Non-Markovian reduced propagator, multiple-time correlation functions, and master equations with general initial conditions in the weak-coupling limit

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In this paper we derive the evolution equation for the reduced propagator, an object that evolves vectors of the Hilbert space of a system S interacting with an environment B in a non-Markovian way. This evolution is conditioned to certain initial and final states of the environment. Once an average over these environmental states is made, reduced propagators permit the evaluation of multiple-time correlation functions of system observables. When this average is done stochastically the reduced propagator evolves according to a stochastic Schrödinger equation. In addition, it is possible to obtain the evolution equations of the multiple-time correlation functions which generalize the well-known quantum regression theorem to the non-Markovian case. Here, both methods, stochastic and evolution equations, are described by assuming a weak coupling between system and environment. Finally, we show that reduced propagators can be used to obtain a master equation with general initial conditions, and not necessarily an initial vacuum state for the environment. We illustrate the theory with several examples.

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I. INTRODUCTION AND MOTIVATION

Quantum open systems are found in many different situations in physics. Usually one is interested in the description of a quantum system (S) in contact with an environment (B) and one needs to compute the expectation values and fluctuations of observables defined for S. Many examples are found in statistical physics, condensed matter, and quantum optics. The dynamics of an atom interacting with an electromagnetic field is a typical example. The atom plays the role of S and the electromagnetic field the role of B.

The dynamics of the system is described by its reduced density matrix, which is obtained by an average of the density matrix of the total system $S \oplus B$ over the environmental degrees of freedom. The dynamics of the reduced density matrix is ruled by some master equation that in the Markovian case is of Lindblad type [1-6].

There is a complementary scheme to the master equation that consists in deriving stochastic Schrödinger equations that evolve state vectors in the Hilbert space of S, conditioned by the dynamics of the environment. The effects of the environment are included in the Schrödinger equation through a time-dependent function that eventually may be considered as a noise, and through the environmental correlation function. The average over many realizations of the noise leads to the computation of the required quantities. Depending on the method used in its derivation, there are many different stochastic equations or unravelings of the reduced density matrix. Some of these stochastic schemes are related to specific detection procedures such as the homodyne and the heterodyne detection [3,4,7-18].

The first master equation for non-Markovian interactions was derived by Redfield in the context of nuclear magnetic resonance [19,20]. The derivation of non-Markovian stochastic Schrödinger equations is more recent [21-25]. In addition, stochastic schemes can be used to derive master equations [23]. It is very common to assume an initial density matrix for the total system (composed of quantum open system and environment) corresponding to a decorrelated state, that is the product of a density matrix of the system times the density matrix of the environment, which can be considered in thermal equilibrium [4].

In all the above-mentioned formulations it is possible to compute the expectation values of S observables in a satisfactory manner. Nonetheless, sometimes it is interesting to compute some fluctuation properties, which are characterized by certain correlation functions of system observables. For instance, in quantum optics the evaluation of the spectrum of an emitting dipole, or the analysis of photon statistics, requires the evaluation of two-time and four-time correlation functions, respectively.

For Markovian interactions, since the seminal work of Lax and Onsager, there is a theory to compute multiple-time correlation functions (MTCFs), the quantum regression theorem (QRT) [4,6,26–28]. Moreover, in the context of stochastic Schrödinger equations there exists a complementary theory of multiple-time correlation functions that agrees with the QRT (see, for instance, Refs. [4,29,30]). It is natural to develop a theory to compute multiple-time correlation functions for systems where non-Markovian effects are relevant. Interesting examples of these systems are atoms immersed in photonic crystals (PCs) [31]. In PCs the refraction index is periodic, which produces Bragg scattering of photons with wavelengths related to the periodicity of the refraction index. As a consequence, these photonic modes do not appear within the crystal, and the dispersion relation of the electromagnetic field displays a band structure interrupted with gaps of forbidden frequencies. In such structured materials the correlation function of the electromagnetic field is highly non-Markovian, particularly within the edges of the bands, and the dynamics of an atom in interaction with such field presents non-Markovian effects. This has been shown in several works that study the evolution of the atomic quantum mean values with master equations [31–34], and with non-Markovian stochastic Schödinger equations [35]. It is therefore pertinent to be able to evaluate other important dynamical quantities of those systems, such as MTCFs.

A theory of non-Markovian MTCFs has been recently introduced in Ref. [36]. In this paper we present a detailed presentation of that theory and propose a master equation for initially correlated states between system and environment.

The paper is organized as follows. Multiple-time correlation functions are defined in Sec. II in terms of the reduced propagator, which evolves the system state vector conditioned to the state of the environment. The evolution equation of this propagator is also derived in this section and the stochastic method for computing MTCFs is explained. In Sec. III the set of evolution equations for MTCFs in the weak-coupling limit is obtained. Two examples are shown in which MTCFs are calculated using two alternative approaches; a stochastic method or a system of differential equations. We analyze an example of a solvable system and a two-level atom in contact with a dissipative environment. In Sec. V the master equation for general initial conditions is obtained within the weak-coupling limit. This equation is applied to the solvable model and to the problem of an atom in contact with a thermal reservoir. Finally, some conclusions and remarks are presented in Sec. VI.

II. MULTIPLE-TIME CORRELATION FUNCTIONS

We consider a class of systems modeled by the Hamiltonian

$$H = H_S + LB^{\dagger} + L^{\dagger}B + H_B$$

= $H_S + \sum_n g_n (La_n^{\dagger} + L^{\dagger}a_n) + \sum_n \omega_n a_n^{\dagger}a_n,$ (1)

where L is an operator that acts in the Hilbert space of the system and a_n and a_n^{\dagger} are the annihilation and creation operators that act on the Hilbert space of the environment. H_I is the Hamiltonian describing the interaction between the system and the environment. The g'_n s are the coupling constants that can be taken as real numbers, and the ω'_n s are the frequencies of the harmonic oscillators that constitute the environment [37]. Throughout the paper we will make expansions in a small coupling parameter g, which gives the difference in magnitude between the interaction Hamiltonian H_I and the so-called free term of the Hamiltonian, $H_0 = H_S$ $+H_B$, in such a way that $g[H_0] = [H_I]$ (where [A] denotes the magnitude of A). Instead of a single coupling system operator L in the interaction Hamiltonian we could consider a set of them, but such generalization is straightforward and does not affect the conclusions we shall derive in this paper.

We are interested in a *N*-time correlation function of system observables in the Heisenberg representation, $\{A_1(t_1), \ldots, A_N(t_N)\} = \mathbf{A}(\mathbf{t})$. These correlation functions are defined as

$$C_{A_1,A_2,\ldots,A_N}(t_1,t_2,\ldots,t_N|\Psi_0) = C_{\mathbf{A}}(\mathbf{t}|\Psi_0)$$
$$= \langle \Psi_0 | A_1(t_1)\cdots A_N(t_N) | \Psi_0 \rangle.$$
(2)

Let us make a comment about the *time ordering* in multipletime correlation functions. In this paper we use the time ordering $t_1 > t_2 > \cdots > t_N$ and denote $\mathbf{t} = \{t_1, \ldots, t_N\}$. A different type of correlations may be defined as $C_{\mathbf{A}',\mathbf{A}}(\mathbf{t}',\mathbf{t}|\Psi_0) = \langle \Psi_0 | A'_N(t'_N) \cdots A'_2(t'_2)A_1(t_1) \cdots A_N(t_N) | \Psi_0 \rangle$, with the time ordering such that $t'_N \leq t'_{N-1} \leq \cdots \leq t'_2 \leq t_1$ and $t_N \leq t_{N-1} \leq \cdots$ $\leq t_2 \leq t_1$. These 2N-1-time correlation functions can correspond to a sequence of measurements performed over the system. Nevertheless, we stress that although we only treat here the correlations of type $C_{\mathbf{A}}(\mathbf{t}|\Psi_0)$ and not of type $C_{\mathbf{A}',\mathbf{A}}(\mathbf{t}',\mathbf{t}|\Psi_0)$, the method derived here can be used *in any* given time ordering for the multiple-time correlation functions.

The initial state of the full system is taken as the tensor product of a system state $|\psi_0\rangle$ and the environment state $|z_0\rangle$, i.e., $|\Psi_0\rangle = |\psi_0\rangle |z_0\rangle$. Let us point out that more general initial states can also be treated. In terms of the evolution operator in the interaction picture U_l , Eq. (2) can be written as

3.7

$$C_{\mathbf{A}}(\mathbf{t}|\Psi_0) = \langle \Psi_0 | \prod_{i=1}^N \mathcal{U}_I^{-1}(t_i, 0) A_i \mathcal{U}_I(t_i, 0) | \Psi_0 \rangle.$$
(3)

The bath is composed of a set (possibly infinite) of harmonic oscillators. A suitable basis to treat this system is a coherent state basis in Bargmann representation. The coherent states in Bargmann representation $|z\rangle$ are eigenstates of the annihilation operator $a|z\rangle = z|z\rangle$. In terms of these coherent states the resolution of the identity is given by $1 = \int d\mu(z)|z\rangle\langle z|$, with $d\mu(z) = \exp(-(|z|^2)/\pi$ [4,38].

