(\omega - \omega_\mathrm{L})^2/2\lambda^2],$$

$$I_\mathrm{L}(\omega) = (1/\pi)\mathrm{Re}[1/(\lambda' - \mathrm{i}(\omega - \omega_\mathrm{L}))], \tag{16}$$

a gaussian with HWHM = $\lambda\sqrt{2 \log 2}$ and a lorentzian with HWHM = λ', respectively. We notice that the confluent hypergeometric function in eq. (13) accounts for the deviation from the lorentzian limit, which is obscured in the common notation with the incomplete gamma function. The Lorentz limit (15) is the phase diffusion process, which is the favoured choice for $\dot{\phi}(t)$ [17,18], since its stochastics can be dealt with exactly [11] due to the δ-correlations and the resulting factorization of averages. It was shown by Kuś [12] however, that also the other extreme case, where the correlation time in (12) becomes very long, can be treated exactly. The photon statistics in the limit (15) are well established [5,13], and here we consider the opposite case (14). It can be proved that the choices (14) and (15) are the only gaussian $\dot{\phi}(t)$ processes, which yield a gaussian and a lorentzian laser profile, so these limits are not merely artificial simplifications.

The moments in the gaussian limit become $\langle \dot{\phi}(t_1) \dots \dot{\phi}(t_{2m}) \rangle = (2m!)\lambda^{2m}/(m!2^m)$ and the odd moments are zero. This can be written as

$$\langle \dot{\phi}(t_1) \dots \dot{\phi}(t_n) \rangle = \int_{-\infty}^{\infty} x^n p(x)\,\mathrm{d}x$$

with

$$p(x) = (1/\lambda\sqrt{2\pi}) \exp(-x^2/2\lambda^2) \qquad (17)$$

for all n. Notice that the laser profile is related to $p(x)$ as $I_G(\omega) = p(\omega - \omega_L)$. If we now expand the time-ordered exponential $Y(t_2, t_1)$ and take the stochastic average with (17), we obtain

$$\langle \sigma(t) \rangle = \int_{-\infty}^{\infty} \mathrm{d}x\, p(x) \times \exp[-\mathrm{i}(L_\mathrm{d} + xL_\mathrm{g} - \mathrm{i}\Gamma - \mathrm{i}\Phi)(t - t_0)]\, \sigma(t_0), \qquad (18)$$

where the initial atomic state $\sigma(t_0)$ is assumed to be non-stochastic. The stationary state of the atom then becomes

$$\overline{\sigma} = \lim_{t\to\infty} \langle \sigma(t) \rangle = \int_{-\infty}^{\infty} \overline{\sigma}(x)\, p(x)\,\mathrm{d}x, \qquad (19)$$

with $\overline{\sigma}(x)$ the unique solution of

$$(L_\mathrm{d} + xL_\mathrm{g} - \mathrm{i}\Gamma - \mathrm{i}\Phi)\overline{\sigma}(x) = 0, \quad \overline{\sigma}(x)^\dagger = \overline{\sigma}(x),$$
$$\mathrm{Tr}\, \overline{\sigma}(x) = 1, \qquad (20)$$

which can be found immediately. We notice that $\overline{\sigma}$ is independent of the arbitrary initial value $\sigma(t_0)$.

A similar method applies to the intensity correlations (9). Expanding all $Y(t_{i+1}, t_i)$ and taking the average yields

$$\langle I_k(t_1, \dots, t_k) \rangle = (\alpha A)^k \int_{-\infty}^{\infty} \mathrm{d}x\, p(x) \times \mathrm{Tr}\, \mathcal{R}\, Z(x, t_k - t_{k-1}) \mathcal{R} \dots \mathcal{R}\, Z(x, t_2 - t_1) \times \mathcal{R}\, \overline{\sigma}(x), \qquad (21)$$

which contains the evolution operator $Z(x, t) = \exp[-\mathrm{i}(L_\mathrm{d} + xL_\mathrm{g} - \mathrm{i}\Gamma - \mathrm{i}\Phi)t]$ and the atomic state $\overline{\sigma}(x)$. If we introduce the function

$$f(x, t) = \alpha A\, \mathrm{Tr}\, P_\mathrm{e}(Z(x, t)P_\mathrm{g}), \qquad (22)$$

the intensity correlations take the form [15]

$$\langle I_k(t_1, \dots, t_k) \rangle = \int_{-\infty}^{\infty} \mathrm{d}x\, p(x) \times f(x, t_k - t_{k-1}) \dots f(x, t_2 - t_1)\, \alpha A \overline{n}_\mathrm{e}(x), \qquad (23)$$

where the excited state population of $\overline{\sigma}(x)$ is

$$\overline{n}_\mathrm{e}(x) = \langle \mathrm{e}|\overline{\sigma}(x)|\mathrm{e}\rangle = \frac{\frac{1}{2}\Omega^2(\frac{1}{2}A + \gamma)}{\Omega^2(\frac{1}{2}A + \gamma) + A[(\Delta + x - \beta)^2 + (\frac{1}{2}A + \gamma)^2]}. \qquad (24)$$

