

Work, dissipation, and fluctuations in nonequilibrium physics

Dynamics of fluctuations in non-Markovian systems

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Abstract

The dynamics of multiple time correlation functions (MTCFs) of a system in contact with a non-Markovian environment is relevant in many different contexts. In this article we study the dynamical equations that satisfy MTCFs of a system and how they are used to characterize its fluctuations. The non-Markovian character of the interaction between the system and its environment leads to dynamical equations different from those derived from the Quantum Regression Theorem. We further discuss a stochastic method to compute MTCFs. To illustrate the theory we apply it to the evaluation of the emission spectrum of an atom in contact with a non-Markovian environment. The emission spectrum strongly depends on the way the environment reacts to the absorption of a photon emitted by the atom. **To cite this article:** *D. Alonso et al., C. R. Physique 8 (2007).*

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Résumé

Dynamique de fluctuations dans les systèmes non-markoviens. La dynamique des fonctions de corrélation à temps multiples (FCTM) d'un système en contact avec un environnement non-markovien est importante dans de nombreux contextes différents. Dans cet article, nous étudions les équations dynamiques satisfaites par les FCTM d'un système et nous montrons comment elles peuvent être utilisées pour caractériser ses fluctuations. Le caractère non-markovien de l'interaction entre le système et son environnement mène à des équations dynamiques différentes de celles dérivées à partir du théorème de régression quantique. Nous discutons de plus une méthode stochastique pour calculer les FCTM. Pour illustrer la théorie, nous l'appliquons à l'évaluation du spectre d'émission d'un atome en contact avec un environnement non-markovien. Le spectre d'émission dépend fortement de la manière dont l'environnement réagit à l'absorption d'un photon émis par l'atome. **Pour citer cet article :** *D. Alonso et al., C. R. Physique 8 (2007).*

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1. Introduction and motivation

Any system can be considered open, in the sense that the degrees of freedom under study are just a subset of a larger set containing many more degrees of freedom. In many circumstances we can assume that the system of interest is isolated, but this approximation is not appropriate in other cases. Furthermore, the fact that the system is considered open leads to interesting physics. Open systems are ubiquitous and have attracted our attention for very long time [1].

With the advances in modern technologies and micro and nano-fabrication techniques the interaction of a particular system with its environment has to be treated at a quantum level. A beautiful example is found in the so-called nanomechanical oscillators [2], which are objects of about 10^{10} atoms that oscillate under the influence of several sources of quantum noise, for instance the thermal environment. The size of the object is so small that the distance between adjacent quantum energies of the oscillator are much bigger than the typical thermal energy of the environment. This leads to the conclusion that the interaction between the system and the environment is fully quantum mechanical [3]. Another example is found in the interaction of atoms or quantum dots with a radiation field (environment) confined in a photonic crystal [4–6].

A crucial step in the description of the dynamics of a quantum open system is to compare the typical time scale associated to the relaxation dynamics of the system (τ_R) with the time scale associated to the environment, which takes into account how fast the energy absorbed by the environment is redistributed among its degrees of freedom, the so-called correlation time (τ_B). If the correlation time is much smaller than τ_R we can assume that the environment reacts almost instantaneously to the interaction with the system. This is the Markovian approximation, which is extremely useful and provides a solid framework in which many situations can be studied [7].

However, the Markovian approximation does not properly describe those situations where the environmental time scale is not small enough in comparison with the typical time scale of the atom. An example of these non-Markovian interactions is an atom in contact with a structured environment, like the modified radiation field that exists within a photonic crystal. Here, the decay of correlations that measure the relaxation of the environment after its interaction with the atom is very slow. Another example is the dynamics of a localized electron spin interacting with an environment of nuclear spins [8]. Hence, the theory of an open quantum system with non-Markovian interactions becomes relevant in many interesting situations [9–12].

In general, the *dynamics* of a system in contact with an environment is characterized by the expectation values of the relevant observables as well as their fluctuations. These fluctuations describe important properties, such as the response of the system to external fields. In the case of atoms in contact with a radiation field these fluctuations characterize properties such as the spontaneous emission, the fluorescence spectrum and the photon statistics of the light emitted by the atom. These fluctuations are given by the so-called multiple-time correlation functions [7].

We see that there is a strong motivation in understanding *open quantum systems* that have a *non-Markovian* character, and, furthermore, in describing not only the dynamics of *expectation values*, but also the dynamics of their *fluctuations* in terms of MTCFs. Here, we shall describe a theory to compute the dynamics of MTCFs in non-Markovian interactions. Such dynamics is described by a set of coupled differential equations that involve all the MTCF of the observables that describe the system. Alternatively, it is possible to compute an specific MTCF by an stochastic scheme. Both methods are equivalent. However, they are in some sense complementary, since they might present advantages and disadvantages compared to each other depending on the particular case to be studied. We shall use the theory to compute the emission spectrum of a simple atom in a non-Markovian environment.

The plan of the article is the following. In Section 2 we shall discuss the formalism to compute MTCFs. We shall derive the set of dynamical equations that satisfy MTCFs in the weak coupling limit. In this section we shall also discuss an stochastic method to compute a particular MTCF. In Section 3 we shall apply the formalism introduced in 2 to the evaluation of the emission spectrum on an atom in a non-Markovian environment. Finally we shall draw the main conclusions.

2. Fluctuations: multiple time correlation functions

We shall consider a model in which the system and the environment interact linearly. The total Hamiltonian that describes the dynamics of the system \mathcal{S} , with Hamiltonian H_S , in interaction with the environment, with Hamiltonian H_B is

$$H_T = H_S + H_B + H_I = H_S + \sum_{\lambda} \omega_{\lambda} a_{\lambda}^{\dagger} a_{\lambda} + \sum_{\lambda} g_{\lambda} (a_{\lambda}^{\dagger} L + L^{\dagger} a_{\lambda}) \quad (1)$$

where the operator L acts on the Hilbert space of the system and $a_{\lambda}, a_{\lambda}^{\dagger}$ are the annihilation and creation operators, respectively, in the environment Hilbert space. The g_{λ} are the coupling constants that can be taken as real numbers, and the ω_{λ} are the frequencies of the harmonic oscillators that constitute the environment.

