# Multiple-Time Correlation Functions for Non-Markovian Interaction: Beyond the Quantum Regression Theorem

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We derive the dynamical equation of the *reduced propagator,* an object that evolves state vectors of the system conditioned to the dynamics of its environment, which is not necessarily in the vacuum state at the initial time. Such a reduced propagator is essential to obtain multiple-time correlation functions (MTCFs). We also study the evolution of MTCFs within the weak-coupling limit and show that the quantum regression theorem is, in general, not satisfied. We illustrate the theory in two different cases: first, solving an exact model, and, second, presenting the results of the numerical integration for a system coupled with a thermal environment through a nondiagonal interaction.

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*Introduction and motivation.*—Many research contexts are focused on the dynamics of a system ($S$) that is affected by an environment ($\mathcal{B}$) from which it cannot be considered *isolated.* Such situations are encountered in statistical physics, condensed matter, and quantum optics. A concrete example is found in the dynamics of an atom ($S$) immersed in an electromagnetic field ($\mathcal{B}$) [1].

The dynamics of the system $S$ is usually described by its reduced density operator which verifies some master equation that can be Markovian or non-Markovian. In the Markovian case, the master equation is of the Lindblad type [1–5]. A complementary scheme to the master equation is the so-called *stochastic Schrödinger equations* (SSE) [1,3,6–9]. In the SSE approach a state vector in the Hilbert space of the system $S$ evolves in time under the influence of a *noise* that takes into account the interaction with the environment. Such evolution defines a *history* or *trajectory.* An average over many histories (for different noise realizations) leads to the reduced density matrix of $S$. In the non-Markovian case, the Redfield master equation was developed within the context of nuclear magnetic resonance [10,11]. In addition, non-Markovian SSE have been established very recently for systems influenced by a structured environment [12–19]. In all these schemes one-time averages are successfully computed. Nonetheless, *multiple-time correlation functions* (MTCFs) are involved in many observable quantities, in particular, the spectrum of the radiating field of an atom which requires two-time averages [1,5].

In the Markovian case, an important tool to compute two-time correlation functions is the quantum regression theorem (QRT) [1,5,20]. The theory of stochastic Schrödinger equations, which was initially elaborated to compute the expectation values of system observables, has been extended [1,21–23] to calculate MTCFs for the Markovian case. In addition, such stochastic methods are in accordance with the results expected from the QRT. It is then natural to develop an equivalent theory of MTCFs for *non-Markovian* interactions within the stochastic Schrödinger equations approach. Of particular interest are those cases where the QRT *is not valid.*

Lately there has been an increasing interest in developing a theory of non-Markovian MTCFs that can be applied to systems with experimental interest. Because of its potential applications, photonic band-gap (PBG) materials constitute an interesting example. In PBG the electromagnetic field displays a band-gap structure, so that an atom interacting with such field displays non-Markovian dynamics near the band edge [24,25]. In this context, there are several works dealing with the dynamics of few-level atoms in PGB materials [24,26,27] where non-Markovian stochastic Schrödinger equations have been successfully applied [28]. Therefore, the theory of MTCFs that we present in this Letter is important from both experimental and theoretical points of view.

*Multiple-time correlation functions.*—A frequently used Hamiltonian model in the study of the dynamics of $S$, with Hamiltonian $H_S$, in interaction with $\mathcal{B}$, described by the Hamiltonian $H_B$, is

$$H = H_S + g\sum_n (g_n L a_n^\dagger + g_n^* L^\dagger a_n) + \sum_n \omega_n a_n^\dagger a_n, \quad (1)$$

where the operator $L$ acts on the Hilbert space of the system, $a_n$ and $a_n^\dagger$ are the annihilation and creation operators on the environment Hilbert space, and $g$ is a suitable perturbation parameter that eventually can be taken equal to one. The $g_n's$ are the coupling constants and the $\omega_n's$ are the frequencies of the harmonic oscillators that constitute the environment [29].

