A formally exact master equation for open quantum systems

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Abstract

We present a succinct and intuitive derivation of a formally exact master equation for general open quantum systems, without the use of an “inverse” map which was invoked in previous works on formally exact master equations. This formalism is applicable to non-Markovian regimes. We derive a second-order equation of motion for the illustrative spin-boson model at arbitrary temperatures, observing non-exponential decoherence and relaxation. Limiting our generic derivation to zero temperature, we also reproduce the result for the special case of a vacuum bath in Phys. Rev. A 81, 042103 (2010).

1 Introduction

A closed quantum system does not interact with external quantum degrees of freedom and its unitary dynamics is describable by the von Neumann equation,

\[ \frac{d}{dt} \rho_{\text{total}}(t) = -i \{ H_{\text{total}}(t), \rho_{\text{total}}(t) \}. \]  

(1)

An open quantum system interacts with external quantum degrees of freedom (the “environment”). \cite{5} There have been extensive studies on open quantum

\textsuperscript{*}This is a first draft of the manuscript. More physical applications of the master equation are being written up.
It is well known that open quantum systems generally do not evolve unitarily and the dynamics of their reduced density matrix \( \rho_S(t) \equiv \text{Tr}_E(\rho_{\text{total}}(t)) \) cannot be adequately described by the von Neumann equation; the scope of our work falls under the master equation approach to open system dynamics, which goes beyond the von Neumann equation and aims to describe non-unitary dynamics. [5]

Broadly speaking, quantum coherence plays an essential role in the field of quantum information [1] and quantum control [2]. The loss of quantum coherence, or “decoherence”, generally arises in open systems, resulting from their interaction with the environment. [8] One focus of open system study is thus on the decoherence aspect, besides other issues such as dissipation. [8]

There have been much works on approximate approaches to open quantum systems, [5] such as the widely used Born approximation and Markovian approximation [8, 9, 10, 11, 12]. However, from a theoretical point of view, these approximate approaches do not adequately reveal the “properties” of open quantum system dynamics. A formally exact master approach makes it possible to gain insights into the “properties” of general open quantum system dynamics, exact to every perturbative order. The closed form of the equation of motion may already provide insights into the nature of such dynamics, without it necessarily being solved. On the practical side, these approximations may be unjustified in and inapplicable to important situations. For example, the Markovian description does not apply to various scenarios of physical, chemical, and/or biological interest. [13, 14, 15] In principle, a formally exact approach makes it possible to go beyond such restrictions and be more widely applicable, including to non-Markovian regimes.

Formally exact approaches to general open quantum system dynamics are studied in [4, 5] with the time-convolutionless projection operator technique. (See [32, 33, 34] for the first proposal of this technique by Shibata et al.) There are also works outside of the field of open quantum systems, but on formally exact approaches to average dynamics of closed quantum systems [6, 7]. All the aforementioned formally exact approaches [4, 5, 6, 7] explicitly invoke some “inverse” in the derivations. Here we hope to dispense with the use of “inverse” in our derivation of the formally exact master equation. Our approach will be direct and “by construction”, rather than starting with some ansatz.

Besides, exact master equations are constructed in [16] for a two-level system decaying to a bath initially in vacuum state, wherein various techniques including the time-convolutionless method are discussed. The works [17, 18] present exact master equations for the case of Gaussian open quantum system dynamics. An exact master equation for quantum Brownian motion is presented in [19] with the influence functional method. The work in [20] shows an exact master equation for electrons in double dot by extending the influence functional method to fermionic environments. There is also a work on post-Markovian master equation through a measurement approach [21].

It is our goal to provide a succinct and intuitive, and yet sound, approach to deriving a formally exact master equation for general open quantum systems, that is, without restrictions on the type of system, environment, or system-
environment interaction. This is the subject of Section 2 of this paper. The formalism developed in Section 2 is then applied to study the spin-boson model as in Section 3 to illustrate the use of the master equation.

2 Theory

2.1 Derivations

Series expansion of full dynamics

We start with the equation of motion for the full system-environment dynamics,

$$i \frac{d}{dt} \rho_{SE}(t) = [H_{SE}(t), \rho_{SE}(t)],$$

where $H_{SE}(t)$ and $\rho_{SE}(t)$ are the interaction Hamiltonian and the full system-environment density matrix in the interaction picture respectively. \cite{3, 5} Following a standard approach to parametrize the Hamiltonian $H_{SE}(t)$ by $\lambda$, \cite{6, 7} we have

$$i \frac{d}{dt} \rho_{SE}(t) = \lambda [H_{SE}(t), \rho_{SE}(t)].$$

The full density matrix $\rho_{SE}(t)$ evolves unitarily,

$$\rho_{SE}(t) = U(t, 0)\rho_{SE}(0)U^\dagger(t, 0),$$

where the uniraty operator $U(t, 0)$ obeys the equation of motion

$$i \frac{d}{dt} U(t, 0) = \lambda H_{SE}(t)U(t, 0).$$

We suppose the unitary operator can be expanded in a power series of $\lambda$: \cite{3, 6, 7}

$$U(t, 0) = \sum_{n=0}^{\infty} \lambda^n U_n(t, 0).$$

Plugging Eq.(6) into Eq.(5), we have

$$i \frac{d}{dt} U_0(t, 0) = 0,$$

$$i \frac{d}{dt} U_n(t, 0) = H_{SE}(t)U_{n-1}(t, 0) \quad (n = 1, 2, \ldots).$$

\footnote{Throughout the paper we formally set $\hbar = 1$ for notational convenience unless otherwise noted.}
Solving the above equations, we have

\[ U_0(t, 0) = I, \]
\[ U_n(t, 0) = -i \int_0^t dt' H_{SE}(t') U_{n-1}(t', 0) \quad (n = 1, 2, \ldots). \]

The full system-environment dynamics can thus be expressed as

\[ \rho_{SE}(t) = \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \lambda^{m+n} U_m(t, 0) \rho_S(0) \otimes \rho_E(0) U_n^\dagger(t, 0). \]

Reduced dynamics

The reduced density matrix of the system is the partial trace of the full density matrix over environmental degrees of freedom [5, 8]

\[ \rho_S(t) = \text{Tr}_E (\rho_{SE}(t)) = \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \lambda^{m+n} \text{Tr}_E (U_m(t, 0) \rho_S(0) \otimes \rho_E(0) U_n^\dagger(t, 0)). \]

For convenience in subsequent derivation, let’s re-write the mapping as

\[ \rho_S(t) = \sum_{k=0}^{\infty} \lambda^k \mathcal{E}_{k,t} (\rho_S(0)) \]
\[ = \rho_S(0) + \sum_{k=1}^{\infty} \lambda^k \mathcal{E}_{k,t} (\rho_S(0)) \]
\[ \equiv (I + \mathcal{E}_t) (\rho_S(0)), \]

where

\[ \mathcal{E}_t (\rho) \equiv \sum_{k=1}^{\infty} \lambda^k \mathcal{E}_{k,t} (\rho), \]
\[ \mathcal{E}_{k,t} (\rho) \equiv \sum_{j=0}^{k} \text{Tr}_E \left( U_{k-j}(t, 0) \rho \otimes \rho_E(0) U_j^\dagger(t, 0) \right) \quad (k = 1, 2, \ldots). \]

Note that because \( \mathcal{E}_t (\rho) = \sum_{k=1}^{\infty} \lambda^k \mathcal{E}_{k,t} (\rho) \sim O(\lambda), \) we know \( \mathcal{E}_t (\rho) \) approaches zero as \( \lambda \to 0. \) Also, by definition, \( \mathcal{E}_t (\rho) \) approaches zero as \( t \to 0. \)

The \( Y_{N,t} \) map

The key to obtaining a formally exact, time-local equation of motion in closed form is the following step. Let’s define a linear map central to our construction:

\[ Y_{N,t} (\rho) \equiv \sum_{n=0}^{N} (-1)^n \mathcal{E}^{(n)}_t (\rho), \]
where $\mathcal{E}_t^{(n)}(\rho) \equiv \mathcal{E}_t(\mathcal{E}_t(\ldots \mathcal{E}_t(\rho))))$ is a composition of $n \mathcal{E}_t$ maps. Then, applying this linear map to the system’s density matrix at time $t$ yields

$$Y_{N,t}(\rho_S(t)) = \sum_{n=0}^{N} (-1)^n \mathcal{E}_t^{(n)}((I + \mathcal{E}_t)(\rho_S(0)))$$

$$= (I + (-1)^{N+1} \mathcal{E}_t^{(N+1)})(\rho_S(0)). \tag{17}$$

Denoting $\rho_S(t) \equiv \rho_t$ and $\rho_S(0) \equiv \rho_0$ for notational convenience, we now have the key equality in our work:

$$\rho_0 = Y_{N,t}(\rho_t) + (-1)^{N+1} \mathcal{E}_t^{(N+1)}(\rho_0)$$

$$= \sum_{n=0}^{N} (-1)^n \mathcal{E}_t^{(n)}(\rho_t) + (-1)^{N+1} \mathcal{E}_t^{(N+1)}(\rho_0). \tag{18}$$

What it does is to express the initial system’s state $\rho_0$ in terms of the system’s state at time $t$ $\rho_t$ (with a residual term $(-1)^{N+1} \mathcal{E}_t^{(N+1)}(\rho_0)$ that can be neglected to certain perturbative orders).

Note that the form of the $Y_{N,t}(\rho)$ map might bear some resemblance to the $[1 - \Sigma(t)]^{-1}$ super-operator in [4, 5], but there is at least one important difference besides others: here we make no use of an inverse map, whereas [4, 5] assumes an inverse.

