Quantum jumps in atomic systems

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We consider a three-level atomic system driven strongly on one transition and weakly on the other. The excited state on the weak transition is assumed to be metastable. We give an analysis of the fluorescence from the strong transition in terms of the elementary probability density $p_{[0,t)}(t_1,t_2,\ldots,t_n)$ which gives us the probability density that exactly n photons are emitted at times $t_1,t_2,\ldots,t_n$ by the atom in the time interval $[0,t)$. We show that $p_{[0,t)}$ essentially factorizes into products of conditional densities $\tilde{c}(\tau)$ that, given a photon is emitted at time zero, the next photon emission occurs at time τ. This enables a simulation of the individual photon emissions to be given which shows directly the existence of prolonged dark windows in the fluorescence corresponding to the shelving of the electron in the metastable state or "quantum jumps."

I. INTRODUCTION

There have recently been a number of discussions in the literature concerning the possibility of observing quantum jumps in atomic systems.[1–4] The system currently under discussion involves a double-resonance scheme illustrated in Fig. 1(a) where two excited states $|1\rangle$ and $|2\rangle$ are connected to a common lower level $|0\rangle$ via a strong and weak transition, respectively. The fluorescent photons from the strong transition are observed. However, an excitation of the weak transition where the electron is temporarily shelved in the metastable level $|2\rangle$ will cause the strong transition to be turned off. It is, therefore, possible to monitor the quantum jumps of the weak transition via the macroscopic signal provided by the fluorescence of the strong transition. In the language of quantum measurement theory,[5] the fluorescence from the strong transition acts as a pointer from which the microscopic quantum state of the atom may be determined. A similar effect may be observed for an atom in the Λ configuration shown in Fig. 1(b).

This idea was first suggested by Dehmelt[6] as a way to detect a weak transition in single-atom spectroscopy. Because the weak-transition linewidth may be exceptionally narrow, this scheme has been proposed for an ultimate laser frequency standard.

Cook and Kimble[1] have argued that since the weak transitions occur randomly in time, the atomic fluorescence intensity has the form of a random telegraph signal. Using a rate-equation approach they have calculated the probability density for the durations τ of darkness (off times) and light (on times) in the fluorescent signal and show them to be exponentially distributed.

The paper by Cook and Kimble has stimulated a number of responses. Schenzle *et al.*[2] and Pegg *et al.*[4] have calculated the conditional probabilities that, given a photon is observed at time t, another photon but not necessarily the next one is observed at time $t+\tau$. From these intensity correlation functions these authors infer the existence of significant dark periods in the fluorescence and hence quantum jumps. Javanainen[3] has calculated the nth-order conditional probabilities of the type described above and shows that the photon counting statistics are equivalent to a Markov jump process between two states. This allows him to deduce that the photon counting experiment should record alternating periods of finite counts and zero counts.

In the present paper we shall present a microscopic model based on calculating the elementary probability density $p_{[0,t)}(t_1,\ldots,t_n)$ which gives us the probability density that exactly n photons are emitted at times $t_1,\ldots,t_n$ by the atom in the time interval $[0,t)$ on the strong transition. In this context we calculate the conditional probabilities $\tilde{c}(\tau)$ that, given a photon has been emitted at $\tau=0$, the *next* photon emission occurs at time τ. This quantity is shown to be the sum of exponentials which for sufficient differential in the time scales describe the periods of light and darkness. Using this conditional probability it is possible to numerically simulate the individual photon emissions. These photon emissions clearly show the presence of emission windows where the electron has been shelved in the metastable level.

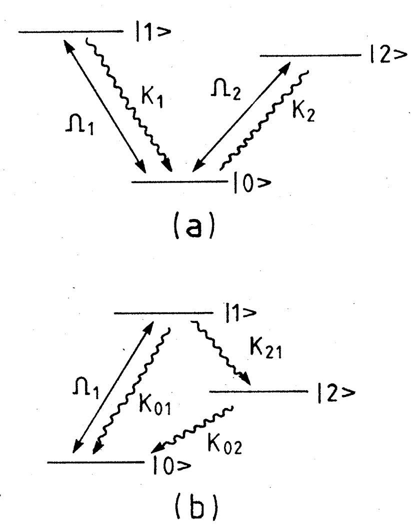

FIG. 1. (a) V system; (b) Λ system.

II. STATISTICS OF PHOTON EMISSION IN RESONANCE FLUORESCENCE FROM TWO- AND THREE-LEVEL SYSTEMS

We consider resonance fluorescence from a three-level system in a V configuration shown in Fig. 1(a). The ground state is denoted by $|0\rangle$ which is coupled to the upper states $|1\rangle$ and $|2\rangle$. Level $|2\rangle$ is assumed to be metastable. The system is driven by two lasers at the "strong" and "weak" transition lines $|0\rangle$-$|1\rangle$ and $|0\rangle$-$|2\rangle$, respectively. Most of the time the atom emits fluorescent photons on the strong transition line $|1\rangle$-$|0\rangle$. According to the suggestion of Cooke and Kimble[1] the "quantum jumps" of the atomic electron from the active two-level system $\{|0\rangle,|1\rangle\}$ to the metastable state $|2\rangle$ is reflected in the presence of emission windows on the strong transition line. In this section we derive general expressions for the elementary probability density $p_{[0,t)}(t_1,\ldots,t_n)$ which gives us the probability density that *exactly* n photons are emitted at times $t_1,\ldots,t_n$ by the atom in the time interval $[0,t)$ on the transition $|1\rangle$-$|0\rangle$. This provides us with a microscopic description of photon emission on the strong line. We show that the elementary probability density factorizes essentially into products of conditional probability densities $\tilde{c}(t_{r+1}|t_r)$ $(r=1,\ldots,n-1)$ (Markov property) which are the probabiilty densities that—given a photon has been emitted at time t_r—the *next* photon is emitted at time t_{r+1}. The conditional probability density $\tilde{c}$ governs the time distributions of photon emissions and are the basis of our understanding and interpretation of the presence of photon emission windows. We emphasize that these elementary probability densities $p_{[0,t)}$ differ from the coincidence probability densities usually considered in the context of calculating intensity correlations (antibunching) for n resonance fluorescence (see Sec. II A below).[1-4,7]

For simplicity we start developing the formalism below by considering the problem of resonance fluorescence from a two-level system (Sec. II A).[8] Generalization to the three-level situation is given in Sec. II B.

A. Two-level system

We consider a two-level atom with ground state $|0\rangle$ and excited state $|1\rangle$ which is driven by a classical light field and coupled to a bath of modes of the radiation field. The Hamiltonian for the combined system atom and radiation field is

$$H=H_{0A}+H_{0F}+H_1(t)\ . \tag{1}$$

Setting $\hbar=1$,

$$H_{0A}=\omega_{10}a_1^\dagger a_1 \tag{2}$$

is the free atomic Hamiltonian with ω_{10} the atomic transition frequency and $a_1=|0\rangle\langle 1|$ the atomic lowering operator. Denoting by $b_{\mathbf{k}\lambda}$ $(b_{\mathbf{k}\lambda}^\dagger)$ the destruction (creation) operator of the mode $\mathbf{k}$ of the radiation field with polarization $\epsilon_{\mathbf{k}\lambda}$ $(\lambda=1,2)$, the Hamiltonian of the free radiation field is

$$H_{0F}=\sum_\lambda \int d^3k\ \omega_{\mathbf{k}\lambda} b_{\mathbf{k}\lambda}^\dagger b_{\mathbf{k}\lambda}\ . \tag{3}$$

