Exact quantum dissipative dynamics under external time-dependent driving fields

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Abstract. An exact and nonperturbative quantum master equation can be constructed via the calculus on the path integral. It results in hierarchical equations of motion for the reduced density operator. Involved also are a set of well-defined auxiliary density operators that resolve not just the system–bath coupling strength but also memory. In this work, we scale these auxiliary operators individually to achieve a uniform error tolerance, as set by the reduced density operator. An efficient propagator is then proposed to the hierarchical Liouville-space dynamics of quantum dissipation. Numerically exact studies are carried out on the dephasing effect on population transfer in the simple stimulated Raman adiabatic passage scheme. We also make assessments on several perturbative theories for their applicabilities in the present system of study.
1. Introduction

The central problem of quantum dissipation theory is to study the dynamics of a quantum system embedded in a quantum thermal bath. The primary quantity of interest here is the reduced density operator, \( \rho(t) \equiv \text{tr}_B \rho_T(t) \), after the bath degrees of freedom are all traced from the total composite density operator. Due to its fundamental importance, quantum dissipation theory has remained an active topic in diversified fields [1]–[10]. The challenge here, from both formulation and numerical aspects, is nonperturbative dissipation, with multiple timescales of memory, under a time-dependent external driving field.

For Gaussian stochastic force, the influence of the bath on the system can be characterized by force–force correlation functions. Exact formalism had then been established via the Feynman–Vernon influence functional approach [1]–[5]. Direct numerical integration methods based on the discretization of the path integral and the summation of the memory correlated terms have been put forward, such as the quasi-adiabatic propagator method [11]–[14] or the real-time quantum Monte Carlo scheme [15]–[19].

The alternative is the differential approach, especially in a linear form to maximize the numerical advantage. It also has an advantage in the study of various dynamics such as the spectroscopic or control problems [20]. The calculus-on-path-integral (COPI) method is hence proposed to construct the differential counterpart of the path integral theory, reported as the hierarchical equations of motion (HEOM) formalism [21]–[30]. This formalism can also be derived via the stochastic description of quantum dissipation [31]–[35]. The COPI algorithm provides a unified approach to the influence of quantum environment ensembles, either canonical or grand canonical, and either bosonic or fermionic [28, 29]. The COPI algorithm also takes into account the combined effects of multiple memory timescales, system–bath coupling strengths and system anharmonicity. The resulting HEOM formalism is therefore nonperturbative in nature, and always converges in principle. Moreover, the HEOM formalism is exact, not just for its propagation equivalent to the path integral theory, but also for the fact that...
the initial correlations between the system and bath can now be incorporated by the steady-state solutions to the HEOM, before external time-dependent fields take effect. Recently, we have further developed a numerical efficient filtering method for the propagation of the HEOM [36, 37].

In this work, we report a HEOM-based study on population transfer with dephasing in the scheme of the stimulated Raman adiabatic passage (STIRAP) [38]. The laser control of dissipative systems has been addressed extensively [39]–[49], but mostly on the basis of weak dissipation treatment. The correlated influence of driving and dissipation is often important, as demonstrated previously [50, 51]. With the aid of numerically exact results, we analyze the dephasing effects on transfer dynamics in relation to the STIRAP mechanism and examine some second-order quantum dissipation theories for their applicabilities in the systems of study.

The remainder of this paper is organized as follows. We present the HEOM formalism together with comments on its numerical implementation in section 2, and the derivations in the appendix. In section 3, we study the dephasing effect on population transfer dynamics in the STIRAP scheme. We report the numerically exact results via the HEOM formalism, followed by discussions in relation to the STIRAP mechanism. In section 4, we present details of the numerical performance of the HEOM results, and make concrete assessments on several approximated quantum dissipation theories. Finally we conclude the paper.

2. HEOM formalism for quantum dissipation

2.1. Description of stochastic bath coupling

The total system-plus-bath Hamiltonian can be written in general as

\[ H_T = H(t) + h_b - \sum_a Q_a \hat{F}_a. \]  

The last term denotes the multi-mode system–bath interactions. The involving system operators \( \{Q_a\} \) are called the dissipative modes, through which the generalized Langevin forces \( \{\hat{F}_a(t) = e^{i h_b t} \hat{F}_a e^{-i h_b t}\} \) from the bath \( (h_b) \) act on the system. For convenience, let the dissipative modes be dimensionless. The time dependence in the system \( H(t) \) arises from external driving fields. Throughout this paper, we denote the inverse temperature \( \beta \equiv 1/(k_B T) \) and set \( \hbar \equiv 1 \).

We treat the Langevin forces as Gaussian stochastic processes. Therefore, their effects on the system are completely characterized by the correlation functions,

\[ C_{ab}(t - \tau) = \langle \hat{F}_a(t) \hat{F}_b(\tau) \rangle_B. \] 

Here, \( \langle \hat{O} \rangle_B = \text{tr}_B(\hat{O} \rho_{eq}^B) \) denotes the thermodynamics average over the canonical ensembles of the bosonic bath. The correlation functions satisfy the symmetry and detailed-balance relations, or equivalently the fluctuation–dissipation theorem [3, 20]:

\[ C_{ab}(t) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \frac{e^{-i\omega t} J_{ab}(\omega)}{1 - e^{-\beta \omega}}, \] 

with \( J_{ab}(\omega) = -J_{ba}(-\omega) = J_{ba}^*(\omega) \) being the bath spectral density functions. The HEOM formalism requires \( C_{ab}(t) \) to be expanded in a certain series form, so that the hierarchy can
be constructed via consecutive time derivatives on the path integral. Various schemes [22, 28, 52, 53] have been proposed to expand $C_{ab}(t)$ in exponential series on the basis of analytical continuation evaluation of equation (3). In particular, the hybrid scheme that also exploits the quadrature integration method is applicable for arbitrary spectral density functions [30].