Our work might be mathematically equivalent to the apparently different work in [4, 5], wherein more complicated theoretical constructs are used, such as the projection operator technique and antichronological time-ordering. In fact, any formulation of a general exact master equation should be mathematically equivalent to any other formulation in every order of the perturbative parameter $\lambda$. In any case, our work is independently constructed, with all the derivation steps naturally motivated and intermediate terms intuitively defined. It is our goal to formulate a succinct and intuitive, and yet sound, approach to deriving a formally exact master equation for general open quantum systems, and we believe that we have achieved this goal with our work.

**Equation of motion**

Taking the time derivative of the system’s reduced density matrix and mak-
ing use of Eq. (18), we have

\[
\frac{d}{dt} \rho_t = \frac{d}{dt} (\mathbb{I} + \mathcal{E}_t) (\rho_0) = \dot{\mathcal{E}}_t (\rho_0) = \dot{\mathcal{E}}_t \left( \sum_{n=0}^{N} (-1)^n \mathcal{E}^{(n)}_t (\rho_t) + (-1)^{N+1} \mathcal{E}^{(N+1)}_t (\rho_0) \right). \tag{19}
\]

Note that in the third equality the \( Y_{N,t} (\rho) \) map as in Eqs. (16, 18) has done the crucial job of re-expressing the right-hand side of the equation in terms of the quantity of interest, namely the system’s state at time \( t \). Therefore, we have

\[
\frac{d}{dt} \rho_t = \sum_{n=0}^{N} (-1)^n \dot{\mathcal{E}}_t \left( \mathcal{E}^{(n)}_t (\rho_t) \right) + (-1)^{N+1} \dot{\mathcal{E}}_t \left( \mathcal{E}^{(N+1)}_t (\rho_0) \right). \tag{20}
\]

Note that, up to this point, no approximation has been made and Eq. (20) is formally exact.

With Eq. (20), we can systematically make approximations, that is, collecting like-order terms in \( \lambda \) and truncating the series as needed. Since \( \mathcal{E}_t (\rho) \sim \mathcal{O} (\lambda) \Rightarrow \dot{\mathcal{E}}_t \left( \mathcal{E}^{(N+1)}_t (\rho_0) \right) \sim \mathcal{O} (\lambda^{N+2}) \), if we want to consider \( M \)th-order approximation, we can always choose \( N \geq M - 1 \), so that the residual term \((-1)^{N+1} \dot{\mathcal{E}}_t \left( \mathcal{E}^{(N+1)}_t (\rho_0) \right)\) may be neglected in our intended approximation and thus its presence in Eq. (20) does not matter. \(^2\)

Idealistically, we may hope to obtain a formally exact, time-local, linear homogeneous differential equation as the equation of motion. This can be formally achieved by taking the \( N \to \infty \) limit on the right-hand side of Eq. (20). Loosely speaking, as \( \lim_{N \to \infty} (-1)^{N+1} \dot{\mathcal{E}}_t \left( \mathcal{E}^{(N+1)}_t (\rho_0) \right) \sim \lim_{N \to \infty} \mathcal{O} (\lambda^{N+2}) \to 0 \), the residual term may be dropped, and we have

\[
\frac{d}{dt} \rho_t = \sum_{n=0}^{N} (-1)^n \dot{\mathcal{E}}_t \left( \mathcal{E}^{(n)}_t (\rho_t) \right), \tag{21}
\]

which is formally a linear homogeneous differential equation, albeit with infinitely many terms. \(^3\) However, note that this \( N \to \infty \) formal treatment and the resulting linear homogeneous differential equation are not necessary for ob-

\(^2\)Loosely speaking, in order for the residual term \((-1)^{N+1} \dot{\mathcal{E}}_t \left( \mathcal{E}^{(N+1)}_t (\rho_0) \right)\) to be negligible compared to lower order terms like \((-1)^N \dot{\mathcal{E}}_t \left( \mathcal{E}^{(N)}_t (\rho_0) \right)\) in Eq. (20), it apparently requires the map \( \mathcal{E}_t (\ldots) \) to be reasonably small. As we discuss earlier, the time-dependent map \( \mathcal{E}_t (\ldots) \to 0 \) as \( t \to 0 \) by definition; also, as the coupling strength approaches zero, the interaction Hamiltonian tends to vanish, thus \( \mathcal{E}_t (\ldots) \to 0 \) as well. Therefore, our approximation should work in the short time and/or weak coupling regimes. We do not extrapolate this approximation to the long time or strong coupling regimes.

\(^3\)Implicit in this discussion is the convergence of the infinite series in Eq. (21). Loosely speaking, in order for the infinite series to converge, it apparently requires the higher order
taining an $M$th-order approximate equation of motion for the system’s reduced
dynamics, the latter of which is all that matters in applications. In other words,
this $N \to \infty$ formal treatment can be dispensed with no practical implications.

2.2 Second-order equation of motion

In many cases, one is interested in the second-order approximate equation
of motion, as it is usually the leading order term that exhibits interesting effects
such as decoherence. For second-order approximation, let $N = 2 - 1 = 1$ in
Eq. (20):

\[
\frac{d}{dt}\rho_t = \dot{\mathcal{E}}_1 (\rho_t) - \dot{\mathcal{E}}_1 (\mathcal{E}_1 (\rho_t)) + O (\lambda^3)
\]

\[
= \left[ \lambda \mathcal{E}_{1,t} (\rho_t) + \lambda^2 \mathcal{E}_{2,t} (\rho_t) + \ldots \right]
\]

\[- \left[ \left( \lambda \mathcal{E}_{1,t} + \lambda^2 \mathcal{E}_{2,t} + \ldots \right) \left( \lambda \mathcal{E}_{1,t} (\rho_t) + \lambda^2 \mathcal{E}_{2,t} (\rho_t) + \ldots \right) \right]
\]

\[+ O (\lambda^3)
\]

\[
= \lambda \mathcal{E}_{1,t} (\rho_t) + \lambda^2 \left[ \mathcal{E}_{2,t} (\rho_t) - \mathcal{E}_{1,t} (\mathcal{E}_{1,t} (\rho_t)) \right] + O (\lambda^3).
\]

(22)

Therefore, the second-order equation of motion is

\[
\frac{d}{dt}\rho_t = \mathcal{L}_{1,t} (\rho_t) + \mathcal{L}_{2,t} (\rho_t),
\]

(23)

where $\mathcal{L}_{1,t} (\rho)$ and $\mathcal{L}_{2,t} (\rho)$ are defined for an arbitrary $\rho$ as

\[
\mathcal{L}_{1,t} (\rho) = \mathcal{E}_{1,t} (\rho),
\]

(24)

\[
\mathcal{L}_{2,t} (\rho) = \mathcal{E}_{2,t} (\rho) - \mathcal{E}_{1,t} (\mathcal{E}_{1,t} (\rho)).
\]

(25)

More specifically, we can work out the formal expressions of $\mathcal{L}_{1,t} (\rho)$ and
$\mathcal{L}_{2,t} (\rho)$ in terms of $H_{SE}(t)$ and $\rho_{E0}$:

\[
\mathcal{L}_{1,t} (\rho) = \mathcal{E}_{1,t} (\rho)
\]

\[= TrE \left[ \dot{U}_1 (t,0) \rho \otimes \rho_{E0} \right] + TrE \left[ \rho \otimes \rho_{E0} U_1^\dagger (t,0) \right]
\]

\[= -i (TrE [H_{SE}(t) \rho \otimes \rho_{E0}] - TrE [\rho \otimes \rho_{E0} H_{SE}(t)]) ;
\]

(26)

\[\text{terms (i.e. } \dot{\mathcal{E}}_1 (\mathcal{E}^{(n)}_t (\rho_t)) \text{ with larger } n \text{) be progressively smaller. As we discuss earlier,}
\]

$\mathcal{E}_t (\ldots) \to 0$ as $t \to 0$; also, $\mathcal{E}_t (\ldots) \to 0$ as coupling approaches zero. As $\mathcal{E}_t (\ldots) \to 0$,
$\dot{\mathcal{E}}_t (\mathcal{E}^{(n)}_t (\rho_t))$ should be progressively smaller for larger $n$, thus our discussion should be valid
in the short time and/or weak coupling regimes. We do not extrapolate this discussion to the
long time or strong coupling regimes.
\( \mathcal{L}_{2,t}(\rho) = \mathcal{E}_{2,t}(\rho) - \mathcal{E}_{1,t}(\mathcal{E}_{1,t}(\rho)) \)
\[ = \text{Tr}_E \{ \dot{U}_2(t,0)\rho \otimes \rho_{E0} + \dot{U}_1(t,0)\rho \otimes \rho_{E0}U_1^\dagger(t,0) \]
\[ + U_1(t,0)\rho \otimes \rho_{E0}U_1^\dagger(t,0) + \rho \otimes \rho_{E0}U_2^\dagger(t,0) \} \]
\[ + i \text{Tr}_E \{ H_{SE}(t)\mathcal{E}_{1,t}(\rho) \otimes \rho_{E0} - \mathcal{E}_{1,t}(\rho) \otimes \rho_{E0}H_{SE}(t) \} \]
\[ = - \int_0^t dt' \text{Tr}_E \{ H_{SE}(t)H_{SE}(t')\rho \otimes \rho_{E0} - H_{SE}(t)\rho \otimes \rho_{E0}H_{SE}(t') \]
\[ - H_{SE}(t')\rho \otimes \rho_{E0}H_{SE}(t) + \rho \otimes \rho_{E0}H_{SE}(t')H_{SE}(t) \} \]
\[ + \int_0^t dt' \text{Tr}_E \{ H_{SE}(t)(\text{Tr}_E [H_{SE}(t')\rho \otimes \rho_{E0} - \rho \otimes \rho_{E0}H_{SE}(t')]) \otimes \rho_{E0} \]
\[ - (\text{Tr}_E [H_{SE}(t')\rho \otimes \rho_{E0} - \rho \otimes \rho_{E0}H_{SE}(t')]) \otimes \rho_{E0}H_{SE}(t) \} \} \quad (27) \]