We will assume the weak coupling approximation, in which the system interaction Hamiltonian H_I is g times smaller than the free Hamiltonian $H_S + H_B$. We will also use, for simplicity, units in which $\hbar = 1$.

The model of linear interaction Hamiltonian is good enough to characterize most of the interactions in quantum optics related to processes in which only one photon is involved, and which can be described within the dipolar approximation. Other models, such as the well known Spin-Boson model are also described by the Hamiltonian (1) [13].

In this article, we shall focus in the dynamics of two-time correlation functions of system observables, higher order correlations can also be evaluated, but to illustrate the theory it is enough to consider up to two-time correlations. Higher order correlations are obtained in [14]. We will use the system of equations derived in [15,16] in the weak coupling limit.

2.1. Two-time correlation function of system observables in the weak coupling limit

In general let us take a set of N system observables $A_1(t_1), A_2(t_2), \dots, A_N(t_N)$ in Heisenberg representation, such that $t_1 > t_2 > \dots > t_N$. If Ψ_0 is the initial state of the total system

$$C_A(t) = \langle \Psi_0 | A_1(t_1) A_2(t_2) \cdots A_N(t_N) | \Psi_0 \rangle$$

is a N -time correlation function of the system. This is the object of our interest.

To begin let us consider the Heisenberg evolution equation for a system observable $A(t) = \mathcal{U}^{-1}(t, 0) A \mathcal{U}(t, 0)$, where $\mathcal{U}(t, 0)$ is the evolution operator with the total Hamiltonian (1),

$$\begin{aligned} \frac{dA(t_1)}{dt_1} &= i\mathcal{U}^{-1}(t_1, 0) [H_T, A] \mathcal{U}(t_1, 0) \\ &= -i[H_S(t_1), A(t_1)] + i \sum_{\lambda} g_{\lambda} (a_{\lambda}^{\dagger}(t_1, 0) [L(t_1), A(t_1)] + [L^{\dagger}(t_1), A(t_1)] a_{\lambda}(t_1, 0)) \end{aligned} \quad (2)$$

We can replace in (2) the formal solution of the evolution equation of the environmental operators,

$$\begin{aligned} da_{\lambda}(t_1, 0)/dt_1 &= i[H_T(t_1), a_{\lambda}(t_1, 0)] = -i\omega_{\lambda} a_{\lambda}(t_1, 0) - ig_{\lambda} L(t_1) \\ a_{\lambda}(t_1, 0) &= e^{-i\omega_{\lambda} t_1} a(0, 0) - ig_{\lambda} \int_0^{t_1} d\tau e^{-i\omega_{\lambda}(t_1 - \tau)} L(\tau) \end{aligned} \quad (3)$$

The single evolution equation (2) becomes,

$$\begin{aligned} \frac{dA(t_1)}{dt_1} &= i[H_S(t_1), A(t_1)] - v^{\dagger}(t_1) [L(t_1), A(t_1)] + \int_0^{t_1} d\tau \alpha^*(t_1 - \tau) L^{\dagger}(\tau) [A(t_1), L(t_1)] \\ &\quad + [L^{\dagger}(t_1), A(t_1)] v(t_1) + \int_0^{t_1} d\tau \alpha(t_1 - \tau) [L^{\dagger}(t_1), A(t_1)] L(\tau) \end{aligned} \quad (4)$$

where we have defined $\alpha(t) = \sum_{\lambda} |g_{\lambda}|^2 e^{-i\omega_{\lambda} t}$, the so called *environment correlation function*. Generally for an environment with a large number of degrees of freedom this function decays in a typical time scale τ_B , the *environment correlation time*. We have also defined the bath operators

$$v^{\dagger}(t_1) = -i \sum_{\lambda} g_{\lambda} a_{\lambda}^{\dagger}(0, 0) e^{i\omega_{\lambda} t_1}, \quad v(t_1) = i \sum_{\lambda} g_{\lambda} a_{\lambda}(0, 0) e^{-i\omega_{\lambda} t_1} \quad (5)$$

From (4) the evolution equation of the quantum mean value of A for an initial state $|\Psi_0\rangle = |\psi_0\rangle|0\rangle$, with $|0\rangle$ the vacuum state for the environment, is equal to

$$\begin{aligned} \frac{d}{dt_1} \langle \Psi_0 | A(t_1) | \Psi_0 \rangle &= i \langle \Psi_0 | [H_S(t_1), A(t_1)] | \Psi_0 \rangle + \int_0^{t_1} d\tau \alpha(t_1 - \tau) \langle \Psi_0 | [L^\dagger(t_1), A(t_1)] L(\tau) | \Psi_0 \rangle \\ &+ \int_0^{t_1} d\tau \alpha^*(t_1 - \tau) \langle \Psi_0 | L^\dagger(\tau) [A(t_1), L(t_1)] | \Psi_0 \rangle \end{aligned} \quad (6)$$

It is important to note that in the dynamical equation (4) the environment operators $v(t)$ and $v^\dagger(t)$ describe the action of the environment on the system; they represent an external driving acting on the system due to its interaction with the environment. These *forces* are related to the correlation function $\alpha(t - t')$, in fact it can be shown that $\langle 0 | v(t) v^\dagger(t') | 0 \rangle = \alpha(t - t')$, therefore the environment correlation function is the autocorrelation function of the environmental *forces* acting on the system. Furthermore, $\langle 0 | v^\dagger(t) | 0 \rangle = \langle 0 | v(t) | 0 \rangle = 0$.