For simplicity, we set $C_{ab}(t) = C_{aa}(t)\delta_{ab}$. In this case, the contributions from different dissipative modes $\{Q_a\}$ are additive. Without loss of generality, we present the formalism explicitly only for the single-dissipative-mode case, $Q_a = Q$. We thus omit the index $a$ for clarity of formulation. We also adopt the super-Drude model,

$$J(\omega) = \frac{\eta \omega}{(\omega/\gamma)^2 + 1}.$$  

The corresponding correlation function can be analytically evaluated as [20, 28, 53]

$$C(t \geq 0) = [v + (\tilde{\nu} + i\tilde{\nu}_t) \gamma t]e^{-\gamma t} + \sum_{m=1}^{M} \tilde{v}_m e^{-\gamma \omega t} + \delta C(t).$$  

All coefficients here are real and given in the appendix (cf equations (A.9) and (A.10)). The first term arises from the pole of the spectral density function, which is of rank two. The second term is from the Matsubara poles, with $\gamma_m \approx 2\pi m/\beta$ being the Matsubara frequency. The last term is the Matsubara residue, which would approach zero if $M \to \infty$. In this work, we adopt the Markovian residue ansatz [25, 35], i.e. $\gamma_m e^{-\gamma \omega t}|_{m>M} \approx \delta(t)$; thus,

$$\delta C(t) \simeq \Delta(\delta(t)); \quad \Delta = \sum_{m=M+1}^{\infty} \tilde{v}_m = \frac{\eta}{\beta} - \frac{v + \tilde{\nu}}{\gamma} - \sum_{m=1}^{M} \tilde{v}_m.$$  

### 2.2. The HEOM formalism

The dynamics quantities in the HEOM formalism are the reduced density operator $\rho(t)$ and a set of auxiliary density operators (ADOs), $\{\rho_n(t)\}$, that hierarchically resolve the memory contents of the bath correlation functions in the exponential series expansion of equation (5). The index $n$ that specifies an $N$th-tier ADO $\rho_n$ consists of a series of nonnegative integers:

$$n \equiv \{n, n', \bar{n}, \bar{n'}, \bar{n}_1, \ldots, \bar{n}_M\}, \quad \text{with} \quad n + n' + \bar{n} + \bar{n'} + \bar{n}_1 + \cdots + \bar{n}_M = N.$$  

Compared to the reduced density operator $\rho(t) \equiv \rho_0(t)$ of primary interest, the specified $\rho_n$ would have the order of $|v|^{n+n'}|\tilde{\nu}_t + i\tilde{\nu}|^{\bar{n}+\bar{n'}} \prod_{m=1}^{M} |\tilde{v}_m|^{\bar{n}_m}$, due to its dependence on the individual components of interaction bath correlation functions in the series expansion of equation (5). These scaling factors will be incorporated properly in the final dimensionless $\rho_n$, in order to validate a filtering algorithm for the numerical efficiency of the HEOM formalism. On the other hand, the indices in the set $n$ of equation (7) cover all accessible derivatives of the Feynman–Vernon influence functional; see the appendix for more details.

The final HEOM formalism is summarized as follows. It has the generic form of

$$\dot{\rho}_n = -[i\mathcal{L}(t) + \Gamma_n + \delta \mathcal{R}]\rho_n + \rho_{n}^{(-a)} + \rho_{n}^{(-)} + \rho_{n}^{(a)}.$$  

Here, $\mathcal{L}(t)\rho_n \equiv [H(t), \rho_n]$, which depends in general on the external driving fields; $\Gamma_n$ is the damping parameter that collects all related exponents, and $\delta \mathcal{R}$ is the residue dissipation
superoperator due to $\delta C(t)$. For the bath correlation function in the series expansion, equation (5) with equation (6), they are given, respectively, by

$$\Gamma_n = (n + n' + \tilde{n} + \tilde{n}') \gamma + \sum_{m=1}^{M} \tilde{n}_m \tilde{y}_m, \quad \delta \mathcal{R} \rho_n = \Delta [Q, [Q, \rho_n]]. \quad (9)$$

Apparently, $\Gamma_0 \equiv \Gamma_n|_{n=0} = 0$.