In general, the interaction Hamiltonian \( H_{SE}(t) \) can be expressed in terms of operators on the system Hilbert space \( \{S_n(t)\} \) and those on the bath Hilbert space \( \{E_n(t)\} \) as [5]

\[ H_{SE}(t) = \sum_n S_n(t) \otimes E_n(t). \quad (28) \]

With this, \( \mathcal{L}_{1,t}(\rho) \) and \( \mathcal{L}_{2,t}(\rho) \) can be re-expressed as:

\[ \mathcal{L}_{1,t}(\rho) = -i \left( \sum_n \text{Tr}_E (S_n(t)\rho \otimes E_n(t)\rho_{E0}) - \sum_n \text{Tr}_E (\rho S_n(t) \otimes \rho_{E0}E_n(t)) \right) \]
\[ = -i \sum_n \text{Tr}_E (\rho_{E0}E_n(t)) [S_n(t), \rho], \quad (29) \]

\[ \mathcal{L}_{2,t}(\rho) = - \int_0^t dt' \sum_m \sum_n (\text{Tr}_E (\rho_{E0}E_m(t)E_n(t')) - \text{Tr}_E (\rho_{E0}E_m(t)) \text{Tr}_E (\rho_{E0}E_n(t'))) \]
\[ [S_m(t), S_n(t')\rho] \]
\[ + \int_0^t dt' \sum_m \sum_n (\text{Tr}_E (\rho_{E0}E_n(t')E_m(t)) - \text{Tr}_E (\rho_{E0}E_n(t')) \text{Tr}_E (\rho_{E0}E_m(t'))) \]
\[ [S_m(t), \rho S_n(t')]. \quad (30) \]

**Main result**

In summary, for an open quantum system interacting with a bath via the Hamiltonian \( H_{SE}(t) = \sum_n S_n(t) \otimes E_n(t) \), the initial state of the bath being \( \rho_{E0} \), the equation of motion for the system’s reduced density matrix \( \rho \) is (up to second order)

\[ \frac{d}{dt} \rho(t) = -i [H_{\text{eff}}(t), \rho(t)] + \mathcal{L}_{2,t}(\rho(t)), \quad (31) \]
where the first-order effective Hamiltonian is
\[
H_{\text{eff}}(t) \equiv \sum_n T_{\rho E} (\rho_{E_0} E_n(t)) S_n(t), \tag{32}
\]
and the second-order term is
\[
\mathcal{L}_{2,t} (\rho) = - \sum_m \sum_n \int_0^t dt' \left( C_{mn}(t, t') [S_m(t), S_n(t') \rho] - C_{nm}(t', t) [S_m(t), \rho S_n(t')] \right), \tag{33}
\]
with the coefficients being
\[
C_{jk}(t, t') \equiv \text{Tr} E \left( \rho_{E_0} E_j(t) E_k(t') \right) - \text{Tr} E \left( \rho_{E_0} E_j(t) \right) \text{Tr} E \left( \rho_{E_0} E_k(t') \right). \tag{34}
\]

Second-order non-Markovian master equations like this are previously studied in the literature. For example, [4, 5] shows a time-convolutionless projection operator approach, wherein Eqs.(9.52, 9.61) of Ref.[5] is a second-order non-Markovian master equation, though with the first-order effective Hamiltonian vanishing due to the vanishing odd moments of the interaction Hamiltonian with respect to the environmental state.

### 2.3 Higher-order equations of motion

With the master equation formalism developed herein, one can systematically investigate an open quantum system’s dynamics to higher orders. For example, if one is interested in the reduced dynamics up to \( M \)-th order, one can first set \( N = M - 1 \) in Eq.(20) to obtain
\[
\frac{d}{dt} \rho_t = \sum_{n=0}^{M-1} (-1)^n \hat{\mathcal{E}}_t \left( \mathcal{E}_t^{(n)} (\rho_t) \right) + (-1)^M \hat{\mathcal{E}}_t \left( \mathcal{E}_t^{(M)} (\rho_0) \right) = \sum_{n=0}^{M-1} (-1)^n \hat{\mathcal{E}}_t \left( \mathcal{E}_t^{(n)} (\rho_t) \right) + \mathcal{O}(\lambda^{M+1}), \tag{35}
\]
then work out the terms \( \hat{\mathcal{E}}_t \left( \mathcal{E}_t^{(n)} (\rho_t) \right) \) according to Eqs.(14, 15),
\[
\mathcal{E}_t (\rho) = \sum_{k=1}^{\infty} \lambda^k \mathcal{E}_{k,t} (\rho), \tag{36}
\]
\[
\mathcal{E}_{k,t} (\rho) = \sum_{j=0}^{k} T_{\rho E} \left( U_{k-j}(t, 0) \rho \otimes \rho_{E_0} (0) U_j^\dagger(t, 0) \right), \tag{37}
\]
with \( U_n(t, 0) \) defined as in Eqs.(9,10), and then collect like order terms up to \( M \)-th order (dropping higher-order contributions) to obtain an equation of the form
\[
\frac{d}{dt} \rho_t = -i [H_{\text{eff}}(t), \rho_t] + \mathcal{L}_{2,t} (\rho_t) + \sum_{n=3}^{M} \mathcal{L}_{n,t} (\rho_t), \tag{38}
\]
with every term \( \mathcal{L}_{n,t} (\rho) \) in Eq.(38) well defined. All these steps can be carried out mechanically.

Non-Markovian master equations of higher orders are also known in the literature. See [4, 5] again, for example, wherein Eqs.(9.41, 9.42, 9.47, 9.51) of Ref.[5] show some higher-order terms of the non-Markovian master equation.

3 Example: Spin-boson model

A two-level system (TLS) interacting with bosonic field modes is extensively studied and widely used in the open quantum systems literature. [5, 8, 27, 4, 14, 16] Here we will use the spin-boson model as an illustrative example for the master equation formalism developed above.

3.1 Problem description

For a two-level system (TLS) interacting with a bosonic field, the total Hamiltonian is (in Schrödinger picture) [5]

\[
H_{total} = \frac{\omega_0}{2} \sigma_z + \sum_k \omega_k b_k^\dagger b_k + \sum_k g_k \left( \sigma_+ b_k + \sigma_- b_k^\dagger \right), \quad (39)
\]

where the first term is the self-Hamiltonian of the TLS (\( \omega_0 \) being the energy spacing), the second term is the self-Hamiltonian of a collection of independent bosonic modes (\( b_k \) and \( b_k^\dagger \) being the annihilation and creation operators of \( k \)-th mode, \( \omega_k \) being its frequency) [5], and the third term is the system-bath interaction (\( g_k \) being the coupling strength between TLS and \( k \)-th field mode, and \( \sigma_+ \) leading to transition from TLS’s ground state to its excited state while \( \sigma_- \) doing the opposite) [5].

Treating \( H_{SE} = \sum_k g_k \left( \sigma_+ b_k + \sigma_- b_k^\dagger \right) \) as a perturbation to the unperturbed Hamiltonian \( H_0 = \frac{\omega_0}{2} \sigma_z + \sum_k \omega_k b_k^\dagger b_k \) and switching to the interaction picture [3] (i.e. the “rotating frame” generated by \( H_0 \)), we have

\[
H_{SE}^{(int-pic)} (t) = \sum_k g_k \left( \sigma_+ b_k e^{-i(\omega_k - \omega_0)t} + \sigma_- b_k^\dagger e^{i(\omega_k - \omega_0)t} \right). \quad (40)
\]

Suppose the bosonic field is initially in the thermal state, that is,

\[
\rho_{E0} = \frac{1}{Z} \exp (-\beta H_{field}), \quad (41)
\]

where \( Z = Tr_{E} \left( \exp (-\beta H_{field}) \right) \) is the partition function and \( \beta = 1/k_B T \) is

\[\text{Hereafter we drop the superscript “interaction picture” for notational convenience and have in mind all operators are in the interaction picture unless otherwise noted.}\]
the inverse temperature. \cite{8, 28, 29} In this example, we have

\[
\rho_{E0} = \Pi_k \otimes \left( \frac{1}{Z_k} \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle m_k| \right) = \frac{1}{Z} \Pi_k \otimes \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle m_k| \right),
\]

(42)

where \(Z_k = \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k}\) and \(Z = \prod_k Z_k\), \(\omega_k\) is the frequency of the \(k\)-th bosonic mode, and \(m_k\) is the number of bosons in the \(k\)-th mode. \cite{8, 28, 29}

### 3.2 Equation of motion

The first-order effective Hamiltonian in the equation of motion (see Appendix A for calculation details) is found to vanish,

\[
H_{eff}^I(t) = 0, \tag{43}
\]

which means the system-bath interaction does not have first-order contribution to the TLS’s reduced dynamics in this case.

