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Photon Statistics of Fluorescence from a Single Three-Level Atom*

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Abstract. A sharp increase in the variance of the photon counting distribution for long collection times indicates an intermittent fluorescence signal, caused by (macroscopic) quantum jumps. Mandel's Q-parameter presents a convenient measure for the deviation of the actual statistics from that of a Poisson process. While the short time limit of $Q(T)$ reflects the quantum statistical properties of individual emission events, the long time behaviour is dominated by the quasi-classical aspects of a random sequence of bright and dark periods. The typical signatures of quantum jumps, as they appear in the statistical description, can be visualized when comparing fluorescence from a two- and a three-level system. The classical aspects become obvious when comparison is made between the properties of a classical random telegraph signal and those of the quantum statistical calculation.

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To emphasize the various peculiar features of quantum mechanical processes, it has been particularly helpful in the past to devise elementary and transparent experiments that involve e.g. only a few atoms. Oversimplified as they were, those experiments illustrated very clearly certain specific aspects of the quantum mechanical formalism and were of invaluable pedagogical importance. However, it was commonly understood that they are forbiddingly unrealistic, so they could never be performed in practice. Nobody in the time of Einstein and Schrödinger was able to prophesy the revolutionary technical developments that would bring those Gedanken-experiments into reach of a laboratory test. Since it became possible to store and to manipulate a small number or even a single ion for an arbitrarily long time in a trap [1], those "impossible" Gedanken-experiments suddenly became feasible.

Among such suggested elementary experiments is the measurement of the randomness of fluorescence photons from a single atom. Particularly attractive was the idea to look for photons from a long-lived state, since here the quantum jumps should occur at convenient time intervals of between 1 and 100 s – the corresponding signal, however, would be extremely weak. In order to raise that signal to a measurable macroscopic level, it was suggested that one couples the forbidden transition via a common level to an allowed transition [2, 3]. The individual quantum jumps from the metastable state could then be monitored by the random appearance and extinction of the strong fluorescence signal. This phenomenon is obviously a unique single atom effect, which is averaged out entirely in the presence of a number of scatterers, since part of the atomic ensemble contributes to fluorescence, while the rest remains shelved in the metastable state.

For the description of this quantum mechanical single-particle effect, it is necessary to formulate the photon statistics in greater detail. A quite general tool for the characterization of the statistical behaviour of light is the photon counting probability $W(n, T, \eta)$ i.e. the probability of registering n events in an observation

* Dedicated to Prof. Dr. Herbert Welling on the occasion of his 60th birthday

interval of duration T. Here η is the quantum efficiency of the entire detection process, which takes into account that not every photon emitted is also recorded. Spontaneous emission is triggered by vacuum fluctuations, therefore the counting probability must be derived in a quantum mechanically consistent way. Cook and Kimble [4] have revived interest in this problem by a heuristic description of the jump probabilities and subsequent papers have treated the problem with various degrees of rigor – from plausible assumptions to attempts at a first-principles derivation [5–15]. Since Dehmelt first suggested this phenomenon in 1975, it was more than 10 years before this effect was demonstrated experimentally [16–18].

While the counting probability $W(n, T, \eta)$ for arbitrary n provides a detailed description, it can hardly ever be obtained analytically for the general case. Even the probability of observing no event $W(n=0, T, \eta)$ already requires the knowledge of the entire hierarchy of multiphoton correlation functions [10]. Provided it is possible to calculate the "no event" probability $W(n=0, T, \eta)$ in analytical form, then it is a straightforward task to find the probability for arbitrary n by differentiation:

$$W(n, T, \eta) = (-1)^n \frac{\eta^n}{n!} \frac{\partial^n}{\partial \eta^n} W(n=0, T, \eta). \tag{1}$$

The analytical form is essential, since we are interested in n varying from 1 to 10^8. A more compact, but less complete characterization of the statistical behaviour is contained in the ensemble averages, such as the moments and the variances of the photon counting rates:

$$\langle n \rangle = \sum_{n=0}^{\infty} n W(n, T, \eta), \tag{2}$$

$$\langle \Delta^2 n \rangle = \sum_{n=0}^{\infty} (n - \langle n \rangle)^2 W(n, T, \eta) \tag{3}$$

or Mandel's Q-parameter

$$Q(T) = \frac{\langle \Delta^2 n \rangle}{\langle n \rangle} - 1. \tag{4}$$

It may appear that in order to calculate those averages we would need the complete probability distribution $W(n, T, \eta)$ anyway, and by summing over n we could only lose most of that information again. Fortunately this is not the case, and there is a less cumbersome way to obtain those averages directly. It is entirely sufficient to know $W(n=0, T, \eta)$ and its first and second order derivatives to calculate the average photon number and its variance. This can be seen by inserting (1) into (2) and (3) and by interpreting the obtained sum as a Taylor series:

$$\langle n \rangle = -\eta \left(\frac{\partial}{\partial \eta} W(0, T) \right)_{\eta=0}, \tag{5}$$

$$\langle \Delta^2 n \rangle = \langle n \rangle + \eta^2 \left[\left(\frac{\partial^2 W(0, T)}{\partial \eta^2} \right)_{\eta=0} - \left(\frac{\partial W(0, T)}{\partial \eta} \right)^2_{\eta=0} \right], \tag{6}$$

$$Q(T) = -\eta \frac{\left[\left(\frac{\partial^2 W(0, T)}{\partial \eta^2} \right)_{\eta=0} - \left(\frac{\partial W(0, T)}{\partial \eta} \right)^2_{\eta=0} \right]}{\left(\frac{\partial W(0, T)}{\partial \eta} \right)_{\eta=0}}. \tag{7}$$

In order to demonstrate how the occurrence of quantum jumps can be read off from the variance, we compare the properties of a driven two-level system which lacks this feature, with the fluorescence signal from a three-level atom. Rate equations are compared with the more involved Bloch treatment to demonstrate the role of coherent superposition states and Rabi oscillations.

