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Quantum stochastic differential equations: an application to the electron shelving effect

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Abstract. By means of a physically interesting example, it is shown how the mathematical theory of quantum stochastic calculus can be used in modelling physical systems. The example is the so-called electron shelving effect. The fluorescent light emitted by a single atom or ion presents periods of darkness if two transitions, one weak and one strong, are simultaneously driven. By a previously developed theory of counting processes, based on quantum stochastic differential equations, the full statistics of the emitted photons is obtained. In particular, the results on the duration of bright and dark periods, previously obtained by Cohen-Tannoudji and Dalibard, are completely confirmed.

1. Introduction

Quantum stochastic calculus (QSC) (see, for instance, [1-3]) was developed originally as a mathematical theory of quantum noise. Afterwards it was realised that it was also useful in other physical applications as in the treatment of input and output channels in quantum systems [4] and in measurement theory in quantum mechanics [5-9]. In the opinion of the writer, QSC is now not only a well developed mathematical theory, but also a flexible tool, very useful in modelling concrete physical systems. Indeed, the aim of this paper is to illustrate this last point, by showing how QSC can be used for treating a physically interesting phenomenon, the so-called *electron shelving effect*.

Today, experimental techniques have reached the point at which it is possible to observe the fluorescent light emitted by a single atom or ion. There is, therefore, the possibility of observing new effects which are completely masked when many emitters are involved. One of these phenomena (electron 'shelving') was proposed by Dehmelt as a very sensitive scheme for detecting very weak transitions in single ions [10, 11].

Consider a three-level atom. Following Cohen-Tannoudji and Dalibard [12], call the states $|g\rangle$ (ground), $|b\rangle$ (blue) and $|r\rangle$ (red) and the corresponding transitions the blue transition ($b \rightarrow g$) and the red one ($r \rightarrow g$). Assume the blue transition to be very strong and the red one very weak. When both transitions are driven by two suitably tuned lasers, we expect the atom to emit blue fluorescent light. But sometimes, when the atom absorbs a red photon, the electron goes into the red state, which has a long lifetime (a fraction of a second), and the fluorescent light stops until the red state decays. Thus we expect to observe bright and dark periods, randomly distributed. In a pictorial language, we say that during a dark period the electron is 'shelved' in the red state. Indeed, experimentalists have succeeded in observing this effect [13-15].

Various papers have been published on the theoretical explanation of this effect [12, 16–22], studying different aspects of the phenomenon and considering various physical situations (different level schemes, coherent–incoherent excitation, etc). For the case of the three-level structure discussed above and for coherent excitation, a good treatment is given by Cohen-Tannoudji and Dalibard [12].

From the theoretical point of view, the problem is to study the statistics of the photons emitted by a three-level system, excited by two coherent sources. A theory of counting processes, well based on measurement theory in quantum mechanics, was developed some time ago by Davies [23] (see also [24–26]). Later, this theory was reformulated [5, 7] by using qsc and the connections with other continuous-measurement schemes [8, 27–29] were exploited.

Other approaches to quantum counting processes have been proposed in the literature, based on perturbation theory and on analogies with classical counting formulae (of which [30] is a typical example). However, the counting formulae derived from these approaches lead to unphysical consequences, such as negative probabilities, if they are applied without regard to the approximations made in their derivations. This is essentially a consequence of the fact that the attenuation of the field due to the detector is not taken into account. These criticisms were raised, for instance, in [25, 26, 31]. For the case of a single-mode free field, by starting from explicit models for the detector, an alternative photon counting formula has been derived in [31–33] which is free from unphysical features. On the other hand, the theory developed in [5, 7, 23–26] always gives rise to well defined probabilities, is not model dependent and has a wide range of applicability. When this theory is particularised to the case of a single-mode free field the results of [31–33] are reobtained.

In this paper we show how the model of [12] can be restated in the language of qsc and how the theory of [5, 7] can be used for obtaining the counting probabilities. In this way the full statistics of the light emitted by the three-level system described above is obtained and, in particular, the length of bright and dark periods is calculated. The results and the physical explanation of the shelving effect given in [12] are completely confirmed by our computations. In this respect, let us stress that our main purpose is not to give new results, but to illustrate a previously developed theoretical formalism by means of a physically interesting example. We hope, however, that the methods presented here might contribute to a better understanding of the behaviour of the system and might help in the treatment of more general situations.

2. Quantum stochastic calculus

In this section, we review, at a very informal level, some of the results of Hudson and Parthasarathy [1–3]. We consider the simplest version of quantum stochastic calculus, namely that one based on the boson Fock space.

Consider N Bose fields $a_j(t)$, $a_j^\dagger(t)$, satisfying the canonical commutation relations

$$[a_j(t), a_i^\dagger(s)] = \delta_{ij}\delta(t-s) \quad [a_j(t), a_i(s)] = 0 \quad (2.1)$$

and choose the Fock representation. This means that the one-particle space is $h_1 \equiv \mathbb{C}^N \otimes L^2(\mathbb{R})$ and the Hilbert space on which the field operators act is the symmetric Fock space over h_1 . We denote the Fock space by Γ and the Fock vacuum by ψ_0 .

We then define the annihilation, creation and gauge (or number) processes by, respectively,

$$\begin{aligned}
 A_t^j &:= \int_0^t ds a_j(s) \\
 A_t^{j*} &:= \int_0^t ds a_j^*(s) \\
 \Lambda_t^j &:= \int_0^t ds a_j^*(s)a_j(s).
 \end{aligned}
 \tag{2.2}$$

A QSC of Itô type, based on the integrators dA_t^j , dA_t^{j*} , $d\Lambda_t^j$ and dt , has been developed by Hudson and Parthasarathy (see, for instance, [2]). The couples A_t^j, A_t^{j*} are non-commutative analogues of independent Wiener processes and the operators Λ_t^j are the main ingredients in the construction of quantum analogues of Poisson processes.

Quantum stochastic calculus obeys very simple formal rules, which can be summarised in the following way.

(a) The fundamental integrators ‘point into the future’, i.e.

$$dA_t^j := A_{t+dt}^j - A_t^j = \int_t^{t+dt} ds a_j(s), \dots$$

and commute with *adapted processes* (roughly speaking, an adapted process $M(t)$ is an operator-valued quantity depending on the fields $A_s^j, A_s^{j*}, \Lambda_s^j$ only for times s less than t (see [2] definition 3.1 and theorem 4.5)).

