Open Quantum Systems Initially Correlated with their Environment

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Most descriptions of open quantum system dynamics assume a product initial state between the system and its environment, as the presence of correlations causes both practical and conceptual difficulties in characterizing the evolution of the open system. In this thesis we study initially correlated states by employing an *operational approach* to open quantum dynamics, which resolves these problems by mapping *initial preparations* to future states. We apply this framework to study the dynamics of systems initially in *global thermal equilibrium* and we distinguish between classical and quantum correlations as classified by the notion of quantum discord. Regarding the former, we solve for the exact dynamics of the spin-boson pure dephasing model and study the effect that initial correlations have on the geometry of the decoherence in the Bloch-sphere, while also presenting an analytical treatment of a general class of pure dephasing models. We then move into the quantum domain and derive a *family of weak-coupling master equations* using second-order perturbation theory in the system-environment coupling. The obtained equation contains a non-negligible inhomogeneous correction term to the conventional *Born-Markov* master equation and is a general theoretical tool that can be applied to many physical models in the weak-coupling regime.

Keywords: Open quantum systems, initial correlations, operational quantum dynamics, dephasing models, master equation.

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Contents

1 Introduction

Understanding the behavior and predicting the dynamics of quantum systems interacting with their environment is critical to the development of many emerging technologies such as quantum metrology [DRC17] and quantum computing [NC01], as in practice no physical system is perfectly isolated from its surroundings. Aiming to predict and control the evolution of open systems calls for methods to effectively describe the evolution of the relevant part only [BP07], since the degrees of freedom composing the environment are typically inaccessible to the experimenter. From a more theoretical point of view, the field of open quantum systems plays an important role in characterizing thermodynamic and stochastic processes in the quantum domain [Str22, MM21], as well as in improving our understanding of the measurement process and the *quantum-to-classical transition* [Zur91, Zur03, Sch07].

One fundamental assumption that is present in most treatments of the dynamics of open quantum systems is that of an initially uncorrelated state between system and environment [BP07, Sch07]. While justified when the interaction begins at a fixed point in time, it is in many situations an unreasonable assumption to make [MSSvL08, DRPP97, MR11]. Accounting for the presence of initial correlations such as quantum entanglement in the global state not only leads to more involved calculations, but more significantly poses conceptual difficulties regarding our ability to characterize the reduced dynamics through well defined mathematical objects. In many cases the dynamical maps describing the evolution of an open system under such circumstances have been found to violate complete positivity [SB01, JSS04], yielding thus quantum states that lack a clear physical interpretation. A fundamental result by Pechukas argued that to account for initial correlations we must either give up linearity or complete positivity [Pec94]. This sparked a long and controversial debate in the field, with many arguing for not-completely positive dynamics and others claiming that dynamical maps under initial correlations are not well defined [Ali95, Pec95]. In more recent years however there has been an increased interest on the topic and a few frameworks to treat this issues have been proposed [Mod12, PSHW19, TSMV21]. In this thesis we will focus on the operational approach to quantum dynamics, introduced by K.Modi in [Mod12], that circumvents the aforementioned problems and results in completely positive dynamics. This is achieved by changing the domain of the map from the set of initial reduced states to the set of possible preparations that can be performed on the system. This framework is applied to study correlations in the global thermal state of the system-bath composite.

This work is organized as follows. In Section 2 we start by briefly introducing the formalism of open quantum systems along with its fundamental mathematical tools, followed by a discussion of the problem of initial correlations and its operational resolution via the formalism of *causal breaks*. Next, classically correlated initial states are explored in Section 3 in the context of pure dephasing models, with a focus on the geometric aspects of the decoherence process in the presence of correlations. In Section 4 we derive a family of perturbative master equations that account for both quantum and classical correlations in the initial state, which is the main result of this thesis. Finally, Section 5 summarizes the results of the previous chapters, while also discussing future outlooks for the project. Supplementary material is included in the Appendix and is referenced at appropriate points throughout the text.

2 Preliminaries

2.1 Open quantum systems

The field of open quantum systems deals with characterizing the dynamics of a physical system S in the presence of a large *environment* or *bath* B, which is assumed to be out of the experimenters control and whose influence is affecting the evolution of the open system non-trivially. It is precisely the presence of an environment that gives rise to the classical world that we experience, where quantum phenomena are strongly suppressed due to effects such as *decoherence* and *dissipation* [Sch07]. Decoherence here is referring to the environmentally induced loss of the distinctive quantum features of a physical system, such as superposition and entanglement, while dissipation is the phenomenon of irreversible loss of energy towards the environment. To model this situation we consider a quantum system composed of two subsystems $\mathcal{H} = \mathcal{H}_S \otimes \mathcal{H}_B$, where $\mathcal{H}_S$ and $\mathcal{H}_B$ stand for the Hilbert spaces of the system and environment respectively. The total S-B evolution is governed by a Hamiltonian of the form

$$H = H_S \otimes \mathbb{1}_B + \mathbb{1}_S \otimes H_B + V_{SB}, \tag{1}$$

where H_S and H_B are the local Hamiltonians and V_{SB} the interaction between the two subsystems. The combined system can always be assumed to be *closed*, and therefore its dynamics are governed by the Liouville–von Neumann equation, $\partial_t \varrho_{SB}(t) = -i[H, \varrho_{SB}(t)]$, where we have assumed units with $\hbar = 1$. The reduced state of each subsystem is obtained by tracing out the complementary degrees of freedom, i.e. $\varrho_S = \mathrm{Tr}_B(\varrho_{SB})$ and $\varrho_B = \mathrm{Tr}_S(\varrho_{SB})$ for system and bath respectively. The evolution of any closed system is unitary and given by

$$\varrho_{SB}(t) = U(t)\varrho_{SB}(0)U^\dagger(t), \tag{2}$$

where $U(t) = e^{-iHt}$ is the unitary evolution operator. Unitary evolution is *time-local*, as the future evolution of the system only depends on the current state of the system. The same cannot be said for the reduced system dynamics, whose behavior is often times a lot more intricate and requires different mathematical tools.

Dynamical maps and master equations

As already discussed, the main focus in the field of open quantum systems is to determine the evolution of the system S, while assuming no control over the environment. In mathematical terms, the task at hand is to determine the *dynamical map* $\mathcal{E} : \mathcal{B}(\mathcal{H}_S) \to \mathcal{B}(\mathcal{H}_S)$ that governs the evolution of ϱ_S, i.e. $\varrho_S(t) = \mathcal{E}[\varrho_S(0)]$. Here $\mathcal{B}(\mathcal{H}_S)$ denotes the space of bounded operators on $\mathcal{H}_S$. The reduced state of the open system can be obtained from eq. (2) by tracing out the environment and reads $\varrho_S(t) = \mathrm{Tr}_B\left(U(t)\varrho_{SB}(0)U^\dagger(t)\right)$. If the system at time $t = 0$ is decorrelated from the environment, i.e. $\varrho_{SB}(0) = \varrho_S(0) \otimes \varrho_B(0)$, then the dynamical map $\mathcal{E}$ is well defined and reads

$$\mathcal{E}[\bullet] = \mathrm{Tr}_B\left(U(t) \bullet \varrho_B(0)U^\dagger(t)\right). \tag{3}$$

Note that the dynamical map only depends on the global unitary evolution and the state of the environment and not on the state it acts upon. As we shall discuss in more detail later, this procedure is no longer applicable for initially correlated states, i.e. when $\varrho_{SB}(0) \neq \varrho_S(0) \otimes \varrho_B(0)$.

The dynamical map as introduced in this section is an exact description of the reduced state dynamics, however it is often impossible to determine in practice. This is largely due

to the lack of control over the environmental degrees of freedom. An alternative (but equivalent) approach, which proves to be especially useful when dealing with approximations, is that of the *quantum master equation*. The most general form of the master equation is

$$\partial_t \varrho_S(t) = -i[H, \varrho_S(t)] + \int_0^t dt' \mathcal{K}(t, t')[\varrho_S(t')], \tag{4}$$

where the first term accounts for the unitary part of the evolution and $\mathcal{K}(t, t')$ is the *memory kernel* [RH12]. The memory kernel accounts for the fact that the environment in general keeps track of the previous interactions with the system. If the contribution of the memory kernel can be argued to be negligible, then future states only depend on the current state of the system and not on an integral over the history of past states $\{\varrho_S(t')\}_{t'=0}^t$, i.e. they are local in time. Such master equations are often being referred to as *Markovian*, however there is a lot of debate regarding the definition of Markovianity in the quantum domain and whether a master equation, which only describes two-time correlations, can even be called Markovian or not[1]. We refrain therefore from using this terminology and refer the reader to [Str22, MM21] for a detailed analysis on the topic. Master equations are widely used in various field of physics, such as quantum optics, condensed matter physics, quantum thermodynamics and many more [BP07, Sch07, Str22, GAFCT10].

Before proceeding to introduce the main topic of this thesis, let us stress that any dynamical map or master equation describing the dynamics of an open system should yield valid quantum states, i.e. they must be trace preserving and guarantee the positivity of $\varrho_S(t)$ for all times, a property known as *complete positivity*. In this context, a quantum map $\Phi_S \in \mathcal{B}(\mathcal{B}(\mathcal{H}_S))$ is said to be completely positive (CP) if

$$\Phi_S \otimes \mathcal{I}_A \varrho_{SA} \geq 0, \text{ for all } \varrho_{SA} \geq 0 \tag{5}$$

Here $\mathcal{I}_A$ denotes the identity in $\mathcal{B}(\mathcal{B}(\mathcal{H}_A))$, where $\mathcal{H}_A$ is an arbitrary ancillary Hilbert space. Finally, we note that the map $\mathcal{E}$ defined in eq. (3) is CP, as a concatenation of CP maps (unitaries and partial trace are both CP).

2.2 The problem of initial correlations

Let us now consider a correlated initial state, which can always be decomposed as $\varrho_{SB}(0) = \varrho_S(0) \otimes \varrho_B(0) + \chi_{AB}(0)$, where χ_{AB} accounts for the correlations and is not a quantum state. Following the same line of thought as in the previous chapter, its evolution can be written as

$$\varrho_S(t) = \tilde{\mathcal{E}}[\varrho_S(0)] = \mathcal{E}[\varrho_S(0)] + \mathcal{J}_{\chi_{AB}}, \tag{6}$$

where $\mathcal{J}_{\chi_{AB}} = \text{Tr}_B\left(U(t)\chi_{AB}U^\dagger(t)\right)$ and $\mathcal{E}$ is defined in (3). Clearly the map $\tilde{\mathcal{E}}$ may no longer be independent of the reduced state of the system. Thus, it does not constitute a map from initial to future states. Furthermore, despite $\mathcal{E}$ being CP, the same is not guaranteed for $\mathcal{J}_{\chi_{AB}}$, leading thus to a potentially not-completely positive (NCP) dynamical map $\tilde{\mathcal{E}}$. This is because for $\varrho_S(0)$ and $\varrho_B(0)$ to be positive, not any χ_{AB} is allowed, restricting thus the domain of the map. A final caveat with this approach is the operational meaning of the map $\tilde{\mathcal{E}}$. In an experimental scenario where one wants to determine $\tilde{\mathcal{E}}$ via *process tomography* [MS10] by letting it act upon a complete set of basis elements of $\mathcal{B}(\mathcal{H}_S)$, the correlation term χ_{AB} would have to be fixed for each input. Given however our lack of

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[1]Markovianity as defined in probability theory refers to *multitime* processes and not just two-point correlations.

control over the environment, there is no operational mechanism to fix these system-bath correlations.

In general, defining a dynamical map $\tilde{\mathcal{E}} : \mathcal{B}(\mathcal{H}_S) \to \mathcal{B}(\mathcal{H}_S)$ is not meaningful for entangled initial states. This can be seen by considering a pair of global initial states $\varrho_{SB}^1(0) \neq \varrho_{SB}^2(0)$ such that $\varrho_S^1(0) = \varrho_S^2(0)$ and $\varrho_B^1(0) = \varrho_B^2(0)$. Their respective reduced states evolve under $\varrho_S(t) = \text{Tr}_B \left(U_{SB}(t) \varrho_{SB}(0) U^\dagger(t) \right)$ and clearly satisfy $\varrho_S^1(t) \neq \varrho_S^2(t)$. This would mean that $\tilde{\mathcal{E}}$ has two distinct outputs for the exact same input, making it thus an *ill-defined* map. Figure 1 depicts the problem of initial correlations schematically.

