Open Quantum System Dynamics: recovering positivity of the Redfield equation via Partial-Secular Approximation

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We show how to recover complete positivity (and hence positivity) of the Redfield equation via a coarse grain average technique. We derive general bounds for the coarse graining time scale above which the positivity of the Redfield equation is guaranteed. It turns out that a coarse grain time scale has strong impact on the characteristics of the Lamb shift term and implies in general non-commutation between the dissipating and the Hamiltonian components of the generator of the dynamical semi-group. Finally we specify the analysis to a two-level system or a quantum harmonic oscillator coupled to a fermionic or bosonic thermal environment via dipole-like interaction.

I. INTRODUCTION

Describing in a proper way the whole time evolution of a quantum system interacting with a thermal environment is of crucial importance for quantum technology advancements and quantum computation [1, 2]. An exact treatment is in general a difficult task because of the exponential growth of the Hilbert space dimension with the number of constituents characterizing the system plus environment universe. Reasonable assumptions (Born and Markov approximations) lead to the Redfield equation which while being quite effective in providing a reasonable description of several quantities of interest in many contexts — for example for excitons interacting with phonon baths with broad spectrum [3] — in general does not guarantee the positivity of the system evolution, nor the more stringent complete positivity condition, i.e. positivity of correlated states of the system with external degrees of freedom [1, 4]. In practical terms this corresponds to the occurrence of non-positive eigenvalues in the system density matrix, resulting in negative expectation probabilities for observables and, consequently, in unphysical predictions. There are different ways of curving this non-positivity. For example in [5, 7] some reasonable assumptions on the bath correlation function are taken into account to handle this problem. Among these procedures the implementation of the secular approximation is a standard approach. It, on top of the Born and Markov approximations, results in the celebrated Gorini-Kossakowski-Lindblad-Sudarshan (GKLS) master equation (Lindblad equation) [5, 10]. However, when more than one characteristic frequency does characterize the internal dynamics of the system, the effectiveness of the secular approximation becomes an issue. For instance its applicability is under current debate when the system is composed by two or more interacting parties, leading to the local vs global discussion on Markovian master equations [11, 13]. In this context the global approximation makes full use of the secular approximation, leading to a steady state solution which is fully consistent with thermodynamics [12]. This is not the case (even if a reconciliation can be achieved by taking into account the work produced by the switches of the environment ancillas in a collisional model approach [15]) in general in a local approximation [14], for which — once assumed the smallness of the appropriate internal energy scales of the system — the secular approximation is not required because the positivity of the system state turns out automatically.

In this paper we show how to recover complete positivity (and hence positivity) of the Redfield equation via a coarse grain average technique that is less drastic and have less impact than the secular approximation. Contrarily to previous works [15–18] we introduce a coarse grain time scale only once the Markov and Born approximation has been fully employed, allowing to study the effects of the secular approximation without introducing spurious contributions. It turns out that this is feasible with a sufficiently large coarse grain time interval that depends on the spectrum of the system and on temperature. An estimation of such critical time scale is derived under rather general assumptions on the system-bath coupling model. Furthermore we show that a finite coarse grain time has strong impact on the structure of the Lamb shift term and that typically it implies non-commutation between the dissipating and the Hamiltonian parts of the generator of the resulting dynamical semi-group.

The sections are organized as follows: In Sec. I we introduce the general formalism of the partial secular approximation [19, 23] when the Redfield equation is taken as starting point [10]. We also discuss the above mentioned non-commutations between the Hamiltonian and dissipating components of the generator arising from the non-secular terms. Then we provide examples by specifying in Sec. II the analysis to a qubit or a quantum harmonic oscillator system coupled to a
fermionic or bosonic bath via dipole-like interaction. Here the non secular terms come from the counter-rotating contributions of the coupling between the bath and the system degrees of freedom. Finally in Sec. IV we derive general bounds for the coarse graining time scale above which the positivity of the Redfield equation is guaranteed.

II. FROM REDFIELD TO GKSL VIA SECULAR APPROXIMATION

In this section we review the microscopic derivation of the Redfield equation and how one can arrive from it to a proper GKSL form via secular approximation. Following Ref. [10] we shall work in a general setting, limiting to a minimum all the assumptions on the system Hamiltonian and on its environment. Readers who are familiar with the field can probably skip this part moving directly to following sections where our main results are presented.

A. Microscopic Model

Let $S$ be a quantum system interacting with and external environment $E$. Following conventional approach we assume the SE compound to be isolated and describe their joint evolution in terms of a global Hamiltonian $H_{SE}$ composed by three terms:

$$H_{SE} = H_S + H_E + H_1,$$

(1)

with $H_S$ and $H_E$ being local contributions, and with $H_1$ being the coupling Hamiltonian which, in full generality, we express as

$$H_1 = \sum_{\alpha=1}^{M} A_\alpha \otimes B_\alpha,$$

(2)

where $A_\alpha$ and $B_\alpha$ are not-null self-adjoint operators acting on $S$ and $E$ respectively, and where the parameter $M$ enumerates the number of non trivial terms entering the decomposition. As input state we take a factorized density matrix of the form

$$\rho_{SE}(0) = \rho_S(0) \otimes \rho_E(0),$$

(3)

with the $\rho_E(0)$ environment component fulfilling the following stationary conditions:

- invariance under the action of the local Hamiltonian, i.e.

$$[\rho_E(0), H_E] = 0;$$

(4)

where hereafter the symbols $[\cdots,\cdots]_\pm$ will be used to represent the commutator $(-)$ and the anti-commutator $(+)$, respectively;

- zero expectation value of the operators $B_\alpha$ entering the coupling Hamiltonian $[2]$, i.e.

$$\text{Tr}_E\{\rho_E(0)B_\alpha\} = 0, \quad \forall \alpha \in \{1, \cdots, M\},$$

(5)

the symbol $\text{Tr}_E\{\cdots\}$ representing the partial trace with respect to the environment degrees of freedom.

As we shall see in the following the condition (5) is essential for dropping first order contributions in the system master equation: it should be stressed that however it is not as stringent as it may looks at first site, as it can always be enforced by properly redefining the free Hamiltonian of $S$.

We hence move in the interaction picture in which the free Hamiltonian of the universe $H_0 = H_S + H_E$ is integrated away, introducing the operators

$$\tilde{H}_1(t) := e^{iH_0 t} H_1 e^{-iH_0 t},$$

(6)

$$\tilde{\rho}_{SE}(t) := e^{iH_0 t} \rho_{SE}(t)e^{-iH_0 t},$$

(7)

$$\tilde{\rho}_S(t) := \text{Tr}_E\{\tilde{\rho}_{SE}(t)\} = e^{iH_0 t} \rho_S(t)e^{-iH_0 t},$$

(8)

with $\rho_{SE}(t)$ the density matrix of $S$ and $E$ at time $t$ and $\rho_S(t) := \text{Tr}_E\{\rho_{SE}(t)\}$ its reduced form describing the corresponding state of $S$ ($\hbar$ having been set equal to $1$). Accordingly the dynamics of the joint system writes $\dot{\tilde{\rho}}_{SE}(t) = -i[\tilde{H}_1(t), \tilde{\rho}_{SE}(t)]$ which, upon formal integration, can be equivalently expressed as

$$\dot{\tilde{\rho}}_{SE}(t) = -i[\tilde{H}_1(t), \rho_{SE}(0)]_\pm - \int_0^t d\tau \left[ \tilde{H}_1(t), \left[ \tilde{H}_1(t-\tau), \tilde{\rho}_{SE}(t-\tau) \right]_\pm \right]_\pm.$$  

(9)

Taking the partial trace with respect to $E$ the left-hand-side of Eq. (9) reduces to the first derivative of $\tilde{\rho}_{S}(t)$ while the first term on the right-hand-side disappears thanks to the cooperative effect of the stationary conditions (4) and (6). The integral contribution on the contrary still exhibits a non-trivial functional dependence on the joint state $\tilde{\rho}_{SE}(t)$ which we treat by invoking the Born (or weak-coupling) approximation, requiring that at first order in the coupling the state of the environment is not affected by the presence of $S$, i.e. writing

$$\tilde{\rho}_{SE}(t_1) \simeq \tilde{\rho}_S(t_1) \otimes \tilde{\rho}_E(0),$$

(10)

all $t_1 \in [0, t]$. Under this condition we hence arrive to the following homogenous equation for $S$,

$$\dot{\tilde{\rho}}_S(t) \simeq \int_0^t d\tau \sum_{\alpha,\beta=1}^{M} c_{\alpha\beta}(\tau) \left( \tilde{A}_\beta(t-\tau) \tilde{\rho}_S(t-\tau) \tilde{A}_\alpha(t) - \tilde{A}_\alpha(t) \tilde{A}_\beta(t-\tau) \tilde{\rho}_S(t-\tau) \right) + \text{h.c.},$$