Introducing the following definitions with \(\omega_{k0} \equiv \omega_k - \omega_0\) for notational convenience,

\[
D_R(t) \equiv \int_0^t dt' \sum_k |g_k|^2 \bar{N}_k \cos(\omega_{k0}(t-t')) , \tag{44}
\]

\[
D_I(t) \equiv \int_0^t dt' \sum_k |g_k|^2 \bar{N}_k \sin(\omega_{k0}(t-t')) , \tag{45}
\]

\[
D'_R(t) \equiv \int_0^t dt' \sum_k |g_k|^2 \left(\bar{N}_k + 1\right) \cos(\omega_{k0}(t-t')) , \tag{46}
\]

\[
D'_I(t) \equiv \int_0^t dt' \sum_k |g_k|^2 \left(\bar{N}_k + 1\right) \sin(\omega_{k0}(t-t')) , \tag{47}
\]

where we have denoted the average occupation number in the \(k\)-th mode of the bath as

\[
\bar{N}_k \equiv Tr_E \left( \rho_{E0} b_k^\dagger b_k \right) = \frac{1}{Z_k} \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} \langle m_k | b_k^\dagger b_k | m_k \rangle, \tag{48}
\]

it can be shown that the second-order term in the equation of motion is (see Appendix A for calculation details)

\[
\mathcal{L}_{2,t}(\rho) = -i \left[ H_{eff}^I(t), \rho \right] - D_R(t) \left( \sigma_- \sigma_+ \rho + \rho \sigma_- \sigma_+ - 2 \sigma_+ \rho \sigma_- \right) \\
- D'_R(t) \left( \sigma_+ \sigma_- \rho + \rho \sigma_+ \sigma_- - 2 \sigma_- \rho \sigma_+ \right), \tag{49}
\]
where the second-order effective Hamiltonian is defined as

\[ H_{II}^{eff}(t) \equiv D_I(t)\sigma_-\sigma_+ - D'_I(t)\sigma_+\sigma_- . \] (50)

With Eq.(43) for the first-order term and Eq.(49) for the second-order term, we can write down the equation of motion up to second order,

\[
\frac{d}{dt}\rho_t = -i \left[ H_{II}^{eff}(t), \rho_t \right] - D_R(t) (\sigma_-\sigma_+\rho_t + \rho_t\sigma_-\sigma_+ - 2\sigma_+\rho_t\sigma_-)
- D'_R(t) (\sigma_+\sigma_-\rho_t + \rho_t\sigma_+\sigma_- - 2\sigma_-\rho_t\sigma_+) ,
\] (51)

where the second-order effective Hamiltonian \( H_{eff}^{II}(t) \) is defined in Eq.(50) and the prefactors \( D_R(t) \), \( D_I(t) \), \( D'_R(t) \), and \( D'_I(t) \) are defined in Eqs.(44, 45, 46, 47) respectively. Non-Markovian master equations like this are previously known in the literature. For example, Eq.(5) of Ref.[31] shows a similar master equation for a TLS, without the rotating wave approximation.

**Decoherence rate**

Loosely speaking, the prefactor \( D_R(t) \) (\( D'_R(t) \)) may be called “decoherence rate,” [8] which determines how fast quantum coherence (as represented by some off-diagonal element of the system’s reduced density matrix in the relevant basis) decays. By examining the formal expression of \( D_R(t) \) (\( D'_R(t) \)) as in Eq.(44) (Eq.(46)),

\[
D_R(t) = \int_0^t dt' \sum_k |g_k|^2 \tilde{N}_k \cos \left( \omega_{k0}(t - t') \right) ,
\] (52)

\[
D'_R(t) = \int_0^t dt' \sum_k |g_k|^2 \tilde{N}_k \cos \left( \omega_{k0}(t - t') \right)
+ \int_0^t dt' \sum_k |g_k|^2 \cos \left( \omega_{k0}(t - t') \right) ,
\] (53)

we have the following observations:

(a) For each occupied bosonic mode \((\tilde{N}_k \neq 0)\), its contribution to the decoherence rate depends linearly on its average occupation number \(\tilde{N}_k\). This linear dependence on occupation number is well known. See also Eq.(3.219) of Ref.[5] for another example of linear dependence on occupation number (albeit at the transition frequency, in the case of a Markovian master equation).

(b.1) For each occupied bosonic mode \((\tilde{N}_k \neq 0)\), its contribution to the decoherence rate is quadratic on its coupling strength to the system \(|g_k|\); and (b.2) in addition to the contributions from occupied modes as discussed in (a) and (b.1), all modes coupled to the system \((g_k \neq 0)\), regardless of being occupied or unoccupied, contribute to the prefactor \( D'_R(t) \) for the last term in Eq.(51), and each coupled mode’s contribution is quadratic on its coupling strength to
the system $|g_k|$. This quadratic dependence on coupling strength is also well known in the literature. For example, the second-order contribution in Eqs.(16, 33) of Ref.[16] shows another example of quadratic dependence, though with the environment initially in the vacuum state.

**Constant decoherence rate**

Generally, decoherence rates $D_R(t)$ ($D'_R(t)$) can depend on time. In many cases, however, decoherence rates are (approximately) time independent. Appendix B shows one way constant decoherence rates can be recovered. 5 (Also note that Markovian master equations usually come with constant decoherence rates, which are extensively studied in the literature. See, for example, Eq.(3.219) of Ref.[5] for a Markovian equation for a TLS.) A constant decoherence rate in turn implies exponential decay in relevant elements of the system’s reduced density matrix $\rho_L$.

**Vacuum limit**

Suppose the bosonic field is initially in the vacuum state, $\rho_{E0} = |0\rangle\langle 0|$. 6 This specific case of a TLS coupled to a bath initially in the vacuum state is previously studied in [16]. In this vacuum limit, the expected occupation number is zero for all bosonic field modes,

$$\bar{N}_k \equiv Tr_E \left( \rho_{E0} b_k^\dagger b_k \right) = 0. \quad (54)$$

---

5Note that the discussions in Appendix B regarding the evaluation of prefactors like $D_R(t)$ are not necessarily rigorous and are meant for heuristic purpose. We follow the treatments and arguments as in references [23, 24, 25], which are supposedly standard practice but are not necessarily always valid. Figures 1-3 are for illustrative purpose and are by no means accurate.

6The vacuum state may be thought of as the “thermal state” at zero temperature. Formally, the vacuum state is diagonal in the occupation number eigenbasis, therefore the derivations leading to Eq.(51) remains valid.
Plugging Eq.(54) into Eqs.(44-47), we have

\[
D_R(t) = 0, \quad (55)
\]

\[
D_I(t) = 0, \quad (56)
\]

\[
D_R'(t) = \int_0^t dt' \sum_k |g_k|^2 \cos (\omega_{k0}(t-t')) = \frac{1}{2} \gamma^{(2)}(t), \quad (57)
\]

\[
D_I'(t) = \int_0^t dt' \sum_k |g_k|^2 \sin (\omega_{k0}(t-t')) = -\frac{1}{2} S^{(2)}(t), \quad (58)
\]

where new parameters \(\gamma^{(2)}(t)\) and \(S^{(2)}(t)\) have been introduced in accordance with the notations in Eqs.(33, 16) of Ref.[16].

Plugging Eqs.(55-58) into Eqs.(50, 51), we obtain the equation of motion describing the reduced dynamics of a TLS coupled to a bosonic field initially in the vacuum state (up to second order):

\[
\frac{d}{dt} \rho_t = -i \left[ -\frac{1}{2} S^{(2)}(t) \sigma_+ \sigma_- + \rho_t \right] - \frac{1}{2} \gamma^{(2)}(t) \left( \sigma_+ \sigma_- \rho_t + \rho_t \sigma_+ \sigma_- - 2 \sigma_- \rho_t \sigma_+ \right) + \frac{1}{2} \left( \sigma_- \rho_t \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_- , \rho_t \} \right). \quad (59)
\]

Comparing Eq.(59) with Eqs.(26, 28, 33, 16) of Ref.[16], we see that our result agrees with the second-order result in [16].

### 3.3 Reduced dynamics

Now we use the second-order master equation Eq.(51) to easily get some quantitative results and gain more insights into the TLS coupled to bosonic field.

#### Differential equations for density matrix elements

To find the equations of motion for the elements \(\rho_{mn}(t)\) of the reduced density matrix \(\rho_t\), we sandwich both sides of Eq.(51) with \(\langle m| \cdots |n \rangle\) for \(m, n = 0, 1\), with the convention that \(|0\rangle\) represents spin-up and \(|1\rangle\) represents spin-down. With \(\sigma_+|0\rangle = 0, \sigma_+|1\rangle = 2|0\rangle, \sigma_-|0\rangle = 2|1\rangle,\) and \(\sigma_-|1\rangle = 0\), it can be shown that the evolution of matrix elements are governed by a system of linear ordinary
differential equations as follows,

\[
\frac{d}{dt} \rho_{00}(t) = -8D_R'(t)\rho_{00}(t) + 8D_R(t)\rho_{11}(t), \\
\frac{d}{dt} \rho_{01}(t) = i(4(D_I(t) + D_I'(t)))\rho_{01}(t) - 4(D_R(t) + D_R'(t))\rho_{00}(t), \\
\frac{d}{dt} \rho_{10}(t) = -i(4(D_I(t) + D_I'(t)))\rho_{10}(t) - 4(D_R(t) + D_R'(t))\rho_{11}(t), \\
\frac{d}{dt} \rho_{11}(t) = 8D_R'(t)\rho_{00}(t) - 8D_R(t)\rho_{11}(t),
\]

(60) (61) (62) (63)

We see that the evolution of off-diagonal element \( \rho_{01}(t) \) is governed by a (linear homogeneous) ordinary differential equation Eq.(61), that is, the dynamics of \( \rho_{01}(t) \) is decoupled from that of the other density matrix elements. The same can be said about \( \rho_{10}(t) \). For the diagonal elements \( \rho_{00}(t) \) and \( \rho_{11}(t) \), they form a system of coupled differential equations.