Overall, we have established that the approach employed to describe the dynamics of initially uncorrelated states is not applicable anymore, as it raises significant questions regarding the positivity of the open system dynamics and the meaning of dynamical maps in this context. In the following chapter we present an operational framework, originally introduced by K.Modi in [Mod12], that resolves these issues by paying proper attention to the effect that *preparations* have on the system dynamics in the presence of system-environment correlations [Mod11]. An extensive review of operational quantum dynamics and its mathematical tools can be found in [MPM17].
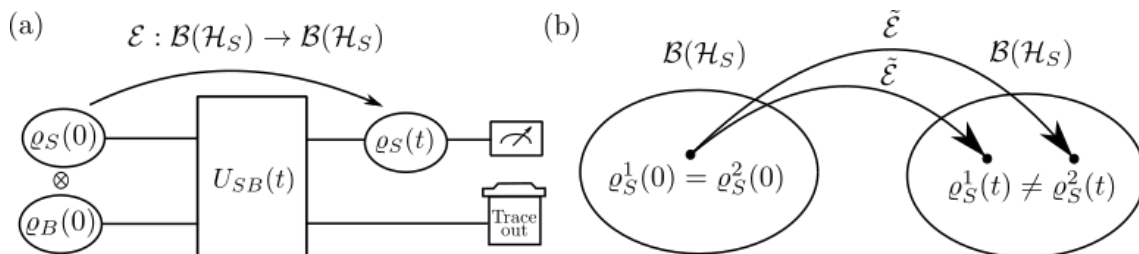

Figure 1: The problem with initial correlations: (a) A dynamical map for the open system is well defined for initially uncorrelated states. (b) In the presence of initial entanglement a dynamical map is ill defined

2.3 Operational solution to the initial correlation problem

Let us consider a preparation $\mathcal{P} \in \mathcal{B}(\mathcal{B}(\mathcal{H}_S))$ acting on the open system such that the global state afterwards is disentangled, i.e. $\varrho_{SB}(0^+) = \mathcal{P} \varrho_{SB}(0) = \varrho_S(0^+) \otimes \varrho_B(0^+)$. Note here that $\mathcal{P}$ is a *superoperator* mapping operators onto operators. Throughout this thesis we denote superoperators by calligraphic capital letters to distinguish them from 'regular' operators that map vectors onto vectors[2]. A naive approach would be to claim that the system-bath composite can always be initialized in such a product state, whose dynamics are then governed by the dynamical map $\mathcal{E}_\mathcal{P}[\bullet] = \text{Tr}_B \left(U(t) \bullet \varrho_B^\mathcal{P}(0^+) U^\dagger(t) \right)$. The issue here is that the state $\varrho_B^\mathcal{P}(0^+)$ depends on the applied preparation and each of the infinite preparations that can be applied on S could lead to a different dynamical map. This approach hints at the solution, by realizing that a preparation applied on the open system has a clear operational meaning, as it is entirely on the control of the experimenter. This motivates the idea of changing the domain of the map from the space of initial states to the space of preparations acting on them. We consider therefore a map of the form

$$
\begin{aligned}
\mathcal{M} &: \mathcal{B}(\mathcal{B}(\mathcal{H}_S)) \to \mathcal{B}(\mathcal{H}_S), \\
\varrho_S(t) &= \mathcal{M}[\mathcal{P}] \varrho_{SB}(0).
\end{aligned}
\tag{7}
$$

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[2]From a mathematical perspective this distinction is not necessary, it helps however in keeping track of the spaces that different objects act upon.

To account for the infinity of possible preparations $\mathcal{P}$ that can be performed on a quantum system, a basis spanning the entire space of operations is required. A natural choice here is a basis consisting of *decorrelating* elements, i.e. preparations whose output is a product state.

Causal breaks

At this point it is convenient to introduce the concept of *causal breaks*, which will form the aforementioned basis of $\mathcal{B}(\mathcal{B}(\mathcal{H}_S))$. A causal break $\mathcal{C}_{ab}$ is an *intervention* in which the state of the system is measured with an *informationally complete* set of projectors $\{\Pi_b\}$[3] and then prepared in a new state τ_a that is independent of the measurement result and of the previous state of the system. This ensures the absence of correlations between the current state the system and its environment, as well as with its previous states. Their effect on a state is given by

$$\mathcal{C}_{ab}\varrho_{SB} = \tau_a \otimes \mathrm{Tr}_S(\Pi_b \varrho_{SB}) \equiv \tau_a \otimes \eta_b, \tag{8}$$

where η_b is the conditional state of the environment. Note that the states η_b are not normalized and their trace is the outcome probability of the associated measurement Π_b. Formally, a causal break is a *quantum instrument* and for $\tau_a = |\psi_a\rangle\langle\psi_a|$ and $\Pi_b = |\psi_b\rangle\langle\psi_b|$ is defined as

$$\mathcal{C}_{ab}[\bullet] = |\psi_a\rangle\langle\psi_b| \bullet |\psi_b\rangle\langle\psi_a| . \tag{9}$$

If both $\{\tau_a\}$ and $\{\Pi_b\}$ form a basis of $\mathcal{B}(\mathcal{H}_S)$, then so does their cross-combination on the space of quantum instruments, $\mathcal{B}(\mathcal{B}(\mathcal{H}_S))$. Any preparation (and any map in general) can then be linearly expanded as

$$\mathcal{P} = \sum_{ab} \gamma_{ab}\mathcal{C}_{ab}, \ \ \gamma_{ab} \in \mathbb{C}. \tag{10}$$

Additionally, if $\mathcal{P}$ is hermiticity preserving, then it is straightforward to verify that the coefficients γ_{ab} must be are real, while trace preservation implies $\sum_{ab} \gamma_{ab} = 1$. Note finally that causal breaks are superoperators, and as such can be represented in various ways. An example of such a representation that can be used to determine the coefficients γ_{ab} for a given $\mathcal{P}$ is given in the Appendix A. A more thorough analysis on causal breaks and their role in characterizing quantum stochastic process is given in [PRRF+18, Str22].

Mapping initial preparations to final states

With the introduction of the causal break we can now resolve the initial correlation problem by defining the *superchannel* or $\mathcal{M}$-map

$$\mathcal{M}[\bullet] = \mathrm{Tr}_B \left(U(t)(\bullet \otimes \mathcal{I}_B)\varrho_{SB}(0)U^\dagger(t) \right), \tag{11}$$

which maps initial preparations $\mathcal{P}$ to future system states

$$\varrho_S(t) = \mathcal{M}[\mathcal{P}]\varrho_{SB}(0). \tag{12}$$

The $\mathcal{M}$-map is operationally well-defined, as it captures all the inaccessible parameters of the system, while its input is entirely in the control of the experimenter. It has been shown in [Mod12] to be both linear and completely positive, circumventing thus Pechukas

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[3]By informationally complete is meant that $\{\Pi_b\}$ is a basis of $\mathcal{B}(\mathcal{H}_S)$.

argument for dropping one of the two. Note here that if $\mathcal{P}$ is trace preserving, then so is $\mathcal{M}$.

Expressing now the preparation in the basis of causal breaks, the problem reduces to solving for the open dynamics of an initially uncorrelated system-bath composite for the d^4 *different initial states* $\varrho_{SB}^{ab}(0^+) = \tau_a(0) \otimes \eta_b(0)$, where $a, b \in \{0, 1, ..., d^2\}$. The respective solutions $\tau_{ab}(t) = \mathcal{M}(\mathcal{C}_{ab})\tau_a(0)$ can then be determined via a dynamical map, an appropriate master equation or any other suitable theoretical or numerical method [Sch14, Ing02, CC14]. The solution for the correlated initial state is then simply given by the linear combination

$$\varrho_S(t) = \sum_{ab} \gamma_{ab}\tau_{ab}(t). \tag{13}$$

This reduction of the problem into a set of problems involving factorisable initial states allows for the extension of existing theoretical and numerical methods into the regime of arbitrary initial correlations. A schematic representation of this approach is given in Fig. 2.
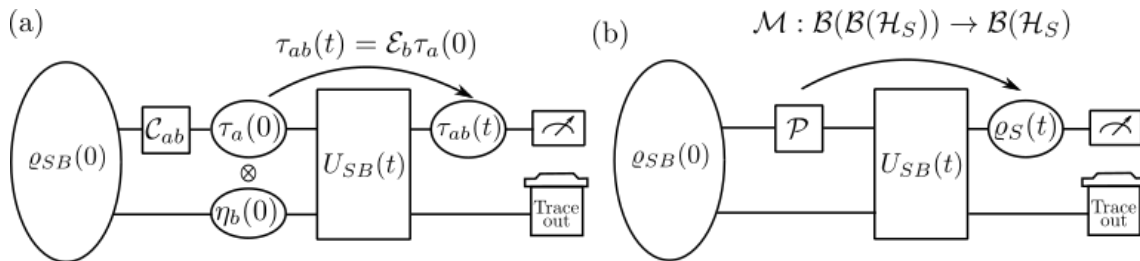

Figure 2: Schematic representation of the operational solution to the problem. (a) A causal break initializes the system in a product state, whose dynamics can be determined via existing methods. (b) Any preparation can be expressed as a linear combination of causal breaks and therefore be mapped via $\mathcal{M}$ onto future states of the system.

In the following section we will apply this formalism to study the effect that classically correlated initial states have on the open system dynamics and how it compares to the case of an initial product state. Throughout this work we utilize quantum discord to distinguish between classical and quantum correlations, an information measure introduced by Ollivier and Zurek in 2001 [OZ01]. Briefly speaking, quantum discord accounts for the fact the even separable states can contain non-classical correlations. This is a consequence of the disruptive nature of measurements in quantum mechanics, which means that even non-entangled bipartite quantum states can contain information that is not accessible via local measurements only. In that sense, zero-discord states are said to be classically correlated. A more detailed discussion on quantum discord can be found in the Appendix B. In Section 4 the $\mathcal{M}$-map formalism will be applied for deriving a set of master equations that account for any type of correlations in the initial state, quantum or classical. Throughout this thesis we focus on the global *thermal* or *Gibbs state*

$$\varrho_{SB}(0) = e^{-\beta H}/Z, \tag{14}$$

where $Z = \text{Tr}(e^{-\beta H})$ is the partition function of the full system and $\beta = 1/T$ is the inverse temperature in units where $k_B = 1$. The thermal state is stationary and is the state that maximizes the von-Neumann entropy $S(\varrho) = -\text{Tr}(\varrho \ln \varrho)$ for a fixed energy $E = \text{Tr}(H\varrho)$. A derivation of its form given in eq.(14) can be found in [Jay57].

3 Classically correlated initial states: Pure dephasing models

3.1 General dephasing models

In this section we study the reduced dynamics for the general class of *pure dephasing models* under the assumption of a correlated initial state. Dephasing models satisfy the condition $[H_S \otimes \mathbb{1}_B, V_{SB}] = 0$ and are therefore characterized by the fact that the energy of the open system is a constant of motion. The effect of the environment in the reduced dynamics is thus purely decoherent, with no energy dissipation taking place, as the populations remain thus constant throughout the evolution of the open system. The Hamiltonian describing this class of models is given by

$$H = H_S \otimes \mathbb{1}_B + \mathbb{1}_S \otimes \sum_k H_k + S \otimes \sum_k B_k, \tag{15}$$

for all H_S, S such that $[H_S, S] = 0$. Here H_k stand for the Hamiltonian of the k-th bath mode, which are assumed to be non-interacting. In this work we will focus in the initial correlations contained in the global thermal state (14). One characteristic feature of this class of models is that their thermal state only contains *classical correlations* between the system and the environment. A proof of this statement can be found in the Appendix C. Moreover, application of a causal break on that stationary state will result in a non-equilibrium state with non-trivial dynamics, which we aim to characterize and compare with the case of an initially uncorrelated state.

The pure dephasing Hamiltonian can be rewritten in the following block-diagonal form

$$H = \sum_m \omega_m \, |m\rangle\langle m| \otimes \left(\mathbb{1}_B + H_B + s_m \sum_k B_k \right) \equiv \sum_m |m\rangle\langle m| \otimes H_B^m. \tag{16}$$

This simple structure of the Hamiltonian allows for some analytical progress regarding the evolution of the reduced state, without further assumptions on the form of the interaction. However, before introducing these more general results, let us first analyze the dynamics of the *exactly solvable* spin-boson pure dephasing model.

