

# Operator Correlations and Quantum Regression Theorem in Non-Markovian Lindblad Rate Equations

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**Abstract** Non-Markovian Lindblad rate equations arise from alternative microscopic interactions such as quantum systems coupled to composite reservoirs, where extra degrees of freedom mediate the interaction between the system and a Markovian reservoir, as well as from systems coupled to complex structured reservoirs whose action can be well approximated by a direct sum of Markovian sub-reservoirs (Budini in Phys. Rev. A 74:053815 2006). The purpose of this paper is two fold. First, for both kinds of interactions we find general expressions for the system operator correlations written in terms of the Lindblad rate propagator. Secondly, we find the conditions under which a quantum regression hypothesis is valid. We show that a non-Markovian quantum regression theorem can only be granted in a stationary regime, being a necessary condition the fulfillment of a detailed balance condition. This result is independent of the underlying microscopic interaction, providing a criterion for the validity of the regression hypothesis in non-Markovian Lindblad-like master equations. As an example, we study the correlations of a two-level system coupled to different kind of reservoirs.

**Keywords** Non-Markovian open quantum system dynamics · Quantum regression theorem

## 1 Introduction

In many areas of physics, one is confronted with the description of small quantum systems interacting with an uncontrollable environment. This situation is well understood when the reduced system dynamics follows a (completely positive) Markovian evolution [1–6].

One of the cornerstones of the theory of Markovian open quantum systems is the quantum regression theorem (QRT). This theorem, originally proposed by Lax [7, 8], allows to calculate multiple-time operators correlation functions from the knowledge of single-time expectation values, which in turn implies the knowledge of the density matrix evolution

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[4–6]. The importance of this theorem comes from the physical information contained in the operator correlations. In fact, in a stationary regime, it is possible to relate the Fourier transform of these objects with the spectrum of the decay process [3]. Furthermore, in radiant systems, the statistic of the scattered field can be described through system operator correlations [4–6].

Another central cornerstone of non-equilibrium quantum Markovian dynamics is the quantum detailed balance condition, which imposes severe symmetry properties on the operator correlations structure. While in classical stochastic processes this condition has a clear meaning in terms of transitions between the available states of the system [9, 10], in quantum dissipative systems this condition relies in the time reversal property of the underlying stationary microscopic Hamiltonian evolution [11–17]. The breakdown of this condition has direct experimental implications [18].

Although the applicability of the Markovian approximation range over many physical situations [1–6], there exist several real systems whose dynamics present strong departures from it. Remarkable examples are the presence of  $1/f$  [19, 20] and random telegraph noise [21, 22] in solid-state qubits, transport through interacting electron systems [23], anomalous intermittent fluorescence in nanocrystal quantum dots [24–31], band gap materials [32, 33], etc.

Consistently with the existence of experimental situations that can not be described by a Markovian evolution, in the context of different approaches recent effort was dedicated to characterize non-Markovian operator correlation dynamics [34–38].

While the description of non-Markovian processes may depend on each specific situation, there exists an increasing interest in describing these kinds of dynamics by introducing memory contributions in standard Lindblad evolutions [39–51]. These non-Markovian-Lindblad-like master equations provide a simple framework for describing strong non-Markovian effects. Nevertheless, these equations do not have associated a rule for calculating system operator correlations. Clearly, these objects can only be well defined by starting from a full Hamiltonian microscopic description.

In Refs. [52, 53] we have demonstrated that a broad class of non-Markovian Lindblad-like master equations can be derived from specific microscopic interactions, such as systems interacting with composite reservoirs, where extra degrees of freedom modulates the interaction with a Markovian reservoir and also from complex environments whose action can be approximated by a direct sum of Markovian sub-reservoirs (generalized Born-Markov approximation). In both cases, the density matrix evolution can be written in terms of a set of auxiliary states whose evolution involve Lindblad contributions with local coupling between all of them, resembling the structure of a classical rate equation. These “*Lindblad rate equations*” has been applied in the description of specific experimental situations such as the radiation pattern of fluorescent systems coupled to complex nano-environments [28–31]. Furthermore, they also arise when modeling dichotomous noise in solid-state qubits [21, 22], in the formulation of Bloch-Boltzman equations [54], in alternative consistent quantum measurement theories [55] and in the description of closed system-environment nano-arrangements [56, 57]. Derivations from pure theoretical conditions have been also explored [58, 59] and applied in the description of non-Markovian spin baths [60].

The goal of this paper is two fold. First, for both kinds of microscopic interactions that lead to a Lindblad rate equation we derive general expressions for the system operator correlations. This result is of fundamental importance for calculating physical observable defined in terms of operator correlation functions. Secondly, on the base of the previous result, the conditions under which a non-Markovian QRT can be formulated is explored. We find that the validity of the regression hypothesis is parallel to the fulfillment of a detailed balance

condition. As this result is independent of the underlying microscopic interaction, it provides a necessary condition for the validity of the regression hypothesis in time-convolution Lindblad-like evolutions.

The paper is outlined as follows. In Sect. 2, the definition and microscopic derivation of Lindblad rate equations are reviewed. The dual operators evolution is also introduced. In Sect. 3, starting from a full microscopic description, we derive the expressions for the operator correlations. In Sect. 4 we find the conditions under which a non-Markovian QRT can be formulated. In Sect. 5 we relate the fulfillment of the QRT in a stationary regime with the microreversibility property of the dissipative dynamics. In Sect. 6 we exemplify our theoretical results by analyzing the correlation dynamics of a two-level system embedded in different non-Markovian environments. In Sect. 7 we give the conclusions.

## 2 Lindblad Rate Equations

In this approach [52, 53], the density matrix of an open quantum system can be written as

$$\rho_S(t) = \sum_R \rho_R(t), \tag{1}$$

where the auxiliary states  $\rho_R(t)$  evolve as

$$\begin{aligned} \frac{d}{dt} \rho_R(t) = & \frac{-i}{\hbar} [H_R^{eff}, \rho_R(t)] - \{D_R, \rho_R(t)\}_+ + F_R[\rho_R(t)] \\ & - \sum_{\substack{R' \\ R' \neq R}} \{D_{R'R}, \rho_R(t)\}_+ + \sum_{\substack{R' \\ R' \neq R}} F_{RR'}[\rho_{R'}(t)]. \end{aligned} \tag{2}$$

The effective Hamiltonian  $H_R^{eff} = H_S + H_R$  is defined in terms of the system Hamiltonian  $H_S$  and a  $R$ -dependent shift contribution  $H_R$ , which is induced by the interaction with the reservoir. On the other hand, the dissipative diagonal superoperator contributions are defined by

$$D_R = \frac{1}{2} \sum_{\alpha, \gamma} a_R^{\alpha\gamma} V_\gamma^\dagger V_\alpha, \quad F_R[\bullet] = \sum_{\alpha, \gamma} a_R^{\alpha\gamma} V_\alpha \bullet V_\gamma^\dagger, \tag{3}$$

while the non-diagonal contributions reads

$$D_{R'R} = \frac{1}{2} \sum_{\alpha, \gamma} a_{R'R}^{\alpha\gamma} V_\gamma^\dagger V_\alpha, \quad F_{RR'}[\bullet] = \sum_{\alpha, \gamma} a_{RR'}^{\alpha\gamma} V_\alpha \bullet V_\gamma^\dagger. \tag{4}$$

With  $\{V_\alpha\}$ , we denote an arbitrary set of system operators. Then, the indexes  $\alpha$  and  $\gamma$  run from one up to  $(\dim \mathcal{H}_S)^2$ , where  $\dim \mathcal{H}_S$  is the system Hilbert space dimension. The (positive defined) matrices  $a_R^{\alpha\gamma}$  and  $a_{R'R}^{\alpha\gamma}$  characterize the rate constants of the dissipative evolution. The initial conditions of (2) read  $\rho_R(0) = P_R \rho_S(0)$ , where the definition of the positive weights  $P_R$  depend on each specific environment and satisfy  $\sum_R P_R = 1$ . The labeling indexes  $R$  run over an arbitrary domain related to the specific environment properties.

### 2.1 Non-Markovian Density Matrix Evolution

While the auxiliary states  $\rho_R(t)$  evolves under a local in time evolution (Markovian), the system density matrix  $\rho_S(t)$  is characterized by a non-Markovian dynamics. In order to get its evolution, we write the evolution equation (2) as

$$\frac{d|\rho(t)\rangle}{dt} = \hat{\mathcal{L}}_H |\rho(t)\rangle + \hat{\mathcal{M}} |\rho(t)\rangle. \tag{5}$$

In order to simplify the notation, we introduced the column vector  $|\rho\rangle \equiv (\rho_1, \rho_2, \dots, \rho_R, \dots)^T$ , where T denote a transposition operation. Furthermore, the hat symbol denote matrices in the  $R$ -space whose elements are superoperators acting on the (auxiliary) system states. The Hamiltonian contribution  $\hat{\mathcal{L}}_H$  is defined by its matrix elements

$$(\hat{\mathcal{L}}_H)_{RR'}[\bullet] = -(i/\hbar)[H_S, \bullet]\delta_{RR'}, \tag{6}$$

while the matrix elements of  $\hat{\mathcal{M}}$  reads

$$\begin{aligned} \hat{\mathcal{M}}_{RR'}[\bullet] = & \delta_{R,R'} \left\{ \frac{-i}{\hbar} [H_R, \bullet] - \{D_R, \bullet\}_+ + F_R[\bullet] \right\} \\ & + F_{RR'}[\bullet] - \delta_{R,R'} \sum_{\substack{R'' \\ R'' \neq R}} \{D_{R''R}, \bullet\}_+. \end{aligned} \tag{7}$$

With this notation, the initial condition reads  $|\rho(0)\rangle = |P\rangle\rho_S(0)$ , where we have introduced the vector  $|P\rangle = (P_1, P_2, \dots, P_R, \dots)^T$ . The system state (1) reads  $\rho_S(t) = (1|\rho(t))$ , where  $|1\rangle$  is the row vector with elements equal to one. Notice that the normalization of the statistical weights it written as  $(1|P) = (P|1) = 1$ .

From (5), the system state can be trivially written in the Laplace domain as

$$\rho_S(u) = \left( 1 \left| \frac{1}{u - (\hat{\mathcal{L}}_H + \hat{\mathcal{M}})} \right| P \right) \rho_S(0), \tag{8a}$$

$$\equiv (1|\hat{\mathcal{G}}(u)|P)\rho_S(0), \tag{8b}$$

where  $u$  is the conjugate variable. Multiplying the right term by the identity operator written in the form  $1/(1|\hat{\mathcal{G}}(u)[u - (\hat{\mathcal{L}}_H + \hat{\mathcal{M}})]|P)$ , it is straightforward to arrive to the non-local evolution

$$\frac{d\rho_S(t)}{dt} = \mathcal{L}_H[\rho_S(t)] + \int_0^t d\tau \mathcal{K}(t - \tau)[\rho_S(\tau)], \tag{9}$$

where  $\mathcal{L}_H[\bullet] = -(i/\hbar)[H_S, \bullet]$ , and the system-superoperator-kernel  $\mathcal{K}(t)$  is defined in the Laplace domain by

$$\mathcal{K}(u)[\bullet] = (1|\hat{\mathcal{G}}(u)|P)^{-1} (1|\hat{\mathcal{G}}(u)\hat{\mathcal{M}}|P)[\bullet]. \tag{10}$$

In general, depending on the underlying structure, the evolution equation (9) involves many different memory functions, each one associated to a Lindblad contribution.

