Journal of Statistical Physics, Vol. 53, Nos. 1/2, 1988

Photon Emission As a Random Event

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Received April 1, 1988

The statistical properties of the random events that correspond to the emission or the detection of a photon spontaneously emitted by a single atom are discussed. This statistics constitutes a full and nontrivial solution of the stochastic theory of random events. The results are most explicit for an atom with two relevant states. The formalism is also generalized to atomic models with nondegenerate states or with more than one driven transition.

KEY WORDS: Random events; photon emission; waiting times.

1. INTRODUCTION

A substantial part of van Kampen's work is devoted to stochastic processes.[1] Most often the basis of the stochastic nature of phenomena is the molecular structure of matter. In that situation, fluctuations of a macroscopic quantity reflect the rapid variation of the underlying microscopic state of the system. In fact, a central goal of statistical mechanics is the construction of macroscopic equations of motion by elimination of the rapid variations within the class of microscopic states.[2] Another class of stochastic processes arises from the probabilistic nature of quantum mechanics, which allows only statistical predictions for the outcome of measurements. For this type of process, the fluctuations in measured quantities do not reflect variations in the state of the system.

In quantum optics, which deals with the interaction of photons with matter, stochastic processes play an important part. A prime example is spontaneous emission of photons. In the case of one or a few atoms or molecules, quantum fluctuations in the number of emitted photons or in the time lapse between successive emissions are crucial.

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0022-4715/88/1000-0417$06.00/0

In the present paper, I apply the stochastic theory of random events to the fluorescent emission of photons by a single atom. This problem has received much attention in the last decade.[3-6] I shall demonstrate that the treatment becomes considerably more transparent when the concept of the waiting-time distribution is applied. Furthermore, I shall show that a slightly more formal treatment allows a direct generalization to the case of an atom in which more than two states are coupled by the radiation field.

2. RANDOM EVENTS

First I briefly review some elements of the stochastic theory of random events, which may be represented as dots on the time axis.[1,7] In subsequent sections I shall apply this theory to the specific case of photon counts, in particular, the photon emissions by a single atom in a radiation field.

2.1. Distribution Function and Statistics

Basic to the description of random events are the distribution functions f_n. These are defined by requiring that[1,3]

$$f_n(t_1, t_2,..., t_n)\, dt_1 \cdots dt_n \tag{2.1}$$

is the probability for an event in the time interval $[t_1, t_1 + dt_1]$,..., and one in the time interval $[t_n, t_n + dt_n]$, for infinitesimal, nonoverlapping intervals. I assume that the events have a vanishing probability to coincide exactly, which implies that the functions f_n contain no delta functions of the type $\delta(t_i - t_j)$. Thus, the probability of having two events in an infinitesimal interval dt is at least of order $(dt)^2$. When N is the number of events in a time interval $[0, T]$, the integrals of the distribution functions are simply related to the factorial moments

$$s_n = \langle N!/(N-n)! \rangle \tag{2.2}$$

by the relations

$$s_n = \int_0^T dt_1\, dt_2 \cdots dt_n\, f_n(t_1, t_2,..., t_n) \tag{2.3}$$

When we introduce the probability p_N for precisely N events in the count interval $[0, T]$, the obvious relations

$$s_n = \sum_{N=n}^{\infty} p_N N!/(N-n)! \tag{2.4}$$

between s_n and p_N may be inverted to the sum rules

$$p_N = \sum_{n=0}^{\infty} (-1)^n s_{n+N}/(N!\, n!) \tag{2.5}$$

These relations are particularly convenient in cases where the evaluation of f_n and thereby of s_n is easier than the evaluation of p_N.

Another convenient tool in the study of the number statistics is the generating function[1]

$$G(\mu) \equiv \langle (1-\mu)^N \rangle = \sum_{N=0}^{\infty} p_N (1-\mu)^N \tag{2.6}$$

(In order to avoid confusion, I remark that usually $1-\mu$ is taken as the argument of the generating function.) An expression for the factorial moments s_n follows immediately after differentiating (2.6) n times with respect to μ, and putting μ equal to one. This leads to the alternative expansion

$$G(\mu) = \sum_{n=0}^{\infty} s_n (-\mu)^n/n! \tag{2.7}$$

Clearly, the full number statistics $\{p_N\}$ is contained in the set of factorial moments $\{s_n\}$, or alternatively in the analytic function G.

2.2. Statistics of Detected Events

In practical cases, the occurrence of an event is detected with a finite probability α, with $0 < \alpha < 1$. For instance, an emitted photon is usually detected by a photomultiplier with a probability α, which reflects the restricted aperture and the limited quantum efficiency of the multiplier. When we introduce in analogy to (2.1) the distribution functions $g_n(t_1, t_2, \ldots, t_n)$ for detected events, then we may write

$$g_n(t_1, t_2, \ldots, t_n) = \alpha^n f_n(t_1, t_2, \ldots, t_n) \tag{2.8}$$

and likewise the factorial moments r_n of these detected events are related to s_n by

$$r_n = \alpha^n s_n \tag{2.9}$$

These equations are subject to the assumption that the detection efficiency is not affected by a previous detection, so that the detector has no appreciable dead time. The generating function $F(\mu)$ specifying the statistics

of the number of detected events is then related to the generating function (2.7) by

$$F(\mu) = G(\alpha\mu) \tag{2.10}$$

An interesting feature of this relation is that the number statistics $\{p_N\}$ of the events or the number statistics $\{q_N\}$ of the detected events may be expressed as derivatives of q_0 with respect to α.[6] According to the definition of F and G, we obtain the identities

$$p_0 = G(1), \qquad q_0 = F(1) = G(\alpha) \tag{2.11}$$

Since p_N and q_N may be expressed as derivatives of G and F with respect to μ, we easily find the general expression

$$q_N = (-\alpha)^N (d/d\alpha)^N q_0/N! \tag{2.12}$$

Substituting $\alpha = 1$ in the right-hand side of (2.12) yields expressions for p_N. This demonstrates that knowledge of the probability q_0 for detecting no events in the count interval as a function of the efficiency α is suffcient to determine the complete number statistics $\{q_N\}$ and $\{p_N\}$.

