Theories of quantum dissipation and nonlinear coupling bath descriptors

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The quest of an exact and nonperturbative treatment of quantum dissipation in nonlinear coupling environments remains in general an intractable task. In this work, we address the key issues toward the solutions to the lowest nonlinear environment, a harmonic bath coupled both linearly and quadratically with an arbitrary system. To determine the bath coupling descriptors, we propose a physical mapping scheme, together with the prescription reference invariance requirement. We then adopt a recently developed dissipaton equation of motion theory [R. X. Xu et al., Chin. J. Chem. Phys. 30, 395 (2017)], with the underlying statistical quasi-particle (“dissipaton”) algebra being extended to the quadratic bath coupling. We report the numerical results on a two-level system dynamics and absorption and emission line shapes. Published by AIP Publishing. https://doi.org/10.1063/1.4991779

I. INTRODUCTION

Quantum dissipation plays crucial roles in many fields of modern science. Various methods have been constructed since 1950s, with the focus on the reduced dynamics of central systems, under the influence of bath environments. Exact theories include the Feynman–Vernon influence functional path integral method\(^1\)–\(^3\) and its differential equivalence, the hierarchical-equations-of-motion formalism.\(^4\)–\(^11\) However, these theories exploit the Gaussian–Wick’s thermodynamical statistics,\(^2\)\(^,\)\(^3\) which is strictly valid only for linear bath couplings. Intrinsically, a linear bath coupling implies a weak backaction of system on environment. The lowest non-Gaussian–Wick’s environment influence requires a quadratic bath coupling. The quest here is closely related to the vibronic optical lineshape and rate problems,\(^12\)–\(^16\) beyond the linear response theory.\(^17\)–\(^19\) The interest here is also closely related to the recent efforts toward quantum computing devices, where the fluctuations of linear noise could be effectively suppressed at optimal operational situations.\(^20\)–\(^24\) The quadratic noise fluctuations would then become the dominant source of decoherence.

There are two issues on the quantum dissipation theories with quadratic coupling environments. One is related to the fact that a non-Gaussian–Wick’s environment cannot be completely characterized with linear bath correlation functions. Actually the existence of nonlinear bath couplings would also alter the appearance of linear coupling descriptors.\(^25\) The characterizations should be physically well supported. In our previous work,\(^25\) we considered a nonlinear solvation model, which however did not preserve the prescription reference invariance properties. We will revisit this issue and propose a proper mapping scheme, together with the aforementioned invariance requirement. The new bath coupling descriptors preserve the given total composite Hamiltonian with different system-and-bath decompositions. Another issue is concerned with the construction of a proper quantum dissipation theory such that the evaluated observable dynamic quantities also preserve the bath reference invariance. Various methods have been proposed, such as the time-dependent variational approaches with Davydov ansatz\(^12\),\(^26\),\(^27\) and the stochastic methods.\(^28\)–\(^30\)

In this work, we adopt the dissipaton-equation-of-motion (DEOM) approach.\(^31\),\(^32\) This is a statistical quasi-particle theory for the dynamical influence of environment that can be either bosonic or fermionic or hard-core bosonic. DEOM describes explicitly not only the reduced system but also the hybrid bath dynamics, which could be measured experimentally, such as the Fano interference,\(^33\)–\(^35\) Herzberg–Teller vibronic coupling dynamics,\(^36\) and transport current noise spectrum.\(^37\) The extended DEOM theory\(^23\) will be presented in Sec. II, where the environment assumes to be a linear-plus-quadratic coupling bath. From the construction point of view, the extended theory applies the well-established linear dissipaton algebra\(^31\),\(^32\) to the evaluation of dissipaton-pair actions. We will elaborate in due course which would resemble an Ehrenfest mean-field type of treatment on quadratic bath couplings.

The main theoretical development of this work will be presented in Sec. III. It is concerned with the nonlinear coupling bath descriptors. To that end, we propose a linear-displacement-mapping (LDM) scheme, together with the reference-bath invariance requirement. It is noticed that the quadratic coupling strength does influence the linear descriptor, but not the other way around. We report the numerical demonstrations in Sec. IV. Indeed, the extended DEOM evaluations on dynamical observables exhibit the required reference-bath invariance properties. Finally, we conclude this paper in Sec. V.
II. DYNAMICAL DISSIPATON THEORY

A. The dissipaton equation of motion theory

In this section, we review the DEOM approach to the quantum dissipative dynamics in the presence of quadratic bath coupling. Without loss of generality, we illustrate the theory for a single dissipative mode case, in which the total system-and-bath composite Hamiltonian of the form

\[ H_T = H_0 + h_B + \hat{Q}_s(\alpha_0 + \alpha_1 \hat{x}_B + \alpha_2 \hat{x}_B^2) . \]  

The system Hamiltonian \( H_0 \) and dissipative operator \( \hat{Q}_s \) are arbitrary. The bath Hamiltonian and solvation coordinate assume

\[ h_B = \frac{1}{2} \sum_j \omega_j (\hat{p}_j^2 + \hat{q}_j^2) \quad \text{and} \quad \hat{x}_B = \sum_j c_j \hat{q}_j . \]  

The solvation coordinate \( \hat{x}_B \) is a macroscopic bath operator and set to be dimensionless, as well as \( \hat{Q}_s \). The \( \alpha \)-parameters, which will be determined in Sec. III via a physical model, are then of energy unit. Throughout this paper, we set \( h = 1 \) and \( \beta = 1/(k_B T) \), with \( k_B \) and \( T \) being the Boltzmann constant and temperature, respectively. Set also \( t \geq 0 \) for the time variable.