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

 

$$C_{\mathbf{A}}(\mathbf{t}|\Psi_0) \equiv \langle \Psi_0|A_1(t_1)\cdots A_N(t_N)|\Psi_0\rangle, \tag{2}$$

with $t_1 > t_2 > \cdots > t_N$ and $\mathbf{t} = \{t_1, \ldots, t_N\}$. 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., $|\Psi_0\rangle = |\psi_0\rangle|z_0\rangle$.

In the partial interaction picture with respect to the environment, the $N$-time correlation function is defined as $C_{\mathbf{A}}(\mathbf{t}|\Psi_0) = \langle \Psi_0|\prod_{i=1}^N \mathcal{U}_I^{-1}(t_i,0)A_i\mathcal{U}_I(t_i,0)|\Psi_0\rangle$, where $\mathcal{U}_I$ is the evolution operator of the system in the interaction picture. A suitable basis to treat the environment $\mathcal{B}$ is a coherent state basis, $|z_1, z_2, \ldots, z_n, \ldots\rangle = |z\rangle$ in the Bargmann representation [1,30]. In this basis the resolution of the identity is given by $1 = \int d\mu(z)|z\rangle\langle z|$ with $d\mu(z) = \prod_i[d^2z_i \exp(-|z_i|^2)/\pi]$ and thus, when inserted in the definition of $C_{\mathbf{A}}(\mathbf{t}|\Psi_0)$, it follows that

$$C_{\mathbf{A}}(\mathbf{t}|\Psi_0) = \int d\mu(z)\langle\psi_0|G^{-1}(0,1)\prod_{i=1}^N A_i G(i,i+1)|\psi_0\rangle, \tag{3}$$

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 $z_i$. Once their time evolution is solved, the time correlation function (3) can be obtained. Therefore, to proceed further we need to derive the equation of motion of the reduced propagator $G(i,i+1)$ by considering its time derivative with respect to $t_i$. Taking into account that the evolution operator $\mathcal{U}_I$ satisfies the Schrödinger equation in the partial interaction picture, after some manipulations we arrive at the equation

$$\begin{aligned}\frac{\partial G(i,i+1)}{\partial t_i} &= (-iH_S + Lz_{i,t_i}^* - L^\dagger z_{i+1,t_i})G(i,i+1)\\ &\quad - L^\dagger\int_{t_{i+1}}^{t_i} d\tau\alpha(t_i-\tau)\langle z_i|\mathcal{U}_I(t_i,t_{i+1})\\ &\quad \times L(\tau,t_{i+1})|z_{i+1}\rangle,\end{aligned} \tag{4}$$

with $L(t',t) = e^{iH_B t}e^{-iH(t-t')}Le^{iH(t-t')}e^{-iH_B t}$, $z_{i,t} = i\sum_n g_n z_{i,n}e^{i\omega_n t}$, $\alpha(t-\tau) = \sum_n |g_n|^2 e^{-i\omega_n(t-\tau)}$, 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 its time autocorrelation function, as it can be verified by computing the average $M[z_{i,t}z_{i,\tau}^*]$ regarding the measure $d\mu(z)$. The integration of Eq. (4) still presents the inconvenience that the matrix element $\langle z_i|\mathcal{U}_I(t_i,t_{i+1}) \times L(\tau,t_{i+1})|z_{i+1}\rangle$ cannot be in general computed exactly or expressed as a function of the reduced propagator. This would convert (4) into an explicit equation for this propagator. Since exact solutions can be obtained only in very exceptional cases, some approximate scheme must be taken.

One possible way is to assume that $\langle z_i|\mathcal{U}_I(t_i,t_{i+1}) \times L(\tau,t_{i+1})|z_{i+1}\rangle = O(z_{i+1}, z_i^*, t_{i+1}, \tau)G(i,i+1)$, where the operator $O$ has to be constructed [31], 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. (4) reads

$$\begin{aligned}\frac{\partial G(i,i+1)}{\partial t_i} &= \Big(-iH_S + Lz_{i,t_i}^* - L^\dagger z_{i+1,t_i} - L^\dagger\\ &\quad \times \int_{t_{i+1}}^{t_i} d\tau\alpha(t_i-\tau)O(z_{i+1}, z_i^*, t_{i+1}, \tau)\Big)\\ &\quad \times G(i,i+1).\end{aligned} \tag{5}$$

Equation (4) or its approximate versions, in particular Eq. (5), 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. They constitute one of the results of this Letter and are the starting point to compute the MTCFs in the non-Markovian case. Indeed, it is possible to solve (3) 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 (3) 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 $|\phi_N\rangle = G(N,N+1)|\psi_0\rangle$ is obtained, 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" leads to the MTCFs defined in (3).

