Photon Statistics and Quantum Jumps: The Picture of the Dressed Atom Radiative Cascade

SERGE REYNAUD, JEAN DALIBARD, AND CLAUDE COHEN-TANNOUDJI

Abstract—The statistics of spontaneous photons emission times in single atom resonance fluorescence are investigated through the radiative cascade of the dressed atom. We calculate the delay function which gives the distribution of the delays between two successive emissions for a coherent as well as an incoherent laser excitation. For a two-level atom, we review in this way various signals concerning the fluorescence intensity (average value, photon counting, fluctuations spectrum, etc.). For a three-level atom, this approach is applied to the analysis of the recently observed phenomenon of intermittent fluorescence and quantum jumps.

I. INTRODUCTION

RESONANCE fluorescence, i.e., resonant absorption and emission of photons by an atom, has been studied for a long time [1]-[3]. Novel phenomena occur when the atomic transition is saturated with an intense, quasi-resonant laser beam: triplet structure of the fluorescence spectrum [4]-[6], temporal antibunching of the fluorescence photons [6]-[7], sub-Poissonian statistics [8], etc. The possibility of trapping a single ion has recently opened the way to new developments. For example, the effect of "intermittent fluorescence" has been observed [9]-[11], giving for the first time a direct evidence at the macroscopic level for the "quantum jumps" of an atom reaching or leaving one of its eigenstates.

The statistical properties of the fluorescence light have been the subject of several theoretical works. In most of these works, the atomic dynamics are described by "optical Bloch equations" [12]-[13]. In spite of its remarkable efficiency for the computation of most signals, this method does not always provide a simple understanding of the statistical properties of the fluorescence light. Since the laser field is treated as a classical field, absorption and emission do not appear explicitly as elementary processes in the equations. A solution to this problem is to treat the laser field as a quantum field. The dynamical equation of the compound system-atom "dressed" by the laser photons-thus directly describes the elementary absorption and emission processes. Statistical properties of the fluorescence light are thus simply understood by considering the "dressed atom radiative cascade," i.e., the dressed atom cascading downwards its energy diagram while emitting fluorescence photons [14]-[16].

Manuscript received October 5, 1987; revised December 5, 1987. The authors are with the Ecole Normale Supérieure et Collège de France, 24, rue Lhomond—F 75231 Paris Cedex 05, France.

The purpose of this paper is to analyze in detail such a picture, and to show that it can provide a quantitative understanding of the statistical properties of the spontaneous emission times of a single atom interacting with a resonant laser beam. Starting from the master equation describing the evolution of the dressed atom density matrix, we will show that several photon statistics signals can be calculated in terms of a single function, the "delay function," which is the probability distribution of the delay between two successive spontaneous emission times. Such a function already has been used for analyzing various effects such as resonance fluorescence [16] or quantum jumps [17]-[19]. It has close connections with the socalled "exclusive" two-time probability density of the general photodetection theory [39], [40]. In this paper, we review various types of signals which can be expressed in terms of this function and we extend previous calculations in order to include the case of an incoherent excitation.

We consider first the simplest case of a two-level atom and we calculate the delay function for a coherent (Section II) and an incoherent excitation (Section III). We then show (Section IV) how various photon statistics signals are related to this function. Finally, we extend the previous analysis to three-level atoms (Section V) and discuss intermittent fluorescence and quantum jumps.

II. COHERENT EXCITATION OF A TWO-LEVEL ATOM

The states of the compound system-atom + laser photons (the "dressed" atom)-are labeled by two quantum numbers: an atomic one (g for the ground state and e for the excited state) and the number n of laser photons. For a quasi-resonant excitation, the two states $|g, n\rangle$ and |e,(n-1) form a nearly degenerate manifold (that we denote \mathcal{E}_n) since their splitting is just the detuning $\delta = \omega_L$ $-\omega_A$ between the laser frequency ω_L and the atomic one ω_A . These two levels are coupled by absorption and stimulated emission processes: the atom can go from g to e(respectively, from e to g) while absorbing (respectively, emitting) a laser photon. On the other hand, spontaneous emission is associated with the coupling of the atom with the vacuum field reservoir and corresponds to transitions between two adjacent manifolds: the atom goes from e to g while emitting a fluorescence photon and the number nof laser photons is conserved.

Fluorescence thus appears as a succession of elemen-

0018-9197/88/0700-1395\$01.00 © 1988 IEEE

IEEE Log Number 8821245.

1396

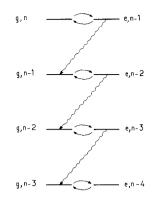


Fig. 1. Energy levels of the system atom + laser photons. The horizontal arrows describe reversible absorption and stimulated emission processes. The wavy arrows describe irreversible spontaneous emission processes.

tary processes (see Fig. 1): starting, for example, from $|g, n\rangle$, the dressed atom can go to $|e, n - 1\rangle$ (absorption process), then jump down to $|g, n - 1\rangle$ (spontaneous emission), etc. This phenomenon is called the "dressed atom radiative cascade" because of the analogy with atomic or nuclear radiative cascades.

