Exact quantum master equation via the calculus on path integrals

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An exact quantum master equation formalism is constructed for the efficient evaluation of quantum non-Markovian dissipation beyond the weak system-bath interaction regime in the presence of time-dependent external field. A novel truncation scheme is further proposed and compared with other approaches to close the resulting hierarchically coupled equations of motion. The interplay between system-bath interaction strength, non-Markovian property, and required level of hierarchy is also demonstrated with the aid of simple spin-boson systems.

The key quantity in quantum statistical dynamics is the reduced density operator, \( \rho(t) \equiv \text{tr}_B \rho(t) \), i.e., the partial trace of the total density operator over the bath space. It is well known that the exact evolution of \( \rho(t) \) can be formulated in terms of path integral functional assuming the harmonic linear coupling bath model. Its numerical implementation is however much tedious in comparison with that of the differential quantum master equation formulations. Tanimura et al.\(^{1,2} \) had constructed exact quantum Fokker-Planck equations from path integral formulations based on a Gaussian-Markovian bath model. More recently, the construction of exact quantum master equations has been achieved by exploiting various stochastic approaches to dissipative dynamics.\(^{3,4,5,6,7,8} \)

In this article, we generalize the Tanimura-Kubo’s method\(^{1,2} \) to arbitrary cases and construct an exact differential formulation of \( \rho(t) \) based on rather simple calculus on the path integral functional. Let us start with the review of the path integral formulation. The total Hamiltonian assumes \( H_T(t) = H(t) - \sum_a Q_a F_a(t) \). Here, \( H(t) \) is the deterministic part governing the coherent motion of the reduced system, and the system-bath interaction is characterized in terms of the coupling between system operators \( \{Q_a\} \) and stochastic bath operators \( \{F_a(t) \equiv e^{i\beta H_{\text{bath}}} F_a e^{-i\beta H_{\text{bath}}} \} \). For an initial \( \rho(t_0) = \rho(t_0) \rho_{\text{bath}}^\text{eq} \), with \( \rho_{\text{bath}}^\text{eq} \propto e^{-\beta H_{\text{bath}}} \) being the thermal bath density operator, we have \( [h \equiv 1 \text{ and } \beta \equiv 1/(k_B T) \) hereafter\]

\[
\rho(t) = \text{tr}_B \left[ U_T(t, t_0; \{F_a(t)\}) \rho(t_0) e^{-\beta H_{\text{bath}}} \times U_T^\dagger(t, t_0; \{F^\dagger_a(t)\}) e^{\beta H_{\text{bath}}} \rho_{\text{bath}}^\text{eq} \right],
\]

where \( U_T(t, t_0; \{F_a(t)\}) \) is the Hilbert-space propagator with the total Hamiltonian \( H_T(t) \). Noting that \( e^{\beta H_{\text{bath}}} F_a(t) e^{-\beta H_{\text{bath}}} = F_a(t - i\beta) \), Eq. (1) can be recast as

\[
\rho(t) = \langle \langle U_T(t, t_0; \{F_a(t - i\beta)\}) \rangle \rangle \rho(t_0) U_T^\dagger(t, t_0; \{F^\dagger_a(t)\})), \quad \text{(2)}
\]

Here, \( \langle \cdot \rangle \equiv \text{tr}_B (\cdot \rho_{\text{bath}}^\text{eq}) \). Denote

\[
\rho(t) \equiv \mathcal{U}(t, t_0) \rho(t_0), \quad \text{(3)}
\]

which defines the dissipative Liouville-space propagator \( \mathcal{U}(t, t_0) \). Let \( \Gamma \equiv \{\alpha, \alpha'\} \) be an arbitrary Liouville-space representation for the reduced system. The path integral formulation for \( \mathcal{U}(\Gamma_i, t; \Gamma_0, t_0) \equiv \langle \langle \mathcal{U}(\Gamma_i(t, t_0)|\Gamma_0) \rangle \rangle \) is then

\[
\mathcal{U}(\Gamma_i, t; \Gamma_0, t_0) = \int_{\Gamma_0}^{\Gamma_i} D\Gamma e^{iS[\alpha]} \mathcal{F}[\alpha, \alpha'] e^{-iS[\alpha']}, \quad \text{(4)}
\]