### 2.2 Operators Dual Evolution

Associated to the density matrix evolution equation (9), it is possible to define a dual evolution, where the time dynamics is carried out by the system operators. As in standard Lindblad equations [1], the relation

$$\text{Tr}_S\{\rho_S(t)A_S\} = \text{Tr}_S\{\rho_S(0)A_S(t)\}, \tag{11}$$

where  $A_S$  is an arbitrary system operator, allows to define the operators dual evolution. We write the system operator as

$$A_S(t) = \sum_R P_R A_R(t) = (P|A(t)), \tag{12}$$

where the auxiliary operators  $A_R(t)$  define the vector  $|A\rangle \equiv (A_1, A_2, \dots, A_R, \dots)^T$ . By using the relation (11), their evolution is defined as

$$\frac{d|A(t)\rangle}{dt} = \hat{\mathcal{L}}_H^\# |A(t)\rangle + \hat{\mathcal{M}}^\# |A(t)\rangle, \tag{13}$$

with  $A_R(0) = A_S(0)$ . The dual Liouville matrix superoperator reads  $\hat{\mathcal{L}}_H^\# = -\hat{\mathcal{L}}_H$  and the matrix elements of  $\hat{\mathcal{M}}^\#$  are given by

$$\begin{aligned} \hat{\mathcal{M}}_{RR'}^\#[\bullet] = & \delta_{R,R'} \left\{ \frac{i}{\hbar} [H_R, \bullet] - \{D_R^\#, \bullet\}_+ + F_R^\#[\bullet] \right\} \\ & + F_{RR'}^\#[\bullet] - \delta_{R,R'} \sum_{\substack{R'' \\ R'' \neq R}} \{D_{R''}^\#, \bullet\}_+, \end{aligned} \tag{14}$$

with the definitions

$$D_R^\# = D_R, \quad F_R^\#[\bullet] = \sum_{\alpha,\gamma} a_R^{\alpha\gamma} V_\gamma^\dagger \bullet V_\alpha, \tag{15}$$

while the non-diagonal contributions read

$$D_{R'R}^\# = D_{R'R}, \quad F_{R'R'}^\#[\bullet] = \sum_{\alpha,\gamma} a_{R'R'}^{\alpha\gamma} V_\gamma^\dagger \bullet V_\alpha. \tag{16}$$

The non-Markovian evolution of  $A_S(t)$  follows from (12) and (13). By writing the dynamics in the Laplace domain as

$$A_S(u) = \left( P \left| \frac{1}{u - (\hat{\mathcal{L}}_H^\# + \hat{\mathcal{M}}^\#)} \right| 1 \right) A_S(0), \tag{17a}$$

$$\equiv (P|\hat{\mathcal{G}}^\#(u)|1)A_S(0), \tag{17b}$$

it is possible to obtain

$$\frac{dA_S(t)}{dt} = \mathcal{L}_H^\#[A_S(t)] + \int_0^t d\tau \mathcal{K}^\#(t - \tau)[A_S(\tau)], \tag{18}$$

where  $\mathcal{L}_H^\# = -\mathcal{L}_H$  and the superoperator kernel  $\mathcal{K}^\#$  is defined by

$$\mathcal{K}^\#(u)[\bullet] = (P|\hat{\mathcal{G}}^\#(u)|1)^{-1} (P|\hat{\mathcal{G}}^\#(u)\hat{\mathcal{M}}^\#|1)[\bullet]. \tag{19}$$

### 2.3 Microscopic Derivation

Lindblad rate equations arise from different kind of microscopic interactions. Here, in order to define the notation of the next sections, we review the derivation from composite bipartite reservoirs. In Appendix A we review the derivation from a generalized Born-Markov approximation [52, 53].

Bipartite composite reservoirs refers to the situation where there exist extra degrees of freedom  $U$  that modulate the interaction (the entanglement) between a system  $S$  and a Markovian reservoir  $B$ . The total microscopic dynamics is defined by the Hamiltonian

$$H_T = H_S + H_U + H_B + H_I. \tag{20}$$

$H_S$  represent the system Hamiltonian.  $H_B$  is the Hamiltonian of the *Markovian* environment. On the other hand,  $H_U$  is the Hamiltonian of the extra degrees of freedom. The interaction Hamiltonian  $H_I$  couples the three involved parts. The total density matrix  $\rho_T(t)$  evolves as

$$\frac{d\rho_T(t)}{dt} = \frac{-i}{\hbar} [H_T, \rho_T(t)]. \tag{21}$$

The system states follows from  $\rho_T(t)$  after tracing out the degrees of freedom of  $B$  and  $U$

$$\rho_S(t) = \text{Tr}_{UB}\{\rho_T(t)\}, \tag{22a}$$

$$= \text{Tr}_{UB}\{\rho_{SU}(t) \otimes \rho_B\}, \tag{22b}$$

$$= \text{Tr}_U\{\rho_{SU}(t)\}. \tag{22c}$$

For writing the second line, we assume that the interaction  $H_I$  weakly couples the Markovian reservoir  $B$  with the systems  $U$  and  $S$ . Therefore, its degrees of freedom can be eliminated through a standard Born-Markov approximation [1–6], which allows to write the total density matrix as the external product between the stationary bath state  $\rho_B$  and the state  $\rho_{SU}(t)$  corresponding the composite system  $SU$ .

By introducing a complete basis  $\{|R\rangle\}$  in the Hilbert space of  $U$ , from (22), the system density matrix reads

$$\rho_S(t) = \sum_R \langle R | \rho_{SU}(t) | R \rangle, \tag{23a}$$

$$\equiv \sum_R \rho_R(t) = (1 | \rho(t)). \tag{23b}$$

Then, we notice that the sum structure equation (1) have a trivial interpretation in terms of a trace operation. Each state  $\rho_R$  correspond to the system dynamics given that the system  $U$  is in the state  $|R\rangle$ .

As the reservoir  $B$  can be traced-out in a Born-Markov approximation, the evolution of  $\rho_{SU}(t)$  can be written in terms of a standard Markovian Lindblad equation,  $(d/dt)\rho_{SU}(t) = L_{SU}[\rho_{SU}(t)]$ . Then, the evolution of each auxiliary state  $\rho_R(t)$  reads

$$\frac{d\rho_R(t)}{dt} = \langle R | L_{SU}[\rho_{SU}(t)] | R \rangle, \tag{24a}$$

$$= \sum_{R'} \hat{\mathcal{L}}_{RR'} \rho_{R'}(t), \tag{24b}$$

where  $\hat{\mathcal{L}}_{RR'}$  define a Lindblad rate equation, i.e.,

$$\hat{\mathcal{L}} = \hat{\mathcal{L}}_H + \hat{\mathcal{M}}. \tag{25}$$

Here,  $\hat{\mathcal{L}}_H$  and  $\hat{\mathcal{M}}$  are defined by (6) and (7) respectively. We remark that the possibility of writing the evolution of  $\rho_R(t)$  as a Lindblad rate equation impose severe symmetry conditions on the superoperator  $L_{SU}$  (or equivalently on the interaction Hamiltonian  $H_I$ ), which must not to couple the coherences and populations of  $U$  [52, 53] (see for example Sect. 6.2 and the interaction Hamiltonian in Ref. [31]).

When the system  $S$  and the extra degrees of freedom  $U$  begin in a uncorrelated state,  $\rho_{SU}(0) = \rho_S(0) \otimes \rho_U(0)$ , where  $\rho_S(0)$  and  $\rho_U(0)$  are arbitrary initial states for both systems, from (23) the initial conditions of the auxiliary states reads  $\rho_R(0) = P_R \rho_S(0)$ , where

$$P_R = \langle R | \rho_U(0) | R \rangle. \tag{26}$$

Therefore, here the weights  $P_R$  are defined by the initial populations of  $U$ .

### 3 Operator Correlations

Operators correlations can only be well defined and calculated by starting from a full microscopic dynamics. Here, we determine the corresponding expressions for the case of bipartite composite reservoirs. In Appendix A we arrive to the same expressions for the case of complex structured reservoirs described in a generalized Born-Markov approximation.

Let us introduce a complete set of operators  $\{A_\mu\}$  of the system, collected into a vector  $\mathbf{A}$ , and consider the *microscopic* expression for expectation values

$$\overline{\mathbf{A}(t)} \equiv \text{Tr}_{SUB}[\mathbf{A}(t)\rho_T(0)], \tag{27}$$

as well as for the correlation functions

$$\overline{O(t)\mathbf{A}(t + \tau)} \equiv \text{Tr}_{SUB}[O(t)\mathbf{A}(t + \tau)\rho_T(0)], \tag{28}$$

where  $O(t)$  is an arbitrary system operator. The time dependence of the operators refers to a Heisenberg representation with respect to the total Hamiltonian (20), i.e.,  $O(t) = \exp[(i/\hbar)tH_T]O(0)\exp[-(i/\hbar)tH_T]$ .

From (22) and (23), trivially we can write the expectation values as an sum of the averages corresponding to each state  $\rho_R(t)$ , i.e.,

$$\overline{\mathbf{A}(t)} = \sum_R \text{Tr}_S[\mathbf{A}\rho_R(t)] \equiv \sum_R \overline{\mathbf{A}(t)}_R \tag{29a}$$

$$= (1|\overline{\mathbf{A}(t)}). \tag{29b}$$

In order to work out the operator correlations, we first express the total initial density matrix as  $\rho_T(0) = \exp[(i/\hbar)tH_T]\rho_T(t)\exp[-(i/\hbar)tH_T]$ . Then, by using the cyclic property of the trace operation, from (28) we obtain

$$\overline{O(t)\mathbf{A}(t + \tau)} = \text{Tr}_S\{\mathbf{A} \text{Tr}_{UB}[O_{SUB}(\tau)]\}, \tag{30}$$

where the operator  $O_{SUB}(\tau)$  satisfies

$$\frac{d}{d\tau} O_{SUB}(\tau) = -\frac{i}{\hbar} [H_T, O_{SUB}(\tau)], \tag{31}$$

with  $O_{SUB}(\tau)|_{\tau=0} = \rho_T(t)O(0) = \rho_{SU}(t)O(0) \otimes \rho_B$ , where the last equality follows from the Markovian property of the reservoir  $B$ . As the operator  $O_{SUB}(\tau)$  evolves as the total density matrix  $\rho_T(t)$ , (21), the dynamics of  $\text{Tr}_{UB}[O_{SUB}(\tau)]$  can also be written as a Lindblad rate equation, delivering

$$\text{Tr}_{UB}[O_{SUB}(\tau)] = \sum_{RR'} (e^{\tau \hat{L}})_{RR'} \rho_{R'}(t) O, \tag{32a}$$

$$= (1|e^{\tau \hat{L}}|\rho(t)O), \tag{32b}$$

where  $\hat{L}$  is defined by (25). From (30), it follows

$$\begin{aligned} \overline{O(t)\mathbf{A}(t+\tau)} &= \sum_{RR'} \text{Tr}_S\{\mathbf{A}(e^{\tau \hat{L}})_{RR'}[\rho_{R'}(t)O]\}, \\ &\equiv \sum_R \overline{O(t)\mathbf{A}(t+\tau)_R}, \\ &= (1|\overline{O(t)\mathbf{A}(t+\tau)}). \end{aligned} \tag{33}$$

This equation give us the desired expression for the operator correlations. It is written in terms of the generator  $\hat{L}_{RR'}$  of the Lindblad rate evolution equation (2). Higher correlations operators can also be obtained in a similar way. For example, using the same steps as before, for arbitrary system operators  $O_1$  and  $O_2$ , it is possible to obtain

$$\begin{aligned} \overline{O_1(t)\mathbf{A}(t+\tau)O_2(t)} &= \sum_{RR'} \text{Tr}_S\{\mathbf{A}(e^{\tau \hat{L}})_{RR'}[O_2\rho_{R'}(t)O_1]\} \\ &\equiv \sum_R \overline{O_1(t)\mathbf{A}(t+\tau)O_2(t)_R}, \\ &= (1|\overline{O_1(t)\mathbf{A}(t+\tau)O_2(t)}). \end{aligned} \tag{34}$$