The interaction part is

$$H_1(t)=-\boldsymbol{\mu}_{10}^*\cdot[\mathscr{E}_R^\dagger(\mathbf{x}=0)+\mathscr{E}_c^*(\mathbf{x}=0,t)]a_1 -\boldsymbol{\mu}_{10}\cdot[\mathscr{E}_R(\mathbf{x}=0)+\mathscr{E}_c(\mathbf{x}=0,t)]a_1^\dagger \tag{4}$$

with $\boldsymbol{\mu}_{10}$ the atomic dipole matrix element,

$$\mathscr{E}_R^\dagger(\mathbf{x})=i\sum_\lambda\int d^3k\left[\frac{\hbar\omega}{2\epsilon_0(2\pi)^3}\right]^{1/2}\epsilon_{\mathbf{k}\lambda}e^{i\mathbf{k}\cdot\mathbf{x}}b_{\mathbf{k}\lambda}^\dagger \tag{5}$$

the positive frequency part of the quantized electromagnetic field; $\mathbf{x}=0$ denotes the position of the atom. The incident laser field is described by a classical light wave with positive frequency part $\mathscr{E}_c(\mathbf{x}=0,t)=\mathscr{E}_1\epsilon e^{-i\omega_1 t}$ with ω_1 being the laser frequency.

We define a reduced atomic density operator in the subspace containing exactly $n=0,1,2,\ldots$ scattered photons according to

$$\rho_A^{(n)}(t)=\mathrm{Tr}_F\{P^{(n)}\rho(t)\} \tag{6}$$

with $\rho(t)$ the density operator of the combined atom-field system,

$$P^{(n)}=\frac{1}{n!}\sum_{\lambda_1}\int d^3k_1\cdots\sum_{\lambda_n}\int d^3k_n b_{\mathbf{k}_1\lambda_1}^\dagger\cdots b_{\mathbf{k}_n\lambda_n}^\dagger|0\rangle \times\langle 0|b_{\mathbf{k}_n\lambda_n}\cdots b_{\mathbf{k}_1\lambda_1} \tag{7}$$

the projection operator onto the n-photon subspace, and Tr_F indicating the trace over the radiation modes. The probability of finding exactly n photons in the field at time t is given by

$$P^{(n)}(t)=\mathrm{Tr}_A\rho_A^{(n)}(t)\equiv\rho_{00}^{(n)}(t)+\rho_{11}^{(n)}(t) \tag{8}$$

with Tr_A a trace over the atomic variables. As has been shown by Mollow[8] (see also Blatt *et al.*[9]), $\rho_A^{(n)}(t)$ obeys the equation

$$\frac{d}{dt}\rho_a^{(n)}=-i(H_{\mathrm{eff}}\rho_A^{(n)}-\rho_A^{(n)}H_{\mathrm{eff}}^\dagger) +\kappa_1 a_1\rho_A^{(n-1)}a_1^\dagger(1-\delta_{n0})\ . \tag{9}$$

Here

$$H_{\mathrm{eff}}=(\omega_{10}-i\tfrac{1}{2}\kappa_1)a_1^\dagger a_1 -[\boldsymbol{\mu}_{10}\cdot\mathscr{E}_c(\mathbf{x}=0,t)a_1^\dagger+\mathrm{H.c.}] \tag{10}$$

is an effective non-Hermitian atomic Hamiltonian with κ_1 the spontaneous decay rate of the upper state in the two-level system. Summing over the n-photon contributions, Eq. (9) reduces to the familiar optical Bloch equations[7,8]

$$\frac{d}{dt}\rho_a=-i(H_{\mathrm{eff}}\rho_A-\rho_A H_{\mathrm{eff}}^\dagger)+\kappa_1 a_1\rho_A a_1^\dagger \tag{11}$$

for the reduced atomic density operator

$$\rho_A(t)=\sum_{n=0}^\infty\rho_A^{(n)}(t)\ . \tag{12}$$

The n-photon density matrix $\rho_A^{(n)}(t)$ is seen to obey an inhomogeneous equation of motion [Eq. (9)] for $n=1,2,\ldots$

with $|0\rangle\langle 0|\kappa_1\rho_{11}^{(n)}(t)$ as a source term. The solution of Eq. (9) can be written in the form

$$\rho_A^{(0)}(t)=S_{t,t_0}\rho_A(t_0=0) \tag{13a}$$

with the initial condition $\rho_A(t_0=0)=|0\rangle\langle 0|$ and

$$\rho_A^{(n)}(t)=\int_{t_0=0}^{t}dt' S_{t,t'}J_1\rho_A^{(n-1)}(t') \quad (n=1,2,\ldots) \tag{13b}$$

where we have defined a time-evolution operator of the homogeneous part of Eq. (9),

$$S_{t,t_0}\rho_A^{(n)}(t_0) = T\left[\exp\left[-i\int_{t_0}^{t}dt' H_{\text{eff}}(t')\right]\right]\rho_A^{(n)}(t_0) \times T\left[\exp\left[i\int_{t_0}^{t}dt' H_{\text{eff}}^{\dagger}(t')\right]\right] \tag{14}$$

with T denoting the time ordering, which can be interpreted as the time evolution confined to a particular subspace containing exactly n photons, i.e., the time evolution between two photon emissions. In addition we have written the source term in Eq. (9) in terms of the operator J_1 acting on $\rho_A^{(n-1)}$ as

$$J_1\rho_A^{(n-1)}=\kappa_1 a_1\rho_A^{(n-1)}a_1^{\dagger}=\kappa_1|0\rangle\langle 0|\rho_{11}^{(n-1)} . \tag{15}$$

Equation (13) describes a situation where atoms in the ground state with n photons in the field are created at time t' at a rate which is the product of the decay rate κ_1 times and the excited-state population $\rho_{11}^{(n-1)}$ with $n-1$ scattered photons in the radiation field. The time evolution following the spontaneous emission (until the next photon is emitted) is described by the non-Hermitian Hamiltonian H_{eff} according to Eq. (13). Solving the hierarchy (13) recursively we obtain for the n-photon probabilities

$$P^{(0)}(t)=\text{Tr}_A\{S_{t,0}\rho_A(0)\} \tag{16a}$$

and

$$P^{(n)}(t)=\int_0^t dt_n\int_0^{t_n}dt_{n-1}\cdots\int_0^{t_2}dt_1\,\text{Tr}_A\{S_{t,t_n}J_1S_{t_n,t_{n-1}}\cdots J_1S_{t_1,0}\rho_A(0)\} \tag{16b}$$

for $n=1,2,\ldots$. Equation (16) may be interpreted in terms of the time evolution of an atom in a time interval $[0,t)$ which emits exactly n photons at the times $t_1,\ldots,t_n$. Each spontaneous emission is accompanied by a reduction of the atomic density operator to a pure atomic state $|0\rangle\langle 0|$ as described by the operator J_1 in Eq. (16) (reduction of the wave packet). The time evolution between the spontaneous emission events is governed by the non-Hermitian Hamiltonian H_{eff} which describes the reexcitation of the atom to the upper state of the atom by the laser field. Equation (16) has a structure which is expected for an n-photon probability from continuous measurement theory as applied to the theory of photon counting by Srinivas and Davies.[10] In particular this theory supports the interpretation of

$$p_{[0,t)}(t_1,\ldots,t_n)=\text{Tr}_A\{S_{t,t_n}J_1\cdots J_1S_{t_1,0}\rho_A(0)\} \tag{17a}$$

as an elementary probability density that the atom emits n photons at times $t_1,\ldots,t_n$ (and no other photons) in the time interval $[0,t)$. According to the definition of S_{t,t_0} and J_1 we can rewrite Eq. (17) as

$$p_{[0,t)}(t_1,\ldots,t_n) =[\tilde{\rho}_{00}(t\,|\,t_n)+\tilde{\rho}_{11}(t\,|\,t_n)]\prod_{r=1}^{n}\kappa_1\tilde{\rho}_{11}(t_r\,|\,t_{r-1}) , \tag{17b}$$

where we have defined two functions

$$\tilde{\rho}_{jj}(t\,|\,t_0)=\langle j|(S_{t,t_0}|0\rangle\langle 0|)|j\rangle \equiv\rho_{jj}^{(0)}(t\,|\,t_0) \quad (j=0,1) . \tag{18}$$