(11)

with $\tilde{A}_\alpha(t) = e^{iH_0 t} A_\alpha e^{-iH_0 t}$ and where $c_{\alpha\beta}(\tau)$ are environment correlation functions defined as

$$c_{\alpha\beta}(\tau) := \text{Tr}_E\{\rho_E(0)e^{iH_0 t} B_\alpha e^{-iH_0 t} B_\beta\},$$

(12)
that exploiting Eq. (14) and the fact that the $B_{αs}$ are self-adjoint operators can be shown to fulfil the condition

$$c_{αβ}^*(τ) = c_{βα}(−τ).$$

(13)

The next assumption concerns the memory properties of the environment. We call $τ_E$ the characteristic width of the environment correlation functions $c_{αβ}(τ)$ and we assume that the time scales $δt$ over which the system $S$ significantly evolves in the interaction picture satisfy the condition $δt \gg τ_E$. This hypothesis justifies the Markov approximation which in Eq. (11) neglects $i$ the τ dependence of the state and $ii$ substitutes the upper extreme of integration with $+∞$, leading to the Redfield equation

$$\dot{ρ}_S(t) = \sum_{ij} \Gamma_{ij}(t) (A_j^\dagger \tilde{ρ}_S(t) A_i - A_i A_j^\dagger \tilde{ρ}_S(t)) + h.c.,$$

(14)

where the last identity has been obtained by decomposing the operators $A_i$ in terms of the eigenvectors of the free system Hamiltonian. Specifically we write

$$A_α = \sum_ω A_{αω},$$

(15)

with

$$A_{αω} := \sum_{ε_1, ε_2} \pi_{ε_1} A_ε A_ε^\dagger = \sum_ε \pi_{ε+ω} A_ε A_ε^\dagger,$$

(16)

where $π_ε$ is the projector associated with the eigenvalue $ε$ of $H_S$, i.e. $H_S = \sum_ε π_ε$. The new variable $ω := ε_1 - ε_2$ spans a range of $G$ different cases, counting all the distinguishable energy gaps of $H_S$ (including the zero energy gap value associated with the terms where $ε_1 = ε_2$). Introducing then the collective indices $i = (α, ω)$ and $j = (β, ω')$ which run over a set of $N = GM$ different entries, and noticing that $A_β - ω = A_β^\dagger$, the Redfield equation (14) can hence be cast as shown in the last line with the $N \times N$ matrix $Γ_{ij}(t)$ given by

$$Γ_{ij}(t) = e^{i(ω−ω')t} Ω_{αβ}(ω'),$$

(17)

where for each value of the energy gap $ω$ the coefficients

$$Ω_{αβ}(ω) := \int_0^∞ dτ c_{αβ}(τ)e^{iωτ},$$

identify an $M \times M$ complex matrix $Ω(ω)$ that is going to play an important role in what follows. Equation (14) can be further simplified by performing a temporal averaging over coarse grain time intervals $Δτ$ which is larger than $τ_E$ but still much smaller than the time scale $δt$ where $\dot{ρ}(t)$ varies appreciably, i.e.

$$τ_E \ll Δt \ll δt.$$

(19)

The last passage is intimately connected with the hypothesis underlying the Markov approximation and, as we shall see in the following, is essential in order to recover the GKSL structure of the generator. In particular using the fact that the coarse graining does not affect $\dot{ρ}(t)$, we can replace (14) with

$$\dot{ρ}_S(t) ≃ \sum_{ij} \Gamma_{ij}^{(Δt)}(t) (A_j^\dagger \tilde{ρ}_S(t) A_i - A_i A_j^\dagger \tilde{ρ}_S(t)) + h.c.,$$

(20)

where now

$$Γ_{ij}^{(Δt)}(t) := \frac{1}{Δt} \int_{t−Δt/2}^{t+Δt/2} ds Γ_{ij}(s) S_{ω−ω'}^{(Δt)},$$

(21)

where we introduced the function

$$S_{ω−ω'}^{(ω)} := sinc[(ω−ω')Δt/2] ,$$

(22)

with $sinc[x] := sin x/x$ being the cardinal sinus.

Next we express the matrix $Γ_{ij}(t)$ in terms of its hermitian and anti-hermitian components writing

$$Γ_{ij}^{(Δt)}(t) = γ_{ij}^{(Δt)}(t)/2 + i η_{ij}^{(Δt)}(t),$$

(23)

with

$$γ_{ij}^{(Δt)}(t) := Γ_{ij}^{(Δt)}(t) + (Γ_{ij}^{(Δt)}(t))^\ast,$$

$$η_{ij}^{(Δt)}(t) := (Γ_{ij}^{(Δt)}(t) − (Γ_{ij}^{(Δt)}(t))^\ast)/2i.$$

(24)

(25)

With this choice the terms on the r.h.s. of Eq. (14) can be expressed as

$$\dot{ρ}_S(t) ≃ −i \left[ H_{LS}^{(Δt)}(t), \tilde{ρ}_S(t) \right] + \sum_{ij} γ_{ij}^{(Δt)}(t) \left( A_j^\dagger ρ_S(t) A_i - \frac{1}{2} [A_i A_j^\dagger, \tilde{ρ}_S(t)] + \right),$$

(26)

where $\{ \cdots, \cdots \}$ in the second line represents the anti-commutator and $H_{LS}^{(Δt)}(t)$ the Lamb shift term

$$H_{LS}^{(Δt)}(t) := \sum_{ij} η_{ij}^{(Δt)}(t) A_i A_j^\dagger.$$

(27)

Going back in Schrödinger picture we can finally remove the time dependence of coefficients $γ_{ij}^{(Δt)}(t)$ and $η_{ij}^{(Δt)}(t)$ obtaining a master equation with constant generator terms

$$\dot{ρ}_S(t) ≃ −i \left[ H_{LS}^{(Δt)}(t), ρ_S(t) \right] + \sum_{ij} γ_{ij}^{(Δt)}(t) \left( A_j^\dagger ρ_S(t) A_i - \frac{1}{2} [A_i A_j^\dagger, ρ_S(t)] + \right),$$

(28)

where now

$$H_{LS}^{(Δt)}(t) := H_{LS}^{(Δt)}(0) = \sum_{ij} η_{ij}^{(Δt)}(t) A_i A_j^\dagger.$$

(29)

(30)
Explicitly, exploiting the symmetry \([13]\), the \(N \times N\) matrices \(\gamma_{ij}^{(\Delta t)}\) and \(\eta_{ij}^{(\Delta t)}\) appearing in these expressions can be shown to correspond to
\[
\gamma_{ij}^{(\Delta t)}(0) = \gamma_{\alpha\omega,\beta\omega'}^{(+)} S_{\omega-\omega'}^{(\Delta t)} \ ,
\eta_{ij}^{(\Delta t)}(0) = \frac{\gamma_{\alpha\omega,\beta\omega'}^{(-)}}{2i} S_{\omega-\omega'}^{(\Delta t)} \ ,
\]
with
\[
\gamma_{\alpha\omega,\beta\omega'}^{(\pm)} := \Omega_{\alpha\beta}(\omega') \pm \Omega_{\beta\alpha}^{*}(\omega) \ .
\]

The last passage needed to put Eq. (28) in GKSL is the diagonalization of \(\gamma_{ij}^{(\Delta t)}\). Such step however works if and only if such matrix is positive semi-definite (or equivalently non-negative), the presence of negative eigenvalues only if such matrix is positive semi-definite (or equivalently non-negative), the presence of negative eigenvalues being formally incompatible with the complete-positivity requirement \([4]\) of the resulting dynamics of \(\rho(t)\). This is the reason for which one introduces the coarse-graining transformation \([21]\). Indeed thanks to the fact that
\[
\lim_{\Delta t \to \infty} S_{\omega-\omega'}^{(\Delta t)} = \delta_{\omega,\omega'} \ ,
\]
as \(\Delta t\) diverges the \(N \times N\) matrix \(\gamma_{ij}^{(\Delta t)}\) reduces to a block diagonal form with respect to the frequency labels,
\[
\eta_{ij}^{(\infty)} := \lim_{\Delta t \to \infty} \gamma_{ij}^{(\Delta t)} = \gamma_{\alpha\omega,\beta\omega}^{(+)} \delta_{\omega,\omega'} \ ,
\]
where for each \(\omega\) the coefficients \(\gamma_{\alpha\omega,\beta\omega}^{(+)}\) identify \(M \times M\) matrices
\[
\gamma^{(+)}(\omega,\omega) := \Omega(\omega) + \Omega(\omega)^{*} \ ,
\]
that are explicitly non-negative (see Appendix \([A]\) for details). The \(\Delta t \to \infty\) limit goes under the name of secular approximation (SA) and it is the last step one typically enforces in order to recover the GKSL form \([10]\): this is a rather drastic approximation, which, formally speaking, is an explicit violation of the upper bound \([19]\) and which forces two main structural constraints on the resulting master equation (ME). Specifically from \([32]\) it follows that under SA also the matrix \(\eta_{ij}^{(\Delta t)}\) gets block diagonal with respect to the gap index \(\omega\) and \(\omega'\),
\[
\eta_{ij}^{(\infty)} := \lim_{\Delta t \to \infty} \eta_{ij}^{(\Delta t)} = \frac{\gamma_{\alpha\omega,\beta\omega}^{(-)}}{2i} \delta_{\omega,\omega'} \ ,
\]
yielding the following properties