Here, \( S[\alpha] \) and \( \mathcal{F}[\alpha, \alpha'] \) denote the action and influence functionals, respectively. The latter is given as [cf. Eq. (4)]

\[
\mathcal{F}[\alpha, \alpha'] = \left\langle \exp_+ \left\{ i \sum_a \int_{t_0}^{t} \text{d}\tau Q_a[\alpha(\tau)] F_a(\tau - i\beta) \right\} \times \exp_- \left\{ -i \sum_a' \int_{t_0}^{t} \text{d}\tau Q_{a'}[\alpha'(\tau)] F_{a'}(\tau) \right\} \right\rangle \quad \text{(5)}
\]

For the stochastic bath operators \( \{F_a(t)\} \) that satisfy the Gaussian statistics, the above equation can be recast in terms of the standard Feynman-Vernon form

\[
\mathcal{F}[\alpha, \alpha'] = \exp \left\{ -\sum_a \int_{t_0}^{t} \text{d}\tau \left\{ Q_a[\alpha(\tau)] - Q_{a'}[\alpha'(\tau)] \right\} \times \left\{ \hat{Q}_a[\alpha(\tau)] - \hat{Q}_{a'}^*[\alpha'(\tau)] \right\} \right\} \quad \text{(6)}
\]

with

\[
\hat{Q}_a[\alpha(t)] = \sum_b \int_{t_0}^{t} \text{d}\tau C_{ab}(t - \tau) Q_b[\alpha(\tau)]. \quad \text{(7)}
\]
Here, $C_{ab}(t - \tau) \equiv \langle F_a(t) F_b(\tau) \rangle$ are the bath correlation functions, which satisfy the symmetry and detailed-balance relations $C^{*}_{ab}(t) = C_{ba}(-t) = C_{ba}(t - i\beta)$.

We are now in the position to derive the exact quantum master equation via a rather simple calculus on the above integral functional formulation. For clarity, we shall in the following consider only the single dissipative mode case, in which the system-bath interaction assumes $H'(t) = -QF(t)$. Consider the time derivative of Eq. (10) term by term. Firstly,

$$\frac{\partial}{\partial t} e^{iS[\alpha]} = -i \int d\alpha H(t, \alpha) e^{iS[\alpha]} \equiv -i H(t) \cdot e^{iS[\alpha]}.$$  

(8)

The last identity defines the notion used in this paper. We have also

$$\frac{\partial}{\partial t} e^{-iS[\alpha']} = i e^{-iS[\alpha']} \cdot H(t).$$  

(9)

Similarly,

$$\frac{\partial}{\partial t} F = -Q \cdot ((\tilde{Q}_t - \tilde{Q}_t^{*}) F) + ([\tilde{Q}_t - \tilde{Q}_t^{*}) F] \cdot Q.$$  

(10)

with

$$\tilde{Q}_t \equiv \int_{t_0}^{t} d\tau C(t - \tau)Q[\alpha(\tau)].$$  

(11)

Together with Eqs. (3) and (4), we obtain

$$\dot{\rho}(t) = -i[H(t), \rho(t)] - i[Q, \rho(t)].$$  

(12)

Here, $\rho_1(t) \equiv \mathcal{U}_1(t, t_0) \rho(t_0)$, or generally,

$$\rho_n(t) \equiv \mathcal{U}_n(t, t_0) \rho(t_0),$$  

(13)

with $\mathcal{U}_n(t, t_0)$ being given in terms of path integral as

$$\mathcal{U}_n(\Gamma_t, t; \Gamma_0, t_0) = \int_{\Gamma_0}^{\Gamma_t} D\Gamma e^{iS[\alpha]} F_n[\alpha, \alpha'] e^{-iS[\alpha']},$$  

(14)

and

$$F_n[\alpha, \alpha'] \equiv (-i)^n (\tilde{Q}_t - \tilde{Q}_t^{*})^n F[\alpha, \alpha'].$$  