It is well known that in the absence of nonlinear bath coupling (\( \alpha_2 = 0 \)), the influence of harmonic environment follows the Gaussian statistics. In other words, the linear bath influence is completely dictated with the Gaussian–Wick’s thermodynamic variables. Set also \( \langle \hat{x}_B(t), \hat{x}_B(0) \rangle_B \) for the free bath coordinate and temperature, respectively. Set also \( t \) to be zero for the time variable.

It is expressed in terms of \( \chi_B^{\langle \cdot \rangle}(\omega) \equiv \text{Im} \chi_B(\omega) \), which must also appear in the linear response theory. In particular, one starts with the bath spectral density

\[ \chi_B^{\langle \cdot \rangle}(\omega) = -\frac{1}{2} \sum_j c_j^2 \delta(\omega - \omega_j) = -\chi_B^{\langle \cdot \rangle}(\omega) . \]  

It is expressed in terms of \( \chi_B^{\langle \cdot \rangle}(\omega) \equiv \text{Im} \chi_B(\omega) \), with \( ^{3,38} \)

\[ \chi_B(\omega) = i \int_0^\infty dt \ e^{i\omega t} \langle \{ \hat{x}_B(t), \hat{x}_B(0) \} \rangle_B . \]  

Here, \( \{ \cdot, \cdot \} \) denotes a commutator, \( \hat{x}_B(t) \equiv e^{iH_0 t} \hat{x}_B e^{-iH_0 t} \), and \( \langle \hat{O} \rangle_B \equiv \text{Tr}_B(\hat{O} e^{-\beta H_B})/\text{Tr}_B(e^{-\beta H_B}) \). We have then

\[ \langle \hat{x}_B(t), \hat{x}_B(0) \rangle_B = \frac{1}{\pi} \int_0^\infty \frac{e^{-i\omega t} \chi_B^{\langle \cdot \rangle}(\omega) \cdot \chi_B^{\langle \cdot \rangle}(\omega)}{1 - e^{-\beta \omega}} . \]  

This is the fluctuation-dissipation theorem. \(^{3,38}\)

As \( H_T \) of Eq. (1) involves a quadratic \( \hat{x}_B^2 \)-coupling, the influence of the harmonic bath is not solely characterized with the linear correlation function \( \langle \hat{x}_B(t) \hat{x}_B(0) \rangle_B \). Additional information will be needed. We will address this issue in Sec. III, together with the \( \alpha \)-parameters that are to be determined with a physical model. Nevertheless, the dynamical influence of the \( \hat{x}_B^2 \)-coupling could be well treated with the dissipaton algebra, in particular the non-Gaussian or generalized Wick’s theorem. \(^{25,31,32}\)

The DEOM theory starts with describing the influence of environment with a finite number of statistically independent quasi-particles, the dissipatons. \(^{31,32}\) No matter whether there are nonlinear bath coupling or not, dissipatons are defined via the linear bath \( \hat{x}_B \)-coupling part only. The effects of nonlinear bath coupling will be taken into account via the generalized (non-Gaussian) Wick’s theorem to be detailed later [cf. Eqs. (21)–(23)].

To proceed, we expand Eq. (5) in an exponential series, \(^{38-40}\)

\[ \langle \hat{x}_B(t) \hat{x}_B(0) \rangle_B = \sum_{k=1}^K \nu_k e^{-\gamma_k t} . \]  

We express its time reversal in the form of \(^{9,38}\)

\[ \langle \hat{x}_B(0) \hat{x}_B(t) \rangle_B = \langle \hat{x}_B(t) \hat{x}_B(0) \rangle_B^\ast = \sum_{k=1}^K \nu_k^\ast e^{-\gamma_k t} , \]  

with \( \nu_k \) being defined via \( \gamma_k \equiv \nu_k^\ast \), which must also appear in Eq. (6). The solvation coordinate can now be expressed in the dissipatons decomposition form \(^{31,32}\)

\[ \hat{x}_B = \sum_{k=1}^K \hat{f}_k , \]  

with

\[ \hat{f}_k(t) = \delta_{jk} \nu_k e^{-\gamma_k t} , \]  

\[ \hat{f}_k(0) = \delta_{jk} \nu_k^\ast e^{-\gamma_k t} . \]

It is easy to verify that both Eqs. (6) and (7) are reproduced. As defined above, dissipatons consist of linear and statistically independent macroscopic bath degrees of freedoms. Each dissipaton is associated with a single exponent that can be complex, for its forward and backward correlation functions. The implication here will be elaborated later; see Eqs. (16) and (17).

Dynamical variables in the DEOM theory are the dissipaton density operators (DDOs), \(^{31,32}\)

\[ \rho^{(n)}(t) \equiv \rho^{(n)}_{\nu_1 \cdots \nu_n}(t) \equiv \text{Tr}_B[\hat{f}_K^{\nu_K} \cdots \hat{f}_1^{\nu_1} \hat{\rho}_T(t)] . \]  

Apparently, the reduced system density operator is just \( \rho_S(t) \equiv \rho^{(0)}(t) \). The indexes \( n \equiv \{ \nu_1 \cdots \nu_n \} \) and \( n = 1 + \cdots + n_K \) specify the configuration in terms of occupations and the total number of dissipatons, respectively. The irreducible notation, \( \langle \cdots \rangle' \), highlights the fact that \( \rho^{(n)}(t) \) defined above is truly an \( n \)-dissipaton quantity. This notation is also followed by \( \langle \hat{f}_k \rangle'^\ast = \langle \hat{f}_k \rangle^\ast \), the identity for Bosonic dissipatons that satisfy the symmetric permutation. Moreover, it will be used in reporting the generalized (non-Gaussian) Wick’s theorems [cf. Eq. (18)], the essential ingredients of dissipaton algebra; see Sec. II B.