The operators correlations take a simple form when expressed in term of the dual super-operators introduced in Sect. 2.2. By using the property

$$\text{Tr}_S\{(A|\hat{L}|B)\} = \text{Tr}_S\{(B|\hat{L}^\#|A)\}, \tag{35}$$

valid for arbitrary R-vectors (of operators)  $|A\rangle$  and  $|B\rangle$ , and where

$$\hat{L}^\# = \hat{L}_H^\# + \hat{\mathcal{M}}^\#, \tag{36}$$

from (33), we get

$$\begin{aligned} \overline{O(t)\mathbf{A}(t+\tau)} &= \text{Tr}_S\{\mathbf{A}(1|e^{\tau \hat{L}}|\rho(t)O)\}, \\ &= \text{Tr}_S\{(\mathbf{A}|e^{\tau \hat{L}}|\rho(t)O)\}, \\ &= \text{Tr}_S\{(\rho(t)O|e^{\tau \hat{L}^\#}|\mathbf{A})\}, \end{aligned} \tag{37}$$



while from (34) it follows

$$\begin{aligned} \overline{O_1(t)\mathbf{A}(t + \tau)O_2(t)} &= \text{Tr}_S\{\langle \mathbf{A} | e^{\tau \hat{\mathcal{L}}} | O_2 \rho(t) O_1 \rangle\}, \\ &= \text{Tr}_S\{\langle O_2 \rho(t) O_1 | e^{\tau \hat{\mathcal{L}}^\#} | \mathbf{A} \rangle\}, \\ &= \text{Tr}_S\{\langle \rho(t) O_1 | [e^{\tau \hat{\mathcal{L}}^\#} | \mathbf{A} ] O_2 \rangle\}. \end{aligned} \tag{38}$$

### 4 Quantum Regression Theorem

For Markovian master equations the QRT [4–6] provides a direct relation between the time evolution of the expectation values of system observable and their corresponding correlation functions. Here, based on the results of the previous section, we will explore the possibility of formulating an equivalent relation when the system evolution is defined through a Lindblad rate equation. Taking in account the results of Appendix A, the next results apply for both underlying microscopic interactions, i.e., composite bipartite reservoirs and the generalized Born-Markov approximation.

#### 4.1 Expectation and Correlation Evolutions

From the previous results, independently of the microscopic interaction, from (29) and (33), we can write the evolution of both, expectation values and operator correlations as

$$\frac{d}{dt} \overline{\mathbf{A}(t)} = \sum_{RR'} \hat{\mathbb{M}}_{RR'} \overline{\mathbf{A}(t)}_{R'}, \tag{39a}$$

$$\frac{d}{d\tau} \overline{O(t)\mathbf{A}(t + \tau)} = \sum_{RR'} \hat{\mathbb{M}}_{RR'} \overline{O(t)\mathbf{A}(t + \tau)}_{R'}. \tag{39b}$$

With blackboard bold letters, we denote matrices acting on the indexes of the vector (of system operators)  $\mathbf{A}$ . The hat symbol denotes their dependence on the  $R$ -indexes. For each pair of indexes  $R$  and  $R'$ , the set of matrices  $\hat{\mathbb{M}}_{RR'}$  are defined by the relation

$$\sum_R \text{Tr}_S\{\mathbf{A} \hat{\mathcal{L}}_{RR'} [O]\} = \sum_R \hat{\mathbb{M}}_{RR'} \text{Tr}_S\{\mathbf{A} O\}, \tag{40}$$

where  $\hat{\mathcal{L}}_{RR'}$  is the generator of the Lindblad rate evolution, (25).

As for the density matrix, it is possible to get a closed non-Markovian evolution for the expectation values and correlations. First, from (39a), the expectation values can be expressed in the Laplace domain as

$$\overline{\mathbf{A}(u)} = (1 | \hat{\mathbb{G}}(u) | P) \overline{\mathbf{A}(0)}, \tag{41}$$

while from (39b) the correlations read

$$\overline{O(t)\mathbf{A}(t + \tau)} = (1 | \hat{\mathbb{G}}(u) | \overline{O(t)\mathbf{A}(t)}), \tag{42}$$

where we have introduced the propagator  $\hat{\mathbb{G}}(u) \equiv (u + \hat{\mathbb{M}})^{-1}$ . After introducing in (41) the identity operator in the form  $\overline{\mathbf{A}(u)} = (1 | \hat{\mathbb{G}}(u) (u + \hat{\mathbb{M}}) | P) (1 | \hat{\mathbb{G}}(u) | P) \overline{\mathbf{A}(0)}$ , we arrive to the closed evolution

$$\frac{d}{dt} \overline{\mathbf{A}(t)} = - \int_0^t dt' \mathbb{K}(t - t') \overline{\mathbf{A}(t')}. \tag{43}$$

Using a similar procedure, from (42), for the correlations we get

$$\frac{d}{d\tau} \overline{O(t)\mathbf{A}(t+\tau)} = - \int_0^\tau dt' \mathbb{K}(\tau-t') \overline{O(t)\mathbf{A}(t+t')} + \mathbf{I}(t, \tau). \tag{44}$$

The matrix kernel  $\mathbb{K}(t)$  fulfills the equation

$$\mathbb{K}(u) = (1|\hat{\mathbb{G}}(u)|P)^{-1} (1|\hat{\mathbb{G}}(u)\hat{\mathbb{M}}|P), \tag{45}$$

while the inhomogeneous term  $\mathbf{I}(t, \tau)$  is defined by

$$\mathbf{I}(t, u) = (1|\hat{\mathbb{G}}(u)|P)^{-1} (1|\hat{\mathbb{G}}(u)|\overline{O(t)\mathbf{A}(t)}) - (1|\overline{O(t)\mathbf{A}(t)}). \tag{46}$$

Besides that (39b) has the same structure as (39a), the inhomogeneous term is only present in the correlation evolution, (44).  $\mathbf{I}(t, \tau)$  arise because, in contrast with the expectation evolutions, the initial condition of each contribution in (39b),  $\overline{O(t)\mathbf{A}(t)}_R = \text{Tr}_S[O\mathbf{A}\rho_R(t)]$ , depend on the auxiliary states  $\rho_R(t)$ . In contrast, as (39a) is defined with initial conditions fixed at  $t = 0$ , its initial condition  $\overline{\mathbf{A}(0)}_R = \text{Tr}_S[\mathbf{A}\rho_R(0)] = P_R \text{Tr}_S[\mathbf{A}\rho_S(0)]$  does not depends on the auxiliary states, which in turn implies that the inhomogeneous term is not present in the averaged evolution equation (43). Consistently, notice that  $\mathbf{I}(t, \tau)$  always vanishes at the initial time, i.e.,  $\mathbf{I}(0, \tau) = 0$ .

Due to the inhomogeneous term  $\mathbf{I}(t, \tau)$ , the QRT is not fulfilled in general. A QRT is only valid when this term vanish, which leads to the condition

$$(1|\hat{\mathbb{G}}(u)|\overline{O(t)\mathbf{A}(t)}) \stackrel{QRT}{=} (1|\hat{\mathbb{G}}(u)|P)(1|\overline{O(t)\mathbf{A}(t)}). \tag{47}$$

We realize that the previous equality is always satisfied for Markovian dynamics, i.e., when the superoperator  $\hat{\mathcal{L}}_{RR'}$  only involve a unique diagonal Lindblad contribution,  $\hat{\mathcal{L}}_{RR'} = \delta_{RR'}\mathcal{L}$  (with arbitrary unitary and dissipative terms). In fact, in this situation at any time it is satisfied  $\rho_R(t)/P_R = \rho_S(t)$ , which immediately implies the validity of (47) at all times recovering the QRT for Markovian dynamics [4–6]. In this case, the expectation and correlation evolutions are defined by local in time evolutions, (43) and (44) with  $\mathbb{K}(t) = \delta(t)\mathbb{K}$  and  $\mathbf{I}(t, \tau) = 0$ .

In the general non-Markovian case,  $\hat{\mathcal{L}}_{RR'} \neq \delta_{RR'}\mathcal{L}$ , the QRT is not valid. Nevertheless, we note that “a non-Markovian QRT can be asymptotically valid if the states

$$\rho_R^\infty \equiv \lim_{t \rightarrow \infty} \frac{\rho_R(t)}{\text{Tr}_S[\rho_R(t)]} \tag{48a}$$

do not depend on index  $R$  and the stationary condition

$$P_R = P_R^\infty \equiv \lim_{t \rightarrow \infty} \text{Tr}_S[\rho_R(t)] \tag{48b}$$

is satisfied.” In fact, under these conditions it follows  $\lim_{t \rightarrow \infty} \overline{O(t)\mathbf{A}(t)} = \lim_{t \rightarrow \infty} \text{Tr}_S\{O(0)\mathbf{A}(0)|\rho(t)\} = \lim_{t \rightarrow \infty} \overline{O(t)\mathbf{A}(t)}|P^\infty$ , and then condition equation (47) is automatically satisfied. However, if the asymptotic states  $\rho_R^\infty$  depends on  $R$  and/or  $P_R \neq P_R^\infty$ , the inhomogeneous term will contribute at all times, even in the asymptotic regime, and the QRT is invalidated. The same condition is valid for higher operators correlations.

In the context of bipartite reservoirs (Sect. 2.3), the condition equation (48a) guarantees that in the stationary regime the system  $S$  and the extra degrees of freedom  $U$  are statistically uncorrelated. Furthermore, (48b) implies that at the initial time, the system  $U$  is in its stationary state, i.e.,  $\rho_U(0) = \rho_U(\infty)$ . On the other hand, we notice that (48b) is always satisfied in diagonal Lindblad rate equations, i.e., equation (2) with  $a_{RR'}^{\alpha\gamma} = 0$ .

## 4.2 Non-Markovian Dynamics

The evolution equations (43) and (44) can be integrated in the Laplace domain. For the expectation values we get

$$\overline{\mathbf{A}(t)} = \mathbb{T}(t)\overline{\mathbf{A}(0)}, \quad (49a)$$

while for the correlations it follows

$$\overline{O(t)\mathbf{A}(t+\tau)} = \mathbb{T}(\tau)\overline{O(t)\mathbf{A}(t)} + \mathbf{F}(t, \tau). \quad (49b)$$

The non-Markovian propagator is defined in the Laplace domain as

$$\mathbb{T}(u) = \frac{1}{u + \mathbb{K}(u)}, \quad (50)$$

and the extra inhomogeneous term is

$$\mathbf{F}(t, \tau) = (1|\hat{\mathbb{G}}(\tau)|\overline{O(t)\mathbf{A}(t)}) - (1|\hat{\mathbb{G}}(\tau)|P)(1|\overline{O(t)\mathbf{A}(t)}). \quad (51)$$

The previous expressions explicitly show that the departure from the condition equation (47) measures the size of the dynamical effects that can not be captured by assuming valid the QRT. In fact, the QRT is fulfilled only when  $\mathbf{F}(t, \tau)$  vanishes. On the other hand, from (49a) and (49b) it is immediate to understand the meaning of the *validity of the non-Markovian QRT only in the stationary regime*: when the conditions (48) are fulfilled, the propagator of the expectation values and the propagator of the stationary correlations are exactly the same, i.e.,  $\mathbb{T}(\tau)$ , while the inhomogeneous term cancel identically,  $\mathbf{F}(\infty, \tau) = 0$ .