It has been suggested – rather convincingly – on the basis of classical arguments that, in the limit $T \to \infty$, $Q(T)$ must vanish in general [19]. The basic idea was that for large collection times in a photon counting experiment, the intensity fluctuations should be averaged out, since the intensity enters only under the time integral. In the quantum mechanical formulation, however, this is clearly not the case, neither for the two- nor for the three-level system. In such a situation one might be tempted to attribute this discrepancy to the action of quantum fluctuations. On closer inspection, however, it appears highly improbable that such a large deviation, especially in the long time limit, should be a consequence of quantum effects. In order to clarify this point, we present an entirely general and classical calculation, based on a Markovian process which demonstrates clearly that there exists no universal long time limit for $Q(T)$. A model of dichotomous noise, which intuitively simulates the intermittent fluorescence classically, is presented in Sect. 2.3 and compared with the quantum result. While the predictions do not agree in the short time limit, they do so quantitatively for long times. This comparison is helpful for the interpretation of the general quantum mechanical result.

1. General Definitions and Results

1.1. *Quantum Mechanical Photon Counting Statistics*

In a photon-counting experiment light falls on the cathode of a photomultiplier creating a sequence of random electrical pulses. The average counting rate is proportional to the intensity of the incident field. In this section the photon counting statistics is determined for systems where the quantum nature of the field as well as of the detection process is relevant. The probability of observing n counting events in a time interval of length T was first evaluated in a quantum mechanically consistent way by Glauber [20] and independently by Kelley and Kleiner [21]

$$W(n,T)=\frac{1}{n!}\operatorname{Tr}\left\{\varrho\hat{T}\left(\eta\int_0^T\gamma b^+(t)b(t)dt\right)^n \times\exp\left(-\eta\int_0^T\gamma b^+(t)b(t)dt\right)\right\}. \tag{8}$$

The intensity of the light is given by $\gamma b^+(t)b(t)$, where $b(t)$ and $b^+(t)$ are the boson operators of the field. γ denotes the spontaneous emission rate of the considered transition. We combine the quantum efficiency η, which is typically of the order 10^{-5}, with γ to give the factor $\lambda=\eta\gamma$. The trace over the statistical operator of the field ϱ corresponds to an averaging over the fluctuations of the field intensity in the classical case. Characteristic for the quantum mechanical nature of expression (8) is also the operator $\hat{T}$, which introduces an ordering of the non-commuting operators $b(t)$ and $b^+(t)$, according to the following rules:
- normal ordering of the operators b^+ and b,
- time ordering of the operators $b(t)$ with time increasing to the left,
- time ordering of the operators $b^+(t)$ with time increasing to the right.

Normal ordering guarantees that the probability of detecting more photons than are contained in the field vanishes. The time ordering is a remnant of the perturbative origin of (8).

Given $W(n,T)$ we are in a position to determine all relevant statistical properties, like the mean value or the variance of the photon number. The mean number of photons counted during an interval T is related to the mean intensity $\gamma\,\langle b^+b\rangle$ by:

$$\langle n\rangle=\sum_{n=0}^{\infty} nW(n,T)=\lambda\int_0^T\langle b^+(t)b(t)\rangle dt. \tag{9}$$

Similarly, the variance of the photon number is related to the two-photon correlation function:

$$\langle\Delta^2 n\rangle=\langle n^2\rangle-\langle n\rangle^2=\langle n\rangle-\langle n\rangle^2 +2\lambda^2\int_0^T dt\int_0^t dt'\langle b^+(t')b^+(t)b(t)b(t')\rangle. \tag{10}$$

The deviation of the statistics from that of a Poisson process is conveniently characterized by Mandel's Q-parameter:

$$Q(T)=\frac{\langle\Delta^2 n\rangle-\langle n\rangle}{\langle n\rangle} =-\langle n\rangle+\frac{2\lambda^2}{\langle n\rangle}\int_0^T dt\int_0^t dt'\langle b^+(t')b^+(t)b(t)b(t')\rangle\geqq -1 \tag{11}$$

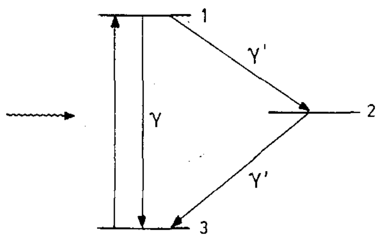

Fig. 1. Energy-level diagram considered in this paper. The ground level $|3\rangle$ is coupled coherently to level $|1\rangle$ that can decay either back to $|3\rangle$ by induced or spontaneous emission or to the metastable state $|2\rangle$ and from there on to $|3\rangle$. We will consider the case where $\gamma\gg\gamma'$

A light field with a stable intensity displays only the Poissonian fluctuations of the measurement process and therefore $Q=0$. Fluctuations of the light intensity, if they are of classical origin, are observed as an additional broadening above the Poisson limit i.e. $Q>0$. Sub-Poissonian statistics with a variance less than that of a Poisson process, i.e. $Q<0$, is a subtle quantum phenomenon, since it is not compatible with the laws of classical statistics.

Under the assumption of stationarity, G_1 is time independent and $G_2(t,t')$ depends only on the time difference $t-t'$:

$$G_1(t)=\langle b^+(t)b(t)\rangle=G_1(0), \tag{12}$$

$$G_2(t,t')=\langle b^+(t')b^+(t)b(t)b(t')\rangle=G_2(|t-t'|). \tag{13}$$

As a consequence, one of the integrals in (9) and (11) can be carried out explicitly and we obtain:

$$\langle n\rangle=\lambda G_1 T, \tag{14}$$

$$Q(T)=\lambda G_1 T\left[\frac{2}{T}\int_0^T\frac{G_2(t)}{G_1^2}\left(1-\frac{t}{T}\right)dt-1\right]. \tag{15}$$

In order to calculate the field correlation functions G_1 and G_2 we have to relate the properties of the field to those of the atomic sources. We will consider here a three-level atom with an allowed and a forbidden transition, where the allowed one is driven resonantly by a laser field (Fig. 1). This is a level configuration which is known to display quantum jumps [5, 7, 12, 15, 22]. In order to emphasize the typical traces left by the jumps in the photon statistics, we will compare the results for the three-level system with those derived for a two-level atom.