2.3. Waiting-Time Distribution

A final quantity of practical interest is the probability distribution $w(t)$ for the time one has to wait for the first event to occur, after time zero. This waiting-time distribution is directly related to the probability $p_0(T)$ for having no events at all during $[0, T]$. Since during this interval there either was no event or at least one, we may write

$$\int_0^T dt\, w(t) + p_0(T) = 1 \tag{2.13}$$

After differentiating (2.13), we find[1]

$$w(T) = -\frac{d}{dT} p_0(T) \tag{2.14}$$

One notices that the waiting-time distribution $w(t)$ is an actual probability distribution that is normalized to unity when at least one event occurs with certainty for any positive time. A similar relation holds for the waiting-time distribution for the first detection.

I emphasize that the relations given in this section are generally valid, regardless of the nature of the events and of the mechanism causing their recurrence.

3. PHOTOELECTRON COUNTING

In a common experimental situation, photon counting is basically the counting of photoelectric pulses that are generated by a weak light field in a photomultiplier. The photopulses correspond to the release of a photoelectron and reflect the arrival of a photon. Ideally, the emission rate of the photoelectrons is proportional to the intensity $I(t)$. For convenience, we express the intensity in units of photoelectric counts per unit time.

In a classical picture, where the intensity $I(t)$ has a well-defined value at any instant, irrespective of the measurement, the obvious form for the distribution function $g_n(t_1, t_2, ..., t_n)$ of detected photoelectric counts is

$$g_n(t_1, t_2, ..., t_n) = \langle\langle I(t_1)\, I(t_2) \cdots I(t_n) \rangle\rangle \tag{3.1}$$

where the double brackets $\langle\langle \cdot \rangle\rangle$ denote an average over the stochastics of the fluctuating intensity. For a count interval $[0, T]$, the factorial moments are given by

$$r_n = \langle\langle P(T)^N \rangle\rangle \tag{3.2}$$

with

$$P(T) = \int_0^T I(t)\, dt \tag{3.3}$$

the integrated intensity. The probabilities q_N for observing N photocounts in the interval are then[8]

$$q_N = \langle\langle [P(T)]^N \exp[-P(T)] \rangle\rangle / N! \tag{3.4}$$

as we easily find after using the generating function

$$F(\mu) = \langle\langle \exp[-\mu P(T)] \rangle\rangle \tag{3.5}$$

One notices that the distribution $\{q_N\}$ has the nature of a stochastic average over Poisson distributions. When the intensity does not fluctuate, $P(T)$ is not a stochastic quantity, and the number of counts is described by the Poisson distribution with the mean value $P(T)$. Fluctuations cause $r_2 - r_1^2$ to be positive, since

$$r_2 - r_1^2 = \langle\langle P(T)^2 \rangle\rangle - \langle\langle P(T) \rangle\rangle^2 \equiv \Delta P^2 \tag{3.6}$$

which is the variance of P. This means that the variance of the number N of photocounts obeys the identity

$$\langle N^2 \rangle - \langle N \rangle^2 = \langle N \rangle + \Delta P^2 \tag{3.7}$$

so that the variance of the photocounts is larger than for a Poisson distribution with the same average number.

Similar expressions can be obtained when the quantum mechanical nature of the radiation field is accounted for. The main difference is that the order of the operators must be properly prescribed. For instance, Eq. (3.4) for the number statistics remains valid when we substitute the proper operator for the intensity, which is proportional to the photon-number operator $\sum b^+ b$, and when both time ordering (higher times to the left) and normal ordering (creation operators b^+ to the left) are imposed in each term after expansion of the exponential.[9] In that case, the arguments leading to (3.6) no longer hold, and the term $r_2 - r_1^2$ may be negative. Put in other words, one may say that the normally ordered variance of P is not necessarily positive. Hence, a photocount statistics with a variance that is smaller than in a Poisson distribution with the same average number cannot be simulated by a classical incident field where fluctuations are used to model the quantum fluctuations. In the next section we shall encounter several simple cases with such a sub-Poissonian photocount statistics.

4. FLUORESCENCE OF A TWO-STATE ATOM

In this section I shall describe the correlated statistics of the fluorescent photons emitted by an atom in a monochromatic radiation field. In fact, this problem has only recently been fully treated. Most treatments took as their starting point the quantum version of Eq. (3.5). I wish to demonstrate that an easier and more transparent description results when one uses the concept of the waiting-time distribution $w(t)$. Furthermore, this formalism allows a direct generalization to cases where more than one transition is driven by external fields, so that various spectral lines in the fluorescence spectrum appear.

4.1. Evolution Equation

Consider an atom in a monochromatic radiation field with a frequency ω that is near an atomic transition frequency ω_0. This transition couples the ground state $|g\rangle$ to an excited state $|e\rangle$, and radiative transitions to other states are assumed to be negligible. Moreover, the two coupled states are treated as nondegenerate. The equation of motion of the reduced density matrix ρ of the atom alone is, to an excellent approximation,[10]

$$d\rho/dt = -\Gamma\rho - (i/\hbar)[H(t), \rho] \tag{4.1}$$

where Γ is the effective relaxation operator for spontaneous decay, and where the Hamiltonian $H(t)$ is

$$H(t) = |e\rangle E_e\langle e| + |g\rangle E_g\langle g| - \mu E(t) \tag{4.2}$$

with E_e and E_g the atomic level energies, μ the atomic dispole operator, and E the electric component of the radiation field. The dipole has non-diagonal elements only, and the field is described as a monochromatic classical field.