## 4.3 Discussion

We remark that the validity of the regression hypothesis for quantum systems is restricted to *weak system-environment interactions* [61–64]. In the strong coupling regime the generalization of the classical Onsager hypothesis [65] is the fluctuation dissipation theorem [61–63]. These well known results may give the impression that the QRT is only valid when a Markovian approximation applies [66]. Here, in contrast we demonstrated that the QRT might be extended to strong non-Markovian system dynamics. There is not any contradiction in our formalism. This conclusion is evident after noting that non-Markovian Lindblad rate equations *also* arise from weak system-environment interactions. This property guarantees the consistency of our approach with all previous results. On the other hand, from Sect. 3, it is simple to realize that the operator correlations and in consequence their associated evolutions rely in the same calculation steps used to derive the density matrix evolution. Therefore, their range of validity is evidently the same.

### 5 Detailed Balance Condition

In the previous section we have demonstrated that the non-Markovian QRT can be assumed valid in an asymptotic regime if the stationary states  $\rho_R^\infty$  does not depends on the index  $R$  [see (48a)] and the initial weights are stationary, i.e.,  $P_R = P_R^\infty$  [see (48b)]. Here, we demonstrate that these constraints can be associated with a quantum detailed balance condition [11–17], which in turn is related with the microreversibility of the underlying microscopic dynamics [11].

While the classical detailed balance condition has a clear interpretation in terms of the available stationary transitions of a stochastic system [9], for quantum dynamics this condition is defined in terms of the time reversal property [11] of the stationary system-bath dynamics. It can be written as an statement of time-symmetry of the stationary operator correlations [11–15]

$$\lim_{t \rightarrow \infty} \overline{O(t + \tau)\mathbf{A}(t)} = \lim_{t \rightarrow \infty} \widetilde{\overline{\mathbf{A}(t + \tau)\tilde{O}(t)}}, \tag{52}$$

where  $\tilde{O}(t)$  and  $\tilde{\mathbf{A}}(t)$  represent time-reversed operators [67].

From (38), the left hand side term of (52) can be written as

$$\lim_{t \rightarrow \infty} \overline{O(t + \tau)\mathbf{A}(t)} = \text{Tr}_S\{\mathbf{A}(\rho_\infty | e^{\tau \hat{\mathcal{L}}^\#} | O)\}, \tag{53}$$

where  $|\rho_\infty\rangle \equiv \lim_{t \rightarrow \infty} |\rho(t)\rangle$ , and the dual superoperator  $\hat{\mathcal{L}}^\#$  is defined by (36). Furthermore, from (38), the right hand side of (52) follows as

$$\begin{aligned} \lim_{t \rightarrow \infty} \widetilde{\overline{\mathbf{A}(t + \tau)\tilde{O}(t)}} &= \text{Tr}_S\{(\tilde{O} \rho_\infty | e^{\tau \hat{\mathcal{L}}^\#} | \tilde{\mathbf{A}})\}, \\ &= \text{Tr}_S\{(\tilde{\mathbf{A}} | e^{\tau \hat{\mathcal{L}}} | \tilde{O} \rho_\infty)\}, \\ &= \text{Tr}_S\{\tilde{\mathbf{A}}(1 | e^{\tau \hat{\mathcal{L}}} | \widetilde{\rho_\infty O})\}, \end{aligned}$$

where we have used (35) and the property  $\widetilde{AB} = \tilde{B} \tilde{A}$ , valid for arbitrary operators  $A$  and  $B$ . By using the time reverse invariance of the trace operation, we can write

$$\begin{aligned} \lim_{t \rightarrow \infty} \widetilde{\overline{\mathbf{A}(t + \tau)\tilde{O}(t)}} &= \widetilde{\text{Tr}_S\{(\tilde{\mathbf{A}} | e^{\tau \hat{\mathcal{L}}} | \widetilde{\rho_\infty O})\}}, \\ &= \text{Tr}_S\{\mathbf{A}(1 | e^{\tau \hat{\mathcal{L}}} | \widetilde{\rho_\infty O})\}, \\ &= \text{Tr}_S\{\mathbf{A}(1 | e^{\tau \tilde{\hat{\mathcal{L}}}} | \widetilde{\rho_\infty O})\}, \end{aligned} \tag{54}$$

where we have introduced the superoperator  $\tilde{\hat{\mathcal{L}}}$  [11–15] defined by  $\widetilde{\exp[\tau \hat{\mathcal{L}}] | O} = \exp[\tau \tilde{\hat{\mathcal{L}}}] | \tilde{O}$ . Then, from (53) and (54), the microreversibility condition equation (52) implies

$$\text{Tr}_S\{\mathbf{A}[(\rho_\infty | e^{\tau \hat{\mathcal{L}}^\#} | O) - (1 | e^{\tau \tilde{\hat{\mathcal{L}}}} | \widetilde{\rho_\infty O})]\} = 0. \tag{55}$$

As the system operators  $\mathbf{A}$  and  $O$  are arbitrary, it follows

$$(\rho_\infty | e^{\tau \hat{\mathcal{L}}^\#} | \bullet) = (1 | e^{\tau \tilde{\hat{\mathcal{L}}}} | \widetilde{\rho_\infty \bullet}). \tag{56}$$

By demanding the validity of this condition at all times  $\tau$ , one arrive to the equivalent conditions

$$\rho_S^\infty = \tilde{\rho}_S^\infty, \tag{57a}$$

$$(\rho_\infty | \hat{\mathcal{L}}^\# | \bullet) = (1 | \tilde{\hat{\mathcal{L}}} | \tilde{\rho}_\infty^\bullet), \tag{57b}$$

where  $\rho_S^\infty \equiv \lim_{t \rightarrow \infty} \rho_S(t)$ .

The conditions (57) [or equivalently (56)] guaranty the fulfillment of the microreversibility condition equation (52). These constraints are written in terms of the generator  $\hat{\mathcal{L}} = \hat{\mathcal{L}}_H + \hat{\mathcal{M}}$  [(25)] of the time evolution of the Lindblad rate dynamics and in general can not be written in terms of the superoperators that define the non-Markovian density matrix evolution equation (9), i.e., in terms of  $\mathcal{L}_H + \mathcal{K}(u)$ . Nevertheless, when the conditions that guaranty the validity of the “non-Markovian QRT” are valid, from (56) it follows

$$\rho_S^\infty (P_R^\infty | e^{\tau \hat{\mathcal{L}}^\#} | 1) [\bullet] = (1 | e^{\tau \tilde{\hat{\mathcal{L}}}} | P_R^\infty) [\tilde{\rho}_S^\infty \bullet], \tag{58}$$

where we have used that  $|\rho_\infty\rangle = \rho_S^\infty | P_R^\infty \rangle$ , i.e., (48a). Furthermore, by using the stationary condition  $P_R = P_R^\infty$  [see (48b)], we realize that the right and left hand sides of (58) can be expressed in terms of the density matrix and operator propagators respectively, i.e., (8) and (17). Then, by demanding the fulfillment of (58) at all times  $\tau$  we can write

$$\rho_S^\infty = \tilde{\rho}_S^\infty, \tag{59a}$$

$$\rho_S^\infty \{ \mathcal{L}_H^\# + \mathcal{K}^\#(u) \} [\bullet] = \{ \tilde{\mathcal{L}}_H + \tilde{\mathcal{K}}(u) \} [\rho_S^\infty \bullet]. \tag{59b}$$

We notice that a similar equation also arises when formulating the detailed balance condition for non-Markovian classical Fokker-Planck equations [14, 15]. Here,  $\mathcal{K}^\#(u)$  is defined by (19) while  $\tilde{\mathcal{K}}(u)$  follows from (10) after replacing all superoperators by their associated time-reverse expressions.

In contrast to the previous conditions [see (57)], (59) does not depends on the underlying dynamics that lead to the non-Markovian system evolution. In fact, it only depends on the superoperator  $\mathcal{L}_H + \mathcal{K}(u)$  that defines the density matrix evolution, (9). In this way, a general relation between the “non-Markovian QRT” and the non-Markovian quantum detailed balance condition can be established. We conclude that, “a necessary condition for the validity of the non-Markovian QRT in the stationary regime is the fulfillment of the non-Markovian quantum detailed balance conditions (59).” As we will show in the next examples, while the fulfillment of (59) guarantees the validity of the condition equation (48a), it does not guarantees the stationary condition (48b). On the other hand, from the previous derivation, it is simple to realize that the relation between the detailed balance condition and the QRT does not apply to quantum Markovian evolutions.

### 6 Examples

Here, we exemplify our theoretical results by studying a two-level system whose dissipative dynamics can be written in terms of different Lindblad rate evolutions. First, we analyze the case of a diagonal equation [see (2) with  $\{a_{R'R}^{\alpha\gamma}\} = 0$ ] representing the action of a complex thermal reservoir. The influence of an external field on the validity of the QRT is analyzed. Secondly, we study the validity of the QRT in a dispersive non-diagonal Lindblad rate equation.

### 6.1 Diagonal Lindblad Rate Equation

The Hamiltonian of the system is given by

$$H_S = \frac{\hbar\omega_A}{2}\sigma_z + H_{ext}, \tag{60}$$

where  $\hbar\omega_A$  is the difference of energy between the two levels, denoted by  $|\pm\rangle$ , and  $\sigma_z$  is the z-Pauli matrix.  $H_{ext}$  represent an external perturbation. The diagonal Lindblad rate equation reads

$$\frac{d\rho_R(t)}{dt} = \mathcal{L}_H[\rho_R(t)] + \gamma_R \mathcal{L}_{th}[\rho_R(t)] + \frac{\gamma_d}{2} \mathcal{L}_d[\rho_R(t)], \tag{61}$$

with  $\mathcal{L}_H[\bullet] = -(i/\hbar)[H_S, \bullet]$ . We take an arbitrary set  $\{\gamma_R, P_R\}$  of diagonal rates and initial weights. On the other hand, the superoperator  $\mathcal{L}_{th}$  is defined by

$$\begin{aligned} \mathcal{L}_{th}[\bullet] &= \frac{1 + n_{th}}{2}([\sigma, \bullet\sigma^\dagger] + [\sigma\bullet, \sigma^\dagger]) \\ &\quad + \frac{n_{th}}{2}([\sigma^\dagger, \bullet\sigma] + [\sigma^\dagger\bullet, \sigma]). \end{aligned} \tag{62}$$

Here,  $\sigma^\dagger$  and  $\sigma$  are the raising and lowering operators acting on the states  $|\pm\rangle$ . The dimensionless constant  $n_{th}$  defines the temperature  $T$  of the environment as  $\exp[-\hbar\omega_A/kT] = n_{th}/(n_{th} + 1)$ , where  $k$  is the Boltzmann constant. In (61), we have also considered the action of an extra Markovian dispersive environment which is introduced through the Lindblad superoperator

$$\mathcal{L}_d[\bullet] = ([\sigma_z, \bullet\sigma_z] + [\sigma_z\bullet, \sigma_z])/2, \tag{63}$$

and the single rate  $\gamma_d$ . For  $n_{th} = 0$ , (61) recover the dynamics analyzed in Refs. [28–30].

#### 6.1.1 Free Decay Dynamics

First we analyze the case without the external perturbation, i.e.,  $H_{ext} = 0$ .