The amplitude of the fluorescence field is determined by the polarization of the atomic source and we find, apart from geometrical factors [23, 24]:

$$G_1=\varrho_{11}^{SS}, \tag{16}$$

$$G_2(t)=\varrho_{11}^{SS}h(t), \tag{17}$$

$$Q(T)=\frac{2\lambda}{T}\int_0^T dt\int_0^t dt' h(t')-\lambda\varrho_{11}^{SS}T, \tag{18}$$

where $h(t)$ is the probability of finding the system in the excited state of the fluorescent transition if it had been prepared in the ground state initially. $h(t)$ is obtained as the solution of the corresponding Bloch equations. The stationary population of the excited state ϱ_{11}^{SS} is the limit of $h(t)$ for $t \to \infty$. Before presenting the details of the calculations in the next section, we first want to discuss some general properties and the limiting behaviour of $Q(T)$.

For small times T, $Q(T)$ is obtained by Taylor series expansion of $h(t)$ in (18):

$$Q(T) = -\lambda \varrho_{11}^{SS} T + \frac{\lambda}{3} \dot{h}(t=0) T^2 . \tag{19}$$

It should be noticed that $Q(T)$ always assumes negative values for small times, corresponding to sub-Poissonian statistics, irrespective of the specific process under consideration. $h(t=0)$ vanishes by definition and since $h(t)$ is positive for all times, $\dot{h}(t=0)$ must be positive as well. The sub-Poissonian character becomes most prominent for times of the order $T = 3\varrho_{11}^{SS}/2\dot{h}(t=0)$ where $Q(T)$ has the value $Q = -3\lambda(\varrho_{11}^{SS})/4\dot{h}(t=0)$.

In the limit of long collection times $T \to \infty$ we find

$$\lim_{T \to \infty} Q(T) = 2\lambda \lim_{z \to 0} \frac{\partial z h(z)}{\partial z}, \tag{20}$$

where $h(z)$ is the Laplace transform of $h(t)$. $h(z)$ is obtained directly from the Bloch equations and does not require their solution, as shown in more detail in Sect. 2.

One might suspect naively that $Q(T \to \infty)$ vanishes in general, since the photon statistics in (8) depends only on the time integral over the intensity. For times T large compared to the characteristic time scales of the intensity fluctuations, the time integral, apart from the norm, becomes identical to the time average. When we identify the time average for a moment with the ensemble average over the intensity, then the trace in (8) is to be taken only over the density operator ϱ and the photon distribution obviously becomes Poissonian [19]. This intuitive line of reasoning, however, is not correct, certainly not for the quantum mechanical case, nor even for the classical one. $Q(T=\infty)$ vanishes only under rather special additional conditions, as can be seen from (20) or equivalently from:

$$\lim_{T \to \infty} \left(\int_0^T [G_2(t) - G_1^2] dt \right) = 0 \quad \text{or} \quad \int_0^\infty [h(\infty) - h(t)] dt = 0. \tag{21}$$

We will demonstrate that for a two-level system these relations are asymptotically satisfied e.g. for strong coherent as well as incoherent pumping, while for a three-level system they have no solution for any relevant choice of parameters.

1.2. Classical Photon Counting Statistics

In order to separate the classical from the quantum behaviour, it is useful to summarize briefly the results obtained from a classical description. The term photon is used here only to indicate the discreteness of the photon counting events and does not relate to the quantized states of the field. It is in this sense that we use the term "classical photon statistics". While the limiting behaviour of $Q(T)$ for small time intervals depends sensitively on the use of the classical or the quantum description, the deviation from the Poisson statistics for long times is observed in both cases. The basic equation (8) simplifies considerably for a classical light field, when we replace the field operator $\gamma b^+(t) b(t)$ by the classical intensity $I(t)$ and $\hat{T}$ by unity

$$W(n, T) = \left\langle \frac{1}{n!} \left(\eta \int_0^T I(t) dt \right)^n \exp\left(-\eta \int_0^T I(t) dt \right) \right\rangle . \tag{22}$$

The brackets denote the ensemble average over the field fluctuations, since $I(t)$ has now the meaning of a classical stochastic variable. The main statistical properties follow from the mean photon number and its variance, similarly to (9) and (10),

$$\langle n \rangle = \eta \int_0^T \langle I(t) \rangle dt , \tag{23}$$

$$Q(T) = \eta \frac{\int_0^T dt \int_0^T dt' \langle I(t) I(t') \rangle - \left(\int_0^T \langle I(t) \rangle dt \right)^2}{\int_0^T \langle I(t) \rangle dt} . \tag{24}$$

We assume that the intensity fluctuations can be characterized by a continuous Markov process described by a Fokker-Planck equation, quite similar to the traditional laser models. $P(I, t) dI$ is the probability of finding the field intensity in the interval between I and $I + dI$ at a time t. $P(I, t)$ is obtained as the solution of an appropriate Fokker-Planck equation:

$$\frac{\partial P}{\partial t} = L(I, \partial/\partial I) P(I, t) , \tag{25}$$

where L is the Fokker-Planck operator for the fluorescence process. We denote the eigenfunction of the Fokker-Planck equation (25) by $P_n(I)$ and the eigenvalues by λ_n, which have the meaning of relaxation rates, $\lambda_n > 0$. Then the transient solution of the Fokker-Planck equation can be written in the form:

$$P(I, t) = \sum_{n=0}^{\infty} c_n P_n(I) \exp(-\lambda_n t) , \tag{26}$$

where c_n is determined by the initial condition. Since we have made use of the Markov assumption, the joint stationary probability density is given by:

$$P(I_1, t_1, I_2, t_2) = \sum_{n=0}^{\infty} P_n(I_1) P_n(I_2) \exp[-\lambda_n (t_1 - t_2)] . \tag{27}$$