By definition these functions satisfy the initial conditions $\tilde{\rho}_{00}(t_0\,|\,t_0)=1$ and $\tilde{\rho}_{11}(t_0\,|\,t_0)=0$. We note that—again following the general argument of Srinivas and Davies—the quantity

$$\tilde{c}(t_1\,|\,t_0)=\frac{\text{Tr}_A\{J_1S_{t_1,t_0}J_1T_{t_0,0}\rho_A(0)\}}{\text{Tr}_A\{J_1T_{t_0,0}\rho_A(0)\}}=\kappa_1\tilde{\rho}_{11}(t\,|\,t_n) \tag{19}$$

may be identified with the conditional probability that, given a photon was emitted at t_0, the *next* photon is emitted at time t_1. Since for this quantity it is of no importance how many photon emissions have occurred at times previous to t_0, a summation over all possible previous counts is involved and hence leads to the appearance of a time-evolution operator

$$T_{t_0,0}\rho_A(0)=\sum_{n=0}^{\infty}\int_0^t dt_n\int_0^{t_n}dt_{n-1}\cdots\int_0^{t_2}dt_1 S_{t,t_n}J_1S_{t_n,t_{n-1}}\cdots J_1S_{t_1,0}\rho_A(0) . \tag{20}$$

Using Eq. (13), Eq. (20) can be expressed as

$$T_{t,t_0}\rho_A(0)=\sum_{n=0}^{\infty}\rho_A^{(n)}(t)\equiv\rho_A(t) . \tag{21}$$

This shows that T_{t,t_0} is identical to the time-evolution operator U for the optical Bloch equations (11),

$$\rho_{\mu\nu}(t)\equiv[T_{t,t_0}\rho_A(t_0)]_{\mu\nu}=\sum_{\sigma,\tau}U_{\mu\nu,\sigma\tau}(t,t_0)\rho_{\sigma\tau}(t_0) \tag{22}$$

with the Greek indices labeling atomic states.

It is not difficult to convince ourselves that Eq. (19) is consistent with the interpretation of $\kappa_1\tilde{\rho}_{11}(t_1\,|\,t_0)$ as a conditional probability density for the emission of the next photon at time t_1 given that one was emitted at time t_0. First we note that in view of

$$\kappa_1\tilde{\rho}_{11}(t\mid t_0)=-\frac{d}{dt}[\tilde{\rho}_{00}(t\mid t_0)+\tilde{\rho}_{11}(t\mid t_0)] \quad (23)$$

we have

$$\int_{t_0}^{t} d\tau\,\tilde{c}(\tau\mid t_0)=1-[\tilde{\rho}_{00}(t\mid t_0)+\tilde{\rho}_{11}(t\mid t_0)] \equiv x\in[0,1] \quad (24)$$

with x a number in the interval between zero and one. If the driving field is monochromatic than $\tilde{\rho}_{00}(t\mid t_0)$ and $\tilde{\rho}_{11}(t\mid t_0)$, and hence $\tilde{c}(t\mid t_0)$, are functions of the time difference only. Equation (21) expresses the intuitively obvious result that the probability of emission of a photon in the time interval $[0,t)$—given the last photon emission occurred at time $t_0=0$—equals the population left in the atom at time t when it evolves according to the Hamiltonian H_{eff}. In particular Eq. (21) implies the normalization condition

$$\int_{t_0}^{\infty} d\tau\,\tilde{c}(\tau\mid t_0)=1\ . \quad (25)$$

To summarize, the elementary probability density $p_{[0,t)}$ can be written in the form

$$p_{[0,t)}(t_1,\ldots,t_n) = \left[1-\int_{t_n}^{t} d\tau\,\tilde{c}(\tau\mid t_n)\right]\prod_{r=1}^{n}\tilde{c}(t_r\mid t_{r-1})\quad (t_0=0) \quad (26)$$

which is the product of conditional densities describing photon emission at times $t_1,\ldots,t_n$ times the probability that no photon is emitted during $[t_n,t)$. Equation (26) expresses the fact that the elementary probability density satisfies the Markov property.

The conditional probability density $\tilde{c}(\tau\mid 0)$ is the basis for simulating sample photon emission data which can be observed in fluorescence of a single atom. Choosing x a random number, equally distributed in the interval between zero and one, gives according to Eq. (21) the (random) decay time t for the time delay between two successive photon emissions. Note that $\tilde{c}(t_0\mid t_0)=0$ which reflects the antibunching property of the emitted light.[11]

What is usually considered in studies of photon count statistics are coincidence probability densities that a photon is emitted at times $t_1,\ldots,t_n$ together with possible emissions in between,

$$h_{[0,t)}(t_1,\ldots,t_n)=\mathrm{Tr}_A\{T_{t,t_n}J_1\cdots J_1T_{t,0}\rho_A(0)\}\ . \quad (27)$$

In particular the conditional probability that given a photon is emitted at t_0 another (not necessarily the next) emission occurs at t_1 is, using Eq. (22), given by

$$c(t_1\mid t_0)=\frac{\mathrm{Tr}_A\{J_1T_{t_1,t_0}J_1T_{t_0,0}\rho_A(0)\}}{\mathrm{Tr}_A\{J_1T_{t_0,0}\rho_A(0)\}} =\kappa_1U_{11,00}(t_1,t_0)\ , \quad (28)$$

which in the case of resonance fluorescence is just the intensity correlation function calculated when studying antibunching.[11] Equation (28) agrees with the result obtained with the help of the quantum fluctuation regression theorem.

B. Three-level theory

Finally, we turn to generalizing the foregoing discussion to describe the photon statistics of resonance fluorescence for the three-level system of Fig. 1(a). To the extent the atomic transition frequencies ω_{10} and ω_{20} are widely separated, it is meaningful to define separate photon count distributions for both photons emitted on the strong and weak transition line. In the following we focus on photon emission probabilities of the strong transition $|1\rangle$-$|0\rangle$ which are quantities of immediate physical interest. Following our treatment in the two-level case we define $P^{(n)}(t)$ as the probability that exactly $n=0,1,\ldots$ photons are emitted on the strong transition line. An expression for $P^{(n)}(t)$ can be derived by again defining a reduced atomic density matrix as in Eq. (6) with the exception that $P^{(n)}$ is a projection operator on a frequency band around ω_{10} which is broad compared with the characteristic frequencies associated with the transition $|0\rangle$-$|1\rangle$ (the Rabi frequency, the spontaneous decay rate and detuning of the laser) but much smaller than $\omega_{10}-\omega_{20}$, the difference between the atomic transition frequencies (Mathematically we interpret this as introducing two heat baths of radiation modes, one for the first and one for the second transition line.) We obtain the equation of motion