The last three terms in equation (8) denote how the specified $N$th-tier ADO $\rho_n$ depends on other ADOs of the same tier, the $(N - 1)$th-tier and the $(N + 1)$th-tier, respectively. For the bath correlation function in equation (5), they are given explicitly by

$$\rho_n^{i,j} = -i [Q, \lambda_n \rho_n] + [Q, \lambda_{n'} \rho_{n'}] - i \left[ Q, \sum_{m=1}^{M} \lambda_{n_m} \rho_{n_m} \right], \quad (10)$$

Here, $\lambda_n = \sqrt{n} |v|$, $\lambda_{n_1} = \sqrt{n_1} |\tilde{v}_m|$, $\tilde{\lambda}_n^{r/i} = \sqrt{n} |\tilde{v}_r/^i|/|v|$, and $\tilde{\lambda}_n^{r/i} = {\gamma} \tilde{\lambda}_n^{r/i} \bar{\rho}_n^n$, with the italic-font indices being from those in $n$ of equation (7). The indices' variations in equation (10) that specify the ADOs participating in the equation of $\rho_n$ are exemplified as follows:

$$\tilde{n} \equiv \{n + 1, n', \tilde{n} - 1, \tilde{n}', \tilde{n}_1, \ldots, \tilde{n}_M \}, \quad n^\pm \equiv \{n \pm 1, n', \tilde{n}, \tilde{n}', \tilde{n}_1, \ldots, \tilde{n}_M \}. \quad (11)$$

Similarly, $\tilde{n}'$ differs from $n$ of equation (7) only by changing $(n', \tilde{n}')$ to $(n' + 1, \tilde{n} - 1)$, while $\tilde{n}^\pm$ differs from $n$ by changing $\tilde{n}_m$ to $\tilde{n}_m \pm 1$, and so on. Also note that $\rho_{\tilde{n}}$ is an $N$th-tier ADO, while $\rho_{n^\pm}$ is of an $(N + 1)$th tier, as inferred from the second identity of equation (7).

The initial conditions to the HEOM in the study of driven dissipative dynamics are obtained via the steady-state solutions to equation (8), before the time-dependent external fields' interactions. For the steady-state solutions satisfying $\bar{\rho}_n^\mp = 0$, equation (8) reduces to a set of linear equations, under the constraint of $\text{Tr} \rho_0 = 1$. The resulting $\rho_n^\mp$ is used as the initial $\rho_n(t_0)$ to the HEOM. The initial system–bath correlations are accounted for by those nonzero initial ADOs.

2.3. Comments on numerical implementation

For the numerical HEOM propagation, we would like to have a certain convenient working index scheme to track the multiple indices, denoted now as an ordered set of $n = \{n_1, \ldots, n_K\}$ that specifies $\rho_n$. Here, we will provide two such schemes. The number of the $N$th-tier ADOs, with $n_1 + \cdots + n_K = N$, is $\binom{N + K - 1}{K - 1}$. In one scheme, the ADOs are arranged as $\rho_n \equiv \rho_{j_0}$ with $j_0$ initialized by $j_{n=0} = 0$ and then

$$j_{n \neq 0} = j_{n_1, \ldots, n_K} = \sum_{N'=0}^{N-1} \binom{N'}{K} + \sum_{k=1}^{K} \sum_{q=0}^{N-s_k} \binom{q}{K-k}; \quad s_k = 1 + n_1 + \cdots + n_k. \quad (12)$$

Let $L$ be the maximum level of the hierarchical tier. The total number of the ADOs $\{\rho_n; 0 \leq N \leq L\}$ is

$$N' = \sum_{N=0}^{L} \binom{N}{K} = \binom{L}{K}. \quad (13)$$

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In another scheme, ADOs can also be arranged as \( \rho_n \equiv \rho_{l_n}; l_n = 0, \ldots, N - 1 \), with

\[
I_n = l_{n_1, \ldots, n_K} = n_1 + \sum_{k=2}^{K} \sum_{q=1}^{n_k} \left\{ L + q - (n_k + \cdots + n_K) \right\}.
\]

Both schemes, equations (12) and (14), allow an easy tracking of the coupled ADOs in the HEOM. The former (equation (12)) is somewhat more convenient in the filtering propagator described later since it does not depend on \( L \).

The major difficulty in implementing the HEOM formalism is its numerical tractability. The number of ADOs, \( N \) of equation (13), itself alone can be huge in the case of strong non-Markovian system–bath coupling and/or low temperature, as large \( L \) and/or large \( K \) implied. Thus, a brute-force implementation is greatly limited by the memory and central processing unit (CPU) capability of computer facility, even for a two-level system where each ADO is a \( 2 \times 2 \) matrix.

To facilitate this problem, Shi et al have recently proposed an efficient numerical filtering algorithm that often reduces the effective number of ADOs by order of magnitude \([36, 37]\). In reality, there is usually only a very small fraction of total ADOs significant to the reduced system dynamics. To validate the accuracy-controlled numerical filtering algorithm, the present HEOM formalism has been scaled properly so that all ADOs \( \{\rho_n(t)\} \) are of a uniform error tolerance. This remarkable feature is suggested by comparing the HEOM theory with the stochastic bath interaction field approach in the case of Gaussian–Markovian dissipation \([37]\). The involving ADOs are just the expansion coefficients, over the normalized harmonic wave functions that are used as the basis set for resolving the diffusive bath field \([37]\). Our numerical HEOM propagator exploits the filtering algorithm \([36]\). It goes simply as follows. If a \( \rho_n(t) \) whose matrix element amplitudes all become smaller than the pre-chosen error tolerance, it is set to be zero. Apparently, the filtering algorithm also automatically truncates the required hierarchy level on-the-fly during numerical propagation. By far, the truncation for the Matsubara expansion still goes by checking convergency.