We introduce the *reduced propagator* as

$$G(z_i^* z_{i+1} | t_i t_{i+1}) = \langle z_i | \mathcal{U}_I(t_i, t_{i+1}) | z_{i+1} \rangle.$$
(4)

This object acts in the Hilbert space of the system and gives the evolution of a vector from t_{i+1} to t_i , conditioned that the environmental coordinates go from z_{i+1} to z_i in the same time interval. Introducing the identity operator in the bath Hilbert space in Eq. (3) and defining $t_0=0$, $t_{N+1}=0$, and $z_{N+1}=z_0$ it follows that

$$C_{\mathbf{A}}(\mathbf{t}|\Psi_{0}) = \int d\mu(z) \langle \psi_{0}| G^{\dagger}(z_{0}^{*}z_{1}|t_{0}t_{1}) \prod_{i=1}^{N} A_{i}G(z_{i}^{*}z_{i+1}|t_{i}t_{i+1})|\psi_{0}\rangle.$$
(5)

If the evolution for the reduced propagators is solved, then the time correlation function can be computed. To obtain the time evolution of the reduced propagator we have to generate its dynamical equation. To that end we have to compute

$$\frac{\partial G(z_i^* z_{i+1} | t_i t_{i+1})}{\partial t_i} = \left\langle z_i \left| \frac{\partial \mathcal{U}_I(t_i, t_{i+1})}{\partial t_i} \right| z_{i+1} \right\rangle.$$
(6)

The evolution operator U_I satisfies the Schrödinger equation in the partial interaction picture,

$$\frac{\partial \mathcal{U}_{I}(t_{i}, t_{i+1})}{\partial t_{i}} = \left(-iH_{S} - i\sum_{n} g_{n}(L^{\dagger}a_{n}e^{-i\omega_{n}t_{i}} + La_{n}^{\dagger}e^{i\omega_{n}t_{i}})\right)$$
$$\times \mathcal{U}_{I}(t_{i}, t_{i+1}). \tag{7}$$

When inserted in Eq. (6) this equation leads to

$$\frac{\partial G(z_i^* z_{i+1} | t_i t_{i+1})}{\partial t_i} = \left(-iH_S - iL \sum_n g_n e^{i\omega_n t_i} z_{i,n}^* \right) G(z_i^* z_{i+1} | t_i t_{i+1})$$
$$-iL^{\dagger} \sum_n g_n e^{-i\omega_n t_i} \langle z_i | a_n \mathcal{U}_I(t_i, t_{i+1}) | z_{i+1} \rangle, \quad (8)$$

where we have used $\langle z | a_n^{\dagger} = \langle z | z_n^*$. This equation is not yet in a useful form to be solved in the system Hilbert space due to the last term on the right-hand side. To proceed further we have to treat the matrix element $\langle z_i | a_n \mathcal{U}_l(t_i, t_{i+1}) | z_{i+1} \rangle$ which it is equal to $\langle z_i | \mathcal{U}_l(t_i, t_{i+1}) a_n(t_i, t_{i+1}) | z_{i+1} \rangle$, with $a_n(t_i, t_{i+1}) = \mathcal{U}_l^{-1}(t_i, t_{i+1}) a_n \mathcal{U}_l(t_i, t_{i+1})$. Integrating the Heisenberg equations of motion for $a_n(t_i, t_{i+1})$ it follows that

$$a_n(t_i, t_{i+1}) = a_n(t_i, t_i) - ig_n \int_{t_{i-1}}^{t_i} d\tau L(\tau, t_{i+1}) e^{i\omega_n \tau}, \qquad (9)$$

with

$$L(\tau, t_{i+1}) = e^{iH_B t_{i+1}} e^{-iH(t_{i+1}-\tau)} L e^{iH(t_{i+1}-\tau)} e^{-iH_B \tau}.$$
 (10)

Gathering the results, Eq. (8) becomes

$$\frac{\partial G(z_i^* z_{i+1} | t_i t_{i+1})}{\partial t_i} = (-iH_S + L z_{i,t_i}^* - L^{\dagger} z_{i+1,t_i}) G(z_i^* z_{i+1} | t_i t_{i+1}) - L^{\dagger} \int_{t_{i+1}}^{t_i} d\tau \alpha(t_i - \tau) \langle z_i | \mathcal{U}_I(t_i, t_{i+1}) L(\tau, t_{i+1}) | z_{i+1} \rangle, \quad (11)$$

where we define the functions

$$z_{i,t} = i \sum_{n} g_n z_{i,n} e^{i\omega_n t},$$
(12)

and

$$\alpha(t-\tau) = \sum_{n} |g_n|^2 e^{-i\omega_n(t-\tau)}.$$
(13)

The function $z_{i,t}$ is a sum over time-dependent coherent states and $\alpha(t-\tau)$ is its time autocorrelation function, as it can be easily verified by computing the average $\mathcal{M}[z_{i,t}z_{i,\tau}^*]$ with respect to the measure $d\mu(z_i) = \prod_n (d^2 z_{i,n} \exp(-|z_{i,n}|^2)/\pi)$. Here and throughout the paper, we define the Gaussian average $\mathcal{M}_i[\cdots] = \int d\mu(z_i)\cdots$. From Eq. (11) we can integrate the reduced propagators with the initial conditions $G(z_i^* z_{i+1} | t_i t_i)$ $= \exp(z_i^* z_{i+1})$. Nonetheless, it is not always possible to compute the matrix element $\langle z_i | \mathcal{U}_i(t_i, t_{i+1}) L(\tau, t_{i+1}) | z_{i+1} \rangle$ exactly and express the result as a function of the reduced propagator, so that Eq. (11) turns into an explicit equation for the reduced propagator. Since only in very exceptional cases can exact solutions be obtained, some approximate scheme has to be taken at this stage. One possible way is to treat $L(\tau, t_{i+1})$ in the weak-coupling limit [23]. In some other circumstances, it is possible to assume that

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

where the operator O has to be constructed [23,35,39]. For this particular case we have

$$\frac{\partial G(z_i^* z_{i+1} | t_i t_{i+1})}{\partial t_i} = (-iH_S + L z_{i,t_i}^* - L^{\dagger} z_{i+1,t_i}) G(z_i^* z_{i+1} | t_i t_{i+1}) - L^{\dagger} \int_{t_{i+1}}^{t_i} d\tau \times \alpha(t_i - \tau) O(z_{i+1} z_i, t, \tau) G(z_i^* z_{i+1} | t_i t_{i+1}).$$
(14)

Equation (11), or its approximate version (14), depends 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. The integration of the equations for the reduced propagators with their initial conditions allows the evaluation of the *N*-time correlation functions previously defined.

Since the environment distribution function is considered constant during the interaction with the system, the equations introduced in this section are linear. This is a good approximation for environments at low temperature. However, its state distribution changes quite sensibly due to the interaction when the environment is at high temperatures [40,41].

It is important to note that because the equation for the reduced propagator corresponds to an initial state of the environment different from the vacuum, it is possible to use it in the evaluation of expectation values of system observables and correlation functions with more general initial conditions that the one usually taken, i.e., $|\Psi_0\rangle = |\psi_0\rangle|0_10_2\cdots 0_n\cdots\rangle = |\psi_0\rangle|vacuum\rangle$. For the sake of simplicity, when computing MTCFs we shall consider the initial vacuum state for the environment. More general environmental initial states will be considered when we compute the master equation for correlated initial conditions.

Traditionally, under the Markovian assumption, the multiple-time correlations are obtained by means of the quantum regression theorem. In the next sections we show how to compute the non-Markovian multiple-time correlation functions by using reduced propagators, and analyze the validity of the quantum regression theorem.

Computing the MTCF: Stochastic sampling and stochastic equations

The solutions of the equations of motion for the reduced propagators, once they are replaced in Eq. (5), are the starting point to compute correlation functions. If we were able to perform the integrations over the coherent state variables z, then we would obtain the MTCF. However, the complex Gaussian integrals over coherent states can only be completely done in the very special case of a solvable model. In general, some approximate or numerical schemes are needed.

One way is to choose at random a set of coherent state coordinates z that are distributed according to the measure $d\mu(z)$. For a single realization, if all other parameters are known, i.e., the bath frequencies ω'_n s and the coupling constants g'_n s, it is possible to construct the functions z_t , Eq. (12), and the correlation function $\alpha(t)$, Eq. (13). The whole information of the system and environment is available, and we just decide to solve the multidimensional integrals over coherent states by a Monte Carlo method. As a consequence, the better is the sampling, the closer we are to the exact solution of the problem, except for the approximations made, if any, in the equations of motion for the reduced propagators as we have already emphasized.

In many applications the detailed information of the environment is not known, i.e., the individual frequencies ω'_n s and coupling constants g'_n s are not accessible. At this point we should keep in mind that the only required information of the environment is its correlation function. If $\alpha(t)$ is known, we can generate a Gaussian distributed set of complex numbers having such correlation function. The synthesis of this complex noise consists in the construction of a bath of oscillators that has the desired correlation function. This would be in a sense a phenomenological description of the interaction between the system and the environment. Once the noise is generated, the equations that rule the dynamics of the system state vectors can be integrated. The average over many realizations of the noise again leads to the MTCF. It turns out that the states of the oscillator that have a major contribution are, first, those closer to the vacuum (at zero temperature), a fact that is encoded in the measure $d\mu(z)$, and, second, the pair of coherent states z_i, z_{i+1} that have a significant overlap $\exp(|z_i^* - z_{i+1}|^2)$. This last point is relevant if we want to have a reliable method of stochastic sampling, and it is a consequence of the initial condition for the reduced propagator. In the case that we consider one time averages of observables, such overlap is irrelevant but becomes important when computing the MTCF.

Once the noise, i.e., the set of coherent states which appears in Eq. (5), has been chosen, the element $\langle \psi_0 | G^{\dagger}(z^*z_1 | 0t_1) \Pi_{i=1}^N A_i G(z_i^*z_{i+1} | t_i t_{i+1}) | \psi_0 \rangle$ can be integrated. If, for instance, we want to compute numerically two-time correlations, it is necessary to make an average with the functions $\langle \psi_0 | G^{\dagger}(0z_1 | 0t_1) A G(z_1^*z_2 | t_1t_2) B G(z_2^{*0} | t_20) | \psi_0 \rangle$ for different noise histories. To construct each function, the following steps must be followed:

(i) Propagate $G(z_2^*0|t_20)|\psi_0\rangle$ from 0 to t_2 , choosing at random the set of $z_{2,n}$ appearing in the function (12). This choice is made according to a Gaussian distribution $d\mu(z)$. In the same way, choose the set of $z_{1,n}$ and evolve the function $|\psi_{t_1}(z_1^*0)\rangle = G(z_1^*0|t_10)|\psi_0\rangle$.

(ii) Apply the matrix B to obtain $|\phi_{t_2}(z_2^*)\rangle = BG(z_2^*0|t_20)|\psi_0\rangle$.