$$\frac{d}{dt}\rho_A^{(n)}=i(H_{\text{eff}}\rho_a^{(n)}-\rho_A^{(n)}H_{\text{eff}}^{\dagger}) +J_2\rho_A^{(n)}+J_1\rho_A^{(n-1)}(1-\delta_{n0})\ , \quad (29)$$

where

$$H_{\text{eff}}=\sum_{j=1}^{2}(\omega_{j0}-\tfrac{1}{2}\kappa_j)a_j^{\dagger}a_j -\sum_{j=1}^{2}[\mu_{j0}\mathscr{E}_{cj}(t)e^{-i\omega_j t}a_j^{\dagger}+\text{H.c.}] \quad (30)$$

is an effective Hamiltonian with $a_j=|0\rangle\langle j|$ the atomic lowering operators, κ_j the spontaneous decay rates, and μ_{j0} the atomic dipole matrix elements; $\mathscr{E}_{cj}(t)$ are (slowly varying) laser amplitudes and ω_j laser frequencies for the first and second transition, respectively ($j=1,2$). The operators J_j are defined by

$$J_j\rho_A^{(n)}=\kappa_j a_j\rho_A^{(n)}a_j^{\dagger}\equiv\kappa_j|0\rangle\langle 0|\rho_{jj}^{(n)}\quad (j=1,2) \quad (31)$$

and describe the collapse of the atomic density operator to a pure ground state $|0\rangle\langle 0|$ following the emission of a photon. Note that the source term $J_1\rho_A^{(n-1)}$ in Eq. (26) describes the creation of atoms in the ground state with n photons having been emitted on the transition $|1\rangle$-$|0\rangle$, while $J_2\rho_A^{(n)}$ corresponds to the recycling of the atomic electron to the ground state after emitting a photon on the weak transition (which, of course, does not change n, the number of photons emitted on the first line).

Equation (23) from our discussion of the two-level system remains valid in the present case except that the conditional densities $\tilde{c}(t\mid t_0)$ are now given by

$$\tilde{c}(t\mid t_0)=\kappa_1\tilde{\rho}_{11}(t\mid t_0) \tag{32}$$

with $\tilde{\rho}_{11}$ defined as the solution of the homogeneous part of Eq. (29), which for coherent driving fields $[\mathscr{E}_{cj}(t)=\text{const}]$ in the rotating frame has the form

$$\begin{aligned}
&\frac{d}{dt}\tilde{\rho}_{00}=\kappa_2\tilde{\rho}_{22}+\tfrac{1}{2}i\sum_{j=1}^{2}\Omega_j\tilde{\rho}_{j0}+\text{c.c.}\;,\\
&\frac{d}{dt}\tilde{\rho}_{jj}=-\kappa_j\tilde{\rho}_{jj}-\tfrac{1}{2}i\Omega_j\tilde{\rho}_{j0}+\text{c.c.}\quad (j=1,2)\;,\\
&\frac{d}{dt}\tilde{\rho}_{10}=(i\Delta_1-\tfrac{1}{2}\kappa_1)\tilde{\rho}_{10}+\tfrac{1}{2}i\Omega_1(\tilde{\rho}_{00}-\tilde{\rho}_{11})-\tfrac{1}{2}i\Omega_2\tilde{\rho}_{12}\;,\\
&\frac{d}{dt}\tilde{\rho}_{20}=(i\Delta_2-\tfrac{1}{2}\kappa_2)\tilde{\rho}_{20}+\tfrac{1}{2}i\Omega_2(\tilde{\rho}_{00}-\tilde{\rho}_{22})-\tfrac{1}{2}i\Omega_1\tilde{\rho}_{21}\;,\\
&\frac{d}{dt}\tilde{\rho}_{12}=(i\Delta_1-i\Delta_2-\tfrac{1}{2}\kappa_1-\tfrac{1}{2}\kappa_2)\tilde{\rho}_{12}-\tfrac{1}{2}i\Omega_2\tilde{\rho}_{10}+\tfrac{1}{2}i\Omega_1\tilde{\rho}_{02}
\end{aligned} \tag{33}$$

with $\tilde{\rho}_{\mu\nu}(t\mid t_0)\equiv\tilde{\rho}_{\mu\nu}(t-t_0)$ satisfying the initial condition $\tilde{\rho}_{\mu\nu}(0)=\delta_{\mu 0}\delta_{\nu 0}$. Here Ω_1,Ω_2 and Δ_1,Δ_2 are the Rabi frequencies and laser detunings for the first and second transitions, respectively. Note that Eq. (33) differs from the usual density matrix equation for a three-level system only by the missing "recycling" term $\kappa_2\tilde{\rho}_{22}$ on the right-hand side of the equation for $\tilde{\rho}_{00}$. From Eqs. (32) and (33) it follows that $\tilde{c}(t\mid t_0)\equiv\tilde{c}(t-t_0)$ is positive ($\tilde{c}\geq 0$) and normalized since

$$\begin{aligned}
\int_0^t d\tau\,\tilde{c}(\tau)&=1-\sum_{j=0}^{2}\tilde{\rho}_{jj}(t)\\
&\equiv 1-\mathrm{Tr}_A\{\tilde{\rho}_A(t)\}=x\in[0,1]\;.
\end{aligned} \tag{34}$$

According to Eqs. (32) and (33) the conditional density has the form

$$\tilde{c}(t\mid 0)=\sum_{j=1}^{9}A_j\lambda_j e^{-\lambda_j t}\quad\left[\sum_{j=1}^{9}A_j=1\right]\;, \tag{35}$$

where the coefficients A_j and the eigenvalues λ_j follow from the solution of Eq. (33).

A finite bandwidth b_1 and b_2 of the first and second laser according to the phase diffusion model is easily incorporated in the present calculation.[12] It can be shown that an ensemble average over phase fluctuations (indicated by the angular brackets) of the elementary probability density $p_{[0,t)}$ again leads to an equation of the form (26) with the conditional densities $\tilde{c}$ replaced by $\langle\tilde{c}(t\mid t_0)\rangle=\kappa_1\langle\tilde{\rho}_{11}(t\mid t_0)\rangle$. The averages $\langle\tilde{\rho}_{\mu\nu}(t\mid t_0)\rangle$ obey the system of equations obtained from Eq. (33) with the substitutions $\frac{1}{2}\kappa_1\to\frac{1}{2}\kappa_1+b_1$, $\frac{1}{2}\kappa_2\to\frac{1}{2}\kappa_2+b_2$, and $\frac{1}{2}\kappa_1+\frac{1}{2}\kappa_2\to\frac{1}{2}\kappa_1+\frac{1}{2}\kappa_1+b_1+b_2$. In the large-bandwidth limit an adiabatic elimination of the off-diagonal elements becomes possible which allows a reduction of the system (33) to a set of rate equations for the average populations.

III. QUANTUM JUMPS IN THREE-LEVEL SYSTEMS

In this section we explicitly calculate and interpret the conditional probability density $\tilde{c}(t\mid t_0)$ and the elementary probability density $p_{[0,t)}$ for the three-level systems shown in Fig. 1. Our starting point is Eqs. (32) and (33). To obtain physical insight we concentrate on two limiting cases where simple analytical solutions are possible: (i) reduction of the system (33) to a set of rate equations in the limit of a broad-bandwidth field for the weak transition and strong saturation of the first transition and (ii) coherent excitation for both transitions.