It will be evident in Sec. II B that DEOM theory can be considered as the dissipaton algebra, together with the Schrödinger equation. The latter refers to

\[ \rho^{(n)}(t) \equiv \rho^{(n)}_{\nu_1 \cdots \nu_n}(t) \equiv \text{Tr}_B[\hat{f}_K^{\nu_K} \cdots \hat{f}_1^{\nu_1} \hat{\rho}_T(t)] , \]  

with the total composite density operator satisfying

\[ \hat{\rho}_T(t) = -i[H_T, \hat{\rho}_T(t)] . \]  

The dissipaton algebra consists of the generalized diffusion equation and the generalized (non-Gaussian) Wick’s theorems. These two components evaluate the bath \( h_B \)-action and the system-bath coupling \( H_{SB} \)-action, via the last two terms of \( H_T \) in Eq. (1), respectively.
The final DEOM formalism in the presence of both linear and quadratic bath couplings reads
\[ \hat{\rho}_{\text{in}}^{(n)} = -i \mathcal{L}_S + \sum_k n_k \gamma_k \hat{\rho}_{\text{in}}^{(n)} - i2\alpha_2 \sum_{k,j} n_k \mathcal{C}_k \hat{\rho}_{\text{in}}^{(n-1)} + \sum_{k,j} A_k \hat{\rho}_{\text{in}}^{(n+1)} + n_k (\delta_{jk} - \delta_{k,n}) \mathcal{B}_{n,j} \hat{\rho}_{\text{in}}^{(n-2)} \]
\[ -i\alpha_1 \sum_k \left( A_k \hat{\rho}_{\text{in}}^{(n+1)} + n_k \mathcal{C}_k \hat{\rho}_{\text{in}}^{(n-1)} \right). \]  \hspace{1cm} (13)

Here, \( \mathcal{L}_S \hat{O} = [\hat{H}_S, \hat{O}] \), with
\[ H_S = H_0 + (\alpha_0 + \alpha_2 (\hat{x}_B^2)) \hat{Q}_S, \]  \hspace{1cm} (14)
and
\[ A \hat{O} \equiv [\hat{Q}_S, \hat{O}], \]  \hspace{1cm} (15a)
\[ B_{kj} \hat{O} \equiv \nu_k \nu_j \hat{Q}_S \hat{O} - \nu^* \nu^* \hat{Q}_S \hat{O}, \]  \hspace{1cm} (15b)
\[ \mathcal{C}_k \hat{O} \equiv \nu_k \hat{Q}_S \hat{O} - \nu^* \hat{Q}_S \hat{O}. \]  \hspace{1cm} (15c)

B. The dissipaton algebra in the DEOM theory

1. The generalized diffusion equation

As defined in Eq. (9), each dissipaton is associated with a single exponent, for its forward and backward correlation functions, Eq. (9). This feature leads to the generalized diffusion equation, \[ \hat{\rho}_{\text{in}} \]
\[ \operatorname{tr}[ \frac{\partial \hat{f}}{\partial t} ]_{\text{in}} \hat{\rho}_T(t) = -\gamma_k \operatorname{tr}[ \hat{f}_k \hat{\rho}_T(t) ]. \]  \hspace{1cm} (16)

This together with \( \left( \frac{\partial \hat{f}}{\partial t} \right)_{\text{in}} = -i[\hat{f}_k, \hat{h}_\text{B}] \) will be used to treat the action of the bath Hamiltonian. The influence of the bath \( \hat{h}_\text{B} \)-action on the DDO is then evaluated as
\[ i \operatorname{tr}[ \hat{\rho}_T^{\hat{f}_k} \cdots \hat{f}_1^{\hat{f}_1} ]_{\text{in}} = \left( \sum_{k=1}^K n_k \gamma_k \right) \hat{\rho}_{\text{in}}^{(n)}. \]  \hspace{1cm} (17)

This is the sum-over-exponents term in Eq. (13).

2. The generalized Wick’s theorems

The Generalized Wick’s Theorem-1 (GWT-1) evaluates the linear bath coupling, where one dissipaton is added each time. It reads \[ \operatorname{tr}[ \hat{\rho}_T^{\hat{f}_k} \cdots \hat{f}_1^{\hat{f}_1} ] = \hat{\rho}_T^{(n+1)} + \sum_{k=1}^K n_k \hat{\rho}_T^{\hat{f}_k \cdots \hat{f}_1^{\hat{f}_1}}. \]  \hspace{1cm} (18)

The expression of \( \operatorname{tr}[ \hat{\rho}_T^{\hat{f}_k} \cdots \hat{f}_1^{\hat{f}_1} ] \) is similar, but with \( \hat{f}_k \) being replaced by \( \hat{g}_k \). Here [cf. Eq. (9)]
\[ \hat{g}_k \equiv \hat{g}(0) \hat{g}(0) \hat{g}(++) = \delta_{k,j} \nu_k, \]  \hspace{1cm} (19)
\[ \hat{g}_k \hat{g}_k^* = \hat{g}(0) \hat{g}(0) \hat{g}(++) = \delta_{k,j} \nu^* \nu^*. \]

The associated index \( n^\pm \) differs from \( n_k \equiv \{n_1 \ldots n_K\} \) by replacing the specified \( n_k \) with \( n_k \pm 1 \). This specifies the \( (n \pm 1) \)-particle DDO, \( \hat{\rho}_{n^\pm}(t) \), in Eq. (18). The \( \hat{Q}_S \hat{x}_B \)-action, with \( \hat{x}_B = \sum_{j=1}^N \hat{f}_j \) [Eq. (8)], can then be readily evaluated, resulting in Refs. 31 and 32
\[ \operatorname{tr}\left[ \hat{\rho}_T^{\hat{f}_k} \cdots \hat{f}_1^{\hat{f}_1} \right] = \sum_{k} A_k \hat{\rho}_{n^\pm}(t) + \sum_{n_k} n_k \mathcal{C}_k \hat{\rho}_{n^\pm}(t). \]  \hspace{1cm} (20)

This contributes to the last term in Eq. (13). The superoperators \( A \) and \( C \) had been defined in Eq. (15).