(iii) Propagate $|\phi_{t_2}(z_2^*)\rangle$ from t_2 to t_1 applying the propagator $G(z_1^*z_2|t_1t_2)$, which gives a wave function $|\phi_{t_1,t_2}(z_2^*, z_1^*, z_2)\rangle = G(z_1^*z_2|t_1t_2)|\phi_{t_2}(z_2^*)\rangle$. It is important to stress that the noises z_{1,t_1} and z_{2,t_1} appearing in the evolution of the propagator are calculated respectively with the sets $\{z_{1,t_1}\}$ and $\{z_{2,t_1}\}$ already chosen. Another important point is to take into account that the propagator has as initial condition $G(z_1^*z_2|t_2t_2) = \exp(z_1^*z_2)$, with $z_1^*z_2 = \sum_n z_{1,n}^* z_{2,n}$. When the number of environmental degrees of freedom is large, such initial condition may be a large number, which makes the convergence of the numerical method slower. This problem can be overcome by sampling the environmental spectral function with only a few values, as it is done in the example shown in Sec. V B.

(iv) Reconstruct

$$\begin{split} \langle \psi_0 | G^{\dagger}(0z_1 | 0t_1) A G(z_1^* z_2 | t_1 z_2) B G(z_2^* 0 | t_2 0) | \psi_0 \rangle \\ &= \langle \psi_{t_1}(z_1) | A | \phi_{t_1, t_2}(z_2^*, z_1^*, z_2) \rangle. \end{split}$$

With these steps, we have obtained a member of the stochastic ensemble of objects $\langle \psi_0 | G^{\dagger}(0z_1 | 0t_1) A G(z_1^*z_2 | t_1t_2) \times BG(z_2^*0 | t_20) | \psi_0 \rangle$ that allows a Monte Carlo sampling of the sum (5). Obviously, the more members we add to the sum, the more accurately will be reproduced the resulting MTCF (in this case a two-time correlation).

III. BEYOND THE QUANTUM REGRESSION THEOREM: SYSTEM OF EQUATIONS FOR TWO-TIME CORRELATIONS IN THE WEAK-COUPLING LIMIT

Once we have the multiple-time correlation functions, we may compute them directly from the stochastic method. Nonetheless, this may turn to be an expensive strategy from the numerical point of view, which is especially true when the number of environmental degrees of freedom needed to correctly describe its correlation function is large. Therefore it may be convenient to have a set of differential equations from which the MTCF may be obtained, and where the stochastic average has been done analytically. In this section, we calculate such a set of coupled differential equations which evolve, up to second order in a convenient perturbation parameter g, the non-Markovian two-time correlations. Throughout this section we denote $V_{t_1}L = \exp\{i\mathcal{L}_S t_1\}L$ $=\exp(iH_{S}t_{1})L\exp(-iH_{S}t_{1})$, where $V_{t_{1}}=\exp\{i\dot{\mathcal{L}}_{S}t_{1}\}$ is the free system Liouville operator, acting on both sides of the immediately contiguous system operator. The method we will follow consists in deriving the stochastic two-time correlation with respect to t_1 ,

$$\frac{d}{dt_{1}} \langle \psi_{0} | G^{\dagger}(z_{1}0|t_{1}0) A G(z_{1}^{*}z_{2}|t_{1}t_{2}) B G(z_{2}^{*}0|t_{2}0) | \psi_{0} \rangle
= \langle \psi_{0} | \left(\frac{d}{dt_{1}} G^{\dagger}(z_{1}0|t_{1}0) \right) A G(z_{1}^{*}z_{2}|t_{1}t_{2}) B G(z_{2}^{*}0|t_{2}0) | \psi_{0} \rangle
+ \langle \psi_{0} | G^{\dagger}(z_{1}0|t_{1}0) A \left(\frac{d}{dt_{1}} G(z_{1}^{*}z_{2}|t_{1}t_{2}) \right) B G(z_{2}^{*}0|t_{2}0) | \psi_{0} \rangle,$$
(15)

and then performing analytically the average over the variables z_1 and z_2 . The first derivative appearing in Eq. (15) corresponds to the Hermitian conjugate of the usual linear stochastic Schrödinger equation, which in interaction image reads as follows:

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$$\frac{d}{dt_1}G(z_1^*0|t_10) = \left\{ -iH_S + Lz_{1,t_1}^* - L^{\dagger} \int_0^{t_1} d\tau \alpha(t_1 - \tau) V_{\tau - t_1}L \right\} G(z_1^*0|t_10) + \mathcal{O}(g^3).$$
(16)

The second term is given by the equation

$$\frac{d}{dt_1}G(z_1^*z_2|t_1t_2) = \left\{-iH_S + Lz_{1,t_1}^* - L^{\dagger}z_{2,t_1} - L^{\dagger}\int_{t_2}^{t_1} d\tau\alpha(t_1 - \tau)V_{\tau - t_1}L\right\}G(z_1^*z_2|t_1t_2) + \mathcal{O}(g^3),\tag{17}$$

which is equal to Eq. (14) once $O(z_{i+1}z_i, t, \tau)$ is replaced by its weak-coupling expansion up to zero order [37]. Inserting the last expressions in Eq. (15), we find that

$$\frac{d}{dt_{1}} \langle \psi_{0} | G^{\dagger}(z_{1}0 | t_{1}0) AG(z_{1}^{*}z_{2} | t_{1}t_{2}) BG(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle
= i \langle \psi_{0} | G^{\dagger}(z_{1}0 | t_{1}0) [H_{S}, A] G(z_{1}^{*}z_{2} | t_{1}t_{2}) BG(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle - \int_{0}^{t_{1}} d\tau \alpha^{*}(t_{1} - \tau) \langle \psi_{0} | G^{\dagger}(z_{1}0 | t_{1}0) V_{\tau-t_{1}} L^{\dagger} LAG(z_{1}^{*}z_{2} | t_{1}t_{2}) \\
\times BG(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle - \int_{t_{2}}^{t_{1}} d\tau \alpha(t_{1} - \tau) \langle \psi_{0} | G^{\dagger}(z_{1}0 | t_{1}0) AL^{\dagger} V_{\tau-t_{1}} LG(z_{1}^{*}z_{2} | t_{1}t_{2}) BG(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle \\
+ z_{1,t_{1}} \langle \psi_{0} | G^{\dagger}(z_{1}0 | t_{1}0) L^{\dagger} AG(z_{1}^{*}z_{2} | t_{1}t_{2}) BG(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle + z_{1,t_{1}}^{*} \langle \psi_{0} | G^{\dagger}(z_{1}0 | t_{1}0) ALG(z_{1}^{*}z_{2} | t_{1}t_{2}) BG(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle \\
- z_{2,t_{1}}^{*} \langle \psi_{0} | G^{\dagger}(z_{1}0 | t_{1}0) AL^{\dagger} G(z_{1}^{*}z_{2} | t_{1}t_{2}) BG(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle.$$
(18)

In order to make the averages, we introduce second-order perturbative expansions of these evolution operators, which are

$$G(z_1^*0|t_10) = \left\{ 1 + \int_0^{t_1} d\tau z_{1,\tau}^* V_{\tau-t_1} L + \int_0^{t_1} d\tau \int_0^{\tau} d\tau' z_{1,\tau}^{**} V_{\tau-t_1} L V_{\tau'-t_1} L - \int_0^{t_1} d\tau \int_0^{\tau} d\tau' \alpha(\tau-\tau') V_{\tau-t_1} L^{\dagger} V_{\tau'-t_1} L \right\} \times G^{(0)}(z_1^*0|t_10) + \mathcal{O}(g^3),$$
(19)

for Eq. (16), where $G^{(0)}(z_1^*0|t_10) = \exp(-iH_s t_1)$ represents the zero order in the perturbative expansion, and

$$G(z_{1}^{*}, z_{2}|t_{1}t_{2}) = \left\{ 1 + \int_{t_{2}}^{t_{1}} d\tau z_{1,\tau}^{*} V_{\tau-t_{1}}L - \int_{t_{2}}^{t_{1}} d\tau z_{2,\tau} V_{\tau-t_{1}}L^{\dagger} - \int_{t_{2}}^{t_{1}} d\tau \int_{t_{2}}^{\tau} d\tau' \alpha(\tau-\tau') V_{\tau-t_{1}}L^{\dagger} V_{\tau'-t_{1}}L + \int_{t_{2}}^{t_{1}} d\tau \int_{t_{2}}^{\tau} d\tau' z_{1,\tau}^{*} z_{1,\tau'}^{*} V_{\tau-t_{1}}L V_{\tau'-t_{1}}L - \int_{t_{2}}^{t_{1}} d\tau \int_{t_{2}}^{\tau} d\tau' z_{1,\tau}^{*} z_{2,\tau'} V_{\tau-t_{1}}L V_{\tau'-t_{1}}L^{\dagger} - \int_{t_{2}}^{t_{1}} d\tau \int_{t_{2}}^{\tau} d\tau' z_{2,\tau} z_{1,\tau'}^{*} V_{\tau-t_{1}}L^{\dagger} V_{\tau'-t_{1}}L \right\} G^{(0)}(z_{1}^{*}, z_{2}|t_{1}t_{2}) + \mathcal{O}(g^{3}),$$

$$(20)$$

for Eq. (17), where now the zero order is $G^{(0)}(z_1^*z_2|t_1t_2) = \exp[-iH_s(t_1-t_2)]\exp(z_1^*z_2)$. The average of the term containing z_{1,t_1} in Eq. (18) satisfies

$$\int d\mu(z_{1}) \int d\mu(z_{2}) z_{1,t_{1}} \langle \psi_{0} | G^{\dagger}(z_{1}0 | t_{1}0) L^{\dagger} A G(z_{1}^{*} z_{2} | t_{1} t_{2}) B G(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle$$

$$= \int d\mu(z_{1}) \int d\mu(z_{2}) \langle \psi_{0} | G^{\dagger}(z_{1}0 | t_{1}0) L^{\dagger} A G(z_{1}^{*} z_{2} | t_{1} t_{2}) B G(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle$$