3.2 Spin-boson pure dephasing model

This model describes the interaction between a two-level system and a bath consisting of harmonic oscillators. The total Hamiltonian[4] reads

$$H = H_S + H_B + V_{SB} = \frac{\omega_0}{2}\sigma_z + \sum_k \omega_k b_k^\dagger b_k + \sigma_z \otimes \sum_k (g_k b_k^\dagger + g_k^* b_k), \tag{17}$$

where ω_0 is the energy gap between the two energy levels of the system, ω_k the frequency of the k-th bath mode and $b_k^\dagger, b_k$ are the *bosonic* creation and annihilation operators respectively. Pure dephasing model are widely used to study noise in many physical systems, such as quantum computers [PSE96, RQJ02] and impurities in crystals [Ski88].

Assuming a product initial state $\bar{\varrho}_{SB}(0) = \tau \otimes \varrho_B^{th}$, where $\varrho_B^{th} = e^{-\beta H_B}/Z_B$ is the local thermal state of the bath, the evolution of the open system is given by

$$\langle i | \, \tau \, | i \rangle \, (t) = \langle i | \, \tau \, | i \rangle \, (0), \quad i = \{0, 1\} \tag{18}$$

$$\langle i | \, \tau \, | j \rangle \, (t) = e^{-\Gamma(t)} \langle i | \, \tau \, | j \rangle \, (0), \quad i \neq j, \tag{19}$$

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[4]We suppress from now on tensor products with the identity for clarity of notation.

where $r_{uc}(t) = e^{-\Gamma(t)}$ is the *uncorrelated decoherence factor* and

$$\Gamma(t) = 8 \sum_k \frac{|g_k|^2}{\omega_k^2} \sin^2(\omega_k t/2) \coth(\beta \omega_k/2) \tag{20}$$

the *decoherence rate*. As expected, we observe stationary populations and exponentially decaying coherences. This factor can be split up in two terms,

$$\Gamma(t) = \Gamma_{vac}(t) + \Gamma_{therm}(t), \tag{21}$$

in order to distinguish between decoherence caused by thermal excitations in the bath and by vacuum fluctuations. A detailed analysis on this topic can be found in [Sch07].

Let us now focus on the correlated, global thermal state $\varrho_{SB}(0) = e^{-\beta H}/Z$ instead and introduce a causal break $\mathcal{C}_{ab}$, such that the state of the combined system is given by

$$\varrho_{SB}^{ab}(0^+) = \sum_{ab} \gamma_{ab} \mathcal{C}_{ab} \varrho_{SB}(0) \equiv \tau_a \otimes \eta_b, \tag{22}$$

where

$$\eta_b = \mathrm{Tr}_S \left(\Pi_b \varrho_{SB}(0) \right) = \frac{1}{Z_{SB}} \langle \psi_b | e^{-\beta H} | \psi_b \rangle. \tag{23}$$

is the *conditional state of the bath* after the causal break and $\Pi_b = |\psi_b\rangle\langle\psi_b|$. The block-diagonal form (16) can be utilized to diagonalize the Hamiltonian and determine η_b, which characterizes the exact dynamics induced on the system by $\mathcal{C}_{ab}$[5]. Clearly the populations will still be stationary, as this is a consequence of $[H_S, V_{SB}] = 0$ and does not depend on the initial state. After several algebraic manipulations and a change of variables to identify the effective bath Hamiltonians

$$H_B^m \equiv H_B^{\pm} = \pm \frac{\omega_0}{2} + \sum_k \omega_k b_k^\dagger b_k \pm \frac{1}{2} \sum_k (g_k b_k^\dagger + g_k^* b_k) \tag{24}$$

in the decomposition (16) as a pair *shifted harmonic oscillators*, the coherences $\langle i| \tau_{ab} |j\rangle (t) = \mathrm{Tr}\left(|j\rangle\langle i| \varrho_{SB}^{ab}(t)\right)$ are found to be

$$\langle i| \tau_{ab} |j\rangle (t) = (\cos\theta(t) + iK_b \sin\theta(t)) e^{-\Gamma(t)} \langle i| \tau_a |j\rangle (0). \tag{25}$$

Above we have introduced the time-dependent *correlation phase*

$$\theta(t) = \sum_k \frac{|g_k|^2}{\omega_k^2} \sin(\omega_k t) \tag{26}$$

and the asymmetry factor

$$K_b = \frac{f_0 e^{-\beta\omega_0/2} - f_1 e^{\beta\omega_0/2}}{f_0 e^{-\beta\omega_0/2} + f_1 e^{\beta\omega_0/2}}, \tag{27}$$

where $f_m = \langle m| \Pi_b |j\rangle\langle i| \Pi_b |m\rangle$, $m = 0, 1$. We should note here that $|K_b| \leq 1$ for all causal breaks, indicating that the absolute value of the coherence in the open system can not surpass at any time the respective value of the uncorrelated case. This is due to the classical nature of the initial correlations, as the presence of entanglement in the initial state can lead to information backflow and generally more complex behavior [SBPV10].

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[5]Note that the global state prior to the causal break satisfies $[\varrho_{SB}(0), H] = 0$ and is a steady state of the model.

Additionally, the effect of each causal break on the decoherence rate is only present in the asymmetry factor K_b and not in the time-dependent correlation phase, leading thus to a straightforward generalization for the case of a general preparation $\mathcal{P} = \sum_{ab} \gamma_{ab} \mathcal{C}_{ab}$. The evolution of the coherences in that case reads

$$\varrho_S^{ij}(t) = \sum_{ab} \gamma_{ab} r_c^b(t) \langle i | \tau_a | j \rangle (0), \tag{28}$$

where we have defined the correlated decoherence factor,

$$r_c^b(t) = \left(\cos \theta(t) + i K_b \sin \theta(t) \right) e^{-\Gamma(t)}. \tag{29}$$

It can be seen that more complex preparations can alter the plane and the amplitude of the oscillations in the decoherence factor, but not its frequency, which is determined from the mode-dependent coupling strength g_k and the the respective frequencies ω_k. Following a similar approach as in eq. (21), an additional term can be introduced to account for the impact of the initial correlations on the decoherence rate for various forms of the system-bath coupling [MMR12]. Instead, we will focus on the observed effect of the *asymmetric decay of the Bloch-vector*, which to our knowledge has not been explored previously. This effect be seen directly in eq. (25) in the presence of an imaginary part in the decoherence factor and is the focus of the following section.

3.3 Geometry of decoherence in the spin-boson pure dephasing model

To gain a more clear physical understanding of this effect, let us express the density matrix as $\tau_{ab}(t) = (\mathbb{1} + \mathbf{f}(t) \cdot \boldsymbol{\sigma})/2$, where $\mathbf{f}(t) = (f_x(t), f_y(t), f_z(t))$ is the *Bloch vector* and $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)^T$ is the vector of Pauli matrices. It is straightforward to compute the expressions for the components of the Bloch-vector as a function of time, which are given by

$$f_x(t) = 2 \cos \theta(t) \langle 0 | \tau_{ab} | 1 \rangle (0) \tag{30}$$

$$f_y(t) = -2 K_b \sin \theta(t) \langle 0 | \tau_{ab} | 1 \rangle (0) \tag{31}$$

$$f_z(t) = 2 \langle 0 | \tau_{ab} | 0 \rangle (0) - 1. \tag{32}$$

Let us now consider a preparation consisting of a single causal break of the form

$$\tilde{\mathcal{P}} \varrho_{SB}(0) = \tau \otimes \text{Tr}_S(\tau \varrho_{SB}(0)), \tag{33}$$

where the system is projected onto $\tau = |\psi\rangle\langle\psi|$, followed by a preparation of the same state. It is the simple structure of the generalization (29) that justifies our decision to focus on that simple case moving forward. The evolution of the Bloch vector components for both product and correlated initial states shows a major difference in the geometry of the decay of the Bloch-vector. While for the uncorrelated case it always decays along its projection to the z-axis following a straight line, in the presence of correlations it experiences an 'oscillations'[6] along the plane x-y plane, before reaching the same steady state. This demonstrates that the initial correlations lead to an asymmetric decay of the Bloch vector, with coherences emerging in directions that were not present in the initial state. An example for two different coupling strengths ($\alpha = \{1, 6\}$) is shown in Fig. 3(a), while Fig. 3(b) shows the effect that initial correlations have in the the decoherence factor.

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[6]They are not oscillations in the strict sense, as $K_b \neq 1$ in general.
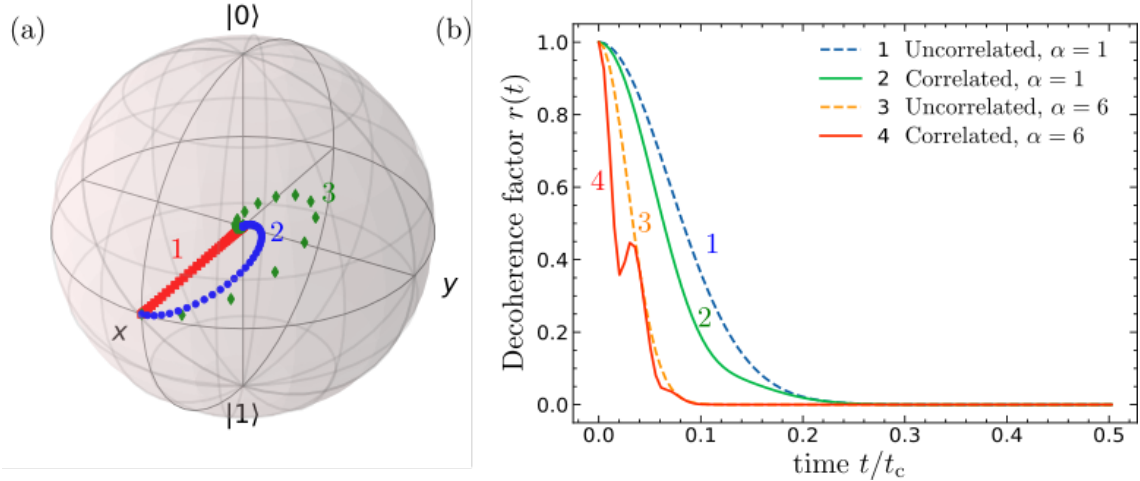
Figure 3: (a) Geometry of decoherence in the Bloch-sphere for (1) initial product state (red-squares), (2) correlated initial state, $\alpha = 1$ (blue-circles), (3) correlated initial state, $\alpha = 6$ (green-rhombus) (b) Decoherence factor for product (dashed lines) and correlated (solid lines) initial states. Plot parameters: $\tau = |+\rangle\langle+|$, $\omega_0 = 2 \cdot 10^{-2}\kappa$, $\beta = 50/\kappa$, $J_0 = 13$, $t_c = \beta/8\pi\kappa$.

Note here that for all plots we have assumed a continuous spectrum for the environment, allowing us to perform the transformation $\sum_k |g_k|^2 \longrightarrow \int_0^\infty d\omega J(\omega)$, where we have introduced the *spectral density*

$$J(\omega) = \sum_k |g_k|^2 \delta(\omega - \omega_k). \tag{34}$$

The spectral density is a positive function defined for $\omega > 0$ that completely characterizes the coupling between a system and its environment and satisfies $J(\omega) \to 0$ for $\omega \to 0$ and $\omega \to \infty$. For all results in this section we have assumed a spectral density that of the form $J(\omega) = 8J_0\omega e^{-\omega/\kappa}$, where J_0 is a dimensionless constant. This spectral density is *ohmic* for small ω, i.e. $J(\omega) \propto \omega$ and has a smooth cutoff, quantified by the parameter κ.

Before proceeding with generalizing and interpreting this phenomenon, let us stress here that this asymmetric decay is not noticeable if the applied preparation satisfies $[\mathcal{P}\varrho_{SB}(0), H] = 0$, as in that case the the decay of the Bloch vector will follow a linear trajectory towards the steady state without exhibiting any assymetries. Finally, if the applied measurement results in $K_b = 1$, then the correction to the decoherence factor due to the initial correlations would correspond to a full rotation, i.e. $e^{i\theta(t)}$. This could have potentially led to a misinterpretation of this phenomenon as being part of the systems unitary dynamics, leading to wrong estimations of the frequency ω_0 of the qubit. A very brief qualitative analysis of this idea in the context of *Ramsey interferometry* [Ram50] is included in the Appendix D. In the following we analyze this phenomenon for the general class of dephasing models, while also providing a physical intuition on its origin.