The GWT-2 is related to the quadratic bath coupling, where a pair of dissipations are added each time. We evaluated it as
\[ \operatorname{tr}\left[ \hat{\rho}_T^{\hat{f}_k} \cdots \hat{f}_1^{\hat{f}_1} \right] = \sum_{k=1}^K \hat{\rho}_T^{\hat{f}_k \cdots \hat{f}_1^{\hat{f}_1}}. \]  \hspace{1cm} (21)

The associated DDO index, \( n_{k^\pm} \), differs from \( n_k \equiv \{n_1 \ldots n_K\} \) on the specified subindexes, \( n_k \) and \( n_j \), that are replaced by \( n_k \pm 1 \) and \( n_j \pm 1 \), respectively. Together with Eqs. (8) and (19), we obtain
\[ \operatorname{tr}\left[ \hat{\rho}_T^{\hat{f}_k} \cdots \hat{f}_1^{\hat{f}_1} \right] = \sum_{k=1}^K \hat{\rho}_T^{\hat{f}_k \cdots \hat{f}_1^{\hat{f}_1}} + \sum_{n_k} n_k \mathcal{C}_k \hat{\rho}_{n^\pm}(t). \]  \hspace{1cm} (22)

The expression of \( \operatorname{tr}\left[ \hat{\rho}_T^{\hat{f}_k} \cdots \hat{f}_1^{\hat{f}_1} \right] \) is similar, but with \( \nu_k \) and \( \nu_j \) being replaced by \( \nu^* \) and \( \nu^* \), respectively. Therefore, \[ \operatorname{tr}\left[ \hat{\rho}_T^{\hat{f}_k} \cdots \hat{f}_1^{\hat{f}_1} \right] = \sum_{k=1}^K \hat{\rho}_T^{\hat{f}_k \cdots \hat{f}_1^{\hat{f}_1}} + \sum_{n_k} n_k \mathcal{C}_k \hat{\rho}_{n^\pm}(t). \]  \hspace{1cm} (23)

with \( A, B, \mathcal{C} \) being defined in Eqs. (15). The first term here, which together with the \( H_0 + \alpha_0 \hat{Q}_S \) from Eq. (1), contributes to \( H_S \) of Eq. (14). The last three terms in Eq. (23) contribute to the \( \alpha_2 \)-terms in Eq. (13).

We have thus completed the derivations of the DEOM formalism, Eq. (13), with the total composite Hamiltonian of Eq. (1). The bridge between Eq. (13) and the Liouville–von Neumann equation (12) is the generalized diffusion equation and the generalized Wick’s theorems presented above. These ingredients of the dissipaton algebra, including the most recent GWT-2, Eq. (21), have all been established. 25,31,32
C. Comments

Some additional features are as follows. In the absence of quadratic coupling ($\alpha_2 = 0$), Eq. (13) reduces to the previous result,\textsuperscript{31,32} which is identical to the hierarchical-equations-of-motion formalism.\textsuperscript{4–9} The latter is a path integral influence functional based theory,\textsuperscript{1} with $\rho_n^{(\sigma=0)}$ being considered as mathematical auxiliaries. The above observations validate the generalized diffusion equation (16) that leads to Eq. (17), and the GWT-1, Eq. (18) that, together with its time-reversal counterpart, leads to Eq. (20). While the conventional diffusion equation\textsuperscript{41} and Wick’s theorem\textsuperscript{4} are concerned only with c-number properties of Gaussian statistics, those two generalizations, Eqs. (16) and (18), go with reduced operators in the system-subspace, with the arbitrary Hamiltonian $H_S$ and arbitrary dissipative mode $Q_S$.

Nevertheless, the GWT-2, Eq. (21), which had been validated in the high-temperature regime,\textsuperscript{25} is subject to further scrutiny. The resulted $\langle f^f_f^f \rangle_n$ [cf. the second term in Eq. (21)] is just the bath ensemble average on the product of the specified pair without time ordering. As inferred from its construction, Eq. (21) would be an Ehrenfest mean-field type of treatment on quadratic bath couplings. We will defer this issue to future study.

III. CHARACTERIZATIONS ON LINEAR AND QUADRATIC BATH COUPLINGS

A. Solvation mode description

To complete the quantum dissipation theory with nonlinear coupling environments, we shall further have physically supported $\alpha$-parameters. In the presence of quadratic bath couplings, the $\alpha$-parameters cannot be characterized solely via the linear response theory. In the following, we adopt the well-established linear solvation model\textsuperscript{18,42–45} and extend it to a unified characterization on both linear and quadratic bath coupling $\alpha$-parameters. In this model, the last term of Eq. (1) arises from the surrounding environment rearrangements in response to the dissipative system operator $Q_S$. The solvation model approach is to rewrite Eq. (1) as

$$H_T = H_0 + h_B + \mathcal{Q}_S(h_B' - h_B),$$

with

$$\delta h_B \equiv h_B' - h_B = \alpha_0 + \alpha_1 \tilde{x}_n + \alpha_2 \tilde{x}_n^2.$$  \hspace{2cm} \text{(25)}

The kinetics energies in $h_B'$ and $h_B$ are the same. The surrounding environment rearrangements, which hybridize with the system dissipative mode $Q_S$, is measured by the potential energy difference; i.e., $\delta h_B = v_n^\prime - v_n$. The total bath involves both $\tilde{x}_n$, defined in Eq. (2), and the secondary bath coordinates, $\tilde{x} \equiv \{\tilde{x}_n\}$.