It is useful to note that, since the equation for the reduced propagator (4) is made for an initial state of the environment different from the vacuum, it allows one to evaluate expectation values and correlation functions of system observables with *more general initial conditions than the one usually taken,* i.e., $|\Psi_0\rangle = |\psi_0\rangle|\text{vacuum}\rangle$ [32].

*Beyond the quantum regression theorem. Weak-coupling limit.*—We have seen how to compute MTCFs with the stochastic method. Nonetheless, this may turn out to be an expensive strategy from the numerical point of view, especially when a large number of environmental degrees of freedom is needed to correctly describe its correlation function $\alpha(t)$. For these cases, we present a set of coupled differential equations which evolves the non-Markovian two-time correlations up to second order in a convenient perturbative parameter $g$ and where the stochastic average has been done analytically.

The method that we follow starts by considering the derivative with respect to $t'$ of the two-time correlation $\langle\psi_0 \mid G^\dagger(z_1 0|t'0)AG(z_1^* z_2|t't)BG_{t,0}(z_2^* 0|t0) \mid \psi_0\rangle$ and then performing analytically the average over the variables $z_1$ and $z_2$.

We then arrive at the following set of differential equations for the two-time correlation functions up to $\mathcal{O}(g^3)$ (we will give the details elsewhere [32]):

$$\frac{d}{dt'}\langle\Psi_0 \mid A(t')B(t) \mid \Psi_0\rangle = \langle\Psi_0|\Big(i\{[H_s, A]\}(t')B(t) + \int_0^{t'} d\tau\alpha^*(t'-\tau)\{V_{\tau-t'}L^\dagger[A, L]\}(t')B(t) + \int_0^{t'} d\tau\alpha(t'-\tau)\{[L^\dagger, A]V_{\tau-t'}L\}(t')B(t) + \int_0^{t} d\tau\alpha(t'-\tau)\{[L^\dagger, A]\}(t')\{[B, V_{\tau-t}L]\}(t)\Big)|\Psi_0\rangle, \tag{6}$$

where $\{ABC\}(t') = \mathcal{U}_I^{-1}(t')ABC\mathcal{U}_I(t') = A(t')B(t')C(t')$. The time dependency denoted by $V_{t'}L \equiv \exp\{i\mathcal{L}_S t'\}L = \exp(iH_S t')L\exp(-iH_S t')$ is the free system Liouville operator, which can be solved easily from the usual Heisenberg equation. In this notation, the initial conditions appearing in (6) are the quantum mean values $\langle\psi_0|\{AB\}\times(t)|\psi_0\rangle$, which can be computed with the usual master equation. It is clear that the QRT does not hold due to the last term in Eq. (6) containing $[L^\dagger, A]$ and $[B, V_{\tau-t}L]$. Please note that this term is zero in the Markovian case, since the corresponding correlation function $\alpha(t'-\tau) = \Gamma\delta(t'-\tau)$ vanishes in the domain of integration from 0 to $t$.

*A solvable example.*—To illustrate the theory proposed in this Letter, we apply it to a simple solvable model described by the Hamiltonian (1) with $L = \sigma_z$ and $H_S = -\frac{\omega}{2}\sigma_z$. This model describes the dynamics of system state vectors towards one of the eigenstates of the system Hamiltonian. In this case $[H_S, L] = 0$ and therefore $O = L$ [see Eq. (5)].