(15)

Note that $\rho_0(t) = \rho(t)$. We have further [cf. Eq. (10)]

$$\frac{\partial}{\partial t} F_n[\alpha, \alpha'] =$$

$$= -i \{ Q \cdot F_{n+1}[\alpha, \alpha'] + F_{n+1}[\alpha, \alpha'] \cdot Q \} - i n \{ C(0) Q \cdot F_{n-1}[\alpha, \alpha'] - C^{*}(0) F_{n-1}[\alpha, \alpha'] \cdot Q \} + n \tilde{F}_n[\alpha, \alpha'],$$

(16)

where

$$\tilde{F}_n[\alpha, \alpha'] \equiv -i F_{n-1}[\alpha, \alpha'] \int_{t_0}^{t} d\tau \{ \dot{C}(t - \tau) Q[\alpha(\tau)] - C^{*}(t - \tau) Q[\alpha'(\tau)] \}. $$  

(17)

We have therefore for $\rho_n; n \geq 0$ [cf. Eqs. (13) and (14)]

$$\dot{\rho}_n(t) = -i[H(t), \rho_n(t)] - i[Q, \rho_{n+1}(t)] - i n \{ C(0) \rho_{n-1}(t) - C^{*}(0) \rho_{n-1}(t) Q \} + n \tilde{\rho}_n(t).$$  

(18)

Here,

$$\tilde{\rho}_n(t) \equiv \tilde{\mathcal{U}}_n(t, t_0) \rho(t_0),$$  

(19)

with $\tilde{\mathcal{U}}_n(t, t_0)$ being given in terms of path integral as

$$\tilde{\mathcal{U}}_n(\Gamma_t, t; \Gamma_0, t_0) = \int_{\Gamma_0}^{\Gamma_t} D\Gamma e^{iS[\alpha]} \tilde{F}_n[\alpha, \alpha'] e^{-iS[\alpha']}.$$  

(20)

Both $\rho_n$ [Eq. (15)] with Eqs. (14) and (15) and $\tilde{\rho}_n$ [Eq. (19)] with Eqs. (14) and (17) are of the $(2n)^{th}$-order in the system-bath interaction as their leading contributions. In Eq. (15), $\rho_n(t)$ depends on $\rho_{n+1}(t)$ and $\tilde{\rho}_n(t)$. The latter however does not belong to the same hierarchy. To proceed, we shall first convert Eqs. (15) into a set of hierarchically coupled equations of motion (EOM), followed by a proper truncation scheme to close the resulting EOM. Note that in some special cases, such as the driven Brownian oscillators 10,11,12 and the pure-dephasing (non-demolishing) dynamics, the exact $\rho_1(t)$ can be obtained in terms of $\rho_0(t) = \rho(t)$; thus the non-hierarchical exact quantum master equation can be constructed.

The hierarchicalization of Eqs. (15) can be carried out with certain forms of bath correlation function. Consider, for example, a Gaussian-Markovian bath with $C(t) = \gamma e^{-\gamma t}$, where $\gamma$ is real and $\eta$ may be complex. In this case, $\rho_n(t) = -\gamma \rho_{n-1}(t)$ and Eqs. (15) become completely hierarchical, recovering the Fokker-Planck equation obtained previously by Tanimura and co-workers.