To highlight the fact that the solvation dynamics, $\tilde{x}_n(t) = e^{i\Delta t} \tilde{x}_n e^{-i\Delta t}$, is a Brownian motion in the bare bath, we recast the reference bath $h_B$ of Eq. (2) in Caldeira–Leggett’s interaction form,\textsuperscript{42}

$$h_B = \frac{1}{2} \omega_n (\tilde{p}_n^2 + \tilde{x}_n^2) + \frac{1}{2} \sum_k \omega_k \left[ \tilde{p}_k^2 + \left( \tilde{x}_k - \frac{\tilde{c}_k}{\omega_k} \tilde{x}_n \right)^2 \right]$$

$$= \frac{1}{2} \omega_n \tilde{p}_n^2 + \frac{1}{2} \sum_k \omega_k \tilde{p}_k^2 + v_n(\tilde{x}_n; \tilde{x}).$$ \hspace{2cm} \text{(26)}

The potential energy here is given by

$$v_n(\tilde{x}_n; \tilde{x}) = \frac{1}{2} \omega_n (\tilde{x}_n \tilde{x}_n^2 - \tilde{x}_n \tilde{x} + \frac{1}{2} \sum_k \tilde{c}_k \tilde{c}_k)$$

$$= \frac{1}{2} \omega_n (\tilde{x}_n \tilde{x}_n^2 - \tilde{x}_n \tilde{x} + \frac{1}{2} \sum_k \tilde{c}_k \tilde{c}_k)$$  \hspace{2cm} \text{(27)}

with $\tilde{x} \equiv \{\tilde{x}_k\}$.

The resultant classical friction kernel function reads\textsuperscript{38}

$$\tilde{c}(t) = \omega_n \sum_k \frac{\tilde{c}_k^2}{\omega_k} \cos(\tilde{\omega}_k t).$$ \hspace{2cm} \text{(28)}

The Langevin force is given by $F(t) = \epsilon \tilde{c} e^{-i\tilde{c} t}$, where $\tilde{h} = \frac{1}{2} \sum_k \tilde{c}_k (\tilde{p}_k^2 + \tilde{x}_k^2)$ is the secondary bath Hamiltonian. The above Brownian oscillator descriptions result in Eq. (4) the well-established relation,\textsuperscript{3,38}

$$\chi_n(\omega) = \frac{\omega_n}{\omega^2_n - \omega^2 - i\omega \zeta(\omega)},$$ \hspace{2cm} \text{(30)}

where

$$\zeta(\omega) \equiv \int_0^\infty dt e^{i\omega t} \tilde{c}(t).$$ \hspace{2cm} \text{(31)}

The relevant $h_B'$ in Eq. (25) is similar as Eqs. (26)–(28),

$$h_B' = \frac{1}{2} \omega_n(\tilde{p}_n^2 + \tilde{x}_n^2) + \frac{1}{2} \sum_k \tilde{\omega}_k \left[ \tilde{p}_k^2 + \left( \tilde{x}_k - \frac{\tilde{c}_k}{\omega_k} \tilde{x}_n \right)^2 \right]$$

$$= \frac{1}{2} \omega_n \tilde{p}_n^2 + \frac{1}{2} \sum_k \tilde{\omega}_k \tilde{p}_k^2 + v_n'(\tilde{x}_n'; \tilde{x}).$$ \hspace{2cm} \text{(32)}

It has the identical kinetic energy as that in Eq. (26), but a different potential energy [cf. Eqs. (27) and (28)],

$$v_n'(\tilde{x}_n'; \tilde{x}') = \frac{1}{2} \omega_n (\tilde{x}_n' + \tilde{n}) \tilde{x}_n'^2 - \tilde{x}_n' \tilde{x}' + \frac{1}{2} \sum_k \omega_k \tilde{x}_k'^2,$$  \hspace{2cm} \text{(33)}

with

$$\tilde{n} = \sum_k \frac{\tilde{c}_k^2}{\omega_k} \text{ and } \tilde{x}' = \sum_k \tilde{c}_k \tilde{x}_k.$$ \hspace{2cm} \text{(34)}

Here,

$$\tilde{x}_n' = (\omega_n^2/\omega_n^2) \tilde{x}_n - \tilde{d}_n$$ \hspace{2cm} \text{(35a)}$$

$$\tilde{x}_k' = (\omega_k^2/\omega_k^2) \tilde{x}_n - \tilde{d}_k.$$ \hspace{2cm} \text{(35b)}

Therefore the two reference bath Hamiltonians differ by

$$\delta h_B = v_n' - v_n = [0] + [I] + [II].$$ \hspace{2cm} \text{(36)}

where

$$[0] = \frac{1}{2} \theta_n (\omega_n' + \tilde{n}) d_n^2 - d_n \sum_k (\omega_n' \tilde{d}_k) \tilde{c}_k \tilde{d}_k'$$

$$+ \frac{1}{2} \sum_k \tilde{c}_k \tilde{d}_k \tilde{d}_k'$$, \hspace{2cm} \text{(37a)}

$$[I] = -\omega_n (\omega_n' + \tilde{n}) d_n \tilde{x}_n + \tilde{x}_n \sum_k (\theta_n' \tilde{d}_k) \tilde{c}_k \tilde{d}_k'$$

$$+ \sum_k \left[ (\omega_n' \tilde{d}_k) \tilde{c}_k \tilde{d}_k' - \tilde{d}_k \tilde{c}_k \tilde{d}_k' \right] \tilde{x}_n,$$ \hspace{2cm} \text{(37b)}

$$[II] = \frac{1}{2} (\omega_n^2 - 1) \omega_n + \theta_n \tilde{n} - \tilde{n} \tilde{x}_n^2 + \frac{1}{2} \sum_k (\tilde{d}_k - 1) \omega_k \tilde{x}_k^2$$

$$- \tilde{x}_n \sum_k \left[ (\theta_n' \tilde{d}_k) \tilde{c}_k - \tilde{c}_k \tilde{x}_n \right].$$ \hspace{2cm} \text{(37c)}
Apparently, [I] and [II] describe the linear and quadratic bath couplings, respectively, whereas \([0] = \alpha_0\). The associating \(\alpha\)-parameters could be determined via the statistical effect of the secondary bath on the solvation coordinate. In other words, one should effectively relate \(\tilde{x}_k\) with \(\check{x}_k\) so that \([I] = \alpha_1 \check{x}_n\) and \([II] = \alpha_2 \tilde{x}_n\).