This radiative cascade can be described quantitatively by a "master equation" derived from general relaxation theory. Such an equation can be written as follows [12], [20]:

$$\frac{d}{dt}\sigma = -\frac{i}{\hbar}[H,\sigma]$$
$$-\frac{\Gamma}{2}(S^{+}S^{-}\sigma + \sigma S^{+}S^{-})$$
$$+\Gamma S^{-}\sigma S^{+} \qquad (2.1)$$

where σ is the density operator of the dressed atom, *H* is the Hamiltonian describing atom and laser photons interacting together (thus including absorption and stimulated emission), Γ is the Einstein coefficient associated with spontaneous emission, and S^+ and S^- are the raising and lowering atomic operators ($S^+ = |e\rangle \langle g|$; $S^- =$ $|g\rangle \langle e|$). Let us consider now the restriction σ_n of the density matrix σ inside the manifold \mathcal{E}_n . Its evolution equation can be written from (2.1) as [16], [18]

$$\frac{d}{dt}\sigma_n = -\frac{i}{\hbar} [H_n, \sigma_n] - \frac{\Gamma}{2} (S^+ S^- \sigma_n + \sigma_n S^+ S^-) + \Gamma S^- \sigma_{n+1} S^+.$$
(2.2)

 H_n is the restriction of the Hamiltonian inside the manifold ϵ_n :

$$H_{n} = n\hbar\omega_{L}|g, n\rangle\langle g, n|$$

$$+ (n\hbar\omega_{L} - \hbar\delta)|e, n - 1\rangle\langle e, n - 1|$$

$$+ \frac{\hbar\Omega}{2}(|g, n\rangle\langle e, n - 1| + |e, n - 1\rangle\langle g, n|)$$
(2.3)

IEEE JOURNAL OF QUANTUM ELECTRONICS, VOL. 24, NO. 7, JULY 1988

where Ω is the so-called Rabi nutation frequency. The first term of (2.2) describes the atom-laser coupling and includes absorption and stimulated emission (terms proportional to Ω in H_n). The second term describes the damping of σ_n due to spontaneous emission. The evolution associated with these two terms is restricted inside the manifold \mathcal{E}_n , while the third term describes the transfer to \mathcal{E}_n due to the damping of \mathcal{E}_{n+1} . This transfer term can be written as

$$\Gamma S^{-} \sigma_{n+1} S^{+} = \Gamma \pi(e, n) |g, n\rangle \langle g, n| \qquad (2.4)$$

where $\pi(e, n)$ is the population of the state $|e, n\rangle$ belonging to the manifold \mathcal{E}_{n+1} .

The solution σ_n of (2.2) obeys the integral equation

$$\sigma_n(t) = \int_0^{\tau} d\tau \mu_n(\tau) \, \Gamma \pi(e, n, t - \tau) \qquad (2.5)$$

where $\mu_n(\tau)$ is defined by

$$\frac{d}{d\tau} \mu_n(\tau) = -\frac{i}{\hbar} [H_n, \mu_n] - \frac{\Gamma}{2} (S^+ S^- \mu_n + \mu_n S^+ S^-) \quad (2.6)$$

$$\mu_n(\tau=0) = |g, n\rangle \langle g, n| \qquad (2.7)$$

(evolution inside the manifold \mathcal{E}_n —without transfer term starting from the initial state $|g, n\rangle$). It appears in (2.5) that the knowledge of the populations $\pi(e, n)$ is sufficient for the determination of the matrices $\sigma_n(t)$. Now, the populations of these states $|e, n\rangle$, $|e, n - 1\rangle \cdots$ are given by integral equations derived by taking the average value of (2.5) in the state $|e, n - 1\rangle$:

$$\pi(e, n-1, t) = \int_0^\infty d\tau W_n(\tau) \ \pi(e, n, t-\tau) \ (2.8)$$

with

$$W_n(\tau) = \Gamma \langle e, n-1 | \mu_n(\tau) | e, n-1 \rangle.$$
(2.9)

Finally, it appears that the problem of the evolution of $\pi(e, n)$ and therefore of σ_n , reduces to the evaluation from (2.6) and (2.7) of the function $W_n(\tau)$.

The previous equations are the formal expression of the qualitative picture of the radiative cascade: for the set of experiments analyzed in this paper (intensity measurements with broad-band photodetectors), absorption and stimulated emission processes correspond to reversible evolutions inside the various manifolds, while spontaneous processes can be considered as irreversible "quantum jumps" from one manifold to another one. The reversible evolution, for example, inside \mathcal{E}_n , can be switched off by an irreversible process which "projects" the system into the state $|g, n - 1\rangle$: after such a "jump," a new reversible evolution begins inside the manifold \mathcal{E}_{n-1} , and so on. It follows that time intervals between successive fluorescence photon emissions are statistically

independent random variables. Any photon statistics signal can therefore be deduced from the knowledge of the delay functions $W_n(\tau)$ which are just the probability distributions of these time intervals. Strictly speaking, the function $W_n(\tau)$ depends on *n* since the Rabi nutation frequency Ω varies as \sqrt{n} . But this dependence can be ignored when the distribution of the values of *n* is a quasiclassical distribution (i.e., when $1 \ll \Delta n \ll \overline{n}$ where \overline{n} and Δn are the mean value and width of the distribution), which is the case for a laser excitation. Therefore, all the delays have the same probability distribution $W(\tau)$.

We now switch to the explicit calculation of $W(\tau)$ from (2.6), (2.7), and (2.9). We first note that the structure of (2.6), involving only a commutator and an anticommutator with the operator μ_n , allows an important simplification: one can reduce (2.6)–(2.7) to equations bearing only on probability amplitudes, instead of working with density matrix equations as usual in relaxation theory. More precisely, we can rewrite (2.9) as

$$W_n(\tau) = \Gamma \left| \left\langle e, n-1 \right| \psi_n(\tau) \right\rangle \right|^2 \qquad (2.10)$$

where $|\psi_n(\tau)\rangle$ is the ket solution of

$$\begin{cases} \frac{d}{dt} \left| \psi_n(\tau) \right\rangle = \left(-\frac{i}{\hbar} H_n - \frac{\Gamma}{2} S^+ S^- \right) \left| \psi_n(\tau) \right\rangle \\ \left| \psi_n(0) \right\rangle = \left| g, n \right\rangle. \end{cases}$$
(2.11)