i) commutation between the Lamb shift Hamiltonian \(H_{LS}^{(\infty)}\) and the free Hamiltonian contribution \(H_{S}\), (see e.g. Eq. \([10]\) below);

ii) commutation between the free Hamiltonian \(H_{S}\) and the dissipative super-operator components \(D^{(\infty)}\) of the dynamical semi-group generator;

iii) under certain hypotheses, commutation between the full Hamiltonian \(H_{S}^{(\infty)}\) super-operator and \(D^{(\infty)}\).

The first property can be easily verified by expanding the coefficients \(\eta_{ij}^{(\Delta t)}\) appearing in Eq. \([30]\) and using the property
\[
H_{S} \pi_{\epsilon} = \pi_{\epsilon} H_{S} = \epsilon \pi_{\epsilon} \ .
\]

Accordingly we get
\[
\left[H_{S}, H_{LS}^{(\Delta t)}\right]_{-} = \sum_{\alpha\beta\omega\omega'} (\omega - \omega') \eta_{\alpha\omega,\beta\omega'}^{(\Delta t)}
\times \sum_{\epsilon} \pi_{\epsilon+\omega} A_{\alpha \epsilon} \pi_{\epsilon} A_{\beta \epsilon} \pi_{\epsilon+\omega'},
\]
which in the SA limit where Eq. \([37]\) forces \(\eta_{\alpha\omega,\beta\omega'}^{(\Delta t)}\) to be proportional to the Kronecker delta \(\delta_{\omega,\omega'}\), gets explicitly null, i.e.
\[
\lim_{\Delta t \to \infty} \left[H_{S}, H_{LS}^{(\Delta t)}\right]_{-} = \left[H_{S}, H_{LS}^{(\infty)}\right]_{-} = 0 \ .
\]

To properly express property ii) let us rewrite the r.h.s. of Eq. \([28]\) in the formal compact way:
\[
\mathcal{L}^{(\Delta t)}[\rho_{S}(t)] := \mathcal{H}^{(\Delta t)}[\rho_{S}(t)] + \mathcal{D}^{(\Delta t)}[\rho_{S}(t)] \ ,
\]
where \(\mathcal{H}^{(\Delta t)} := H_{S} + H_{LS}^{(\Delta t)}\) and \(\mathcal{D}^{(\Delta t)}\) represent the Hamiltonian and dissipative contributions to the super-operator \(\mathcal{L}^{(\Delta t)}\) which generates the system dynamics, i.e.
\[
\mathcal{H}_{S}[\cdots] := -i \left[H_{S}, \cdots\right]_{-} \ ,
\]
\[
\mathcal{H}_{LS}^{(\Delta t)}[\cdots] := -i \left[H_{LS}^{(\Delta t)}, \cdots\right]_{-} \ ,
\]
\[
\mathcal{D}^{(\Delta t)}[\cdots] := \sum_{ij} \gamma_{ij}^{(\Delta t)} \left(A_{j}^{\dagger} \cdots A_{i} - \frac{1}{2} \left[A_{i} A_{j}^{\dagger}\right]_{+}\right) \ .
\]

The commutation between \(\mathcal{H}_{S}\) and \(\mathcal{D}^{(\infty)}\), i.e.
\[
\left[H_{S}, \mathcal{D}^{(\infty)}\right]_{-} := \mathcal{H}_{S} \circ \mathcal{D}^{(\infty)} - \mathcal{D}^{(\infty)} \circ \mathcal{H}_{S} = 0 \ ,
\]
with "\(\circ\)" being the composition of super-operators, can then be proven by inspection, exploiting that, by construction, the operators \(A_{\alpha\omega}\) are eigen-operators of \(H_{S}\), i.e.
\[
[H_{S}, A_{\alpha\omega}]_{-} = \omega A_{\alpha\omega} \ .
\]

We discuss now the point iii). Despite Eqs. \([40]\) and \([45]\) the generators \(\mathcal{H}_{LS}^{(\infty)}\) and \(\mathcal{D}^{(\infty)}\) in general don’t commute (and consequently \(\mathcal{H}_{S}^{(\infty)}\) and \(\mathcal{D}^{(\infty)}\) neither) if we don’t enforce some specific hypotheses. Similarly to the property in Eq. \([16]\), a sufficient condition for the commutator
\[
\left[H_{LS}^{(\infty)}, \mathcal{D}^{(\infty)}\right]_{-}
\]
to be zero is to have the operators \(A_{\alpha\omega}\) eigen-operators of \(H_{S}^{(\infty)}\) with eigenvalues \(f(\omega)\), the last being an odd function of \(\omega\), i.e.
\[
[H_{LS}^{(\infty)}, A_{\alpha\omega}]_{-} = f(\omega) A_{\alpha\omega} ,
\]
\[
f(-\omega) = -f(\omega) \ .
\]

This is verified for instance when \((a)\) both the gaps \(\omega\) and the energies \(\epsilon\) are non-degenerate, i.e. for a given
energy gap \( \omega \) we associate one and only one pair \((\epsilon_1, \epsilon_2)\), with \( \epsilon_i \) non-degenerate eigenvalues of \( H_S \) or \( \langle b \rangle \) when the energy levels are of the type \( \epsilon_n = n\omega_0 \) — implying that the energies are non-degenerate, but the gaps are — and the only effect of \( H'_{LS}^{(\infty)} \) is a renormalization of the characteristic energy \( \omega_0 \). Examples \((a) \) and \((b) \) will be presented in the next Section. Here we just notice that when condition in Eq. (47) holds, because of Eq. (45), we achieve commutation also between the Hamiltonian and the dissipator:

\[
[H_{S}^{(\infty)}, D^{(\infty)}] = 0 .
\] (48)

Remarkably, as we shall see explicitly in the next Section, going beyond the SA by working with finite values of the coarse graining time \( \Delta t \), in general one has

\[
[H_{S}, H_{LS}^{(\Delta t)}] \neq 0 ,
\] (49)

and

\[
[H_{S}, D^{(\Delta t)}] \neq 0 .
\] (50)

Furthermore when Eq. (48) is satisfied within SA, the breaking of commutation rules in Eqs. (49), (50) can induce non-commutation also between \( H_{S}^{(\Delta t)} \) and \( D^{(\Delta t)} \), i.e.

\[
[H_{S}^{(\Delta t)}, D^{(\Delta t)}] \neq 0 .
\] (51)

III. PARTIAL SECULAR APPROXIMATION

While effective in transforming the Redfield equation into a GKSL dynamical semigroup, hence restoring the complete-positivity of the resulting dynamics, the SA is not strictly necessary. As a matter of fact in many models of physical interest, it is possible to arrive to a proper GKSL form also by adopting a Partial Secular Approach (PSA) where the coarse graining step is performed over time scales \( \Delta t \) which are finite. Explicit examples will be presented in this section, while in Sec. [V] a set of sufficient conditions that allows one to determine the range of such special coarse graining times, will be given in a rather general context. As we shall see under PSA, while the complete-positivity of the Redfield equation is maintained, three main structural modifications can occur, namely the loss of the commutation relations i), ii) and iii) detailed in the previous section.

