

Distribution of photocounts in nonlinear resonance fluorescence

D. F. Smirnov and A. S. Troshin

Leningrad Pedagogical Institute

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The distribution function determined by the field correlators of all orders, of the photocounts in nonlinear resonance atom fluorescence induced by quasimonochromatic radiation is investigated. The extreme cases of long and short observation times are considered. Results of a numerical calculation are presented. Some distribution features connected with the photon antibunching and the feasibility of experiments are discussed.

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1. Nonlinear resonance fluorescence (NRF) is presently the object of intense experimental and theoretical study, being a typical example of the nonlinear photoresponse of atomic-molecular systems. The dynamics of the system under resonant excitation of light manifests itself in the spectrum, in the correlation functions of the field, and in the statistics of NRF photocounts.

Observation of antibunching of photons in NRF, a phenomenon theoretically predicted in Refs. 3-5, was reported in Refs. 1 and 2. The antibunching is due to the time delay of the successive acts of photon scattering by one and the same atom. This delay reflects the dynamics of the population of the excited level of the atom in the field of an intense light wave. It manifests itself already in the fourth-order correlation function of the field of the scattered radiation: the term corresponding in this function to the single-atom contribution vanishes at equal times.

In this paper we investigate the distribution function $p(n, T)$ of the photocounts n during the observation time T for stationary scattering of intense monochromatic light by a two-level atom. This function is determined by the correlators of all orders of the field of the scattered radiation. We consider in greater detail the case of negative correlations (antibunching) and compare the results with the Poisson distribution, which corresponds to the coherent state of the radiation field.

2. It is convenient to calculate the characteristic function $Q(\lambda, T)$ of the distribution $p(n, T)$, which is defined by an expansion in the vicinity of the point $\lambda = 1$

$$Q(\lambda, T) = \sum_{n=0}^{\infty} (1-\lambda)^n p(n, T), \quad (1)$$

in the form of a series near the point $\lambda = 0$:

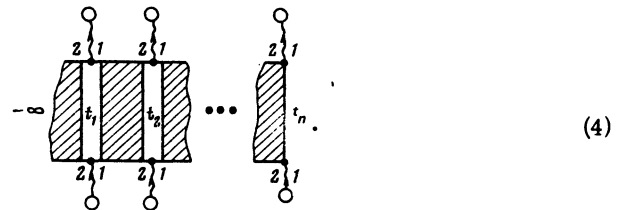
$$Q(\lambda, T) = \sum_{n=0}^{\infty} \frac{(-\lambda)^n}{n!} Q_n(T). \quad (2)$$

Here $Q_n(T)$ are the factorial moments of the distribution $p(n, T)$; they are directly connected with the correlators of the field of the scattered radiation incident on the photodetector⁶:

$$Q_n(T) = q^n \int dS_1 \dots \int dS_n \int_0^T dt_1 \dots \int_0^T dt_n \times \langle E_{\alpha}^{-}(\mathbf{r}_1, t_1) E_{\beta}^{-}(\mathbf{r}_2, t_2) \dots E_{\beta}^{+}(\mathbf{r}_n, t_n) E_{\alpha}^{+}(\mathbf{r}_1, t_1) \rangle. \quad (3)$$

Here q is the quantum yield of the photodetector and $E_{\alpha}^{\pm}(\mathbf{r}, t)$ are the NRF field operators in the Heisenberg representation. The integration is over the surface of the photodetector, which is assumed to be spherical with a center in the scattering region and with a radius much larger than the size of this region. Summation is carried out over the repeated indices of the Cartesian components.

Generalizing the procedure of Ref. 5, we can represent the correlator $\langle \dots \rangle$ in (3) in the form of the diagram



In the diagram (4), each shaded block represents a diagonal element $\rho_{22}^{(11)}(\tau)$ of the atom density matrix in a strong field—the upper indices denote the initial condition at $\tau = 0$ with 1 and 2 standing for the ground and excited levels, respectively). The circles in the upper part of the diagram are set in correspondence with the operators $E^{+}(\mathbf{r}_i, t_i)$; and in the lower part with the operators $E^{-}(\mathbf{r}_i, t_i)$. For the form of the photon propagators (wavy lines) see Ref. 5. After integrating in (3) over the surface of the photodetector, we obtain

$$Q_n(T) = (q\alpha\gamma)^n n! \int_0^T dt_n \int_0^{t_n} dt_{n-1} \dots \int_0^{t_2} dt_1 \bar{\rho}_{22} \times \rho_{22}^{(11)}(t_2 - t_1) \rho_{22}^{(11)}(t_3 - t_2) \dots \rho_{22}^{(11)}(t_n - t_{n-1}). \quad (5)$$