It thus appears from (2.10) that $W_n(\tau)$ is equal to the decay rate from the level $|e, n - 1\rangle$ at time τ , knowing that the dressed atom is in level $|g, n\rangle$ at time 0. Now, we note that $|\psi_n(\tau)\rangle$ evolves only in the manifold \mathcal{E}_n , so that it can be written

$$\left|\psi_{n}(\tau)\right\rangle = a_{0}(\tau)\left|g,n\right\rangle + a_{1}(\tau)\left|e,n-1\right\rangle \quad (2.12)$$

where the evolution of a_0 and a_1 is obtained from (2.11):

$$\begin{cases} i\dot{a}_0 = \frac{\Omega}{2} a_1 \\ i\dot{a}_1 = \frac{\Omega}{2} a_0 - i\left(\frac{\Gamma}{2} - i\delta\right) a_1. \end{cases}$$
(2.13)

Solving these equations, one finds in the simple case of an exactly resonant excitation ($\delta = 0$)

$$W(\tau) = \Gamma \frac{\Omega^2}{\lambda^2} \sin^2\left(\frac{\lambda\tau}{2}\right) \exp\left(-\frac{\Gamma\tau}{2}\right)$$
 (2.14)

with

$$\lambda^2 = \Omega^2 - \frac{\Gamma^2}{4} \tag{2.15}$$

(these expressions correspond to the case $\lambda^2 > 0$; otherwise, one has to change λ^2 into $-\lambda^2$ and the sine function into the hyperbolic sine). It is worth giving also the Laplace transform $\tilde{W}(p)$ of $W(\tau)$:

$$\tilde{W}(p) = \frac{\Gamma \Omega^2}{(2p + \Gamma) \left[p(p + \Gamma) + \Omega^2 \right]}.$$
 (2.16)

Before discussing how the photon statistics signals can

be deduced from these expressions, we now turn to the case of an incoherent excitation.

III. INCOHERENT EXCITATION OF A TWO-LEVEL ATOM

In this section, we suppose that the atom is irradiated by a resonant *broad-band* laser. If the spectral distribution of the laser light is large enough, relaxation theory can be used to describe not only the atom-vacuum field interaction, but also the atom-laser interaction. We obtain in this manner the following rate equations for the populations of the "dressed" states $|g, n\rangle$ and $|e, n - 1\rangle$ where *n* is the *total* number of laser photons (summed over all frequencies):

$$\frac{d}{dt}\pi(g,n) = \Gamma'(\pi(e,n-1) - \pi(g,n)) + \Gamma\pi(e,n)$$
(3.1a)

$$\frac{d}{dt}\pi(e, n-1) = \Gamma'(\pi(g, n) - \pi(e, n-1)) - \Gamma\pi(e, n-1).$$
(3.1b)

Absorption and stimulated emission processes are described by transitions inside a given manifold \mathcal{E}_n (term proportional to Γ'), while spontaneous emission processes are described by transitions between adjacent manifolds (terms proportional to Γ). The absorption and stimulated emission rate Γ' is the product of the Einstein *B* coefficient by the power spectrum of the laser at the atomic frequency, while the spontaneous emission rate Γ is the Einstein *A* coefficient. As we study the case of a broadband laser, we will consider that the distribution of *n* is a quasi-classical distribution $(1 \ll \Delta n \ll \overline{n})$ and we will therefore ignore the variation of Γ' with *n*.

Equations (3.1) describe the radiative cascade in the case of an incoherent excitation. Since the spontaneous emission terms have the same formal structure as for coherent excitation, one can solve this equations in the same manner. In particular, their solutions obey the integral equation

$$\pi(g, n, t) = \int_0^\infty d\tau \rho(g, n, \tau) \,\Gamma \pi(e, n, t - \tau)$$
(3.2a)

$$\pi(e, n - 1, t) = \int_0^\infty d\tau \rho(e, n - 1, \tau) \,\Gamma \pi(e, n, t - \tau)$$
(3.2b)

where the populations ρ are defined by

$$\frac{d}{d\tau}\rho(g, n, \tau) = \Gamma'(\rho(e, n-1) - \rho(g, n))$$
(3.3a)

$$\frac{d}{d\tau}\rho(e, n-1, \tau) = \Gamma'(\rho(g, n) - \rho(e, n-1))$$
$$-\Gamma\rho(e, n-1) \qquad (3.3b)$$

$$\rho(g, n, 0) = 1; \, \rho(e, n - 1, 0) = 0. \quad (3.4)$$

This equation describes the evolution inside the manifold \mathcal{E}_n , without a transfer term from the upper one, starting from the initial state $|g, n\rangle$ [compare to eqs. (2.6), (2.7)]. Finally, the populations of the states $|e, n\rangle$, $|e, n - 1\rangle$, \cdots obey the integral equations deduced from (3.2b):

$$\pi(e, n - 1, t) = \int_0^\infty d\tau W(\tau) \ \pi(e, n, t - \tau) \quad (3.5)$$

with

$$W(\tau) = \Gamma \rho(e, n-1, \tau). \tag{3.6}$$

An important difference with the case of a coherent excitation is that these equations can no longer be reduced to amplitude equations. This is associated with the fact that absorption and stimulated emission are now irreversible processes. Solving these equations, one finds the following expressions for the delay function $W(\tau)$ and its Laplace transform $\tilde{W}(p)$:

$$W(\tau) = \Gamma \frac{\Gamma'}{\kappa} \operatorname{sh}\left(\frac{\kappa\tau}{2}\right) \exp\left(-\frac{\Gamma+2\Gamma'}{2}\tau\right) \quad (3.7)$$

with

$$\kappa = \sqrt{\Gamma^2 + 4\Gamma'^2} \tag{3.8}$$

and

$$\tilde{W}(p) = \frac{\Gamma\Gamma'}{p(p+\Gamma) + \Gamma'(2p+\Gamma)}.$$
 (3.9)

IV. PHOTON STATISTICS SIGNALS

We have shown in the previous sections that the succession of spontaneous emission times is a stochastic point process characterized by the delays between successive emissions. These delays are statistically independent random variables and all have the same probability distribution $W(\tau)$. It follows that any photon statistics signal, i.e., a signal in which only the intensity of the fluorescence field is measured, can be calculated from this "delay function" $W(\tau)$.