(b) The fundamental integrators satisfy simple *multiplication rules* (see [2] equation (7.1)):

$$\begin{aligned}
 dA_t^j dA_t^{i*} &= \delta_{ji} dt & dA_t^j d\Lambda_t^i &= \delta_{ji} dA_t^j \\
 d\Lambda_t^j dA_t^{i*} &= \delta_{ji} dA_t^{i*} & d\Lambda_t^j d\Lambda_t^i &= \delta_{ji} d\Lambda_t^j
 \end{aligned}
 \tag{2.3}$$

and all the other products involving $dA_t^j, dA_t^{j*}, d\Lambda_t^j$ and dt vanish.

The Hudson and Parthasarathy theory allows us, in particular, to give meaning to ‘stochastic Schrödinger equations’. Let h_0 be an Hilbert space (representing the Hilbert space of some system S) and consider the following quantum stochastic differential equation (QSDE) for operators in $h_0 \otimes \Gamma$:

$$dU_t = \left(\sum_j (-R_j^\dagger dA_t^j + R_j dA_t^{j*} - \frac{1}{2} R_j^\dagger R_j dt) - iH dt \right) U_t \tag{2.4a}$$

$$U_0 = \mathbb{1} \tag{2.4b}$$

where $R_j, H \in \mathcal{B}(h_0)$ (bounded operators on h_0) and $H = H^\dagger$. Here and in the following we identify R_j with $R_j \otimes \mathbb{1}$, A_t^j with $\mathbb{1} \otimes A_t^j$, and so on. The solution U_t of this equation exists and is unique; $\{U_t, t \geq 0\}$ is an adapted process and, for any $t \geq 0$, U_t is a unitary operator on $h_0 \otimes \Gamma$ (see [2], § 7). Note that, since U_t is an adapted process and by rule (a) above, we have that $[U_t, dA_t^j] = [U_t, dA_t^{j*}] = 0$.

The formal solution of (2.4) can be written as [7]

$$U_t = \tilde{T} \exp \int_0^t \left(-iH ds + \sum_j (-R_j^\dagger dA_s^j + R_j dA_s^{j*}) \right) \tag{2.5}$$

where \bar{T} denotes the time-ordered product. Indeed, this equation gives

$$dU_t \equiv U_{t+dt} - U_t = \left[\exp\left(-iH dt + \sum_j (-R_j^\dagger dA_t^j + R_j dA_t^{j\dagger})\right) - 1 \right] U_t$$

$$= \sum_{n=1}^\infty \frac{1}{n!} \left(-iH dt + \sum_j (-R_j^\dagger dA_t^j + R_j dA_t^{j\dagger})\right)^n U_t.$$

Up to order dt , the terms with $n > 2$ vanish, the term with $n = 1$ gives $-iH dt + \sum(-R_j^\dagger dA_t^j + R_j dA_t^{j\dagger})$ and, by the multiplication rules (2.3), the term with $n = 2$ gives $-\frac{1}{2} \sum R_j^\dagger R_j dt$. Therefore, (2.4a) is recovered; from this and the fact that U_t , as given by (2.5), satisfies the initial condition (2.4b), then (2.5) gives the unique solution of (2.4).

From expression (2.5) the unitarity of U_t is apparent. The quantity U_t can be interpreted as the evolution operator for the system S interacting with the fields $a_j(t)$, in the interaction picture with respect to the free dynamics of the fields. The operator H represents the ‘free’ Hamiltonian for system S and the expression $i\sum(R_j dA_t^{j\dagger} - R_j^\dagger dA_t^j)$ gives the interaction between system S and fields. The term $-\frac{1}{2}\sum R_j^\dagger R_j dt$ is a correction term due to the fact that we are using a stochastic calculus of Itô type; it appears only in differential equation (2.4a) and not in its solution (2.5). Formally, it is possible to introduce also a QCS of Stratonovich type [4]; in this case no correction term would also appear in the differential equation for the evolution operator U_t .

Let ρ be the initial state for system S (ρ is a statistical operator on h_0) and let the initial state for the fields be the Fock vacuum. The expression

$$\rho(t) := \text{Tr}_r\{U_t(\rho \otimes |\psi_0\rangle\langle\psi_0|)U_t^\dagger\}$$

(where Tr_r is the partial trace over Fock space) represents the reduced dynamics of system S . It can be shown [2-4, 7] that $\rho(t)$ satisfies exactly the *quantum master equation*

$$(d/dt)\rho(t) = L\rho(t) \tag{2.6}$$

$$L\rho = -i[H, \rho] + \frac{1}{2} \sum_j ([R_j, \rho R_j^\dagger] + [R_j \rho, R_j^\dagger]). \tag{2.7}$$

Another important result is the so-called *quantum regression theorem*, which says that certain time-ordered multitime correlation functions for system operators can be re-expressed by means of the reduced dynamics $\exp(Lt)$ (see [4] § IV.D).

In a pictorial language, what we have done in this section is the following. We have an open system S interacting with its environment. To describe this situation we make certain approximations. First, the physical environment is replaced by some Bose fields $a_j(t)$. Then, the interaction between system S and fields is chosen to be so singular that, when the degrees of freedom of the fields are eliminated, the reduced dynamics of system S exactly obeys a quantum master equation. Moreover, the quantum regression theorem holds, not as an approximation, but as an exact result. The approximations usually needed for eliminating the ‘memory’ terms from the equation for the reduced dynamics are made directly on the dynamics of the total system (system S and environment). Quantum stochastic calculus is the mathematical theory that allows us to give a rigorous meaning to the resulting equations for the dynamics of the total system.

3. The model

Consider a three-level system driven by two lasers. The laser fields are treated classically. With respect to the notations of the introduction, we denote by $|0\rangle$ the ground

state $|g\rangle$, by $|1\rangle$ the blue excited state $|b\rangle$ and by $|2\rangle$ the red excited state $|r\rangle$. In the rotating-wave approximation and after removing the explicit time dependence, the Hamiltonian of the system is given by

$$H = \sum_{\nu=1}^2 [\frac{1}{2}\Omega_{\nu}(|0\rangle\langle\nu| + |\nu\rangle\langle 0|) - |\nu\rangle\Delta_{\nu}\langle\nu|] \tag{3.1}$$

where the quantities Ω_{ν} ($\Omega_{\nu} > 0$) are called Rabi frequencies and the quantities Δ_{ν} are called detuning parameters (cf [12]).