$$= \int d\mu(z_{1}) \int d\mu(z_{2}) z_{1,t_{1}} \langle \psi_{0} | G^{\dagger(0)}(0 z_{1} | 0 t_{1}) \left\{ 1 + \int_{0}^{t_{1}} d\tau z_{1,\tau} V_{\tau-t_{1}} L^{\dagger} \right\} L^{\dagger} A \left\{ 1 + \int_{t_{2}}^{t_{1}} d\tau z_{1,\tau}^{*} V_{\tau-t_{1}} L - \int_{t_{2}}^{t_{1}} d\tau z_{2,\tau} V_{\tau-t_{1}} L^{\dagger} \right\}$$

$$\times G^{(0)}(z_{1}^{*} z_{2} | t_{1} t_{2}) B \left\{ 1 + \int_{0}^{t_{1}} d\tau z_{1,\tau}^{*} V_{\tau-t_{1}} L \right\} G^{(0)}(z_{2}^{*} 0 | t_{2} 0) | \psi_{0} \rangle + \mathcal{O}(g^{3}), \qquad (21)$$

where we have inserted the perturbative expansions of the propagators (19) and (20) up to first order in g, since the term is at least of first order due to the presence of the first-order quantity z_{1,t_1} . The Gaussian integrals are in fact multidimensional integrals over the coordinates of each harmonic oscillator of the environment. Four types of them have to be computed,

$$\int d\mu(z_{2,n}) \int d\mu(z_{1,n}) z_{1,n} z_{1,n} e^{z_{1,n}^{*} z_{2,n}} = 0,$$

$$\int d\mu(z_{2,n}) \int d\mu(z_{1,n}) z_{1,n} z_{1,n}^{*} e^{z_{1,n}^{*} z_{2,n}} = 1,$$

$$\int d\mu(z_{2,n}) \int d\mu(z_{1,n}) z_{1,n} z_{2,n}^{*} e^{z_{1,n}^{*} z_{2,n}} = \int d\mu(z_{1,n}) z_{1,n} z_{1,n}^{*} = 1,$$

$$\int d\mu(z_{2,n}) \int d\mu(z_{1,n}) z_{1,n} z_{2,n} e^{z_{1,n}^{*} z_{2,n}} = 0,$$
(22)

which gives rise to the following result:

$$\int d\mu(z_{1}) \int d\mu(z_{2}) z_{1,t_{1}} \langle \psi_{0} | G^{\dagger}(0z_{1} | 0t_{1}) \\ \times L^{\dagger}AG(z_{1}^{*}z_{2} | t_{1}t_{2}) BG(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle \\ = \mathcal{M}_{1,2} [\langle \psi_{0} | G^{\dagger}(0z_{1} | 0t_{1}) L^{\dagger}AG(z_{1}^{*}z_{2} | t_{1}t_{2}) BG(z_{2}^{*}0 | t_{2}0) | \psi_{0} \rangle] \\ = \int_{t_{2}}^{t_{1}} d\tau \alpha(t_{1} - \tau) \langle \psi_{0} | e^{iH_{S}t_{1}} L^{\dagger}AV_{\tau-t_{1}} L e^{-iH_{S}(t_{1}-t_{2})} Be^{-iH_{S}t_{2}} | \psi_{0} \rangle \\ + \int_{0}^{t_{2}} d\tau \alpha(t_{1} - \tau) \langle \psi_{0} e^{iH_{S}t_{1}} | L^{\dagger}Ae^{-iH_{S}(t_{1}-t_{2})} \\ \times BV_{\tau-t_{1}} L e^{-iH_{S}t_{2}} | \psi_{0} \rangle + \mathcal{O}(g^{3}).$$
(23)

In the last expression we introduce the notation $\mathcal{M}_i[\mathcal{F}] = \int d\mu(z_i)\mathcal{F}$ to denote the multidimensional integrals over the Gaussian variables z_i , where \mathcal{F} is any function of them. Since the quantities $e^{iH_St_1}$, $e^{-iH_S(t_1-t_2)}$, and $e^{-iH_St_2}$ represent the Gaussian averages over z_1 and z_2 of the first-order term of the perturbative expansion of $G^{\dagger}(0z_1|0t_1)$, $G(z_1^*z_2|t_1t_2)$, and $G(z_2^*0|t_20)$, respectively, we conclude that the last terms can be written as

$$\mathcal{M}_{1,2}[z_{1,t_{1}}\langle\psi_{0}|G^{\dagger}(0z_{1}|0t_{1})L^{\dagger}AG(z_{1}^{*}z_{2}|t_{1}t_{2})BG(z_{2}^{*}0|t_{2}0)|\psi_{0}\rangle]$$

$$=\int_{t_{2}}^{t_{1}}d\tau\alpha(t_{1}-\tau)\langle\Psi_{0}|\{L^{\dagger}AV_{\tau-t_{1}}L\}(t_{1})B(t_{2})|\Psi_{0}\rangle$$

$$+\int_{0}^{t_{2}}d\tau\alpha(t_{1}-\tau)\langle\Psi_{0}\{L^{\dagger}A\}(t_{1})$$

$$\times\{BV_{\tau-t_{1}}L\}(t_{2})|\Psi_{0}\rangle+\mathcal{O}(g^{3}).$$
(24)

The brackets involving a group of operators indicate that the evolution affects all of them, $\{ABC\}(t_1) = U_I^{-1}(t_1, 0)ABCU_I(t_1, 0) = A(t_1)B(t_1)C(t_1)$. A similar procedure can be used to perform the averages of the terms with z_{1,t_1}^* and z_{2,t_1} in Eq. (18), which are such that

$$\mathcal{M}_{1,2}[z_{1,t_1}^*\langle\psi_0|G^{\dagger}(0z_1|0t_1)ALG(z_1^*z_2|t_1t_2)BG(z_2^*0|t_20)|\psi_0\rangle] = \int_0^{t_1} d\tau \alpha^*(t_1-\tau)\langle\Psi_0|\{V_{\tau-t_1}L^{\dagger}AL\}(t_1)B(t_2)|\Psi_0\rangle, \quad (25)$$

and

$$\mathcal{M}_{1,2}[z_{1,t_1}^*\langle\psi_0|G^{\dagger}(0z_1|0t_1)L^{\dagger}AG(z_1^*z_2|t_1t_2)BG(z_2^{*}0|t_20)|\psi_0\rangle] = \int_0^{t_2} d\tau \alpha(t_1-\tau)\langle\Psi_0|\{AL^{\dagger}\}(t_1)\{BV_{\tau-t_2}\}(t_2)|\Psi_0\rangle.$$
(26)

Inserting Eqs. (24)–(26) in Eq. (18), we get the following equation for two-time correlations:

$$\frac{d}{dt_{1}} \langle \Psi_{0} | A(t_{1}) B(t_{2}) | \Psi_{0} \rangle
= i \langle \Psi_{0} | \{ [H_{S}, A] \}(t_{1}) B(t_{2}) | \Psi_{0} \rangle + \int_{0}^{t_{1}} d\tau \alpha^{*}(t_{1} - \tau)
\times \langle \Psi_{0} | \{ V_{\tau - t_{1}} L^{\dagger} [A, L] \}(t_{1}) B(t_{2}) | \Psi_{0} \rangle + \int_{t_{2}}^{t_{1}} d\tau \alpha(t_{1} - \tau)
\times \langle \Psi_{0} | \{ [L^{\dagger}, A] V_{\tau - t_{1}} L \}(t_{1}) B(t_{2}) | \Psi_{0} \rangle + \int_{0}^{t_{2}} d\tau \alpha(t_{1} - \tau)
\times \langle \Psi_{0} | \{ [L^{\dagger}, A] \}(t_{1}) \{ B V_{\tau - t_{2}} L \}(t_{2}) | \Psi_{0} \rangle + \mathcal{O}(g^{3}). \quad (27)$$

Equation (27) represents the set of κ^{κ} evolution equations for the correlation of system observables defined with a basis of κ operators. For a two-level system $\kappa=3$ and the dimension of the operators in their matricial representation is 2.

In order to make use of Eq. (27) to calculate two-time correlations, it is necessary to evolve initially the single mean value evolution equations up to time t_2 . This is necessary since the initial value for the two-time correlations is given by $\langle \Psi_0 | A(t_2)B(t_2) | \Psi_0 \rangle = \langle \Psi_0 | C(t_2) | \Psi_0 \rangle$, where C=AB. We now show that non-Markovian multiple-time correlation functions do not obey the quantum regression theorem. This theorem, valid for Markovian interactions, states that the coefficients of the evolution of *N*-time correlation functions are the same as those for the single-time evolution. In the non-Markovian case, this evolution is given by

$$\frac{d}{dt_{1}} \langle \Psi_{0} | A(t_{1}) | \Psi_{0} \rangle
= i \langle \Psi_{0} | \{ [H_{S}, A] \}(t_{1}) | \Psi_{0} \rangle
+ \int_{0}^{t_{1}} d\tau \alpha^{*}(t_{1} - \tau) \langle \Psi_{0} | \{ V_{\tau - t_{1}} L^{\dagger} [A, L] \}(t_{1}) | \Psi_{0} \rangle
+ \int_{0}^{t_{1}} d\tau \alpha(t_{1} - \tau) \langle \Psi_{0} | \{ [L^{\dagger}, A] V_{\tau - t_{1}} L \} | \Psi_{0} \rangle + \mathcal{O}(g^{3}).$$
(28)

Let us take the two-time correlation function equation (27), and rearrange its last term as

$$\int_{0}^{t_{2}} d\tau \alpha(t_{1} - \tau) \langle \Psi_{0} | \{ [L^{\dagger}, A] \}(t_{1}) \{ BV_{\tau - t_{2}} L \}(t_{2}) | \Psi_{0} \rangle$$

$$= \int_{0}^{t_{2}} d\tau \alpha(t_{1} - \tau) \langle \psi_{0} | \{ [L^{\dagger}, A] \}(t_{1}) \{ [B, V_{\tau - t_{2}} L] \}(t_{2}) | \Psi_{0} \rangle$$

$$+ \int_{0}^{t_{2}} d\tau \alpha(t_{1} - \tau) \langle \psi_{0} | \{ [L^{\dagger}, A] \}(t_{1}) \{ V_{\tau - t_{2}} LB \}(t_{2}) | \Psi_{0} \rangle.$$
(29)