**6.1.1.1 Non-Markovian Density Matrix Evolution** In an interaction representation with respect to  $\hbar\omega_A\sigma_z/2$ , from (61) the density matrix evolution equation (9) can be written as  $d\rho_S(t)/dt = \int_0^t d\tau \mathcal{K}(t - \tau)[\rho_S(\tau)]$ , with the superoperator

$$\mathcal{K}(u)[\bullet] = \frac{1}{1 + 2n_{th}}k_{\parallel}(u)\mathcal{L}_{th}[\bullet] + \frac{k'_{\perp}(u)}{2}\mathcal{L}_d[\bullet]. \tag{64}$$

The memory kernel functions are defined in the Laplace domain by

$$k_{\parallel}(u) = \left(P \left| \frac{\gamma_{\parallel}}{u + \gamma_{\parallel}} \right. \right) \left(P \left| \frac{1}{u + \gamma_{\parallel}} \right. \right)^{-1}, \tag{65a}$$

$$k_{\perp}(u) = \left(P \left| \frac{\gamma_{\perp}}{u + \gamma_{\perp}} \right. \right) \left(P \left| \frac{1}{u + \gamma_{\perp}} \right. \right)^{-1}, \tag{65b}$$

with  $k'_{\perp}(u) = k_{\perp}(u) - k_{\parallel}(u)/2$ . For shortening the notation, we used the  $R$ -vector notation  $|f(\gamma)\rangle \equiv (f(\gamma_1), f(\gamma_2), \dots, f(\gamma_R), \dots)^T$ , with the rates  $\gamma_{\parallel R} \equiv \gamma_R(1 + 2n_{th})$  and  $\gamma_{\perp R} \equiv \gamma_{\parallel R}/2 + \gamma_d$ .

**6.1.1.2 Quantum Detailed Balance Condition** In order to check the condition (59), we note that the evolution defined by (64) leads to the stationary state,  $\rho_S^\infty = \lim_{t \rightarrow \infty} \rho_S(t)$ ,

$$\rho_S^\infty = \Pi_+^{eq} |+\rangle\langle +| + \Pi_-^{eq} |-\rangle\langle -|, \tag{66}$$

where the stationary populations  $\Pi_+^{eq}$  and  $\Pi_-^{eq}$  are defined by  $\Pi_+^{eq} / \Pi_-^{eq} = n_{th} / (n_{th} + 1)$  and  $\Pi_+^{eq} + \Pi_-^{eq} = 1$ . Due to the time reversal invariance of Hamiltonian eigenvectors this state satisfies  $\tilde{\rho}_S^\infty = \rho_S^\infty$ . Then, it is easy to prove that (59) is satisfied identically.

**6.1.1.3 Quantum Regression Theorem** As the Lindblad rate equation (61) is diagonal (the set of states  $\{\rho_R(t)\}$  are not coupled between them) at all times it is satisfied  $P_R = \text{Tr}_S\{\rho_R(t)\}$ , which implies that the condition (48b) is trivially fulfilled. In this case, the fulfillment of the quantum detailed balance condition (59) guarantees the validity of the QRT in the stationary regime. Consistently, the normalized stationary states of (61) must not depend on  $R$  [condition equation (48a)]. In fact, it is simple to prove that  $\lim_{t \rightarrow \infty} \{\rho_R(t) / \text{Tr}_S[\rho_R(t)]\} = \rho_S^\infty$ .

**6.1.1.4 Expectation Values and Operator Correlations** The density matrix evolution defined by (64) is equivalent to the non-Markovian Bloch equation

$$\frac{dS_X(t)}{dt} = - \int_0^t d\tau k_\perp(t - \tau) S_X(\tau), \tag{67a}$$

$$\frac{dS_Y(t)}{dt} = - \int_0^t d\tau k_\perp(t - \tau) S_Y(\tau), \tag{67b}$$

$$\frac{dS_Z(t)}{dt} = - \int_0^t d\tau k_\parallel(t - \tau) [S_Z(\tau) - S_Z^\infty], \tag{67c}$$

where  $S_j(t) \equiv \text{Tr}_S\{\rho_S(t)\sigma_j\}$  are the expectation values of the Pauli matrices  $\sigma_j$ , and  $S_Z^\infty \equiv \Pi_+^{eq} - \Pi_-^{eq}$ . In order to deal with diagonal matrices, we analyze the operator correlations in the base  $\mathbf{A} = \{\sigma_x, \sigma_y, (\sigma_z - S_Z^\infty), \mathbf{I}\}$ , where  $\mathbf{I}$  is the  $(2 \times 2)$  identity matrix. Then, the propagator for operator expectation values,  $\overline{\mathbf{A}}(t) = \mathbb{T}(t)\overline{\mathbf{A}}(0)$ , can be written as

$$\mathbb{T}(t) = \text{diag}\{h_\perp(t), h_\perp(t), h_\parallel(t), \mathbf{I}\}. \tag{68}$$

Here, we defined the functions  $h_\perp(u) = [u + k_\perp(u)]^{-1}$  and  $h_\parallel(u) = [u + k_\parallel(u)]^{-1}$ , which can be written in the time domain as

$$h_\perp(t) = (P | \exp[-\gamma_\perp t]), \quad h_\parallel(t) = (P | \exp[-\gamma_\parallel t]). \tag{69}$$

On the other hand, the extra inhomogeneous term [see (51)] that defines the operator correlations,  $\overline{O(t)\mathbf{A}(t + \tau)} = \mathbb{T}(\tau)\overline{O(t)\mathbf{A}(t)} + \mathbf{F}(t, \tau)$ , can be written as

$$\mathbf{F}(t, \tau) = \mathbb{T}_\perp(t, \tau)\mathbf{F}_\perp + \mathbb{T}_\parallel(t, \tau)\mathbf{F}_\parallel, \tag{70}$$

where we have defined the vectors

$$\mathbf{F}_\perp = \text{Tr}_S[O\mathbf{A}\rho_S^-(0)], \tag{71a}$$

$$\mathbf{F}_\parallel = \text{Tr}_S[O\mathbf{A}\{\rho_S^+(0) - \rho_S^\infty\}], \tag{71b}$$

with  $\rho_S^\pm(0) \equiv [\rho_S(0) \pm \sigma_z \rho_S(0) \sigma_z]/2$ .<sup>1</sup> We note that  $\mathbf{F}_\parallel$  measure the departure of the initial populations from the equilibrium values  $\Pi_\pm^{eq}$ , while  $\mathbf{F}_\perp$  measure the departure of the initial system coherences (in the base  $|\pm\rangle$ ) from their null stationary value. On the other hand, the time dependence of  $\mathbf{F}(t, \tau)$  is defined by the matrices

$$\mathbb{T}_\perp(t, \tau) = \text{diag}\{f_\perp(t, \tau), f_\perp(t, \tau), f_0(t, \tau), 0\}, \tag{72a}$$

$$\mathbb{T}_\parallel(t, \tau) = \text{diag}\{f_0(t, \tau), f_0(t, \tau), f_\parallel(t, \tau), 0\}, \tag{72b}$$

with the definitions

$$f_\perp(t, \tau) = h_\perp(t + \tau) - h_\perp(t)h_\perp(\tau), \tag{73a}$$

$$f_\parallel(t, \tau) = h_\parallel(t + \tau) - h_\parallel(t)h_\parallel(\tau), \tag{73b}$$

$$f_0(t, \tau) = e^{-\tau\gamma_d} f_\parallel(t, \tau/2). \tag{73c}$$

These functions measure the transient departure from the validity of the QRT. Only when the decay behaviors are exponential, they vanish identically and the QRT is valid at all times. This situation only happens when the evolution is Markovian. On the other hand, with the previous definitions, it is easy to check that (70) satisfies  $\lim_{t \rightarrow \infty} \mathbf{F}(t, \tau) = 0$ , i.e., in the non-Markovian case the QRT is only valid for calculating stationary correlations.

**6.1.1.5 Transient Decay Behaviors** In order to illustrate the previous results, we specify the properties of the diagonal rates  $\gamma_{\parallel R}$  as well as the weights  $P_R$ . We choose

$$\gamma_{\parallel R} = \gamma_0 \exp[-bR], \quad P_R = \frac{(1 - e^{-a})}{(1 - e^{-aN})} \exp[-aR], \tag{74}$$

where  $R \in [0, N - 1]$ ,  $\gamma_0$  scale the rates, and the dimensionless constants  $b$  and  $a$  measure the exponential decay of the diagonal rates and their associated weights. The relevant parameters of this set are

$$\gamma_p \equiv (P|\gamma_\parallel), \quad \beta_p \equiv \frac{(P|\gamma_\parallel^2) - (P|\gamma_\parallel)^2}{(P|\gamma_\parallel)}, \quad \alpha \equiv \frac{a}{b}. \tag{75}$$

Here,  $\gamma_p$  can be interpreted as an average of the rates  $\{\gamma_{\parallel R}\}$  over the set of probabilities  $\{P_R\}$ . In fact,  $\gamma_p = \sum_R P_R \gamma_{\parallel R}$ . Then, the rate  $\beta_p$  correspond to the ‘‘normalized dispersion’’ of the set  $\{\gamma_{\parallel R}\}$ . On the other hand, we remark that the rates (74) lead to transient system decay behaviors characterized by power law dependences (with exponent  $\alpha$ ) [53] allowing to fit specific experimental situations [28–30].

In Fig. 1 we plot the transient decay behavior of the correlation

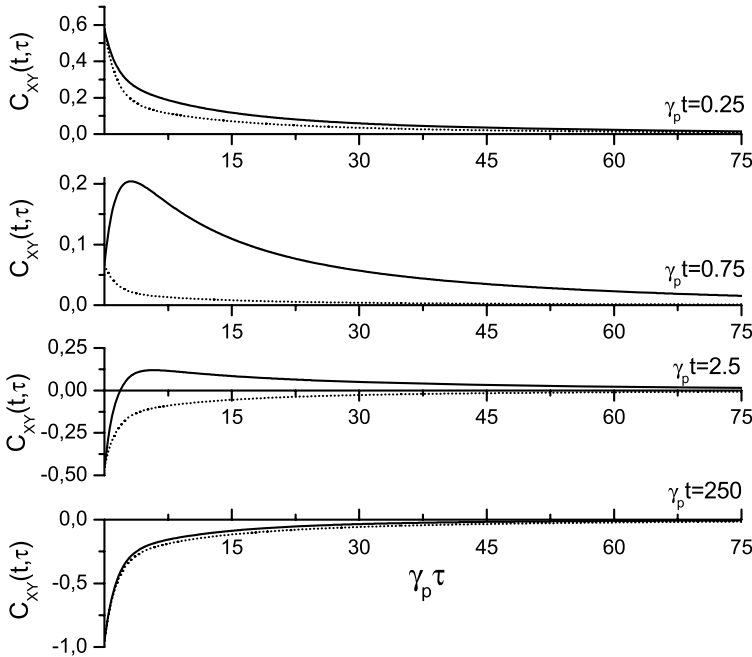
$$C_{XY}(t, \tau) \equiv \overline{\sigma_x(t)\sigma_y(t + \tau)} / i, \tag{76}$$

which from (68) and (70) can be written as

$$C_{XY}(t, \tau) = \{h_\perp(\tau)S_Z(t) + f_0(t, \tau)[S_Z(0) - S_Z^\infty]\}, \tag{77}$$

<sup>1</sup>The density matrix dynamics corresponding to the evolution generated by (64) can be written as  $\rho_S(t) = h_\parallel(t)\rho_S^+(0) + h_\perp(t)\rho_S^-(0) + [1 - h_\parallel(t)]\rho_S^\infty$ . This expression is used when deriving (70).