For the case of two-time correlation functions we now calculate the following evolution equation,

$$\begin{aligned} \frac{dA(t_1)B(t_2)}{dt} &= i\mathcal{U}^{-1}(t_1)[H_T, A]\mathcal{U}(t_1)B(t) = i[H_S(t_1), A(t_1)]B(t_2) \\ &+ i \sum_{\lambda} g_{\lambda} (a_{\lambda}^{\dagger}(t_1, 0)[L(t_1), A(t_1)]B(t_2) + [L^{\dagger}(t_1), A(t_1)]a_{\lambda}(t_1, 0)B(t_2)) \end{aligned} \quad (7)$$

The idea again is to eliminate the dependency on the environmental operators once the average over the total system state is performed. First, we replace the analytical solution of the creation operator $a_{\lambda}^{\dagger}(t_1, 0)$, so that the term $a_{\lambda}^{\dagger}(0, 0)$ appears on the left hand side of the expression and can be eliminated when applying the vacuum initial state. Second, we move the annihilation operator to the right hand side by doing the following,

$$\begin{aligned} a_{\lambda}(t_1, 0)B(t_2) &= \mathcal{U}^{-1}(t_2)a_{\lambda}(t_1, t_2)B\mathcal{U}(t_2) \\ &= \mathcal{U}^{-1}(t_2)e^{-i\omega_{\lambda}(t_1-t_2)}a_{\lambda}(0, 0)B\mathcal{U}(t_2) - ig_{\lambda} \int_{t_2}^{t_1} d\tau e^{-i\omega_{\lambda}(t_1-\tau)}L(\tau)B(t_2) \\ &= B(t_2)a_{\lambda}(t_2, 0) - ig_{\lambda} \int_{t_2}^{t_1} d\tau e^{-i\omega_{\lambda}(t_1-\tau)}L(\tau)B(t_2) \end{aligned} \quad (8)$$

where we have used

$$a_{\lambda}(t_1, t_2) = e^{-i\omega_{\lambda}(t_1-t_2)}a_{\lambda}(t_2, t_2) - ig_{\lambda} \int_{t_2}^{t_1} d\tau e^{-i\omega_{\lambda}(t_1-\tau)}L(\tau, t_2) \quad (9)$$

with $a_{\lambda}(t_2, t_2) = a_{\lambda}(0, 0) \equiv a_{\lambda}$, and $[B, a_{\lambda}(0, 0)] = 0$. We now insert the solution of $a_{\lambda}(t_2, 0)$, which is of the form (3), in the former expression and obtain

$$a_{\lambda}(t_1, 0)B(t_2) = e^{-i\omega_{\lambda}t_1}B(t_2)a_{\lambda}(0, 0) - ig_{\lambda} \int_0^{t_2} d\tau e^{-i\omega_{\lambda}(t_1-\tau)}B(t_2)L(\tau) - ig_{\lambda} \int_{t_2}^{t_1} d\tau e^{-i\omega_{\lambda}(t_1-\tau)}L(\tau)B(t_2) \quad (10)$$

Replacing (10) in (7), and considering the solution of $a_{\lambda}^{\dagger}(t_1, 0)$, we obtain the following

$$\begin{aligned} \frac{dA(t_1)B(t_2)}{dt_1} &= i[H_S(t_1), A(t_1)]B(t_2) - v^\dagger(t_1)[L(t_1), A(t_1)]B(t_2) \\ &- \int_0^{t_1} d\tau \alpha^*(t_1 - \tau)L^\dagger(\tau)[L(t_1), A(t_1)]B(t_2) + [L^\dagger(t_1), A(t_1)]B(t_2)v(t_1) \\ &+ \int_{t_2}^{t_1} d\tau \alpha(t_1 - \tau)[L^\dagger(t_1), A(t_1)]L(\tau)B(t_2) + \int_0^{t_2} d\tau \alpha(t_1 - \tau)[L^\dagger(t_1), A(t_1)]B(t_2)L(\tau) \end{aligned} \quad (11)$$

The evolution of the quantum mean value $\langle A(t_1)B(t_2) \rangle$ is again obtained by applying the total initial state on both sides of the former expression. When such initial state is $|\psi_0\rangle|0\rangle$, we obtain the following

$$\begin{aligned} \frac{d\langle \Psi_0|A(t_1)B(t_2)|\Psi_0 \rangle}{dt_1} &= i\langle \Psi_0|[H_S(t_1), A(t_1)]B(t_2)|\Psi_0 \rangle \\ &+ \int_0^{t_1} d\tau \alpha^*(t_1 - \tau) \langle \Psi_0|L^\dagger(\tau)[A(t_1), L(t_1)]B(t_2)|\Psi_0 \rangle \\ &+ \int_{t_2}^{t_1} d\tau \alpha(t_1 - \tau) \langle \Psi_0|[L^\dagger(t_1), A(t_1)]L(\tau)B(t_2)|\Psi_0 \rangle \\ &+ \int_0^{t_2} d\tau \alpha(t_1 - \tau) \langle \Psi_0|[L^\dagger(t_1), A(t_1)]B(t_2)L(\tau)|\Psi_0 \rangle \end{aligned} \quad (12)$$

Eqs. (6) and (12) represent the evolution of quantum mean values and two-time correlations respectively, obtained *without the use of any approximation*. However, it is clear that these equations are open, in the sense that quantum mean values depend on two-time correlations, while two-time correlations depend on three-time correlations. In general, when no approximations are made, N -time correlation depend on $(N + 1)$ -time correlations, what gives rise to a hierarchy structure of MTCFs as described in [14].