We now have to introduce the possibility of emission of fluorescent light. Our strategy will be to replace the true electromagnetic field by the Bose fields $a_j(t)$ introduced in the previous section and to describe the dynamics of the system atom-fields by a QSDE of the type (2.4a). The physical approximations that allow this replacement are essentially: (i) the interaction atom-field is taken to be linear in the field operators; (ii) the rotating-wave approximation is made; (iii) the field spectrum is assumed flat and the coupling constant is independent of frequency (see [4] p 3762).

In the interaction picture with respect to the free dynamics of the electromagnetic field and in the dipole approximation, the interaction between an atom and the electromagnetic field is $-e\mathbf{r} \cdot \mathbf{E}(\mathbf{r}, t)$, where \mathbf{r} is the position of the electron and \mathbf{E} is the quantum electric field (approximation (i) above).

If the atomic transition frequencies are well separated, any transition is essentially coupled to different modes of the field and can be treated independently from the others. Consider the $|0\rangle \rightleftharpoons |1\rangle$ transition and let $\psi_{\nu}(\mathbf{r})$, $\nu = 0, 1$, be the wavefunctions of the two states. In the rotating-wave approximation ((ii) above), the interaction Hamiltonian becomes

$$H_{10} = -e|1\rangle \int d^3\mathbf{r} \psi_1^*(\mathbf{r}) \mathbf{r} \cdot \mathbf{E}^+(\mathbf{r}, t) \psi_0(\mathbf{r}) \langle 0| + \text{HC}$$

where $\mathbf{E}^+(\mathbf{r}, t)$ is the positive-frequency electric field operator. Then, let $b_j(\omega)$ be a set of modal annihilation operators, such that $[b_j(\omega), b_i^{\dagger}(\omega')] = \delta_{ij}\delta(\omega - \omega')$, where $\omega \geq 0$ is a continuous index representing energy and j a discrete index. For instance, by using an expansion in spherical harmonics, j is the triple (l, m, σ) , where $\sigma = 1, 2$ is a polarisation index. By using these operators, $\mathbf{E}^+(\mathbf{r}, t)$ can be written as

$$\mathbf{E}^+(\mathbf{r}, t) = \sum_j \frac{1}{\sqrt{2\pi}} \int_0^{+\infty} d\omega \mathbf{F}_j(\mathbf{r}; \omega) e^{-i\omega t} b_j(\omega). \tag{3.2}$$

The explicit expression of the coefficients $\mathbf{F}_j(\mathbf{r}; \omega)$ is not important here. By setting

$$f_j^{10}(\omega) := -e \int d^3\mathbf{r} \psi_1^*(\mathbf{r}) \mathbf{r} \cdot \mathbf{F}_j(\mathbf{r}; \omega) \psi_0(\mathbf{r})$$

the interaction Hamiltonian becomes

$$H_{10} = |1\rangle \sum_j \frac{1}{\sqrt{2\pi}} \int_0^{+\infty} d\omega e^{-i\omega t} f_j^{10}(\omega) b_j(\omega) \langle 0| + \text{HC}$$

and, by going into the interaction picture with respect to the free dynamics of the atom, we obtain

$$H_{10} = |1\rangle \sum_j \frac{1}{\sqrt{2\pi}} \int_{-\omega_1}^{+\infty} d\omega e^{-i\omega t} f_j^{10}(\omega + \omega_1) b_j(\omega + \omega_1) \langle 0| + \text{HC}$$

where ω_1 is the frequency of the considered transition.

Now, the approximation (iii) above consists of taking $f_j^{10}(\omega)$ constant in a neighbourhood of $\omega = \omega_1$ and zero elsewhere, so that we can replace $f_j^{10}(\omega + \omega_1)$ by $f_j^{10}(\omega_1)$ (coupling constant independent of frequency). Then we let the interval of integration go from $-\infty$ to $+\infty$ (flat spectrum of the field). In this way, the interaction Hamiltonian becomes

$$H_{10} = |1\rangle\langle 0| \sum_j f_j^{10}(\omega_1) a_j(t) + \text{HC}$$

where

$$a_j(t) := \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} d\omega e^{-i\omega t} b_j(\omega + \omega_1).$$

It turns out that the fields $a_j(t)$ satisfy the commutation rules (2.1). This shows that, in the approximations considered, the atom-field interaction is of the type of the interaction term appearing in (2.5) and (2.4a).

Summing up, the evolution operator for our system is given by (2.5) and the evolution equation by (2.4a), where H is given by (3.1) and

$$\begin{aligned} R_j &= \lambda_j |0\rangle\langle 1| & j \in I_1 \\ R_j &= \lambda_j |0\rangle\langle 2| & j \in I_2. \end{aligned} \tag{3.3}$$

Here we have split the field modes into two disjoint sets I_1 and I_2 , due to the fact that the two transitions are coupled to essentially independent modes of the field.

The previous comments are intended to give a rough justification of the use of (2.4a), (3.1) and (3.3) for describing the dynamics of our system (atom and electromagnetic field). However, to convince oneself that the approximations introduced are sensible, the best way is to consider the reduced dynamics of the atom and to show that it is given by the usual Bloch equations. Indeed, as we have seen, (2.4a) implies exactly the master equation (2.6). In our case the Liouville operator (2.7) turns out to be

$$L\rho = -i\tilde{H}\rho + i\rho\tilde{H}^\dagger + |0\rangle\left(\sum_{\nu=1}^2 \Gamma_\nu \langle \nu | \rho | \nu \rangle\right)\langle 0| \tag{3.4}$$

where

$$\begin{aligned} \tilde{H} &:= H - \frac{1}{2}i \sum_{\nu=1}^2 |\nu\rangle\Gamma_\nu\langle \nu| \\ &\equiv \sum_{\nu=1}^2 \left[\frac{1}{2}i\Omega_\nu (|0\rangle\langle \nu| + |\nu\rangle\langle 0|) - |\nu\rangle(\Delta_\nu + \frac{1}{2}i\Gamma_\nu)\langle \nu| \right] \end{aligned} \tag{3.5}$$