3.4 Geometry of decoherence in general dephasing models

In this chapter we extend our analysis on the geometric aspects of decoherence onto the broader category of dephasing models characterized by the Hamiltonian (15). To do so, we once again consider a preparation consisting of a single causal break as in (33). The global state is then given by $\varrho_{SB}(0^+) = \tau(0) \otimes \eta(0)$, where

$$\eta(0) = \frac{1}{Z} \sum_m |\langle\psi|m\rangle|^2 e^{-\beta H_B^m} = \frac{1}{Z} \sum_m |\langle\psi|m\rangle|^2 e^{-\beta\omega_m} e^{-\beta(H_k + s_m B_k)} \tag{35}$$

13

is the conditional state of the bath. The matrix elements of the reduced system evolve then according to

$$\varrho_S^{ij}(t) = \langle i|\tau|j\rangle\,(0)\frac{1}{Z}\sum_m |\langle\psi|m\rangle|^2 e^{-\beta\omega_m}\prod_k \mathrm{Tr}_B\left(\tilde{U}_i e^{-\beta(H_k + s_m B_k)}\tilde{U}_j^\dagger\right), \qquad (36)$$

where $\tilde{U}_i = \mathcal{T}_\leftarrow \exp\left[-is_i\int_0^t dt'\,B(t')\right]$ and s_i are the eigenvalues of the system operator S. A step-by-step derivation of this exact result is included in the Appendix E. Overall, we observe a similar but more complicated structure in comparison to the spin-boson model, which is a special case of the general model (15). In the following we analyze the origins of the asymmetric decay of the Bloch vector by highlighting the differences between the initial bath states $\eta(0)$ and ϱ_B^{th}.

Interpretation

It is precisely the partition of the conditional bath-state (35) into a *mixture of thermal states*, each with an effective Hamiltonian H_B^m, that causes the asymmetry observed in the decay of the coherences. Furthermore, the choice of projection $\Pi = |\psi\rangle\langle\psi|$ determines the contribution that each of the effective thermal states $e^{-\beta(H_B^m)}/Z$ has in the total mixture. The spacing between the energy levels of the open system determines the constant energy shift between each state in the partition of conditional bath states, while the eigenvalues s_m of the system operator S in the common basis between S and H_S determine the *energy spacing* between the modes in each effective Hamiltonian. In the case of the spin-boson pure dephasing model studied previously, the former contributions are encoded solely in the stationary K-factor, while the latter are encoded in the time-dependent phase $\theta(t)$, having a more significant impact in the evolution of the coherences. As an analogy, the observed asymmetry in the decay of the Bloch-factor can be thought of as a consequence of the system interacting probabilistically with multiple distinct baths, each one 'pulling' the Bloch-vector towards a different direction. This concept is illustrated schematically in Figure 4 for the case of the spin-boson model.
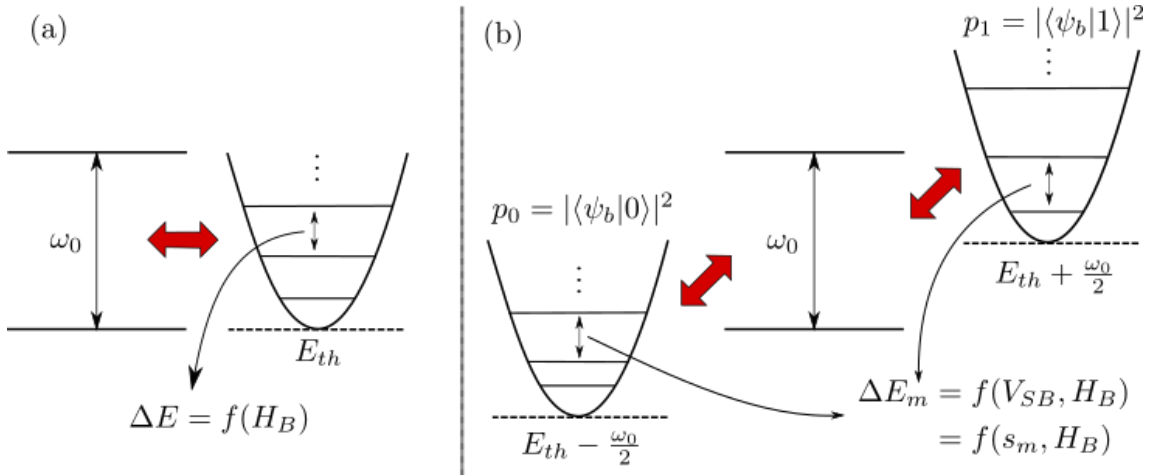
Figure 4: Schematic representation of the process leading to the observed geometry of decoherence in the Bloch-sphere for (a) product initial state: the qubit is interacting with a single bath (b) Correlated initial state, the qubit is interacting with a statistical mixture of two energy-shifted baths generated by different effective Hamiltonians H_B^m, resulting thus in different energy spacing ΔE for each of the respective spectra.

4 A family of weak-coupling master equations for initial correlations

In this section we treat a fully general Hamiltonian describing a system S weakly coupled to its environment B and analyze the reduced system dynamics by deriving a second-order master equation that accounts for possible initial correlations (quantum or classical) between system and environment. More specifically, in order to properly account for initial correlations we are deriving a *family of master equations* $\{\mathcal{ME}\}_{ab}$, one for each causal break $\mathcal{C}_{ab}$ in the set that spans the space of possible preparations in S. One master equation is derived for each *decorrelated* state $\varrho_{SB}^{ab}(0^+) = \mathcal{C}_{ab}\varrho_{SB}^{ab}(0) = \tau_a(0) \otimes \eta_b(0)$. We show that this family of master equations contains an inhomogeneous term that depends on the initial state of the system. For a given preparation $\mathcal{P} = \sum_{ab}\gamma_{ab}\mathcal{C}_{ab}$, the state of the system at time t is then given by

$$\varrho_S(t) = \sum_{ab} \gamma_{ab}\tau_{ab}(t), \tag{37}$$

where $\tau_{ab}(t)$ is the solution of the corresponding equation $\{\mathcal{ME}\}_{ab}$. For the sake of clarity we will first consider a *single* master equation and switch to the causal break formulation of the problem once the structure of the master equation has become clear.

4.1 Setting up the problem

Starting with a general Hamiltonian $H = H_S + H_B + \lambda V_{SB}$, where λ accounts for the strength of the interaction, we move into the interaction picture with respect to $H_0 = H_S + H_B$, thus writing $\tilde{\varrho}_{SB}(t) = e^{i(H_S+H_B)t}\varrho_{SB}e^{-i(H_S+H_B)t}$. The combined system-environment evolution is then given by $\partial_t\tilde{\varrho}_{SB}(t) = -i\lambda[\tilde{V}_{SB}(t), \tilde{\varrho}_{SB}(t)]$. After integrating both sides, iteratively inserting the result back to the equation for the combined state evolution and tracing out the environmental degrees of freedom we obtain

$$\partial_t\tilde{\varrho}_S(t) = -i\lambda\mathrm{Tr}_B\left([\tilde{V}_{SB}(t), \varrho_{SB}(0)]\right) - \lambda^2\mathrm{Tr}_B\left(\left[\tilde{V}_{SB}(t), \int_0^t dt'[\tilde{V}_{SB}(t'), \tilde{\varrho}_{SB}(t')]\right]\right), \tag{38}$$

an integrodifferential equation for the reduced state $\tilde{\varrho}_S$ which is still formally exact. This implies that knowledge of the whole history of states $\{\varrho_S(t')\}_{t'=0}^t$ is required for the equation to be solved, making it thus a *time non-local* differential equation. This is practically impossible and indicates that approximations have to be made to proceed further with the task of characterizing the reduced dynamics. Additionally, the evolution of the system depends on the state of the environment at those times too, something incompatible with the spirit of open quantum systems, where one assumes no access the environment.

The standard procedure at this point is to assume no initial correlations, i.e. $\varrho_{SB}(0) = \varrho_S(0) \otimes \varrho_B(0)$, with the bath being in thermal equilibrium, and invoke the Born, first and second Markov approximations, as well as the secular approximation to arrive at a *time-local*, first-order differential equation for ϱ_S [KCK08]. Instead, we will assume global thermal equilibrium, i.e.

$$\varrho_{SB}(0) = \varrho_{SB}^{th} = e^{-\beta(H_0+\lambda V_{SB})}/Z_{SB}, \tag{39}$$

where $Z_{SB} = \mathrm{Tr}\left(e^{-\beta(H_0+\lambda V_{SB})}\right)$ is the partition function of the combined system, and perform second-order perturbation theory in the coupling strength λ to investigate the effect of the initial correlations in the structure of the weak coupling master equation. In general the state (39) is not of product form in the presence of system-bath interaction. The type of the interaction determines whether the correlations contained in it are of classical or quantum nature. Along the way we will not perform the *second Markov* and *secular approximations*, for reasons that will be clarified at appropriate points in the derivation.

First order-correction to the global Gibbs state

Let us begin with the first order approximation of the global Gibbs state, by making use of the identity[7]

$$e^{tX} = e^{tY} + \int_0^t dt' e^{(t-t')Y}(X-Y)e^{t'X}. \tag{40}$$

Substituting now $X = -H_0 - \lambda V_{SB}$, $Y = -H_0$ and $t = \beta$ and then utilizing the exact same identity again for the factor $e^{t'X}$ results in

$$e^{-\beta H} = e^{-\beta H_0} - \lambda \int_0^\beta d\beta' e^{-(\beta-\beta')H_0}V_{SB}e^{-\beta'H_0} + \mathcal{O}(\lambda^2). \tag{41}$$

We expand the global Gibbs state in a Taylor series up to first order in the coupling-strength, i.e.

$$\varrho_{SB}(0) = \varrho_S^{th} \otimes \varrho_B^{th} + \lambda\varrho_{corr} + \mathcal{O}(\lambda^2), \tag{42}$$

since a first order approximation in the initial state leads to second order terms in the master equation (38). Here $\varrho_S^{th} = e^{-\beta H_S}/Z_S$ and $\varrho_B^{th} = e^{-\beta H_B}/Z_B$ are the *local* equilibrium states. For the first order correction we obtain

$$\varrho_{corr} = -\frac{1}{Z_0}\int_0^\beta d\beta' e^{-(\beta-\beta')H_0}V_{SB}e^{-\beta'H_0} + \frac{\mathrm{Tr}\left(\int_0^\beta d\beta' e^{-(\beta-\beta')H_0}V_{SB}e^{-\beta'H_0}\right)}{Z_0^2}e^{-\beta H_0} \tag{43}$$

where $Z_0 = \mathrm{Tr}\left(e^{-\beta H_0}\right)$ is the partition function of the unperturbed Hamiltonian. Note that when derivating with respect to λ to determine ϱ_{corr} one needs to account for the fact that both $e^{-\beta H}$ and Z_{SB} depend on the coupling strength. Moreover any terms proportional to λ in the derivative have been neglected, as they would be absorbed in $\mathcal{O}(\lambda^2)$ in the Taylor expansion (42).

4.2   Causal-break formulation of the master equation

At this point we shift our focus to the family of decorrelated initial states $\varrho_{SB}^{ab}(0^+)$, parametrized by the action of the causal breaks $\mathcal{C}_{ab}$ on the initial state (42). They are given by

$$\varrho_{SB}^{ab}(0^+) = \tau_a(0) \otimes \left(\mathrm{Tr}_S(\Pi_b\varrho_S(0))\varrho_B^{th} + \lambda\mathrm{Tr}_S\left(\Pi_b\varrho_{corr}\right)\right) + \mathcal{O}(\lambda^2)$$
$$= \tau_a(0) \otimes (p_b\varrho_B(0) + \lambda\chi_b(0)) + \mathcal{O}(\lambda^2) \tag{44}$$

where we have defined

$$p_b = \mathrm{Tr}_S(\Pi_b\varrho_S^{th}) \tag{45}$$

and

$$\chi_b(0) = \mathrm{Tr}_S\left(\Pi_b\varrho_{corr}\right). \tag{46}$$

Inserting these states in the exact master equation (38) yields

$$\partial_t\tilde{\tau}_{ab}(t) = -i\lambda p_b\mathrm{Tr}_B\left(\left[\tilde{V}_{SB}(t), \tau_a(0) \otimes \varrho_B^{th}\right]\right) - i\lambda^2\mathrm{Tr}_B\left(\left[\tilde{V}_{SB}(t), \tau_a(0) \otimes \chi_b(0)\right]\right)$$
$$- \lambda^2\mathrm{Tr}_B\left(\left[\tilde{V}_{SB}(t), \int_0^t dt'[\tilde{V}_{SB}(t'), \tilde{\varrho}_{SB}^{ab}(t')]\right]\right) + \mathcal{O}(\lambda^3), \tag{47}$$

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[7]This expression can be proven by simply multiplying with e^{-tY} form the left and then differentiating with respect to time.

where the first two terms depend on the global initial state and the third is the canonical term governing the reduced state dynamics for product initial states. After certain approximations the third term is also known as the *dissipator*, as it is in most cases the dominant term accounting for the effect of the environment on the system dynamics. From now on we omit the $\mathcal{O}(\lambda^3)$ term and any expression for a master equation should be understood to be approximate and to pertain to the weak-coupling regime.