A. An application to qubit and harmonic oscillator models

The method of the PSA can be applied in the case of a single qubit or quantum harmonic oscillator (QHO) coupled to a fermionic or bosonic bath via dipole-like interaction. As in Eq. (1), the Hamiltonian of the total system \( H_{SE} = H_S + H_E + H_1 \) is composed of three terms with

\[
H_S = \omega_0 \hat{\zeta} \hat{\zeta} ,
\] (52)

\[
H_E = \sum_k \omega_k c_k^\dagger c_k ,
\] (53)

\[
H_1 = \sum_k \gamma_k (c_k^\dagger + c_k)(\zeta + \zeta^\dagger) .
\] (54)

Equations (52) and (53) describe the free Hamiltonians of the system and of the environment respectively and Eq. (54) is the system-environment interaction. The ladder operators of the system \( \zeta \) and \( \zeta^\dagger \) and the ones of the environment \( c_k \) and \( c_k^\dagger \) respect the following commutation rules:

\[
\zeta \zeta^\dagger - s \zeta^\dagger \zeta = 1
\] (55)

\[
c_k c_k^\dagger - q c_k^\dagger c_k = \delta_{k,k'}
\] (56)

\[
c_k c_{k'} - q c_{k'} c_k = 0.
\] (57)

In this formalism \( s = 1 (-1) \) implies that the system \( S \) is a QHO (qubit) and \( q = 1 (-1) \) implies that the environment \( E \) is a bosonic (fermionic) thermal bath. Under these assumptions the Redfield Eq. (14) reduces to:

\[
\dot{\rho}_S(t) = \int_0^\infty dt \epsilon(t) \times \\
\{ \hat{A}(t - \tau) \rho_S(t) \hat{A}(t) - \hat{A}(t) \rho_S(t) \hat{A}(t - \tau) \} + \text{h.c.} .
\] (58)

Here we have no index \( \alpha \) since the interaction in Eq. (54) is a single tensor product \((M=1)\) of two hermitian operators \( A \) and \( B \), the first on the system and the second on the bath: \( A = \zeta + \zeta^\dagger, B = \sum_k \gamma_k (c_k + c_k^\dagger) \). This leads to a single bath correlation function \( c(\tau) \) (see Eq. (12)) which is convenient to split into two terms:

\[
c(\tau) := \langle \hat{B}(\tau) \hat{B} \rangle = c_1(\tau) + c_2(\tau),
\] (59)

with

\[
c_1(\tau) := \sum_k \gamma_k^2 n_k e^{-i\omega_k \tau},
\] (60)

\[
c_2(\tau) := \sum_k \gamma_k^2 (q n_k + 1) e^{-i\omega_k \tau},
\] (61)

with \( n_k := \langle c_k c_k^\dagger \rangle \) being the occupation number at wave vector \( k \), following the Bose-Einstein (Fermi-Dirac) distribution for \( q = 1 \) \((q = -1)\) :

\[
n_k = \frac{1}{e^{\beta \omega_k} - q}.
\] (62)

Notice that the value of \( q \) determines also the sign of the term into \( c_2(\tau) \) (see Eq. (61)), which is responsible for the stimulated emission in the standard SA. The expression of the system operator \( A \) in interaction picture, \( \hat{A}(t) = \zeta e^{-i\omega_0 t} + \zeta^\dagger e^{i\omega_0 t}, \) makes explicit its eigenstate representation: \( A = \sum_{\omega \in \{-\omega_0, \omega_0\}} A_{\omega}, \) with \( A_{-\omega_0} = \zeta \) and \( A_{\omega_0} = \zeta^\dagger \) and also the value of \( G=2 \). In what follows, for
brevity, the components $\pm \omega_0$ will be indicated simply by $\pm$ so that $A_{\omega_0} := \zeta^\dagger := \zeta_+ + A_{-\omega_0} := \zeta := \zeta_-$ Under PSA we obtain the following Shr"{o}dinger picture master equation:

$$\dot{\rho}_S(t) = -i [H_S + H_{LS}(\Delta t), \rho_S(t)]_+ + \sum_{\omega \omega'} \gamma^{(\Delta t)}_{\omega \omega'} \left( \zeta^\dagger \rho_S(t) \zeta - \frac{1}{2} \left[ \zeta \zeta^\dagger, \rho_S(t) \right]_+ \right),$$

where

$$H_{LS}(\Delta t) = \sum_{\omega \omega'} \eta^{(\Delta t)}_{\omega \omega'} \zeta^\dagger \zeta_\omega \zeta^\dagger \zeta_\omega.$$

(64)

According to Eqs. (22-25) of the general formalism, the matrices $\gamma^{(\Delta t)}_{\omega \omega'} = \Gamma^{(\Delta t)}_{\omega \omega'} + \Gamma^{(\Delta t)}_{\omega' \omega'}$ and $\eta^{(\Delta t)}_{\omega \omega'} = 1/(2i)(\Gamma^{(\Delta t)}_{\omega \omega'} - \Gamma^{(\Delta t)}_{\omega' \omega'})$ are obtained from the hermitian and anti-hermitian parts of the $2 \times 2$ matrix

$$\Gamma^{(\Delta t)}_{\omega \omega'} = S^{(\Delta t)}_{\omega \omega'} \int_0^\infty d\tau c(\tau) e^{i\omega' \tau},$$

(65)

where now, because $\omega, \omega' \in \{-\omega_0, \omega_0\}$, we can write

$$S^{(\Delta t)}_{\omega \omega'} = \delta_{\omega \omega'} + (1 - \delta_{\omega \omega'}) \operatorname{sinc}(\omega_0 \Delta t).$$

(66)

As discussed at the end of the previous section the complete-positivity properties of the master equation (63) are directly linked to the spectrum of the matrix $\gamma^{(\Delta t)}$. To evaluate its entries we pass to the continuous frequency counterpart of model, introducing the density of states of the thermal bath $D_\epsilon := \sum_k \delta(\omega_k - \epsilon)$, an the associated spectral density $\rho_\epsilon := D_\epsilon \gamma_\epsilon^2 = \sum_k \delta(\omega_k - \epsilon) \gamma_k^2$, obtaining

$$\gamma_{-\epsilon} = \kappa_{\omega_0} n_{\omega_0},$$

$$\gamma_{\epsilon} = \kappa_{\omega_0} (1 + q n_{\omega_0}),$$

(67)

(68)

which explicitly do not depends upon $\Delta t$, and

$$\gamma_{-\epsilon}^{(\Delta t)} = \gamma_{\epsilon}^{(\Delta t)} = \frac{(q + 1) n_{\omega_0} + 1) \kappa_{\omega_0}}{2} - i \mathcal{I} \operatorname{sinc}(\omega_0 \Delta t),$$

(69)

where $\kappa_\epsilon := 2 \pi \rho_\epsilon$ is the system decay rate, and where

$$\mathcal{I} := \frac{\omega_0}{\pi} \int_0^\infty d\epsilon \frac{(q + 1) n_{\omega_0} + 1) \kappa_\epsilon}{\epsilon^2 - \omega_0^2},$$

(70)

the symbol "$\mathcal{I}$" indicating that we are considering the principal value of the integral.

Finally we report the components of the Lamb shift matrix that will be useful in what follows:

$$\eta_{-\epsilon} = \mathcal{I}_-, \quad \eta_{\epsilon} = \mathcal{I}_+,$$

(71)

$$\eta_{-\epsilon}^{(\Delta t)} = \frac{i}{4} (\gamma_{\epsilon} - \gamma_{-\epsilon}) + \frac{1}{2} (\mathcal{I}_- + \mathcal{I}_+) \gamma^{(\Delta t)}_{-\epsilon},$$

$$\eta_{\epsilon}^{(\Delta t)} = \eta_{-\epsilon}^{(\Delta t)}.$$
and the dissipator becomes
\[
D(\Delta t)[\cdots] = \gamma_- \left(f_-^\dagger \cdots f_- - \frac{1}{2} \left[f_- f_-^\dagger, \cdots \right]_+ \right)
+ \gamma_+ \left(f_+^\dagger \cdots f_+ - \frac{1}{2} \left[f_+ f_+^\dagger, \cdots \right]_+ \right),
\]
where for easy of notation we dropped any reference to the functional dependence upon \(\Delta t\) of the terms that appears on the right-hand-side term. It is worth noticing that in general for finite values of \(\Delta t\), one has that \(f_+\) is not the adjoint counterpart of \(f_-\), i.e. \(f_+ \neq f_-^\dagger\), the identity instead holding in the full SA limit where \(\mathcal{U} = 1\).