I introduce a transformed density matrix $\sigma(t)$ for the atom by the definitions

$$\begin{aligned} \sigma_{ee} &= \rho_{ee}, & \sigma_{eg} &= \rho_{eg} e^{i\omega t} \\ \sigma_{gg} &= \rho_{gg}, & \sigma_{ge} &= \rho_{ge} e^{-i\omega t} \end{aligned} \tag{4.3}$$

and ignore as usual the rapidly oscillating terms in the resulting equation of motion for σ. The elements of σ obey the equations

$$\begin{aligned} \frac{d}{dt}\sigma_{ee} &= -A\sigma_{ee} + \frac{i}{2}\Omega(\sigma_{eg} - \sigma_{ge}) \\ \frac{d}{dt}\sigma_{gg} &= A\sigma_{ee} + \frac{i}{2}\Omega(\sigma_{ge} - \sigma_{eg}) \\ \frac{d}{dt}\sigma_{eg} &= -\left(\frac{1}{2}A - i\Delta\right)\sigma_{eg} + \frac{i}{2}\Omega(\sigma_{gg} - \sigma_{ee}) \\ \frac{d}{dt}\sigma_{ge} &= -\left(\frac{1}{2}A + i\Delta\right)\sigma_{ge} + \frac{i}{2}\Omega(\sigma_{ee} - \sigma_{gg}) \end{aligned} \tag{4.4}$$

where $\Delta = \omega - \omega_0$ is the detuning of the radiation frequency from resonance, and where Ω is the Rabi frequency, defined as the dipole matrix element times the field amplitude divided by $\hbar$. These equations have the same general form as the Bloch equations for the magnetic moment of a spin 1/2 under the influence of a static and a radiofrequency magnetic field, as in magnetic resonance experiments. For these reasons, Eqs. (4.4) are often termed the optical Bloch equations.

4.2. Photon Distribution Functions and Waiting Time

For a given solution $\sigma(t)$ of (4.4), the rate of spontaneous emission is equal to $A\sigma_{ee}(t)$, and this quantity plays the role of the fluorescence intensity $\langle\langle I(t)\rangle\rangle$. Obviously, spontaneous emission of a photon is accompanied by a transition of the atom from $|e\rangle$ to $|g\rangle$, so that immediately after an

emission the atom is with certainty in the ground state. When we start a count interval at an instant $t=0$ where emission just took place, or where the field was just switched on, we may write for the one-photon distribution function

$$f_1(t) = A\sigma_{ee}(t) \tag{4.5}$$

which is the probability density for another photon emission at time t_1. The initial condition is that $\sigma_{gg}(0)=0$, and the other matrix elements vanish at $t=0$. After this emission at time t, the atom has returned to the ground state, and the probability for a subsequent emission builds up in the same way as after the initial time zero. In this way, it is easy to understand that the n-fold distribution function f_n for subsequent photon emissions at time $t_1 < t_2 < \cdots < t_n$ is equal to[3,5,6]

$$f_n(t_1, t_2, \ldots, t_n) = f_1(t_n - t_{n-1}) f_1(t_{n-1} - t_{n-2}) \cdots f_1(t_2 - t_1) f_1(t_1) \tag{4.6}$$

It is now simple to derive the full photon statistics, at least in Laplace transform.[6] I denote as

$$\hat{f}_1(v) = \int_0^\infty dt\, e^{-vt} f_1(t) \tag{4.7}$$

the Laplace transform of f_1, and I similarly introduce the Laplace transforms of the factorial moments $s_n(t)$, the number probabilities $p_N(t)$, and the generating function $G(\mu, T)$. After using (4.6), (4.7), and (2.3), we obtain

$$\hat{s}_n(v) = \frac{n!}{v} [\hat{f}_1(v)]^n \tag{4.8}$$

This gives for the Laplace transform of the generating function (2.7)

$$\hat{G}(\mu, v) = \frac{1}{v} \frac{1}{1 + \mu \hat{f}_1(v)} \tag{4.9}$$

which gives, after use of (2.6), for the number probabilities

$$\hat{p}_N(v) = \frac{1}{v} \frac{[\hat{f}_1(v)]^N}{[1 + \hat{f}_1(v)]^{N+1}} \tag{4.10}$$

For completeness we also give the waiting-time distribution. If we take the Laplace transform of (2.14), and if we use that $p_0(0)=1$, $p_0(\infty)=0$, we find

$$\hat{w}(v) = \frac{\hat{f}_1(v)}{1 + \hat{f}_1(v)} \tag{4.11}$$

4.3. Operator Expressions

It is illuminating to give a formal operator expression for f_1 and w. This will also allow us to generalize the results of this section. First we write the optical Bloch equations (4.4) as an operator equation

$$d\sigma/dt = L\sigma \tag{4.12}$$

for the density matrix σ. Next we separate the evolution operator L according to

$$L = L_0 + S \tag{4.13}$$

where the action of L_0 is defined by the right-hand sides of (4.4), with the first term of the right-hand side of the second equation omitted. This omitted term, which defines the operator S, describes the gain in the ground-state population due to spontaneous emission. If one introduces the projectors

$$P_e = |e\rangle\langle e|, \qquad P_g = |g\rangle\langle g| \tag{4.14}$$

then S is expressed by its action on a density matrix σ

$$S\sigma = AP_g \operatorname{Tr} \sigma P_e \tag{4.15}$$

In the quantum electrodynamic description of spontaneous emission, this operator gives the effect on the atom at instants where the photon number of the fluorescence modes is increased by one. Hence the operator S may be properly called the spontaneous emission operator. The terms proportional to the spontaneous decay rate A contributing to L_0 describe the decay of the atomic density matrix pertaining to a fixed number of fluorescent photons.[4]