3. Effect of dephasing on population transfer via STIRAP

3.1. Numerical results

The STIRAP is celebrated as an efficient and robust method for population transfer \([38]\). It is characterized by its counterintuitive field configuration. For a three-level \( \Lambda \)-system as shown in figure 1, the Stokes pulse proceeds the pump pulse and the intermediate state remains effective in dark. The STIRAP mechanism \([38]\) is rooted at the existence of the coherent population trapping state under the two-photon resonance (TPR) condition, \( \omega_S - \omega_P = \epsilon_1 - \epsilon_3 \), in the \( \Lambda \)-system. Dephasing destroys this condition in terms of resonance and/or the existence of a coherent population trapping state. The effect of dephasing on the simple STIRAP scheme has been studied extensively, but with approximations. These include phenomenological/perturbative methods \([44]–[46]\), or classical/stochastical bath treatments \([47]–[49]\).

We revisit the dephasing effect on the simple STIRAP-based population transfer, as the exact dissipative dynamics are now established with the present HEOM formalism. We also examine three schemes of the second-order approximation \([20], [52]–[55]\): (i) the Redfield theory, which neglects the correlated driving-and-dissipation effect; (ii) the complete second-order chronological ordering prescription (CS–COP), which is the conventional time-nonlocal quantum master equation, including the field-dressed dissipation contribution, and equivalent to
the present HEOM truncated at the first tier, and (iii) the correlated driving–dissipation equations (CODDE), in which the driving field-free part of dissipation superoperator is time-local, while the field-dressed part is time-nonlocal. Neglecting the latter leads it to the Redfield theory. The total Hamiltonian under the rotating wave approximation assumes

$$H_T(t) = \Omega_P(t) \hat{D}_P + \Omega_S(t) \hat{D}_S + \sum_j \left[ \frac{p_j^2}{2m_j} + \frac{1}{2} m_j \omega_j^2 \left( x_j - \sum_a c_{aj} Q_a \right)^2 \right]. \quad (15)$$

Here, \( \hat{D}_P \equiv |1\rangle\langle 2| + |2\rangle\langle 1| \) and \( \hat{D}_S \equiv |2\rangle\langle 3| + |3\rangle\langle 2| \), while \( \Omega_P(t) \) and \( \Omega_S(t) \) denote the Rabi frequencies of the resonant pump and Stokes fields, respectively. The dissipative mode \( Q_a = |a\rangle\langle a| \) is responsible for dephasing. The interaction spectral density function \( J_a(\omega) = (\pi/2) \sum_j \left[ c_{aj}^2/(m_j \omega_j) \right] \delta(\omega - \omega_j) \) assumes super-Drude as equation (4). The system Hamiltonian is then

$$H(t) = \Omega_P(t) \hat{D}_P + \Omega_S(t) \hat{D}_S + \sum_{a=1}^3 \delta \epsilon_a |a\rangle\langle a|. \quad (16)$$

The Caldeira–Leggett renormalization energy \([56, 57]\) is \( \delta \epsilon_a = \frac{1}{2} \int_0^\infty d\omega J_a(\omega) / \omega = \frac{1}{4} \eta_a \gamma_a \) for the super-Drude model (equation (4)). In the STIRAP configuration, it would relate to the effective detuning at short-time of the pump or Stokes field, as inferred from the analytical result of a driven Brownian oscillator \([20, 58]\). We set the pump and Stokes fields to be of the same Gaussian shape, \( \Omega_P(t + t_p) = \Omega_S(t + t_s) = A \exp[-t^2/(w^2)] \), but center them at \( t_p = 200 \beta \) and \( t_s = -200 \beta \), respectively, and counter-intuitively. The driving strength and inverse duration parameters are set to be \( \beta A = 0.1 \) and \( \beta w = 0.005 \). The corresponding dissipation-free transfer dynamics is shown in figure 1(b). As here the bath influence is considered to be pure-dephasing in the absence of fields, the initial system is just chosen to be completely on the \( |1\rangle \) state and all the ADOs are zero. For the effect of the bath, we set the coupling strength \( \eta = 0.64 \) (cf equation (4)), and consider both the Markovian and non-Markovian cases as follows.

The Markovian transfer dynamics, under the influence of the single dephasing mode of either \( Q_1 = |1\rangle\langle 1| \) or \( Q_2 = |2\rangle\langle 2| \), is exemplified in figure 2, with \( \beta \gamma = 5 \). We observe that

Figure 1. (a) A schematic view of the STIRAP of a three-level \( \Lambda \) system. (b) Population transfer under the STIRAP scheme for the dissipation-free gas phase. Time is in the unit \( \beta \). See parameters in the text.
Figure 2. Evolutions of $\rho_{33}$ and $\rho_{22}$ (in insets) via the exact HEOM (solid), CODDE (dash), CS–COP (dot) and the Redfield equation (thin-solid) for single-dissipative-mode case: (a) $Q_1 = |1\rangle\langle 1|$ and (b) $Q_2 = |2\rangle\langle 2|$. The system–bath coupling strength $\eta = 0.64$. The parameter $\beta\gamma = 5$ exemplifies the Markovian condition. Time is in the unit $\beta$.

Figure 3. Same as figure 2 except that the parameter $\beta\gamma = 0.5$ exemplifies the non-Markovian condition. Time is in the unit $\beta$.

(i) the $Q_1$-mode effect shown in figure 2(a) leads to all three populations about $1/3$ after the driving; (ii) the $Q_2$-mode effect shown in figure 2(b) is less sensitive than its $Q_1$ counterpart, achieving a higher transfer efficiency, despite it being only about $0.55$.