A plot of the upper bound on the r.h.s. of the above expressions is reported in Fig. 1 as function of \(k_B T/\omega_0\), both for bosonic [Panel (a)] and for fermionic baths [Panel (b)] for a decay rate of the form
\[
\kappa_{\epsilon} = \kappa_0 \epsilon \exp(-\epsilon/\omega_c).
\]
that behaves ohmically for small energies — \(\kappa_{\epsilon} \sim \epsilon\) for \(\epsilon \ll \omega_c\) — and decays exponentially with the cutoff energy \(\omega_c\). From Fig. 1 we deduce that at low temperature the full SA \((\Delta t \to \infty)\) is necessary for ensuring positivity; this is a general behaviour that does not depend upon the special form of the decay rate we choose for the plot. Indeed for \(\beta \to \infty\) the right-hand side of Eq. (77) always nullifies forcing us to take \(\Delta t \to \infty\) in order to satisfy the inequality; for non-zero temperature values instead, finite values of \(\Delta t\) are admitted such that the associated PSA master equation is well behaved. Furthermore we note that the only difference between the bosonic (Fig. 1a) and the fermionic (Fig. 1b) cases comes just from the principal value integral \(\mathcal{I}\). The last turns out to be independent of temperature only for fermions, see Eq. (70). Finally in Fig. 2 we compare the actual threshold value of Eq. (77) with the value provided by the estimation of Eq. (130) associated with the sufficient positivity condition we derive in Sec. IV under general assumption on the system dynamics, which for examples we study here assumes the form
\[
|\text{sinc}(\omega_0 \Delta t)| \leq \frac{2\kappa_{\omega_0}\omega_0}{\sqrt{\omega_0^2 + 4\Delta^2} + \sqrt{(\omega_0^2 + 4\kappa_{\omega_0})^2 + 4\Delta^2}},
\]
where \(\mathcal{I}_-\) are the same as in Eq. (72) [notice that \(\mathcal{I} = \mathcal{I}_- - \mathcal{I}_+\), see Eq. (70)]. We infer that Eq. (81) underestimates the threshold value at low temperatures and gives better results at high temperatures.

### C. Commutativity

As anticipated in the end of Sec. IVA the main structural consequence of the implementation of the PSA is the breaking of the commutation rules either at the level of the operators than at the level of the super-operators entering in Eq. (63). We distinguish now the cases of qubit and QHO and in particular we concentrate on three main aspects: i) the value of the commutator between the Lamb shift and the free Hamiltonian of the system, ii) the change of the commutation rules at the level of super-operators, iii) the effects of the non-secular terms in the dynamics. About item ii) we remark that both for qubit and for QHO we are in the hypotheses of Eq. (47) for which Eq. (48) is satisfied.
1. Qubit

In the case of qubit ($s = -1$) the ladder operator $\zeta$ is the operator $\sigma_- = |0\rangle \langle 1|$, with $|0\rangle$ and $|1\rangle$ being the eigenvectors of the Pauli matrix $\sigma_-$ corresponding to the eigenvalues $-1$ and $1$, respectively. Because $\sigma_+^2 = 0$, the Hamiltonian of the system is modified just by a change of the two level spacing of the system and is independent of $\Delta t$, i.e.

$$H_S^{(\Delta t)} = \tilde{\omega} \sigma_+ \sigma_-, \quad \tilde{\omega} := \omega_0 + \eta_+ - \eta_-,$$

(82)

with $\eta_-$ and $\eta_+$ as in Eq. (71). Accordingly for all allowed PSA values $\Delta t$, this grants commutation between the free and LS contributions of the full Hamiltonian, i.e.

$$[H_{LS}^{(\Delta t)}, H_S]_-= 0.$$

(84)

On the contrary the dissipator \(^{(63)}\) acquires non-secular terms which depend on $\Delta t$:

$$\mathcal{D}^{(\Delta t)}[\rho_S(t)] = \gamma_{--} \left( \sigma_- \rho_S(t) \sigma_- - \frac{1}{2} \{ \sigma_- \sigma_+, \rho_S(t) \} \right) + \gamma_{++} \left( \sigma_+ \rho_S(t) \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho_S(t) \} \right) + \gamma_{-+} (\sigma_- \rho_S(t) \sigma_+ + \gamma^{(\Delta t)} \sigma_+ \rho_S(t) \sigma_-),$$

with $\gamma_{-+}$ as in Eq. (69). Hence at the level of superoperators, it is interesting to observe that the Hamiltonian and the dissipating parts of the generator of the dynamical semi-group don’t commute under PSA:

$$\left[ \mathcal{D}^{(\Delta t)}, H_S^{(\Delta t)} \right] = -2i\tilde{\omega} \left( \gamma^{(\Delta t)} \sigma_- \rho \sigma_- - \text{h.c.} \right).$$

(85)

Notice that consistently the commutator in Eq. (85) nullifies in the limit $\Delta t \to \infty$, i.e. within the SA. Analogous considerations hold about the term $\left[ \mathcal{D}^{(\Delta t)}, H_S \right]_-$ once replaced $\tilde{\omega}$ with the bare frequency $\omega_0$.

We show now an example of non-positive semi-definite evolution by considering as initial state the pure vector $|\psi(0)\rangle_S := (|0\rangle + |1\rangle)/\sqrt{2}$. Its dynamics is described by Eq. (63), which in a more explicit form reads as

$$\frac{d}{dt} \rho_{S00} = \gamma_{++} (1 - \rho_{S00}) - \gamma_{--} \rho_{S00}, \quad \frac{d}{dt} \rho_{S10} = -i\tilde{\omega} \rho_{S10} - \frac{1}{2} (\gamma_{++} + \gamma_{--}) \rho_{S10} + \gamma^{(\Delta t)} \rho^{(\Delta t)} S_{11},$$

(86)

(87)

(88)

with

$$s := \gamma_{++} - \gamma_{--}, \quad d := \gamma_{++} + \gamma_{--},$$

$$\tilde{\omega}_{\Delta t} := \sqrt{\tilde{\omega}^2 - |\gamma^{(\Delta t)}|^2}. \quad \quad \quad (90)$$

We plot the results in Fig. 3 for different values of $S_{2\omega}$, corresponding to the SA (i.e. $S_{2\omega} = 0$); to the PSA at positivity threshold (bound of Eq. (77)); and to the Redfield regime (i.e. $S_{2\omega} = 1$). Panels (a), (b) and (c) of the figure show that the PSA — when compared to the SA — implies corrections on the off-diagonal terms of $\rho_S(t)$ only, while leaving unchanged the diagonal ones

and the steady state of the system, i.e.

$$\rho_S(\infty) = \rho_{SS} := \begin{pmatrix} n_f(\omega_0) & 0 \\ 0 & 1 - n_f(\omega_0) \end{pmatrix},$$

(91)

where $n_f(\omega_0)$ is the Fermi-Dirac occupation number, $\frac{1}{n_f(\omega_0)} := \frac{1}{1 - \frac{1}{1 - n_f(\omega_0)}}$. As evident from the plots PSA somehow interpolates between SA and the R-B behaviours, retaining part of the fast oscillations of the latter which are instead washed away by the former. In Panel (d) of Fig. 3 it is instead plotted the determinant of $\rho_S(t)$. For short time scales Redfield implies non-positive evolution being $\text{Det}[\rho_S(t)] < 0$, whilst positivity is main-
where the parameters $d$, $s$ and $\omega_{\Delta t}$ are again defined as in Eq. (90). Being
\[
\lambda^{(\Delta t)}(0) = 0, \quad \hat{\lambda}^{(\Delta t)}(0) = \frac{1}{2} \left( s - \sqrt{d^2 + 4|\lambda^{(\Delta t)}|} \right),
\]
we obtain that the first derivative $\hat{\lambda}^{(\Delta t)}(0) \geq 0$ when $|\gamma^{(\Delta t)}_{++}|^2 \geq \gamma^{(\Delta t)}_{--}$ and, consequently, at short time scales it happens that $\lambda^{(\Delta t)}(t) \geq 0$. About the steady state, for any value of $S^{(\Delta t)}_{2\omega_0}$, $\rho_{CJ}(\infty) = \rho_\beta \odot \frac{1}{2} 1_A$ with $\rho_\beta$ being again the Gibbsian state of Eq. (91).