In the last term of Eq. (29), it should be taken into account that, up to second order in g,

$$\int_{0}^{t_{2}} d\tau \alpha(t_{1} - \tau) \langle \psi_{0} | \{ [L^{\dagger}, A] \}(t_{1}) \{ V_{\tau - t_{2}} LB \}(t_{2}) | \Psi_{0} \rangle$$

$$= \int_{0}^{t_{2}} d\tau \alpha(t_{1} - \tau) \alpha(t_{1} - \tau) \langle \Psi_{0} | \{ [L^{\dagger}, A] V_{\tau - t_{1}} L \}(t_{1}) B(t_{2}) | \Psi_{0} \rangle.$$
(30)

Inserting Eq. (29) in Eq. (27), the second-order evolution equation of the two-time correlation function can also be expressed as

$$\frac{d}{dt_{1}} \langle \Psi_{0} | A(t_{1}) B(t_{2}) | \Psi_{0} \rangle
= i \langle \Psi_{0} | \{ [H_{S}, A] \}(t_{1}) B(t_{2}) | \Psi_{0} \rangle + \int_{0}^{t_{1}} d\tau \alpha^{*}(t_{1} - \tau)
\times \langle \Psi_{0} | \{ V_{\tau-t_{1}} L^{\dagger}[A, L] \}(t_{1}) B(t_{2}) | \Psi_{0} \rangle + \int_{0}^{t_{1}} d\tau \alpha(t_{1} - \tau)
\times \langle \Psi_{0} | \{ [L^{\dagger}, A] V_{\tau-t_{1}} L \}(t_{1}) B(t_{2}) | \Psi_{0} \rangle + \int_{0}^{t_{2}} d\tau \alpha(t_{1} - \tau)
\times \langle \Psi_{0} | \{ [L^{\dagger}, A] \}(t_{1}) \{ [B, V_{\tau-t_{2}} L] \}(t_{2}) | \Psi_{0} \rangle + \mathcal{O}(g^{3}). \quad (31)$$

The first three terms of the last equation are analogous to the non-Markovian evolution of the quantum average of A, Eq. (28). The last term containing $[L^{\dagger}, A]=0$ and $[B, V_{\tau-t}L]=0$ is responsible of the break of the validity of the quantum regression theorem. As expected, this term is zero in the Markovian case, since the corresponding correlation function $\alpha(t_1-\tau)=\Gamma \delta(t_1-\tau)$ vanishes in the domain of integration from 0 to t_2 . Notice that in both Eqs. (31) and (28), the time dependencies of the operators on the system's Hamiltonian, given by V_{t_1} , are especially simple in the present case, since the operators are expressed in the atomic basis in which H_S is diagonal. In summary, the previous equations lead to the computation of the MTCF and they contain the conditions under which the QRT remains valid in the weak-coupling limit.

Computing the MTCF: System of equations for two-time correlations in the weak-coupling limit

In order to make use of Eq. (31) to calculate two-time correlations, it is necessary to evolve initially the quantum

mean value evolution equations (28) up to time t_2 . This is necessary since the initial value for the two-time correlations is given by $\langle \Psi_0 | A(t_2)B(t_2) | \Psi_0 \rangle = \langle \Psi_0 | C(t_2) | \Psi_0 \rangle$, where C=AB.

It is also important to notice that some correlations that formally obey the QRT might give rise to solutions of Eq. (31) that differ from those given by the QRT. This happens in particular for those correlations that depend on others that do not obey the QRT. A good example is in the calculus of the emission spectra of quantum open systems, which is one of the most common applications of two-time correlation functions. Since it is considered as the Fourier transform of the correlation of its coupling operators L and L^{\dagger} , in this case B=L and the quantum regression theorem formally applies. However, care should be taken, since the evolution given by the QRT is not always correct: for instance, for $L=\sigma_x$ the correlation $C_{\sigma_x \sigma_x}$ formally obeys the QRT, but its evolution differs from that given by the QRT since it depends on the correlation $C_{\sigma_x \sigma_x}$ that does not obey the QRT.

The choice of the stochastic method or the system of equations for computing the MTCF has to be made according to the particular problem. For a system with a large number of degrees of freedom \mathcal{F} , it is generally more convenient to use the stochastic method, since in the system of equations the dimension of the matrices grows with $\mathcal{F}^2/2$.

The same happens when *N*-time correlation functions have to be computed for κ large, since in that case the stochastic method permits us to compute only the particular correlation function that is needed, and not the whole set of κ^{κ} correlations that appears in Eqs. (31).

For the particular case of quantum mean values of system operators, an accurate comparison of the performance of both methods (stochastic and master equation) can be found in Ref. [42], where is studied the time needed to numerically compute the quantum mean value of a certain operator both with the master equation (T_{ME}) and with the stochastic sampling (T_{SSE}) , the last one within a certain standard error. The relation between both times is such that $T_{ME}/T_{SSE} \propto \mathcal{F}^{1+x}$, where x is a parameter that depends on the operator, but generally is equal to 0 or 1. Evidently, the former is still a rough relation. In a more precise calculus, the time T_{SSE} is found, also in Ref. [42], to be dependent on the number of trajectories κ) that are required to reproduce the result with a certain accuracy.

IV. SOME EXAMPLES

A. A solvable model

To illustrate the theory proposed in this paper, we apply it to a simple solvable model. Consider the Hamiltonian (1) with $H_S = (\omega_S/2)\sigma_z$ and $L = \sigma_z$. This model describes the dynamics of system state vectors towards one of the eigenstates of the system Hamiltonian. It turns out that because $[H_S, L]$ =0, then O=L in Eq. (14). The reduced propagator is diagonal and it is explicitly given by

$$G(z_{1}^{*}z_{2}|t_{1}t_{2}) = \exp\left(-i\frac{\omega_{s}}{2}(t_{1}-t_{2})\sigma_{z}+L\int_{t_{2}}^{t_{1}}d\tau z_{1,\tau}^{*}-L^{\dagger}\int_{t_{2}}^{t_{1}}d\tau z_{2,\tau} - LL^{\dagger}\int_{t_{2}}^{t_{1}}d\tau\int_{t_{2}}^{\tau}ds\alpha(\tau-s)+z_{1}^{*}z_{2}\right).$$
(32)

We first look at the expectation values of system observables, since they will be useful in our discussion [23]. Let us consider an observable A that has, in the basis of eigenvectors of σ_z , the matrix representation

$$A = \begin{pmatrix} 0 & \alpha \\ \beta & 0 \end{pmatrix}.$$

For an initial state $|\Psi_0\rangle = |\psi_0\rangle|$ vacuum \rangle , the expectation value of A is formally given by

$$\langle A \rangle = M[\langle \psi_0 | G^{\dagger}(0z|0t) A G(z^*0|t0) | \psi_0 \rangle].$$
(33)

From Eqs. (32) and (33), performing the average over the environment degrees of freedom it follows that

$$\begin{split} \langle A \rangle &= e^{-2\int_0^t d\tau \int_0^\tau ds (\alpha(\tau-s)+\alpha^*(\tau-s)))} \\ &\times \{ \alpha \langle \psi_{01} | \psi_{01} \rangle e^{i\omega_S t} + \langle \psi_{02} | \psi_{02} \rangle e^{-i\omega_S t} \}, \end{split}$$

where we have taken a normalized initial system state $|\psi_0\rangle = |\psi_{01}\rangle + |\psi_{02}\rangle$. In the same manner, it follows for $\langle \sigma_z \rangle$

$$\langle \sigma_z \rangle = \langle \psi_{01} | \psi_{01} \rangle - \langle \psi_{02} | \psi_{02} \rangle.$$

Now we turn our attention to the multiple-time correlation functions. In particular, we compute *two-time* correlations. Higher-order time correlations can be treated in the same vein and they do not require any new consideration. Let us consider the observables

$$A = \begin{pmatrix} 0 & \alpha \\ \beta & 0 \end{pmatrix}; \quad B = \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

Their two-time correlation function (5) is

$$C_{AB}(t_1t_2|\Psi_0) = \mathcal{M}_{1,2}[\langle \psi_0 | G^{\dagger}(0z_1|0t_1)AG(z_1^*z_2|t_1t_2)BG(z_2^*0|t_20)|\psi_0\rangle].$$
(34)

If we insert Eq. (32) into Eq. (34) the following expression is obtained:

$$C_{AB}(t_{1}t_{2}|\Psi_{0}) = e^{-2\int_{0}^{t_{1}} d\tau \int_{0}^{t_{1}} ds \,\alpha(\tau-s)} \{-\alpha \langle \psi_{01} | \psi_{02} \rangle e^{i\omega_{S}t_{1}} + \beta \langle \psi_{02} | \psi_{01} \rangle e^{-i\omega_{S}t_{1}} \}.$$
(35)

As in the previous cases, once the environment correlation function is known, the time correlation function can be directly computed. In the case in which $A=B=\sigma_z$, we have $C_{\sigma_z\sigma_z}=0$.

We now calculate the evolution equations for the twotime correlation functions and investigate their relation to the QRT. To this purpose, we first consider the time derivative of the mean averages $\langle \sigma_i \rangle$, i=(x,y,z) from the general form (33), which results in the following set of coupled linear differential equations:

$$\frac{\partial \langle \sigma_x \rangle}{\partial t} = -D(t) \langle \sigma_x \rangle - \omega_S \langle \sigma_y \rangle,$$
$$\frac{\partial \langle \sigma_y \rangle}{\partial t} = -D(t) \langle \sigma_y \rangle + \omega_S \langle \sigma_x \rangle,$$
$$\frac{\partial \langle \sigma_z \rangle}{\partial t} = 0,$$

with

$$D(t) = 2 \int_0^t d\tau [\alpha(t-\tau) + \alpha^*(t-\tau)].$$
 (36)

In the same manner, the following set of equations for the two-time correlation functions is obtained by deriving Eq. (35):

$$\frac{C_{\sigma_x \sigma_z}}{\partial t_1} = -D(t_1)C_{\sigma_x \sigma_z} - \omega_S C_{\sigma_y \sigma_z},$$
$$\frac{C_{\sigma_y \sigma_z}}{\partial t_1} = -D(t_1)C_{\sigma_y \sigma_z} + \omega_S C_{\sigma_y \sigma_z},$$
$$\frac{C_{\sigma_z \sigma_z}}{\partial t_1} = 0.$$

Comparing Eqs. (36) and (37), we could get the tentative conclusion that the QRT is valid for this model. However, as discussed in the previous sections, the validity of the QRT follows from the fact that in this case we have $[V_{\tau-t_2}L,B] = 0$, and therefore the last term of Eq. (31) is zero. To illustrate that the validity of the QRT cannot be considered as general, we study the time correlation $C_{\sigma_x \sigma_y}(t_1 t_2)$ for which neither [L,B]=0 nor $[L^{\dagger},A]=0$. We start by considering the general antidiagonal system operators $A = \{\{0, \alpha\}, \{\beta, 0\}\}$ and $B = \{\{0, \alpha'\}, \{\beta', 0\}\}$.