As a first example, we want to deduce the mean value and the fluctuations of the number m of photons emitted during a given time t. We first study a slightly different problem: what is the distribution for the variable t which is the sum of m successive delays (time between the emission of some photon m_0 and the emission of the photon $m_0 + m$)? As successive delays are independent variables, one obtains

$$\bar{t} = m\bar{\tau}; \,\Delta t^2 = m\Delta\tau^2 \tag{4.1}$$

where $\overline{\tau}$ and $\Delta \tau$ are the mean value and dispersion of one delay τ [which can be calculated from the distribution $W(\tau)$]. When *m* is a large number, one also knows that the distribution $p_m(t)$ of *t* is a Gaussian distribution (central limit theorem):

$$p_m(t) \sim \exp\left[-(t - m\bar{\tau})^2/(2m\Delta\tau^2)\right].$$
 (4.2)

Coming back to the original problem, one can obtain the distribution $q_t(m)$ of the number *m* of photons emitted during a fixed time *t* by inferring that it is also a Gaussian distribution at the limit $t \gg \overline{\tau}$ and by identifying the arguments of the exponential functions. From

$$\frac{\left(t-m\bar{\tau}\right)^2}{2m\Delta\tau^2} = \frac{\left(m-t/\bar{\tau}\right)^2}{2m\Delta\tau^2/\bar{\tau}^2}$$
(4.3)

one deduces the mean value and the dispersion of the distribution $q_t(m)$:

$$\overline{m} = t/\overline{\tau} \tag{4.4}$$

$$\Delta m^2 = \overline{m} \Delta \tau^2 / \overline{\tau}^2. \tag{4.5}$$

Equation (4.4) means that the mean value I of the intensity is just the inverse of the mean delay:

$$I = \overline{m}/t = 1/\overline{\tau}.$$
 (4.6)

One can calculate $\overline{\tau}$ from the expression of $W(\tau)$ or, in a simpler manner, from the expression of $\tilde{W}(p)$ which can be considered as the characteristic function of the distribution $W(\tau)$:

$$\bar{\tau} = \left[-\frac{d}{dp} \,\tilde{W}(p) \right]_{(p=0)}.\tag{4.7}$$

One thus recovers the usual result in the coherent case:

$$I = \Gamma \Omega^2 / (\Gamma^2 + 2\Omega^2)$$
 (4.8a)

and in the incoherent case:

$$\bar{I} = \Gamma \Gamma' / (\Gamma + 2\Gamma'). \tag{4.8b}$$

Equation (4.5) shows that the variance Δm^2 is proportional to the mean value \overline{m} . The Poisson statistics correspond to the particular case where the delay dispersion is equal to the mean delay. This would occur, for example, if the delay function had an exponential form. But it clearly appears that the statistics can be sub-Poissonian or super-Poissonian depending on the ratio $\Delta \tau/\overline{\tau}$. The statistics are usually characterized by the Q factor [21] defined through

$$\Delta m^2/\overline{m} = 1 + Q = \Delta \tau^2/\overline{\tau}^2. \tag{4.9}$$

Computing the delay variance from

$$\Delta \tau^2 = \left[\frac{d^2}{dp^2} \log \tilde{W}(p) \right]_{(p=0)}$$
(4.10)

one recovers the well-known result for the Q factor in the coherent case [21]. For example, for $\delta = 0$, we get

$$Q = -6\Gamma^{2}\Omega^{2}/(\Gamma^{2} + 2\Omega^{2})^{2}.$$
 (4.11a)

This corresponds to a photon noise reduction which can reach the value 1 + Q = 1/4 for $\Gamma^2 = 2\Omega^2$. A less known result is that photon noise reduction also occurs in the case of an incoherent excitation where one finds

$$Q = -2\Gamma\Gamma' / (\Gamma + 2\Gamma')^2 \qquad (4.11b)$$

reaching 1 + Q = 3/4 for $\Gamma = 2\Gamma'$. As usual, these factors correspond to the statistics of photon emissions.

The Q factors associated with the statistics of photon detections are often much smaller since they have to be multiplied by the probability for an emitted photon to be effectively detected.

There exist other signals characterizing the fluctuations of the fluorescence intensity. The photon correlation signal has been studied in great detail, experimentally [6], [7] as well as theoretically [12], [22], [23] since it reveals the effect of photon antibunching. It is interesting to see how this signal is related to the picture of the dressed atom radiative cascade.

The correlation signal $C(t, t + \tau)$ is associated with the detection of a first photon at time t and of a second one at time $t + \tau$. As discussed previously, the emission of the first fluorescence photon "projects" the atom-laser photons system into some state $|g, n - 1\rangle$. One thus expects that the correlation signal is given by

$$C(t, t + \tau) = \overline{I}J(\tau) \tag{4.12}$$

where \overline{I} is the mean intensity (associated with the first emission) and $J(\tau)$ is the transient intensity emitted by the system starting at $\tau = 0$ from this state $|g, n - 1\rangle$. The function $J(\tau)$ is thus different from $W(\tau)$: $W(\tau)$ corresponds to the emission of the *next* photon at time τ , while $J(\tau)$ corresponds to the emission of any photon (not necessarily the next one) at time τ . So, $W(\tau)$ is proportional to the population of the state $|e, n\rangle$ at time τ , knowing that the system is projected into $|g, n + 1\rangle$ at time 0 [see (2.9)], while $J(\tau)$ is proportional to the sum of all the populations of the states $|e, n'\rangle$ (with $n' \leq n$) with the same conditions. One can therefore deduce from (2.8) the following relation between J and W [16], [19]:

$$I = W + W \otimes W + W \otimes W \otimes W + \cdots \quad (4.13)$$

(the symbol \otimes represents the convolution product) which has a very simple meaning since W corresponds to the first emission, $W \otimes W$ to the second one, and so on. This equation leads to an algebraic relation between the Laplace transforms \tilde{J} and \tilde{W} [16], [19]:

$$\tilde{J} = \tilde{W} + (\tilde{W})^2 + (\tilde{W})^3 + \cdots = \frac{\tilde{W}}{1 - \tilde{W}}.$$
 (4.14)

One can check that these results are identical to those already known for the correlation signal.

We will conclude this section by studying the noise spectrum of the fluorescence intensity $S_I(\omega)$ which could, in principle, be measured by entering into a spectrum analyzer the fluctuations of the fluorescence intensity.

One can show that this signal is directly related to the Fourier transform of $J(\tau)$:

$$S_{I}(\omega) = S_{0}(1 + Q(\omega)) \qquad (4.15)$$

with

$$Q(\omega) = \tilde{J}(i\omega) + \tilde{J}(-i\omega) \qquad (4.16)$$

where S_0 is the standard photon noise and is proportional to the mean intensity \overline{I} . The limit $\omega \to 0$ gives the results already discussed for the usual Q factor since counting photons during a long time is equivalent to analyzing at zero frequency the fluorescence intensity. But it is also possible to reduce photon noise at frequencies different from zero. A particularly interesting case corresponds to a coherent excitation at the high-intensity limit:

$$W(\tau) = \Gamma \sin^2\left(\frac{\Omega \tau}{2}\right) \exp\left(-\frac{\Gamma \tau}{2}\right)$$
 (4.17)

(see (2.13) and (2.14) with $\Omega >> \Gamma$). The delay function $W(\tau)$ exhibits a damped oscillation at frequency Ω , the so-called "optical Rabi nutation." As a consequence, the factor $Q(\omega)$ presents a peak structure around Ω . From (4.14)-(4.17), one finds that this peak corresponds to a photon noise reduction which can reach the value $1 + Q(\omega) = 1/3$ (for $\omega = \Omega$; $\Omega >> \Gamma$). Note that, as for zero frequency, Q has to be multiplied by the photon detection efficiency in order to get the factor corresponding to the experimental effect.

V. INTERMITTENT FLUORESCENCE OF A THREE-LEVEL ATOM: THE QUANTUM JUMPS

The picture of the radiative cascade of the dressed atom is, of course, not limited to two-level systems. For example, this picture is very convenient for studying the phenomenon of intermittent fluorescence recently observed on a three-level system. Such an effect occurs when a system with three levels g, e_B , e_R in a V configuration [see Fig. 2(a)] is simultaneously excited by two laser waves respectively resonant with the "blue" transition $g - e_B$ and the "red" transition $g - e_R$, the red transition being much weaker than the blue one. The fluorescence signal, represented in Fig. 2(b), then exhibits bright periods, with many fluorescence photons, alternating with long dark periods during which no photon is emitted.

The existence of this effect has been initially suggested by Dehmelt [24] who used the following picture: the atom excited by the "blue" laser emits many fluorescence photons on the $g - e_B$ transition, with a rate $\simeq \Gamma_B/2$ if the transition is saturated (Γ_B^{-1} is the radiative lifetime of e_B). However, when the "red" laser is resonant with the g – e_R transition, the atom can also absorb a red photon and jump into level e_R . This "shelving" interrupts the blue fluorescence (dark period) until the atom jumps back from e_R to g either by spontaneous emission (time constant Γ_R^{-1}) or by stimulated emission. Consequently, the resonance of the red laser and the absorption of a single red photon results in the absence of a very large number of blue photons. This scheme therefore allows a very sensitive detection of the weak resonance, with an amplification factor Γ_B/Γ_R which reaches 10⁸ in the recent experimental results.

This scheme also leads to a unique feature as pointed out by Cook and Kimble [25]: one can observe "by eye" the quantum jumps of a microscopic device. In absence of the red laser, it is indeed possible to observe by eye the blue fluorescence of a single trapped ion [9]-[11]. When the red laser is applied, one can then look "in real time"

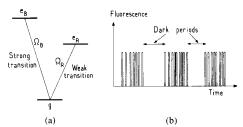


Fig. 2. (a) Three-level atom with two transitions starting from the ground state, one strong $(g - e_B)$ and one weak $(g - e_R)$. (b) Random sequence of pulses given by a photodetector recording the fluorescence of a single atom. The dark periods correspond to the shelving of the atom on the metastable level e_R .

at the atom jumping from g to e_R (beginning of a dark period) or from e_R to g (beginning of a bright period).

After the proposal of Cook and Kimble, several theoretical treatments have been proposed for this effect [17]– [18], [26]–[31]. Among them, the dressed atom approach appears as a simple and efficient way to predict the various characteristics of the signal [17], [18]. In this approach, fluorescence is interpreted as a radiative cascade of the dressed atom, and dark periods are due to random interruptions of this cascade. All the information concerning these dark periods can therefore be deduced from the delay function $W(\tau)$ characterizing the probability distribution of the time interval between two successive fluorescence photons [17]–[18]. In the two next subsections, we will outline the calculation of $W(\tau)$ for both coherent and incoherent laser excitation, and we will discuss the main physical results.