The non-Markovian transfer dynamics is exemplified in figure 3, with $\beta\gamma = 0.5$. In comparison with the Markovian counterparts, we observe that (iii) the $Q_1$-mode case behaves about the same, (iv) but the $Q_2$-mode results in a higher transfer yield, increasing to about $0.73$ via the exact calculation. We have also calculated the influences of $Q_3 = |3\rangle\langle 3|$ for both Markovian and non-Markovian cases. The results (not shown here) are similar to those of $Q_1$, except for some small oscillations.

Two double-mode ($Q_1 + Q_2$ and $Q_1 + Q_3$ uncorrelated) non-Markovian dephasing dynamics are shown in figures 4(a) and (b), respectively. They are insensitive to the non-Markovian parameter, and both reach final equal-populations, based on the numerically exact
results. Comments on the approximated schemes, the CODDE, CS–COP and Redfield theory, presented in figures 2–4, will be given later; see section 4.2.

3.2. Discussions

The above observations can be understood by the well-established STIRAP mechanism [38]. The $Q_1$-mode, which associates with the fluctuation of level $|1\rangle$, easily destroys the TPR condition, as described at the beginning of section 3. Thus, it ends up with observed equal populations in all accessible levels by the strong fields, as consistent with the analysis in [44]. The similarity between $Q_3$ and $Q_1$ influences is also explained. The same reason further accounts for the case of uncorrelated two-mode ($Q_1 + Q_2$ or $Q_1 + Q_3$) dephasing, as depicted in figure 4. It is anticipated that when $\gamma \ll w$ (termed as the linear adiabatic limit below), the equal-population will be broken to be in favor of $|3\rangle$, due to the marginally partial fulfillment of the TPR condition.

On the other hand, the $Q_2$-mode is associated with the fluctuation of the intermediate level $|2\rangle$. It alone does not affect the TPR condition. However, this condition, based on the numerically exact results shown in this work, is not sufficient to retain the coherent population trapping state, chosen ad hoc earlier for the dephasing-free STIRAP scenario in figure 1(b). It is anticipated that the coherent population trapping state may be recovered in the aforementioned linear adiabatic limit. This is in line with observation-(iv), where the non-Markovian population transfer with single $Q_2$-mode dephasing (figure 3(b)) is of higher efficiency than its Markovian counterpart (figure 2(b)). The previous study based on perturbative dephasing dynamics [44] has also shown that the single $Q_2$-mode does not affect the transfer efficiency in the linear adiabatic limit. Nevertheless, STIRAP in the presence of complex dephasing, if 100% transfer is ever achievable, would require dynamics feedback control of pump or Stokes laser frequency [59]. This would involve chirp and realize STIRAP in a nonlinear adiabatic condition, rather than the linear simplification considered here.
4. Assessments on theoretical methods and concluding remarks

4.1. Numerical performance of the HEOM formalism

The numerical performance of the HEOM formalism with filtering is summarized in table 1, for the systems reported in the three figures’ (b)-panels. The CPU time is for a single Intel(R) Xeon(R) processor@3.00 GHz to calculate the exact result in each (b)-panel for the time period $-1200\beta < t < 2000\beta$, with the time step $dt = 0.01\beta$ using the fourth-order Runge–Kutta propagator. $N_{\text{max}}$ denotes the largest number of active ADOs and $L_{\text{max}}$ the highest tier level, that has ever survived in the entire time span of the numerical propagation. The filtering error tolerance is chosen to be $10^{-6}$, following our previous work [36]. We input $M = 6$ for the number of Matsubara terms being explicitly included, which has been tested to give converged results of $\rho(t) = \rho_0(t)$ in all calculations. The total number $N$ of mathematical ADOs follows equation (13) and is given inside parentheses. The effect of filtering is clearly seen. The number of active ADOs with filtering is insensitive to the input $M$, as long as it is large enough. In the present study, the number of active ADOs reaches $N_{\text{max}}$ only during the period of about $-250\beta < t < 500\beta$ and grows up or drops down dramatically outside that period with the fields turning on or getting over. Apparently, $N_{\text{max}}$ increases with the number of dissipative modes.

At least one ($L_{\text{max}}$)th-tier ADO actively participates during the HEOM propagation. Its leading contribution to the reduced density operator is of $(2L_{\text{max}})$th order in the system–bath interaction. Physically, $L_{\text{max}}$ is closely related to the modulation $\kappa$-parameter [27], introduced originally by Kubo for motional narrowing problem [6]. This dimensionless parameter is determined via $\kappa = \gamma / \sqrt{\nu}$, or similarly, for each individual exponential component in equation (5). The last two columns of table 1 are the modulation parameters $\kappa$ and $\tilde{\kappa}_m = 1$ of the leading Matsubara term. The modulation $\kappa$-parameter relation to the value of $L_{\text{max}}$ [27] can be clearly seen. In both the Markovian and non-Markovian cases of the present study, $\tilde{\kappa}_m = \tilde{\gamma}_m / \sqrt{\tilde{\nu}_m}$ monotonically increases with $m$, cf equation (A.10). Actually, the Matsubara series truncation $M$ in equation (5) can be estimated via its reaching the fast modulation condition, $\tilde{\kappa}_M \gg 1$. As the temperature decreases, $\tilde{\kappa}_m$ becomes smaller, which eventually causes the value of $L_{\text{max}}$ to be pretty large. The present HEOM construction is based on the Matsubara series expansion, which may no longer be numerically implementable in the extremely low temperature regime. Alternative expansion method such as the hybrid scheme [30] is needed for the required HEOM construction.