Here α is a coefficient that depends on the details of the experimental geometry (see Ref. 5), $\bar{\rho}_{22}$ is the stationary value of the population of the excited level. Expression (5) is a multiple convolution; this makes

it easy to obtain its Laplace transform:

$$Q_n(s) = n! \frac{\bar{p}}{s^2} \rho(s)^{n-1}, \quad n \geq 1; \quad Q_0(s) = 1/s, \quad (6)$$

where $\bar{p} = q\alpha\gamma\bar{p}_{22}$ is the mean value of the stationary photocurrent, and $\rho(s) = q\alpha\gamma\rho_{22}^{(11)}(s)$ (for simplicity we use the same symbol for the original and its Laplace transform, making only the argument change $T \rightarrow s$). Using (6), we carry out a formal summation of the series (2) in the Laplace representation:

$$Q(\lambda, s) = \frac{1}{s} - \frac{\lambda\bar{p}}{s^2} \frac{1}{1+\lambda\rho(s)}. \quad (7)$$

From the condition for the normalization of the distribution $p(n, T)$ it follows that the Laplace transform $Q(\lambda, s)$ of the generating function (1) exists under the condition $\text{Re}(s) > 0$ and is analytic in λ , at least in the open circle $|1 - \lambda| < 1$. On the other hand, the function (7) appears as a sum of a series that converges under the condition $|\lambda\rho(s)| < 1$. From the explicit form of $\rho_{22}^{(11)}(s)$ [see Eqs. (10) and (11) below] it is clear that $\rho_{22}^{(11)}(s)$ becomes arbitrarily small if the region of permissible values of s is shifted far enough to the right. Therefore for any $\varepsilon > 0$ it is possible to satisfy the inequality $|\lambda\rho(s)| < 1$ in the circle $|\lambda| < 1 + \varepsilon$, confining oneself to the region $\text{Re}(s) > A(\varepsilon)$ in which $|\rho(s)| < (1 + \varepsilon)^{-1}$. The function $Q(\lambda, s)$ obtained in the form (7) under the indicated limitation, can be expanded in the ε -vicinity of the point $\lambda = 1$ in the series (1) for the Laplace transforms. We thus obtain

$$p(n, s) = \frac{\bar{p}}{s^2} \frac{\rho(s)^{n-1}}{[1+\rho(s)]^{n+1}}, \quad n \geq 1, \quad (8)$$

$$p(0, s) = \frac{1}{s} - \frac{\bar{p}}{s^2} \frac{1}{1+\rho(s)}.$$

The same result can be obtained directly by expressing $p(n, s)$ in terms of $Q_i(s)$ using the formula

$$p(n, s) = \frac{1}{n!} \sum_{m=0}^{\infty} \frac{(-1)^m}{m!} Q_{n+m}(s). \quad (9)$$

The series (9), with account taken of (6), converges again under the condition $|\rho(s)| < 1$. But the Laplace transform $p(n, s)$ of any of the sought functions $p(n, T)$ exists and is analytic in the entire $\text{Re}(s) > 0$ plane [since $p(n, T) \leq 1$]. Inasmuch as $p(n, s)$ is determined in the region $\text{Re}(s) > A(\varepsilon)$ (see above) by expressions (8), and the function (8) are meromorphic, we arrive at the conclusion that they represent $p(n, s)$ everywhere at $\text{Re}(s) > 0$.

We present now the expressions for $\rho_{22}^{(11)}(s)$ and \bar{p}_{22} , which follow from the solution of the system of equations for the elements of the density matrix of a two-level atom in a strong monochromatic field (see, e.g., Ref. 5):

$$\rho_{22}^{(11)}(s) = D(s)^{-1} V_0^2 (2s + \gamma); \quad (10)$$

$$D(s) = s \left[s^2 + 2\gamma s^2 + \left(\frac{5\gamma^2}{4} + \nu_0^2 + 4V_0^2 \right) s + \gamma \left(\frac{\gamma^2}{4} + \nu_0^2 + 2V_0^2 \right) \right], \quad (11)$$

$$\bar{p}_{22} = \lim_{s \rightarrow 0} \{s\rho_{22}^{(11)}(s)\} = \frac{V_0^2}{\gamma^2/4 + \nu_0^2 + 2V_0^2}. \quad (12)$$

Here $\nu_0 = \omega_0 - \omega_{21}$ is the detuning from resonance, $V_0 = |d_{21} \cdot \mathbf{E}_0|/2\hbar$, ω_0 is the frequency, \mathbf{E}_0 is the amplitude of the incident light wave, and ω_{21} and d_{21} are the

frequency and the dipole moment of the transition of the atom.