Consider another model in which the bath spectral density assumes $J(\omega) \propto \omega/(\omega^2 + \gamma^2)^2$, and the resulting bath correlation is found to be of $C(t) = \nu_0 e^{-\nu t} + \nu_1 e^{-\nu t}$, if the Matsubara terms can be neglected. Here, $\gamma$ is real and $\nu_0$ and $\nu_1$ are complex. The hierarchically coupled EOM equivalent to Eq. (15) can readily be constructed as follows. For the present model of $C(t)$, we have $\dot{Q}_t = \nu_0 \dot{Q}_0 + \nu_1 \dot{Q}_1$, [cf. Eq. (11)], where $\dot{Q}_0 = \int_{t_0}^{t} d\tau (t - \tau) e^{-\gamma(t-\tau)} Q[\alpha(\tau)]$ and $\dot{Q}_1 = \int_{t_0}^{t} d\tau e^{-\gamma(t-\tau)} Q[\alpha(\tau)]$. Consequently, $F_n$ of Eq. (15) can be expressed in terms of $F_{kj}^{k'j'} = \tilde{Q}_0^{k'}(Q_0)_{k'}^{j'} \tilde{Q}_1^{j'} F$, with $k + j + k' + j' = n$. Introduce now $\rho_{kj}^{k'j'}$ similarly as Eqs. (15) and (17) but with $F_n$ there being replaced by $F_{kj}^{k'j'}$. Note that $\partial Q_0/\partial t = \dot{Q}_1 - \gamma \dot{Q}_0$ and $\partial Q_1/\partial t = Q_1 - \gamma \dot{Q}_1$. The hierarchical EOM for $\rho_{kj}^{k'j'}$;
with $\rho_{00}^\gamma \equiv \rho$, can then be obtained as

$$\kappa_{kj}^{\gamma j} = -i[H(t), \rho_{kj}^{\gamma j}] - (k + j + k' + j')\gamma\rho_{kj}^{\gamma j}$$

$$- [Q, \nu_0 \rho_{kj}^{\gamma j} - \nu_0 \rho_{kj}^{\gamma j}]$$

$$- [Q, \nu_1 \rho_{kj}^{\gamma j} - \nu_1 \rho_{kj}^{\gamma j}]$$

$$+ k \rho_{kj-1,j+1} + k' \rho_{kj}^{\gamma j}$$

$$+ j \rho_{kj}^{\gamma j} - j \rho_{kj}^{\gamma j} Q. \tag{21}$$

Obviously, $\rho_{kj}^{\gamma j}$ is of the $[2(k + j + k' + j')]^{\text{th}}$-order in the system-bath interaction as its leading contribution. In general, the bath correlation function $C(t)$ can be expressed in terms of a series of exponential expansion, and thus, by following the similar procedure as shown above, one can construct a hierarchy of coupled EOM based on Eq. (13).

To complete the formulation, we propose in the following a novel truncation scheme and compare it with other two existing approaches to close the hierarchical EOM equivalent to Eq. (13). As shown earlier $\rho_0(t)$ in Eq. (13) is of the same order as $\rho_n(t)$, we need only to approximately anchor $\rho_n$ in terms of $\rho_{n<N}$, such that the system-bath interactions are accounted rigorously for up to $(2N)^{\text{th}}$ order, but partially for higher-order contributions. To do that, let us first recast Eq. (15) as

$$\rho_{n<N}= -i(\tilde{Q}_{n} - \tilde{Q}_{n}'')
\rho_{n-1},$$

and then approximate the involving $\tilde{Q}_i$ with its bath-free-evolution counterpart; i.e.,

$$\tilde{Q}_{\text{trun}}(t) \equiv \int_{0}^{t} dt C(t - \tau)\mathcal{G}(t, \tau)Q. \tag{22}$$

Here, $\mathcal{G}(t, \tau)$ denotes the dissipation-free propagator that satisfies $\partial \mathcal{G}(t, \tau)/\partial t = -i\mathcal{L}(t)\mathcal{G}(t, \tau)$, with the reduced system Liouvillean $\mathcal{L}(t) \equiv [H(t), \cdot]$. The resulting truncation scheme proposed here amounts therefore to [cf. Eqs. (13) and (14)]

$$\rho_N(t) \approx -i \left[\tilde{Q}_{\text{trun}}(t)\rho_{N-1}(t) - \rho_{N-1}(t)\tilde{Q}_{\text{trun}}(t)\right]. \tag{23}$$

The above truncation leads to a time-local evolution for the anchoring $\rho_N$, and the resulting EOM is in a partial ordering prescription (POP).