At this stage we consider that the system and the interaction are weakly coupled. If we define $V_t A B = e^{iH_S t} A e^{-iH_S t} B$ and $A(t) = e^{iH_T t} A e^{-iH_T t}$, we can write the weak coupling approximation of (6) and (12) up to *second order* in the coupling constant (see [15,16] for details).

Then we obtain the following equation for quantum mean values,

$$\begin{aligned} \frac{d}{dt_1} \langle \Psi_0|A(t_1)|\Psi_0 \rangle &= i\langle \Psi_0|\{[H_S, A]\}(t_1)|\Psi_0 \rangle + \int_0^{t_1} d\tau \alpha^*(t_1 - \tau) \langle \Psi_0|\{V_{\tau-t_1} L^\dagger[A, L]\}(t_1)|\Psi_0 \rangle \\ &+ \int_0^{t_1} d\tau \alpha(t_1 - \tau) \langle \Psi_0|\{[L^\dagger, A]V_{\tau-t_1} L\}(t_1)|\Psi_0 \rangle \end{aligned} \quad (13)$$

and for two-time correlations

$$\begin{aligned} \frac{d}{dt_1} \langle \Psi_0|A(t_1)B(t_2)|\Psi_0 \rangle &= i\langle \Psi_0|\{[H_S, A]\}(t_1)B(t_2)|\Psi_0 \rangle \\ &+ \int_0^{t_1} d\tau \alpha^*(t_1 - \tau) \langle \Psi_0|\{V_{\tau-t_1} L^\dagger[A, L]\}(t_1)B(t_2)|\Psi_0 \rangle \\ &+ \int_0^{t_1} d\tau \alpha(t_1 - \tau) \langle \Psi_0|\{[L^\dagger, A]V_{\tau-t_1} L\}(t_1)B(t_2)|\Psi_0 \rangle \\ &+ \int_0^{t_2} d\tau \alpha(t_1 - \tau) \langle \Psi_0|\{[L^\dagger, A]\}(t_1)\{[B, V_{\tau-t_2} L]\}(t_2)|\Psi_0 \rangle \end{aligned} \quad (14)$$

As noted in [15,16], while the first two terms of (14) are analogous to those of (13), the equation for two-time correlations contains an additional term that does not appear in the evolution of quantum mean values. Since the Quantum Regression Theorem (QRT) [17,18,7] states that the evolution of two-time correlations has the same coefficients as the evolution of one-time correlations, these equations show that the QRT theorem is not satisfied for non-Markovian

interactions. A detailed discussion on the QRT may be found in [19–21]. For Markovian interactions the last term of (14) vanishes, since the corresponding correlation function $\alpha(t_1 - \tau) = \Gamma\delta(t_1 - \tau)$ is zero in the domain of integration from 0 to t_2 , so that the QRT is valid in this case. Therefore, the equation (14) shall be used in general to evaluate the evolution of non-Markovian two-time correlations.

In this scheme Eqs. (13) and (14) are just the first two equations of a full hierarchy of equations that form a closed system in the sense that N -time correlation function depend at most on other N -time correlations [14].

Alternatively to (13) and (14) it is possible to compute a particular multiple time correlation function with an stochastic scheme. We shall summarize the main results of this procedure in the following section.

2.2. Stochastic scheme

If we take the partial interaction picture with respect to the environment, the N -time correlation function is defined as

$$C_A(\mathbf{t}|\Psi_0) = \langle \Psi_0 | \prod_{i=1}^N \mathcal{U}_I^{-1}(t_i, 0) A_i \mathcal{U}_I(t_i, 0) | \Psi_0 \rangle$$

where \mathcal{U}_I is the evolution operator of the system in the interaction picture. Since the environment is composed of a set of harmonic oscillators, a convenient choice is a coherent state basis, $|z_1, z_2, \dots, z_n, \dots\rangle = |z\rangle$ in the Bargmann representation [7,22]. In this basis the resolution of the identity is given by $1 = \int d\mu(z) |z\rangle\langle z|$ with $d\mu(z) = \prod_i (d^2z_i \exp(-|z_i|^2)/\pi)$. Within this representation,

$$C_A(\mathbf{t}|\Psi_0) = \int d\mu(z) \langle \psi_0 | G^{-1}(0, 1) \prod_{i=1}^N A_i G(i, i+1) | \psi_0 \rangle \tag{15}$$

with $t_0 = 0, t_{N+1} = 0$ and $z_{N+1} = z_0$. We have introduced the *reduced propagators*

$$G(i, i+1) \equiv G(z_i^* z_{i+1} | t_i t_{i+1}) = \langle z_i | \mathcal{U}_I(t_i, t_{i+1}) | z_{i+1} \rangle$$

which act on the system Hilbert space and give the evolution of system state vectors from t_{i+1} to t_i , given that in the same time interval the environment coordinates go from z_{i+1} to z_i . It is clear then that once their time evolution of the reduced propagators is solved, the time-correlation function (15) can be obtained. It can be shown that the reduced propagator satisfies the evolution equation [15]

$$\begin{aligned} \frac{\partial G(i, i+1)}{\partial t_i} &= (-iH_S + Lz_{i,t_i}^* - L^\dagger z_{i+1,t_i}) G(i, i+1) \\ &\quad - L^\dagger \int_{t_{i+1}}^{t_i} d\tau \alpha(t_i - \tau) \langle z_i | \mathcal{U}_I(t_i, t_{i+1}) L(\tau, t_{i+1}) | z_{i+1} \rangle \end{aligned} \tag{16}$$

with

$$\begin{aligned} L(t', t) &= e^{iH_B t'} e^{-iH(t-t')} L e^{iH(t-t')} e^{-iH_B t'} \\ z_{i,t} &= i \sum_n g_n z_{i,n} e^{i\omega_n t} \\ \alpha(t - \tau) &= \sum_n |g_n|^2 e^{-i\omega_n(t-\tau)} \end{aligned}$$