4.2. Assessments on three second-order approximated theories

With the exact results, we can now make concrete assessments on the three second-order approximated schemes: the Redfield theory, CS-COP, and CODDE, exploited in the numerical demonstrations. The dissipative modes $\{Q_a = |a\rangle\langle a|\}$ considered in section 3 are all of pure

Table 1. Performance of the HEOM formalism with filtering.

| Filter | CPU (min) | $N_{\text{max}}$ | $L_{\text{max}}$ | $\kappa$ | $\tilde{\kappa}_1$ |
|--------|----------|-----------------|-----------------|--------|----------------|
| Figure 2(b) | 932 | 19765 (8.44 x 10^6) | 17 | 0.95 | 1.28 |
| Figure 3(b) | 15 | 400 (9.24 x 10^4) | 9 | 1.24 | 348 |
| Figure 4(b) | 266 | 3664 (1.00 x 10^7) | 9 | 1.24 | 348 |

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dephasing, in the absence of external fields. The Redfield theory would be exact if there were no correlated driving-and-dissipation effect [20, 54]. Therefore, the non-Markovian dynamics manifest here as correlated driving and dissipation. Apparently, the Redfield theory, by its Markovian nature, is independent of the width $\gamma$-parameter of bath spectral density. Observed is also the fact that the schemes of approximation are sensitive to the $Q_2$-mode rather than the $Q_1$- or $Q_3$-mode dephasing. This fact is also easily understood by considering their relations to the STIRAP mechanism as discussed earlier. Further remarks on the approximated theories for their applicabilities in the systems of study are as follows.

The CS–COP theory [20, 53, 55] is overall most unsatisfactory, despite it containing formally a description of memory and driving-and-dissipation correlation. Even in the non-Markovian $Q_2$-mode case of figure 3(b), where it appears to be superior than the Redfield theory, the CS–COP results in a decreased transfer efficiency from its Markovian counterpart (cf figure 2(b)). This is qualitatively contradictory to the physical anticipation, as discussed earlier.

The CODDE [20, 53, 55] appears to be the most favorable perturbation theory. It gives the best approximated transfer dynamics in all cases presented in section 3, except the one to be discussed soon. Its overall superiority is also true in the driven Brownian oscillator systems [20, 51]. The CODDE is actually a modified Redfield theory, with the inclusion of correlated driving-and-dissipation effects. The involving field-dressed dissipation kernel is time-nonlocal but constructed with a partial ordering resummation, rather than the chronological ordering prescription that characterizes the CS–COP [20, 53, 55].

The only exception is the Markovian $Q_1$-mode case shown in figure 2(a), where the Redfield dynamics is almost exact. The reason for this exception is also accountable. As we mentioned earlier, the non-Markovian dynamics manifest as the correlated driving and dissipation. This correlated effect diminishes in both fast- and slow-modulation regimes, as inferred from the exact and analytical results of driven Brownian oscillator systems [58]. This conclusion can be carried over to the present system of study, as suggested here. Apparently, the identical value of $\beta \gamma = 5$, adopted in the two cases of figure 2, acquires the fast-modulation limit for the $Q_1$-mode, but not yet for the $Q_2$-mode. In the latter case, the CODDE resumes its superiority.

4.3. Closing remarks

In summary, we have presented a hierarchical Liouville-space approach, which is exact and also quite tractable numerically, to general quantum dissipation systems under external driving fields. The ADOs are all of a uniform error tolerance, as that of the reduced density operator. We comment on the numerical facilitation of the multiple-index assignment and the filtering algorithm. We numerically study the dephasing effects on the population transfer, with a fixed simple STIRAP configuration, and present a concrete assessment on various approximation schemes.

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Appendix. Construction of HEOM via the COPI approach

This appendix gives details of the COPI approach to the HEOM formalism. It starts with the influence functional in path integral. Let \(|\alpha\rangle\) be a basis set in the system subspace and set \(\alpha \equiv (\alpha, \alpha')\). Denote the evolution of the reduced system density operator in the \(\alpha\)-representation by

\[
\rho(\alpha, t) \equiv \rho(\alpha, \alpha', t) \equiv \int d\alpha_0 \mathcal{U}(\alpha, t; \alpha_0, t_0) \rho(\alpha_0, t_0).
\]

Here, the reduced Liouville-space propagator is

\[
\mathcal{U}(\alpha, t; \alpha_0, t_0) = \int_{\alpha_0[\alpha_0]} \mathcal{D}\alpha e^{iS[\alpha]} \mathcal{F}[\alpha] e^{-iS[\alpha']}.\]

The effects of the bath on the reduced system are contained completely in the influence functional \(\mathcal{F}\). For Gaussian stochastic forces \(\hat{F}_a(t)\) from a fluctuating bath, it assumes the Feynman–Vernon form \([1]\), which can be recast as \([27]–[29]\)

\[
\mathcal{F}[\alpha] = \exp \left\{ - \int_{t_0}^{t} d\tau \sum_a A_a[\alpha(\tau)] B_a(\tau; \{\alpha\}) \right\},
\]

with

\[
A_a[\alpha(t)] = Q_a[\alpha(t)] - Q_a[\alpha'(t)],
\]

\[
B_a(t; \{\alpha\}) = B_a(t; \{\alpha\}) - B'_a(t; \{\alpha\}),
\]

and

\[
B_a(t; \{\alpha\}) = \sum_b \int_{t_0}^{t} d\tau C_{ab}(t - \tau) Q_b[\alpha(\tau)],
\]

\[
B'_a(t; \{\alpha\}) = \sum_b \int_{t_0}^{t} d\tau C^*_{ab}(t - \tau) Q_b[\alpha'(\tau)].
\]

Here, \(C_{ab}(t)\) is the bath correlation function, defined by equation \((2)\). The functional \(A_a\) (equation \((A.4)\)) depends only on the local time and its operator level form is just the commutator of the dissipative mode \(Q_a\).