$$\Gamma_\nu := \sum_{j \in I_\nu} |\lambda_j|^2 \quad \nu = 1, 2. \tag{3.6}$$

In terms of the matrix elements $\rho_{\nu\eta} := \langle \nu | \rho | \eta \rangle = \rho_{\eta\nu}^*$, (2.6) becomes

$$\begin{aligned} \dot{\rho}_{00} &= \sum_{\nu=1}^2 \left[\frac{1}{2}i\Omega_\nu (\rho_{0\nu} - \rho_{0\nu}^*) + \Gamma_\nu \rho_{\nu\nu} \right] \\ \dot{\rho}_{\nu\nu} &= -\frac{1}{2}i\Omega_\nu (\rho_{0\nu} - \rho_{0\nu}^*) - \Gamma_\nu \rho_{\nu\nu} \quad \nu = 1, 2 \\ \dot{\rho}_{01} &= \frac{1}{2}i\Omega_1 (\rho_{00} - \rho_{11}) - \frac{1}{2}i\Omega_2 \rho_{12}^* - (\frac{1}{2}\Gamma_1 + i\Delta_1)\rho_{01} \\ \dot{\rho}_{02} &= \frac{1}{2}i\Omega_2 (\rho_{00} - \rho_{22}) - \frac{1}{2}i\Omega_1 \rho_{12} - (\frac{1}{2}\Gamma_2 + i\Delta_2)\rho_{02} \\ \dot{\rho}_{12} &= -\frac{1}{2}i\Omega_2 \rho_{02} + \frac{1}{2}i\Omega_2 \rho_{01}^* + [i(\Delta_1 - \Delta_2) - \frac{1}{2}(\Gamma_1 + \Gamma_2)]\rho_{12}. \end{aligned}$$

These equations show that the reduced dynamics of the atom is the usual one and, in particular, that the quantities Γ_ν represent the total transition rates for the blue and the red transitions. In the following we shall see that we do not need to solve these equations for studying the characteristics of bright and dark periods.

Finally, let us stress that it is also possible to treat the lasers in a quantum way by using the same Bose fields that describe the emission process. One has simply to take a suitable coherent state as initial state for the fields [34]. However, this procedure does not imply any difference for the dynamics of the atom ((2.6), (3.4) and (3.5) are not changed). The true approximation is not in the use of a classical description for the lasers, but in replacing the true electromagnetic field with the Bose fields $a_j(t)$. However, this approximation is the usual one (it gives the standard Markovian master equation for the density matrix of the atom) and it is justified as long as 'memory' contributions do not play any important role.

4. The detection process

We now have to treat the counting process of blue and red photons. We assume there are two detectors at the same distance D from the atom, one counting the blue photons that reach it and the other the red photons. For the moment we do not consider the interaction between atom and field.

First we have to describe the propagation of the electromagnetic field from the atom position to the position of the counters. We can choose the modal expansion (3.2) in such a way that the index j also contains the direction of propagation. Only the photons with the right direction of propagation will reach the counter. Therefore, the set I_1 of field modes for the blue photons can be split into two disjoint sets I_1^a and I_1^b . If $j \in I_1^a$, the field $a_j(t)$ represents a field (carrying blue photons) that has the right direction of propagation for reaching the counter for blue photons; if $j \in I_1^b$, then $a_j(t)$ does not reach any counter. The same applies for the 'red' fields $a_j(t)$, $j \in I_2$, with I_2 being split into I_2^a and I_2^b .

In our approximations, the propagation of the electromagnetic field is very simple. The field $a_j(t)$ is at the atom position at the time t . The corresponding field at a distance D along the direction specified by j will, at time t , be $\tilde{a}_j(D; t) = a_j(t - D/c)$; we have only to take into account the retardation due to the time of flight of the photons. Indeed, a justification of this statement can be found in [35] where a different propagation problem is studied, but the approximations are similar to ours (note the Dirac delta $\delta(t - t')$ in the commutation rules (2.16) in [35]). The final result is that propagation involves pure translation in time (see [35] p 660).

From the above considerations, the operator that gives the number of photons arriving at the detector for blue photons in the time interval (t_1, t_2) is

$$\begin{aligned}
 N_1(t_1, t_2) &:= \sum_{j \in I_1^a} \int_{t_1}^{t_2} dt \tilde{a}_j^\dagger(D; t) \tilde{a}_j(D; t) \\
 &= \sum_{j \in I_1^a} \int_{t_1}^{t_2} dt a_j^\dagger(t - D/c) a_j(t - D/c) \equiv \sum_{j \in I_1^a} (\Lambda_{t_2 - D/c}^j - \Lambda_{t_1 - D/c}^j) \quad (4.1)
 \end{aligned}$$

where the operators Λ_t^j are given by (2.2). Analogously, one defines the number operator $N_2(t_1, t_2)$ for red photons. Note that, by the commutation rules (2.1), we have $[\Lambda_t^j, \Lambda_s^k] = 0$ and, therefore, all the operators $N_\nu(t_1, t_2)$ commute, also for different times.

Consider now an ideal photoemissive counter. This detector performs essentially a measurement of the number operator, so that we can assume the probabilities of counts to be given by the quantum expectation values of the projection-valued measure associated with the commuting self-adjoint operators (4.1). Because infinitely many self-adjoint operators are involved, it is easier to work with the Fourier transform of this projection-valued measure, which is given (cf [7] equations (4.1), (4.3) and (4.6)) by the unitary operator

$$V_t[\mathbf{k}] = \exp\left(i \sum_{\nu=1}^2 \int_0^t k_\nu(s) dN_s^\nu\right)$$

where $\mathbf{k}(t) \equiv (k_1(t), k_2(t))$ is a two-component, real-valued test function and we have set

$$N_t^\nu := N_\nu(D/c, t + D/c) \equiv \sum_{j \in I_t^\nu} \Lambda_j^\nu. \tag{4.2}$$

The operator $V_t[\mathbf{k}]$ is the analogue of the characteristic function of a probability measure and can be called a *characteristic operator*. It satisfies the QSDE (see [7] § IV)

$$dV_t[\mathbf{k}] = \left(\sum_{\nu=1}^2 (e^{ik_\nu t} - 1) dN_t^\nu \right) V_t[\mathbf{k}] \quad V_0[\mathbf{k}] = \mathbb{1}. \tag{4.3}$$