In comparison, the corresponding chronological ordering prescription (COP) of $(2N)^{\text{th}}$-order truncation can be realized via setting $\rho_{N+1} = 0$, as it effectively leads to a time-ordered, memory description of $\rho_N(t)$ in terms of $\rho_{n<N}(\tau \leq t)$. It is easy to verify that the conventional second-order memory-kernel and time-local formulations, just the special cases of the present COP-truncation ($\rho_2 = 0$) and POP-truncation [Eq. (23) with $N = 1$], respectively.

Consider now the truncation scheme proposed by Tanmura and coworkers in which the fast modulation or Markovian anzatz is assumed applicable at the anchor level. In this case, $\tilde{Q}_{\text{trun}}(t)$ of Eq. (22) reduces to $\tilde{Q}_{\text{trun}}^\text{COP} = \tilde{C}Q$, where $\tilde{C} = \int_{0}^{\infty} dt C(\tau)$. This scheme is essentially identical to that proposed by Tanmura and co-workers, but is obtained here for general $C(t)$ without invoking the Gaussian-Markovian bath model. Unlike the $N$-level POP [Eq. (23) with Eq. (22)] or COP ($\rho_{N+1} = 0$) truncation, the $N$-level fast-modulation truncation scheme is not a rigorous $(2N)^{\text{th}}$ order, but rather $(2N - 2)^{\text{th}}$-order formulation. We shall come back to this point later in terms of the practical efficiencies of the aforementioned three truncation schemes; see discussions following Fig. 1. Obviously, all these schemes become exact when $N \to \infty$.

To investigate the efficiencies of involving truncation schemes for the study of non-Markovian dissipation with arbitrary system-bath interaction, we consider here a simple spin-boson system: $H = \frac{1}{2} \Omega \sigma_z$, with $Q = \sigma_z$ and $C(t) = \Delta^2 e^{-\gamma t}$. In the following, we adopt the integrated coupling strength $\Gamma \equiv \int_{0}^{\infty} dt C(t) = \Delta^2/\gamma$ and the dimensionless modulation parameter $\kappa \equiv \gamma/\Delta$ to characterize the nature of system-bath coupling, and

$$s_N \equiv \frac{1}{\Gamma} \max_{\{t\}} \left\{ \int_{t}^{\infty} dt A_N(t) - A_N(\infty) \right\}^2,$$

with $T = 2\pi/\Omega$, to calibrate the finite $N$-truncation induced error in evaluation of a given test quantity $A(t)$, which will be chosen to be $\rho_{22}(t) - \rho_{11}(t)$ for demonstration. The initial condition is set to be $\rho_{kk}(0) = \delta_{kk} x; k = 1, 2, 3$.

Depicted in Fig. 1 are the resulting error indicators $s_N$ for the aforementioned three truncation schemes, demonstrated as functions of anchoring index $N$ at various values of system-bath coupling strength and modulation parameter: (a) $\Gamma = \Omega$, $\kappa = 1$; (b) $\Gamma = \Omega$, $\kappa = 0.1$; (c) $\Gamma = 10\Omega$, $\kappa = 1$; and (d) $\Gamma = 10\Omega$, $\kappa = 0.1$. This figure clearly demonstrates the following features: (i) The anchor index $N$ depends not only on the system-bath coupling strength $\Gamma$, but more importantly on the modulation parameter $\kappa$; (ii) In the slow modulation ($\kappa \ll 1$) limit, the low order truncation may not be sufficient even in the weak coupling ($\Gamma \ll \Omega$) regime; (iii) As inferred from their constructions, these three truncation schemes are in principle of the same quality in the fast modulation limit; see Fig. 1(c). Considering further the computation efforts involved, while the anchor $\rho_N$ in the POP or fast-modulation truncation is expressed directly by Eq. (23) or its Markovian counterpart, respectively, the $\rho_N^\text{COP}$ shall however be propagated via coupled EOM. Thus, the POP truncation scheme proposed in the work is overall the best, including the case studied in Fig. 1(b).