**General solutions for coherence**

We can solve the homogeneous linear ODE for the off-diagonals \( \rho_{01}(t) \) and \( \rho_{10}(t) \), [26]

\[
\rho_{01}(t) = \rho_{01}(0) \exp \left(i \int_0^t dt' A(t' + D_I'(t')) \right) \\
\quad \exp \left(- \int_0^t dt' A(t' + D_R'(t')) \right), \\
\rho_{10}(t) = \rho_{10}(0) \exp \left(-i \int_0^t dt' A(t' + D_I'(t')) \right) \\
\quad \exp \left(- \int_0^t dt' A(t' + D_R'(t')) \right).
\]

(64) (65)

As we can see, the first term in Eq.(61) with a pure imaginary prefactor results in a phase shift of \( \rho_{01}(t) \), as is manifest in the first exponential factor of the solution Eq.(64); the second term in Eq.(61) with a real prefactor results in a decay in the amplitude of \( \rho_{01}(t) \), as is manifest in the second exponential factor of Eq.(64). The same can be said about \( \rho_{10}(t) \). Focusing on the amplitude of \( \rho_{01}(t) \) (\( \rho_{10}(t) \)), we see that

\[
|\rho_{01}(t)| = |\rho_{01}(0)| \exp \left(- \int_0^t dt' A(t' + D_R'(t')) \right), \\
|\rho_{10}(t)| = |\rho_{10}(0)| \exp \left(- \int_0^t dt' A(t' + D_R'(t')) \right).
\]

(66) (67)

Thus we see that the coherence \( \rho_{01}(t) \) (\( \rho_{10}(t) \)) between the system’s energy
eigenlevels decay in this case.

**General solutions for populations**

To solve for the diagonals $\rho_{00}(t)$ and $\rho_{11}(t)$, that is, the spin-up and spin-down populations, we may make use of the unit trace property of density matrix, namely $\rho_{00}(t) + \rho_{11}(t) = 1$. Plugging $\rho_{11}(t) = 1 - \rho_{00}(t)$ into Eq.(60), we obtain a linear inhomogeneous ODE for $\rho_{00}(t)$,

$$\frac{d}{dt}\rho_{00}(t) = -8D_R(t)\rho_{00}(t) + 8D_R(t)(1 - \rho_{00}(t)),$$  \hspace{0.5cm} (68)

the solution to which is \[26\]

$$\rho_{00}(t) = \rho_{00}(0) \exp \left( -\int_{0}^{t} dt' 8(D_R(t') + D'_R(t')) \right) + \exp \left( -\int_{0}^{t} dt' 8(D_R(t') + D'_R(t')) \right) \times \int_{0}^{t} dt' 8D_R(t') \exp \left( \int_{0}^{t} dt'' 8(D_R(t'') + D'_R(t'')) \right).$$  \hspace{0.5cm} (70)

The spin-down population may also be obtained accordingly,

$$\rho_{11}(t) = 1 - \rho_{00}(t).$$  \hspace{0.5cm} (71)

**High temperature limit**

If the bath starts at (extremely) high temperature, the average number of bosons in the field modes are large, \[29\] that is, $\bar{N}_k \equiv Tr_E \left( \rho_E h_k^\dagger h_k \right) \gg 1$, in which case we can treat $\bar{N}_k + 1 \equiv \bar{N}_k$ in Eq.(46) for $D'_R(t)$,

$$D'_R(t) \equiv \int_{0}^{t} dt' \sum_k |g_k|^2 (\bar{N}_k + 1) \cos (\omega_{k0}(t - t'))$$

$$\approx \int_{0}^{t} dt' \sum_k |g_k|^2 \bar{N}_k \cos (\omega_{k0}(t - t'))$$

$$= D_R(t),$$  \hspace{0.5cm} (72)
and in Eq.(47) for $D'_I(t)$,

$$D'_I(t) \equiv \int_0^t dt' \sum_k |g_k|^2 (\tilde{N}_k + 1) \sin (\omega_{k0}(t-t'))$$

$$\simeq \int_0^t dt' \sum_k |g_k|^2 \tilde{N}_k \sin (\omega_{k0}(t-t'))$$

$$= D_I(t). \quad (73)$$

Plugging Eqs.(72, 73) into Eqs.(64, 65), we see that the coherence between energy eigenlevels will evolve as

$$\rho_{01}(t) = \rho_{01}(0) \exp \left( i 8 \int_0^t dt' D_I(t') \right) \exp \left( -8 \int_0^t dt' D_R(t') \right), \quad (74)$$

$$\rho_{10}(t) = \rho_{10}(0) \exp \left( -i 8 \int_0^t dt' D_I(t') \right) \exp \left( -8 \int_0^t dt' D_R(t') \right), \quad (75)$$

with the amplitudes decaying according to

$$|\rho_{01}(t)| = |\rho_{01}(0)| \exp \left( -8 \int_0^t dt' D_R(t') \right), \quad (76)$$

$$|\rho_{10}(t)| = |\rho_{10}(0)| \exp \left( -8 \int_0^t dt' D_R(t') \right). \quad (77)$$

Plugging Eqs.(72, 73) into Eq.(70), we see that the spin-up population evolves as

$$\rho_{00}(t) = \rho_{00}(0) \exp \left( -16 \int_0^t dt' D_R(t') \right) \right)$$

$$+ \frac{1}{2} \exp \left( -16 \int_0^t dt' D_R(t') \right) \int_0^t dt' 16 D_R(t') \exp \left( 16 \int_0^t dt'' D_R(t'') \right)$$

$$= \rho_{00}(0) \exp \left( -16 \int_0^t dt' D_R(t') \right)$$

$$+ \frac{1}{2} \exp \left( -16 \int_0^t dt' D_R(t') \right) \exp \left( 16 \int_0^t dt'' D_R(t'') \right) |t'_0 = 0)$$

$$= \rho_{00}(0) \exp \left( -16 \int_0^t dt' D_R(t') \right) + \frac{1}{2} \left( 1 - \exp \left( -16 \int_0^t dt' D_R(t') \right) \right)$$

$$= \frac{1}{2} + \left( \rho_{00}(0) - \frac{1}{2} \right) \exp \left( -16 \int_0^t dt' D_R(t') \right). \quad (78)$$

From Eq.(78), we may make two observations about the population at the high temperature limit:

(a) If we start at $\rho_{00}(0) = \frac{1}{2}$, it will stay at $\rho_{00}(t) = \frac{1}{2}$ subsequently. In other
words, \( \rho_{00}(t) = \frac{1}{2} \) is a steady state solution.

(b) Regardless of the initial spin-up population, even for \( \rho_{00}(0) \neq \frac{1}{2} \), as long as sufficient time passes by so that the factor \( \exp \left( -16 \int_0^t dt' D_R(t') \right) \) gets close enough to vanishing,\(^7\) we may say the spin-up population approaches the steady state solution \( \rho_{00} = \frac{1}{2} \). By Eq.(71), the spin-down population will also be \( \rho_{11} = 1 - \rho_{00} = \frac{1}{2} \) in this case.

These observations are consistent with statistical mechanics - at the high temperature limit, the energy eigenlevels should be equally populated at equilibrium. [30]

**Low temperature limit**

If the bath starts at zero temperature, where the average number of bosons in the field modes are zero, [29] that is, \( \bar{N}_k \equiv Tr_E \left( \rho_{EE} b_k^\dagger b_k \right) = 0 \), the coefficients of the linear differential equations become

\[
D_R(t) = \int_0^t dt' \sum_k |g_k|^2 \bar{N}_k \cos(\omega_{k0}(t-t')) = 0, \tag{79}
\]

\[
D_I(t) = \int_0^t dt' \sum_k |g_k|^2 \bar{N}_k \sin(\omega_{k0}(t-t')) = 0, \tag{80}
\]

\[
D'_R(t) = \int_0^t dt' \sum_k |g_k|^2 (\bar{N}_k + 1) \cos(\omega_{k0}(t-t')) \\
= \int_0^t dt' \sum_k |g_k|^2 \cos(\omega_{k0}(t-t')) \\
= D'_R(t), \tag{81}
\]

\[
D'_I(t) = \int_0^t dt' \sum_k |g_k|^2 (\bar{N}_k + 1) \sin(\omega_{k0}(t-t')) \\
= \int_0^t dt' \sum_k |g_k|^2 \sin(\omega_{k0}(t-t')) \\
= D'_I(t). \tag{82}
\]

Plugging Eqs.(79-82) into Eqs.(64, 65), we see that the coherence will now

\(^7\)Suppose that it is within the domain of applicability of our master equation formalism, namely reasonably short time and/or weak coupling, and that the second-order approximate equation of motion still holds.
evolve as
\[
\rho_{01}(t) = \rho_{01}(0) \exp \left( i 4 \int_0^t dt' D_0^0(t') \right) \exp \left( -4 \int_0^t dt' D_0^0(t') \right), \quad (83)
\]
\[
\rho_{10}(t) = \rho_{10}(0) \exp \left( -i 4 \int_0^t dt' D_0^0(t') \right) \exp \left( -4 \int_0^t dt' D_0^0(t') \right); \quad (84)
\]
and their amplitudes decaying according to
\[
|\rho_{01}(t)| = |\rho_{01}(0)| \exp \left( -4 \int_0^t dt' D_0^0(t') \right), \quad (85)
\]
\[
|\rho_{10}(t)| = |\rho_{10}(0)| \exp \left( -4 \int_0^t dt' D_0^0(t') \right). \quad (86)
\]

Plugging Eqs.(79-82) into Eq.(70), we see that the spin-up population now evolves as
\[
\rho_{00}(t) = \rho_{00}(0) \exp \left( -8 \int_0^t dt' D_0^0(t') \right). \quad (87)
\]

From Eq.(87), we may make two observations about the population at zero temperature:
(a) If we start at \( \rho_{00}(0) = 0 \), it will stay at \( \rho_{00}(t) = 0 \). In other words, \( \rho_{00}(t) = 0 \) is a steady state solution, and thus by Eq.(71) \( \rho_{11}(t) = 1 - \rho_{00}(t) = 1 \), that is, all populations being in spin-down (the energy ground state).
(b) Regardless of the initial spin-up population, even for \( \rho_{00}(0) \neq 0 \), as long as sufficient time passes by so that the factor \( \exp \left( -8 \int_0^t dt' D_0^0(t') \right) \) gets close enough to vanishing,\(^8\) we may say the spin-up population approaches the steady state solution \( \rho_{00} = 0 \), which also implies \( \rho_{11} = 1 - \rho_{00} = 1 \) by Eq.(71).