A. Incoherent excitation

1. V system

Following Cook and Kimble, a simple analytical treatment is possible in the limit where the transition $|0\rangle$-$|1\rangle$ is strongly saturated and the weak excitation of $|2\rangle$ is incoherent with a transition rate W_{02} (assuming $b_2\gg\Omega_1$). We define $\mathscr{P}_-^{(n)}=\tilde{\rho}_{00}^{(n)}(t)+\tilde{\rho}_{11}^{(n)}(t)$ and $\mathscr{P}_+^{(n)}=\tilde{\rho}_{22}^{(n)}(t)$ as the probabilities that n photons have been emitted on the atomic transition $|1\rangle$-$|0\rangle$ and the electron is in the two-level system $\{|0\rangle,|1\rangle\}$ or state $|2\rangle$, respectively. From Eq. (29) we derive

$$\frac{d}{dt}\mathscr{P}_+^{(n)}=-R_-\mathscr{P}_+^{(n)}+-R_+\mathscr{P}_-^{(n)}\;, \tag{36a}$$

$$\frac{d}{dt}\mathscr{P}_-^{(n)}=R_-\mathscr{P}_+^{(n)}-(\tfrac{1}{2}\kappa_1+R_+)\mathscr{P}_-^{(n)}+\tfrac{1}{2}\kappa_1\mathscr{P}_-^{(n-1)}(1-\delta_{n0}) \tag{36b}$$

with $R_+=\frac{1}{2}W_{02}$ the excitation rate $\{|0\rangle,|1\rangle\}$-$|2\rangle$ and $R_-=\kappa_2+W_{02}$ the decay rate from the metastable state back to the active two-level system. Note that

$$P^{(n)}(t)=\mathscr{P}_+^{(n)}(t)+\mathscr{P}_-^{(n)}(t) \tag{37}$$

is the probability of finding n scattered photons of frequency $\approx\omega_{10}$ in the field. The derivation of Eq. (36) implies that $1/\kappa_1$, $1/R_\pm$, and $1/\kappa_2$ are the slowest time scales in our problem.

Summing over n in Eq. (36) we obtain the rate equations

$$\frac{d}{dt}\mathscr{P}_+(t)=-R_-\mathscr{P}_+(t)+R_+\mathscr{P}_-(t)\;, \tag{38a}$$

$$\frac{d}{dt}\mathscr{P}_-(t)=R_-\mathscr{P}_+(t)-R_+\mathscr{P}_-(t)\;, \tag{38b}$$

where $\mathscr{P}_\pm$ are the occupation numbers of the electron in the system $\{|0\rangle,|1\rangle\}$ and state $|2\rangle$, respectively. Note that Eq. (38) is independent of κ_1.

To assist in the interpretation of Eq. (36) we solve this equation with the result

$$\mathscr{P}_\pm^{(0)}(t)=\tilde{p}_{\pm -}(t) \tag{39a}$$

for $n=0$ and

$$\mathscr{P}_\pm^{(n)}(t)=\int_0^t dt'\,\tilde{p}_{\pm -}(t-t')\tfrac{1}{2}\kappa_1\mathscr{P}_-^{(n-1)}(t') \tag{39b}$$

for $n=1,2,\ldots$. Here $\tilde{p}_{\pm -}(t)$ are solutions of the homogeneous part of Eq. (36) [compare Eq. (33)],

$$\frac{d}{dt}\tilde{p}_{+-}(t)=-R_-\tilde{p}_{+-}+R_+\tilde{p}_{--}(t)\;, \tag{40a}$$

$$\frac{d}{dt}\tilde{p}_{--}(t)=-R_-\tilde{p}_{+-}(t)-(\tfrac{1}{2}\kappa_1+R_+)\tilde{p}_{--}(t) \tag{40b}$$

with the initial conditions $\tilde{p}_{--}(0)=1$ and $\tilde{p}_{+-}(0)=0$. We identify $\tilde{p}_{+-}(t-t')$ with the probability that the electron—starting at time t' in the system $\{|0\rangle,1\rangle\}$—is excited to state $|2\rangle$ and has not yet emitted another photon on the strong transition line; $\tilde{p}_{--}(t-t')$ is the analogous probability for the electron staying in $\{|0\rangle,|1\rangle\}$. Thus we interpret Eq. (39) as describing the creation of atoms in the (ground state of the) active two-level system at a rate $\frac{1}{2}\kappa_1\mathcal{P}_-^{(n-1)}$ at time t', which is subsequently redistributed by the incident field between $\{|0\rangle,|1\rangle\}$ and $|2\rangle$.

Moments of the photon numbers are readily calculated generating from Eq. (36) by defining generation functions. In particular we derive the intuitively obvious result

$$\frac{d}{dt}\langle n\rangle\equiv\sum_{n=0}^{\infty}n\frac{d}{dt}P^{(n)}(t)=\tfrac{1}{2}\kappa_1R_-/(R_++R_-)\equiv\kappa_1\bar{\rho}_{11} \tag{41}$$

for the mean emission rate $|1\rangle$-$|0\rangle$ with $\bar{\rho}_{11}=\frac{1}{2}\mathcal{P}_-$ the population of $|1\rangle$. In a similar way we can show

$$\Delta n^2/\langle n\rangle\equiv(\langle n^2\rangle-\langle n\rangle^2)/\langle n\rangle=1 \tag{42}$$

which indicates that there are no deviations from Poissonian statistics in the rate-equation approximation.

Following analogous arguments to those given in Sec. II leading there to Eqs. (17) and (19) we find the corresponding similar expressions

$$\tilde{p}_{[0,t)}(t_1,\ldots,t_n)=[\tilde{p}_{+-}(t-t_n)+\tilde{p}_{--}(t-t_n)]\times\prod_{r=1}^{n}\tfrac{1}{2}\kappa_1\tilde{p}_{--}(t_r\,|\,t_{r-1}) \tag{43}$$

and

$$\tilde{c}(\tau)=\tfrac{1}{2}\kappa_1\tilde{p}_{--}(\tau)\ . \tag{44a}$$

Again following the reasoning of Sec. II, it is easy to prove that $\tilde{c}(\tau)$ as given by (44a) is a genuine normalized conditional probability, too, and that the Markov property (26) is still valid. Inserting the solution $p_{--}(\tau)$ we find

$$\tilde{c}(\tau)=A\lambda_+e^{-\lambda_+\tau}+B\lambda_-e^{-\lambda_-\tau} \tag{44b}$$

with

$$A=1-B=[\lambda_+-(R_++R_-)]/(\lambda_+-\lambda_-)\quad(A,B\geq 0) \tag{44c}$$

and

$$\lambda_\pm=\tfrac{1}{2}(R_++R_-+\tfrac{1}{2}\kappa_1)\pm[\tfrac{1}{4}(R_++R_-+\tfrac{1}{2}\kappa_1)^2-R_-\tfrac{1}{2}\kappa_1]^{1/2}\ , \tag{44d}$$

which clearly is of the form (35). The mean time between the successive photon emissions is

$$\tau_1=\int_0^\infty d\tau\,\tau\tilde{c}(\tau)=1/(\kappa_1\bar{\rho}_{11}) \tag{45}$$

which is just the inverse mean emission rate (41) for $W_{02}=0$ (no excitation of level $|2\rangle$) we have $\tilde{c}(\tau)=\frac{1}{2}\kappa_1e^{-\kappa_1\tau/2}$; this is the expected exponential decay law with a rate $\kappa_1\bar{\rho}_{11}=\frac{1}{2}\kappa_1$. Note that the antibunching property of the fluorescence light [which implies $\tilde{c}(0)=0$, see Ref. 11] is lost in the present approximation which assumes that the populations of $|0\rangle$ and $|1\rangle$ equalize instantaneously, after an emission event has prepared the electron in the ground state.