If the three-level system is also considered, the operators Λ_j^ν and N_t^ν , that give the number of photons before the interaction, have to be substituted by the analogous operators in the Heisenberg picture

$$\Lambda_t^{j\text{out}} := U_t^\dagger \Lambda_j^\nu U_t, \quad N_t^{\nu\text{out}} := U_t^\dagger N_t^\nu U_t,$$

which give the number of photons after the interaction between fields and atom. Correspondingly, the operator $V_t[\mathbf{k}]$ has to be replaced by [7]:

$$V_t^{\text{out}}[\mathbf{k}] := U_t^\dagger V_t[\mathbf{k}] U_t.$$

Now consider the quantity

$$\Phi_t[\mathbf{k}] := \text{Tr}_{h_0 \otimes \Gamma} [V_t^{\text{out}}[\mathbf{k}] (\rho \otimes |\psi_0\rangle\langle\psi_0|)] \tag{4.4}$$

where ρ is the initial state of the atom and ψ_0 (Fock vacuum) is the initial state of the fields. The quantity $\Phi_t[\mathbf{k}]$ is the quantum expectation value of the characteristic operator $V_t^{\text{out}}[\mathbf{k}]$ and, by definition of this operator, it represents the Fourier transform of the probabilities of the counts. More technically, (4.4) gives the characteristic functional of a counting process. This process fully describes the statistics of the photon counts. This point is discussed in [7] (cf also [5]); for the notion of a characteristic functional see, for instance, [36].

By using (2.4a) and (4.3), it is possible to eliminate the fields from the description. The procedure is similar to that giving the master equation (2.6). One obtains (cf [7] equations (3.20), (3.22), (3.28) and (4.8))

$$\Phi_t[\mathbf{k}] = \text{Tr}_{h_0} (G_t[\mathbf{k}] \rho) \tag{4.5}$$

where $G_t[\mathbf{k}]$ is a linear map on the space spanned by the statistical operators on h_0 , satisfying the equation

$$\frac{d}{dt} G_t[\mathbf{k}] = (L + K(\mathbf{k}(t))) G_t[\mathbf{k}] \quad G_0[\mathbf{k}] = \mathbb{1}. \tag{4.6}$$

The quantities L and $K(\mathbf{k})$ are linear maps on the same space as $G_t[\mathbf{k}]$; L is defined by (3.4) and $K(\mathbf{k})$ turns out to be given by

$$K(\mathbf{k})\rho := \sum_{\nu=1}^2 (e^{i\mathbf{k}\nu} - 1)\varepsilon_\nu \Gamma_\nu |0\rangle\langle\nu|\rho|\nu\rangle\langle 0|$$

where

$$\varepsilon_\nu := \frac{1}{\Gamma_\nu} \sum_{j \in I_\nu} |\lambda_j|^2.$$

From definition (3.6) of Γ_ν , we have $0 \leq \varepsilon_\nu \leq 1$. As will be apparent from the following developments, ε_1 is the efficiency of the counter for the blue photons and ε_2 is the efficiency of the counter for red photons. In particular, $\varepsilon_2 = 0$ represents the case of no counter for red photons.

It is possible to prove [5] that, once the fields are eliminated, a counting process in the sense of [23–25] is obtained.

From now on we shall forget the retardation D/c (see (4.1) and (4.2)) that simply gives a constant time shift in all the detection times.

5. The statistics of the photon counting process

As stated in the previous section, the characteristic functional (4.4) or (4.5) contains the whole statistics of the counting process and any statistical information on the emitted light can be extracted from it. This functional can be rewritten in such a way that the structure of a counting process is apparent. By setting

$$\tilde{L}\rho := -i\tilde{H}\rho + i\rho\tilde{H}^\dagger + \sum_{\nu=1}^2 (1 - \varepsilon_\nu)\Gamma_\nu |0\rangle\langle\nu|\rho|\nu\rangle\langle 0| \tag{5.1}$$

$$J(t)\rho := \sum_{\nu=1}^2 |e^{i\mathbf{k}_\nu(t)} \varepsilon_\nu \Gamma_\nu |0\rangle\langle\nu|\rho|\nu\rangle\langle 0|$$

one has

$$L + K(\mathbf{k}(t)) = \tilde{L} + J(t).$$

The solution of (4.6) can now be written as an expansion in $J(t)$ (Dyson series):

$$\begin{aligned} G_t[\mathbf{k}] = & Y(t) + \sum_{m=1}^\infty \int_0^t dt_m \int_0^{t_m} dt_{m-1} \dots \int_0^{t_2} dt_1 Y(t - t_m) J(t_m) \\ & \times Y(t_m - t_{m-1}) J(t_{m-1}) \dots Y(t_2 - t_1) J(t_1) Y(t_1) \end{aligned}$$

where

$$Y(t) := \exp(\tilde{L}t). \tag{5.2}$$

Now the characteristic functional (4.5) becomes

$$\begin{aligned} \text{Tr}_{h_0}(G_t[\mathbf{k}]\rho) = & P_t(0|\rho) + \sum_{m=1}^\infty \sum_{\nu_1=1}^2 \dots \sum_{\nu_m=1}^2 \int_0^t dt_m \int_0^{t_m} dt_{m-1} \dots \int_0^{t_2} dt_1 \\ & \times \exp\left(i \sum_{r=1}^m \mathbf{k}_{\nu_r}(t_r)\right) p_t(\nu_m, t_m; \nu_{m-1}, t_{m-1}; \dots; \nu_1, t_1 | \rho) \end{aligned} \tag{5.3}$$

where

$$P_t(0|\rho) := \text{Tr}_{h_0}(Y(t)\rho) \tag{5.4}$$

$$p_t(\nu_m, t_m; \dots; \nu_1, t_1|\rho) := P_{t-t_m}(0|Q_0) W_{\nu_m}(t_m - t_{m-1}) W_{\nu_{m-1}}(t_{m-1} - t_{m-2}) \times \dots \times W_{\nu_2}(t_2 - t_1) \varepsilon_{\nu_1} \Gamma_{\nu_1} \langle \nu_1 | (Y(t_1)\rho) | \nu_1 \rangle \tag{5.5}$$

$$Q_0 := |0\rangle\langle 0| \tag{5.6}$$

$$W_\nu(t) := \varepsilon_\nu \Gamma_\nu \langle \nu | (Y(t)Q_0) | \nu \rangle. \tag{5.7}$$