With these definitions it is straightforward to express the moments and variances of the photon counting statistics in terms of the eigenvalue solutions. We find

$$G_1(t)=G_1(0)=W_0\,, \tag{28}$$

$$G_2(t)=W_0^2+\sum_{n=1}^{\infty} W_n^2 e^{-\lambda_n t} \tag{29}$$

and

$$Q(T)=\frac{2\eta}{W_0}\sum_{n=1}^{\infty}\frac{W_n^2}{\lambda_n}\left[1-\frac{1}{T\lambda_n}(1-e^{-\lambda_n T})\right], \tag{30}$$

where we used the following definition:

$$W_n=\int_0^{\infty} xP_n(x)dx \qquad n=0,1,2,\ldots. \tag{31}$$

Now it is possible to discuss the properties of $Q(T)$ and its asymptotic behaviour for an arbitrary classical process. The only restriction is that the intensity fluctuations are assumed to be Markovian. We find in the long time limit:

$$\lim_{T\to\infty} Q(T)=\frac{2\eta}{W_0}\sum_{n=1}^{\infty}\frac{W_n^2}{\lambda_n}>0\,, \tag{32}$$

and for small times T

$$Q(T)=\frac{\eta}{W_0}\sum_{n=1}^{\infty} W_n^2 T\geqq 0\,. \tag{33}$$

When comparing the classical results here with the quantum mechanical expressions above we find the following significant differences:

– $Q\geqq 0$ for all T, i.e. a classical process never exhibits sub-Poissonian statistics.

– For small times $Q(T)$ also approaches zero linearly, but with a positive slope in contrast to the quantum mechanical result.

– For large times $Q(T)$ assumes a finite positive value and there is no general reason why this asymptotic value should vanish. In certain limiting cases it might be numerically small but still non-zero. Obviously for low quantum efficiencies $\eta \ll 1$ the Poisson statistics of the measurement process dominates and leaves one with a small value for Q. Also when all relaxation rates λ_n tend to diverge, the deviation from a Poisson process can become negligible. In general, however, the behaviour for large times still depends on the details of the process and no universal behaviour is obtained in this limit. The intuitive picture that the intensity fluctuations are averaged out by the time integration in (22), leaving us qualitative with the result of a coherent field, is not correct, not even in the classical limit. It is the short time regime where quantum effects are to be seen, while the long time behaviour is of more classical nature. As we will see below, the occurrence of quantum jumps is indicated by a large positive value of $Q(T)$ for long times – this follows from the quantum, as well as the classical description.

2. Mandel's Q-Parameter for Two- and Three-Level Systems

With the general formulas derived in the previous section, we are now in a position to calculate the Q-parameter explicitly for an externally driven three-level system that exhibits quantum jumps. The key to these results is the solution of the atomic dynamics under the influence of coherent driving fields, i.e. the three-level Bloch equations. In limiting cases the solutions can be obtained in analytical form, while for the general case one has to resort to numerical methods. If coherence effects do not play an essential role, one may simplify the calculation by making use of the rate equation approximation. The Q-parameter, which provides only an estimate for the noisiness of the field, nevertheless allows one to distinguish normal Poissonian fluorescence from the occurrence of quantum jumps. The typical traces of intermittent fluorescence are easily recognized when we compare the three-level system where the jumps occur, with a driven two-level system. Since the latter results have been derived previously [25], we only summarize them here in the present notation.

2.1. Two-Level System

Rate Equations. We assume a two-level atom which is driven coherently with a rate R. The excited level $|1\rangle$ decays spontaneously with a rate γ into the ground state $|0\rangle$. The population of state $|1\rangle$ changes in time according to

$$h(t)=\varrho_{11}^{SS}(1-e^{-(2R+\gamma)t})\,, \tag{34}$$

when it was initially prepared in the ground state. ϱ_{11}^{SS} is the stationary population of state $|1\rangle$:

$$\varrho_{11}^{SS}=\lim_{t\to\infty} h(t)=\frac{R}{2R+\gamma}\,. \tag{35}$$

$Q(T)$ can be calculated according to (18) with the help of (34) and (35):

$$Q(T)=-\frac{2R\lambda}{(2R+\gamma)^2}\frac{1}{T(2R+\gamma)}\left[e^{-(2R+\gamma)T}-1+(2R+\gamma)T\right]. \tag{36}$$

Obviously, $Q(T)$ is negative for all times T. Thus even the simplest description of a two-level system by the "classical" rate equations already shows non-classical behaviour – namely sub-Poisson statistics – for the photon counting events. This is due to the non-classical way of calculating the ensemble averages above, where time and normal ordering must be taken into account.

In this approximation, the time derivative of $Q(T)$ is also negative and so $Q(T)$ is a monotonically decreasing function. For small times T we find:

$$Q(T)=-\lambda T\left(\varrho_{11}^{SS}-\frac{RT}{3}\right), \tag{37}$$

while for large times, Q approaches the asymptotic value:

$$Q_\infty=\lim_{T\to\infty} Q(T)=-\frac{2R\lambda}{(2R+\gamma)^2}. \tag{38}$$

This example, in agreement with the general expressions above, shows a linear decrease for small times and a non-vanishing asymptotic value for large times. In the limit of strong saturation $Q\to 0$ and the fluorescence signal becomes Poissonian for all times. The dynamics becomes less trivial when we allow for coherence effects and discuss the statistics in terms of the Bloch dynamics.