Analysis of the first order term and redefining the Hamiltonian

Let us now address the first order term and whether it can be set to zero or not, as is the case for the standard master equation, where initial correlations are neglected [BP07]. Starting our considerations from the Schrödinger picture, we express the interaction in the decomposition $V_{SB} = \sum_k A_k \otimes B_k$ and compute

$$\text{Tr}_B[V_{SB}, \tau_a(0) \otimes \varrho_B^{th}] = \sum_k [A_k, \tau_a(0)]\text{Tr}_B(B_k\varrho_B^{th}). \tag{48}$$

One can always redefine the system and interaction terms in the Hamiltonian, such that $\sum_k \langle B_k \rangle \equiv \sum_k \text{Tr}_B(B_k\varrho_B^{th})$ vanishes, by simply choosing $T_S = \sum_k \langle B_k \rangle A_k$ and setting

$$V'_{SB} = V_{SB} - T_S \otimes \mathbb{1}_B \quad \text{and} \quad H'_S = H_S + T_S \otimes \mathbb{1}_B, \tag{49}$$

where T_S represents a constant energy shift in the system Hamiltonian. This transformation leaves the total Hamiltonian unchanged, while at the same time guaranteeing

$$\text{Tr}_B\left(V'_{SB}\tau_a(0) \otimes \varrho_B^{th}\right) = \sum_k A_k\tau_a(0)\text{Tr}_B\left(B_k\varrho_B^{th} - \langle B_k \rangle\right) \tag{50}$$

$$= \sum_k A_k\tau_a(0)\text{Tr}_B\left(\langle B_k \rangle - \langle B_k \rangle\right) = 0. \tag{51}$$

We conclude therefore that the first order term in the master equation (47) can always be set to be zero, irrespective of the initially prepared state $\tau_a(0)$, simplifying thus the calculations significantly. Remains now to show that this procedure can be carried over in the interaction picture as well. We check therefore whether

$$\text{Tr}_B\left(\tilde{V}_{SB}(t)\varrho_B^{th}\right) = \sum_k \text{Tr}_B\left((B_k - \langle B_k \rangle)e^{-iH_Bt}\varrho_B^{th}e^{iH_Bt}\right) = 0, \tag{52}$$

which is only satisfied if

$$[\varrho_B^{th}, H_B] = 0. \tag{53}$$

In our case $\varrho_B^{th} = e^{-\beta H_B}/Z_B$ and the condition (53) is clearly satisfied, meaning that the first-order term in (47) can be safely neglected. We can also conclude that this is the case for the whole family of master equations $\{\mathcal{ME}\}_{ab}$, as the shift operator T_S that makes the term vanish is independent of a and b.

It is important to stress however that this result is directly related to our assumption of an initial global thermal state and cannot be generalized to arbitrary initial correlations. Had the zero-order term in the expansion of $\varrho_{SB}(0)$ not been proportional to ϱ_B^{th}, then the corresponding term in the master equation would have to be taken into account normally. In the following chapter we investigate whether the same procedure can be applied for the term involving $\chi_b(0)$ as well.

4.3 Inhomogeneous correction term due to initial correlations

We begin by writing the correlation term (43) in the more convenient form

$$\varrho_{corr} = -\varrho_{IC} + \frac{\text{Tr}(\varrho_{IC})}{Z_0^2} e^{-\beta H_S} \otimes e^{-\beta H_B}, \tag{54}$$

where we have introduced

$$\varrho_{IC} = \frac{1}{Z_0} \int_0^\beta d\beta' e^{-(\beta-\beta')H_0} V_{SB} e^{-\beta' H_0}. \tag{55}$$

The conditional state of the bath can then be written as

$$\chi_B(0) = -\text{Tr}_S(\Pi_b \varrho_{IC}) + p_b \frac{\text{Tr}(\varrho_{IC})}{Z_S} \varrho_B^{th}. \tag{56}$$

From this we see that the second term is proportional to ϱ_B^{th} and thus will not contribute to the dynamics, as the corresponding term in the master equation can be set to zero by the exact same shift operator T_S discussed previously, enabling us to define

$$\chi_b'(0) = \text{Tr}_S(\Pi_b \varrho_{IC}). \tag{57}$$

The only contribution remaining is therefore due to the term proportional to $\text{Tr}_S(\Pi_b \varrho_{IC})$ and is of the form $i\lambda^2/Z_0 \left[\tilde{V}_{SB}(t), \tau_a(0) \otimes \chi_B'(0) \right]$. For this term to be safely neglected for any reduced system state, one would need to redefine things such that

$$\sum_{k,l} \text{Tr}_B \left(B_k e^{-(\beta-\beta')H_0} B_l e^{-\beta' H_B} \right) = 0 \tag{58}$$

and additionally verify that this translates into the interaction picture as well, guaranteeing that condition (58) is satisfied for all times. For this to hold in the interaction picture, the correction term must commute with the bath Hamiltonian, as was shown in the previous paragraph (see equations (52)-(53)). Here however we have

$$\left[H_B, \int_0^\beta d\beta' e^{-(\beta-\beta')H_0} V_{SB} e^{-\beta' H_0} \right] = \int_0^\beta d\beta' e^{-(\beta-\beta')H_0} [H_B, V_{SB}] e^{-\beta' H_0} \neq 0, \tag{59}$$

since in general the interaction does not commute with the bath Hamiltonian. This indicates that this term cannot be eliminated from the master equation, as there is no unique way of redefining the system and interaction Hamiltonians that make it vanish for all times. Overall the master equation at this point reads

$$\partial_t \tilde{\tau}_{ab}(t) = i\lambda^2 \text{Tr}_B \left([\tilde{V}_{SB}(t), \tau_a(0) \otimes \chi_b'(0)] \right) - \lambda^2 \text{Tr}_B \left(\left[\tilde{V}_{SB}(t), \int_0^t dt' [\tilde{V}_{SB}(t'), \tilde{\varrho}_{SB}^{ab}(t')] \right] \right), \tag{60}$$

where $\chi_b'(0)$ contains all the information pertaining to the effect that initial correlations have on the dynamics of the system. We conclude therefore that allowing for correlations in the initial density matrix modifies the master equation by contributing an inhomogeneous term.

Remark

An interesting effect that can be seen in the structure of eq. (60) is that even if the first order correction on the global Gibbs state for the *reduced system* density matrix vanishes, i.e. $\text{Tr}_B(\varrho_{corr}) = 0 \Rightarrow \chi_b'(0) = 0$ (as is the case for many models, the spin-boson model amongst them [GRT00]), this does not imply that initial correlations do not affect the dynamics of the open system. This is because the commutator and the partial trace operations do not commute. In other words $\chi_b'(0) = 0$ does not imply $\text{Tr}_B([\tilde{V}_{SB}(t), \tau_a(0) \otimes \chi_b'(0)]) = 0$, indicating that initial correlations cannot be neglected.

Born-Markov approximations

So far we have managed to get some insights on the effect of the initial correlations in the structure of the master equation, however we still have to deal with the issue of the time non-local nature of the equation, as well as with its dependence on the global state $\tilde{\varrho}_{SB}(t)$. The treatment of these issues is standard textbook material in the field of open quantum systems [BP07, RH12] and for weak coupling master equations they are typically resolved by first invoking the *Born approximation* and then the *first Markov approximation*. In short, the Born approximation assumes the bath to be in thermal equilibrium throughout the systems evolution, imposing thus

$$\tilde{\varrho}_{SB}^{ab}(t) \approx p_b \tau_{ab}(t) \otimes \varrho_B^{th} + \mathcal{O}(\lambda), \tag{61}$$

where any corrections due to $\mathcal{O}(\lambda)$ can be neglected in the weak-coupling regime, as they would contribute third-order terms in the master equation. Meanwhile, the Markov approximation assumes a *memoryless* environment and performs the change of variables $t-t' \to t'$ to replace the retarded time density matrix $\tilde{\tau}_{ab}(t')$ in the integral with the *current* density matrix $\tilde{\tau}_{ab}(t)$, yielding thus a *time-local* equation. A more detailed discussion on the physical intuition behind these approximations, along with some mathematical insights, is given in the Appendix F.

At this point we opted to not perform the *second Markov approximation*, which replaces $\int_0^t dt'$ with $\int_0^\infty dt'$ and leads to a time-independent dissipator $\tilde{\mathcal{D}}$ at the cost of lower accuracy for very short times. The reason is that we aim to study the effect of the initial correlations on the system dynamics, whose effect is expected to be stronger at transient times and to decrease as the correlations due to the system-bath interaction build up over time. This of course will lead to a time-dependent dissipator $\tilde{\mathcal{D}}_t$.

Moreover, we also choose to not invoke the *secular approximation*. The secular approximation involves averaging over the fast-oscillating terms in the equation, and formally is very similar to the *rotating-wave approximation* typically performed in deriving Hamiltonians in the field of quantum optics, such as the Jaynes-Cummings one [JC63]. The main reason for performing the secular approximation is to bring the master equation in *Lindblad form* [RH12], which guarantees the positivity of the density matrix at future times (the converse is not true however, a master equation that is not of Lindblad form does not necessarily have negative solutions). It has been observed however that for short time scales the master equation obtained via the secular approximation is not justified, sometimes failing completely to capture the system dynamics in the weak coupling regime [SSSE18]. Furthermore, it has been demonstrated that the loss positivity does not manifest in the validity regime of the equation and it has been argued that it should even be embraced as a witness for the breakdown of the weak-coupling assumption [HS20].

4.4 Final form of the modified master equation

The master equation after the aforementioned approximations reads

$$\partial_t \tilde{\tau}_{ab}(t) = -\lambda^2 p_b \tilde{\mathcal{D}}_t[\tilde{\tau}_{ab}(t)] + i\lambda^2 \tilde{\Phi}_t[\tau_a(0)], \tag{62}$$

where[8]

$$\tilde{\mathcal{D}}_t[\bullet] = \sum_{k,l} \int_0^t d\tau \Big[C_{kl}(\tau) \Big(\tilde{A}_k(t) \tilde{A}_l(t-\tau) \bullet - \tilde{A}_l(t-\tau) \bullet \tilde{A}_k(t) \Big) + \text{h.c.} \Big], \tag{63}$$

---

[8]The abbreviation h.c. here stands for the hermitian conjugate.

is the time-dependent dissipator,

$$C_{kl}(t, \tau) = \langle \tilde{B}_k(t - \tau) B_l \rangle_{\varrho_B^{th}} = C_{kl}(t - \tau) \tag{64}$$

are the so-called *bath correlation functions* and

$$\tilde{\Phi}_{b,t}[\bullet] = \sum_k \langle \tilde{B}_k(t) \rangle_{\chi_b'(0)} \left[A_k(t), \bullet \right] \tag{65}$$

is the correction term accounting for the initial correlations in the global Gibbs state. It is straightforward to check that operator $\tilde{\Phi}_t$ preserves the trace and the hermiticity of the reduced density matrix $\tilde{\varrho}_S$. This modified master equation is an inhomogeneous, first order differential equation whose formal solution reads

$$\tilde{\tau}_{ab}(t) = G(t, 0)\tau_\alpha(0) + \int_0^t ds\, G(t, s)\tilde{\Phi}_{b,s}[\tau_\alpha(0)], \tag{66}$$

where

$$G(t, s) = \mathcal{T}_\leftarrow \exp\left(p_b \int_s^t dt'\tilde{\mathcal{D}}_{t'} \right) \tag{67}$$

is the solution to the homogeneous equation $\partial_t \tilde{\tau}_{ab}(t) = p_b\tilde{\mathcal{D}}_t[\tau_{ab}(t)]$.