The structure (5.3) is typical for the characteristic functional of a regular point process (roughly speaking, a point process for which the probability density of two or more counts at the same time vanishes). The quantity (5.4) is the probability of having no count in the interval $(0, t]$ when the initial state of the atom is ρ . The quantities (5.5) are the probability densities of counting a photon of type ν_1 at time t_1 , a photon of type ν_2 at time $t_2, \dots, (0 < t_1 < t_2 < \dots < t_m < t)$ and no other count in the interval $(0, t]$; these quantities are usually called elementary (or exclusive) probability densities (EPD) [25, 26]. The factorised structure of the EPD is a characteristic of the model we are considering and it renders the study of the counting process very simple.

From the quantities (5.4) and (5.5) the full statistics of the counting process can be obtained. For instance, the probability of detecting n blue photons and no red ones in the interval $(0, t]$ is given by

$$P_t(n, 1|\rho) = \int_0^t dt_n \int_0^{t_n} dt_{n-1} \dots \int_0^{t_2} dt_1 p_t(1, t_n; \dots; 1, t_1|\rho).$$

It is interesting to consider the EPD in an interval $(t_0, t]$, conditioned by having counted a photon of type ν_0 at time t_0 . From the structure (5.5) we obtain for these conditional EPD

$$p_t(\nu_m, t_m; \dots; \nu_1, t_1|\nu_0, t_0) = p_{t-t_0}(\nu_m, t_m - t_0; \dots; \nu_1, t_1 - t_0|Q_0). \tag{5.8}$$

In other words, after a count (of a blue or red photon) the atom is in the ground state. This result is due to the structure of the interaction atom-field. Moreover, from (5.5)–(5.7) we have

$$W_\nu(t) = p_t(\nu, t|Q_0)$$

and, therefore, by taking into account (5.8), we see that $W_\nu(t)$ is the probability density of detecting a photon of type ν at time $t_0 + t$ and no other photon between t_0 and $t_0 + t$, given a count at time t_0 .

By defining

$$\Theta(t)\rho := \exp(-i\tilde{H}t)\rho \exp(i\tilde{H}^\dagger t)$$

$$S\rho := \sum_{\nu=1}^2 (1 - \varepsilon_\nu) \Gamma_\nu |0\rangle\langle \nu| \rho | \nu\rangle\langle 0|$$

we have from (5.1) and (5.2) that $Y(t)$ satisfies the integral equation

$$Y(t) = \Theta(t) + \int_0^t dt' Y(t-t') S\Theta(t').$$

By putting this expression into (5.7) we obtain

$$W_\nu(t) = \varepsilon_\nu \bar{W}_\nu(t) + \sum_{\eta=1}^2 (1 - \varepsilon_\eta) \int_0^t dt' \bar{W}_\eta(t') W_\nu(t-t') \quad (5.9)$$

where

$$\bar{W}_\nu(t) := \Gamma_\nu \langle \nu | (\Theta(t) Q_0) | \nu \rangle \equiv \Gamma_\nu \langle \nu | \exp(-i\tilde{H}t) | 0 \rangle^2. \quad (5.10)$$

Note that, when all the emitted photons are detected ($\varepsilon_1 = \varepsilon_2 = 1$), the quantities $W_\nu(t)$ and $\bar{W}_\nu(t)$ coincide. Therefore, from the meaning of $W_\nu(t)$, it follows that $\bar{W}_\nu(t)$ is the probability density of the *emission* of a photon of type ν at time $t + t_0$, given that the previous emission was at time t_0 . The quantities $W_\nu(t)$ and $\bar{W}_\nu(t)$ satisfy the equations

$$\int_0^{+\infty} dt \sum_{\nu=1}^2 \bar{W}_\nu(t) = 1 \quad (5.11a)$$

$$\int_0^{+\infty} dt \sum_{\nu=1}^2 W_\nu(t) = 1. \quad (5.11b)$$

Indeed, the first quantity above is the probability that at least a photon is emitted in the interval $(0, +\infty)$ and the second one is the probability that at least a photon is detected in $(0, +\infty)$; on physical grounds, these probabilities must be one. The formal proof runs as follows. We have

$$\begin{aligned} \sum_{\nu=1}^2 \bar{W}_\nu(t) &= i \operatorname{Tr}_{h_0} [(\tilde{H} - \tilde{H}^*)(\Theta(t) Q_0)] = -\frac{d}{dt} \operatorname{Tr}_{h_0} (\Theta(t) Q_0) \\ \sum_{\nu=1}^2 \int_0^{+\infty} dt \bar{W}_\nu(t) &= 1 - \lim_{t \rightarrow +\infty} \operatorname{Tr}_{h_0} [\Theta(t) Q_0]. \end{aligned} \quad (5.12)$$

In the next section we shall show that the limit on the RHS of (5.12) vanishes and this proves (5.11a). Then (5.11b) is a consequence of (5.9).

Consider now the probability of no counts up to time t . From (3.5), (5.1), (5.2) and (5.4), we obtain

$$\begin{aligned} P_0(0 | \rho) &= 1 \\ \frac{d}{dt} P_t(0 | \rho) &= \operatorname{Tr}_{h_0} (\tilde{L} Y(t) \rho) = -\sum_{\nu=1}^2 \varepsilon_\nu \Gamma_\nu \langle \nu | (Y(t) \rho) | \nu \rangle. \end{aligned}$$

By taking $\rho = Q_0$ and recalling (5.7) and (5.11b), these equations give

$$P_t(0 | Q_0) = 1 - \sum_{\nu=1}^2 \int_0^t dt' W_\nu(t') = \sum_{\nu=1}^2 \int_t^{+\infty} dt' W_\nu(t').$$

Analogously, the probability $\bar{P}(t)$ of no emission up to time t , given an emission at time zero, is

$$\bar{P}(t) = 1 - \sum_{\nu=1}^2 \int_0^t dt' \bar{W}_\nu(t') = \sum_{\nu=1}^2 \int_t^{+\infty} dt' \bar{W}_\nu(t'). \quad (5.13)$$