Bloch Equations. The two-level Bloch equations can be solved by Laplace transformation and we obtain

$$h(z)=\frac{2\chi^2(z+1/T_2)}{z[4\chi^2(z+1/T_2)+(z+1/T_1)(\Delta^2+(z+1/T_2)^2)]}, \tag{39}$$

where T_1 and T_2 are the longitudinal and transverse relaxation times and Δ is the detuning parameter. The Rabi frequency χ is proportional to the electric dipole moment μ and the electric field strength E

$$\chi=\frac{\mu E}{2\hbar}. \tag{40}$$

The calculations can be simplified for resonant excitation $\Delta=0$ and for the case where spontaneous emission is the only decay mechanism; then $1/T_1=\gamma$ and $1/T_2=\gamma/2$. In this case $h(z)$ simplifies:

$$h(z)=\frac{2\chi^2}{z[4\chi^2+(z+\gamma)(z+\gamma/2)]}. \tag{41}$$

In order to invert the Laplace transformation, only a quadratic eigenvalue equation has to be solved.

The stationary solution can be determined from $h(z)$ by taking the limit:

$$\varrho_{11}^{SS}=\lim_{z\to 0} zh(z)=\frac{2\chi^2}{4\chi^2+\gamma^2/2}. \tag{42}$$

Equations (41) and (42) are inserted into (18) and we obtain

$$Q(T)=Q_\infty\left[1+\frac{4\chi^2-\frac{7}{4}\gamma^2}{4\chi^2+\gamma^2/2}\left(\frac{1-e^{-\frac{3}{4}\gamma T}\cos\zeta T}{\frac{3}{2}\gamma T}\right.\right.$$
$$\left.\left.-\frac{3}{2}\frac{4\chi^2-\gamma^2/4}{4\chi^2-\frac{7}{4}\gamma^2}e^{-\frac{3}{4}\gamma T}\frac{\sin\zeta T}{\zeta T}\right)\right] \tag{43}$$

where oscillations occur for $\chi>\gamma/8$ with a frequency:

$$\zeta=\sqrt{4\chi^2-\gamma^2/16}. \tag{44}$$

An asymptotic nonvanishing value is obtained for $T\to\infty$

$$Q_\infty=-\frac{6\lambda\gamma\chi^2}{(4\chi^2+\gamma^2/2)^2}. \tag{45}$$

Only under the additional assumption of saturation i.e. $\chi\gg\gamma$ is the Poisson property recovered in the long time limit.

For small times T the signal again shows sub-Poissonian behaviour and decreases linearly starting from $Q(T=0)=0$

$$Q(T)=-\frac{2\lambda\chi^2}{4\chi^2+\gamma^2/2}T+\frac{\lambda\chi^2}{6}T^3. \tag{46}$$

As a general result for the two-level system, we notice that $Q(T)\leqq 0$ for all T, and only Poisson or sub-Poisson statistics is observed. This holds for the rate equations as well as for the coherent dynamics and agrees qualitatively with experimental results [26, 27]. In contrast to this behaviour we expect that the enormous fluctuations of an intermittent signal must lead to an enhancement of noise in the long time limit and therefore Q should assume large positive values.

2.2. Three-Level System

Rate Equations. A resonantly driven three-level system can be described approximately by rate equations, which give a first insight into its dynamic behaviour. Since they are much easier to handle than the more appropriate Bloch equations, we will start with this simplified case first. The rate equations for the populations of the three atomic levels (Fig. 1) read

$$\begin{aligned}\dot\varrho_{11}&=-(\gamma_{12}+\gamma_{13}+R)\varrho_{11}+R\varrho_{33},\\ \dot\varrho_{22}&=\gamma_{12}\varrho_{11}-\gamma_{23}\varrho_{22},\\ \dot\varrho_{33}&=(R+\gamma_{13})\varrho_{11}+\gamma_{23}\varrho_{22}-R\varrho_3,\end{aligned} \tag{47}$$

where R is the laser-induced transition rate into the state $|1\rangle$ and the spontaneous emission rates are γ_{12}, γ_{13}, and γ_{23}. For simplification we assume that $\gamma_{12}=\gamma_{23}$ and abbreviate:

$$\gamma=\gamma_{13},\quad \gamma'=\gamma_{12}=\gamma_{23}. \tag{48}$$

In all other limiting cases the population is either stuck in level $|2\rangle$ or does not stay there long enough to efficiently quench the fluorescence from the allowed transition and quantum jumps disappear.

The probability $h(t)$ of finding the system in the upper level $|1\rangle$, if it has been in the ground state $|3\rangle$ at $t=0$, can be found by solving the set of differential equations (47) through Laplace transformation

$$h(z)=\frac{1}{z}\frac{R(z+\gamma')}{R\gamma'+(z+\gamma')(z+2R+\gamma+\gamma')}. \tag{49}$$

The steady state population may then be obtained from $h(z)$ as

$$\varrho_{11}^{SS}=\lim_{t\to\infty}\varrho_{11}(t)=\lim_{z\to 0} zh(z)=\frac{R}{3R+\gamma+\gamma'}. \tag{50}$$

To invert the Laplace transformation the poles of $h(z)$ are needed

$$\begin{aligned} &z_0=0,\\ &z_{1,2}=\tfrac{1}{2}[-(2R+\gamma+2\gamma')\\ &\qquad\pm\sqrt{(2R+\gamma+2\gamma')^2-4\gamma'(3R+\gamma+\gamma')}] \end{aligned} \tag{51}$$

and the transient evolution of $Q(T)$ is governed by the time scales set by those poles:

$$\begin{aligned} Q(T)=\frac{2\lambda R}{T(z_1-z_2)}\Bigg[&\frac{\gamma'+z_1}{z_1^3}(e^{z_1T}-1-z_1T)\\ &-\frac{\gamma'+z_2}{z_2^3}(e^{z_2T}-1-z_2T)\Bigg]. \end{aligned} \tag{52}$$

From this general result we directly obtain all the desired limiting cases. For small times T we find:

$$Q(T)=-\lambda T\left(\varrho_{11}^{SS}-\frac{RT}{3}\right). \tag{53}$$

$Q(T)$ decreases again linearly similar to the two-level result, but here the absolute value of the slope is smaller because the population is shared by three levels.