From the expression (4.5) for $f_1(t)$ we find immediately the formal expression

$$f_1(t) = \operatorname{Tr} S e^{Lt} P_g \tag{4.16}$$

since in Eq. (4.5) the initial value for σ was taken to be P_g. This form (4.16) reveals the physical significance of $f_1(t)$ as the probability density for a photon emission at time t, with the condition that the atom was in the ground state at time zero. A similar form may be found for the waiting-time distribution $w(t)$. From the mere fact that L_0 describes the evolution of σ in a period where no spontaneous emission occurs, one may anticipate the result

$$w(t) = \operatorname{Tr} S e^{L_0 t} P_g \tag{4.17}$$

This is confirmed when we take the Laplace transform of (4.16), and when we use the formal expansion in powers of S

$$\frac{1}{v-L}=\frac{1}{v-L_0}+\frac{1}{v-L_0}S\frac{1}{v-L_0}+\cdots \tag{4.18}$$

From the explicit action of S as defined in Eq. (4.15), we observe that the Laplace transforms of (4.16) and (4.17) are related by the geometrical series

$$\hat{f}_1(v)=\hat{w}(v)+[\hat{w}(v)]^2+[\hat{w}(v)]^3+\cdots \tag{4.19}$$

This result is equivalent to (4.11), which confirms that (4.17) is indeed a correct expression for $w(t)$. The expansion (4.19) simply indicates that a photon emitted at a selected instant of time t may be the first, or the second, or the third,..., after the initial time zero. This relation (4.19) between the function f_1 and the waiting-time distribution w holds generally, provided that the state of the atom directly after a spontaneous emission is uniquely defined. This is the case when the lower state $|g\rangle$ is nondegenerate.[12]

In the present case without external perturbations such as collisions or light fluctuations, the evolution of w is easier to calculate than that of f_1. The reason is that the evolution operator L_0 appearing in (4.17) transforms a projector P_g into another operator of the form $|\psi(t)\rangle\langle\psi(t)|$. Therefore, only the evolution of state vectors is needed for the evolution of w, whereas the operator L, which specifies f_1, operates essentially on the Liouville space of density matrices. In more general cases this is also true for w.

Now that we have definitely identified L_0 as the evolution operator between spontaneous emissions and S as the operator describing spontaneous emission, it should be obvious that the density matrix

$$\sigma(t)=e^{Lt}P_g \tag{4.20}$$

may be separated as

$$\sigma(t)=\sum_{N=0}^{\infty}\sigma_N(t) \tag{4.21}$$

where σ_N is the contribution to σ from the evolution history in which precisely N photons have been emitted. Therefore σ_N is the Nth order of (4.20) in the expansion in powers of S. In Laplace transform this gives

$$\hat{\sigma}_N(v)=\frac{1}{v-L_0}\left(S\frac{1}{v-L_0}\right)^N P_g \tag{4.22}$$

Obviously, the trace of (4.20) is the Laplace transform of $p_N(t)$, and we find

$$\hat{p}_N(v) = \hat{p}_0(v)[(\hat{w}(v)]^N \tag{4.23}$$

with

$$\hat{p}_0(v) = \mathrm{Tr}\,\frac{1}{v - L_0} P_g \tag{4.24}$$

Equation (4.23) reflects the fact that in the case of N photon emissions in a count interval $[0, T]$, we have to wait N times for the first next photon, and in the remaining time after the Nth emission, no photon emission should occur.

4.4. Explicit Expressions

It is not difficult to obtain explicit expressions for the Laplace transform of f_1, after taking the Laplace transform of Eqs. (4.4) with the initial condition $\sigma(0) = P_g$. Then $\hat{f}_1(v)$ is equal to the Laplace transform of $A\sigma_{ee}$. The result is[11]

$$\hat{f}_1(v) = \frac{1}{v} \frac{\frac{1}{2}A\Omega^2(v + \frac{1}{2}A)}{(v + A)[(v + \frac{1}{2}A)^2 + \Delta^2] + \Omega^2(v + \frac{1}{2}A)} \tag{4.25}$$

Likewise, we obtain an expression for $\hat{w}(v)$ if we omit the first term on the right-hand side of the second equation (4.4), which is equivalent to omitting the operator S. The result is[13]

$$\hat{w}(v) = \frac{\frac{1}{2}A\Omega^2(v + \frac{1}{2}A)}{v(v + A)[(v + \frac{1}{2}A)^2 + \Delta^2] + \Omega^2(v + \frac{1}{2}A)^2} \tag{4.26}$$

One readily checks that Eq. (4.11) holds.

Figures 1 and 2 present the time behavior of f_1 and w for resonant excitation ($\Delta = 0$) and for two values of Ω/A. These plots illustrate that both f_1 and w vanish for time zero, which reflects that after an emission at time zero the atom needs a finite recovery time before a subsequent emission can occur. The photons seem to repel each other in time and have a reduced probability to follow each other within a time lapse that is smaller than both A^{-1} and Ω^{-1}. This is the phenomenon of photon antibunching.[14] For a Rabi frequency Ω that is larger than $A/2$, both f_1 and w display oscillations at this Rabi frequency. For smaller values of Ω, the atom is effectively overdamped. For larger times $t \gg A^{-1}$, f_1 approaches the value of the steady-state intensity, since the memory of the initial condition has died out. The waiting-time distribution w is normalized to unity, and it approaches zero for $t \gg A^{-1}$.