These observations are consistent with statistical mechanics - at zero temperature, the equilibrium population should be all in the ground state. [30]

4 Conclusions
We develop a formally exact master equation for open quantum systems in a succinct and intuitive way. Our derivation is direct and “by construction”. In particular, it dispenses with the use of an “inverse” map, which was used by previous derivations of formally exact master equations. Applying our formalism to the spin-boson model at arbitrary temperature, we observe non-exponential decoherence and relaxation characteristic of non-Markovian behaviors. The equation of motion obtained herein, albeit a second-order approximation, yields the right

\(^8\)Suppose that it is within the domain of applicability of our master equation formalism, namely reasonably short time and/or weak coupling, and that the second-order approximate equation of motion still holds.
steady state solution, in agreement with standard statistical mechanical pre-
dictions. The formalism can be applied to study more physical examples and
further explore its usefulness. For example, it can be used to study the dynamics
of two atoms in an optical cavity, which could have implications on two-atom
entanglement [22]. Higher-order equations of motion can also be obtained me-
chanically using Eqs.(35-38) to study corrections to second-order dynamics.

Appendix A

To derive the equation of motion for the TLS’s reduced density matrix, we first
caste the full interaction Hamiltonian Eq.(40) into the form of Eq.(28):

\[ H_{SE}(t) = S_1 \otimes E_1(t) + S_2 \otimes E_2(t), \]  

(88)

where the system operators are defined as

\[ S_1 \equiv \sigma_+, \]  

(89)

\[ S_2 \equiv \sigma_-, \]  

(90)

and we have absorbed the time dependence into the bath operators,

\[ E_1(t) \equiv \sum_k g_k e^{-i(\omega_k - \omega_0)t} b_k, \]  

(91)

\[ E_2(t) \equiv \sum_k g_k e^{i(\omega_k - \omega_0)t} b_k^\dagger. \]  

(92)

Hereafter we shall denote \( \omega_{k0} \equiv \omega_k - \omega_0 \) for convenience.

First-order term in the equation of motion

To evaluate the first-order term of the equation of motion, plugging Eqs.(89-92)
into Eq.(32) yields

\[ H_{eff}(t) = Tr_E (\rho_{E0} E_1(t)) S_1 + Tr_E (\rho_{E0} E_2(t)) S_2 \]

\[ = \sum_k g_k e^{-i\omega_{k0}t} Tr_E (\rho_{E0} b_k) S_1 \]

\[ + \sum_k g_k e^{i\omega_{k0}t} Tr_E (\rho_{E0} b_k^\dagger) S_2. \]  

(93)
The prefactor $Tr_E (\rho_E b_k)$ for an arbitrary $k$-th mode can be evaluated as

$$Tr_E (\rho_E b_k) = Tr_E \left( \frac{1}{Z_k} \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k \rangle \langle m_k| b_k \right)$$

$$\prod_{k' \neq k} Tr_{E k'} \left( \frac{1}{Z_{k'}} \sum_{m_{k'}=0}^{\infty} e^{-m_{k'} \beta \omega_{k'}} |m_{k'} \rangle \langle m_{k'}| \right)$$

$$= \frac{1}{Z_k} \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} \langle m_k | b_k | m_k \rangle$$

$$= 0. \quad (94)$$

where $Tr_{E k} (\ldots)$ denotes the partial trace over the $k$-th bosonic mode. Similarly, the prefactor $Tr_E (\rho_E b_k^\dagger)$ for an arbitrary $k$-th mode is

$$Tr_E (\rho_E b_k^\dagger) = Tr_E \left( \frac{1}{Z_k} \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k \rangle \langle m_k| b_k^\dagger \right)$$

$$= \frac{1}{Z_k} \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} \langle m_k | b_k^\dagger | m_k \rangle$$

$$= 0. \quad (95)$$

Thus the first-order effective Hamiltonian vanishes,

$$H_{eff}^I (t) = 0, \quad (96)$$

which means the system-bath interaction does not have first-order contribution to the TLS’s reduced dynamics in this case.

**Second-order term in the equation of motion**

To evaluate the second-order term of the equation of motion, plugging Eqs. (89-92) into Eq. (33) yields

$$\mathcal{L}_{2,t} (\rho) = - \sum_{m=1,2} \sum_{n=1,2} \int_0^t dt' \left( C_{mn} (t, t') [S_m, S_n \rho] - C_{nm} (t', t) [S_m, \rho S_n] \right), \quad (97)$$

where the system operators $\{S_1, S_2\}$ are now time-independent and the coefficients are defined as in Eq. (34),

$$C_{jk} (t, t') \equiv Tr_E (\rho_{E0} E_j(t) E_k(t')) - Tr_E (\rho_{E0} E_j(t)) Tr_E (\rho_{E0} E_k(t')). \quad (98)$$
To simplify Eq.(98), we note that
\[
\text{Tr}_E (\rho_{E0}E_1(t)) = \sum_k g_k e^{-i\omega_k t} \text{Tr}_E (\rho_{E0}b_k) = 0, \quad (99)
\]
\[
\text{Tr}_E (\rho_{E0}E_2(t)) = \sum_k g_k e^{i\omega_k t} \text{Tr}_E (\rho_{E0}b_k^\dagger) = 0, \quad (100)
\]
where we have made use of Eqs.(94, 95). Therefore, the coefficients are now
\[
C_{jk}(t,t') = \text{Tr}_E (\rho_{E0}E_j(t)E_k(t')). \quad (101)
\]
Let’s now evaluate Eq.(97) term by term.

For the term with \(m = n = 1\), the first coefficient is
\[
C_{11}(t,t') = \text{Tr}_E (\rho_{E0}E_1(t)E_1(t')) = \sum_k \sum_{k'} g_k g_{k'} e^{-i\omega_k t} e^{-i\omega_{k'} t'} \text{Tr}_E (\rho_{E0}b_k b_{k'}). \quad (102)
\]
We will show that the last factor \(\text{Tr}_E (\rho_{E0}b_k b_{k'})\) vanishes for arbitrary \((k, k')\).

First, for the case of \(k \neq k'\):
\[
\text{Tr}_E (\rho_{E0}b_k b_{k'}) = \text{Tr}_E \left( \prod_k \left( \frac{1}{Z_k Z_{k'}} \sum_{m_K=0}^{\infty} e^{-m_k \beta \omega_k} |m_K\rangle \langle m_K| b_k b_{k'} \right) \right)
\]
\[
= \frac{1}{Z_k Z_{k'}} \text{Tr}_E \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle m_k| b_k \right) \text{Tr}_{E_{k'}} \left( \sum_{m_{k'}=0}^{\infty} e^{-m_{k'} \beta \omega_{k'}} |m_{k'}\rangle \langle m_{k'}| b_{k'} \right)
\]
\[
= \frac{1}{Z_k Z_{k'}} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} \langle m_k| b_k |m_k\rangle \right)
\]
\[
\left( \sum_{m_{k'}=0}^{\infty} e^{-m_{k'} \beta \omega_{k'}} \langle m_{k'}| b_{k'} |m_{k'}\rangle \right)
\]
\[
= 0, \quad (103)
\]
because \(\langle m_k| b_k |m_k\rangle \propto \langle m_k + 1| m_k\rangle = 0\) vanishes for an arbitrary \(k\); second, for
the case of $k = k'$:

$$
\text{Tr}_E (\rho_E b_k b_k) = \frac{1}{Z_k} \text{Tr}_E \left( \sum_{m_k=0}^\infty e^{-m_k \beta \omega_k} |m_k\rangle \langle b_k b_k| \right)
$$

$$
= \frac{1}{Z_k} \left( \sum_{m_k=0}^\infty e^{-m_k \beta \omega_k} \langle m_k| b_k b_k |m_k\rangle \right)
$$

$$
= 0,
$$

(104)

because $\langle m_k| b_k b_k |m_k\rangle \propto \langle m_k+2| m_k\rangle = 0$ vanishes for an arbitrary $k$. Therefore, we have shown

$$
C_{11}(t, t') = 0.
$$

(105)

Similarly, the second coefficient is

$$
C_{11}(t', t) = \text{Tr}_E (\rho_E E_1(t') E_1(t))
$$

$$
= \sum_k \sum_{k'} g_k g_k' e^{-i \omega_k t'} e^{-i \omega_{k'} t} \text{Tr}_E (\rho_E b_k b_{k'})
$$

$$
= 0.
$$

(106)

Therefore, the term for $m = n = 1$ in Eq.(97) vanishes.

Similarly, for the term with $m = n = 2$, the first coefficient is

$$
C_{22}(t, t') = \text{Tr}_E (\rho_E E_2(t) E_2(t'))
$$

$$
= \sum_k \sum_{k'} g_k g_k' e^{i \omega_k t} e^{i \omega_{k'} t'} \text{Tr}_E \left( \rho_E \hat{b}_k \hat{b}_{k'}^\dagger \right)
$$

$$
= 0,
$$

(107)

as it can be similarly shown that $\text{Tr}_E \left( \rho_E \hat{b}_k \hat{b}_{k'}^\dagger \right) = 0$ for arbitrary $(k, k')$. Likewise, the second coefficient can be shown to vanish, $C_{22}(t', t) = 0$. Therefore, the term for $m = n = 2$ in Eq.(97) vanishes.