and the initial condition $G(i, i+1) = \exp(z_i^* z_{i+1})$. Thus the function $z_{i,t}$ is a sum of time dependent coherent states, and $\alpha(t - \tau)$ is their time autocorrelation function, as it can be verified by computing the average $M[z_{i,t} z_{i,\tau}^*]$ regarding the measure $d\mu(z)$. In general the integration of Eq. (16) cannot be computed exactly or expressed as a function of the particular reduced propagator that is evolved. Rather, the evolution of a certain propagator depends on a sum over other propagators. To see that, let us express the last term of Eq. (16) in the following way,

$$\begin{aligned} \langle z_i | \mathcal{U}_I(t_i, \tau) L \mathcal{U}_I(\tau, t_{i+1}) | z_{i+1} \rangle &= \mathcal{M}_I[\langle z_i | \mathcal{U}_I(t_i, \tau) | z_l \rangle L \langle z_l | \mathcal{U}_I(\tau, t_{i+1}) | z_{i+1} \rangle] \\ &= \mathcal{M}_I[G(z_i^* z_l | t_i \tau) L G(z_l^* z_{i+1} | \tau t_{i+1})] \end{aligned} \tag{17}$$

where in the second line we have inserted $1 = \int \frac{d^2z}{\pi} e^{-|z|^2} |z\rangle\langle z|$, and we have defined

$$\mathcal{M}_I[\dots] = \int d\mu(z_I) \dots \quad (18)$$

In that notation, Eq. (16) can be rewritten as

$$\begin{aligned} \frac{\partial G(z_i^* z_{i+1} | t_i t_{i+1})}{\partial t_i} &= (-iH_S + Lz_{i,t_i}^* - L^\dagger z_{i+1,t_i}) G(z_i^* z_{i+1} | t_i t_{i+1}) \\ &\quad - L^\dagger \int_{t_{i+1}}^{t_i} d\tau \alpha(t_i - \tau) \mathcal{M}_I[G(z_i^* z_I | t_i \tau) L G(z_I^* z_{i+1} | \tau t_{i+1})] \end{aligned} \quad (19)$$

In this equation, the last term expresses how the dissipation at time t depends on previous trajectories of other system propagators [23].

It is clear that Eq. (16) cannot in general be expressed in terms of the particular propagator evolved $G(i, i + 1)$, and hence it is not a closed equation for this propagator. Only in very exceptional cases this can be done in an exact way, while in the majority of the systems it is necessary to perform some approximations to close the equation.

One possible approximation is to assume that

$$\langle z_i | \mathcal{U}_I(t_i, t_{i+1}) L(\tau, t_{i+1}) | z_{i+1} \rangle = O(z_{i+1} z_i, t_{i+1}, \tau) G(i, i + 1)$$

where the operator O has to be constructed [24], for instance by treating $L(\tau, t_{i+1})$ in the weak coupling limit. In terms of $O(z_{i+1} z_i, t_{i+1}, \tau)$ Eq. (16) reads

$$\frac{\partial G(i, i + 1)}{\partial t_i} = \left(-iH_S + Lz_{i,t_i}^* - L^\dagger z_{i+1,t_i} - L^\dagger \int_{t_{i+1}}^{t_i} d\tau \alpha(t_i - \tau) O(z_{i+1} z_i, t_{i+1}, \tau) \right) G(i, i + 1) \quad (20)$$

Eqs. (16) or (20), depend on two time dependent functions, z_{i,t_i}^* and z_{i+1,t_i} , which take into account the *history* of the environment and lead to a conditioned dynamics of the system with respect to the environment dynamics. They constitute the starting point to compute the MTCFs in the non-Markovian case stochastically within a Monte Carlo method choosing the variables z_i randomly according the distribution $d\mu(z)$. For a single realization, a value of the integrand appearing in (15) can be obtained; first, evolving $|\psi_0\rangle$ from $(t_{N+1} = 0, z_{N+1} = z_0)$ to (t_N, z_N) so that a vector $|\phi_N\rangle = G(N, N + 1)|\psi_0\rangle$; second, applying A_N to $|\phi_N\rangle$ so that we get $|\tilde{\phi}_N\rangle = A_N|\phi_N\rangle$, third, evolving $|\tilde{\phi}_N\rangle$ with $G(N - 1, N)$, and so on. The process continues until the vector $|\phi_1\rangle = G(1, 2)|\tilde{\phi}_2\rangle$ is obtained and finally used to compute $\langle \psi_1 | A_1 | \phi_1 \rangle$, with $|\psi_1\rangle = G(0, 1)|\psi_0\rangle$. In the end, the sum over many of these *histories* respect to the measure $d\mu(z)$ leads to the MTCFs defined in (15).

Notice that since the equation for the reduced propagator (16) is made for an initial state of the environment different from vacuum, it can be used to compute the expectation values and correlation functions of system observables with more general initial conditions than the one usually taken, i.e. $|\Psi_0\rangle = |\psi_0\rangle|\text{vacuum}\rangle$ [12].

The choice of the stochastic method or the system of equations for computing the MTCF has to be made according to the particular problem. When a N -time correlation function has to be computed with the second method, the system of equations will contain all possible correlations of the matrices \mathcal{Y} that form a basis for the QOS. The correlation of other system observables can be computed by combining correlations of this basic set of observables. In turn, the stochastic method allows us to compute only the particular correlation function that is needed, and not the whole set of \mathcal{Y}^N correlations that appears interrelated in the set of differential equations. Hence, if the system has a large number of freedom, so that \mathcal{Y} is a large set, the stochastic method is in general more convenient.