For large times T we find

$$Q(T)=Q_\infty-\frac{C}{T}, \tag{54}$$

where

$$Q_\infty=\frac{2\lambda R}{\gamma'}\frac{R-\gamma'}{(3R+\gamma+\gamma')^2}, \tag{55}$$

$$C=\frac{2\lambda R[R(2R+\gamma)+\gamma'(3R-\gamma')]}{\gamma'^2(3R+\gamma+\gamma')^3}. \tag{56}$$

Both parameters C and Q_∞ are positive for $R>\gamma'$ and $\gamma>\gamma'$. This means that Q is no longer decreasing monotonically, and there is a change of sign in $Q(T)$. The sub-Poissonian behaviour displayed for small times turns into super-Poissonian fluctuations for large times. The variance of the photon number increases dramatically when T is increased beyond γ'^{-1}, because the number of photons counted over such a time interval can differ widely from shot to shot. While during an uninterrupted fluorescence period up to 10^8 photons/s are counted, it is also possible that during the entire measurement interval not a single event is observed because the electron was shelved in the metastable state.

Bloch Equations. The Bloch equations for a three-level system driven by a coherent field in resonance with the strong transition $|1\rangle-|3\rangle$ are of the form:

$$\begin{aligned} &\dot{u}_{23}=-\chi v_{12},\\ &\dot{v}_{23}=-\chi u_{12},\\ &\dot{u}_{12}=\chi v_{23},\\ &\dot{v}_{12}=\chi u_{23},\\ &\dot{u}_{13}=0,\\ &\dot{v}_{13}=-2\chi w_{13},\\ &\dot{w}_{12}=\chi v_{13},\\ &\dot{w}_{13}=2\chi v_{13},\\ &\dot{w}_{23}=\chi v_{13}, \end{aligned} \tag{57a–i}$$

where u_{ij}, v_{ij}, and w_{ij} are related to the density matrix elements for $i,j=1,2,3$ by

$$\begin{aligned} &u_{ij}=\varrho_{ij}+\varrho_{ji},\\ &v_{ij}=-i(\varrho_{ij}-\varrho_{ji}),\\ &w_{ij}=\varrho_{ii}-\varrho_{jj}. \end{aligned} \tag{58a–c}$$

The Rabi frequency is defined as in (40), where μ is now the electric dipole moment of the strong transition. In (57a–i) the damping terms have not yet been included. Since we are primarily interested in the dynamics of the diagonal matrix element $\varrho_{11}(t)$ and since the equations (57a–e) decouple from the others, it is sufficient to consider only (57f–i). We assume that the electron does not decay out of the three levels:

$$\varrho_{11}+\varrho_{22}+\varrho_{33}=1, \tag{59a}$$

so that we are finally left with only three equations for the variables ϱ_{11}, ϱ_{33}, and v_{13}

$$\begin{aligned} &\dot{v}_{13}=-2\chi(\varrho_{11}-\varrho_{33})-\frac{v_{13}}{T_2},\\ &\dot{\varrho}_{11}=\chi v_{13}-(\gamma_{12}+\gamma_{13})\varrho_{11},\\ &\dot{\varrho}_{33}=-\chi v_{13}+\gamma_{13}\varrho_{11}+\gamma_{23}(1-\varrho_{11}-\varrho_{33}); \end{aligned} \tag{59b–d}$$

T_2 is the transverse relaxation time between the states $|1\rangle$ and $|3\rangle$. Again for simplification we choose:

$$\gamma=\gamma_{13},\quad \gamma'=\gamma_{12}=\gamma_{23}. \tag{60}$$

$h(z)$ is obtained again by solving (59b–d) through Laplace transformation:

$$h(z)=\frac{1}{z}\frac{2\chi^2(z+\gamma')}{2\chi^2\gamma'+(z+\gamma')[4\chi^2+(z+\gamma+\gamma')(z+T_2^{-1})]}. \tag{61}$$

The steady state population follows from $h(z)$ as in (50)

$$\varrho_{11}^{SS}=\frac{2\chi^2}{6\chi^2+(\gamma+\gamma')T_2^{-1}}. \tag{62}$$

The results from the rate equation approximation are also included here and can be reproduced from this more general treatment by taking the limit $T_2\to 0$, while keeping the transition rate $2\chi^2 T_2=R$ constant. Henceforth we shall assume that spontaneous emission is the only decay channel and take $1/T_2=\gamma/2$.

The nonvanishing eigenvalues follow from the solution of a cubic equation representing the denominator of $zh(z)$. In terms of these poles, $Q(T)$ can formally be written as:

$$\begin{aligned}Q(T)=Q_\infty-\frac{C}{T}+&\frac{4\lambda\chi^2}{(z_1-z_2)(z_1-z_3)(z_2-z_3)}\\ &\times\left[\frac{(z_2-z_3)(z_1+\gamma')}{z_1^2}\frac{e^{z_1T}}{z_1T}+\frac{(z_3-z_1)(z_2+\gamma')}{z_2^2}\right.\\ &\left.\times\frac{e^{z_2T}}{z_2T}+\frac{(z_1-z_2)(z_3+\gamma')}{z_3^2}\frac{e^{z_3T}}{z_3T}\right],\end{aligned} \tag{63}$$

where Q_∞ and C are already determined without explicit knowledge of the poles z_i:

$$Q_\infty=\frac{4\lambda\chi^2[2\chi^2-\gamma'(\frac{3}{2}\gamma+\gamma')]}{\gamma'[6\chi^2+\frac{1}{2}\gamma(\gamma+\gamma')]^2}, \tag{64}$$

$$C=\frac{4\lambda\chi^2[8\chi^4+\chi^2(\gamma^2+7\gamma\gamma'+10\gamma'^2)-\gamma'^2(\frac{7}{4}\gamma^2+\frac{5}{2}\gamma\gamma'+\gamma'^2)]}{\gamma'^2[6\chi^2+\frac{1}{2}\gamma(\gamma+\gamma')]^3}. \tag{65}$$