In our previous work,\(^25\) we considered a linear-displacement-mapping (LDM) ansatz, assuming that the secondary bath was of \(\tilde{x}'_k = \tilde{x}_k\); i.e., \(\tilde{\theta}_k \equiv \tilde{\omega}'_k/\tilde{\omega}_k = 1\) and \(\tilde{d}_k = 0\) in Eq. (35b). Proposed in this work is an alternative set of LDM rules, based on the exact results of pure-linear displacements’ case where \(d_k \neq 0\); see Sec. III B. A new LDM ansatz is followed, which together with the reference-bath invariance requirement determines the \(\alpha\)-parameters in Sec. III C. We shall see then that for the completeness of the present coupling bath descriptors, one has to choose \(\tilde{\theta}_k = 1\) for Eq. (35b). We comment on the final results in conjunction with the DEOM evaluations in Sec. III D.

B. Mapping rules via the linear-displacement limit

For the rules of LDM, we consider in this subsection the results of pure-linear displacements’ \((\alpha_2 = 0)\) limit, where \(\omega'_\alpha = \omega_\alpha\) and \(\tilde{\omega}'_k = \tilde{\omega}_k\), cf. Eq. (35). Let us start with the original form of bath Hamiltonian, \(h_B\), of Eq. (2), and

\[
(h'_n - h_n)_{\alpha_2 = 0} = \frac{1}{2} \sum_j \omega_j [\hat{q}_j - d_j^2 - \hat{q}_j^2]
\]

\[
= \frac{1}{2} \sum_j \omega_j d_j^2 - \sum_j \omega_j d_j \hat{q}_j.
\]

(38)

Let the last term be

\[
\sum_j \omega_j d_j \hat{q}_j \equiv (2\lambda_\omega)^{\frac{1}{2}} \sum_j c_j \hat{q}_j = (2\lambda_\omega)^{\frac{1}{2}} \check{x}_n.
\]

(39)

Here, \(\check{x}_n = \sum_j c_j \hat{q}_j\), as defined in Eq. (2); therefore,

\[
\omega_j d_j = (2\lambda_\omega)^{\frac{1}{2}} c_j.
\]

(40)

The first term in Eq. (38) is then

\[
\frac{1}{2} \sum_j \omega_j d_j^2 = \lambda_\omega n \sum_j \frac{c_j^2}{\omega_j} = \frac{\lambda_\omega n}{\pi} \int_{-\infty}^{\infty} d\omega \chi_n^{(-)}(\omega).
\]

(41)

The last identity is obtained via Eq. (3). Consider now the Kramers–Kronig relation,\(^3,38\)

\[
\chi^{(+)}_B(\omega) = -\frac{1}{\pi} \text{P} \int_{-\infty}^{\infty} dw \frac{\chi^{(-)}_B(\omega')}{\omega' - \omega},
\]

between \(\chi^{(+)}_B(\omega) = \text{Re} \chi_B(\omega)\) and \(\chi^{(-)}_B(\omega) = \text{Im} \chi_B(\omega)\). Here \(\text{P}\) denotes the principle part. As \(\chi^{(-)}_B(\omega = 0) = 0\), we have

\[
\chi_B(0) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \frac{\chi^{(-)}_B(\omega)}{\omega} = \frac{1}{\omega_B}.
\]

(43)

The last identity is obtained by using Eq. (30). Substituting Eq. (43) into Eq. (41), we obtain

\[
\lambda = \frac{1}{2} \sum_j \omega_j d_j^2.
\]

(44)

Together with Eq. (39), we obtain Eq. (38) the expression

\[
(h'_n - h_n)_{\alpha_2 = 0} = \lambda - (2\lambda_\omega)^{\frac{1}{2}} \check{x}_n.
\]

(45)

Turn to the solution mode description, Eq. (26), in which the overall bath Hamiltonian \(h_B\) is partitioned into the solution mode and the secondary bath environments. In the case of pure-linear displacements, where \(\omega'_\alpha = \omega_\alpha\) and \(\tilde{\omega}'_k = \tilde{\omega}_k\), Eq. (35) becomes

\[
\check{x}_n' = \check{x}_n - d_n, \quad \tilde{x}_n' = \tilde{x}_n - \tilde{d}_n.
\]

(46)

Here, \(d_n = \sum_j c_j d_j\), as implied in \(\check{x}_n = \sum_j c_j \hat{q}_j\) via Eq. (2). Using Eqs. (40) and (44), we have

\[
d_n = \sum_j c_j d_j = \frac{1}{(2\lambda_\omega)^{\frac{1}{2}}} \sum_j \omega_j d_j^2 = (2\lambda/\omega_\alpha)^{\frac{1}{2}}.
\]

(47)