In the following section, we analyze the spontaneous emission spectra of a two level system, where the basis is $\{\sigma_x, \sigma_y, \sigma_z\}$, so that $\mathcal{Y} = 3$. Since it is a system with a low number of degrees of freedom, we evaluate the two-time correlation functions that are needed by using the system of equations. The equivalence between the two methods, system of equations and the stochastic one, is studied in [12,15,16].

3. Example: an atomic emission spectrum

To illustrate the theory let us discuss the particular example of the spontaneous emission of an atom, which is computed by using the evolution of a two-time correlation of the atomic coupling operator with the field L .

In this section, we derive the formula necessary to obtain the emission spectra of a set of two level atoms in non-Markovian interaction with the surrounding radiation field. We follow a well known photodetection model of experiment, the *gedanken spectrum analyzer*, that provides an operational definition of the spectral profile [25]. The Hamiltonian of the emitting atom (with levels $|1\rangle$ and $|2\rangle$) is given by

$$H_S = -\frac{\omega_{12}}{2}(\sigma_{22} - \sigma_{11}) = \frac{\omega_{12}}{2}\sigma_z \quad (21)$$

where $\sigma_{i,j} = |i\rangle\langle j|$, with $\{i, j\} = 1, 2$, are the atomic pseudospin operators in the atomic basis. The total Hamiltonian H_R that describes the emitting atom and the radiation field is given by $H_R = H_S + H_B + \sum_{\lambda} g_{\lambda}(L^{\dagger}a_{\lambda} + a_{\lambda}^{\dagger}L)$. In order to detect the emitted radiation, suppose we have a detecting atom placed at \mathbf{r} with Hamiltonian $H_{\mathcal{D}} = \Omega\sigma_z/2$, where Ω is its rotating frequency. The Hamiltonian of the total system (detector atom, emitting atom and radiation field) is

$$H = H_{\mathcal{D}} + H_R + W \quad (22)$$

Here the coupling between the detecting atom $H_{\mathcal{D}}$ and the emitting atom and the radiation field H_R is dipolar and it is given by a Hamiltonian W , which in interaction image with respect to the detector is given by

$$W(t) = [\sigma_{21}\mathbf{d}^{\mathcal{D}} \cdot \mathbf{E}^{(+)}(\mathbf{r}, t)e^{i\Omega t} + \sigma_{12}\mathbf{d}^{\mathcal{D}} \cdot \mathbf{E}^{(-)}(\mathbf{r}, t)e^{-i\Omega t}] \quad (23)$$

where we have considered $d_{21}^{\mathcal{D}}\hat{\mathbf{d}}^{\mathcal{D}} = d_{12}^{\mathcal{D}}\hat{\mathbf{d}}^{\mathcal{D}} = \langle 1|\mathbf{D}|2\rangle = \mathbf{d}^{\mathcal{D}}$. The superindex \mathcal{D} reminds that these are the components of the detector's dipole. It is important to note here that the field operators $\mathbf{E}^{(+)}$ and $\mathbf{E}^{(-)}$ correspond to the radiating atoms and the radiation field described by H_R . The positive part of the field at the position \mathbf{r} is defined as

$$\mathbf{E}^{(+)}(\mathbf{r}, \mathbf{r}_0, t) = \sum_{\lambda} \epsilon_{\lambda} A_{\lambda}(\mathbf{r}) a_{\lambda}(\mathbf{r}_0, t) \mathbf{e}_{\lambda} \quad (24)$$

and $\mathbf{E}^{(-)}(\mathbf{r}, \mathbf{r}_0, t) = [\mathbf{E}^{(+)}(\mathbf{r}, \mathbf{r}_0, t)]^{\dagger}$ [26]. In the last expression (and from now on) we have added explicitly the dependency on the position \mathbf{r}_0 of the source dipole (or emitting atom) where the field originates. The quantity $\epsilon_{\lambda} = \sqrt{\frac{\omega_{\lambda}}{2\epsilon_0 v}}$, with v the quantization volume. In terms of the coupling strengths we find that $g_{\lambda} \equiv g_{\lambda}(\mathbf{r}) = \epsilon_{\lambda} A_{\lambda}(\mathbf{r}) \mathbf{d} \cdot \mathbf{e}_{\lambda}$.

A shutter is placed between the radiating atom and the detector. In that way, only at the time T in which the shutter is open the radiation does illuminate the detector. In order to excite the detector, the time of observation T needs to be much bigger than the inverse of the natural width Γ of the detecting atom excited level. In addition, T should be larger than the reciprocal of the spectral width $1/\Gamma$ of the emitting atom. With this set up, the spectral distribution of the fluorescence light, $P(\Omega, T)$, is defined as the probability of excitation of the detecting atom at the time of observation T , i.e.

$$P(\Omega, T) = Tr_{R, \mathcal{D}}(|2\rangle\langle 2|\rho(T)), \quad (25)$$

where $\rho(T)$ is the density matrix of the total system at time T . Replacing the Taylor expansion of the density matrix $\rho(T)$ for $\rho(T) \approx \rho(0)$, and after some manipulations, $P(\Omega, T)$ is obtained as

$$P(\Omega, T) = \int_0^T dt \int_0^T dt' e^{i\Omega(t-t')} g^{(1)}(\mathbf{r}, \mathbf{r}_0; t, t') \quad (26)$$

where the average $\langle \dots \rangle = Tr_R(\rho_R \dots)$, and we have defined

$$g^{(1)}(\mathbf{r}, \mathbf{r}_0; t, t') = \langle \mathbf{d}^{\mathcal{D}} \cdot \mathbf{E}^{(-)}(\mathbf{r}, \mathbf{r}_0, t) \mathbf{d}^{\mathcal{D}} \cdot \mathbf{E}^{(+)}(\mathbf{r}, \mathbf{r}_0, t') \rangle \quad (27)$$