In the limit of long counting intervals $Q(T)$ approaches

$$Q(T)=Q_\infty-\frac{C}{T}, \tag{66}$$

while for short intervals we find the following dependence

$$Q(T)=-\lambda\varrho_{11}^{SS}T+\frac{\lambda\chi^2}{6}T^3. \tag{67}$$

The results simplify considerably when the spontaneous transition rates become substantially different $\gamma'\ll\gamma,\chi$, which is actually the physically most interesting case:

$$\begin{aligned}Q(T)=4\lambda\chi^2&\left\{\frac{2\chi^2}{\gamma'(6\chi^2+\gamma^2/2)^2}-\frac{\chi^2(8\chi^2+\gamma^2)}{\gamma'^2(6\chi^2+\gamma^2/2)^3}\right.\\ &\times\frac{1-e^{z_1T}}{T}+\frac{e^{-\frac{3}{4}\lambda T}}{T(4\chi^2+\gamma^2/2)^3}\\ &\times\left[\frac{9\gamma(\chi^2-\gamma^2/16)}{\zeta}\sin\zeta T\right.\\ &\left.\left.+\left(4\chi^2-\frac{7}{4}\gamma^2\right)\cos\zeta T\right]\right\},\end{aligned} \tag{68}$$

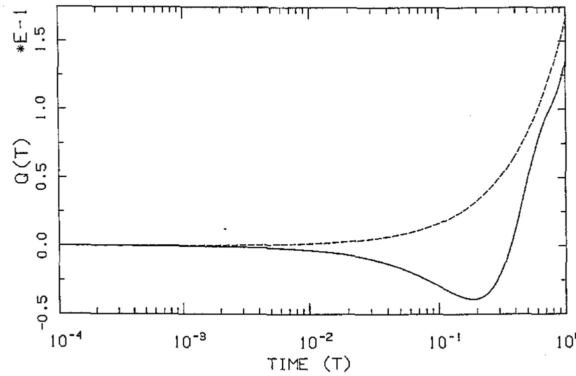

Fig. 2. The deviation from Poissonian statistics $Q(T)$ for time intervals T smaller than the lifetime of the metastable state γ'^{-1} for $\chi=5$ and $\gamma'=10^{-4}$ in units where $\gamma=\lambda=1$. The quantum mechanical result (solid curve) shows the typical sub-Poissonian behaviour. The swerve is a relic of the Rabi oscillations that are straightened out almost entirely by the sharp increase of the fluctuations. The dashed curve represents the classical result for $Q(T)$ (Sect. 2.3 of text)

where

$$z_1=-\frac{12\chi^2+\gamma^2}{8\chi^2+\gamma^2}\gamma', \tag{69}$$

$$\zeta=\sqrt{4\chi^2-\gamma^2/16}. \tag{70}$$

Very compact results are obtained when we further assume that the allowed transition is strongly saturated $\chi\gg\gamma,\gamma'$:

$$\begin{aligned}Q(T)=&\frac{2\lambda}{9\gamma'}\frac{e^{-\frac{3}{2}\gamma'T}-1+\frac{3}{2}\gamma'T}{\frac{3}{2}\gamma'T}\\ &+\frac{\lambda e^{-\frac{3}{4}\lambda T}}{4\chi^2T}\left(\cos 2\chi T+\frac{9\gamma}{8\chi}\sin 2\chi T\right).\end{aligned} \tag{71}$$

The asymptotic value $Q(T=\infty)$ in this case is:

$$Q(T=\infty)=\frac{2\lambda}{9\gamma'}, \tag{72}$$

which, in contrast to the two-level case, does not vanish for $\chi\to\infty$ but is quite large when quantum jumps, i.e. $\gamma\gg\gamma'$, occur. In this final form the Q-parameter displays the initial sub-Poisson statistics, quite similar to the results obtained for the two-level system; see Fig. 2. Then for longer times a sharp increase occurs, indicating a violently fluctuating signal. The coherent oscillations, which are quite visible for the two-level case, are barely to be seen here, since they are ironed out by the steep increase of the Q-function; see Fig. 3.

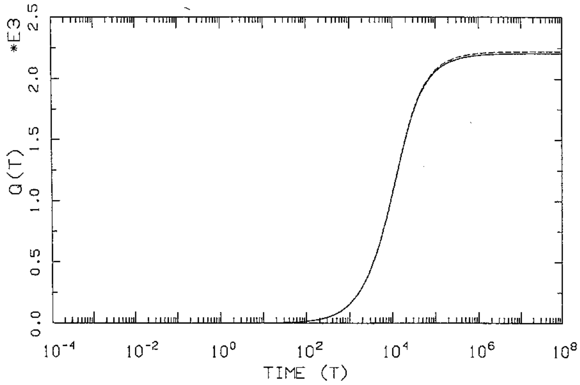

Fig. 3. $Q(T)$ as in Fig. 2 including also large time intervals. Notice that the Q-axis is compressed by a factor 10^4 compared to Fig. 2. The negative values for small time intervals are therefore invisible on this scale. Classical (dashed curve) and quantum mechanical (solid curve) results agree for time intervals of order γ'^{-1} and more