3. From expressions (8) it is easy to show that

$$\bar{n} = \langle n \rangle = q\alpha\gamma\bar{p}_{22}T = \bar{p}T, \quad (13)$$

$$\sigma^2 = \langle (n - \bar{n})^2 \rangle = \bar{n}[1 + \xi(T)]; \quad (14)$$

$$\xi(T) = q\alpha\gamma \frac{2}{T} \int_0^T (T - \tau) [\rho_{22}^{(11)}(\tau) - \bar{p}_{22}] d\tau. \quad (15)$$

The result (14) for the dispersion of the number of photocounts was obtained and discussed in Ref. 7. It is also possible to obtain the asymmetry of the distribution $p(n, T)$, defined by the formula

$$\gamma_1 = \langle (n - \bar{n})^3 \rangle / \sigma^3. \quad (16)$$

In the general case the expression for γ_1 turns out to be too cumbersome. A lucid result is obtained for sufficiently long observation time

$$T \gg \max\{\gamma^{-1}, |\nu_0|^{-1}, V_0^{-1}\}. \quad (17)$$

Under this condition

$$\gamma_1 = (1 + 6r + 6r^2) / (1 + 2r)^3. \quad (18)$$

Here

$$r = \lim_{s \rightarrow 0} r(s), \quad r(s) = \rho(s) - \frac{\bar{p}}{s}, \quad (19)$$

$$r = \frac{1}{2} \lim_{T \rightarrow \infty} \xi(T)$$

$$= q\alpha\gamma \int_0^{\infty} [\rho_{22}^{(11)}(\tau) - \bar{p}_{22}] d\tau.$$

The asymmetry (18) can be smaller than for the corresponding Poisson distribution [i.e., less than $(\bar{n})^{-1/2}$], and becomes negative at $r \leq -0.17$.

By way of example, the figure shows the result of a numerical calculation [the inverse Laplace transform of expressions (8)] for concrete values of the parameters. We call attention to the narrowing of the distribution (by almost a factor of 2) compared with the Poisson distribution, and the faster decrease at $n > \bar{n}$. It can be stated that the realization of the photocurrent pulses is in this case more uniform than the realization of the Poisson process; this corresponds to antibunching of the photons in time.

4. In the case of a sufficiently long observation time, when $\bar{n} \equiv \bar{p}T \gg 1$, it is possible to obtain analytically rather simple approximate representations of the distribution function $p(n, T)$.

It is easy to verify that complete neglect of the photon correlation, namely the replacement $r(s) \rightarrow 0$, i.e., $\rho_{22}^{(11)}(\tau) \rightarrow \bar{p}_{22}$ [see Eqs. (19) and (8) or (6) and (9)] leads to a Poisson distribution. Let us obtain an approximate expression for $p(n, T)$ under the condition of relatively small correlations. When taking the inverse Laplace transforms of expressions (6) for $Q_n(s)$ we confine ourselves to the pole $s=0$ and leave out the terms with the derivatives $r^{(m)}(s)_{s=0}$. It can be shown that this corresponds to the condition $n \ll \bar{n}/|r|$ [see Eqs. (19)]. We then obtain in the indicated region of n approximate expressions for the factorial

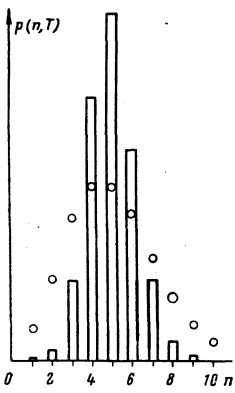


FIG. 1. Distribution $p(n, T)$ of the photocounts. Columns—results of numerical Laplace inverse transformation of the functions (8); $\bar{n} = 5$, $\nu_0 = 0$, $V_0^2 = \gamma^2/8$ [see Eqs. (10)–(12)]; circles—Poisson distribution at $\bar{n} = 5$; $q\alpha = 1$.

moments $Q_n(T)$ (we do not present it here for the sake of brevity). To find the generating function $Q(\lambda, T)$ from formula (2) it is necessary to take into account a sufficiently large number M of the factorial moments. Namely, since a rough estimate yields $Q_n(T) \sim (\bar{\rho}T)^n = \bar{n}^n$, we must choose $M \gg \bar{n}$. This requirement with respect to the numbers of the approximately obtained factorial moments $Q_n(T)$, together with the limitation $n \ll \bar{n}/|\tau|$, leads to the condition $|\tau| \ll 1$. We then obtain for the generating function

$$Q(\lambda, T) = \exp\left\{-\frac{\bar{n}}{1+\lambda r}\right\}, \quad \bar{n} = \bar{\rho}T. \quad (20)$$