Finally, by exploiting (5.9), we obtain

$$P_t(0 | Q_0) = \bar{P}(t) + \sum_{\nu=1}^2 (1 - \varepsilon_\nu) \int_0^t dt' \bar{W}_\nu(t') P_{t-t'}(0 | Q_0). \quad (5.14)$$

This equation has a simple physical meaning. The quantity $\bar{W}_\nu(t') dt'$ is the probability of the emission of a photon of type ν between t' and $t' + dt'$, $1 - \varepsilon_\nu$ is the probability of failing to detect this photon, and $P_{t-t'}(0|Q_0)$ is the probability of no detection between t' and t . Therefore, (5.14) states that the probability of no detection up to time t is equal to the probability of no emission up to time t plus the probability that some photons be emitted before t but not detected. Equations (5.9) and (5.14) connect the emission process to the detection process by taking into account the efficiency of the detectors. Although in our approach they are naturally derived from an operator equation for $Y(t)$, they do not have a quantum origin and could also be derived from purely classical arguments. Moreover, we have considered detectors that count all the photons reaching them; but an intrinsic efficiency of the detectors can be easily taken into account by a smaller value of the parameter ε_ν .

6. Dark and bright periods

In [12] it is shown that the characteristics of bright and dark periods are controlled by the probability $P_t(0|Q_0)$ of no count. However, we expect that the main properties of bright and dark periods do not depend on the efficiency of the counters, at least if the efficiency of the detector for blue photons is not too bad (see equation (11) of [12] and the related comments). Indeed, going back to the intuitive explanation given in the introduction, to detect or not to detect the rare red photons cannot change the length of bright and dark periods and to miss a part of the blue photons merely has the effect of lowering the intensity of the signal during the bright periods. In principle, a rigorous proof of this statement would be based on (5.14); however, computations are not quite so simple. In any case, from now on we shall consider only the emission process, as in [12].

Let us start by computing the functions $\bar{W}_\nu(t)$. If we set

$$\exp(-i\tilde{H}t)|0\rangle = \sum_{\nu=0}^2 a_\nu(t)|\nu\rangle$$

then (5.10) gives

$$\bar{W}_\nu(t) = \Gamma_\nu |a_\nu(t)|^2 \quad \nu = 1, 2. \tag{6.1}$$

By definition (3.5) of \tilde{H} , we have that coefficients $a_\nu(t)$ satisfy the equations (cf [12] equations (14))

$$\begin{aligned} \dot{a}_0(t) &= -\frac{1}{2}i\Omega_1 a_1(t) - \frac{1}{2}i\Omega_2 a_2(t) \\ \dot{a}_1(t) &= -\frac{1}{2}i\Omega_1 a_0(t) + (i\Delta_1 - \frac{1}{2}\Gamma_1) a_1(t) \\ \dot{a}_2(t) &= -\frac{1}{2}i\Omega_2 a_0(t) + (i\Delta_2 - \frac{1}{2}\Gamma_2) a_2(t) \end{aligned} \tag{6.2}$$

with the initial condition $a_\nu(0) = \delta_{\nu,0}$. The solution of these equations can be obtained by the Laplace transform technique and can be written as

$$\begin{aligned} a_1(t) &= -\frac{1}{2}i\Omega_1 \left(\frac{z_0 - i\Delta_2 + \frac{1}{2}\Gamma_2}{(z_2 - z_0)(z_1 - z_0)} e^{z_1 t} + \frac{z_1 - i\Delta_2 + \frac{1}{2}\Gamma_2}{(z_0 - z_1)(z_2 - z_1)} e^{z_1 t} + \frac{z_2 - i\Delta_2 + \frac{1}{2}\Gamma_2}{(z_1 - z_2)(z_0 - z_2)} e^{z_2 t} \right) \\ a_2(t) &= -\frac{1}{2}i\Omega_2 \left(\frac{z_0 - i\Delta_1 + \frac{1}{2}\Gamma_1}{(z_2 - z_0)(z_1 - z_0)} e^{z_1 t} + \frac{z_1 - i\Delta_1 + \frac{1}{2}\Gamma_1}{(z_0 - z_1)(z_2 - z_1)} e^{z_1 t} + \frac{z_2 - i\Delta_1 + \frac{1}{2}\Gamma_1}{(z_1 - z_2)(z_0 - z_2)} e^{z_2 t} \right) \end{aligned}$$

with an analogous expression for $a_0(t)$. The z_ν are the three roots of the characteristic equation of system (6.2). It can be shown that $\text{Re}(z_\nu) < 0$; this implies that the limit in the RHS of (5.12) vanishes and (5.11a) is proved.

We are interested in the case in which very different characteristic times appear. Indeed, when

$$\Gamma_1 \gg \Gamma_2 \quad \Gamma_1 \gg \Omega_2 \quad \Omega_1 \gg \Omega_2 \quad \Omega_1^2 \gg \Gamma_1 \Gamma_2 \quad \Delta_1 = 0 \quad (6.3)$$

the three roots z_ν are approximately given by

$$z_1 \approx -\frac{1}{4}\Gamma_1 + \frac{1}{2}(\frac{1}{4}\Gamma_1^2 - \Omega_1^2)^{1/2} \quad (6.4a)$$

$$z_2 \approx -\frac{1}{4}\Gamma_1 - \frac{1}{2}(\frac{1}{4}\Gamma_1^2 - \Omega_1^2)^{1/2} \quad (6.4b)$$

$$z_0 = -\frac{1}{2}\Gamma_2 + i\Delta_2 - \zeta \quad (6.5a)$$

$$\zeta \approx \frac{1}{2}\Omega_2^2 \left(\frac{\Gamma_1 \Omega_1^2 + 2i\Delta_2(\Omega_1^2 - 4\Delta_2^2 - \Gamma_1^2)}{(\Omega_1^2 - 4\Delta_2^2)^2 + 4\Delta_2^2 \Gamma_1^2} \right) \quad (6.5b)$$

where z_1 and z_2 are real or complex numbers according to the sign of $\Gamma_1^2 - 4\Omega_1^2$. From (6.3) we have that $|\text{Re}(z_0)| \ll |\text{Re}(z_{1,2})|$.