Let us consider the two-time correlation of $A = \{\{0, \alpha\}, \{\beta, 0\}\}$ and $B = \{\{1, 0\}, \{-1, 0\}\} = \sigma_z$. For an initial system state $|\psi_0\rangle = \psi_{01}|+\rangle + \psi_{02}|-\rangle$ after computing all the Gaussian integrals, Eq. (3) reads as follows:

$$C_{AB}(t't|\Psi_0) = e^{-2I_{00}^{t't'}[\alpha]}\{\beta\psi_{02}^*\psi_{01}e^{-i\omega t'} - \alpha\psi_{01}^*\psi_{02}e^{i\omega t'}\}, \tag{7}$$

with the definition $I_{ac}^{bd}[\alpha] \equiv \int_a^b d\tau \int_c^d ds\alpha(\tau - s)$. In the case $A = B = \sigma_z$, we have $C_{\sigma_z\sigma_z} = 1$. Another type of two-time correlation function corresponds to the observables $A = \{\{0, \alpha\}, \{\beta, 0\}\}$ and $B = \{\{0, \alpha'\}, \{\beta', 0\}\}$ and is given by

$$C_{AB}(t't|\Psi_0) = e^{\tilde{D}(t't)}\{\alpha\beta'|\psi_{01}|^2e^{i\omega(t'-t)} + \alpha'\beta|\psi_{02}|^2e^{-i\omega(t'-t)}\}, \tag{8}$$

with $\tilde{D}(t't) = I_{00}^{t'\tau}(\alpha^*) + I_{tt}^{t'\tau}(\alpha) + I_{00}^{t\tau}(\alpha) + I_{0t}^{t't'}(\alpha) - I_{t0}^{t't}(\alpha) - I_{00}^{t't}(\alpha)$. Equation (6) establishes a set of coupled differential equations among two-time correlation functions. In the first case, the QRT holds because $C_{AB}$ belongs to a set of two-time correlation functions for which the last term on the right-hand side (RHS) of (6) is zero. This occurs for $C_{\sigma_x\sigma_z}$, $C_{\sigma_y\sigma_z}$. In the second type of correlation functions, the last term on the RHS of (6) is not zero, and the QRT is not valid.

Figure 1 shows the two-time correlation functions $C_{\sigma_x\sigma_z}$ and $C_{\sigma_x\sigma_y}$, with two oscillators in the environment and parameters $g_1, g_2 = g = 1$ and $\omega_1 = 6, \omega_2 = 2$. The initial system state taken in all computations throughout this Letter is $|\Psi_0\rangle = |\psi_0\rangle|\text{vacuum}\rangle$ with $|\psi_0\rangle = [(1+2i)|+\rangle + (1+i)|-\rangle]/\sqrt{7}$. It is clear from this figure that the QRT does not apply for $C_{\sigma_x\sigma_y}$, since $[L, B] \neq 0$ and $[L^\dagger, A] \neq 0$.

*An example of dissipative system: The spin-boson model.*—Let us now apply the theory previously derived to the case of a system with $H_S = -\frac{\omega}{2}\sigma_z$, $L = \sigma_x$, and a dissipative interaction. Within the perturbative approximation, the operator $O(z, t, \tau)$ can be replaced by its zero order perturbative expansion, $V_{\tau-t}L = \sigma_{12}\exp\{i\omega(t-\tau)\} + \sigma_{21}\exp\{-i\omega(t-\tau)\}$. We chose a thermal environment characterized by the spectral strength $J(\Omega) = \frac{\Omega^3}{\Omega_c^2}e^{-\Omega/\Omega_c}$, where $\Omega_c$ is a cutoff frequency [15], here fixed to $\Omega_c = 1$. In terms of $J(\Omega)$, the environment correlation function, is given as $\alpha(t) = \int_0^\infty d\Omega J(\Omega)[\coth(\frac{\Omega\beta}{2})\cos(\Omega t) - i\sin(\Omega t)]$, where the inverse temperature $\beta = (\kappa_B T)^{-1}$ is chosen according to the energy of the bath, which is assumed to be much higher than the energy of the subsystem [15]. Because of numerical restrictions, we use the Fourier expansion of such a correlation function, $\alpha(t-\tau) = \sum_{m=-\nu/2}^{\nu/2} C(m)e^{-i\pi m(t-\tau)/T}$. Because of that, the coefficients $C(m) = \frac{1}{2T}\int_{-T}^{T} dt\alpha(t)e^{i\pi mt/T}$ can be sampled with only six of the most significant values of frequency. $T$ is the time window considered in the series expansion of the correlation function and $g_m = \sqrt{C(m)}$.