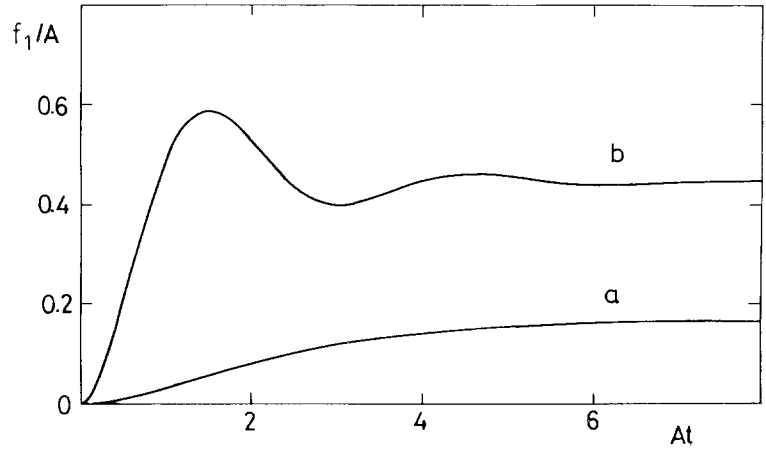

Fig. 1. Behavior of the one-photon distribution f_1 for excitation at resonance. (a) $\Omega = \frac{1}{2}A$; (b) $\Omega = \frac{1}{2}A\sqrt{17}$.

The antibunching property of the emitted photons can easily lead to sub-Poissonian statistics,[3] in the sense mentioned in the previous section. As is obvious from Eq. (4.6) for the n-fold distribution function, the antibunching does not arise merely at time zero but it gives a zero probability density when two emission times t_i and t_{i+1} approach each other. Hence this remains true when the atom has resided many lifetimes in the excited state, so that the density matrix σ has reached its steady state. This should remind us of the essential statistical significance of the density matrix. The observation of an emitted photon at time t_i constitutes a quan-

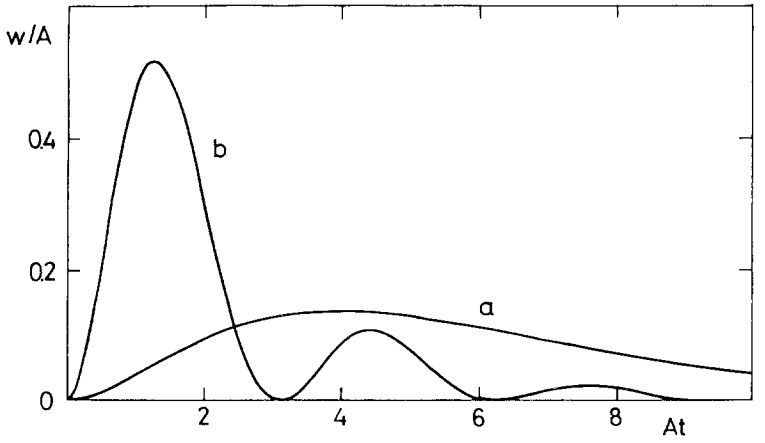

Fig. 2. Behavior of the waiting-time distribution w for the same situations as in Fig. 1.

tum mechanical measurement, which has the effect of projecting the atom in its ground state, even though the form of the density matrix tells us that the atom is in a superposition state.

4.5. Detected Photons

Suppose that each emitted photon has a probability α to be detected by a photomultiplier. The arguments of Section 2.2 apply directly, and the n-fold distribution function g_n of detected photons is simply α^n times the function f_n specified in (4.6). Hence g_n factorizes into a product of n functions g_1, with

$$g_1(t) = \alpha \operatorname{Tr} S e^{Lt} P_g \tag{4.27}$$

This determines completely the correlated statistics of photon detections, and in full analogy to Section 4.2, we find that the number statistics $\{q_N\}$ of the detected photons and the waiting-time distribution u for the first detection are given in Laplace transform by the right-hand side of (4.10) and (4.11) when we replace $\hat{f}_1$ by

$$\hat{g}_1(v) = \alpha \hat{f}_1(v) \tag{4.28}$$

Hence, by using (4.28) and the inverse equation of (4.11), we obtain the relation between the waiting-time distribution $u(t)$ for the first detected photon and the waiting-time distribution $w(t)$ for the first emitted photon, both in Laplace transformation. The result is

$$\hat{u}(v) = \frac{\alpha \hat{w}(v)}{1 - (1-\alpha)\,\hat{w}(v)} \tag{4.29}$$

The operator notation for $u(t)$, in analogy to (4.17), is

$$u(t) = \alpha \operatorname{Tr} S \exp\{[L_0 + (1-\alpha)S]t\}\, P_g \tag{4.30}$$

5. FLUORESCENCE OF MORE COMPLEX ATOMIC MODELS

The formalism of the previous section is by no means restricted to the simple two-state model of an atom. In this section I shall briefly discuss several more complicated level schemes.