The remarkable feature $\tilde{c}(\tau)$ is that it is the sum of two exponential functions. Of particular interest is the case when the two decay rates λ_+ and λ_- differ substantially. This happens, for example, when $\kappa_1\gg R_\pm$, so that

$$\lambda_+\approx\tfrac{1}{2}\kappa_1+R_+,\quad\lambda_-\approx R_-\ ,\qquad B=1-A=R_+/\tfrac{1}{2}\kappa_1 \tag{46}$$

to lowest order in $R_\pm/\frac{1}{2}\kappa_1$. The fast decay rate $\frac{1}{2}\kappa_1$ is associated with the rapid emission of photons on the transition $|1\rangle$-$|0\rangle$. The slow rate R_- is the sum of the spontaneous and induced emission rates $|2\rangle$-$|0\rangle$. As we show below, it is the presence of the slowly decaying exponential function in $\tilde{c}(\tau)$ [Eq. (44b)] which is responsible for the appearance of emission windows on the strong transition line.

In Fig. 2 we plot the logarithm of $\tilde{c}(\tau)$ as a function of $\frac{1}{2}\kappa_1\tau$ for $W_{02}=0$ (curve a), $\kappa_2=W_{02}=10^{-1}\kappa_1$ (curve b), and $\kappa_2=W_{02}=10^{-2}\kappa_1$ with $\kappa_1=2$ (curve c). In Fig. 3 the logarithm of the quantity

$$\int_\tau^\infty\tilde{c}(\tau')d\tau'=\tilde{p}_{--}(\tau)+\tilde{p}_{+-}(\tau)=Ae^{-\lambda_+\tau}+Be^{-\lambda_-\tau}\equiv|x\in[0,1]| \tag{47}$$

which is the probability that the emission time is in the interval $[\tau,\infty)$ (i.e., longer than τ) is plotted for the same parameters. In both figures the existence of two decay rates is clearly apparent.

According to Eq. (47) the probability of decay in the time interval $[\tau,\infty)$ equals the population left at time τ in the three-level system, $\tilde{p}_{--}(\tau)+\tilde{p}_{+-}(\tau)$. We can define

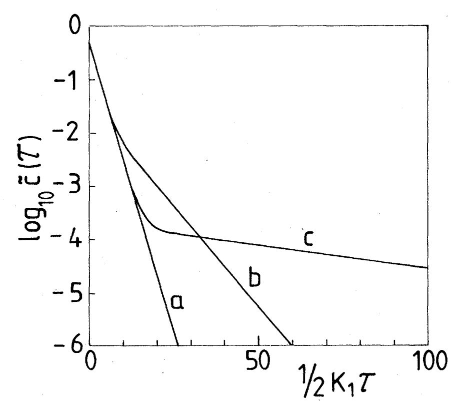

FIG. 2. The conditional probability density $c(\tau)$ that—given a count has occurred at time $\tau=0$—the *next* one will occur at time τ is plotted as a function of $\kappa_1\tau$ in a logarithmic scale for the parameters $W_{02}=\kappa_2=0$ (curve a), $W_{02}=\kappa_2=10^{-1}\kappa_1$ (curve b), and $W_{02}=\kappa_2=10^{-2}\kappa_1$ (curve c).

a critical decay time τ_c,

$$\tau_c = \ln(A/B)/(\lambda_+ - \lambda_-) \approx 2\ln(\kappa_1/2R_+)/\kappa_1 , \quad (48)$$

which marks the transition region in Fig. 3 which is characterized by λ_+ and λ_-, respectively. For $\lambda_+ \gg \lambda_-$ we, therefore, identify the probability that an *emission window* of duration longer than τ_c occurs with the *population trapped in the metastable state* $|2\rangle$:

$$\int_\tau^\infty \tilde{c}(\tau')d\tau' \approx \tilde{p}_{+-}(\tau) \quad (\tau \gg \tau_c) . \quad (49)$$

On the basis of Eq. (47) we can calculate samples of photon emission data of a single atom. Given a sequence of random numbers x which are uniformly distributed in the interval [0,1], we obtain a sequence of random decay times according to Eq. (47). We may visualize this process of simulating decay times graphically in Fig. 3 by choosing a sequence of uniform random numbers on the ordinate and reading off the corresponding decay times from the x axis. Figure 4 shows examples of such numerical simulations which clearly demonstrate the existence of emission windows, i.e., that bright and dark periods of light emissions alternate.

The definition of on and off times of the light in Fig. 4 is meaningful only to the extent the decay times λ_+ and λ_- differ significantly. For $\kappa_1 \gg R_\pm$ we may identify $A \approx 1$ and $B \ll 1$ in Eq. (44) as the probability that—given a photon has been emitted at time $\tau=0$—the next photon will decay according to the exponential decay laws $\frac{1}{2}\kappa_1\exp(-\frac{1}{2}\kappa_1\tau)$ or $R_-\exp(-R_-\tau)$, respectively. In other words, A and B are the probabilities for a fast decay as if from a strongly driven isolated two-level system $\{|0\rangle, |1\rangle\}$ or for the occurrence of an emission window, due to the electron being shelved in the metastable state $|2\rangle$, respectively. Clearly the probability distributions for windows of length τ is

$$P_{\text{off}}(\tau) = Be^{-R_-\tau} . \quad (50)$$

Since the occurrence of only one rapid decay according to

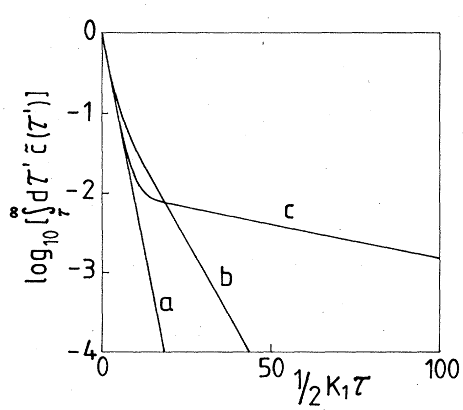

FIG. 3. Case of incoherent excitation: The probability that the time elapsed between two emissions lies in the interval $[\tau, \infty)$ is shown as a function of $\kappa_1\tau$ in a logarithmic scale for the parameters $W_{02}=\kappa_2=0$ (curve a), $W_{02}=\kappa_2=10^{-1}\kappa_1$ (curve b), and $W_{02}=\kappa_2=10^{-2}\kappa_1$ (curve c).

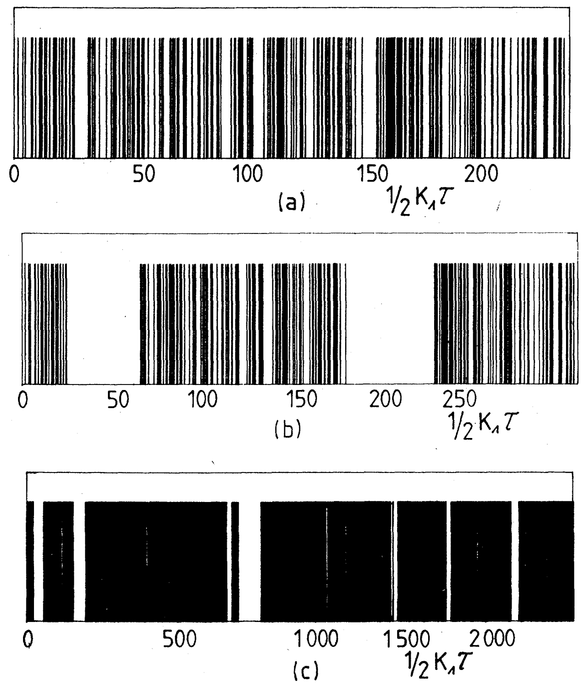

FIG. 4. Simulations of the photon emission on the transition $|0\rangle$-$|1\rangle$. A vertical line (of arbitrarily chosen height) corresponds to the emission of one photon. The number of simulations in the respective plots is denoted by n. (a) $n=250$; $\kappa_1=1$, $\kappa_2=0$, $W_{02}=0$, i.e., photon emission in absence of the metatable level $|2\rangle$. (b) $n=250$; $\kappa_1=1$, $\kappa_2=0.01$, $W_{02}=0.01$. (c) $n=2000$; $\kappa_1=1$, $\kappa_2=0.01$, $W_{02}=0.01$.