2. Quantum harmonic oscillator

Here the system operator $\zeta$ is the annihilation operator $a$. The Lamb shift arises from the PSA counter-duality provided by the Choi-Jamiolkowski (CJ) isomorphism \[1\] \[25\] \[20\]. Given a quantum channel $\Phi_S(t)$ which maps $\rho_\beta(0) \rightarrow \rho_S(t)$, considering the situation in which $S$ is maximally entangled with an ancilla $A$ having the same dimensionality as $S$, the CJ-state $\rho_{CJ}(t)$ is obtained by applying the channel locally on $S$. In this case it reads as
\[
\rho_{CJ}(t) := (\Phi_S(t) \otimes i d_A)(|\psi\rangle \langle \psi|_{SA}), \quad (92)
\]
where the initial state $|\psi\rangle_{SA}$ is the maximally entangled state
\[
|\psi\rangle_{SA} := \frac{1}{\sqrt{2}}(|00\rangle_{SA} + |11\rangle_{SA}). \quad (93)
\]
Hence $\rho_{CJ}(t)$ is a $4 \times 4$-matrix having the following block form:
\[
\rho_{CJ}(t) = \frac{1}{2} \begin{pmatrix} \rho_S(t; |1\rangle \langle 1|) & \rho_S(t; |1\rangle \langle 0|) \\ \rho_S(t; |0\rangle \langle 1|) & \rho_S(t; |0\rangle \langle 0|) \end{pmatrix}, \quad (94)
\]
with $\rho_S(t; |i\rangle \langle j|)$ being the $2 \times 2$-matrix obtained by the solution of the ME Eq. (63) under the initial condition $\rho_S(0) = |i\rangle \langle j|$. The occurrence of negative eigenvalues of $\rho_{CJ}(t)$ encodes the non-complete positivity (non-CP) of the map $\Phi_S(t)$. In Fig. 4 we plot the four eigenvalues of $\rho_{CJ}(t)$ as function of time for different values of $\gamma^{(\Delta t)}$. Non-CP manifests at short time scales as soon as the threshold value of Eq. (17) is overcome. This can be understood by looking at the analytic expression of the eigenvalue $\lambda^{(\Delta t)}(t)$ corresponding to the black full lines in Fig. 4.
FIG. 4. (Color online) Qubit interacting with a bosonic bath (q = 1). We plot the four eigenvalues of ρ_{\text{CP}}(t) as function of time in units 1/ω_{0} for different values of S_{2ω_{0}}^{(Δt)}. Panel (a): S_{2ω_{0}}^{(Δt)} = 0 (SA); Panel (b): S_{2ω_{0}}^{(Δt)} = 0.628 (PSA at positivity threshold S_{2ω_{0}}^{(Δt)} = S_{2ω_{0}}^{(ω_{0})}); Panel (c): S_{2ω_{0}}^{(Δt)} = 1 (Redfield). Then we move across the threshold value to appreciate the crossover between CP and non-CP evolution. Panel (d): S_{2ω_{0}}^{(Δt)} = 0.621 (S_{2ω_{0}}^{(Δt)} slightly below S_{2ω_{0}}^{(ω_{0})}); Panel (e): S_{2ω_{0}}^{(Δt)} = 0.628 (S_{2ω_{0}}^{(ω_{0})} = S_{2ω_{0}}^{(ω_{0})}); Panel (f): S_{2ω_{0}}^{(Δt)} = 0.634 (S_{2ω_{0}}^{(ω_{0})} slightly above S_{2ω_{0}}^{(ω_{0})}). Notice the different axes scales with respect to the first three Panels. We choose the following values of the master equation parameters: k_{B}T = 0.5ω_{0}, κ_{0} = 2, ω_{c} = 5ω_{0}.

Here the dissipator reads as follows:

\[\mathcal{D}^{(Δt)}[ρ_{S}(t)] = γ_{−}\left(a^\dagger ρ_{S}(t)a - \frac{1}{2}\{aa^\dagger, ρ_{S}(t)\}\right) + γ_{++}\left(αρ_{S}(t)a^\dagger - \frac{1}{2}\{a^\dagger a, ρ_{S}(t)\}\right)\]

\[+γ^{−}a^\dagger(αρ_{S}(t)a - \frac{1}{2}\{a^2, ρ_{S}(t)\}) + γ^{++}a^\dagger(ρ_{S}(t)a^\dagger - \frac{1}{2}\{a^{\dagger 2}, ρ_{S}(t)\})\]

and the PSA implies again a breaking of commutation rules at the level of super-operators. Specifically, as anticipated in Eqs. (50) and (51), both the bare and the full Hamiltonians don’t commute with the dissipator:

\[\left[D^{(Δt)}, H_{S}\right]_{−}(ρ) = -iω_{0}\left[γ^{−}\left(2αρa - a^{2}ρ - ρa^{2}\right) - \text{h.c.}\right], \quad (101)\]

\[\left[D^{(Δt)}, H_{S}^{(Δt)}\right]_{−}(ρ) = -i\left[\bar{ω}γ^{−}\left(2αρa - a^{2}ρ - ρa^{2}\right) - \text{h.c.}\right] - 2i\left[γ^{−}\left(2αρa + a^\dagger aρ - ρa^\dagger a - ρ\right) - \text{h.c.}\right]. \quad (102)\]

About the effects on the dynamics, the presence of squeezing introduces a coupling between the second momenta of the system:

\[\langle a^2 \rangle = -2i\left(\bar{ω}\langle a^2 \rangle + 2η^{−}\langle a^\dagger a \rangle + η^{−} \langle a^\dagger a \rangle - γ^{−} - γ^{−}\right)\langle a^2 \rangle, \quad (103)\]

\[\langle a^\dagger a \rangle = 2i\left(η^{−}\langle a^\dagger a \rangle - η^{−}\langle a^\dagger a \rangle - γ^{++} + γ^{−}\right)\langle a^\dagger a \rangle + γ^{−}. \quad (104)\]

In Fig. 5 we plot \(\langle a^\dagger a(t)\rangle, \text{Re}[\langle a^2(t)\rangle], \text{Im}[\langle a^2(t)\rangle]\) choosing a bosonic bath (q = 1) and considering as initial state
of $\gamma(\Delta t)$ is non-negative, i.e.

$$\Lambda_{\text{min}}(\Delta t) \geq 0.$$  \hspace{1em} (106)

For small values of $N$, Eq. (106) turns out to be the proper way to go as explicitly verified in the previous section. However, as $N$ increases determining $\Lambda_{\text{min}}(\Delta t)$ can be problematic. In what follows we hence present an alternative, computational less demanding, approach which allows one to characterize the set of suitable $\Delta t$, by only focusing on the properties of the $M \times M$ blocks $\Omega(\omega)$ defined in Eq. (13). The main result of this analysis is the identification of a critical threshold $\Delta t_c$ above which the coarse graining time $\Delta t$ is guaranteed to yield a positive semi-definite $\gamma(\Delta t)$, i.e.

$$\Delta t \geq \Delta t_c \implies \gamma(\Delta t) \geq 0.$$  \hspace{1em} (107)

Specifically indicating with $||\Omega(\omega)||_\infty$ the operator norm of $\Omega(\omega)$, i.e.

$$||\Omega(\omega)||_\infty := \sup_{\theta} \sqrt{\frac{||\Omega(\omega)^+||_\infty^2 + ||\Omega(\omega)^-||_\infty^2}{|\theta(\omega)|}},$$  \hspace{1em} (108)

and with $\lambda_{\text{min}}(\omega)$ the minimum eigenvalue of its Hermitian component $\gamma(\omega, \omega)$ defined in Eq. (30) (which is non-negative by construction), in the next subsection we shall prove that one can identify $\Delta t_c$ with the quantity

$$\Delta t_c := 2(G - 1) \max_{\omega, \omega'} \left(\frac{||\Omega(\omega)||_\infty + ||\Omega(\omega')||_\infty}{|\omega - \omega'| \lambda_{\text{min}}(\omega)}\right),$$  \hspace{1em} (109)

or with its more compact, version

$$\Delta t_c := \frac{4(G - 1) ||\Omega||_{\text{max}}}{\nu_{\text{min}} \lambda_{\text{min}}},$$  \hspace{1em} (110)

where

$$\nu_{\text{min}} := \min_{\omega, \omega'} |\omega - \omega'|,$$  \hspace{1em} (111)

is minimum among all the gaps differences. As $\Delta t_c^{(2)}$ is always larger than $\Delta t_c^{(1)}$, it provides a worst estimation of the real critical threshold $\Delta t_c$. Still Eq. (110) is more informative as it makes explicit that $\Delta t_c$ should scale as the inverse of the minimal difference $\nu_{\text{min}}$. An estimation of the critical time $\Delta t_c$ that is provably better, but more involved than $\Delta t_c^{(1)}$, is finally given by the quantity

$$\Delta t_c^{(0)} := \max_{\omega} \left(\frac{2}{Q(\omega) K(\omega) \lambda_{\text{min}}(\omega)}\right),$$  \hspace{1em} (112)

obtained by the functions

$$Q(\omega) := \sum_{\omega', \omega' \neq \omega} \frac{|\omega - \omega'|}{||R(\omega)||_\infty + ||R(\omega')||_\infty},$$  \hspace{1em} (113)

$$q_{\omega'}(\omega) := \frac{1}{||R(\omega)||_\infty + ||R(\omega')||_\infty} Q(\omega'),$$  \hspace{1em} (114)

and

$$K(\omega) := \sum_{\omega', \omega' \neq \omega} \frac{1}{q_{\omega'}(\omega')},$$  \hspace{1em} (115)
A. Derivation of the bounds via matrix dilution

Here we explicitly show that both the terms Eq. (109) and (112) are suitable choices for the critical time $\Delta t_c$ entering Eq. (107).