In the lorentzian limit, the correlation functions (23) factorize, but this does not hold any more in the gaussian limit. The function $f(x, t)$ behaves as

$$f(x, t) = \alpha A(\tfrac{1}{2}\Omega t)^2 + O(t^3) \quad (t \to 0),$$
$$f(x, \infty) = \alpha A \overline{n}_\mathrm{e}(x), \qquad (25)$$

and especially $f(x, 0) = 0$, which is again the antibunching for small time-delays. The photon counting rate $\langle I_1(t) \rangle$ follows from (23) and becomes

$$I_1 = \langle I_1(t) \rangle = \int_{-\infty}^{\infty} I_1(x)\, p(x)\,\mathrm{d}x = \alpha A \overline{n}_\mathrm{e}$$

with

$$I_1(x) = \alpha A \bar{n}_e(x), \quad (26)$$

and after substitution of the explicit result (24), the integral can be evaluated in terms of error functions.

The statistics can be obtained from the correlation functions by standard methods [14]. If we denote by $P_n(t)$ the probability to detect n photons in $[0, t]$, we can introduce the factorial moments by

$$S_0(t) = 1,$$

$$S_k(t) = \sum_{n=k}^{\infty} n(n-1) \ldots (n-k+1) P_n(t), \quad k = 1, 2, \ldots, \quad (27)$$

which are determined by the intensity correlations

$$S_k(t) = k! \int_0^t \mathrm{d}t_k \times \int_0^{t_k} \mathrm{d}t_{k-1} \ldots \int_0^{t_2} \mathrm{d}t_1 \langle I_k(t_1, \ldots, t_k) \rangle, \quad k \geqslant 1. \quad (28)$$

With (23) we find $S_k(t)$ and with the inverse of (27)

$$P_n(t) = \frac{1}{n!} \sum_{k=0}^{\infty} \frac{(-1)^k}{k!} S_{n+k}(t), \quad n = 0, 1, 2, \ldots \quad (29)$$

we obtain the probabilities. The results can be cast in a simple form in the Laplace domain [16]. With

$$\tilde{f}(x,s) = \int_0^{\infty} \mathrm{e}^{-st} f(x,t)\,\mathrm{d}t = \frac{\alpha A}{s} \times \frac{\frac{1}{2}\Omega^2(\frac{1}{2}A + \gamma + s)}{\Omega^2(\frac{1}{2}A+\gamma+s) + (A+s)[(\Delta + x - \beta)^2 + (\frac{1}{2}A + \gamma + s)^2]} \quad (30)$$

we find explicitly for the statistics

$$\tilde{S}_0(s) = 1/s,$$

$$\tilde{S}_k(s) = \frac{k!}{s^2} \int_{-\infty}^{\infty} \mathrm{d}x\, p(x) I_1(x) \tilde{f}(x,s)^{k-1} \quad (k > 0) \quad (31)$$

$$\tilde{P}_0(s) = \frac{1}{s} - \frac{1}{s^2} \int_{-\infty}^{\infty} \mathrm{d}x\, p(x) \frac{I_1(x)}{1 + \tilde{f}(x,s)},$$

$$\tilde{P}_n(s) = \frac{1}{s^2} \int_{-\infty}^{\infty} \mathrm{d}x\, p(x) \frac{I_1(x) \tilde{f}(x,s)^{n-1}}{(1 + \tilde{f}(x,s))^{n+1}} \quad (n > 0) \quad (32)$$

in terms of $\tilde{f}(x, s)$. This determines the full statistics.

Let us now discuss some implications of these results, in order to establish the significance of the calculations. Statistical quantities, which are amenable to experiment, are for instance the average and the variance of the photon number distribution. They can be expressed in the factorial moments as $\mu(t) = S_1(t)$, $\sigma^2(t) = S_2(t) + S_1(t) - S_1(t)^2$. With (31) for $k = 1, 2$, transformed to the time domain,

$$S_1(t) = I_1(t),$$

$$S_2(t) = 2 \int_{-\infty}^{\infty} \mathrm{d}x\, p(x) I_1(x) \int_0^t (t - \tau) f(x, \tau)\,\mathrm{d}\tau, \quad (33)$$

we obtain for the difference

$$\sigma^2(t) - \mu(t) = -2I_1 \int_{-\infty}^{\infty} \mathrm{d}x\, p(x) I_1(x) \int_0^t (t-\tau)\kappa(x,\tau)\,\mathrm{d}\tau \quad (34)$$