Now, following [12], we write (see (5.13) and (6.1))

$$\begin{aligned} \bar{P}(t) &= \sum_{\nu=1}^2 \int_t^{+\infty} dt' \Gamma_\nu |a_\nu(t')|^2 \\ &= \int_t^{+\infty} dt' (F_{\text{short}}(t') + F_{\text{long}}(t')) \end{aligned} \quad (6.6)$$

where

$$F_{\text{long}}(t) = \frac{\Gamma_1 \Omega_1^2 |\zeta|^2 + \Gamma_2 \Omega_2^2 \frac{1}{2} |\Gamma_1 + z_0|^2}{4|(z_1 - z_0)(z_2 - z_0)|^2} \exp(2 \text{Re}(z_0)t)$$

and in $F_{\text{short}}(t)$ we have grouped all the other terms, which have a much shorter decaying time. Let us set now

$$\Pi := \int_0^{+\infty} dt F_{\text{long}}(t)$$

then (6.6) and (5.13) imply

$$\int_0^{+\infty} dt F_{\text{short}}(t) = 1 - \Pi.$$

By using the results (6.4) and (6.5) we obtain the approximate expressions:

$$F_{\text{short}}(t) \approx \frac{\Gamma_1 \Omega_1^2}{|\Gamma_1^2 - 4\Omega_1^2|} |e^{z_1 t} - e^{z_2 t}|^2$$

$$F_{\text{long}}(t) = \Pi |2 \text{Re}(z_0)| \exp[2 \text{Re}(z_0)t]$$

$$\begin{aligned} \Pi &\approx \frac{\Omega_2^2}{(\Omega_1^2 - 4\Delta_2^2)^2 + 4\Delta_2^2 \Gamma_1^2} \frac{1}{|2 \text{Re}(z_0)|} \\ &\quad \times \left(\Gamma_1 \Omega_1^2 \Omega_2^2 \frac{\Gamma_1^2 \Omega_1^4 + 4\Delta_2^2 (\Omega_1^2 - 4\Delta_2^2 - \Gamma_1^2)^2}{[(\Omega_1^2 - 4\Delta_2^2)^2 + 4\Delta_2^2 \Gamma_1^2]^2} + \Gamma_2 (\Gamma_1^2 + 4\Delta_2^2) \right). \end{aligned} \quad (6.7)$$

Moreover, (6.3) and (6.7) give $\Pi \ll 1$.

In [12] it is shown that the structure (6.6) for $\bar{P}(t)$ implies the existence of bright and dark periods. The argument is as follows. The quantity $1 - \bar{P}(t)$ is the probability of at least an emission in $(0, t]$. Then the quantity

$$p(t) := \frac{d}{dt}[1 - \bar{P}(t)] = F_{\text{short}}(t) + F_{\text{long}}(t)$$

is the probability density for the waiting time Δt between two emissions.

We introduce now a time delay θ such as

$$|2 \operatorname{Re}(z_2)|^{-1} \ll \theta \ll |2 \operatorname{Re}(z_0)|^{-1} \quad (6.8)$$

and consider the time interval Δt between successive emissions as 'short' if $\Delta t < \theta$ and as 'long' if $\Delta t > \theta$. The probability of having a short waiting time between two emissions is

$$P(\Delta t < \theta) = \int_0^\theta dt p(t) = \int_0^\theta dt F_{\text{short}}(t) \simeq \int_0^{+\infty} dt F_{\text{short}}(t) = 1 - \Pi$$

and, similarly, the probability of a long waiting time is

$$P(\Delta t > \theta) \simeq \int_0^{+\infty} dt F_{\text{long}}(t) = \Pi.$$

Note that these probabilities are independent from θ as far as (6.8) holds. We can say that the 'short' waiting times are distributed with a probability density $(1 - \Pi)^{-1} F_{\text{short}}(t)$ and the 'long' ones with a probability density $\Pi^{-1} F_{\text{long}}(t)$. Therefore, the mean duration of the short intervals is given by

$$T_{\text{short}} = \frac{1}{1 - \Pi} \int_0^{+\infty} dt t F_{\text{short}}(t) = \Gamma_1 \Omega_1^{-2} + 2\Gamma_1^{-1} \quad (6.9)$$

and the mean duration of the long intervals is

$$T_{\text{long}} = \frac{1}{\Pi} \int_0^{+\infty} dt t F_{\text{long}}(t) = |2 \operatorname{Re}(z_0)|^{-1}. \quad (6.10)$$

When many photons are emitted, separated by short time intervals, the intensity of the signal registered by the detector is approximately constant, with small fluctuations due to shot noise, and we have a bright period. When two photons are emitted instead, separated by a long time interval, a dark period is registered. The mean duration τ_B of a bright period is given by $T_{\text{short}} \bar{N}$, where \bar{N} is the average number of consecutive short intervals. The durations of the intervals between two emissions are uncorrelated variables, because after any emission the electron goes into the ground state (cf (5.8) and the following comments). Therefore, the probability of having N short intervals followed by a long one is $(1 - \Pi)^N \Pi$. If we want at least one short interval in the sequence, we have to divide this probability by $1 - \Pi$. Finally, we have

$$\bar{N} = \sum_{N=1}^{\infty} N (1 - \Pi)^{N-1} \Pi = 1/\Pi$$

and the mean duration τ_B of bright periods is

$$\tau_B = T_{\text{short}}/\Pi. \quad (6.11)$$

Analogously, the mean duration τ_D of dark periods is

$$\tau_D = T_{\text{long}}/(1 - \Pi) \simeq T_{\text{long}} \quad (6.12)$$

When (6.4), (6.5), (6.7), (6.9)–(6.12) are particularised to the cases considered in [12] ($\Omega_1 \ll \Gamma_1$ or $\Omega_1 \gg \Gamma_1$), exactly the same results are obtained. In [12] it is also shown that it is possible to obtain spectroscopic information on the weak transition by plotting the ratio τ_D/τ_B against the detuning Δ_2 . Our presentation of the shelving effect shows that the explanation given by Cohen-Tannoudji and Dalibard can be founded in a well developed theory of counting processes and that, in principle, any other statistical property of the emitted light could be computed. Moreover, our procedure can be easily applied also to the case of incoherent excitation.

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