The functional \(B_a\) (equation \((A.5)\)) does however contain memory, which is resolved via the COPI algorithm of consecutive time derivatives on all memory-contained functionals. To construct a close set of HEOM via the COPI algebra, a proper expansion of \(C_{ab}(t)\) is needed such as the exponential series, while maintaining the fluctuation-dissipation theorem of equation \((3)\). A super-Drude parametrization scheme and the resulting HEOM formalism have been presented in our previous work \([28]\). A hybrid scheme, exploiting the analytical continuation and quadrature integration methods to evaluate the fluctuation–dissipation theorem, has also been proposed \([30]\).

To illustrate the COPI algorithm, consider the super-Drude model:

\[
J_{ab}(\omega) = \frac{\eta_{ab}\omega + i\eta'_{ab}\omega^2}{[(\omega/\gamma_{ab})^2 + 1]^2}.
\]
All the parameters are real and satisfy the symmetry relations of $\eta_{ab} = \eta_{ba}$, $\gamma_{ab} = \gamma_{ba}$ and $\eta_{ab}' = -\eta_{ba}'$, as inferred from the symmetry relations implied in $J_{ab}(\omega)$. The resulting correlation functions via equation (3) are

$$C_{ab}(t \geq 0) = \left[(\nu_r + i\nu_i) + (\nu_r' + i\nu_i')\gamma_{ab}\right] e^{-\gamma_{ab}t} + \sum_{m=1}^{M} \tilde{\nu}_m e^{-\tilde{\eta}_m t} + \delta C_{ab}(t).$$  \hspace{1cm} (A.8)

The second term arises from the Matsubara poles, with $\tilde{\nu}_m = 2\pi m/\beta$ being the Matsubara frequencies, and $\tilde{\nu}_m^{ab} = -i(2/\beta)J_{ab}(-i\tilde{\nu}_m) = (\tilde{\nu}_m^{ab})^*$ is real, as inferred from the symmetry relation of the spectral density function in analytical continuation. The first term arises from the pole of the spectral density function of rank 2. The involving coefficients are summarized as follows \cite{20, 28, 53}:

$$\nu_i^{ab} = \eta_{ab}'\gamma_{ab}^3/4, \quad \nu_i = -\frac{1}{4}(\eta_{ab} + \eta_{ab}'\gamma_{ab})\gamma_{ab}^2, \quad \nu_i' = -\nu_i'\cot(\beta\gamma_{ab}/2),$$

$$\nu_i'' = -\nu_i''\cot(\beta\gamma_{ab}/2) - \nu_i''(\beta\gamma_{ab}/2) \csc^2(\beta\gamma_{ab}/2),$$

and

$$\tilde{\nu}_m^{ab} = -\frac{2(\eta_{ab}\tilde{\nu}_m + \eta_{ab}'\tilde{\nu}_m)}{\beta[(\tilde{\nu}_m/\gamma_{ab})^2 - 1]^2} \equiv \tilde{\gamma}_m^{ab}, \quad \text{with} \quad \tilde{\eta}_m^{ab} = -\frac{(\eta_{ab} + \eta_{ab}'\tilde{\nu}_m)/(m\pi)}{[(\tilde{\nu}_m/\gamma_{ab})^2 - 1]^2}. \hspace{1cm} (A.9)$$

The residue $\delta C_{ab}(t)$ can be approximated via the Markovian ansatz, i.e. $\delta C_{ab}(t) \simeq \Delta_{ab}\delta(t)$; cf equation (6).

To proceed, we denote for every distinct exponent terms in equation (A.8),

$$B_{ab}(t; \{\alpha\}) \equiv \int_{t_0}^{t} d\tau \ e^{-\gamma_{ab}(t-\tau)} Q_b[\alpha(\tau)],$$

$$\tilde{B}_{ab}(t; \{\alpha\}) \equiv \int_{t_0}^{t} d\tau \ \gamma_{ab}(t-\tau) e^{-\gamma_{ab}(t-\tau)} Q_b[\alpha(\tau)],$$

$$\tilde{B}_{m}^{ab}(t; \{\alpha\}) \equiv \sum_{b} \tilde{\eta}_m^{ab} \int_{t_0}^{t} d\tau \ e^{-\tilde{\gamma}_m^{ab}(t-\tau)} Q_b[\alpha(\tau)]; \quad m = 1, \ldots, M. \hspace{1cm} (A.11)$$

They are related to the influence generating functionals as (cf equation (A.6))

$$B_{ab} \equiv -i(B_{ab} - B'_{ab}), \quad B'_{ab} \equiv B_{ab} + B'_{ab},$$

$$\tilde{B}_{ab} \equiv -i(\tilde{B}_{ab} - \tilde{B}'_{ab}), \quad \tilde{B}'_{ab} \equiv \tilde{B}_{ab} + \tilde{B}'_{ab},$$

$$\tilde{B}_{m}^{ab} \equiv -i(\tilde{B}_{m}^{ab} - \tilde{B}_{m}^{ab}), \quad m = 1, \ldots, M. \hspace{1cm} (A.12)$$