Thus we are left with the cross terms with $(m = 1, n = 2)$ and $(m = 2, n = 1)$ in Eq.(97):

$$
L_{2,t} (\rho) = - \int_0^t dt' \left\{ C_{12}(t, t') \left[ S_1, S_2 \rho \right] - C_{21}(t', t) \left[ S_1, \rho S_2 \right] + C_{21}(t, t') \left[ S_2, S_1 \rho \right] - C_{12}(t', t) \left[ S_2, \rho S_1 \right] \right\}
$$

$$
= - \int_0^t dt' \left\{ C_{12}(t, t') (\sigma_+ \sigma_- - \sigma_- \rho \sigma_+) + C_{12}(t', t) (\rho \rho_+ \sigma_- - \sigma_- \rho \sigma_+) + C_{21}(t, t') (\sigma_- \sigma_+ - \sigma_+ \rho \sigma_-) + C_{21}(t', t) (\rho \sigma_- \sigma_- - \sigma_- \rho \sigma_-) \right\},
$$

(108)

where in the second equality we have rearranged the order of the terms. The prefactor of each term in Eq.(108) will be evaluated as follows.
For the first term,

\[
C_{12}(t,t') = Tr_E (\rho E_1(t) E_2(t'))
= \sum_k \sum_{k'} g_k g_{k'} e^{-i\omega_k t} e^{i\omega_{k'} t'} Tr_E \left( \rho E_0 b_k^{\dagger} b_{k'} \right),
\]
(109)

where the factor \(Tr_E \left( \rho E_0 b_k^{\dagger} b_{k'} \right)\) is, for \(k \neq k'\):

\[
Tr_E \left( \rho E_0 b_k^{\dagger} b_{k'} \right) = \frac{1}{Z_k Z_{k'}} Tr_{E_k} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle m_k| b_k \right)
\]

\[
= \frac{1}{Z_k Z_{k'}} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle b_k^{\dagger} | b_k \right)
\]

\[
= 0,
\]
(110)

and for \(k = k'\):

\[
Tr_E \left( \rho E_0 b_k^{\dagger} b_k \right) = \frac{1}{Z_k} Tr_{E_k} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle b_k^{\dagger} b_k | m_k \right)
\]

\[
= \frac{1}{Z_k} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle b_k^{\dagger} b_k | m_k \right)
\]

\[
= \frac{1}{Z_k} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle b_k^{\dagger} b_k | m_k \right) + \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k}
\]

\[
= \bar{N}_k + 1,
\]
(111)

where we have denoted the average occupation number in the \(k\)-th mode of the bath as

\[
\bar{N}_k \equiv Tr_E \left( \rho E_0 b_k^{\dagger} b_k \right) = \frac{1}{Z_k} \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} \langle m_k | b_k^{\dagger} b_k | m_k \rangle;
\]
(112)
plugging Eqs. (110, 111) into Eq. (109) yields

\[
C_{12}(t, t') = \sum_k \sum_{k'} g_k g_{k'} e^{-i\omega_k t} e^{i\omega_{k'} t'} \text{Tr}_E \left( \rho_{E0} b_k b_{k'}^\dagger \right)
\]

\[
= \sum_k |g_k|^2 e^{-i\omega_k (t-t')} \text{Tr}_E \left( \rho_{E0} b_k b_{k}^\dagger \right)
\]

\[
= \sum_k |g_k|^2 \left( \bar{N}_k + 1 \right) e^{-i\omega_k (t-t')}
\]

\[
= \sum_k |g_k|^2 \left( \bar{N}_k + 1 \right) \left( \cos (\omega_k (t - t')) - i \sin (\omega_k (t - t')) \right)\] \hspace{1cm} (113)

Similarly, for the second term,

\[
C_{12}(t', t) = \text{Tr}_E \left( \rho_{E0} E_1(t') E_2(t) \right)
\]

\[
= \sum_k \sum_{k'} g_k g_{k'} e^{-i\omega_k t'} e^{i\omega_{k'} t} \text{Tr}_E \left( \rho_{E0} b_k b_{k'}^\dagger \right),
\]

\[
= \sum_k |g_k|^2 e^{i\omega_k (t-t')} \text{Tr}_E \left( \rho_{E0} b_k b_{k}^\dagger \right)
\]

\[
= \sum_k |g_k|^2 \left( \bar{N}_k + 1 \right) e^{i\omega_k (t-t')}
\]

\[
= \sum_k |g_k|^2 \left( \bar{N}_k + 1 \right) \left( \cos (\omega_k (t - t')) + i \sin (\omega_k (t - t')) \right)\] \hspace{1cm} (114)

where in the third and fourth equalities we have made use of Eqs. (110, 111).

For the third term,

\[
C_{21}(t, t') = \text{Tr}_E \left( E_2(t) E_1(t') \rho_{E0} \right)
\]

\[
= \sum_k \sum_{k'} g_k g_{k'} e^{i\omega_k t} e^{-i\omega_{k'} t'} \text{Tr}_E \left( \rho_{E0} b_k b_{k'}^\dagger \right),
\]

\[
= \sum_k \sum_{k'} g_k g_{k'} e^{i\omega_k t} e^{-i\omega_{k'} t'} \text{Tr}_E \left( \rho_{E0} b_k b_{k'}^\dagger \right), \hspace{1cm} (115)
\]

where the factor \( \text{Tr}_E \left( \rho_{E0} b_k b_{k'}^\dagger \right) \) is, for \( k \neq k' \):
\[
\text{Tr}_E \left( \rho_{E0} b_k^\dagger b_{k'} \right) = \frac{1}{Z_k Z_{k'}} \text{Tr}_{E_k} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle m_k| b_{k'}^\dagger \right)
\]
\[
\text{Tr}_{E_{k'}} \left( \sum_{m_{k'}=0}^{\infty} e^{-m_{k'} \beta \omega_{k'}} |m_{k'}\rangle \langle m_{k'}| b_k \right)
\]
\[
= \frac{1}{Z_k Z_{k'}} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} \langle m_k| b_{k'}^\dagger |m_{k'}\rangle \right)
\]
\[
\left( \sum_{m_{k'}=0}^{\infty} e^{-m_{k'} \beta \omega_{k'}} \langle m_{k'}| b_k |m_k\rangle \right)
\]
\[
= 0,
\]
(116)

and for \( k = k' \):
\[
\text{Tr}_E \left( \rho_{E0} b_k^\dagger b_k \right) = \frac{1}{Z_k} \text{Tr}_{E_k} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} |m_k\rangle \langle m_k| b_k^\dagger b_k \right)
\]
\[
= \frac{1}{Z_k} \left( \sum_{m_k=0}^{\infty} e^{-m_k \beta \omega_k} \langle m_k| b_k^\dagger b_k |m_k\rangle \right)
\]
\[
= \bar{N}_k; \tag{117}
\]

plugging Eqs.(116, 117) into Eq.(115) yields
\[
C_{21}(t, t') = \sum_k \sum_{k'} g_k g_{k'} e^{i \omega_{k0} t} e^{-i \omega_{k'0} t'} \text{Tr}_E \left( \rho_{E0} b_k^\dagger b_{k'} \right)
\]
\[
= \sum_k |g_k|^2 e^{i \omega_{k0} (t - t')} \text{Tr}_E \left( \rho_{E0} b_k^\dagger b_k \right)
\]
\[
= \sum_k |g_k|^2 \bar{N}_k e^{i \omega_{k0} (t - t')}
\]
\[
= \sum_k |g_k|^2 \bar{N}_k \left( \cos \left( \omega_{k0} (t - t') \right) + i \sin \left( \omega_{k0} (t - t') \right) \right). \tag{118}
\]
Similarly, for the fourth term,

\[
\mathcal{C}_{21}(t', t) = Tr_E \left( E_2(t')E_1(t)\rho_{E0} \right)
\]

\[
= \sum_k \sum_{k'} g_k g_{k'} e^{i\omega_{k0} t} e^{-i\omega_{k'0} t'} Tr_E \left( \rho_{E0} b_k^\dagger b_{k'} \right)
\]

\[
= \sum_k |g_k|^2 e^{-i\omega_{k0}(t-t')} Tr_E \left( \rho_{E0} b_k^\dagger b_k \right)
\]

\[
= \sum_k |g_k|^2 \bar{N}_k e^{-i\omega_{k0}(t-t')}
\]

\[
= \sum_k |g_k|^2 \bar{N}_k \cos (\omega_{k0}(t-t')) - i \sin (\omega_{k0}(t-t')) ,
\]

where in the third and fourth equalities we have made use of Eqs.(116, 117).