Moreover, in the absence of quadratic couplings, one should also have \(c_k^2 = c_k\); cf. Eq. (32) versus Eq. (26). Consequently, Eqs. (37) become \([II]_{\alpha_2 = 0} = 0\) and

\[
[0]_{\alpha_2 = 0} = \frac{1}{2} (\omega_\alpha + \tilde{\eta}) d_n^2 - d_n \tilde{D} + \frac{1}{2} \tilde{d}_T \tilde{\omega} \tilde{d},
\]

\[
[II]_{\alpha_2 = 0} = - (\omega_\alpha + \tilde{\eta}) d_n \check{x}_n + \check{x}_n \tilde{D} + d_n \check{X} - \tilde{d}_T \tilde{\omega} \check{x}.
\]

(48)

Here, \(\check{x} = \{\check{x}_k\}\) and \(\tilde{d} = \{\tilde{d}_k\}\) are the column vectors; the superscript \(T\) denotes the transpose; \(\tilde{\omega}\) is a diagonal matrix of frequencies \(\{\tilde{\omega}_k\}\), and [cf. \(\check{X}\) in Eq. (28)]

\[
\tilde{D} = \sum_k c_k \tilde{d}_k.
\]

(49)

Note that \(\check{X} - \tilde{D} = \sum_k \tilde{c}_k (\check{x}_k - \tilde{d}_k)\), which appears as the secondary bath analog of \(\check{x}_n - d_n = \sum_j c_j (\hat{q}_j - d_j)\).

Now from the fact that Eq. (45) is statistically equivalent to \(h'_n - h_n = [0]_{\alpha_2 = 0} + [I]_{\alpha_2 = 0}\), via Eq. (48), we have

\[
2\lambda = (\omega_\alpha + \tilde{\eta}) d_n^2 - 2d_n \check{D} + \tilde{d}_T \tilde{\omega} \tilde{d},
\]

\[
(2\lambda_\omega)^{\frac{1}{2}} \check{x}_n = (\omega_\alpha + \tilde{\eta}) d_n \check{x}_n - \check{x}_n \tilde{D} - d_n \check{X} + \tilde{d}_T \tilde{\omega} \check{x}.
\]

Together with Eq. (47), these two expressions are just

\[
0 = \tilde{\eta} d_n^2 - 2d_n \check{D} + \tilde{d}_T \tilde{\omega} \tilde{d},
\]

\[
0 = \tilde{\eta} d_n \check{x}_n - \check{x}_n \tilde{D} - d_n \check{X} + \tilde{d}_T \tilde{\omega} \check{x}.
\]

(50)

These basic requirements would suggest the following rules of mapping:

\[
\check{D} \Rightarrow \tilde{\eta} d_n, \quad \tilde{d}_T \tilde{\omega} \tilde{d} \Rightarrow \tilde{\eta} d_n^2,
\]

\[
\check{X} \Rightarrow \tilde{\eta} \check{x}_n, \quad \tilde{x}_n \tilde{\omega} \check{x} \Rightarrow \tilde{\eta} \check{x}_n, \quad \tilde{d}_T \tilde{\omega} \check{x} \Rightarrow \tilde{\eta} d_n \check{x}_n.
\]

(51)

It seems that only the first two were independent, which would lead to the third mapping and the last two mappings, respectively.

C. The LDM ansatz and reference-bath invariance

The LDM rules [Eq. (51)] are not sufficient for achieving Eq. (37) the required expressions; i.e., \([0] = \alpha_0\), \([I] = \alpha_1 \check{x}_n\), and \([II] = \alpha_2 \tilde{x}_n\), respectively. Adopt further

\[
\varphi = (\tilde{c}_k^* \tilde{c}_k) \tilde{\omega}_k^2, \quad \tilde{\theta}_k \equiv \tilde{\omega}_k^2/\tilde{\omega}_k.
\]

(52)

These two parameters will be determined later; see Eq. (65) and the comments there. The above ansatz, Eq. (52), leads to...
Eq. (37) the expressions,
\[
0 = \frac{1}{2} \theta_0 (\omega_n + \tilde{\eta}) d_0^2 - (\theta_0 \tilde{\eta} \tilde{\varphi}) \frac{1}{2} d_0 \tilde{d} + \frac{\tilde{\varphi}}{2} d^T \tilde{\omega} d
\]
\[
\Rightarrow \frac{1}{2} \theta_0 (\omega_n + \tilde{\eta}) d_0^2 + (\theta_0 \tilde{\eta} \tilde{\varphi}) \frac{1}{2} \tilde{d} \tilde{d} + \frac{\tilde{\varphi}}{2} \tilde{d}^T \tilde{\omega} d_0^2,
\]
\[
[53a]

[II] = - \theta_0 (\omega_n + \tilde{\eta}) d_0 \tilde{d} + (\theta_0 \tilde{\eta} \tilde{\varphi}) \frac{1}{2} (\tilde{d} \tilde{d} + \tilde{d} \tilde{d}^T)
\]
\[
\Rightarrow - \theta_0 (\omega_n + \tilde{\eta}) d_0 \tilde{d} + \frac{1}{2} [2 \theta_0 (\tilde{\eta} \tilde{\varphi}) \frac{1}{2} - \tilde{d} \tilde{d} + \tilde{d} \tilde{d}^T] d_0 \tilde{d},
\]
\[
[53b]

\alpha_1 = -(2 \lambda \omega_n) \frac{1}{4} \Theta,
\]
\[
\alpha_2 = \frac{\omega_n}{2} (\Theta - 1),
\]
\[
\text{where } \Theta = \theta_0^2 + (\tilde{\eta} / \omega_n) [\tilde{d} - (\theta_0 \tilde{\eta} \tilde{d} / \tilde{\varphi}) \frac{1}{4}]^2.
\]
\[
[55]

\tilde{h}_n + \tilde{Q}_b (\tilde{h}_n - h_n) = h_n + (1 - \tilde{Q}_b) (h_n - h_n). [56]

Comparing Eqs. (59) and (60), we obtain
\[
\Theta = (\Theta')^{-1} = \theta_0^2.
\]
\[
[63]

We have thus determined the \( \alpha \)-parameters in both Eqs. (54) and (60), which stand for the reference-h\(_B\) and the reference-h\(_B'\) based descriptions, respectively.