Shown in Fig. 2 are the approximate results of the test quantity $\rho_{22}(t) - \rho_{11}(t)$, obtained via the POP (solid), the COP (dash), and the fast-modulation (dot) truncation schemes at the specified value of $N$ in each panel such that the error tolerance of $10^{-3.5}$ is met by the minimum $s_N$ among these three truncation schemes. Included in each panel of this figure is also the exact result (thick-solid) that can be obtained via any truncation scheme with a sufficiently large $N$. Note that Fig. 2(b) is depicted for $26 < t < 27$ (in the unit of $2\pi/\Omega$) where the difference between various curves is relatively large. The
overall superiority of POP-truncation scheme is again highlighted. We have also showed (iv) The behavior of $\rho(t)$ in short time, long time, and Markovian regimes may be accounted for properly by the low order $\rho_n$: (v) The relatively high order $\rho_n$ are required for the intermediate time, and the slow modulation regimes; (vi) Note that $\kappa \gg \Gamma$ in the slow modulation ($\kappa \ll 1$) limit [cf. Fig. 2(b)].

To summarize, we have derived an exact quantum master equation formalism via a simple calculus on the Feynman-Vernon influence functional path integrals. It is valid for arbitrary external field driven reduced dynamics under non-Markovian system-bath interaction beyond the weak coupling regime. The present derivation constitutes an alternative approach to the exact quantum master equation, Eq. (13), which is identical to that obtained recently by Shao via rather advanced stochastic differential equation algebra. It is also noticed that Shi and Geva had also recently constructed a formally exact quantum dissipation theory. It however depends practically on the path-integral evaluation of the Nakajima-Zwanzig dissipation kernel, despite the fact that is expressed in terms of force-force correlation function to be evaluated with correlated system-bath ensemble.

We have also proposed a novel truncation scheme; i.e., the POP-scheme of Eq. (24), which is shown to be overall superior to the two existing truncation approaches. This superiority is of much more implication than that demonstrated in the numerical examples of this work, as in general $C(t)$ contains multi-exponential terms, especially at low temperature regime, and the relevant number of $\rho_n$ with $n \leq N$ [cf. Eq. (21)] in the hierarchical EOM increases quasi-exponentially with the truncation anchor $N$. The numerical demonstrations are also of rich physical implications on the interplay between system-bath coupling strength, non-Markovian property, and the required order of truncation, which had been summarized in details in Comments (i)–(vi) following Fig. 1 and Fig. 2. Especially, low-order quantum dissipation theories should be used with care if non-Markovian dynamics is important.

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FIG. 1: The error parameters $s_N$ versus the anchor $N$, in the evaluations of $\rho_{22}(t) - \rho_{11}(t)$ via the POP, COP, and fast-modulation truncation schemes. The selected system-bath coupling strength and modulation parameters are: (a) $\Gamma = \Omega$, $\kappa = 1$; (b) $\Gamma = \Omega$, $\kappa = 0.1$; (c) $\Gamma = 10\Omega$, $\kappa = 1$; and (d) $\Gamma = 10\Omega$, $\kappa = 0.1$.

FIG. 2: The evolutions of $\rho_{22}(t) - \rho_{11}(t)$ implied in Fig. 1 with the specified anchor $N$ in each panel being chosen such that the tolerance of $10^{-3.6}$ is satisfied by the minimum $s_N$ among the three truncation schemes. The exact evolutions can be obtained with any schemes truncated at sufficiently high level. The curves for the exact and the POP almost overlap with each other in each of the four panels.

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Fig. 1

(a) $\Gamma = \Omega, \kappa = 1$

(b) $\Gamma = \Omega, \kappa = 0.1$

(c) $\Gamma = 10\Omega, \kappa = 1$

(d) $\Gamma = 10\Omega, \kappa = 0.1$

Log $N_s$ vs. Log $N$

- POP
- COP
- Fast modulation

Note: The diagram shows a comparison of different coupling strengths and their effects on the system parameters.
Fig. 2

(a) $N=4$
- Exact
- POP
- COP
- Fast modulation

(b) $N=9$
- $\Gamma=\Omega, \kappa=0.1$

(c) $N=5$
- $\Gamma=10\Omega, \kappa=1$

(d) $N=11$
- $\Gamma=10\Omega, \kappa=0.1$

Time (unit: $2\pi/\Omega$)