The analytical derivation of their two-time correlation function leads to

$$C_{AB}(t_1t_2) = e^{D(t_1t_2)} \{ \alpha \beta' \langle \psi_{01} | \psi_{01} \rangle e^{i\omega_S(t_1-t_2)}$$

+ $\alpha' \beta \langle \psi_{02} | \psi_{02} \rangle e^{-i\omega_S(t_1-t_2)} \},$ (37)

where we have defined

$$\begin{split} \widetilde{D}(t_1 t_2) &= \int_0^{t_1} d\tau \int_0^{\tau} ds \, \alpha^*(\tau - s) + \int_{t_2}^{t_1} d\tau \int_{t_2}^{\tau} ds \, \alpha(\tau - s) \\ &+ \int_0^{t_2} d\tau \int_0^{\tau} ds \, \alpha(\tau - s) + \int_0^{t_1} d\tau \int_{t_2}^{t_1} ds \, \alpha(\tau - s) \\ &- \int_{t_2}^{t_1} d\tau \int_0^{t_2} ds \, \alpha(\tau - s) - \int_0^{t_1} d\tau \int_0^{t_2} ds \, \alpha(\tau - s). \end{split}$$

For $C_{\sigma_x \sigma_y}(t_1 t_2)$ we have



FIG. 1. (Color online) In the figure different two-time correlation functions are displayed. The upper figure corresponds to the real part of $C_{\sigma_x \sigma_z}$ and the lower to the real part of $C_{\sigma_x \sigma_y}$. In the upper figure, the solid line corresponds to the analytical result (35) that in *this case* coincides with the result expected from the quantum regression theorem. The dotted line is the result of an average over 10^2 trajectories and the dot-dashed line to an average over 10^5 trajectories. In the lower figure, the long-dashed line is the result given by the QRT, which is compared to the exact result given by Eq. (37) (solid line). Clearly the QRT is not valid for $C_{\sigma_x \sigma_y}$. The dotted and dot-dashed lines are the result of an average over 10^2 and 10^4 trajectories, respectively.

$$C_{\sigma_x \sigma_y}(t_1 t_2) = e^{-D(t_1 t_2)} \{ i \langle \psi_{01} | \psi_{01} \rangle e^{i \omega_S(t_1 - t_2)} - i \langle \psi_{02} | \psi_{02} \rangle e^{-i \omega_S(t_1 - t_2)} \},$$
(38)

with its derivative with respect to t_1 being

$$\frac{\partial C_{\sigma_x \sigma_y}(t_1 t_2)}{\partial t_1} = \frac{\partial \tilde{D}(t_1 t_2)}{\partial t_1} C_{\sigma_x \sigma_y}(t_1 t_2) - \omega_S C_{\sigma_x \sigma_x}(t_1 t_2).$$
(39)

We can see from this last equation that the time derivative of the correlation function $C_{\sigma_x \sigma_y}$ does not satisfy the quantum regression theorem, a result that illustrates the conclusion drawn in the previous section and the fact that the theory here introduced applies for both Markovian and non-Markovian cases. Equation (39) has been computed by deriving the exact solution (38), but it can also be obtained from the general equation (31). Although this last equation is derived under the weak coupling assumption, it is also valid for the present model, since the expression O=L is also obtained under a perturbative expansion of the operator.

Figures 1 and 2 show respectively the real and imaginary parts of two-time correlation functions of the system (1), $C_{\sigma_x \sigma_z}$ and $C_{\sigma_x \sigma_y}$. The exact result (38) is compared to the one obtained through the stochastic propagators as explained in Sec. II. When the number of trajectories in the stochastic ensemble is large enough, both results are the same, which proves the validity of the stochastic method. The number of oscillators of the environment has been set equal to 2. The parameters used were $g_1, g_2 = g = 1$ and $\omega_1 = 6$, $\omega_2 = 2$. The



FIG. 2. (Color online) The figure displays the same as Fig. 1, but now for the imaginary parts of $C_{\sigma_x \sigma_z}$ and $C_{\sigma_x \sigma_y}$. In the upper figure, again the analytical result (35) coincides with the result expected from the quantum regression theorem (both in the solid line). Dotted and dot-dashed lines correspond to the result of an average over 10^2 and 10^5 trajectories, respectively. In the lower figure, the analytical result (solid line) differs from that given by the QRT (long-dashed). The dotted and the dot-dashed lines are the result of an average over 10^2 and 10^4 trajectories, respectively.

initial system state taken was $|\Psi_0\rangle = |\psi_0\rangle|00\rangle$ with $|\psi_{01}\rangle = (1 + 2i)/\sqrt{7}|+\rangle$ and $|\psi_{02}\rangle = (1+i)/\sqrt{7}|-\rangle$. It is clear also from the figures that the QRT does not apply for $C_{\sigma_x \sigma_y}$, since $[V_{\tau-t}L, B] \neq 0$ and $[L^{\dagger}, A] \neq 0$.

B. A dissipative system

We now compute two-time correlations for a two level system with $H_S = (\omega_S/2)\sigma_z$ and a dissipative interaction with $L = \sigma_{12}$. Within the perturbative approximation, the operator $O(t, \tau)$ can be replaced by its zero-order perturbative expansion, $V_{\tau-t}L = \sigma_{12} \exp\{i\omega_S(t-\tau)\}$, where ω_S is the system rotating frequency. We propose the following correlation function:

$$\alpha^{appx}(t-\tau) = \sum_{m=-\nu/2}^{\nu/2} C(m) e^{-i\pi m(t-\tau)/T_{max}},$$
(40)

with the coefficients

$$C(m) = \frac{1}{2T_{max}} \int_{-T_{max}}^{T_{max}} dt \alpha(t) e^{i\pi m t/T_{max}},$$
(41)

which represents the Fourier series for the function $\alpha(t) = (\Gamma/2)\exp\{-\Gamma|t|\}$. In these equations, T_{max} is the time window in which the correlation function is expanded in the series. The superindex *appx* in Eq. (40) stands for the fact that the correlation function is an approximation of $\alpha(t)$, as long as the number of oscillators entering in the sum is finite and small. The more members we add in the sum, the closer is the solution to the exponential decaying correlation function, and the larger we can fix the recurrence time T_{max} . However, it is observed that for only $\nu=8$ oscillators, we can already choose parameters (g, Γ) and the environmental correlation time τ_c) such that the second order two-time correlations present a decaying behavior before the recurrence time.



FIG. 3. Two-time correlation $C_{xx} = C_{\sigma_x \sigma_x}$ for the coupling $L = \sigma_{12}$, and the dissipative Fourier series of the exponential correlation function with $\nu = 8$ oscillators. The initial condition is the same as in the former figure, $|\psi_0\rangle = [(1+2i)|+\rangle + (1+i)|-\rangle]/\sqrt{7}$, and the parameters are: $\omega_S = 0.1$, $\Gamma = 1$, perturbative parameter g = 0.2, recurrence time $T_{max} = 40$, and initial time for the correlation $t_2 = 1$. The thick solid line represents the solution of the system (31), whereas long-dashed, short-dashed, and solid lines give the result of the stochastic method for 10^4 , 10^6 , and 10^7 trajectories, respectively. The solid line almost overlaps the solution of the system of equations. An increasing agreement with the system curve is observed as the number of trajectories grows.

We choose $t_2=1$, g=0.5, and $\Gamma=1$, so that the decaying behavior can already be observed in the range $t_1-t_2=\tau=50$ as displayed in Fig. 3, which represents the correlation $C_{\sigma_x\sigma_x}$. In this figure, we compare the result obtained from the evolution equation (31) with the ensemble averaged stochastic evolution, given by Eq. (5) for different number of trajectories. The last one is obtained by following the steps described in Sec. II.

Let us now consider a very large number of oscillators in Eq. (40), i.e., the correlation function $\alpha(t-\tau)=(\Gamma/2)\exp\{-\Gamma|t-\tau|\}$, in order to study the validity of the QRT for the $C_{\sigma_x\sigma_x}$. This is a typical case in which the last term of Eq. (31) does not vanish, so that the result of the QRT differs to the one of Eq. (31), as displayed in Fig. 4.