5.1. Weak Coupling to Another Lower State

We consider the level scheme sketched in Fig. 3, where the excited state $|e\rangle$ can decay not only to the ground state $|g\rangle$, but also to another,

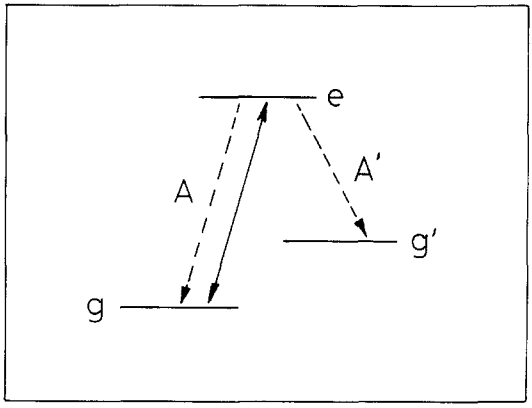

Fig. 3. Level scheme with a weakly coupled third state. Solid lines indicate stimulated transitions, broken lines indicate spontaneous decay.

lower state $|g'\rangle$. The spontaneous emission rate from $|e\rangle$ to $|g'\rangle$ is called A'. The Bloch equations for this modified level scheme follow from Eqs. (4.4) if we substitute $A + A'$ for A in the equations of motion for σ_{ee}, σ_{eg}, and σ_{ge}. The equation for σ_{gg} [the second equation in (4.4)] remains unchanged. Alternatively, we can say that the evolution equation (4.12) still holds with the separation (4.13) for the evolution operator L, where S still has the same form (4.15), but now we replace A by $A + A'$ in L_0. Then the formal results (4.6), (4.16), and (4.17) remain valid with this modified operator L_0 and L, and also the results (4.23) and (4.24) for the photon number statistics hold. The explicit expressions (4.25) and (4.26) for $\hat{f}_i$ and $\hat{w}$ are altered, although their relation (4.11) still holds. We now find

$$\hat{w}(v) = \frac{\frac{1}{2}A\Omega^2(v + \frac{1}{2}A + \frac{1}{2}A')}{v(v + A + A')[(v + \frac{1}{2}A + \frac{1}{2}A')^2 + A^2] + \Omega^2(v + \frac{1}{2}A + \frac{1}{2}A')^2} \tag{5.1}$$

From the scheme in Fig. 3 it is clear that the atom will always end up in the state $|g'\rangle$. The solution of the modified Bloch equations decays to zero, and eventually the population $\sigma_{g'g'}$ of the third state should go to one, whatever the normalized initial condition was. This population is governed by the evolution equation

$$\frac{d}{dt}\sigma_{g'g'} = A'\sigma_{ee} \tag{5.2}$$

Suppose now that the transition from $|e\rangle$ to $|g'\rangle$ is very weak, so that

$$A' \ll A \tag{5.3}$$

This is the case when this transition is dipole forbidden. Then the fluorescence will eventually stop, but only after the emission of many fluorescent photons.

We assume that at time zero the atom enters the field in its ground state $|g\rangle$. If we look at the fluorescent emission on the slow time scale of the weak transition, we see a continuous flow of light with a time-dependent intensity. In order to decide what this time-dependent intensity will look like, we first consider the photon distribution function $f_1(t)$. In view of the large difference in time scale for the rapid radiative transitions between $|e\rangle$ and $|g\rangle$, and the slow decay from $|e\rangle$ to $|g'\rangle$, the behavior of f_1 becomes particularly simple. For short times $t \ll A'^{-1}$, there has been no time for spontaneous decay to the state $|g'\rangle$, and $f_1(t)$ must be the same as in the case of a two-state atom, which has (4.11) as its Laplace transform. This function approaches the limiting value

$$\lim_{t \to \infty} f_1(t) = \lim_{v \to 0} v\hat{f}_1(v) = Ap_e \tag{5.4}$$

with

$$p_e = \frac{\Omega^2/4}{A^2 + A^2/4 + \Omega^2/2} \tag{5.5}$$

the steady-state fraction of excited atoms in the two-state model. By summing the evolution equations for σ_{ee} and σ_{gg}, we find the equation

$$\frac{d}{dt} n_s = -A'\sigma_{ee} \tag{5.6}$$

with

$$n_s = \sigma_{ee} + \sigma_{gg} \tag{5.7}$$

the total population of the strongly coupled states. For times $t \gg A^{-1}$, the strong coupling forces σ_{gg} and σ_{ee} to their steady-state ratio, so that we may write

$$\sigma_{ee} = p_e n_s \tag{5.8}$$

Substituting (5.8) in (5.6) leads to a simple exponential decay of n_s at the rate $A'p_e$. In fact, this is a simple case of elimination of rapid variables, which is a topic to which van Kampen has much contributed.[2] The long-time behavior for $t \gg A^{-1}$ of f_1 is therefore

$$f_1(t) = Ap_e e^{-A'p_e t} \tag{5.9}$$

Hence the one-photon distribution function has a slow exponential decay, and it may be tempting to take this as proof that the fluorescent

intensity at the slow time scale also displays this exponential decrease to zero. However, we should recall that $f_1(t)$ is a probability density, which can only be safely compared with experiment if we consider either an ensemble of many atoms or, equivalently, many runs with a single atom.

In order to decide what a typical single measurement of the time-dependent fluorescent intensity will look like, it is instructive to look at the waiting-time distribution, which is determined by (5.1). The short-time behavior of $w(t)$ corresponds to large values of v, where the contribution of A' is very small. Hence the small-time behavior of w differs negligibly from the two-state case. However, the time integral of w is equal to the limit of $\hat{w}$ for $v \to 0$, and we find

$$\int_0^\infty dt\, w(t) = \frac{A}{A + A'} \tag{5.10}$$

which is slightly smaller than unity. This means that at each photon emission there is a small probability $A'/(A + A')$ that this photon has been the last one and that the fluorescence has stopped. On the other hand, when any photon is emitted, the distribution of the time lapses between this emission and the next one is the same as after the very first photon. This implies that the time-dependent fluorescence has an intensity that has the same value Ap_e as in the two-state case, until it suddenly stops at a random instant. This random instant has the rate $A'p_e$. Only when we observe a large number of atoms will the total fluorescence exhibit the exponential decay at the rate $A'p_e$. In this case, the observation of the fluorescence intensity constitutes a measurement that decides whether or not the atom is still in the strongly coupled states. The distinction between $f_1(t)$ and the time-dependent intensity in a single run is illustrated in Fig. 4. This random