**Fig. 1** Transient decay behavior of  $C_{XY}(t, \tau)$ , (77). The parameters of the environment are  $b = 2.15$ ,  $a = ab$ ,  $\alpha = 1/2$ ,  $N = 5$ ,  $n_{th} = 0$ , and  $\gamma_d/\gamma_p = 0.02$ . The dispersion rate results  $\beta_p/\gamma_p = 0.4$ . The *dotted lines* correspond to the QRT. From top to bottom, we set  $\gamma_p t = 0.25, 0.75, 2.5$ , and  $250$

with  $S_Z(t) = S_Z^\infty + h_{\parallel}(t)[S_Z(0) - S_Z^\infty]$ . We have chosen a zero temperature reservoir,  $n_{th} = 0$ , characterized by (74). As initial condition for the system, we take the pure state  $|+\rangle$ . Thus,  $S_Z^\infty = -1$  and  $S_Z(0) = 1$ . Notice that the initial value of each plot describe the decay of the initial condition from the upper to the lower state. In fact  $C_{XY}(t, 0) = S_Z(t)$ .

We also plotted the correlation behavior that follows by assuming valid the QRT, i.e., (49b) with  $\mathbf{F}(t, \tau) = 0$ . As can be seen from the graphics, the predictions of the QRT are asymptotically valid in the stationary regime, where the function  $f_0(t, \tau)$  vanish identically. In fact, the correlation behavior predicted by the QRT follows from (77) after replacing  $f_0(t, \tau) \rightarrow 0$ .

The transient deviations from the QRT are proportional to the departure of the system decay behavior from an exponential one. This departure arises from the competence between the exponential decay introduced by the rate  $\gamma_d$  and the non-Markovian effects induced by the random rate dispersion  $\beta_p$ . From (77) it is evident that the dispersive rate  $\gamma_d$  introduces a global exponential decay. Thus, in general, by increasing this rate, the transient deviation from the QRT are diminished. On the other hand, an increasing of  $\beta_p$  implies a strong deviation from an exponential decay.

### 6.1.2 Decay under the Action of an External Field

We consider the external Hamiltonian  $H_{ext} = (\hbar\Omega/2)(\sigma^\dagger e^{-i\omega_A t} + \sigma e^{+i\omega_A t})$ . Then, the Hamiltonian dynamics can be associated with a spin subject to a resonant external magnetic field [3] or with a two-level optical transition driven by a resonant laser field [4]. We

notice that in an interaction representation with respect to  $\hbar\omega_A\sigma_z/2$ , the system Hamiltonian does not depend on time,  $H_S \rightarrow \hbar\Omega\sigma_x/2$ . In this case, the expectation values of the Pauli matrices evolve as

$$\frac{dS_X(t)}{dt} = - \int_0^t d\tau \Gamma_X(t - \tau) S_X(\tau), \tag{78a}$$

$$\begin{aligned} \frac{dS_Y(t)}{dt} = & -\Omega S_Z(t) - \int_0^t d\tau \{ \Gamma_Y(t - \tau) S_Y(\tau) \\ & + \Upsilon(t - \tau) [S_Z(\tau) - S_Z^\infty] \}, \end{aligned} \tag{78b}$$

$$\begin{aligned} \frac{dS_Z(t)}{dt} = & \Omega S_Y(t) + \int_0^t d\tau \{ \Upsilon(t - \tau) S_Y(\tau) \\ & - \Gamma_Z(t - \tau) [S_Z(\tau) - S_Z^\infty] \}. \end{aligned} \tag{78c}$$

In Appendix B we give the exact expressions for the kernels  $\Gamma_j(t)$ ,  $j = x, y, z$ , and  $\Upsilon(t)$ , as well as the expression for the non local superoperator  $\mathcal{K}(u)$ , (10). We remark that independently of the set of rates  $\{\gamma_{\parallel R}\}$  and weights  $\{P_R\}$ , the kernels that define the evolution equation (78) depend explicitly on the parameter  $\Omega$ .

The normalized stationary state [see (48a)] corresponding to the evolution of each state  $\rho_R(t)$ , equation (61) with  $\mathcal{L}_H[\bullet] \rightarrow \hbar\Omega[\sigma_x, \bullet]/2$ , reads

$$\rho_R^\infty = \frac{1}{2} \left\{ \mathbf{I} + \frac{\gamma_{\parallel R} [\Omega\sigma_y - \gamma_{\perp R}\sigma_z]}{(1 + 2n_{th})[\gamma_{\parallel R}\gamma_{\perp R} + \Omega^2]} \right\}. \tag{79}$$

This expression explicitly depends on  $R$  if  $\Omega \neq 0$ . Then, even when the condition (48b) is satisfied, when the system is subject to the action of the external field the QRT is not fulfilled, even in the asymptotic regime. Consistently, the superoperator  $\mathcal{K}(u)$  [see (B.1)] does not satisfy (59). As the QRT is not fulfilled, the operators correlations must be calculated from the underlying microscopic Hamiltonian dynamics, i.e., from the expressions obtained in Sect. 3.

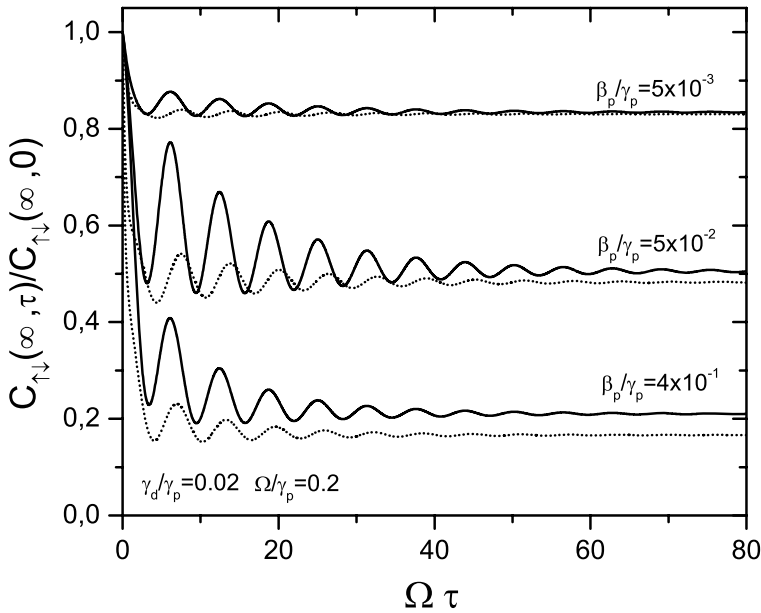
In the next figures we characterize the correlation

$$\begin{aligned} C_{\uparrow\downarrow}(t, \tau) & \equiv \overline{\sigma^\dagger(t)\sigma(t + \tau)}, \\ & = \{C_{xx}(t, \tau) + C_{yy}(t, \tau)\}/4 \\ & \quad - i\{C_{xy}(t, \tau) - C_{yx}(t, \tau)\}/4 \end{aligned} \tag{80}$$

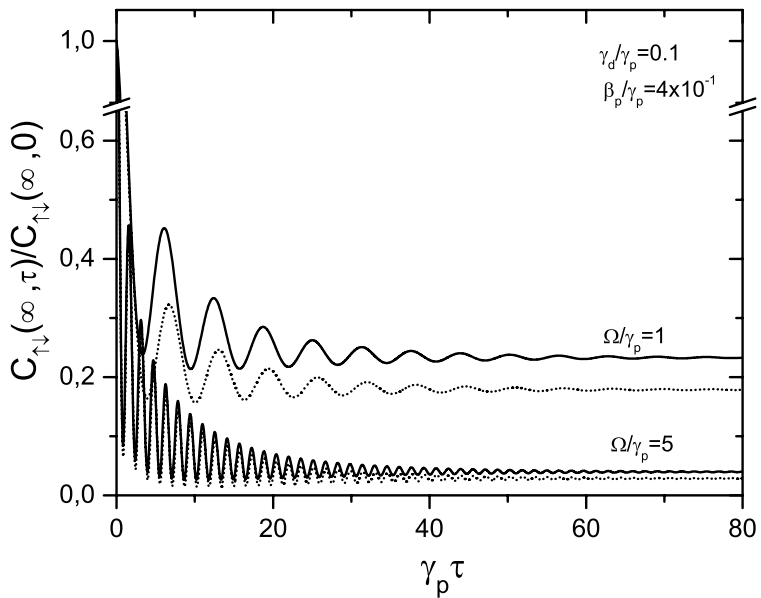
where  $C_{jk}(t, \tau) \equiv \overline{\sigma_j(t)\sigma_k(t + \tau)}$  are the correlations of the Pauli matrices. Each contribution  $C_{jk}(t, \tau)$  can be determine from (42) or equivalently (49b). On the other hand, if one assume the validity of the QRT in the stationary regime, the correlations are defined by (49b) with  $\mathbf{F}(t, \tau) \rightarrow 0$ .

In Fig. 2 we plot the stationary decay  $C_{\uparrow\downarrow}(\infty, \tau)/C_{\uparrow\downarrow}(\infty, 0)$ , where  $C_{\uparrow\downarrow}(\infty, 0) = [1 + S_Z(\infty)]/2$ , for different values of the rate  $\beta_p$ . We notice that both the decay behaviors and the stationary values differ from the QRT predictions. As can be seen in the graphic, the difference in the correlation behavior grows by increasing the dispersion rate  $\beta_p$ .

In Fig. 3 we plot  $C_{\uparrow\downarrow}(\infty, \tau)$  for different values of the field intensity  $\Omega$ . The deviations with respect to the QRT are diminished by increasing  $\Omega$ . Even more, in the limit of high intensity, the dynamical deviations vanish.



**Fig. 2** Stationary decay behavior of  $C_{\uparrow\downarrow}(t, \tau)$ . From top to bottom, the parameters of the complex environment are  $b = 10.6, 6.05,$  and  $2.15$ . In all cases we take  $a = \alpha b, \alpha = 1/2, N = 5, n_{th} = 0,$  and  $\gamma_d/\gamma_p = 0.02$ . The intensity is  $\Omega/\gamma_p = 0.2$ . The *dotted lines* correspond to the QRT



**Fig. 3** Stationary decay behavior of  $C_{\uparrow\downarrow}(t, \tau)$  for different values of the intensity parameter. From top to bottom, we take  $\Omega/\gamma_p = 1$  and  $5$ . In both cases the parameters of the complex environment are  $b = 2.15, a = \alpha b, \alpha = 1/2, N = 5, n_{th} = 0,$  and  $\gamma_d/\gamma_p = 0.1$ . The *dotted lines* correspond to the decay predicted by the QRT

The characteristic behaviors shown in the two previous figures can be analytically characterized by analyzing the asymptotic value of the correlation (80), i.e., from  $C_{\uparrow\downarrow}(\infty, \infty) = \lim_{\tau \rightarrow \infty} C_{\uparrow\downarrow}(t, \tau)$ . It can be written as

$$C_{\uparrow\downarrow}(\infty, \infty) = \frac{1}{(1 + 2n_{th})^2} \left( P \left| \left[ \frac{\Omega \gamma_{\parallel} / 2}{\gamma_{\parallel} \gamma_{\perp} + \Omega^2} \right]^2 \right. \right), \tag{81}$$

while by assuming valid the QRT we get

$$C_{\uparrow\downarrow}^{QRT}(\infty, \infty) = \frac{1}{(1 + 2n_{th})^2} \left( P \left| \frac{\Omega \gamma_{\parallel} / 2}{\gamma_{\parallel} \gamma_{\perp} + \Omega^2} \right. \right)^2. \tag{82}$$

Thus, as a measure of the departure from the validity of the QRT *in the stationary regime*, we introduce the quantity  $\mathcal{E} \equiv C_{\uparrow\downarrow}(\infty, \infty) - C_{\uparrow\downarrow}^{QRT}(\infty, \infty)$ . A general characterization of this object can be given in a small and high intensity limits. First, in the *weak intensity regime*,  $\Omega \ll \{\gamma_{\parallel R}\}$ , we can approximate

$$\mathcal{E} \approx \frac{\Omega^2 / 4}{(1 + 2n_{th})^2} [(P|\gamma_{\perp}^{-2}) - (P|\gamma_{\perp}^{-1})^2] + O(\Omega^3), \tag{83}$$

where  $(\gamma_{\perp}^{-1})_R = (\gamma_{\parallel R} / 2 + \gamma_d)^{-1}$ . Consistently,  $\mathcal{E}$  goes to zero in the limit of small intensity  $\Omega$ . On the other hand, the term in brackets is proportional to the dispersion of the rate  $\gamma_{\perp}^{-1}$ , which in turn is proportional to the dispersion rate  $\beta_p$ , (75), and ‘‘higher moments.’’ Furthermore, by increasing the dispersive rate  $\gamma_d$ , each contribution in (83) diminish, which in turn means that the predictions of the QRT approach the exact dynamics.