as the first order correlation of the projection of the emitted field in the direction of the dipole. In the last expression, the operators $\mathbf{d}^{\mathcal{D}} \cdot \mathbf{E}^{(-)}(\mathbf{r}, \mathbf{r}_0, t)$ and $\mathbf{d}^{\mathcal{D}} \cdot \mathbf{E}^{(+)}(\mathbf{r}, \mathbf{r}_0, t')$ should be replaced by their expression in terms of the system operators L^{\dagger} and L respectively. This is done by inserting in (24), and in its complex conjugated, the solution (3) for $a_{\lambda}^{\dagger}(t, 0)$ and $a_{\lambda}(t', 0)$ respectively. Taking into account that the quantum vacuum field, corresponding to the term with $a_{\lambda}^{\dagger}(0, 0) a_{\lambda}(0, 0)$, does not contribute to photodetection signals, since the field is in the vacuum state $|0\rangle$, then

$$P(\Omega, T) = \int_0^T dt \int_0^T dt' e^{i\Omega(t-t')} \left\{ \int_0^t d\tau \int_0^{t'} d\tau' \alpha^*(t-\tau) \alpha(t'-\tau') \langle L^{\dagger}(\tau) L(\tau') \rangle \right\} \quad (28)$$

where we have assumed that the coupling constants of the detector and the atom to the field are equal and that the spatial dependence in (27) can be neglected. A more general treatment can be found in [27].

This formula emphasizes the role of the system fluctuations $\langle L^\dagger(\tau)L(\tau') \rangle$ in the observable quantities as is the case of the power spectrum of the emitted light.

Here it has been assumed that there is no spatial dependency of the environment correlation function. More details of the derivation are found in [27].

In the Markov case, the environmental correlation is a delta function, $\alpha(t - \tau) = \Gamma\delta(t - \tau)$, and the last formula is just

$$P(\Omega, T) = \Gamma^2 \int_0^T dt \int_0^T dt' e^{i\Omega(t-t')} \langle L^\dagger(t)L(t') \rangle \quad (29)$$

which in the equilibrium, and with the observation time $T \rightarrow \infty$ leads to the usual expression for the power spectrum [7].

In the Markov case the system correlations $\langle L^\dagger(0)L(\tau) \rangle$ can be computed with the Quantum Regression Theorem. In the non-Markovian case, we cannot assume that the correlation function is a delta, and it is necessary to use the original formula (28) for the spectrum, and the system of equations (14) in order to compute the system correlations.

3.1. A two-level atom with non-Markovian interaction

Once the equations to evaluate the dynamical quantities of the system, as well as its emission spectrum have been presented, we illustrate them with the particular example of a two-level system described by $H_S = \frac{\omega_S}{2}\sigma_z$, and an interaction operator $L = \sigma_{12}$. As correlation function, we use a Gaussian, $\alpha(t) = g^2\gamma \exp(-\gamma^2 t^2)$, which becomes equal to a δ -function in the limit of large γ . In that way, the variation of γ from large to small values permits us to study the transition between the Markovian and the non-Markovian interaction. The evolution of quantum mean values of system operators, given by (6), is illustrated in Fig. 1 for different values of γ .

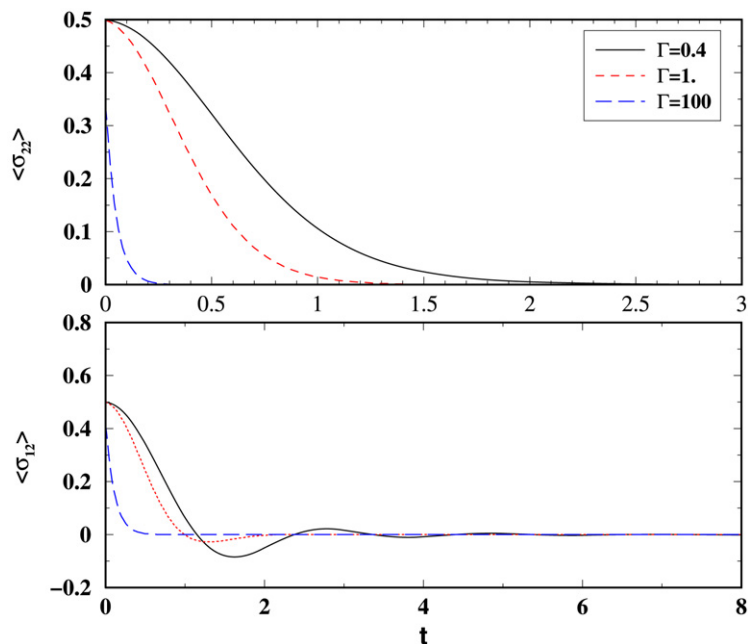


Fig. 1. The upper figure represents the evolution of the atom upper level population, while the lower displays the evolution of the atomic coherence $\langle \sigma_{12} \rangle$. For small γ the environmental correlation time τ_C becomes larger, so that the atomic quantum mean values suffer the non-exponential decaying typical of non-Markovian interactions. On the other hand, τ_C is negligible in contrast with the atomic relaxation time when $\gamma = 100$, so that no memory effects appear in the dynamics and the atomic excited level population and coherences decay exponentially to zero.