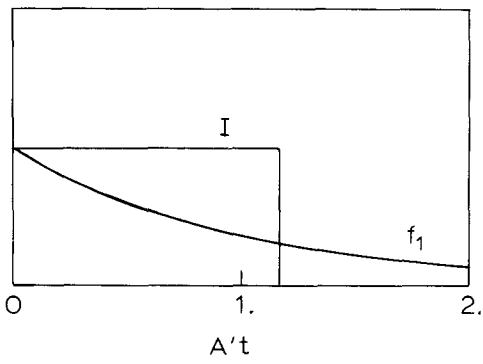

Fig. 4. Plot of one-photon distribution $f_1(t)$ and a typical single run of the time-dependent intensity of fluorescence for the three-state system.

termination of fluorescence is in fact a simple example of a quantum jump.[15]

The same type of reasoning can also be applied in the case that the weak transition between $|e\rangle$ and $|g'\rangle$ is also driven by a radiation field.[16] Then the stream of fluorescent photons can be switched on again by the absorptive excitation on the weak transition. The result is a random switching on and off of the fluorescent intensity on a macroscopic time scale. This phenomenon has received much attention theoretically.[17] It has also been observed experimentally for a single atomic ion in a radio-frequency trap.[18]

5.2. Degenerate States

In most practical cases, the atomic levels coupled by a radiation field are degenerate, and then the Zeeman substates must be accounted for. For linearly polarized radiation, we get transitions as sketched in Fig. 5. This will complicate the explicit form of the Bloch equations (4.4).[19] When we indicate with the indices g and e the multiplets of states in the two levels, the quantities σ_{ee}, σ_{eg}, etc., are submatrices. Nevertheless, the rough structure of (4.4) is retained. The terms with Ω should now contain a multiplication with a dipole submatrix. In particular, the formal separation (4.13) of the evolution operator remains useful. The spontaneous emission operator S maps the excited-state submatrix σ_{ee} onto the ground-state submatrix σ_{gg} and gives the gain in σ_{gg} due to spontaneous emission of an atom in an excited state described by the submatrix σ_{ee}.

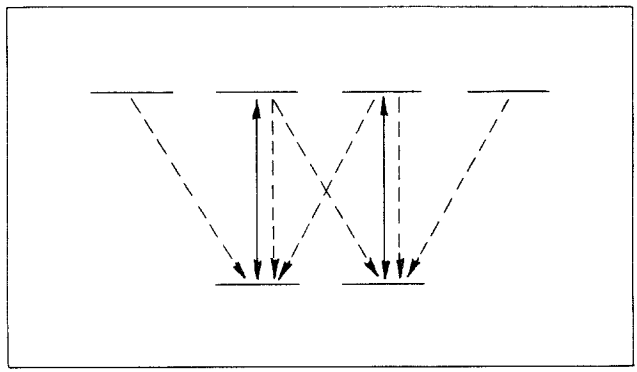

Fig. 5. Indication of stimulated (solid lines) and spontaneous (broken lines) transitions in a system with two degenerate levels. The indicated states represent Zeeman sublevels. Stimulated transitions with polarized light represent more restrictive selection rules than do spontaneous transitions.

The factorization (4.6) of the distribution functions no longer holds when the ground level is degenerate. Nevertheless, many results of Section 4 can be easily generalized to this case, and I mention some examples.

The distribution function f_n takes the general operator form (for $t_1 < t_2 < \cdots < t_n$)

$$f_n(t_1, t_2, \ldots, t_n) = \mathrm{Tr}\, S\, e^{L(t_n - t_{n-1})} S \cdots e^{L(t_2 - t_1)} S e^{Lt_1} \sigma_0 \tag{5.11}$$

where σ_0 is the initial state at time zero. The waiting-time distribution for the first photon after time zero is still given by (4.17) when we replace P_g by σ_0. The photon number probabilities are given by

$$\hat{p}_N(v) = \mathrm{Tr} \frac{1}{v - L_0} \left(S \frac{1}{v - L_0} \right)^N \sigma_0 \tag{5.12}$$

but the factorization (4.23) is not always valid. A similar relation holds for the factorial moments

$$\hat{s}_n(v) = n!\, \mathrm{Tr} \frac{1}{v - L} \left(S \frac{1}{v - L} \right)^n \sigma_0 \tag{5.13}$$

These relations may serve as a starting point for explicit calculations in specific situations. At the same time, they are useful in themselves to illustrate the interdependence of the various quantities concerning the correlated statistics of photoemissions. Similar results for the statistics of detected photons with a limited efficiency α can be directly found with the methods indicated in Section 4.5. The formalism also serves to answer more complex questions, which involve not only the number or the instant of photon emissions, but also the state of the atom. For instance, the density matrix

$$\hat{\sigma}_N = \frac{1}{v - L_0} \left(S \frac{1}{v - L_0} \right)^N \sigma_0 \tag{5.14}$$

may serve to calculate the probability that precisely N photons have been emitted and the atom at time t is in a certain state. Thus, this quantity is a density matrix normalized to a number probability.