In the *high intensity regime*,  $\Omega \gg \{\gamma_{\parallel R}\}$  we get

$$\mathcal{E} \approx \frac{\Omega^{-2} / 4}{(1 + 2n_{th})^2} [(P|\gamma_{\parallel}^2) - (P|\gamma_{\parallel})^2] + O(\Omega^{-3}). \tag{84}$$

In this limit,  $\mathcal{E}$  is also proportional to the dispersion rate  $\beta_p$  (bracket contribution). On the other hand, this expression implies that by increasing  $\Omega$ , the validity of the QRT is asymptotically recuperated. This result is consistent with the fact that at high intensity values [12] the stationary states  $\rho_R^{\infty}$  can be approximated by  $\rho_R^{\infty} \approx I/2$ , which as expected do not depend on index  $R$ . Furthermore, in this limit the microreversibility condition equation (59) is also recovered.

### 6.2 Non-Diagonal Lindblad Rate Equation

Here, we analyze the case of a non-diagonal Lindblad rate equation that arise from an underlying composite bipartite reservoir. For simplicity, we assume that the extra degrees of freedom  $U$  are characterized by a bidimensional Hilbert space defined by the states  $|a\rangle$  and  $|b\rangle$  ( $R = a, b$ ), with  $H_U = E_a |a\rangle\langle a| + E_b |b\rangle\langle b|$ . The two-level system  $S$  has eigenvectors  $|\pm\rangle$ , with  $H_S = \hbar \omega_A \sigma_z / 2$ , where  $\sigma_z$  is the z-Pauli matrix defined in the base  $|\pm\rangle$ . In an interaction with respect to  $H_S + H_U$ , the evolution of the joint density matrix  $\rho_{SU}(t)$  [see (24)] reads

$$\frac{d\rho_{SU}(t)}{dt} = L_{SU}[\rho_{SU}(t)], \tag{85}$$

where the superoperator  $L_{SU}$  is defined as

$$L_{SU} = \gamma_a L_a + \gamma_b L_b + \gamma_{ab} L_{ab} + \gamma_{ba} L_{ba}. \tag{86}$$

The set  $\{\gamma_a, \gamma_b, \gamma_{ab}, \gamma_{ba}\}$  are characteristic rates and each Lindblad contribution is defined by

$$L_i[\bullet] = \frac{1}{2}([V_i, \bullet V_i^\dagger] + [V_i \bullet, V_i^\dagger]), \tag{87}$$

with the operators

$$V_a = \sigma_z \otimes |a\rangle \langle a|, \quad V_{ab} = \sigma_z \otimes |a\rangle \langle b|, \tag{88a}$$

$$V_b = \sigma_z \otimes |b\rangle \langle b|, \quad V_{ba} = \sigma_z \otimes |b\rangle \langle a|. \tag{88b}$$

From (85) it is possible to prove that the populations and coherences of  $U$  does not couples between them. Consistently, the system density matrix evolution can be written as a Lindblad rate equation

$$\begin{aligned} \frac{d}{dt} \rho_a(t) = & -\gamma_a[\rho_a(t) - \sigma_z \rho_a(t) \sigma_z] \\ & - \gamma_{ba} \rho_a(t) + \gamma_{ab} \sigma_z \rho_b(t) \sigma_z, \end{aligned} \tag{89a}$$

$$\begin{aligned} \frac{d}{dt} \rho_b(t) = & -\gamma_b[\rho_b(t) - \sigma_z \rho_b(t) \sigma_z] \\ & - \gamma_{ab} \rho_b(t) + \gamma_{ba} \sigma_z \rho_a(t) \sigma_z \end{aligned} \tag{89b}$$

with initial condition  $\rho_a(0) = P_a \rho_S(0)$  and  $\rho_b(0) = P_b \rho_S(0)$ . The initial weights are defined by  $P_a = \langle a | \rho_U(0) | a \rangle$ , and  $P_b = \langle b | \rho_U(0) | b \rangle$ , satisfying  $P_a + P_b = 1$ . Equations (89) lead to the non-Markovian evolution  $d\rho_S(t)/dt = \int_0^t d\tau \mathcal{K}(t - \tau)[\rho_S(\tau)]$  with the superoperator

$$\mathcal{K}[\bullet] = k(u)(-\bullet + \sigma_z \bullet \sigma_z), \tag{90}$$

defined by  $h(u) = 1/[u + k(u)]$ , where

$$h(u) = \frac{u + P_a(\gamma_b + \gamma_{ab} - \gamma_{ba}) + P_b(\gamma_a + \gamma_{ba} - \gamma_{ab})}{\gamma_{ab}(u + \gamma_b) + \gamma_{ba}(u + \gamma_a) + (u + \gamma_a)(u + \gamma_b)}. \tag{91}$$

The stationary state associated to (90) reads

$$\rho_S^\infty = \begin{pmatrix} \langle + | \rho_S(0) | + \rangle & 0 \\ 0 & \langle - | \rho_S(0) | - \rangle \end{pmatrix}. \tag{92}$$

On the other hand, the kernel superoperator satisfies  $\mathcal{K} = \mathcal{K}^\# = \tilde{\mathcal{K}}$ . Then, it is easy to demonstrate that the conditions that guarantee microreversibility, (59), are satisfied. Nevertheless, even in the stationary regime here the QRT is in general not valid.

In order to check the validity of the QRT, (48), first we calculate the stationary states  $\rho_R^\infty \equiv \lim_{t \rightarrow \infty} \rho_R(t) / \text{Tr}_S[\rho_R(t)]$ . We get  $\rho_a^\infty = \rho_b^\infty = \rho_S^\infty$ . Therefore, the condition equation (48a) is satisfied. In order to check the stationary condition equation (48b), we obtain the evolution of  $P_R(t) = \text{Tr}_S[\rho_R(t)]$ . These objects define the populations of  $U$ . In fact,  $P_R(t) = \langle R | \rho_U(t) | R \rangle$ , where  $\rho_U(t) = \text{Tr}_S[\rho_{SU}(t)]$ . From (89) we get

$$\frac{d}{dt} P_a(t) = -\gamma_{ba} P_a(t) + \gamma_{ab} P_b(t), \tag{93a}$$

$$\frac{d}{dt} P_b(t) = -\gamma_{ab} P_b(t) + \gamma_{ba} P_a(t), \tag{93b}$$

where the initial conditions read  $P_R(0) = P_R$ . The stationary values of these classical rate equations read

$$P_a^\infty = \frac{\gamma_{ab}}{\gamma_{ab} + \gamma_{ba}}, \quad P_b^\infty = \frac{\gamma_{ba}}{\gamma_{ab} + \gamma_{ba}}. \quad (94)$$

Therefore, the QRT is valid in the stationary regime only when at the initial time the system  $U$  is in its stationary state, i.e.,  $P_a = P_a^\infty$  and  $P_b = P_b^\infty$ .

In order to check the previous result, we calculate the expectation values and stationary correlators of the vector of Pauli operators  $\mathbf{A} \equiv \{\sigma_x, \sigma_y, \sigma_z, I\}$ . We get

$$\overline{\mathbf{A}(t)} = \mathbb{T}(t)\overline{\mathbf{A}(0)}, \quad (95a)$$

$$\lim_{t \rightarrow \infty} \overline{O(t)\mathbf{A}(t + \tau)} = \mathbb{T}_\infty(\tau) \lim_{t \rightarrow \infty} \overline{O(t)\mathbf{A}(t)}, \quad (95b)$$

where the matrix propagators read

$$\mathbb{T}(t) = \text{diag}\{h(t), h(t), 1, 1\}, \quad (96a)$$

$$\mathbb{T}_\infty(t) = \text{diag}\{h_\infty(t), h_\infty(t), 1, 1\}. \quad (96b)$$

Here,  $h_\infty(t)$  is defined as  $h(t)$  in (91) with the replacements  $P_a \rightarrow P_a^\infty$  and  $P_b \rightarrow P_b^\infty$ . Consistently, when  $P_a = P_a^\infty$  and  $P_b = P_b^\infty$ , i.e., when at the initial time the total reservoir begin in its stationary state, the QRT is valid in the stationary regime. On the other hand, if the system Hamiltonian include a ‘‘hopping contribution,’’  $H_S \rightarrow (\hbar/2)[\omega_A\sigma_z + \Delta\sigma_x]$ , the microreversibility condition equation (59) is broken and then, independently of the initial weights, the QRT is invalidated at all times.

## 7 Summary and Conclusions

In this paper, we have characterized the operator correlations associated to open quantum systems whose dissipative evolution can be described through a Lindblad rate equation. Independently of the underlying microscopic interaction, the final expressions, (33) and (34), are written in terms of the propagator associated to the Lindblad rate evolution. The correlators adopt a simple form, (37) and (38), when written in terms of dual superoperators, resembling the expressions corresponding to a standard Markovian environment.

It is usually understood that the quantum regression hypothesis is only valid for Markovian dynamics. In contrast, here by using the previous results, we explored the possibility of establishing a non-Markovian QRT. We have found that operator correlations may evolve as the system expectation values only in a stationary regime. The equality of both decay dynamics is valid when two conditions are satisfied. First, the normalized stationary states must be all the same, (48a), and secondly the initial weight of each auxiliary state must correspond to its stationary value, (48b). When any of these conditions is not valid, the QRT is not fulfilled at any time.

We have also analyzed the conditions that guarantee the fulfillment of a detailed balance condition, which for open quantum systems can be written as a symmetry property of the stationary operators correlations, (52). While the fulfillment of this condition can in general be expressed as a symmetry property of the Lindblad rate evolution, (57), from our analysis we deduced that when the conditions that guarantee the validity of the (non-Markovian) QRT in the stationary regime are valid, the microreversibility condition can be written in terms

of the non-local superoperator that defines the system density matrix evolution, (59). As the final constraint is independent of the underlying microscopic interaction, we concluded that the microreversibility symmetry equation (59) provides a necessary condition for the validity of the QRT (in the stationary regime) in non-Markovian (time-convolution) Lindblad-like evolutions [39–51]. In contrast, for Markovian Lindblad evolutions, independently of the fulfillment of the detailed balance condition, the (Markovian) QRT is always valid in both the transient and stationary time regimes.

We exemplified our finding by analyzing the operator correlations of a two-level system whose decay can be written in terms of different Lindblad rate equations. In contrast with Markovian dynamics, we have showed that the validity of the (non-Markovian) QRT in the stationary regime strongly depends on the system unitary evolution. In general, the departure from the predictions of the QRT not only implies differences in the correlation decay behaviors, but also in their asymptotic values.