2.3. *The Random Telegraph Process*

Calculations for a classical process in Sect. 1.2 showed that $Q(T=0)$ vanishes and $Q(T)$ increases monotonically. By taking into account the fact that the fluorescence signal consists of bright periods with weak Poissonian noise alternating with periods of darkness, it seems natural to simulate such a process by a random telegraph signal in an entirely classical way. This allows us to specify $Q(T)$ for a classical process further and to compare the results with those of the quantum mechanical calculation. In this model the intensity of the fluorescence signal is assumed to realize only two different values $I=I_+$ or $I=0$ and a single trajectory jumps randomly between these two states. The probability per unit time for switching from I_+ to $I=0$ is taken to be the rate λ_1, while λ_2 characterizes the inverse transition. The time evolution of the conditional probabilities $P(I_+,t)$ and $P(0,t)$ is then governed by two elementary rate equations

$$\frac{\partial}{\partial t}P(I_+,t)=-\lambda_1 P(I_+,t)+\lambda_2 P(0,t), \tag{73}$$

$$\frac{\partial}{\partial t}P(0,t)=\lambda_1 P(I_+,t)-\lambda_2 P(0,t), \tag{74}$$

which preserve normalization:

$$P(I_+,t)+P(0,t)=1. \tag{75}$$

The system of differential equations (73) and (74) is easily solved for an arbitrary initial condition: for $t=0$ let $I=I_0$ where I_0 is either I_+ or 0:

$$P(I,t)=\left[\frac{\lambda_2}{\lambda_1+\lambda_2}\delta_{I_+,I}+\frac{\lambda_1}{\lambda_1+\lambda_2}\delta_{0,I}\right] \times[1-e^{-(\lambda_1+\lambda_2)t}]+\delta_{I,I_0}e^{-(\lambda_1+\lambda_2)t}. \tag{76}$$

The stationary solution is obtained for $t\to\infty$

$$P_s(I)=\frac{\lambda_2}{\lambda_1+\lambda_2}\delta_{I_+,I}+\frac{\lambda_1}{\lambda_1+\lambda_2}\delta_{0,I}. \tag{77}$$

Actually $P(I,t)=P(I,t/I_0,0)$ is the conditional probability for observing the value $I=I_+$ or 0 at a time t provided that the signal had a value I_0 at the earlier time $t=0$. The stationary and the conditional probability allow us to calculate the moments and correlation functions in explicit form. For the Q parameter we find for arbitrary T:

$$Q(T)=\eta I_+\frac{2\lambda_1}{(\lambda_1+\lambda_2)^2}\left(1+\frac{e^{-(\lambda_1+\lambda_2)T}-1}{(\lambda_1+\lambda_2)T}\right). \tag{78}$$

By expanding the exponential function for small times $T(\lambda_1+\lambda_2)\ll 1$ we find:

$$Q(T)=\eta I_+\frac{\lambda_1}{\lambda_1+\lambda_2}T\left[1-\frac{1}{3}(\lambda_1+\lambda_2)T\right], \tag{79}$$

a result which is again positive, in contrast to what is found from a quantum mechanical calculation.
In the opposite limit $T(\lambda_1+\lambda_2)\gg 1$ we obtain

$$Q(T)=Q_\infty\left(1-\frac{1}{(\lambda_1+\lambda_2)T}\right), \tag{80}$$

with the asymptotic value

$$Q_\infty=\eta I_+\frac{2\lambda_1}{(\lambda_1+\lambda_2)^2}. \tag{81}$$

In order to compare the classical results here with the previous quantum mechanical expressions, we have to relate the coefficients λ_1, λ_2, and I_+ to the spontaneous emission rates γ and γ'. For simplicity we consider here only the case of saturation. In this limit the average intensity during fluorescence is given by $I_+=\frac{1}{2}\gamma$, which should not be confused with the ensemble average of $I(t)$ which is $\langle I\rangle=\frac{1}{3}\gamma$. The transition probabilities λ_1 and λ_2 should be identified with the inverse of the mean duration of the bright ($\propto 2/\gamma'$) and the dark periods ($\propto 1/\gamma'$). With this replacement we find:

$$Q(T)=\frac{2\lambda}{9\gamma'}\left(1+\frac{e^{-\frac{3}{2}\gamma' T}-1}{\frac{3}{2}\gamma' T}\right). \tag{82}$$

This model was merely designed to simulate the intermittent behaviour of the quantum jump signal in an intuitive classical way. Therefore it is not surprising that it agrees quite well with the correct quantum mechanical description in the long time limit; see Fig. 3 and (71). However, since it is based on an entirely classical approach, the quantum features of the short time regime are not reproduced correctly by the somewhat naive assumption of a telegraph signal; see Fig. 2. The purpose of this undertaking was to confirm from a different point of view our previous interpretation of $Q(T)$, i.e. that the sharp increase of $Q(T)$ for long times is an indication of the onset of random intermittencies.

3. Conclusions

We now return to our initial question: Is it possible to characterize the statistical behaviour of fluorescence from a driven three-level system in a simple way? While the most complete description of the photon statistics is provided by the counting probability $W(n, T)$, it is in general a formidable task to calculate $W(n, T)$ in analytical form. In a perturbative way, however, this can be done in various limits [22] but it is still an involved treatment. In order to decide whether quantum jumps occur or not, and to determine their characteristic properties, it is entirely sufficient to use averages over the counting distribution. As demonstrated at the beginning, for calculating low order moments or variances, it is not necessary to be in possession of the entire probability distribution. By comparing the results for two-level systems with the corresponding ones for three levels, we have identified the typical signatures of quantum jumps, i.e. the sharp increase in the fluctuations occurring when the experimental collection time is of the order of the lifetime of the metastable state or longer. Whereas the long time behaviour of Q differentiates between the continuous and the intermittent fluorescence, the short time behaviour is quite analogous for two- and three-level systems. This indicates quite clearly that during the emission phase, where the electron undergoes rapid transitions along the allowed path, the presence of the metastable state is of little influence. In this sense, the Q-parameter, being only a qualitative measure for the random fluctuations of the field, nevertheless contains enough quantitative information to distinguish between continuous emission and the occurrence of quantum jumps.

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