in terms of the normalized correlation function $\kappa(x, \tau)$, defined by the relation $f(x, \tau) = I_1(1 - \kappa(x, \tau))$. It was already indicated in the beginning of this paper, that it is interesting to investigate whether $\sigma^2(t) - \mu(t)$ might become negative, a feature called sub-poissonian statistics. This situation appears as the normalized quantity

$$Q(t) = \frac{\sigma^2(t) - \mu(t)}{\mu(t)^2} = \frac{S_2(t) - S_1(t)^2}{S_1(t)^2} \geqslant -1 \quad (35)$$

is smaller than zero. For classical radiation fields, we have $Q(t) \geqslant 0$, but for arbitrary quantum fields we have $Q(t) \geqslant -1$. For small times we find

$$Q(t) = -1 + O(t^2), \quad (t \to 0) \quad (36)$$

and the extreme limit $Q(0) = -1$ is a direct consequence of the anti-bunching, just as in the lorentzian limit. If we now consider the limit $t \to \infty$, we obtain from (34)

$$Q(\infty) = \int_{-\infty}^{\infty} \mathrm{d}x\, p(x) \left(\frac{I_1(x) - I_1}{I_1} \right)^2 \geqslant 0 \qquad (37)$$

so for a gaussian laser field we can never have sub-poissonian statistics, whereas for a lorentzian laser line, we found sub-poissonian statistics for almost every set of parameters. This reveals the crucial dependence of the statistics on the mechanism of laser-line broadening. The sign of $Q(\infty)$ does not depend only on the bandwidth of the laser, but also on the stochastics of $\dot{\phi}(t)$. From (35) and (37) we also notice that $\sigma^2(t) = \bar{O}(t^2)$ for $t \to \infty$, but for a lorentzian laser field we had $\sigma^2(t) = \bar{O}(t)$, so the nature of the variance of the distribution has dramatically changed. It can be shown in general, that this is a consequence of the fact that the averaged intensity correlation $\langle I_2(t_1, t_2) \rangle$ does not factorize in $\langle I_2(t_1, t_2) \rangle = I_1^2$ for $t_2 \gg t_1$.

The definition of a normalized measure for $\sigma^2(t) - \mu(t)$, the Q-factor (35), is not the same as in our previous paper, where we took $Q_{pr}(t) = [\sigma^2(t) - \mu(t)]/\mu(t)$. For a lorentzian laser line we found that $Q_{pr}(t = \infty)$ remains finite, because $\sigma^2(t) = \bar{O}(t)$, but for the gaussian laser line we have $Q(t)$ finite. Because of the relation $Q(t) = Q_{pr}(t)/\mu(t)$, this Q-factor will approach zero in the lorentzian case in the limit $t \to \infty$. In the transient region $t < \infty$, the definition (35) is more natural. This $Q(t)$ is independent of α, the detector parameter, so it can be considered as a field quantity. Furthermore, the ultimate value $Q(0) = -1$ is a reflection of the anti-bunching for small time delays in the photon statistics, a feature which is hidden in the property $Q_{pr}(0) = -1$.

References

[1] H.J. Carmichael and D.F. Walls, J. Phys. B9 (1976) 1199.
[2] H.J. Kimble and L. Mandel, Phys. Rev. A13 (1976) 2123.
[3] L. Mandel, Optics Lett. 4 (1979) 205.
[4] R. Short and L. Mandel, Phys. Rev. Lett. 51 (1973) 384.
[5] H.F. Arnoldus and G. Nienhuis, Optica Acta 30 (1983) 1573.
[6] N.G. van Kampen, Stochastic processes in physics and chemistry (North-Holland, Amsterdam, 1981).
[7] A.T. Georges and P. Lambropoulos, Phys. Rev. A20 (1979) 991.
[8] P. Zoller, G. Alber and R. Salvador, Phys. Rev. A24 (1981) 398.
[9] R.I. Jackson and S. Swain, J. Phys. B15 (1982) 4375.
[10] M. Abramowitz and I. Stegun, Handbook of mathematical functions (Dover, New York, 1965).
[11] R.F. Fox, J. Math. Phys. 13 (1972) 1196.
[12] M. Kuś, Optica Acta 31 (1984) 261.
[13] S. Singh, Optics Comm. 44 (1983) 254.
[14] D. Lenstra, Phys. Rev. A26 (1982) 3369.
[15] A.T. Georges, Optics Comm. 38 (1981) 274.
[16] R.J. Cook, Phys. Rev. A23 (1981) 1243.
[17] H.J. Kimble and L. Mandel, Phys. Rev. A15 (1977) 689.
[18] L. Mandel, Optics Comm. 39 (1981) 163.