$\frac{1}{2}\kappa_1 e^{-\kappa_1\tau/2}$ cannot be a macroscopic "on period" of the fluorescence, one rather has to integrate all rapid decays with no emission window in between to form a unit, i.e., an on period of *overall* length τ. Making use of the elementary probability densities (43) we find, for the probability distribution for such on periods of duration τ,

$$P_{\text{on}}(\tau) = \sum_{n=0}^{\infty} \int_0^\tau dt_n \int_0^{t_n} dt_{n-1} \cdots \int_0^{t_2} dt_1 \tilde{p}_{[0,t)}(t_1, \ldots, t_n)_F , \quad (51)$$

where the index F ("fast") denotes the requirement that all time intervals between two photon counts have to be smaller than τ_c, i.e., that we have no window in between. Upon insertion of the approximate expression

$$\tilde{p}_{[0,\tau)}(t_1, \ldots, t_n)_F \approx Ae^{-\lambda_+\tau} \prod_{r=1}^{n} A\lambda_+ e^{-\lambda_+(t_r - t_{r-1})} \quad (52)$$

for $|t_r - t_{r-1}| < \tau_c$ and $r=1,\ldots,n$ the summing of the right-hand side of Eq. (51) yields the result

$$P_{\text{on}}(\tau) = Ae^{-B\lambda_+\tau} \approx Ae^{-R_+\tau} . \quad (53)$$

In the limit $\kappa_1 \gg R_\pm$ the probability distribution for durations τ among dark or light periods is thus given by

$$\mathscr{P}_{\rm off}(\tau)=e^{-R_-\tau} \text{ and } \mathscr{P}_{\rm on}(\tau)=e^{-R_+\tau}, \tag{54}$$

respectively, a result which agrees with the predictions of Cook and Kimble[1] which is based on the *assumption* that the on and off times are associated with the hopping (quantum jump) of the electron between $\{|0\rangle,|1\rangle\}$ and $|2\rangle$.

2. Λ system

Similar arguments can be given for the Λ system depicted in Fig. 1(b). Quantum jumps in this system have recently been discussed by Schenzle *et al.*,[2] Javanainen,[3] and Pegg *et al.*[4] The transition $|0\rangle$-$|1\rangle$ is again driven by a strong classical field $\mathscr{E}_{\rm cl}$. The level $|1\rangle$ decays spontaneously at a rate κ_{01} to the ground state or at a rate κ_{21} to a metastable state $|2\rangle$ which itself decays to the ground state at the rate κ_{02}. We assume κ_{01} to be much larger than both other decay rates.

In this case Eq. (29) has to be replaced by

$$\frac{d}{dt}\rho_A^{(n)}=i(H_{\rm eff}\rho_a^{(n)}-\rho_A^{(n)}H_{\rm eff}^\dagger)+J_{02}\rho_A^{(n)} +J_{21}\rho_A^{(n)}+J_{01}\rho_A^{(n-1)}(1-\delta_{n0}) \tag{55}$$

with

$$H_{\rm eff}=\sum_{\nu\,(\in I)}(\omega_\nu-\tfrac{1}{2}\kappa_\nu)a_\nu^\dagger a_\nu -[\mu_{01}\mathscr{E}_{\rm cl}(t)e^{-i\omega_1 t_a}a_{01}^\dagger+\text{H.c.}], \tag{56}$$

where $I=\{(0,1),(0,2),(2,1)\}$ in a notation similar to the one adopted before. An analogous procedure as for the V configuration, justified by a large Rabi frequency or a broadband field on the transition $|0\rangle$-$|1\rangle$ leads upon replacement of $R_\pm\to\hat{R}_\pm$ to the same rate equations for the probabilities $\mathscr{P}_\pm^{(n)}$ defined as before where

$$\hat{R}_+=\tfrac{1}{2}\kappa_{21} \text{ and } \hat{R}_+=\kappa_{02}. \tag{57}$$

Comparing $R_\pm$ and $\hat{R}_\pm$ we notice that, of course, they differ inasmuch as the transition $|0\rangle$-$|2\rangle$ is not driven any more, thus there is no depopulation of $|0\rangle$ to $|2\rangle$, but there is new loss rate $|1\rangle$-$|2\rangle$ at a rate κ_{21} instead. (Note that $\hat{R}_\pm\ll\kappa_{01}$.) With this simple replacement all conclusions drawn for the V configuration, in particular the existence of windows in the emission in the observed fluorescence of transition $|0\rangle$-$|1\rangle$, hold for the Λ system, too.

B. Coherent excitation

Quantum mechanics implies the existence of (coherent) superposition states of the atom. For incoherent excitation the fluctuations in the light field destroy the quantum-mechanical coherence. This is implict in the reduction of density-matrix-type equations to a system of rate equations for the atomic populations (Sec. III A). These rate equations can be interpreted in terms of a (classical) probability of finding the electron in one of the atomic states. The main result of the preceding section was the prediction of the existence of photon emission windows on the strong line for incoherent excitation which we identified with the electronic population being trapped in the metastable state $|2\rangle$. It is, therefore, interesting to investigate to what extent excitation by a coherent (monochromatic) field (which allows atomic coherences to develop) might change the conclusions of Sec. III A.

We consider again the case of a V configuration [Fig. 1(a)] where the first laser is tuned to exact resonance ($\Delta_1=0$), the Rabi frequency Ω_1 is much larger than κ_1, κ_2, and Ω_2 and both lasers are monochromatic. A large Rabi frequency Ω_1 implies fast Rabi oscillations of the atomic populations between levels $|0\rangle$ and $|1\rangle$. Due to the ac Stark splitting of the atomic transition line $|0\rangle$-$|1\rangle$ the population of the metastable state $|2\rangle$ as a function of Δ_2 will show peaks for $\Delta_2=\pm\frac{1}{2}\Omega_1$. This corresponds to tuning the second laser into resonance with one of the dressed eigenstates of the transition $|0\rangle$-$|1\rangle$. For large Ω_1 the time evolution of $\tilde{\rho}_{\mu\nu}(t)$ [Eq. (33)] is conveniently calculated in a basis of these dressed states. Defining dressed-state populations

$$\begin{aligned}\tilde{\rho}_{++}&=\tfrac{1}{2}(\tilde{\rho}_{00}+\tilde{\rho}_{01}+\tilde{\rho}_{10}+\tilde{\rho}_{11}),\\ \tilde{\rho}_{--}&=\tfrac{1}{2}(\tilde{\rho}_{00}-\tilde{\rho}_{01}-\tilde{\rho}_{10}+\tilde{\rho}_{11})\end{aligned} \tag{58a}$$

and coherences

$$\begin{aligned}\tilde{\rho}_{\pm2}&=2^{-1/2}(\tilde{\rho}_{02}\pm\tilde{\rho}_{12}),\\ \tilde{\rho}_{+-}&=\tfrac{1}{2}(\tilde{\rho}_{00}+\tilde{\rho}_{01}-\tilde{\rho}_{10}-\tilde{\rho}_{11})\end{aligned} \tag{58b}$$