V. DISSIPATIVE MASTER EQUATION WITH GENERAL INITIAL CONDITIONS IN THE WEAK-COUPLING LIMIT

As mentioned in the Introduction, the propagator with evolution given by Eq. (14) permits us to calculate master equations with general initial conditions. Suppose, for instance, that we have a *pure* initial state for the total density matrix,

$$\rho_{tot}(0) = |\Psi_0\rangle \langle \Psi_0|$$

= $\int d\mu(z_0) \int d\mu(z'_0) |z_0\rangle |\psi_0(z^*_0)\rangle \langle \psi_0(z'_0)|\langle z'_0|.$ (42)

This state should be normalized as $\rho_{tot}(0) = |\Psi_0\rangle\langle\Psi_0|/\langle\Psi_0|\Psi_0\rangle$. For simplicity, we will omit this normalization factor in the calculus that follows, although it will be taken into account in the initial condition of the examples



FIG. 4. Two-time correlation $C_{xx}=C_{\sigma_x\sigma_x}$ for the coupling $L=\sigma_{12}$ and the dissipative correlation function $\alpha(t-\tau) = -(\Gamma/2)\exp\{-\Gamma|t-\tau|\}$. The initial condition is the same as in the former figure and the parameters are $\omega_S=0.1$, $\Gamma=1$, perturbative parameter g=0.4, and initial time for the correlation $t_2=10$. The solid line represents the solution of the system (31) and the long-dashed line gives the result expected with the QRT. Since the last term in Eq. (31) is nonzero, both results are different from each other and the QRT is not valid.

we show in the next sections. At time t the total density matrix can be expressed as

$$\rho_{tot}(t) = \int d\mu(z_0) \\ \times \int d\mu(z'_0) \mathcal{U}_I(t,0) |z_0\rangle |\psi_0(z^*_0)\rangle \langle\psi_0(z'_0)|\langle z'_0|\mathcal{U}_I^{-1}(t,0).$$
(43)

The reduced density matrix of the system, which is defined as $\rho_s = \text{Tr}_B \{\rho_{tot}\}$ is equal to

$$\rho_s(t) = \int d\mu(z_0) \int d\mu(z'_0) \rho_s(z'_0 z'_0 z'_0 z_0 | t), \qquad (44)$$

where we have made the following definition:

$$\rho_{s}(z_{0}'^{*}z_{0}'z_{0}'z_{0}|t) = \int d\mu(z_{1}) \ G(t0|z_{1}^{*}z_{0})\rangle|\psi_{0}(z_{0}^{*})\rangle$$
$$\times \langle\psi_{0}(z_{0}')|G^{\dagger}(t0|z_{0}'^{*}z_{1}).$$
(45)

Once $\rho_s(z_0'^* z_0' z_0' z_0 | t)$ is known, the sum (44) can be performed to obtain the reduced density operator. This may be done for each particular initial condition, which is given by the set of values $|\psi_0(z_0^*)\rangle$ for each z_0 . Hence it is interesting to compute the evolution equation of $\rho_s(z_0'^* z_0' z_0^* z_0 | t)$, since it represents the most general object needed to compute ρ_s . Such evolution,

$$\frac{d\rho_s(z_0'^* z_0' z_0^* z_0|t)}{dt} = \frac{d}{dt} \mathcal{M}_1[G(z_1^* z_0|t0)|\psi(z_0)\rangle \\ \times \langle \psi(z_0')|G^{\dagger}(z_0'^* z_1|t0)],$$
(46)

can be obtained within the perturbative approximation by using Eq. (17) for the double propagator. In that way equation Eq. (46) becomes

$$\frac{d\rho_{s}(z_{0}^{t*}z_{0}^{\prime}z_{0}z_{0}z_{0})|t)}{dt} = -i[H_{s}, \rho_{s}(z_{0}^{t*}z_{0}^{\prime}z_{0}z_{0}|t)] - z_{0,t}L^{\dagger}\rho_{s}(z_{0}^{t*}z_{0}^{\prime}z_{0}z_{0}|t) - z_{0,t}^{*}\rho_{s}(z_{0}^{t*}z_{0}^{\prime}z_{0}z_{0}|t)L + \int_{0}^{t} d\tau\alpha(t-\tau)L^{\dagger}V_{\tau-t}L\rho_{s}(z_{0}^{t*}z_{0}^{\prime}z_{0}z_{0}|t) - \int_{0}^{t} d\tau\alpha^{*}(t-\tau)\rho_{s}(z_{0}^{t*}z_{0}^{\prime}z_{0}z_{0}|t)V_{t-\tau}L^{\dagger}L + \mathcal{M}_{1}[z_{1,t}^{*}LG(z^{*}z_{0}|t0)|\psi_{0}(z_{0}^{*})\rangle\langle\psi_{0}(z_{0}^{\prime})|G^{\dagger}(z_{0}^{t*}z_{1}|t0)] + \mathcal{M}_{1}[z_{1,t}G(z_{1}^{*}z_{0}|t0)|\psi_{0}(z_{0}^{*})\rangle\langle\psi_{0}(z_{0}^{\prime})|G^{\dagger}(z_{0}^{t*}z_{1}|t0)L^{\dagger}] + \mathcal{O}(g^{3}),$$

$$(47)$$

where we stress again that the initial condition should be normalized as

$$\rho_s(z_0'^* z_0' z_0^* z_0 | 0) = |\psi_0(z_0^*)\rangle \langle \psi_0(z_0') | / \langle \psi_0(z_0') | \psi_0(z_0^*) \rangle.$$

In order to obtain the master equation up to second order in the perturbative parameter, we can use the perturbative expansion (20) considering now that $t_2=0$, $t_1=t$, and the proper labels for the noises. Following the same procedure as in Sec. III, the average of the noise term z_t is the following:

$$\mathcal{M}_{1}[z_{1,t}G(z_{1}^{*}z_{0}|t0)|\psi_{0}(z_{0}^{*})\rangle\langle\psi_{0}(z_{0}')|G^{\dagger}(z_{0}'^{*}z_{1}|t0)L^{\dagger}]$$

$$= z_{0,t}\rho_{s}(z_{0}'^{*}z_{0}'z_{0}'z_{0}|t)L^{\dagger}$$

$$+ \int_{0}^{t}d\tau\alpha(t-\tau)V_{\tau-t}L\rho_{s}(z_{0}'^{*}z_{0}'z_{0}z_{0}|t)L^{\dagger} + \mathcal{O}(g^{3}).$$
(48)

In the last expression, we have replaced the first order in a perturbative expansion of the density operator,

$$\rho_{s}^{(1)}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}^{*}z_{0}|t) = \rho_{s}^{(0)}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}^{*}z_{0}|t) + \int_{0}^{t} d\tau z_{0,\tau}^{\prime*} [V_{\tau-t}L, \rho_{s}^{(0)}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}^{*}z_{0}|t)] + \int_{0}^{t} d\tau z_{0,\tau} [\rho_{s}^{(0)}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}^{*}z_{0}|t), V_{\tau-t}L^{\dagger}],$$

$$(49)$$

by $\rho_s(z_0'^* z_0' z_0' z_0 | t)$, since it appears in terms which are at least of first order in g. Note that the last expression is in terms of the zero-order density operator,

$$\rho_s^{(0)}(z_0'^* z_0' z_0 z_0 z_0 | t) = e^{-iH_S t} |\psi_0(z_0)\rangle \langle \psi_0(z_0') | e^{iH_S t} e^{z_0'^* z_0}.$$
 (50)

In the same way we have

$$\mathcal{M}_{1}[z_{1,t}^{*}LG(z^{*}z_{0}|t0)|\psi_{0}(z_{0}^{*})\rangle\langle\psi_{0}(z_{0}^{\prime})|G^{\dagger}(z_{0}^{\prime*}z_{1}|t0)]$$

$$= z_{0,t}^{\prime*}L\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}^{*}z_{0}|t) + L\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime*}z_{0}^{*}z_{0}|t)$$

$$\times \int_{0}^{t} d\tau \alpha^{*}(t-\tau)V_{\tau-t}L^{\dagger} + \mathcal{O}(g^{3}).$$
(51)

Inserting Eqs. (48) and (51) in Eq. (47), we have the following second-order equation for $\rho_s(z_0^{*}z_0^{*}z_0^{*}z_0|t)$:

$$\frac{d\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}^{\prime}z_{0}|t)}{dt} = -i[H_{s},\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}|t)] + z_{0,t}[\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}|t),L^{\dagger}] \\
+ z_{0,t}^{\prime*}[L,\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}|t)] + \int_{0}^{t}d\tau\alpha(t-\tau) \\
\times [V_{\tau-t}L\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}|t),L^{\dagger}] + \int_{0}^{t}d\tau\alpha^{*}(t-\tau) \\
\times [L,\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}^{\ast}z_{0}|t)V_{\tau-t}L^{\dagger}] + \mathcal{O}(g^{3}).$$
(52)

Suppose now that we have an initial *mixed state* for the total system,

$$\rho_{tot}(0) = \int d\mu(z_0) \mathcal{J}(z_0, z_0^*) |\Psi_0(z_0^*)\rangle \langle \Psi_0(z_0)|, \qquad (53)$$

where $\mathcal{J}(z_0, z_0^*)$ is the statistical probability for the member $|\Psi_0(z_0^*)\rangle$ of the statistical ensemble. Then, the reduced density matrix of the system, $\rho_s = \text{Tr}_B\{\rho_{tot}\}$ is equal to

$$\rho_s(t) = \int d\mu(z_0) \mathcal{J}(z_0 z_0^*) \rho_s(z_0^* z_0 | t), \qquad (54)$$

with the following definition:

$$\rho_{s}(z_{0}^{*}z_{0}|t) = \int d\mu(z_{1}) \ G(t0|z_{1}^{*}z_{0}) \rangle |\psi_{0}(z_{0}^{*})\rangle \langle\psi_{0}(z_{0})|G^{\dagger}(t0|z_{0}^{*}z_{1}),$$
(55)

which is equal to Eq. (45), but with $z'_0 = z_0$. Once such replacement is made, Eq. (52) represents the evolution of $\rho_s(z_0^*z_0|t)$. As in the pure case, provided that the whole set of initial conditions $|\psi_0(z_0^*)\rangle$ is known for the problem, as well as the probability distribution $\mathcal{J}(z_0, z_0^*)$ which describes its frequency in the mixture, then the sum (54) can be performed with the solutions $\rho_s(z_0^*z_0|t)$.

It is clear that Eq. (52) is the essential piece for calculating the evolution equation of single mean values for general initial conditions, both for an initial pure state and a statistical mixture. In the next sections, we are going to compute the evolution of $\rho_s(z_0'^*z_0'z_0^*z_0|t)$ with two examples, the solvable model with $L \propto H_s$, and the spin boson model. The evolution of $\rho_s(z_0^*z_0|t)$ is just a particular case of the former one. The idea is to verify Eq. (52) by comparing its results with those obtained by performing a numerical sampling over stochastic trajectories of the reduced propagator, following the relation (46).