Analysis of the initial correlation term

In this section we evaluate the initial correction term (65) in the master equation explicitly. We begin with

$$\langle \tilde{B}_k(t) \rangle_{\chi_b'(0)} = \frac{1}{Z_0} \sum_l \int_0^\beta d\beta'\, \mathrm{Tr}_S \left(\Pi_b e^{-(\beta - \beta')H_S} A_l e^{-\beta' H_S} \right) \mathrm{Tr}_B \left(\tilde{B}_k(t) e^{-(\beta - \beta')H_B} B_l e^{-\beta' H_B} \right). \tag{68}$$

We insert now the identity $\mathbb{1}_X = e^{-(\beta - \beta')H_X} e^{(\beta - \beta')H_X}, X = S, B$ after S_l and before $\tilde{B}_k(t)$ respectively to simplify the expression. After some algebra and by utilizing the cyclicity of the trace, as well as that $Z_0 = Z_S Z_B$, the correction term can be expressed in the more compact form

$$\tilde{\Phi}_{b,t}[\tau_a(0)] = i\sum_{k,l} \int_0^\beta d\beta'\, \langle \Pi_b \tilde{A}_l(-i(\beta - \beta')) \rangle_{\varrho_S^{th}} \langle \tilde{B}_k(t - i(\beta - \beta') B_l) \rangle_{\varrho_B^{th}} [\tilde{A}_k(t), \tau_a(0)], \tag{69}$$

where the interaction picture operators are now evaluated for complex times. The second expectation value can be recognized as the bath correlation functions evaluated at *complex times*, i.e. $C_{kl}(t - i(\beta - \beta'))$. This is an interesting result, as it shows that the whole effect of the bath is still contained at the same two-time correlation functions as in the case of an initial product state. Functions evaluated at complex times are known in quantum field theory and statistical mechanics as *Wick-rotated* functions [Wic54]. This is an indication of how one could go about interpreting this result, it is however beyond the scope of this thesis. The correction term can now be written as

$$\tilde{\Phi}_{b,t}[\tau_a(0)] = \sum_{k,l} \int_0^\beta d\beta'\, \langle \Pi_b \tilde{A}_l(-i(\beta - \beta')) \rangle_{\varrho_S^{th}} C_{kl}(t - i(\beta - \beta'))[\tilde{A}_k(t), \tau_a(0)]. \tag{70}$$

We introduce now the time- and temperature-dependent *initial correlation function*

$$H_{b,k}(t,\beta) = \sum_l \int_0^\beta d\beta' \, \langle \Pi_b \tilde{A}_l(-i(\beta - \beta')) \rangle_{\varrho_S^{th}} \, C_{kl}(t - i(\beta - \beta')) \tag{71}$$

and write

$$\tilde{\Phi}_{b,t}[\tau_a(0)] = \sum_k H_{b,k}(t,\beta)[\tilde{A}_k(t), \tau_a(0)]. \tag{72}$$

Schödinger Picture

All of the results so far are given in the interaction picture with respect to H_0. We transform now back to the Schrödinger picture and write

$$\partial_t \tau_{ab}(t) = -i[H_S, \tau_{ab}(t)] + e^{-iH_S t} \partial_t \tilde{\tau}_{ab}(t) e^{iH_S t}. \tag{73}$$

After some straightforward algebra, the Schrödinger picture master equation reads

$$\partial_t \tau_{ab}(t) = -i[H_S, \tau_{ab}(t)] - p_b \lambda^2 \mathcal{D}_t[\tau_{ab}(t)] + i\lambda^2 \Phi_{b,t}[\tilde{\tau}_a(0; -t)], \tag{74}$$

where

$$\mathcal{D}_t[\tau_{ab}(t)] = \sum_{k,l} \int_0^t dt' \, (C_{kl}(t')[A_k, A_l(-t')\tau_{ab}(t)] + C_{lk}(-t')[\tau_{ab}(t)A_l(-t'), A_k]) \tag{75}$$

and

$$\Phi_{b,t}[\tilde{\tau}_a(0; -t)] = \sum_{k,l} H_{b,k}(t,\beta)[A_k, \tilde{\tau}_a(0; -t)]. \tag{76}$$

The negative time argument in $\tilde{\tau}_a(0; -t) = e^{-iH_S t}\tau_a(0)e^{iH_S t}$ comes from the transformation back to the Schrödinger picture. This suggests that the influence of the initial correlations on the future state of the system can be related to how the prepared state $\tau_a(0)$ would have evolved backwards in time, had the system been isolated. For a more intuitive expression we define now the operators

$$E_k(t) = \int_0^t dt' \sum_l C_{kl}(t')A_l(-t'), \tag{77}$$

$$F_k(t) = \int_0^t d\tau \sum_l C_{lk}(-t')A_l(-t') \tag{78}$$

and write finally

$$\partial_t \tau_{ab}(t) = -i[H_S, \tau_{ab}(t)] - p_b \lambda^2 \sum_k ([A_k, E_k(t)\tau_{ab}(t)] + [\tau_{ab}(t)F_k(t), A_k])$$
$$+ i\lambda^2 \sum_k H_{b,k}(t,\beta)[A_k, \tilde{\tau}_a(0; -t)]. \tag{79}$$

This concludes the derivation of the family of master equations $\{\mathcal{ME}\}_{ab}$, which consists of d^4 decoupled, inhomogeneous differential equations. As expected they are non-local in time, since they contain a dependence on the initial state, a consequence of the correlated initial state. Importantly, we have shown that the whole effect of the bath is still encoded in the two-time bath correlation functions, except now evaluated at complex times. Let us emphasize once again that this master equations $\{\mathcal{ME}\}_{ab}$ are not in Lindblad form yet and thus do not guarantee the positivity of $\varrho_S(t)$. What is guaranteed however is that any

observed negativities are only caused by the breakdown of the weak-coupling assumption and not by the presence of entanglement in the initial state. Overall, the developed formalism constitutes a general theoretical tool can be applied to study the effect of initial correlations in the evolution and the thermodynamics of an open system for a variety of physical models, as long as the weak-coupling approximation is satisfied. Appendix G includes analytical expressions of the modified master equation and its characteristic functions for the paradigmatic *dissipative spin-boson model* [LCD$^+$87], while also outlining how the derived formalism can be benchmarked in that case.

5 Conclusions and outlook

In this thesis we analyzed the dynamics of open quantum systems in the presence of correlations in the global initial state by employing an operational approach to the problem and paying attention to the effect that local initial preparations can have on the evolution of the open system. We focused specifically on the correlations contained in the global thermal state of the system-bath composite, as it constitutes a natural assumption for the initial state of a system at rest.

The influence of classical correlations in the initial state has been explored by solving for the exact dynamics of the spin-boson pure dephasing model. The study reveals an impact on the geometry of decoherence in the Bloch sphere, which showcases an oscillating behavior that differs significantly from the uncorrelated case, with coherences emerging in multiple directions of the sphere as the system approaches its steady state. This analysis was also extended to the more general class of dephasing models.

Next, a family of weak-coupling master equations to treat initial correlations in the global thermal state of a fully general Hamiltonian was derived. The master equation applies to both classical and quantum correlations and contains an inhomogeneous correction term compared to the Born-Markov master equation that is derived for product initial states. The resulting master equation is non-local in time, as it contains a dependence on the initial state of the system. Moreover it has been demonstrated that the effect of the initial correlations on the bath is completely captured by the *Wick-rotated* ?? bath correlation functions. The developed formalism constitutes a general and practical tool that can be implemented to study the effect of initial entanglement on the dynamics of many relevant models.

Finally, there are several future directions for this project. Regarding the phenomenon of asymmetric decoherence of the Bloch-vector for dephasing models, it can be investigated in the context of Ramsey experiments, as the neglection of initial correlations could have potentially affected the frequency estimation of qubits in several experiments. A short description of this idea is included in the Appendix D. The most promising path this project can take however is the application of the derived master equation formalism for appropriate models, such as the dissipative spin-boson model, in order quantify the effect of initial correlations on the system dynamics, while also analyzing the different effect that quantum and classical correlations can cause. The results would indicate whether the assumption of an initial product state, which to this day underlies most theoretical descriptions of open systems, is justified for said models or not. Additionally, the formalism can also be applied to study the thermodynamics of initially correlated open systems and understand whether any negativities in the entropy production rate of the system constitute a quantum effect or not [SE19]. From a more theoretical point of view, it would also be of interest to interpret the complex time argument in the bath correlation functions appearing in the master equation.

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Appendix

A Representation of superoperators

Superoperators admit many representations and is the theoriticians choice which one to employ, depending on her/his purpose. Here we present a convenient way for representing any superoperator $\mathcal{A}$ that is especially useful for numerical applications. An example of such a use is to determine the unique set of coefficients γ_{ab}, such that $\mathcal{A} = \sum_{ab} \gamma_{ab} \mathcal{C}_{ab}$, where A is any quantum operation. This enables us to The idea is to represent A as a $d^2 \times d^2$ matrix $\hat{\mathcal{A}}$, by first vectorizing the density matrix as a vector of d^2 length, i.e.

$$\varrho = \sum_{kl} \varrho_{kl} |k\rangle\langle l| \longleftrightarrow |\varrho\rangle\rangle = \sum_{kl} \varrho_{kl} |kl\rangle\rangle. \tag{80}$$

Using the fact that the action of any superoperator on an operator can always be decomposed as $\mathcal{A}\varrho = \sum_k X_k \varrho Y_k$ for some $\{X_k\}$, $\{Y_k\}$, the matrix representation of $\mathcal{A}$ is then given by

$$\hat{\mathcal{A}} = \sum_k X_k \otimes Y_k^T. \tag{81}$$

We express now the action of a causal break on a quantum state in the aforementioned decomposition,

$$\begin{aligned}
\mathcal{C}_{ab}\varrho &= |\psi_a\rangle\langle\psi_a| \operatorname{Tr}\{|\psi_b\rangle\langle\psi_b| \varrho\} \\
&= |\psi_a\rangle\langle\psi_a| \langle\psi_b| \varrho |\psi_b\rangle \\
&= (|\psi_a\rangle \langle\psi_b|)\varrho(|\psi_b\rangle \langle\psi_a|),
\end{aligned}$$

and identify $X = |\psi_a\rangle \langle\psi_b|$ and $Y = |\psi_b\rangle \langle\psi_a|$. A single term was needed for the decomposition (??) in this case. The corresponding matrix representation $\hat{\mathcal{C}}_{ab}$ is then given by

$$\hat{\mathcal{C}}_{ab} = |\psi_\alpha\rangle \langle\psi_\beta| \otimes (|\psi_\alpha\rangle \langle\psi_\beta|)^*. \tag{82}$$

Any map $\mathcal{A}$ can now be linearly expanded in the basis formed by the causal breaks (82), i.e.

$$\mathcal{A} = \sum_{ab} \gamma_{ab} |\psi_a\rangle \langle\psi_b| \otimes (|\psi_a\rangle \langle\psi_b|)^*, \ \gamma_{ab} \in \mathbb{C}, \tag{83}$$

which is a set of d^4 linear equations that uniquely determine the coefficients γ_{ab}.

B Classical and quantum correlations: Quantum Discord

Quantum systems can be correlated in more ways than we are used to in the classical world. The most famous example amongst them is quantum entanglement and, at a first glance, one may be tempted to draw the line between quantum and classical correlations at the border between separable and entangled states. However, the disruptive nature of measurements in the quantum domain indicates that even separable states can contain correlations that are not classical in nature. Quantum discord is an information measure that accounts for this fact by quantifying the amount of information in a bipartite system that is not retrievable by local measurements only. It is defined as the difference between the mutual information $I(A : B) = S(A) + S(B) - S(AB)$, which accounts for all correlations present in a state ϱ_{AB}, and the quantity

$$J(B|E_a) = S(B) - S(B|\{E_a\}), \tag{84}$$

where $\{E_a\}$ is a POVM acting on A with outcome probabilities $p_a = \text{Tr}(E_a \otimes \mathbb{1}_B \varrho_{AB})$, and $S(X) = -\sum_X \varrho_X \log \varrho_X$ is the von Neumann entropy of the state. In the classical world, where measurements are not basis-dependent and do not disturb the state system, this quantity is equal to the mutual information $I(A : B)$, i.e.

$$J_{cl} = S(B) - S(B|A) = S(B) - \sum_a p_a S(B|a), \tag{85}$$

where $S(B|a) = -\sum_b p_{b|a} \log p_{b|a}$.