we find that for $\Omega_1\gg\kappa_{1,2}$ and $\Delta_2\approx\frac{1}{2}\Omega_1$ the equations (33) decouple approximately according to

$$\begin{aligned}\frac{d}{dt}\tilde{\rho}_{++}&=-\tfrac{1}{2}\kappa_1\tilde{\rho}_{++}+i\tfrac{1}{2}\Omega_2'(\tilde{\rho}_{2+}-\tilde{\rho}_{+2}),\\ \frac{d}{dt}\tilde{\rho}_{+2}&=(-i\Delta_2'-\tfrac{1}{4}\kappa_1-\tfrac{1}{2}\kappa_2)\tilde{\rho}_{+2}+i\tfrac{1}{2}\Omega_2'(\tilde{\rho}_{22}-\tilde{\rho}_{++}),\\ \frac{d}{dt}\tilde{\rho}_{22}&=-\kappa_2\tilde{\rho}_{22}-i\tfrac{1}{2}\Omega_2'(\tilde{\rho}_{2+}-\tilde{\rho}_{+2})\end{aligned} \tag{59}$$

and

$$\frac{d}{dt}\tilde{\rho}_{--}=-\tfrac{1}{2}\kappa_1\tilde{\rho}_{--}, \tag{60}$$

$$\frac{d}{dt}\tilde{\rho}_{+-}=(i\Omega_1-\tfrac{1}{2}\kappa_1)\tilde{\rho}_{+-}. \tag{61}$$

Equations (59) describe the coupling of the resonant dressed-state component with level $|2\rangle$ with $\Delta_2'=\Delta_2-\frac{1}{2}\Omega_1$ and $\Omega_2'=\Omega_2/\sqrt{2}$.

For $\kappa_1\gg\Omega_2,\kappa_2$ and $\Delta_2'=0$ it is straightforward to integrate Eqs. (59)–(61). We obtain for the probability that the atom emits a photon in the time interval $[\tau,\infty)$ provided the last photon was emitted at time $\tau=0$,

$$\int_\tau^\infty \tilde{c}(\tau')d\tau'\approx e^{-\kappa_1\tau/2}-W'_{+2}/4\kappa_1 e^{-\kappa_1\tau/4} +W'_{+2}/\kappa_1 e^{-(\kappa_2+W'_{+2})\tau} \tag{62}$$

with $W'_{+2}=\frac{1}{4}\Omega_2^2/(\frac{1}{4}\kappa_1)$ an induced transition rate from the dressed state $|+\rangle$ to level $|2\rangle$. In Eq. (62) only the

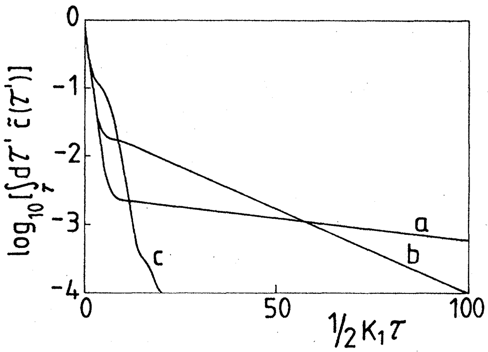

FIG. 5. Case of coherent excitation: The probability that the time elapsed between two emissions lies in the interval $[\tau,\infty)$ is plotted as a function of $\frac{1}{2}\kappa_1\tau$ on a logarithmic scale for the parameters $\kappa_1=2$, $\Omega_1=20$, $\Delta_1=0$, $\kappa_2=0.1$, $\Delta_2=0$, $\Omega_2=0.1$ (curve a), 0.3 (curve b), and 1 (curve c).

dominant (leading) terms of each of the prefactors and decay rates of each of the exponential functions according to a series expansion in $\kappa_{1,2}/\Omega_1 \ll 1$ has been written out; in addition we have dropped rapidly oscillating terms. Note that the last term in Eq. (62) is again a slowly decaying exponential function which is responsible for the appearance of emission windows even in the case of coherent excitation. Comparison with Eq. (47) shows that the first term in Eq. (60) is connected with the decay of the atomic population in $\{|0\rangle,|1\rangle\}$ while the second term [which was missing in Eq. (47)] is due to atomic coherences which are seen to *decay* with a rate $\kappa_1/4$. For long times $\kappa_1\tau \gg 1$ Eq. (54) implies

$$\int_\tau^\infty \tilde{c}(\tau')d\tau' \approx \tilde{\rho}_{22}(\tau) \quad (\kappa_1\tau \gg 1) \tag{63}$$

i.e., the appearance of emission windows is again related to the electron being trapped in the metastable state.

In Fig. 5 we show the probability (62) as a function of $\frac{1}{2}\kappa_1\tau$ for $\kappa_1=2$, $\Omega_1=20$, $\Delta_2=0$, $\kappa_2=0.01$, $\Delta_2'=0$, and $\Omega_2=0.1$ (curve a), 0.3 (curve b), and 1 (curve c). The curves were calculated numerically as solutions of Eq. (33) assuming that both lasers are monochromatic. The existence of a long-time tail in Fig. 5 is clearly visible.

IV. CONCLUSIONS

We have analyzed the photoemission from a three-level atom driven strongly on one transition and weakly on the other. The atom is taken to be in the V configuration and the excited state on the weak transition is assumed to be metastable. We give an analysis of the fluorescence from the strong transition in terms of the elementary probability density $p_{[0,t)}(t_1,\ldots,t_n)$ which gives the probability density that *exactly* n photons are emitted at times $t_1,\ldots,t_n$ by the atom in the time interval $[0,t)$. These techniques are equivalent to those of the continuous measurement theory developed by Srinivas and Davies.[10]

We have calculated the conditional probability density $\tilde{c}(\tau)$ that given a photon is emitted at time zero the *next* photon on the strong line is emitted at time τ. This is in contrast to calculations of $g^{(2)}(\tau)$, the conditional probability that given a photon is emitted at time zero another photon but not necessarily the next one is emitted at time τ.

The probability that the emission time is in the interval $[t,\infty)$ is then $\int_t^\infty d\tau\,\tilde{c}(\tau)=x$. On the basis of this relation we can calculate a sequence of photon emissions from a single atom. Given a sequence of random numbers $x_1,x_2,\ldots$ which are uniformly distributed in the interval [0,1] we obtain a sequence of random decay times $t_1,t_2,\ldots$. A plot of the individual photon emissions as a function of time shows directly the existence of prolonged dark windows in the fluorescence corresponding to the shelving of the electron in the metastable state or "quantum jumps."

The condition for the observation of prolonged dark windows in the fluorescence is that the decay rate on the strong transition greatly exceeds the decay and excitation rates of the weak transition. Similar conclusions hold for the atom in the Λ configuration. The dark windows in the emission also occur for coherent driving fields when the laser on the weak transition is detuned to compensate for the Rabi splitting of the levels.

Note added. After this work was completed we became aware of the paper by Cohen-Tannoudji and Dalibard.[13] They introduce for coherent excitation a conditional probability density for the "emission of the next photon" which is analogous to our conditional probability $\tilde{c}(\tau)$ for the "emission of the next photon on the strong line." A calculation of the complete photon count statistics has been reported in unpublished work by Schenzle and Brewer.[14] Recently, experimental evidence for the existence of quantum jumps has been reported by Nagourney *et al.*,[15] Sauter *et al.*,[16] and Bergquist *et al.*[17]

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