A. A solvable model

We illustrate Eq. (52) by applying it to the solvable model presented in Sec. IV A. This equation is the essential piece for calculating the evolution equation of single mean values for general initial conditions. First of all, we calculate the mean value of an observable A that has, in the basis of eigenvectors of σ_z , the matrix representation

$$A = \begin{pmatrix} 0 & \alpha \\ \beta & 0 \end{pmatrix}.$$

Taking as the initial values for the states of the total system $|\Psi_0(z_0^*)\rangle = |\psi_0(z_0^*)\rangle|z_0\rangle$ and $|\Psi_0(z_0')\rangle = |\psi_0(z_0'^*)\rangle|z_0'\rangle$, the average of A over these two vectors is given by

$$\langle \Psi_0(z'_0) | A | \Psi_0(z^*_0) \rangle = \mathcal{M}_1[\langle \psi_0(z'_0) | G^{\dagger}(z'_0 z_1 | 0t) A G(z^*_1 z_0 | t0) \\ \times | \psi_0(z^*_0) \rangle].$$
(56)

From Eq. (32), performing the average over the environment degrees of freedom z_1 we obtain

$$\langle A \rangle = e^{-2\int_0^t d\tau \int_0^\tau ds (\alpha(\tau-s) + \alpha^*(\tau-s))} \Big\{ \alpha \langle \psi_{0,1} | \psi_{0,2} \rangle e^{i\omega t + 2\int_0^t d\tau (z_{0,\tau} - z_{0,\tau}'^*)} \\ + \beta \langle \psi_{02} | \psi_{01} \rangle e^{-i\omega t - 2\int_0^t d\tau (z_{0,\tau} - z_{0,\tau}'^*)} \Big\},$$
(57)

where we have taken a normalized initial system state $|\psi_0(z_0^{\prime*})\rangle = |\psi_0\rangle = |\psi_{01}\rangle + |\psi_{02}\rangle$. In the same manner it follows for $\langle \sigma_z \rangle$ that

$$\langle \sigma_z \rangle = \langle \psi_{01} | \psi_{01} \rangle - \langle \psi_{02} | \psi_{02} \rangle.$$
(58)

Figure 5 represents Eq. (57) for $A = \sigma_i$, i=x, y, and Eq. (58) compared to the result obtained using the double propagator (17). When the number of stochastic trajectories included in the ensemble mean (46) is large enough, both results coincide. The correlation function is obtained for two oscillators with coupling parameters $g_1=g_2=g=0.1$, frequencies $\omega_1=6$ and $\omega_2=2$, and for an atom with rotating frequency $\omega_s=4$.

B. A dissipative example: Spin-boson model

We now calculate the expectation value $\langle \Psi_0 | A | \Psi_0 \rangle$, where A is an operator belonging to the Hilbert space of a spin which is coupled to a thermal bath of harmonic oscillators, as described by the spin-boson model [21,37]. We consider the interaction Hamiltonian (1) with a perturbative coupling operator $L=\sigma_x$. Within this model, the bath can be characterized by the spectral strength

$$J(\omega) = \frac{\omega^3}{\omega_c^2} e^{-\omega/\omega_c},$$
(59)



FIG. 5. (Color online) Evolution of single mean values of $\text{Tr}_{S}(\rho_{s}(z_{0}'^{*}z_{0}'z_{0}|t)\sigma_{i})$, with $i=\{x, y, z\}$ and normalized initial condition $\rho_{s}(z_{0}'^{*}z_{0}'z_{0}|0)=\text{Tr}_{B}(|\psi_{0}\rangle|z_{0}\rangle\langle z_{0}'|\langle \psi_{0}|/\langle \psi_{0}|\langle z_{0}'|z_{0}\rangle|\psi_{0}\rangle)$ $=|\psi_{0}\rangle\langle \psi_{0}|/\langle \psi_{0}|\psi_{0}\rangle$. We have taken initial value already normalized, $|\psi_{0}\rangle=[(1+2i)]-\rangle+(1+i)|+\rangle]/\sqrt{7}$. The average of Eq. (47) is numerically computed using the evolution (17) for an ensemble of 100 trajectories (dotted line) and 10³ (long dashed line). The last result is practically equal to that of the analytical solutions of Eqs. (57) and (58) (solid line).

where ω_c is a cutoff frequency [21], here chosen as $\omega_c=1$. The correlation function of the noise generated by a thermal bath is given in terms of $J(\omega)$ as

$$\alpha(t) = \int_0^\infty d\omega J(\omega) \left[\coth\left(\frac{\omega\beta}{2}\right) \cos(\omega t) - i\sin(\omega t) \right].$$
(60)

The inverse temperature $\beta = (\kappa_B T)^{-1}$ is chosen according to the energy of the bath, and considering that the energy of the subsystem is very small compared to it [21]. As noted when introducing the reduced propagator, when the environment is at high temperatures the noise statistics evolves quite significantly in time, so that a nonlinear equation needs to be considered in order to take this into account.

At low temperatures, however, the state distribution of the bath (i.e., the noise distribution) remains quite close to a Gaussian during the interaction, and linear equations are good enough. Since we are considering low temperatures, $\beta = 10$, the linear stochastic equations for the propagators presented in this paper are suitable. In order to reduce the computational effort, we propose a Fourier series of Eq. (60) of the form (40), with only m=6 oscillators. The correlation function obtained is a good approximation of Eq. (60), up to T=5, if we first compute the function C(m) with a high number of oscillators, and then sample it with m=6 values of frequency. By doing that, the six frequencies of oscillators entering in the sum (40) will correspond to some of the most representative values of the coefficients. The comparison between Eq. (60) and the approximate correlation for six oscillators is shown in Fig. 6.



FIG. 6. Thermal correlation function $\alpha(t)$, given by Eq. (60) with $\beta = 10$ (solid line), compared to its Fourier series α^{appx} given by Eq. (40) with only m=6 oscillators (long-dashed line). Despite the number of oscillators is very small, the approximation is quite good. This is due to the fact that the frequencies of these oscillators have been selected between the most significant ones of the function C(m).

For the thermal correlation function, we show in Fig. 7 the evolution of $\text{Tr}_{S}(\rho_{s}(z_{0}'^{*}z_{0}'z_{0}|t)\sigma_{i})$, with $i=\{x, y, z\}$. It can be observed how the average of Eq. (47), which is numerically computed using the evolution equation (17), approaches the result of Eq. (52) when a large enough ensemble of trajectories is used. Again, this is a numerical verification of the averages of the variable z_{t} , which has been analytically performed using a perturbative expansion of the propagators.

VI. CONCLUSIONS

We have derived in this paper the evolution equation of the reduced propagator that evolves vectors in the Hilbert space of the system conditioned to the initial and final states of the environment. Thanks to that, the reduced propagator is used, first, to derive a theory of non-Markovian multipletime correlations functions (MTCFs) and second to derive a master equation with general initial conditions.

Concerning the first point, we show how to obtain multiple-time correlation functions with the reduced propagator within a Monte Carlo method, so that it becomes a stochastic propagator. Furthermore, we derive the set of coupled differential equations that satisfy the two-time correlation functions in the weak-coupling limit. Such equations are more general that the quantum regression theorem, in the sense that they are valid in the non-Markovian case, when such a theorem is in general no longer applicable. They coincide with the QRT only for certain particular cases of couplings and correlations, and also within the Markovian limit in which $\alpha(t) = \Gamma \delta(t)$. We apply the theory of MTCF to two examples. The first is a solvable model for which we compute the two-time correlation functions explicitly, and show that they are equal to the result numerically obtained using stochastic propagators. In the second example, a system that



FIG. 7. (Color online) Evolution of $\text{Tr}_{S}(\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0})|t)\sigma_{i})$, with $i=\{x, y, z\}$ and normalized initial condition $\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0})|0\rangle = |\psi_{0}\rangle\langle\psi_{0}|/\langle\psi_{0}|\psi_{0}\rangle$. We have taken the value $|\psi_{0}\rangle = [(1+2i)]-\rangle + (1+i)|+\rangle]/\sqrt{7}$. The average of Eq. (47) is numerically computed using the evolution (17) for an ensemble of 14×10^{3} (dotted line) and 18×10^{6} (long dashed line) trajectories. The last result is practically equal to that of the general condition perturbative master equation (51) (solid line). The correlation function is obtained from the model (59) with six oscillators in the Taylor expansion (40), coupling parameter g=0.1, and rotating frequency $\omega_{S}=0.1$.

interacts with a bath with exponentially decaying correlation function through a nondiagonal coupling, we have compared the differential equations for two-time correlations with the result numerically obtained with the stochastic propagators. In both cases we have shown the validity of the equations derived in this work.

Concerning the calculus of master equations with general initial conditions, we first consider an initial pure state for the total system, $|\Psi_0\rangle = \int d\mu(z_0) |\psi_0(z_0^*)\rangle |z_0\rangle$, instead of the usual one $|\Psi_0\rangle = |\psi_0\rangle |0\rangle$. We derive the evolution equation of an object

$$\rho_{s}(z_{0}^{\prime*}z_{0}^{\prime}z_{0}^{\ast}z_{0}|t) = \int d\mu(z_{1})G(t0|z_{1}^{*}z_{0})|\psi_{0}(z_{0}^{\ast})\rangle$$
$$\times \langle \psi_{0}(z_{0}^{\prime})|G^{\dagger}(t0|z_{0}^{\prime*}z_{1})$$

such that $\rho_s(t) = \int d\mu(z_0) \int d\mu(z'_0) \rho_s(z'_0 z'_0 z_0 z_0 | t)$. Hence once the set of $|\psi_0(z_0^*)\rangle$ is known for each problem, the reduced density operator can be computed. According to its definition, the equation obtained for $\rho_s(z'_0 z'_0 z_0 | t)$ can be verified with the stochastic evolution of the reduced propagators, and then performing their ensemble average over z_1 . This is done for two examples, the exact model already described and a dissipative model of a spin coupled to a thermal environment. The reduced density matrix for an initial mixed state for the total system can be computed by using $\rho_s(z'_0 z'_0 z'_0 z'_0 | t)$ with $z'_0 = z_0$.

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