Now, for convenience, let’s introduce the following notations:

\[
D_R(t) \equiv \int_0^t dt' \sum_k |g_k|^2 \bar{N}_k \cos (\omega_{k0}(t-t')) ,
\]

\[
D_I(t) \equiv \int_0^t dt' \sum_k |g_k|^2 \bar{N}_k \sin (\omega_{k0}(t-t')) ,
\]

\[
D'^R(t) \equiv \int_0^t dt' \sum_k |g_k|^2 (\bar{N}_k + 1) \cos (\omega_{k0}(t-t')) ,
\]

\[
D'^I(t) \equiv \int_0^t dt' \sum_k |g_k|^2 (\bar{N}_k + 1) \sin (\omega_{k0}(t-t')) ,
\]

with which the prefactors can be rewritten as

\[
\int_0^t dt' \mathcal{C}_{12}(t', t') = D'^R(t) - iD'^I(t) ,
\]

\[
\int_0^t dt' \mathcal{C}_{12}(t', t) = D'^R(t) + iD'^I(t) ,
\]

\[
\int_0^t dt' \mathcal{C}_{21}(t', t) = D_R(t) + iD_I(t) ,
\]

\[
\int_0^t dt' \mathcal{C}_{21}(t', t) = D_R(t) - iD_I(t) .
\]

Plugging the prefactors Eqs.(124-127) into Eq.(108) and combining terms with like prefactors, we have

\[
\mathcal{L}_{2,t} (\rho) = -D_R(t) \left( \sigma_-\sigma_\rho + \rho \sigma_-\sigma_+ - 2\sigma_+\rho \sigma_- \right)
\]

\[
- D'_R(t) \left( \sigma_+\sigma_-\rho + \rho \sigma_+\sigma_- - 2\sigma_-\rho \sigma_+ \right)
\]

\[
- i \left( D_I(t) \left[ \sigma_-\sigma_+, \rho \right] - D'_I(t) \left[ \sigma_+\sigma_-, \rho \right] \right) .
\]

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We can further evaluate the prefactors in Eq.(51), for example, Appendix B where the second-order effective Hamiltonian is defined as

\[ H_{eff}^I(t) = D_I(t)\sigma_-\sigma_+ - D_I'(t)\sigma_+\sigma_-. \] (130)

Appendix B

We can further evaluate the prefactors in Eq.(51), for example,

\[ D_R(t) = \int_0^t dt' \sum_k |g_k|^2 \tilde{N}_k \cos(\omega_{k0}(t - t')) \]
\[ = \int_0^t dt' \sum_k |g_k|^2 \tilde{N}_k \text{Re} \left( e^{i\omega_{k0}(t-t')} \right). \] (131)

Integrand

First, let’s examine the integrand in Eq.(131) \( \sum_k |g_k|^2 \tilde{N}_k \cos(\omega_{k0}(t - t')) = \sum_k |g_k|^2 \tilde{N}_k \text{Re} \left( e^{i\omega_{k0}(t-t')} \right) \) as a function of \( t' \), as shown in Figure 1. The integrand is peaked around \( t' = t \), loosely because of the following reasons. On the one hand, at \( t' = t \), the factor \( \cos(\omega_{k0}(t - t)) = 1 \) for all \( k \)'s, therefore the sum \( \sum_k |g_k|^2 \tilde{N}_k \cos(\omega_{k0}(t - t')) \) consists of positive terms \( |g_k|^2 \tilde{N}_k \), all of which add up constructively, leading to the peak at \( t' = t \). On the other hand, at \( t' \neq t \), the factor \( \cos(\omega_{k0}(t - t')) \) oscillates across various \( k \)'s, therefore contributions from various terms with different \( k \)'s tend to cancel out each other. Loosely speaking, the larger \( |t - t'| \) is, the more oscillatory the factor \( \cos(\omega_{k0}(t - t')) \) becomes with respect to different \( k \)'s, the more “destructively” the various terms \( |g_k|^2 \tilde{N}_k \cos(\omega_{k0}(t - t')) \) interfere with one another, the smaller the sum \( \sum_k |g_k|^2 \tilde{N}_k \cos(\omega_{k0}(t - t')) \) becomes. This loosely explains the shape of the integrand \( \sum_k |g_k|^2 \tilde{N}_k \cos(\omega_{k0}(t - t')) \) as a function of \( t' \). (See [24] for similar discussions regarding the peak of the integrand \( \sum_k |g_k|^2 \tilde{N}_k \text{Re} \left( e^{i\omega_{k0}(t-t')} \right) \).)

Integral for short time

Next, evaluating the integral \( D_R(t) = \int_0^t dt' \sum_k |g_k|^2 \tilde{N}_k \cos(\omega_{k0}(t - t')) \) from 0 to \( t \) amounts to finding the area under the curve from \( t' = 0 \) to \( t' = t \), as represented by the shaded area in Figures 2 and 3.

For very short time, as shown in Figure 2, the shaded area increases (almost) linearly with \( t \). This is because the curve (i.e. the integrand as a differentiable
function of \( t' \) is flat in the neighborhood of its maximum \( t' = t \).

**Integral for long time - constant decoherence rate**

For longer time, as shown in Figure 3, the shaded area stays (almost) constant despite the increase of \( t \), because the left tail of the curve has a negligible area. Therefore, we may legitimately extend the lower limit of the integral from \( t' = 0 \) to \( t' = -\infty \) (almost) without changing the shaded area. (See [24] for similar discussions on extending the limit of the integral to infinity.) In doing so, we formally make \( D_R(t) \) a constant:

\[
D_R(t) = \int_0^t dt' \sum_k |g_k|^2 \bar{N}_k \cos(\omega_{k0}(t - t'))
\]

\[
= \int_{-\infty}^t dt' \sum_k |g_k|^2 \bar{N}_k \cos(\omega_{k0}(t - t'))
\]

\[
= \int_{-\infty}^t dt' \sum_k |g_k|^2 \bar{N}_k \text{Re} \left( e^{i\omega_{k0}(t-t')} \right)
\]

\[
= \text{Re} \left( \int_{-\infty}^t dt' \sum_k |g_k|^2 \bar{N}_k e^{i\omega_{k0}(t-t')} \right)
\]

\[
= \sum_k |g_k|^2 \bar{N}_k \text{Re} \left( \int_{-\infty}^t dt' e^{i\omega_{k0}(t-t')} \right)
\]

\[
= \sum_k |g_k|^2 \bar{N}_k \text{Re} \left( -\int_{-\infty}^0 d\tau e^{i\omega_{k0}\tau} \right)
\]

\[
= \sum_k |g_k|^2 \bar{N}_k \text{Re} \left( \int_{0}^{+\infty} d\tau e^{i\tau\omega_{k0}} \right)
\]

\[
= \sum_k |g_k|^2 \bar{N}_k \pi \delta (-\omega_{k0})
\]

\[
= \pi \sum_k |g_k|^2 \bar{N}_k \delta (\omega_0 - \omega_k), \quad (132)
\]

where in the sixth line we have made the change of variable \( \tau = t - t' \) and in the eighth line we have invoked the equality \( \int_0^{+\infty} dk e^{-ikx} = \pi \delta(x) - i \text{Pr}_{2}^{\frac{1}{2}} \).

[23, 24] To facilitate further calculation of Eq. (132), we follow the treatment in
and invoke the following change of variable - for an arbitrary function $f(k)$:

\[
\sum_k f(k) = \sum_k \Delta k f(k) = \sum_k \frac{\Delta k}{\Delta \omega} \Delta \omega f(k) = \sum_k \frac{\Delta k}{\Delta \omega} \Delta \omega f[k(\omega)] = \int \rho(\omega) d\omega f[k(\omega)],
\]

(133)

where $\rho(\omega) \equiv \Delta k / \Delta \omega$ is the density of states per energy/frequency. Thus we have:

\[
D_R(t) \cong \pi \sum_k |g_k|^2 \tilde{N}_k \delta(\omega_0 - \omega_k) = \pi \int_{-\infty}^{+\infty} \rho(\omega) d\omega |g_{k(\omega)}|^2 \tilde{N}_{k(\omega)} \delta(\omega_0 - \omega) = \pi \rho(\omega_0) |g_{k(\omega_0)}|^2 \tilde{N}_{k(\omega_0)},
\]

(134)

where $k(\omega_0)$ indicates the $k$-th bosonic mode that has frequency $\omega = \omega_0$. In a similar fashion, the other prefactor $D'_R(t)$ is found to be:

\[
D'_R(t) \cong \pi \rho(\omega_0) |g_{k(\omega_0)}|^2 (\tilde{N}_{k(\omega_0)} + 1).
\]

(135)

Note that both prefactors become (almost) constant in this case.

Thus, in the longer time regime, the second-order equation of motion becomes:

\[
\frac{d}{dt} \rho_t \cong \text{unitary term} - D_R (\sigma_- \sigma_+ \rho_t + \rho_t \sigma_- \sigma_+ - 2\sigma_+ \rho_t \sigma_-) - D'_R (\sigma_- \sigma_- \rho_t + \rho_t \sigma_+ \sigma_- - 2\sigma_- \rho_t \sigma_+),
\]

(136)

where the constant decoherence rate is $D_R \equiv \pi \rho(\omega_0) |g_{k(\omega_0)}|^2 \tilde{N}_{k(\omega_0)}$ ($D'_R \equiv \pi \rho(\omega_0) |g_{k(\omega_0)}|^2 (\tilde{N}_{k(\omega_0)} + 1)$). This form is consistent with the Markovian master equation for a TLS as in Eq.(3.219) of Ref.[5].

Therefore, for a TLS interacting with multiple bosonic modes at a broad spectrum of frequencies, we have recovered the (almost) constant decay/decoherence rate in the longer time regime. A constant decay/decoherence rate also implies exponential decay in the relevant density matrix element(s).

\footnote{We ignore the treatment of the unitary term here, because the main purpose of our discussion is on the issue of decay/decoherence.}
Figure 1. The integrand $\sum_k |g_k|^2 \bar{N}_k \cos(\omega_{k0}(t - t'))$ as a function of $t'$ is peaked at $t' = t$.

Figure 2. For small $t$, the shaded area grows (almost) linearly with $t$, because the curve, being a differentiable function of $t'$, is flat in the neighborhood of its maximum $t' = t$. 
Figure 3. For large $t$, the shaded area stays (almost) constant, because the left tail of the integral for $t' < 0$ is negligible.

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