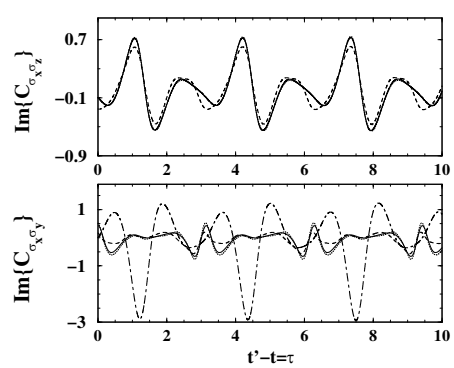


FIG. 1. In the top panel appears the imaginary part of $C_{\sigma_x\sigma_z}$. The solid line represents the analytical result (7), which in *this case* equals the quantum regression theorem result. Dashed and dotted lines represent the average stochastic result with $10^2$ and $10^5$ trajectories, respectively. The lower panel represents the imaginary part of $C_{\sigma_x\sigma_y}$. Comparing the result of the QRT (dot-dashed line), with the exact result given by (8) (solid line), it is clear that in this case QRT is not valid. The dashed and dotted lines represent an average $10^2$ and $10^4$ trajectories, respectively.

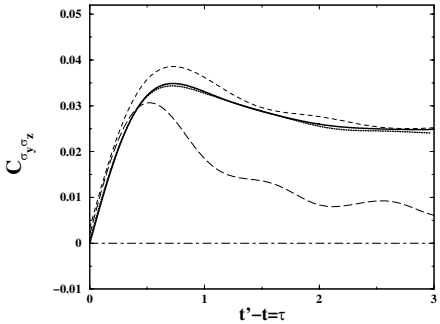


FIG. 2. Two-time correlation function $C_{\sigma_y\sigma_z}(t', t)$ for the coupling $L = \sigma_x$, and the Fourier series of the thermal correlation function with $\nu = 6$ oscillators. The parameters are $\omega = 0.1$, perturbative parameter $g = 0.1$, recurrence time $T = 5$, and initial time for the correlation $t = 1$. The solid line represents the solution of the system (6), whereas the long-dashed, dashed, and dotted lines give, respectively, the result of the stochastic method for $\kappa = 1 \times 10^6$, $8 \times 10^7$, and $5 \times 10^8$ trajectories. An increasing agreement with the system curve [Eq. (6)] is observed as the number of trajectories grows. The result derived from the QRT, displayed in the dot-dashed line, differs completely from (6), since in this case $[L^\dagger, A] \neq 0$ or $[B, V_{\tau-t}L] \neq 0$.

Figure 2 shows the decaying behavior with $\tau = t' - t$ of the correlation $C_{\sigma_y\sigma_z}$ and compares the evolution given by (6) and the stochastic method.

In conclusion, we present a theory for non-Markovian MTCFs from stochastic Schrödinger equations. The starting point of the theory is the evolution equation for the reduced propagator, which evolves vectors in the Hilbert space of the system $S$ conditioned to the dynamics of the environment. Remarkably, such an equation depends on two different histories of the bath. The MTCFs can be obtained from the reduced propagator. Furthermore, we have derived a set of coupled differential equations that satisfy the two-time correlation functions in the weak-coupling limit. This set of equations is a generalization of the quantum regression theorem and shows the conditions in which this theorem is not valid. We have illustrated the theory by applying it to two systems: For a solvable model, we have computed the two-time correlation functions explicitly, displaying an example in which the QRT is fulfilled and one in which it is not. When the system has a nondiagonal interaction with a thermal bath we have numerically integrated the MTCFs. Following the procedures here established, higher order time correlation functions might be calculated [32]. Besides its intrinsic theoretical interest, we believe that this work is relevant to the description of the dynamics of small systems, such as atoms immersed in photonic crystals as well as other situations where non-Markovian effects are significant.

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