We start by observing that by expanding the indexes $i$ and $j$, Eq. (105) can be conveniently casted in the following form

$$\sum_{\omega} \bar{u}^\dagger(\omega) \cdot \gamma(\omega,\omega) \cdot \bar{u}(\omega)$$

$$+ \sum_{\omega,\omega' \neq \omega} S_{\omega,\omega'}^{(\Delta t)} \bar{u}^\dagger(\omega) \cdot \gamma(\omega,\omega') \cdot \bar{u}(\omega') \geq 0,$$

where for given $\omega$ and $\omega'$,

$$\gamma(\omega,\omega') := \Omega(\omega) + \Omega(\omega'),$$

represents the $M \times M$ matrix with elements provided by the terms $s_{\omega,\omega'}^{(+)\gamma}$ of Eq. (33), and where $\bar{u}(\omega)$ is the $M$-dimensional vector defined by the components of $\bar{u}$ associated with the corresponding block $\omega$, i.e. $\bar{u} = (\bar{u}(\omega_1), ..., \bar{u}(\omega_G))^T$.

It is worth observing that the first contribution of Eq. (116) corresponds to the term one would get when enforcing secular approximation (i.e. enforcing the $\Delta t \rightarrow \infty$ limit): accordingly, for all choices of $\bar{u}$ this term can always be guaranteed to be non negative, i.e.

$$\sum_{\omega} \bar{u}^\dagger(\omega) \cdot \gamma(\omega,\omega) \cdot \bar{u}(\omega) \geq 0.$$  \hfill (118)

Problems on the contrary can arise from the second contribution which involves the off-diagonal blocks $\gamma(\omega,\omega')$ with $\omega \neq \omega'$. To treat them we adopt the following dilution technique dividing the contribution coming from the diagonal block term $\omega = \omega'$ into fractions which are then added to the terms associated with the off-diagonal blocks $\omega \neq \omega'$. Specifically, for each given $\omega$ let us introduce a set of numbers $\{p_{\omega'}^{(+)\omega}\}_{\omega'}$ such that

$$\begin{cases}
p_{\omega'}^{(+)\omega} \geq 0, & \omega \neq \omega', \\
\sum_{\omega' : \omega \neq \omega'} p_{\omega'}^{(+)\omega} = 1.
\end{cases}$$  \hfill (119)

They form $G$ sets of probabilities with $G - 1$ entries, which we shall employ as free parameters in our analysis and which allow us to rewrite (116) in the following symmetrized form

$$\sum_{\omega,\omega' \neq \omega} \left\{ p_{\omega'}^{(+)\omega} \bar{u}^\dagger(\omega) \cdot \gamma(\omega,\omega) \cdot \bar{u}(\omega) + p_{\omega'}^{(+)\omega} \bar{u}^\dagger(\omega') \cdot \gamma(\omega',\omega') \cdot \bar{u}(\omega') \right\} + 2 S_{\omega,\omega'}^{(\Delta t)} \text{Re} \left[ \bar{u}^\dagger(\omega) \cdot \gamma(\omega,\omega') \cdot \bar{u}(\omega') \right] \geq 0,$$  \hfill (120)

where we grouped together all the contributions of all the couples $\omega$ and $\omega' \neq \omega$, and used the fact that $S_{\omega,\omega'}^{(\Delta t)}$ is invariant under exchange of $\omega$ and $\omega'$, and the identity $\gamma(\omega,\omega') = [\gamma(\omega,\omega')]^\dagger$.

Now a sufficient condition ensuring that Eq. (120) holds for all $\bar{u}$, can be obtained by forcing each one of such contributions to verify the same property. More specifically, we can claim that the matrix $\gamma^{(\Delta t)}$ is non-negative at least for those $\Delta t$ such that, there exists a proper choice of the probabilities $\{p_{\omega'}^{(+)\omega}\}_{\omega'}$ for which

$$\mathcal{F}_{\omega,\omega'}^{(\Delta t)}(\bar{u}(\omega), \bar{u}(\omega')) := p_{\omega'}^{(+)\omega} \bar{u}^\dagger(\omega) \cdot \gamma(\omega,\omega) \cdot \bar{u}(\omega) + p_{\omega'}^{(+)\omega} \bar{u}^\dagger(\omega') \cdot \gamma(\omega',\omega') \cdot \bar{u}(\omega')$$

$$+ 2 S_{\omega,\omega'}^{(\Delta t)} \text{Re} \left[ \bar{u}^\dagger(\omega) \cdot \gamma(\omega,\omega') \cdot \bar{u}(\omega') \right] \geq 0,$$  \hfill (121)

for all possible choices of $\omega, \omega', \bar{u}(\omega), \bar{u}(\omega')$. Next step is to construct a lower bound for the quantity $\mathcal{F}_{\omega,\omega'}^{(\Delta t)}(\bar{u}(\omega), \bar{u}(\omega'))$. For this purpose we begin observing that, indicating with $\lambda_{\text{min}}(\omega)$ the minimum eigenvalue of the matrix $\gamma^{(+)\omega}(\omega,\omega)$, we have

$$\bar{u}^\dagger(\omega) \cdot \gamma^{(+)\omega}(\omega,\omega) \cdot \bar{u}(\omega) \geq |\bar{u}(\omega)|^2 \lambda_{\text{min}}(\omega),$$  \hfill (122)

with $|\bar{u}(\omega)|$ being the norm of the vector $\bar{u}(\omega)$. Then by using Eq. (117), the Cauchy-Schwartz inequality, and the fact that for generic $\bar{u}$ one has $|\bar{u}^\dagger(\omega) \cdot \Omega(\omega) \cdot \bar{u}(\omega)| \leq |\bar{u}(\omega)| \|\Omega(\omega)\|_\infty$, we observe that

$$\left| \text{Re} \left[ \bar{u}^\dagger(\omega) \cdot \gamma^{(+)\omega}(\omega,\omega') \cdot \bar{u}(\omega') \right] \right| \leq |\bar{u}^\dagger(\omega) \cdot \gamma^{(+)\omega}(\omega,\omega') \cdot \bar{u}(\omega')| \leq |\bar{u}^\dagger(\omega) \cdot \Omega(\omega) \cdot \bar{u}(\omega')| + |\bar{u}^\dagger(\omega') \cdot \Omega(\omega') \cdot \bar{u}(\omega)|$$

$$\leq |\bar{u}(\omega')| \|\bar{u}(\omega')\| \left( \|\Omega(\omega)\|_\infty + \|\Omega(\omega')\|_\infty \right).$$  \hfill (123)
which implies
\[ 2 S_{\omega,\omega'}^{(\Delta t)} \cdot \text{Re} \left[ \overline{\bar{u}}(\omega) \cdot \gamma^{(+)}(\omega, \omega') \cdot \bar{u}(\omega') \right] \geq -2 \left| S_{\omega,\omega'}^{(\Delta t)} \right| \cdot \left| \text{Re} \left[ \overline{\bar{u}}(\omega) \cdot \gamma^{(+)}(\omega, \omega') \cdot \bar{u}(\omega') \right] \right| \]
\[ \geq -2 \left| S_{\omega,\omega'}^{(\Delta t)} \right| \cdot |\overline{\bar{u}}(\omega)| \cdot \left( ||\Omega(\omega)||_{\infty} + ||\Omega(\omega')||_{\infty} \right). \]

Replacing hence \[ (122) \] and \[ (124) \] into the definition of \( \mathcal{F}_{\omega,\omega'}^{(\Delta t)}(\bar{u}(\omega), \bar{u}(\omega')) \) we arrive to establish the following bound
\[ \mathcal{F}_{\omega,\omega'}^{(\Delta t)}(\bar{u}(\omega), \bar{u}(\omega')) \geq \mathcal{F}_{\omega,\omega'}^{(\Delta t)}(\bar{u}(\omega), \bar{u}(\omega')) \]

with \( \mathcal{F}_{\omega,\omega'}^{(\Delta t)}(\bar{u}(\omega), \bar{u}(\omega')) \) being the function
\[ \mathcal{F}_{\omega,\omega'}^{(\Delta t)}(\bar{u}(\omega), \bar{u}(\omega')) := \lambda_{\min}(\omega) \cdot \left( |\overline{\bar{u}}(\omega)|^2 + |\overline{\bar{u}}(\omega')|^2 \right) \geq 2 S_{\omega,\omega'}^{(\Delta t)} \cdot |\overline{\bar{u}}(\omega)| \cdot \left( ||\Omega(\omega)||_{\infty} + ||\Omega(\omega')||_{\infty} \right). \]