The present results provide a step forward in the understanding of open quantum systems dynamics. In fact, our results provide general criteria for characterizing operator correlations associated to a broad class of non-Markovian quantum master equations.

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## Appendix A: Generalized Born-Markov Approximation

Lindblad rate equations also arise when a generalized Born-Markov approximation applies [52, 53]. In this situation an open system  $S$  interacts with a complex structured reservoir  $B$  whose total microscopic dynamics is defined by the Hamiltonian

$$H_T = H_S + H_B + H_I. \quad (\text{A.1})$$

Here,  $H_S$  and  $H_B$  correspond to the system and bath Hamiltonians respectively. The contribution  $H_I$  describes their mutual interaction. The system density matrix follows after eliminating the environment degrees of freedom,  $\rho_S(t) = \text{Tr}_B\{\rho_T(t)\}$ , where the total density matrix  $\rho_T(t)$  evolves as

$$\frac{d\rho_T(t)}{dt} = \frac{-i}{\hbar}[H_T, \rho_T(t)] \equiv L_T[\rho_T(t)]. \quad (\text{A.2})$$

The generalized Born-Markov approximation applies to complex structured environments whose action over the system can be well approximated by a “direct sum” of sub-reservoirs, each one being able to induce by itself a Markovian system dynamics. This assumption applies, for example, in reservoirs characterized by a band structure of eigenvalues [30]. The direct sum structure, in contrast with the standard Born-Markov approximation, allows to write the total density matrix as a mixed state

$$\rho_T(t) = \sum_R \rho_R(t) \otimes \frac{\mathcal{E}_R}{\text{Tr}_B\{\mathcal{E}_R\}}, \quad (\text{A.3})$$

where  $\mathcal{E}_R$  is given by

$$\mathcal{E}_R \equiv \Pi_R \rho_B \Pi_R, \quad (\text{A.4})$$

with  $\rho_B$  being the stationary state of the total bath, while the system states  $\rho_R(t)$  are defined by

$$\rho_R(t) \equiv \text{Tr}_B\{\Pi_R \rho_T(t) \Pi_R\}. \tag{A.5}$$

Here, we have introduced a set of projectors  $\Pi_R$  which provides an orthogonal decomposition of the unit operator  $[I_B]$  in the Hilbert space of the bath,  $\sum_R \Pi_R = I_B$ , with  $\Pi_R \Pi_{R'} = \Pi_R \delta_{R,R'}$ . Each projector define the Hilbert space associated to each Markovian sub-environment. Therefore, they are diagonal in the eigenbasis of  $\rho_B$ , which implies  $\sum_R \mathcal{E}_R = \rho_B$ .

From (A.3), the system density matrix follows as

$$\rho_S(t) = \text{Tr}_B\{\rho_T(t)\} = \sum_R \rho_R(t). \tag{A.6}$$

The evolution of each state  $\rho_R(t)$ , up to second order in the system-environment interaction strength, from (A.2) can be written as

$$\frac{d\rho_R(t)}{dt} \simeq \int_0^\infty dt' \text{Tr}_B\{L_T(t)L_T(t')\rho_T(t)\}, \tag{A.7a}$$

$$= \sum_{R'} \hat{\mathcal{L}}_{RR'} \rho_{R'}(t) \tag{A.7b}$$

where  $\rho_T(t)$  is defined by (A.3) and  $L_T(t)$  is the total Liouville superoperator in an interaction representation with respect to  $H_S + H_B$ . As demonstrated in Ref. [52, 53], the matrix elements of the superoperator  $\hat{\mathcal{L}}$  define a Lindblad rate equation, i.e.,  $\hat{\mathcal{L}} = \hat{\mathcal{L}}_H + \hat{\mathcal{M}}$ , where  $\hat{\mathcal{L}}_H$  and  $\hat{\mathcal{M}}$  are defined by (6) and (7) respectively.

For an uncorrelated initial state,  $\rho_T(0) = \rho_S(0) \otimes \rho_B$ , the auxiliary states satisfies the initial condition  $\rho_R(0) = P_R \rho_S(0)$ , where the statistical weights are defined by

$$P_R = \text{Tr}_B\{\mathcal{E}_R\} = \text{Tr}_B\{\Pi_R \rho_B\} \tag{A.8}$$

which consistently satisfy  $\sum_R P_R = 1$ .

### A.1 Operators correlations

As in Sect. 3, the microscopic derivation of the system operator correlations can also be done in the context of the generalized Born-Markov approximation. First, the operator expectation values read

$$\overline{\mathbf{A}(t)} = \text{Tr}_{SB}[\mathbf{A}(t)\rho_T(0)], \tag{A.9}$$

while the correlation functions follows from

$$\overline{O(t)\mathbf{A}(t+\tau)} \equiv \text{Tr}_{SB}[O(t)\mathbf{A}(t+\tau)\rho_T(0)]. \tag{A.10}$$

Both expressions only involve a trace over the complex environment  $B$  and the system  $S$ . Here, the time dependence of the operators refers to a Heisenberg representation with respect to the total Hamiltonian equation (A.1).

As before, we can trivially write the expectation values as an average over the solutions corresponding to each state  $\rho_R(t)$ ,



$$\overline{\mathbf{A}(t)} = \sum_R \text{Tr}_S[\mathbf{A}\rho_R(t)] \equiv \sum_R \overline{\mathbf{A}(t)}_R, \tag{A.11}$$

$$= (1|\overline{\mathbf{A}(t)}). \tag{A.12}$$

Furthermore, by expressing the initial system-reservoir state  $\rho_T(0)$  in terms of  $\rho_T(t)$ , it is possible to write the correlations as

$$\overline{O(t)\mathbf{A}(t+\tau)} = \text{Tr}_S\{\mathbf{A} \text{Tr}_B[O_{SB}(\tau)]\}, \tag{A.13}$$

where the operator  $O_{SB}(\tau)$  satisfies

$$\frac{d}{d\tau} O_{SB}(\tau) = -\frac{i}{\hbar}[H_T, O_{SB}(\tau)], \tag{A.14}$$

with  $O_{SB}(\tau)|_{\tau=0} = \rho_T(t)O(0)$ . This system-bath operator evolves as the total density matrix, (A.2). On the other hand, (A.3) allows us to write the initial condition as  $O_{SB}(\tau)|_{\tau=0} \approx \sum_R[\rho_R(t)O(0)] \otimes \mathcal{E}_R/\text{Tr}_B\{\mathcal{E}_R\}$ . Therefore, the reduced dynamics of  $O_{SB}(\tau)$  can also be described through a Lindblad rate equation, delivering

$$\text{Tr}_B[O_{SB}(\tau)] = \sum_{RR'}(e^{\tau\hat{\mathcal{L}}})_{RR'}\rho_{R'}(t)O(0), \tag{A.15}$$

$$= (1|e^{\tau\hat{\mathcal{L}}}|\rho(t)O(0)). \tag{A.16}$$

By introducing this expression in (A.13) one recover (33). Therefore, independently of the underlying microscopic interaction that lead to the Lindblad rate equation, the operator correlations assume the same structure when writing in term of the evolution generator  $\hat{\mathcal{L}}$ . The same property is valid for higher operator correlations, (34).

### Appendix B: Non-Markovian System Density Matrix Evolution

Here, we characterize the density matrix evolution corresponding to the example developed in Sect. 6.1 with  $\Omega \neq 0$ . The superoperator  $\mathcal{K}(u)$  [see (10)] associated to the evolution equation (61) can be written as a non-diagonal non-local Lindblad-like superoperator

$$\mathcal{K}(u)[\bullet] = \mathcal{L}_H(u)[\bullet] + \frac{1}{2} \sum_{\alpha\beta} a_{\alpha\beta}(u)([V_\alpha, \bullet V_\beta^\dagger] + [V_\alpha \bullet, V_\beta^\dagger]), \tag{B.1}$$

with the operators  $\{V_\alpha\}_{\alpha=1,2,3} = \{\sigma, \sigma^\dagger, \sigma_z\}$ . The Hamiltonian contribution reads

$$\mathcal{L}_H(u)[\bullet] = -i\frac{\Upsilon(u)}{2}[\sigma_x, \bullet], \tag{B.2}$$

and the matrix elements  $a_{\alpha\beta}(u)$  are defined by

$$a_{11}(u) = \Pi_-^{eq} \Gamma_Z(u), \tag{B.3a}$$

$$a_{22}(u) = \Pi_+^{eq} \Gamma_Z(u), \tag{B.3b}$$

$$a_{33}(u) = \frac{1}{4}\{\Gamma_X(u) + \Gamma_Y(u) - \Gamma_Z(u)\}, \tag{B.3c}$$

$$a_{12}(u) = a_{21}(u) = -\frac{1}{2}\{\Gamma_X(u) - \Gamma_Y(u)\}, \quad (\text{B.3d})$$

$$a_{13}(u) = a_{23}(u) = -i\frac{\Upsilon(u)}{4(1 + 2n_{th})}, \quad (\text{B.3e})$$

$$a_{31}(u) = a_{32}(u) = i\frac{\Upsilon(u)}{4(1 + 2n_{th})}. \quad (\text{B.3f})$$

Without the external excitation,  $\Omega = 0$ , the superoperator  $\mathcal{K}(u)$  reduce to (64). The stationary state reads

$$\rho_S^\infty = \frac{1}{2} \left\{ \mathbb{I} + \frac{\Omega \Gamma_Z \sigma_y - [\Gamma_Y \Gamma_Z + \Upsilon(\Upsilon + \Omega)] \sigma_z}{(1 + 2n_{th})[\Gamma_Y \Gamma_Z + (\Upsilon + \Omega)^2]} \right\}, \quad (\text{B.4})$$

with the notation  $\Gamma_J \equiv \Gamma_J(u)|_{u=0}$ . Consistently, after some algebra, it is possible to write this state as an addition of the corresponding Markovian stationary states, i.e.,  $\rho_S^\infty = (P|\rho^\infty)$ , where  $\rho_R^\infty$  is defined by (79).

The matrix  $a_{\alpha\beta}(u)$  and the Hamiltonian contribution  $\mathcal{L}_H(u)$  are defined in terms of the kernels that define the evolution equation (78). For arbitrary set  $\{\gamma_{\parallel R}, P_R\}$ , they can be written as

$$\Gamma_X(u) = k_\perp(u), \quad (\text{B.5a})$$

$$\Gamma_Y(u) = D \left\{ (u + C) \left[ \frac{B}{2} + (u + \gamma_d) \right] + \Omega^2 \right\} + \gamma_d, \quad (\text{B.5b})$$

$$\Gamma_Z(u) = 2D \left\{ (u + B) \left[ \frac{C}{2} + (u + \gamma_d) \right] + \Omega^2 \right\}, \quad (\text{B.5c})$$

$$\Upsilon(u) = D(C - B)\Omega, \quad (\text{B.5d})$$

where  $D$  denotes the function

$$D(u) = \frac{B(u)/2}{[u + B(u)][u + B(u)/2 + \gamma_d] + \Omega^2}. \quad (\text{B.6})$$

The extra function  $B$  and  $C$  are defined by

$$B(u) = \frac{(P|T(u)\gamma_\parallel)}{(P|T(u))}, \quad C(u) = \frac{(P|T(u)\gamma_\parallel^2)}{(P|T(u)\gamma_\parallel)} \quad (\text{B.7})$$

where we have introduced the function

$$T_R(u) = \frac{1/2}{(u + \gamma_{\parallel R})(u + \gamma_{\perp R}) + \Omega^2}. \quad (\text{B.8})$$

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