In the quantum realm however the choice of measurement $\{E_a\} = \{M_a^\dagger M_a\}$ affects the quantum state non-trivially, making thus the quantity J measurement dependent. To lift this dependence one can maximize over all possible POVM's and define

$$J(B|A) = \max_{\{E_a\}} J(B|\{E_a\}). \tag{86}$$

The information measure $J(B|A)$ quantifies therefore the amount of *classical* correlations between A and B and is interpreted as the amount of information one can gain about subsystem B by only performing local measurement on subsystem A. Note that in general $J(B|A) \neq J(A|B)$. The quantum discord of ϱ_{AB} is then defined as the the difference between total and classical correlations, i.e.

$$D(B|A) = I(A : B) - J(B|A). \tag{87}$$

The discord $D(B|A)$ is non-negative and vanishes if and only if ϱ_{AB} is a classical-quantum state, i.e.

$$D(B|A) = 0 \iff \varrho_{AB} = \sum_a p_a \Pi_a \otimes \varrho_{B|a}, \tag{88}$$

where $\{\Pi_a\}$ is a set of rank-one projectors satisfying $\Pi_i \Pi_j = \delta_{ij} \Pi_j$ and $\sum_a \Pi_a = \mathbb{1}$. States of this form are oftened referred to as *classical* or *zero-discord* states. Overall, quantum discord accounts for all non-classical correlations and only corresponds to entanglement for pure states. In other words discord can be non-zero also for separable, but non-product, mixed states.

Some interesting facts

- *Nullity condition*: $D(B|A) = 0 \implies [\varrho_A \otimes \mathbb{1}_B, \varrho_{AB}] = 0$.

- *Local operations can generate quantum correlations*: The classicality of a state is preserved if and only if the channel $\mathcal{C}$ preserves vanishing commutators, i.e. iff

$$[\mathcal{C}(\varrho), \mathcal{C}(\tau)] = 0 \quad \forall \quad [\varrho, \tau] = 0. \tag{89}$$

- *Negative entropy production rate and non-classical correlations*: If the time-derivative of the entropy of the reduced state at time t is non-zero, then the correlations between system and environment at that time are quantum in nature. This is satisfied if and only if the nullity condition from above is satisfied. This emphasizes that the quantum correlations play an important role for decoherence, as their presence is a necessary condition for it to occur. Moreover it has been proven that

$$\left| \frac{d}{dt} S(\varrho_S) \right| \leq ||H_{\text{int}}|| \; ||[(\log \varrho_S \otimes \mathbb{1}_B), \varrho_{SB}]||_1, \tag{90}$$

showing that the correlations impose bounds on the entropy production rate of the system. This property can be used study the different thermodynamic properties of initial states with quantum vs. classical correlations.

- *Trace-distance quantum-correlations witness*: Assuming the system is subjected to a measurement in its own basis, i.e. $\varrho_S \longrightarrow \sum_s \Pi_s \varrho_S \Pi_s$, it has been shown that the discord is zero if and only if the state is invariant under that measurement, i.e.

$$D(B|S) = 0 \iff \varrho_{SB} = \sum_s \Pi_s \otimes \mathbb{1}_B \varrho_{SB} \Pi_s \otimes \mathbb{1}_B. \tag{91}$$

If one compares the state prepared by $\{\Pi_s\}$ with a freely evolved state (no measurement) and observes differences, it can be concluded that the initial system-environment correlations are quantum. For the case of dephasing models this would correspond to a measurement in the z-basis, which would not induce dynamics on the Gibbs state of the whole system.

A thorough analysis on the classical or quantum nature of correlations can be found in [MBC$^+$12].

C Correlations in the thermal state of pure-dephasing models

One important characteristic of these models is that their global thermal state only contains classicall correlations between the system and its environment. More specifically, the global Gibbs state of a general dephasing is of the form

$$\varrho_{SB}(0) = \frac{1}{Z_{SB}} e^{-\beta H} = \frac{1}{Z_{SB}} \sum_m |m\rangle\langle m| \otimes e^{-\beta H_B^m} \tag{92}$$

$$= \sum_m \frac{Z_m}{Z_{SB}} |m\rangle\langle m| \otimes \frac{1}{Z_m} e^{-\beta H_B^m} = \sum_m p_m \Pi_m \otimes \varrho_{B,m}, \tag{93}$$

where we have defined $Z_m = \text{Tr}(e^{-\beta H_B^m})$, $p_m = \frac{Z_m}{Z_{SB}}$ and $\varrho_{B,m} = \frac{1}{Z_m} e^{-\beta H_B^m}$. This state is classical-quantum in the sense of (88). We can conclude therefore that the global gibbs states of all pure dephasing models satisfy

$$D_{PS}(B|S) = 0. \tag{94}$$

We can also verify the aforementioned nullity condition,

$$[\varrho_S \otimes \mathbb{1}_B, \varrho_{SB}] = \left[\sum_m p_m |m\rangle\langle m| \otimes \mathbb{1}_B, \sum_m p_m |m\rangle\langle m| \otimes \varrho_{B,m} \right] \tag{95}$$

$$= \sum_m p_m^2 |m\rangle\langle m| \otimes \varrho_{B.m} - \sum_m p_m^2 |m\rangle\langle m| \otimes \varrho_{B.m} = 0. \tag{96}$$

Furthermore, it is straightforward to check whether a causal break constitutes a classicality preserving channel, i.e. if

$$[\mathcal{C}(\varrho), \mathcal{C}(\xi)] = 0 \quad \forall [\varrho, \xi] = 0. \tag{97}$$

Starting with the left-hand side

$$[\mathcal{C}(\varrho), \mathcal{C}(\tau)] = \left[\tau_\alpha \otimes \text{Tr}_A(\Pi_\beta \otimes \mathbb{1}_B \varrho), \tau_\alpha \otimes \text{Tr}_A(\Pi_\beta \otimes \mathbb{1}_B \xi) \right]$$

$$= \tau_\alpha \sum_{m,n} \langle m| \Pi_\beta |m\rangle \langle n| \Pi_\beta |n\rangle [\varrho_{B,m}, \xi_{B,m}].$$

Taking into account that $[\varrho, \xi] = 0 \implies [\varrho_{B,m}, \xi_{B,m}] = 0$. We conclude therefore that causal breaks preserve the classicality of a state as defined in (88). This is not surprising given the decorrelating of the causal break.

D Initial correlations in Ramsey experiments

As is the case in most theoretical treatments, initial correlations are often being disregarded in experimental studies too. Based on the previous analysis of the geometry of decoherence in a two-level system, we briefly discuss here how this effect may have been misinterpreted as some short of a "Lamb-shift", i.e. an additional term in the system Hamiltonian. Naturally, this would contribute to the overall error of the estimated parameters. Let us consider the situation where the system is modelled to be closed, evolving thus under a Hamiltonian $H_0 = \omega_0 \sigma_z / 2$. Under this assumption of unitary dynamics, any oscillations detected in the system are associated to the eigenfrequency ω_0. However, We have demonstrated that the presence of correlations induces some oscillations on the system, which if the system is assumed to be isolated can be misaccounted when determining ω_0. To illustrate this, let us rewrite eq. (29) as

$$r_c(t) = \left(1 - K_b \cos\theta(t) + K_b e^{i\theta(t)}\right) e^{-\Gamma(t)}. \tag{98}$$

The correlation phase $\theta(t)$ and the associated asymmetry factor K_b would then find their way into the closed description of the qubit dynamics in the form of a Hamiltonian

$$H' \approx \left(\frac{\omega_0}{2} - K_b \frac{d\theta}{dt}\right) \sigma_z. \tag{99}$$

We focus now on the case of Ramsey interferometry, a method that is commonly utilized to determine the frequency ω_0 of a two-level system and is often the first part of many experiments involving qubits. The method utilizes an electromagnetic field oscillating at frequency ω that couples externally to the two-level system. The frequency ω_0 is determined by monitoring transition probability

$$P(\Delta) = \cos^2\left(\frac{\Delta T}{2}\right), \tag{100}$$

where $\Delta = \omega - \omega_0$ is the detuning between the field and the system and T the time of interaction. The probability $P(\Delta)$ is maximized for $\Delta = 0$, i.e. $\omega_0 = \omega(P(\Delta) = \max)$. For H' however the eigenfrequency should rather be computed by

$$\omega_0 = 2K_b \frac{d\theta}{dt} + \omega(P(\Delta) = \max). \tag{101}$$

To summarize, this analysis demonstrates how measuring the frequency of a qubit via the Ramsey method by disregarding the open nature of its dynamics can cause an error that is due to the initial correlations between the qubit and its environment. Let us mention that this analysis has not been extended further and therefore the order of magnitude of this effect remains unknown.

E Exact evolution of general dephasing models

The Hamiltonian (15) in the interaction picture is given by $H(t) = S \otimes B(t)$, where

$$B(t) = \sum_k e^{iH_k t} B_k e^{-iH_k t}, \tag{102}$$

and the corresponding time evolution operator reads

$$U_{SB}(t) = \mathcal{T}_{\leftarrow} \exp\left(-i \int_0^t dt' S \otimes B(t')\right), \tag{103}$$

where $\mathcal{T}_\leftarrow$ is the *chronological* time-ordering operator. The Taylor expansion of (103) yields

$$U_{SB}(t) = \sum_n \frac{1}{n!} S^n \otimes \tilde{B}_n(t), \tag{104}$$

where $\tilde{B}_n(t) \equiv \mathcal{T}_\leftarrow \left(-i \int_0^t dt' B(t')\right)^n$. The global state after a preparation consisting of a single causal break is performed on the system is $\varrho_{SB}(0^+) = \tau \otimes \eta(0)$, where

$$\eta(0) = \mathrm{Tr}_S \left(\Pi_\psi \otimes \mathbb{1}_B \varrho_{SB}(0)\right), \tag{105}$$

is the conditional state of the bath after the causal break and $\Pi = |\psi\rangle\langle\psi|$. After some algebra the density matrix of the subsystem S at time t is found to be

$$\varrho_S(t) = \sum_{n,k} \frac{1}{n!k!} S^n \tau S^k \otimes \mathrm{Tr}_B \left(\tilde{B}_n(t)\eta_b \tilde{B}_k^\dagger(t)\right), \tag{106}$$

where the adjoint operator of $\tilde{B}_n$ is *anti-chronologically* ordered, i.e.

$$\tilde{B}_n^\dagger(t) = \mathcal{T}_\rightarrow \left(i \int_0^t dt' B(t')\right)^n. \tag{107}$$