Now we define the ADOs via

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

where

$$\mathcal{U}_n(\alpha, t; \alpha_0, t_0) \equiv \int_{\alpha_0=t_0}^{\alpha=t} D\alpha \ e^{iS[\alpha]} \mathcal{F}_n[\alpha] e^{-iS[\alpha]},$$

with

$$
\mathcal{F}_n = s_n \left\{ \prod_{a,b} [(B_{ab})^{\eta_{ab}}(B'_{ab})^{\eta_{ab}}(\tilde{B}_{ab})^{\tilde{\eta}_{ab}}(\tilde{B}'_{ab})^{\tilde{\eta}_{ab}}] \prod_{a,m} (\tilde{B}_{m}^{ab})^{\tilde{\eta}_m^{ab}} \right\} \mathcal{F}. \hspace{1cm} (A.15)
$$

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The scaling factor \( s_n \) is defined as

\[
s_n = \left\{ \prod_{a,b} \frac{|v_{ab}^* n_{ab}^*| v_{ab} |v_{ab}'| \nu_{ab} |\bar{v}_{ab}| \bar{v}_{ab}'| \bar{\nu}_{ab}'|}{n_{ab}^* n_{ab}' \bar{\nu}_{ab} n_{ab} \bar{v}_{ab}'} \right\}^{1/2} \prod_{a,m} \frac{\gamma_{ab}^* \nu_{ab}}{\gamma_{ab} \nu_{ab}^*}.
\]  

(A.16)

The ADOs \( \{ \rho_n \} \) defined in this way are dimensionless and possess uniform error tolerance to support the filtering method, as described in section 2.3. Since \( \eta_{aa}' = 0, v_{aa}' = 0 \); we set \( v_{aa}' = v_{aa} \) for the scaling factor \( s_n \) of equation (A.16) and hereafter. The index set for this complex multi-dissipative modes case is now

\[
n = \{ n_{ab}, n_{ab}', \bar{n}_{ab}, \bar{n}_{ab}', \bar{n}_{1}, \ldots, \bar{n}_{M} \}.
\]  

(A.17)

The HEOM can now be derived by taking the time derivative on \( \rho_n \). Note that the COPI method is not just a time derivative technique, but it provides a way of resolving the bath memory effect of the influence functional in the operative level. The final results are

\[
\dot{\rho}_n = -[\mathcal{I}(t) + \Gamma_n + \delta \mathcal{R}] \rho_n + \rho_n^{(-)} + \rho_n^{(-)} + \rho_n^{[\eta]},
\]  

(A.18)

where

\[
\Gamma_n \equiv \sum_{a,b} \left( n_{ab} + \bar{n}_{ab} + n_{ab}' + \bar{n}_{ab}' \right) \gamma_{ab} + \sum_{a,m} \bar{n}_{ab}^m \gamma_{ab},
\]  

(A.19)

\[
\delta \mathcal{R} \dot{\rho} = \sum_{a,b} \Delta_{ab} \{ Q_a, [Q_b, \dot{\rho}] \}.
\]  

(A.20)

The details of the swap term \( \rho_n^{(-)} \), the tier-down \( \rho_n^{(-)} \) and tier-up \( \rho_n^{[\eta]} \) terms are

\[
\rho_n^{(-)} = \sum_{a,b} \gamma_{ab} \left( (n_{ab} + 1) \bar{n}_{ab} \bar{v}_{ab} / v_{ab} | \rho_{\delta n_{ab}} + (n_{ab} + 1) \bar{n}_{ab}' \bar{v}_{ab}' / v_{ab}' | \rho_{\delta n_{ab}}' \right),
\]  

(A.21)

\[
\rho_n^{(-)} = -i \sum_{a,b} \sqrt{n_{ab}} \sqrt{v_{ab}} \{ Q_a, \rho_{\delta n_{ab}} \} + \sum_{a,b} \sqrt{n_{ab}} \sqrt{v_{ab}} \{ Q_a, \rho_{\delta n_{ab}} \}
\]  

(A.22)

\[
\rho_n^{[\eta]} = -i \sum_{a,b} \left\{ v_{ab} \sqrt{(n_{ab} + 1) / v_{ab}} \left[ Q_a, \rho_{\delta n_{ab}} \right] + v_{ab} \sqrt{(n_{ab}' + 1) / v_{ab}'} \left[ Q_a, \rho_{\delta n_{ab}}' \right] \right. 
\]  

(A.23)

\[
+ \sqrt{v_{ab} / (n_{ab} + 1) / v_{ab} \left[ Q_a, \rho_{\delta n_{ab}} \right] + \sqrt{v_{ab}' / (n_{ab}' + 1) / v_{ab}' \left[ Q_a, \rho_{\delta n_{ab}}' \right]}
\]  

\[
- i \sum_{a \neq b} v_{ab} \sqrt{(n_{ab} + 1) / v_{ab}} \left[ Q_a, \rho_{\delta n_{ab}} \right] - i \sum_{a \neq b} v_{ab} \sqrt{(n_{ab}' + 1) / v_{ab}'} \left[ Q_a, \rho_{\delta n_{ab}}' \right]
\]  

The index variations here are similar to those described in equation (11), which is just the single-mode simplification. Equation (10) is thus readily obtained.

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