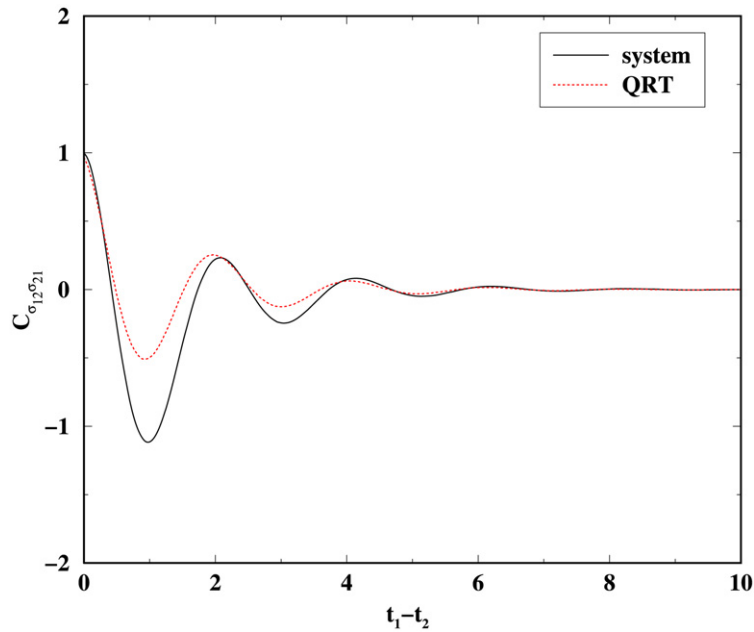


Fig. 2. We display the evolution of the two-time correlation $C_{\sigma_{12}\sigma_{21}} = \langle \Psi_0 | \sigma_{12}(t_1) \sigma_{21}(t_2) | \Psi_0 \rangle$ for $t_2 = 10$. Since for this correlation the last term of (14) is non-zero, the QRT does not apply. This is clearly shown in the picture when comparing the results predicted by this theorem (dotted lines) with that predicted by (14) (solid line).

Let us now represent the solution of (14), compared to that predicted by the Quantum Regression Theorem. As displayed in Fig. 2, when the correlation of operators is chosen in such a way that the last term of (14) does not vanish, then there is a clear departure from the result of the QRT. As stated above, this last term is typical from non-Markovian interactions, since it represents a dependency of the correlation $C_{AB}(t_1, t_2)$ on the past history that goes from 0 to t_2 .

Let us now use formulas (28) and (29) to compute the non-Markovian and Markovian spectra respectively for $\omega_{12} = 1$ (see Fig. 3). The non-Markovian case corresponds to γ chosen sufficiently small so that the correlation function decays within a non-zero correlation time. Since we are dealing with spontaneous emission processes, in which the correlation functions $\langle L^\dagger(t)L(t') \rangle$ relax to zero, we choose the observing time of the detector $T > T_{CA}$, where T_{CA} is the relaxation time of the two-time correlation. Notice that when a laser is tuned to the atomic rotating frequency, then the two-time correlations do not decay to zero, so that the condition $T > T_{CA}$ is not enough to obtain a stationary spectrum. In this case it is necessary to define the spectra in a stationary limit $T \rightarrow \infty$.

4. Conclusions

In this article we have discussed a set of dynamical equations that rule the dynamics of multiple time correlation functions of a system in contact with an environment, in the case that such interaction is non-Markovian. In such systems the evolution equations do not satisfy the Quantum Regression Theorem, and the dynamical equations proposed can be considered as a generalization of the QRT to the non-Markovian case. We have applied the equations to the evaluation of the emission spectrum of an atom in contact with an environment in which non-Markovian features are present. The emission spectrum depends both on the fluctuations of atom observables and on the environmental correlation function, that reflects the finite reaction time of the environment to the absorption of a photon emitted by the atom. In our model, we have considered that the detector has the same nature as the emitting system. As expected in the non-Markovian regime, the emission spectrum does not have a Lorentzian profile as it would be the case for a Markovian interaction. The theory presented has been also applied to the evaluation of the fluorescence spectrum of an atom in a photonic crystal [27].

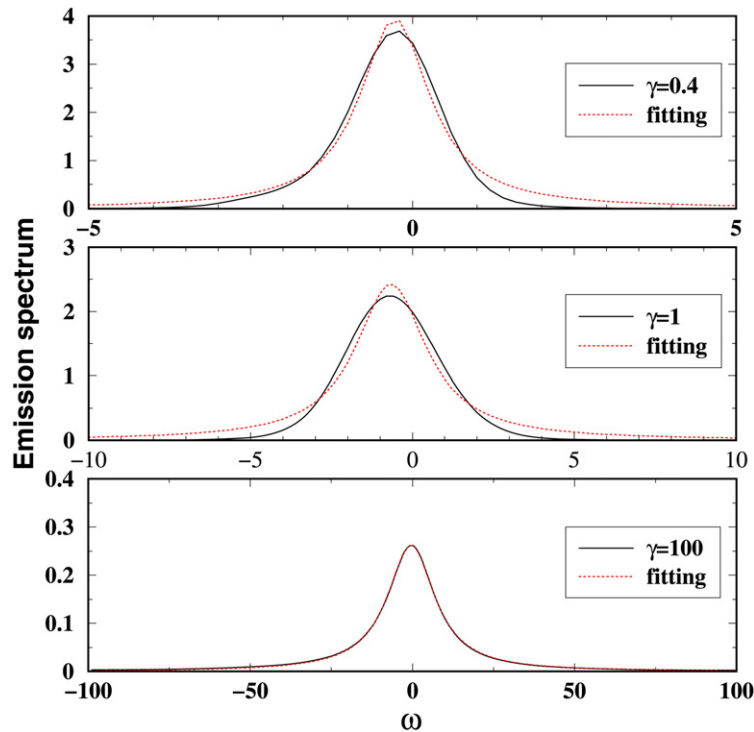


Fig. 3. The spontaneous emission spectrum is computed with the formula (28) for several values of γ , and by choosing $T > T_A$, where T_A is the atomic relaxation time. In order to observe the departure from the Lorentzian profile typical for Markovian interactions, the result is numerically fitted with a Lorentzian function. When γ is small, so that non-Markovian effects are important in the atomic dynamics, the Lorentzian fitting is not appropriate, which means that the use of the formula (28) is necessary to compute the spectrum. For $\gamma = 100$ the interaction is practically Markovian, and the corresponding spectra corresponds perfectly to a Lorentzian.

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