The elements of the density matrix of the system $\mathcal{S}$ are given by $\varrho_S^{ij}(t) = \mathrm{Tr}_S \left(|j\rangle\langle i| \, \varrho_S(t)\right)$, which after expanding the system operator in its eigenbasis, $S = \sum_i s_a |a\rangle\langle a|$, gives the evolution

$$\varrho_S^{ij}(t) = \sum_{n,k} \frac{1}{n!k!} \sum_{a,b} \mathrm{Tr}_S \left(|j\rangle\langle i| \, s_a^n s_b^k |a\rangle \, \langle a|\tau|b\rangle \, \langle b|\right) \mathrm{Tr}_B \left(\tilde{B}_n(t)\eta(0)\tilde{B}_k^\dagger(t)\right)$$

$$= \sum_{n,k} \frac{1}{n!k!} \sum_{a,b} s_a^n s_b^k \, \langle a|\tau|b\rangle \, \delta_{ia}\delta_{jb} \mathrm{Tr}_B \left(\tilde{B}_n(t)\eta(0)\tilde{B}_k^\dagger(t)\right)$$

$$= \sum_{n,k} \frac{1}{n!k!} \sigma_i^n \sigma_j^k \, \langle i|\tau|j\rangle \, \mathrm{Tr}_B \left(\tilde{B}_n(t)\eta(0)\tilde{B}_k^\dagger(t)\right)$$

$$= \langle i|\tau|j\rangle \, \mathrm{Tr}_B \left(\sum_n \frac{1}{n!} s_i^n \tilde{B}_n(t)\eta(0) \sum_k \frac{1}{k!} \sigma_j^k \tilde{B}_k^\dagger\right). \tag{108}$$

$$\tag{109}$$

The expression inside the trace can be simplified further by

$$\sum_n \frac{1}{n!} s_i^n \left(\mathcal{T}_\leftarrow \left(-i \int_0^t dt' B(t')\right)\right) = \mathcal{T}_\leftarrow \left(\sum_n \frac{1}{n!} s_i^n \left(-i \int_0^t dt' B(t')\right)\right) \tag{110}$$

due to the operator S being stationary in the interaction picture. Finally this can be compressed in a form containing exponential operators,

$$\varrho_S^{ij}(t) = \langle i|\tau|j\rangle \, (0) \mathrm{Tr}_B \left(\mathcal{T}_\leftarrow \exp\left[-is_i \int_0^t dt' B(t')\right] \eta(0) \mathcal{T}_\rightarrow \exp\left[is_j \int_0^t dt' B(t')\right]\right), \tag{111}$$

and the full density matrix is given by $\varrho_S(t) = \sum_{i,j} \varrho_S^{ij}(t) |i\rangle\langle j|$. By the cyclicity of the partial trace and due to $\tilde{U}_i = \tilde{U}_j$ for $i = j$, it is straightforward to verify that the populations remain constant. For the global Gibbs state $\varrho_{SB}(0) = e^{-\beta H}/Z$, the conditional bath state can be written as

$$\eta(0) = \sum_m |\langle\psi|m\rangle|^2 e^{-\beta H_B^m}. \tag{112}$$

Defining now the evolution operator

$$\tilde{U}_i = \mathcal{T}_{\leftarrow} \exp\left[-is_i \int_0^t dt' B(t')\right], \tag{113}$$

we obtain the compact expression

$$\varrho_S^{ij}(t) = \langle i|\tau|j\rangle\,(0) \sum_m |\langle\psi|m\rangle|^2 \mathrm{Tr}_B\left(\tilde{U}_i e^{-\beta H_B^m}\tilde{U}_j^\dagger\right), \tag{114}$$

which can be further simplified to

$$\varrho_S^{ij}(t) = \langle i|\tau|j\rangle\,(0)\frac{1}{Z}\sum_m |\langle\psi|m\rangle|^2 e^{-\beta\omega_m} \prod_k \mathrm{Tr}_B\left(\tilde{U}_i e^{-\beta(H_k + s_m B_k)}\tilde{U}_j^\dagger\right). \tag{115}$$

F Born and Markov approximations

In this chapter we discuss the physical intuition behind the approximations that have been performed to arrive at the master equation given in eq. (62).

Born approximation

First, let us discuss the so-called *Born* approximation. This approximation states that for weak system-bath interactions the combined state can be approximated by a product state, up to first order, i.e.

$$\tilde{\varrho}_{SB}^{ab}(t) \approx p_b \tau_{ab}(t) \otimes \varrho_B(t) + \mathcal{O}(\lambda). \tag{116}$$

Accounting for higher order terms in this expansion would only contribute third order terms in the master equation. Furthermore we assume that $\varrho_B(t) \approx \varrho_B(0) = \varrho_B^{th}$, motivated by $[\varrho_B^{th}, H_B] = 0$. It's important to stress this approximation is not physical, but helps in the derivation of the "correct" master equation. This can be seen from the fact that there is no way that the combined system can be in a product state in the presence of an interaction. A more rigorous derivation of the master equation that involves the Nakajima-Zwanzig *projection operator techniques* [Nak58, Zwa60] and circumvents this unphysical assumption can be found in [RH12]. This yields

$$\partial_t\tilde{\tau}_{ab}(t) = i\lambda^2 \mathrm{Tr}_B\left(\left[\tilde{V}_{SB}(t), \tau_a(0) \otimes \chi_b'(0)\right]\right) \tag{117}$$

$$- p_b\lambda^2 \int_0^t dt'\,\mathrm{Tr}_B\left(\left[\tilde{V}_{SB}(t), [\tilde{V}_{SB}(t'), \tilde{\tau}_{ab}(t') \otimes \varrho_B^{th}]\right]\right). \tag{118}$$

Substituting $\tilde{V}_{SB}(t) = \sum_k \tilde{A}_k(t) \otimes \tilde{B}_k(t)$, the second term reads

$$\sum_{k,l} \int_0^t d\tau\,\mathrm{Tr}_B\left(\left[\tilde{A}_k(t) \otimes \tilde{B}_k(t), [\tilde{A}_l(t') \otimes \tilde{B}_l(t'), \tilde{\tau}_{ab}(t') \otimes \varrho_B^{th}]\right]\right) \tag{119}$$

$$\tag{120}$$

We can define now the bath correlation functions

$$C_{kl}(t, t') = \mathrm{Tr}_B\left(\tilde{B}_k(t)\tilde{B}_l(t')\varrho_B^{th}\right) \equiv \left\langle \tilde{B}_k(t)\tilde{B}_l(t')\right\rangle_{\varrho_B^{th}}. \tag{121}$$

Due to the stationarity of ϱ_B^{th} this can also be written in the form

$$C_{kl}(t, t') = \left\langle \tilde{B}_k(t - t')B_l\right\rangle_{\varrho_B} = C_{kl}(t - t') \tag{122}$$

by simply substituting $t \to t - t'$. This shows that the environment correlation functions only depend on the time difference $t - \tau$ instead of the absolute time t. The second term is then

$$\sum_{k,l} \int_0^t dt' \left[C_{kl}(t-t') \Big(\tilde{A}_k(t) \tilde{A}_l(t') \tilde{\tau}_{ab}(t') - \tilde{A}_l(t') \tilde{\tau}_{ab}(t') \tilde{A}_k(t) \Big) \right.$$
$$\left. + C_{lk}(t'-t) \Big(\tilde{\tau}_{ab}(t') \tilde{A}_l(t') \tilde{A}_k(t) - \tilde{A}_k(t) \tilde{\tau}_{ab}(t') \tilde{A}_l(t') \Big) \right]. \qquad (123)$$

Markov approximation

At this point we perform the so-called *Markov* approximation, by assuming a "*memoryless*" environment, meaning that any self-orrelations induced on the bath B due to the interaction with the system S decay at a timescale $t'_{corr} \ll t'_S$, where t'_S is a timescale during which the state of the system $\tilde{\tau}_{ab}(t)$ (in the interaction picture) has changed significantly. By *memoryless* is meant that the environment is modelled as not keeping track of previous interactions with the system, as they decay very fast in comparison with the involved physical time-scales, so that each "step" of the evolution is effectively independent of the previous ones. This is were the name *Markovian* comes from, a concept originally introduced in classical probability theory to model stochastic processes whose future state only depends on the current state of the system. In more mathematical terms this means that the bath self-correlation functions are sharply peaked around $t - t' = 0$. This enables us to exchange the retarder reduced density matrix $\tilde{\tau}_{ab}(t')$ in the integral for the current density matrix, $\tilde{\tau}_{ab}(t)$, making thus this term *time-local*. This is justified because of the assumption that the reduced density matrix of the system does not change significantly during the time-scale t'_{corr}, during which the correlation functions C_{kl} vanish. After substituting $t - t' \to t'$ the term is in the form

$$\int_0^t dt' \sum_{k,l} \left[C_{kl}(t') \Big(\tilde{A}_k(t) \tilde{A}_l(t-t') \tilde{\tau}_{ab}(t) - \tilde{A}_l(t-t') \tilde{\tau}_{ab}(t) \tilde{A}_k(t) \Big) \right.$$
$$\left. + C_{lk}(-t') \Big(\tilde{\tau}_{ab}(t) \tilde{A}_l(t-t') \tilde{A}_k(t) - \tilde{A}_k(t) \tilde{\tau}_{ab}(t) \tilde{A}_l(t-t') \Big) \right]. \qquad (124)$$

G   Master equations for the dissipative spin-boson model

In this section we apply the developed formalism for the paradigmatic spin-boson model, described by the Hamiltonian

$$H = \frac{\omega_0}{2} \sigma_z - \frac{\Delta}{2} \sigma_x + \sum_k \omega_k b_k^\dagger b_k + \frac{1}{2} \sigma_z \sum_k (g_k b_k^\dagger + g_k^* b_k), \qquad (125)$$

where Δ is a parameter known as the *asymmetry* energy. This model, also known as the Caldeira-Legett model, is amongst the most studied models in the field of open systems and was first introduced in [LCD+87] It describes the interaction of a two-level system (or *qubit*) with a bath of harmonic oscillators (or *bosonic field modes*) and plays an important role in the study of decoherence, macroscopic quantum coherence and in the physical implementation of quantum information processing [Sch07].

Besides the many applications of the spin-boson model across various fields of physics, there are several reasons for choosing this specific model for our analysis, the most important one being the presence of non-classical correlations in the global Gibbs state of this

model. Additioanlly, setting the asymmetry energy to zero, i.e. $\Delta = 0$, reduces this model to the already discussed pure-dephasing spin-boson model, providing a straightforward way to benchmark our formalism. We should note here that since the global Gibbs state is stationary under the Hamiltonian (125), the choice of $\mathcal{P} = \mathcal{I}_S$ should show (approximately, since the master equation (79) is not exact) no dynamics, providing thus an additional test for the validity of our formalism.

In order to solve the required master equations numerically, we first need to characterize the coupling between the system and the environment. This is typically done in terms of the spectral density $J(\omega)$. To introduce this function properly, let us evaluate the bath correlation function[9], which was computed to be

$$C(t) = \sum_k |g_k|^2 \left(\coth\left(\beta\omega_k/2\right)\cos(\omega_k t) - i\sin(\omega_k t)\right) \tag{126}$$

and once again assume a continuous spectrum for the environment, performing thus the transformation $\sum_k |g_k|^2 \longrightarrow \int_0^\infty d\omega J(\omega)$. The bath correlation function can then be expressed as

$$C(t) = \nu(t) - i\kappa(t), \tag{127}$$

where $\nu(t)$ and $\kappa(t)$ are known as the *noise* and *dissipation kernels* respectively.

All the remaining terms in the master equation can be expressed in terms of the spectral density as well. For the initial correlation function we obtain

$$\begin{aligned}
H_{k,b}(t,\beta) = &\int_0^\infty d\omega J(\omega) \int_0^\beta d\beta' \langle \Pi_b \tilde\sigma_z(-i(\beta-\beta')) \rangle_{\varrho_S^{th}} \\
&\times \left[\coth(\beta\omega/2)\cos(\omega t - i(\beta-\beta')) - i\sin(\omega t - i(\beta-\beta')) \right], \\
= &\int_0^\beta d\beta' \langle \Pi_b \tilde\sigma_z(-i(\beta-\beta')) \rangle_{\varrho_S^{th}} \{\nu(t - i(\beta-\beta')) - i\kappa(t - i(\beta-\beta'))\}. \quad (128)
\end{aligned}$$

where $\tilde\sigma_z(t) = \sigma_z \cos(\Delta t/2) - \sigma_y \sin(\Delta t/2)$.

The integral over the temperatures in (128) is analytically computable, however results in a very long expression which we omit presenting here. Overall the family of master equations characterizing the initially correlated spin-boson model reads

$$\begin{aligned}
\partial_t \tau_{ab}(t) = &-i[H_S, \tau_{ab}(t)] - p_b \int_0^t dt' \left(\nu(t')\Big[\sigma_z, [\tilde\sigma_z(-t'), \tau_{ab}(t)]\Big] - \kappa(t')\Big[\sigma_z, \{\sigma_z(-t'), \tau_{ab}(t)\}\Big] \right) \\
&+ \int_0^\beta d\beta' \langle \Pi_b \tilde\sigma_z(-i(\beta-\beta')) \rangle_{\varrho_S^{th}} \Big[\nu(t - i(\beta-\beta')) - i\kappa(t - i(\beta-\beta')) \Big] [\sigma_z, \tau_a(0; -t)].
\end{aligned}$$
$$(129)$$

Besides the spectral density, which remains to be chosen (often times it is of interest to explore different functions), all other relevant functions of the model have been computed analytically. Thus, the family of master equations given in (129) can be solved numerically for any given preparation $\mathcal{P}$ using an appropriate technique, such as the Runge-Kutta method.

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[9]For the spin-boson model the interaction is of the type $V_{SB} = A \otimes B$, yielding a single correlation fucntion, $C(t)$.