From Eq. \[ (125) \] it then follows that a sufficient condition for Eq. \[ (121) \] is the positivity of the function \( \mathcal{F}_{\omega,\omega'}^{(\Delta t)}(\bar{u}(\omega), \bar{u}(\omega')) \), which by looking at \[ (126) \] can be guaranteed by imposing the function \( B_{\omega,\omega'}^{(\Delta t)} \) to be smaller than \( A_{\omega,\omega'} \) and \( A_{\omega',\omega} \), i.e.
\[ B_{\omega,\omega'}^{(\Delta t)} \leq \min\{A_{\omega,\omega'}, A_{\omega',\omega}\}, \]
which can be casted in the equivalent form
\[ |S_{\omega,\omega'}^{(\Delta t)}| \leq \frac{\lambda_{\min}(\omega)}{||\Omega(\omega)||_{\infty} + ||\Omega(\omega')||_{\infty}}. \]
by exploiting the symmetry \( B_{\omega,\omega'}^{(\Delta t)} = B_{\omega',\omega}^{(\Delta t)} \). Noticing that from Eq. \[ (22) \] we have \( |S_{\omega,\omega'}^{(\Delta t)}| \leq 2/(||\omega - \omega'||\Delta t) \), the latter can then be replaced by the (stronger) requirement
\[ \Delta t \geq \frac{2}{\lambda_{\min}(\omega)} \cdot \frac{||\Omega(\omega)||_{\infty} + ||\Omega(\omega')||_{\infty}}{||\omega - \omega'|| \lambda_{\min}(\omega)}. \]

To summarize, any coarse graining time \( \Delta t \) admitting a set of probability functions \( \{p_{\omega'}^{(\omega)}\}_{\omega'} \) for which the inequality \[ (131) \] holds for all \( \omega \) and \( \omega' \), ensures the fulfillment of Eq. \[ (120) \], hence the non-negativity of the matrix \( \gamma^{(\Delta t)} \) (notice that if \( \lambda_{\min}(\omega) = 0 \) for some \( \omega \), Eq. \[ (131) \] can still be used: simply it implies that \( \Delta t \) has to be infinite). Alternatively, we can say that for each assigned choice of the dilution probabilities \[ (119) \] the fulfillment of the inequality Eq. \[ (120) \] allows us to identify a coarse graining time \( \Delta t \) that implies the non-negativity of \( \gamma^{(\Delta t)} \). Taking for instance \( \{p_{\omega'}^{(\omega)}\}_{\omega'} = \{1/G - 1\} \), \( \forall \omega' \neq \omega \) equation \[ (131) \] becomes
\[ \Delta t \geq (G - 1) \cdot \frac{||\Omega(\omega)||_{\infty} + ||\Omega(\omega')||_{\infty}}{|\omega - \omega'| \lambda_{\min}(\omega)}. \]
Using a general formalism we found sufficient conditions to guarantee the complete positivity of the Redfield equation, among which a tight bound on the coarse grain time interval. Furthermore we explicitly show that non-secular terms can determine non commutation between the Hamiltonian and the dissipating parts of the master equation. We thus provide examples by applying the partial secular approximation to a qubit or harmonic oscillator interacting with a fermionic or bosonic thermal environment via dipole-like interaction.

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Appendix A: Positivity of the matrix \( \gamma^{(+)}(\omega, \omega) \)

We discuss here the positivity of the secular blocks \( \gamma^{(+)}_{\alpha\omega, \beta\omega} \) of the matrix \( \gamma^{(+)}_{\alpha\omega, \beta\omega} \) of Eq. (31), by following the demonstration given in [10]. Such blocks are the ones with equal frequencies and their entries read as

\[
\gamma^{(+)}_{\alpha\omega, \beta\omega} = \int_{-\infty}^{+\infty} c_{\alpha\beta}(\tau) e^{i\omega\tau} d\tau ,
\]

(A1)

with \( c_{\alpha\beta}(\tau) \) being the bath correlation functions given in Eq. (12). We should prove that

\[
\sum_{\alpha\beta} u^*_\alpha(\omega) \gamma^{(+)}_{\alpha\omega, \beta\omega} u_\beta(\omega) \geq 0 \tag{A2}
\]

for any \( \tilde{u}(\omega) \in \mathbb{C}^M \). The above expression is actually the Fourier transform of a function \( f(\tau) \):

\[
\sum_{\alpha\beta} u^*_\alpha(\omega) \gamma^{(+)}_{\alpha\omega, \beta\omega} u_\beta(\omega) = \int_{-\infty}^{+\infty} e^{i\omega\tau} f(\tau) d\tau \tag{A3}
\]

with

\[
f(\tau) := \langle \Theta^{+}(\tau) \Theta(0) \rangle , \tag{A4}
\]

From the positivity of the matrix \( f_{lm} \) it follows that the Fourier transform of \( f(\tau) \) is always non-negative (Bochner’s theorem) and hence the positivity of the matrix \( \gamma^{(+)}_{\alpha\omega, \beta\omega} \) is guaranteed (see Eq. (A3)).

This can be understood by thinking integrals as summations. Indeed the Fourier transform in the right-hand side of Eq. (A3) can be written in a form which is analogous to the left-hand side of Eq. (A7) which we know to be a positive quantity:

\[
\int_{-\infty}^{+\infty} ds' e^{i\omega s'} f(s') = \frac{1}{2T} \int_{-T}^{+T} dl \int_{-\infty}^{+\infty} ds u^*(s)f(s-l)u(l) \geq 0 ,
\]

(A9)

with \( u(\tau) := e^{-i\omega\tau} \) and for any \( T \).

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[1] M. A. Nielsen and I. L. Chuang, Quantum Computation and Quantum Information: 10th Anniversary Edition, Cambridge University Press (2010).
[2] M.F. Riedel, D. Binosi, R. Thew, and T. Calarco, Quantum Sci. and Tech. 2, 030501 (2017).
[3] M. Yang, and G. R. Fleming, Chem. Phys. 282, 163 (2002).
[4] A. S. Holevo, Quantum Systems, Channels, Information: A Mathematical Introduction, De Gruyter (2012).
[5] K. Ptaszynski, and M. Esposito, arXiv:1901.01093 (2019).
[6] B. Palmieri, D. Abramavicius, and S. Mukamel, J. Chem. Phys. 130, 204512 (2009).
[7] G. Kiršanskas, M. Frančekić, and A. Wacker, Phys. Rev. B 97, 035432 (2018).
[8] G. Lindblad, Comm. Math. Phys. 48, 119 (1976).
[9] V. Gorini, A. Kossakowski, and E.C.G. Sudarshan, J. Math. Phys. 17, 821 (1976).
[10] H. P. Breuer and F. Petruccione, The Theory of Open Quantum Systems (Oxford University Press on Demand, 2007).
[11] P. P. Hofer, M. Perarnau-Llobet, L. D. M. Miranda, G. Haack, R. Silva, J. B. Brask, and N. Brunner, New J. Phys. 19, 123037 (2017).
[12] J. O. Gonzalez, L. A. Correa, G. Nocerino, J. P. Palao, D. Alonso, and G. Adesso, Open Systems & Information Dynamics 24, 1740010 (2017).
[13] G. De Chiara, G. Landi, A. Hewgill, B. Reid, A. Ferraro, A. J. Roncaglia, and M. Antezza, New J. of Phys. 20, 113024 (2018).

[14] A. Levy, and R. Kosloff, Europhys. Lett. 107, 20004 (2014).

[15] D. A. Lidar, Z. Bihary, and K. B. Whaley, Chem. Phys. 268, 35 (2001).

[16] C. Majenz, T. Albash, H. P. Breuer, and D. A. Lidar, Phys. Rev. A 88, 012103 (2013).

[17] D. A. Lidar, arXiv:1902.00967 (2019).

[18] A. Stokes, A. Kurcz, T.P. Spiller, and A. Beige, Phys. Rev. A 85, 053805 (2012).

[19] G. Schaller, and T. Brandes, Phys. Rev. A 78, 022106 (2008).

[20] G. Schaller, P. Zedler, and T. Brandes, Phys. Rev. A 79, 032110 (2009).

[21] J. D. Cresser, and C. Facer, arXiv:1710.09939 (2017).

[22] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, Atom-Photon Interactions: Basic Processes and Applications, Wiley-VCH (2008).

[23] S. Seah, S. Nimmrichter, and V. Scarani, Phys. Rev. E 98, 012131 (2018).

[24] A. G. Redfield, IBM Journal of Research and Development 1, 19 (1957).

[25] M. D. Choi, Can. J. Math. 24, 520 (1972).

[26] A. Jamiołkowski, Rep. Math. Phys. 3, 275 (1972).