Coherent Laser Excitation

For a coherent excitation, the calculation of the delay function is done in a way very similar to the procedure presented in Section II. Here, the states of the dressed atom are bunched in three-dimensional manifolds $\epsilon(n_B, n_R)$ and can be written in the absence of atom-laser fields coupling

$$\begin{cases} |\varphi_0\rangle = |g, n_B, n_R\rangle \\ |\varphi_1\rangle = |e_B, n_B - 1, n_R\rangle \\ |\varphi_2\rangle = |e_R, n_B, n_R - 1\rangle \end{cases}$$
(5.1)

where n_B and n_R are the numbers of blue and red laser photons. The delay function $W(\tau)$ is then given [cf. (2.10), (2.12)]

$$W(\tau) = \Gamma_B |\langle e_B, n_B - 1, n_R | \psi(\tau) \rangle|^2 + \Gamma_R |\langle e_R, n_B, n_R - 1 | \psi(\tau) \rangle|^2$$
(5.2)

where $|\psi(\tau)\rangle$ is the ket solution of

$$\begin{cases} \frac{d}{dt} \left| \psi(\tau) \right\rangle = \left(-\frac{i}{\hbar} H_{n_B n_R} - \frac{\Gamma_B}{2} S_B^+ S_B^- - \frac{\Gamma_R}{2} S_R^+ S_R^- \right) \left| \psi(\tau) \right\rangle \\ \left| \psi(0) \right\rangle = \left| g, n_B, n_R \right\rangle. \end{cases}$$
(5.3)

The Hamiltonian $H_{n_Bn_R}$ is the restriction, inside the manifold $\mathcal{E}(n_B, n_R)$ of the atom-laser fields Hamiltonian, generalizing (2.3), and $S^+_{B(R)}$ and $S^-_{B(R)}$ are the raising and lowering operators:

$$\begin{cases} S_{B(R)}^{-} = |g\rangle \langle e_{B(R)}| \\ S_{B(R)}^{+} = |e_{B(R)}\rangle \langle g|. \end{cases}$$
(5.4)

When $|\psi(\tau)\rangle$ is written in the form

$$\left|\psi(\tau)\right\rangle = \sum_{i=0}^{2} a_{i}(\tau) \left|\varphi_{i}\right\rangle \qquad (5.5)$$

the equations of motion for the a_i coefficients read

$$\begin{cases}
i\dot{a}_0 = \frac{\Omega_B}{2}a_1 + \frac{\Omega_R}{2}a_2 \\
i\dot{a}_1 = \frac{\Omega_B}{2}a_0 - \frac{i\Gamma_B}{2}a_1 \\
i\dot{a}_2 = \frac{\Omega_R}{2}a_0 - \frac{i\Gamma_R}{2}a_2
\end{cases} (5.6)$$

where Ω_B and Ω_R represent the blue and red Rabi frequencies. Both blue and red lasers are here supposed to be resonant. A straightforward calculation then leads to the delay function

$$W(\tau) = \Gamma_B \left| a_1(\tau) \right|^2 + \Gamma_R \left| a_2(\tau) \right|^2 \qquad (5.7a)$$

which can be written, as soon as Γ_R and Ω_R are much smaller than Γ_B and Ω_B , as

$$W(\tau) = W_{\text{short}}(\tau) + W_{\text{long}}(\tau). \quad (5.7b)$$

 $W_{\text{short}}(\tau)$ is equal to the delay function found if only the blue laser is present (2.13), while $W_{\text{long}}(\tau)$ is an additional term, evolving with a time constant τ_{long} much longer than the time constants of W_{short} . This evolution with two time constants is a clear signature for the existence of dark and bright periods: in a bright period, successive photons are separated by an average time τ_{short} deduced from W_{short} (4.8a). In the limit of a weak blue laser excitation ($\Omega_B \ll \Gamma_B$), we get from (4.8a)

$$\tau_{\rm short}^{-1} = \Omega_B^2 / \Gamma_B. \tag{5.8}$$

On the other hand, the long time constant τ_{long} , characterizing the length of dark periods, is given by

$$\tau_{\text{long}}^{-1} = \Gamma_R + \Gamma_B \Omega_R^2 / \Omega_B^2.$$
 (5.9)

As expected, τ_{long}^{-1} represents the departure rate from e_R to g induced by spontaneous emission [first term of (5.9)] and by stimulated emission [second term of (5.9)]. These results are then in perfect agreement with the picture given by Dehmelt: the atom cycles on the $g - e_B$ transition during bright periods, and randomly jumps on the shelf e_R (dark period) before falling back on g either by spontaneous emission or by stimulated emission.

Incoherent Laser Excitation

For an incoherent excitation, the procedure of Section III can be duplicated in a straightforward way. The delay function writes

$$W(\tau) = \Gamma_B \rho_B(\tau) + \Gamma_R \rho_R(\tau) \qquad (5.10)$$

where we have put [cf. (3.6)]

$$\begin{cases} \rho_B(\tau) = \rho(e_B, n_B - 1, n_R, \tau) \\ \rho_R(\tau) = \rho(e_R, n_B, n_R - 1, \tau) \\ \rho_g(\tau) = \rho(g, n_B, n_R, \tau). \end{cases}$$
(5.11)

The populations ρ_B , ρ_R , ρ_g obey the differential system

$$\begin{cases}
\dot{\rho}_B = \Gamma'_B(\rho_g - \rho_B) - \Gamma_B \rho_B \\
\dot{\rho}_R = \Gamma'_R(\rho_g - \rho_R) - \Gamma_R \rho_R \\
\dot{\rho}_g = \Gamma'_B(\rho_B - \rho_g) + \Gamma'_R(\rho_R - \rho_g) \\
\rho_g(0) = 1 \qquad \rho_B(0) = \rho_R(0) = 0.
\end{cases}$$
(5.12)

 Γ'_B and Γ'_R are the rates for absorption and stimulated emission on the blue and red transitions (Γ_R , $\Gamma'_R << \Gamma_B$, Γ'_B). The delay function $W(\tau)$, calculated from (5.10), can again be written

$$W(\tau) = W_{\text{short}}(\tau) + W_{\text{long}}(\tau) \qquad (5.12)$$

where $W_{\text{short}}(\tau)$ is just equal to the delay function calculated for the two-level system $g - e_B$ alone in Section III [cf. (3.7)]. The additional term W_{long} , with a long time constant τ_{long} , is the signature of the existence of dark periods. The average length of these dark periods is equal to

$$\tau_{\text{long}}^{-1} = \Gamma_R + \Gamma_R'. \tag{5.14}$$

Here, again, Dehmelt's picture for quantum jumps is fully confirmed by these analytical results.