5.3. Various Transitions

I now consider an atom with more than two levels that are coupled by radiative transitions. An example is indicated in Fig. 6. Several monochromatic radiation fields drive various transitions. By making a transformation similar to (4.3), the transformed density matrix σ obeys an

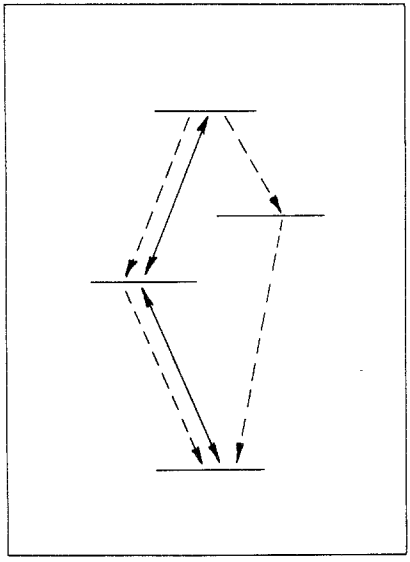

Fig. 6. Example of a system with several driven transitions (solid lines). Fluorescent emission can occur on four transitions (broken lines) indicating spontaneous decay.

evolution equation like (4.12) with a time-independent evolution operator. This is possible in many cases of practical interest. (The condition is that each pair of states is coupled by not more than one frequency, and in the scheme of levels connected by driven transitions there are no closed loops.) In the resulting evolution equations for the diagonal terms, spontaneous emission from higher levels give rise to gain terms, just like the first term in the second equation (4.4). These terms can be denoted by operators $S(i \to j)$, with

$$S(i \to j) = A_{ij} P_j \operatorname{Tr} \sigma P_i \tag{5.15}$$

Here i and j indicate states, A_{ij} is the spontaneous decay rate from state $|i\rangle$ to the lower lying state $|j\rangle$, and P_i and P_j are projectors on these states. Each pair $i \to j$ of states corresponding to a spontaneous transition gives rise to a line in the fluorescence spectrum. The transition frequencies are supposed to differ by a value large compared with radiative transition rates, so that the spectral lines can be resolved within a time $\Delta\omega^{-1}$ that is negligible compared with the radiative lifetimes. Then the different photons can be assigned to a transition in an essentially instantaneous way and we can forget about the inherent time uncertainty.

We label the transition $i \to j$ by the index β and we write the evolution equation for the transformed density matrix σ in the form

$$\frac{d}{dt}\sigma = L\sigma \tag{5.16}$$

with

$$L = L_0 + \sum_{\beta} S_\beta \tag{5.17}$$

where L_0 contains no gain terms, due to spontaneous emission. Then we can describe in the same spirit as in Section 4 the statistical properties of the emitted photons of the various transitions. For instance, when $f_n(t_1\beta_{2x}; t_2\beta_2;...; t_n\beta_n)$ is the probability density for the emission of a photon on the transition β_1 at t_1,..., and on the transition β_n at t_n, we find the formal result for $t_1 < t_2 < \cdots < t_n$,

$$\begin{aligned} &f_n(t_1\beta_1; t_2\beta_2;...; t_n\beta_n) \\ &\quad = \mathrm{Tr}\, S_{\beta_n} e^{L(t_n - t_{n-1})} \cdots S_{\beta_2} e^{L(t_2 - t_1)} S_{\beta_1} e^{Lt_1} P_g \end{aligned} \tag{5.18}$$

where it is assumed that the atom entered the fields at time zero in the ground state. If we replace L by L_0 in (5.18), the resulting expression denotes the probability density for a β_1 photon at time t_1, a β_2 photon at time t_2,..., a β_n photon at time t_n, and no other photons between time zero and t_n. As a result of the specific form (5.15) for the operators S_β, the expression (5.18) factorizes into functions of the type

$$f_1(\beta'\beta, t) = \mathrm{Tr}\, S_{\beta'} e^{Lt} P_{j_\beta} \tag{5.19}$$

which is the probability density for the emission of a photon in the transition β' at time t after an earlier emission of a photon on the transition β at time zero. This condition ensures that the evolution on time zero starts at the lower state $|j_\beta\rangle$ of the transition β. Likewise, we obtain the expression

$$w(\beta'\beta, t) = \mathrm{Tr}\, S_{\beta'} e^{L_0 t} P_{j_\beta} \tag{5.20}$$

for the probability density that after an emission of a β photon at time zero one has to wait a time t for the next photon and that this next photon occurs on the transition β'. After these examples, the reader will have no difficulty in giving analogous expressions for probability densities of any type (photon emissions at given instants on a specified transition, or, irrespective of the transition, with or without the condition of no photon emissions in between).

The two-label functions $f_1(\beta'\beta)$ and $w(\beta'\beta)$ form matrices with a dimension equal to the number of spontaneous transitions. The relation (4.11) may now be generalized into a similar relation between the Laplace transform of these matrices. Denote the matrices $F(t)$ and $W(t)$ by the matrix elements

$$F_{\beta'\beta}(t) = f_1(\beta'\beta, t), \qquad W_{\beta'\beta}(t) = w(\beta'\beta, t) \tag{5.21}$$

Then one proves after generalizing the arguments of Section 4

$$\hat{F}(v) = \hat{W}(v)/[1 - \hat{W}(v)] \tag{5.22}$$

Formal expressions for the mixed number statistics $p(\{n_\beta\})$ can easily be found from the general results.

6. CONCLUSIONS

I have demonstrated that the statistical properties of spontaneous photon emission by a single atom can be accurately and systematically described by the stochastic theory of random events. The distribution functions can be evaluated explicitly in practical cases, which gives rise to closed analytical expressions for the number statistics. At the same time the close connection between these distribution functions and the waiting-time distribution is recognized as resulting from the fact that the atomic state after a photon emission is well determined. From the point of view of quantum mechanical measurement theory, the detection of a photon may be regarded as a measurement of the atomic energy level, where the vacuum of the radiation field is part of the measurement device. The observation effectively interrupts the coherent oscillation of the atomic dipole, which can only be understood in terms of a superposition state.

The formalism developed for a two-state atomic model allows generalization to situations where the coupled levels are degenerate or when more than two states are coupled by the presence of several monochromatic fields. Each of these cases is highly relevant in actual experimental situations.

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