Under the same restrictions ($n \ll \bar{n}/|\tau|$, $|\tau| \ll 1$) we obtain from (20) on the basis of the definition (1)

$$p(n, T) = \exp\left\{-\frac{\bar{n}}{1+r}\right\} \left(\frac{r}{1+r}\right)^n L_n^{(-1)}\left(-\frac{\bar{n}}{r(1+r)}\right). \quad (21)$$

Here $L_n^{(-1)}(x) = L_n(x) - L_{n-1}(x)$, where $L_n(x)$ is a Laguerre polynomial. We note that the upper limit of the zeros of the polynomial $L_n^{(-1)}(x) = x_0 \approx 4n$ (Ref. 8). Therefore formula (21) is valid also at $r < 0$ (antibunching condition) and $|\tau| \ll 1$ in a sufficiently large region of values of n , containing \bar{n} .

We obtain now an expression for $p(n, T)$, which is valid for all values of r , but only in the central region $|n - \bar{n}| \ll \bar{n}$ under the condition $\bar{n} \gg 1$. We write down the inverse Laplace transform of expression (8) for $p(n, s)$ in the form

$$p(n, T) = p(\eta, \bar{n}) = \frac{1}{\bar{n}} \frac{1}{2\pi} \int_{-\infty}^{\infty} dx \exp\{ix\} \quad (22)$$

$$\times \exp\left\{\bar{n}(1+\eta) \left[\ln\left(1 + \frac{ix}{\bar{n}} r(i\bar{\rho}x/\bar{n})\right) - \ln\left(1 + \frac{ix}{\bar{n}} + \frac{ix}{\bar{n}} r(i\bar{\rho}x/\bar{n})\right) \right]\right\}.$$

Here $\eta = (n - \bar{n})/\bar{n}$; we made the substitutions $s = ix/T$ and $n \pm 1 \rightarrow n = \bar{n}(1 + \eta)$. If $|\eta| \leq (n)^{-1/2}$, then the vital region in the integral (22) is $|x| \leq (\bar{n})^{1/2}$, since the poles s_k of the function $r(s)$ lie in the left half-plane and $|\operatorname{Re}(s_k)|$ if of the order of γ [Eq. (11)], and we can assume in (22) that $|r(\dots)| \leq 1$. We expand both logarithms in (22) in powers of x/\bar{n} and retain the quadratic terms [by leaving only the linear terms we would obtain $p(n, T) = \delta(n - \bar{n})$]. We arrive ultimately at the Gaussian distribution [in the region $|n - \bar{n}| \leq (n)^{1/2}$]:

$$p(n, T) \approx [2\pi\bar{n}(1+2r)]^{-1/2} \exp\left\{-\frac{(n-\bar{n})^2}{2\bar{n}(1+2r)}\right\} \quad (23)$$

[we recall that here $r = r(0)$, Eqs. (19)]. The result (23) agrees with the central limit theorem. The number n of the photocounts during the time $T \gg \tau_c$, where $\tau_c \approx |\tau|/\bar{\rho}$ is the characteristic photon-correlation time, can be regarded as the sum of the large number of almost independent random quantities n_k —the numbers of the photocounts in the time intervals T_k ($\tau_c \ll T_k \ll T$). The pair correlation of the photons manifests itself in this approximation in the photocount-distribution variance, which coincides with the exact value (14).

5. We consider in conclusion the possibility of experimentally determining the function $p(n, T)$ for the NRF of one atom in experiments of the type performed in Refs. 1 and 2 with an extremely rarefied atomic beam. Many-atom interference effects and fluctuations of the number of atoms in the region of their interaction with the laser radiation can alter substantially for the form of the distribution $p(n, T)$ compared with that obtained above. Therefore each individual measurement of the number of photocounts must be carried out in the reliable presence of one atom in the interaction region. The most direct method of separating “single-atom” situations is control of the intensity, or more accurately of the total number of photocounts during the transit time. The time intervals T of the samplings are chosen inside the time during which the atom traverses the region of interaction with the laser beam (T_0). In the statistical analysis one takes into account only those intervals T_0 for which it has turned out that $|n(T_0) - \bar{n}(T_0)| < [n(T_0)]^{1/2}$ (here $\bar{n}(T_0) = \bar{\rho}T_0$ is the expected number of photocounts from one atom). Such an organization of the experiment seems realistic in connection with the development of the technique of laser detection of individual atoms.⁹ In principle it is possible also to use the features of the correlation of the intensity in the NRF of a system of two or more atoms. For example, recording of excess coincidences of photocounts during a time on the order of the reciprocal Doppler width of the line, at large photon-gathering angles, would be evidence of the presence of more than one atom in the interaction region.

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