VI. CONCLUSION

We have shown in this paper how a single function, the delay function $W(\tau)$, can provide a quantitative understanding of various photon statistics signals. It must be noted, however, that such a simplification does not necessarily apply to all situations. All the experiments analyzed in this paper are characterized by the fact that the emission of a photon "projects" the atom in a well-defined state, the ground state g. Therefore, after each emission, the dressed atom starts from a well-defined state of a given manifold \mathcal{E}_n , and the study of its subsequent evolution in \mathcal{E}_n gives unambiguously the distribution of the delay between this first emission and the next one. One could, however, consider situations where the emission of a photon does not necessarily project the atom in a welldefined state. This occurs, for example, when the atom can decay to several lower states. It is then clear that the statistics of emission times cannot be analyzed with a single delay function. Another example is the study of temporal correlations between the photons emitted in the various components of the fluorescence triplet of a two-level atom. The dressed state in which the system is projected depends on the line in which the photon is emitted.

For all these more complex situations, the picture of the radiative cascade of the dressed atom appears to still be quite useful. For example, the study of the radiative cascade on the basis of dressed states allows a quantitative understanding of the temporal correlations between the photons emitted in the two sidebands of the fluorescence triplet [15], [16], [32], [33]. We can also mention the "black resonances" of Gozzini [34]–[36] which appear in three-level systems with a \land configuration (one excited level and two ground state levels). In the dressed atom approach, these resonances are interpreted as being due to a stopping of the radiative cascade occurring when the dressed atom decays in a stable trapping level [37]–[38].

REFERENCES

- A. Einstein, "Zur Quanten-theorie der Strahlung," Physikal Zeitschrift, vol. 18, pp. 121-128, 1917.
- [2] P. A. M. Dirac, "The quantum theory of the emission and absorption of radiation," Proc. Roy. Soc., vol. 114, pp. 243-265, 1927.
- [3] V. Weisskopf and E. Wigner, "Berechnung der natürlichen Linenbreite auf Grund der Diracschen Lichttheorie," Zeitschrift Phys., vol. 63, pp. 54-73, 1930.
- [4] F. Schuda, C. R. Stroud, and M. Hercher, "Observation of the resonant Stark effect at optical frequencies," J. Phys., vol. B7, pp. L 198-L 202, 1974.
- [5] R. E. Grove, F. Y. Wu, and S. Ezekiel, "Measurement of the spectrum of resonance fluorescence from a two-level atom in an intense monochromatic field," *Phys. Rev.*, vol. A15, pp. 227-233, 1977.
- [6] J. D. Cresser, J. Hager, G. Leuchs, M. Rateike, and H. Walther, "Resonance fluorescence of atoms in strong monochromatic laser fields," in *Dissipative Systems in Quantum Optics, Topics in Current Physics*, Vol. 27, R. Bonifacio, Ed. Berlin: Springer-Verlag, 1982, pp. 21-59.
- [7] M. Dagenais and L. Mandel, "Investigation of two-time correlations in photon emissions from a single atom," *Phys. Rev.*, vol. A18, pp. 2217-2228, 1978.
- [8] R. Short and L. Mandel, "Observation of sub-Poissonian photon statistics," Phys. Rev. Lett., vol. 51, pp. 384-387, 1983.
- [9] W. Nagourney, J. Sandberg, and H. Dehmelt, "Shelved optical electron amplifier: Observation of quantum jumps," *Phys. Rev. Lett.*, vol. 56, pp. 2797-2799, 1986.
- [10] Th. Sauter, W. Neuhauser, R. Blatt, and P. E. Toschek, "Observation of quantum jumps," *Phys. Rev. Lett.*, vol. 57, pp. 1696–1698, 1986.
- [11] J. C. Bergquist, R. B. Hulet, W. M. Itano, and D. J. Wineland, "Observation of quantum jumps in a single atom," *Phys. Rev. Lett.*, vol. 57, pp. 1699-1702, 1986.
- [12] C. Cohen-Tannoudji, "Atoms in strong resonant fields," Frontiers in Laser Spectroscopy, Les Houches, session XXVII, 1975, R. Balian, S. Haroche, and S. Liberman, Eds. Amsterdam: North-Holland, 1977, pp. 3-104.
- [13] B. R. Mollow, "Theory of intensity dependent resonance light scattering and fluorescence," in *Progress in Optics XIX*, E. Wolf, Ed. Amsterdam: North-Holland, 1981, pp. 1-43.
- [14] C. Cohen-Tannoudji and S. Reynaud, "Dressed-atom description of resonance fluorescence and absorption spectra of a multi-level atom in an intense laser beam," J. Phys., vol. B10, pp. 345-363, 1977. See also same authors, "Dressed atom approach to resonance fluorescence," in Multiphoton Processes, J. Eberly and P. Lambropoulos, Eds. New York: Wiley, 1978, pp. 103-118.
- [15] —, "Atoms in strong light-fields: Photon antibunching in single atom fluorescence," *Phil. Trans. Roy. Soc. London*, vol. A293, pp. 223-237, 1979.
- [16] S. Reynaud, "La fluorescence de résonance: Étude par la méthode de l'atome habillé," Annales de Physique, vol. 8, pp. 315-370, 1983.
- [17] C. Cohen-Tannoudji and J. Dalibard, "Single-atom laser spectroscopy. Looking for dark periods in fluorescence light," *Europhys. Lett.*, vol. 1, pp. 441-448, 1986.
- [18] P. Zoller, M. Marte, and D. F. Walls, "Quantum jumps in atomic systems," *Phys. Rev.*, vol. A35, pp. 198-207, 1987.
- [19] M. S. Kim, P. L. Knight, and K. Wodkiewicz, "Correlations be-

tween successively emitted photons in resonance fluorescence," Opt. Commun., vol. 62, pp. 385-388, 1987.

- [20] G. S. Agarwal, "Quantum statistical theories of spontaneous emission and their relation to other approaches," in Springer Tracts in Modern Physics, Vol. 70, 1974.
- [21] L. Mandel, "Sub-Poissonian photon statistics in resonance fluorescence," Opt. Lett., vol. 4, pp. 205-207, 1979.
- [22] H. Carmichael and D. F. Walls, "Proposal for the measurement of the resonant Stark effect by photon correlation techniques," J. Phys., vol. B9, pp. L 43-L 46, 1976, and "A quantum-mechanical master equation treatment of the dynamical Stark effect," J. Phys., vol. B9, pp. 1199-1219, 1976.
- [23] H. J. Kimble and L. Mandel, "Theory of resonance fluorescence," Phys. Rev., vol. A13, pp. 2123-2144, 1976.
- [24] H. G. Dehmelt, "Mono-ion oscillator as potential ultimate laser frequency standard," IEEE Trans. Instrum. Meas., vol. IM-31, pp. 83-87, 1982; "Proposed $10^{14} \Delta \nu > \nu$ laser fluorescence spectroscopy on T1⁺ mono-ion oscillator II," Bull. Amer. Phys. Soc., vol. 20, p. 60, 1975.
- [25] R. J. Cook and H. J. Kimble, "Possibility of direct observation of
- quantum jumps, " Phys. Rev. Lett., vol. 54, pp. 1023-1026, 1985. [26] J. Javanainen, "Possibility of quantum jumps in a three-level system," Phys. Rev., vol. A33, pp. 2121-2123, 1986.
- [27] D. T. Pegg, R. Loudon, and P. L. Knight, "Correlations in light emitted by three-level atoms," Phys. Rev., vol. A33, pp. 4085-4091, 1986.
- [28] F. T. Arecchi, A. Schenzle, R. G. De Voe, K. Jungman, and R. G. Brewer, "Comment on the ultimate single-ion laser frequency standard," Phys. Rev., vol. A33, pp. 2124-2126, 1986.
- [29] A. Schenzle, R. G. De Voe, and R. G. Brewer, "Possibility of quantum jumps," *Phys. Rev.*, vol. A33, pp. 2127–2130, 1986.
- [30] A. Schenzle and R. G. Brewer, "Macroscopic quantum jumps in a single atom," Phys. Rev., vol. A34, pp. 3127-3142, 1986.
- [31] H. J. Kimble, R. J. Cook, and A. L. Wells, 'Intermittent atomic fluorescence,' Phys. Rev., vol. A34, pp. 3190-3195, 1986.
- [32] P. A. Apanasevich and S. J. Kilin, "Photon bunching and antibunching in resonance fluorescence," J. Phys., vol. B12, pp. L 83-L 86, 1979.
- [33] A. Aspect, G. Roger, S. Reynaud, J. Dalibard, and C. Cohen-Tannoudji, "Time correlations between the two sidebands of the reso-

nance fluorescence triplet," Phys. Rev. Lett., vol. 45, pp. 617-620, 1980.

- [34] G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, "An experimental method for the observation of R.F. transitions and laser beat resonances in oriented Na vapour," 11 Nuovo Cimento, vol. 36B, pp. 5-20, 1976.
- [35] J. E. Thomas, P. R. Hemmer, S. Ezekiel, C. C. Leiby, R. H. Picard, and C. R. Willis, "Observation of Ramsey fringes using a stimulated resonance Raman transition in a sodium atomic beam," *Phys. Rev.* resonance Raman transition in a sodium atomic beam, Lett., vol. 48, pp. 867-870, 1982.
- [36] G. Janik, W. Nagourney, and H. G. Dehmelt, "Doppler-free optical spectroscopy on the Ba⁺ mono-ion oscillator," J. Opt. Soc. Amer., vol. B2, pp. 1251-1257, 1985.
- [37] P. M. Radmore and P. L. Knight, "Population trapping and dispersion in a three-level system," J. Phys., vol. B15, pp. 561-573, 1982. [38] J. Dalibard, S. Reynaud, and C. Cohen-Tannoudji, "La cascade ra-
- diative de l'atome habillé," in Interaction of Radiation with Matter, a volume in honor of Adriano Gozzini, Scuola Normale Superiore, Pisa, Italy, 1987, pp. 29-48.
- [39] P. L. Kelley and W. H. Kleiner, "Theory of electromagnetic field measurement and photoelectron counting," Phys. Rev., vol. 136, pp. A 316-A 334, 1964.
- [40] F. Davidson and L. Mandel, "Photoelectric correlation measurements with time to amplitude converter," J. Appl. Phys., vol. 39, pp. 62-66, 1968.

Serge Revnaud, photograph and biography not available at the time of publication.

Jean Dalibard, photograph and biography not available at the time of publication.

Claude Cohen-Tannoudji, photograph and biography not available at the time of publication