To conclude this subsection, we should also point out that, for the completeness of description, we have to set \( \tilde{\theta}_k = \omega' / \omega_k = 1 \). In fact, Eq. (63) leads to the second terms in Eq. (55) and its equivalent Eq. (61) be zero. It results in a single relation, \( \theta_0 \varphi = \tilde{\varphi}^3 \), for two undetermined parameters. The uncertainty here reflects also in \( \chi'_B (\omega) = \omega_0 / [\omega_0^2 - \omega^2 - \omega \zeta' (\omega)] \), the reference-h\(_B'\) counterpart to Eq. (30), with [cf. Eqs. (29) and (31)]
\[
\zeta' (\omega) \equiv \int_0^\infty dt e^{i \omega t} \omega \sum_k \zeta^2 \cos(\omega_k t) = \tilde{\varphi}^2 \zeta (\omega / \tilde{\varphi}). [64]
\]

D. Discussions and comments

Let us summarize the present nonlinear solvation model description in conjunction with the DEOM evaluations. First of all, Eqs. (54) and (60), with Eq. (63), are
\[
\alpha_0 = \lambda \theta_0^2, \quad \alpha_1 = -(2 \lambda \omega_n) \frac{1}{4} \Theta,
\]
\[
[66]

\alpha_2 = \frac{\omega_n}{2} (\theta_0^2 - 1),
\]
\[
[67]

\text{and}
\]
\[
\alpha_0' = \lambda, \quad \alpha_1' = (2 \lambda \omega_n / \theta_0)^\frac{1}{4},
\]
\[
\alpha_2' = \frac{\omega_n}{2 \theta_0} (1 - \theta_0^2).
\]
\[
[67]

\text{As } \tilde{d}_n \text{ is a stochastic variable, the sign of } \alpha_1 \text{ (or } \alpha_1') \text{ does not matter, but that of } \alpha_2 \text{ makes difference. The quadratic coupling strength is dictated by } \theta_0 \equiv \omega_0 / \omega_n \text{ that enters also explicitly into } \alpha_0, \alpha_1, \text{ and } \alpha_1'. \text{ The above two sets of } \alpha \text{-parameters, Eqs. (66) and (67), are used in the reference-h}_B \text{ and the reference-h}_B' \text{ based prescriptions, respectively. In terms of the total composite Hamiltonian, that is [cf. Eq. (56)]}
\]
\[
H_T = H_0 + h_n + \tilde{Q}_b (\alpha_0 + \alpha_1 \tilde{d}_n + \alpha_2 \tilde{d}_n^2)
\]
\[
= H_0 + h_n' + (1 - \tilde{Q}_b) (\alpha_0' + \alpha_1' \tilde{d}_n + \alpha_2' \tilde{d}_n^2).
\]
\[
[68]

\text{The DEOM theory presented in Sec. II follows the } h_B \text{-based description. The involving dissipation terms } \{\tilde{d}_n\} \text{ arise from Eq. (5), with } \chi_B (\omega) = \text{Im } \chi_B (\omega) \text{ of Eq. (30). Similarly, the } h_B' \text{-based counterpart goes by}
\]
\[
\Theta' \equiv \theta_0^2 + (\tilde{\eta} / \omega_n) [\tilde{d} - (\theta_0 \tilde{\eta} \tilde{d} / \tilde{\varphi}) \frac{1}{4}]^2.
\]
\[
[61]

\text{Apparently, } \theta_n' \theta_n' = \tilde{\varphi}^2 = \varphi \varphi' = 1, \text{ and [cf. Eq. (47)]}
\]
\[
\lambda' = \frac{1}{2} \omega_n'^2 \frac{1}{4} \theta_n \omega_n (\theta_n'^2 \frac{1}{2} \theta_n^2) = \theta_n'^2 \lambda.
\]
\[ \chi'_b(\omega) = \frac{\omega_\theta}{\omega_{\theta}^2 - \omega^2 - i\omega\zeta(\omega)}, \quad (69) \]

with \( \zeta(\omega) = \zeta(\omega) \) [cf. Eqs. (64) and (65)] being identical to that in Eq. (30). The dissipations \( \langle \dot{\rho}_b \rangle \) now go with \( \chi''_b(\omega) = \text{Im} \chi_b(\omega) \) being used in the fluctuation-dissipation theorem [cf. Eq. (5)]. The \( h'_b \)-based DEOM counterparts to Eqs. (13)–(15) are then followed, by replacing \( \hat{Q}_b \) with \( 1 - \hat{Q}_b \) and the \( \alpha \)-parameters with their \( \alpha' \)-counterparts, as specified in Eq. (68). As demonstrated later, the present DEOM evaluations on dynamical observables, such as the optical spectroscopies, agree well with the reference-bath invariance requirement.

Finally, we consider the diffusive limit, with the solvation mode in the \( h_0 \)-based and the \( h'_b \)-based descriptions being characterized, respectively, by

\[ \chi_d(\omega) = \frac{1}{\gamma_0 - i\omega} \quad \text{and} \quad \chi'_d(\omega) = \frac{1}{\gamma_0' - i\omega}. \quad (70) \]

This is the limit of \( \zeta(\omega) \approx \zeta(\omega = 0) \equiv \zeta \gg \omega_\theta, \omega'_b \), but \( \omega_{\theta}^2 / \zeta \equiv \gamma_0 \) and \( \omega_{\theta}^2 / \zeta \equiv \gamma_0' \) being finite. Let

\[ \theta_0 \equiv \gamma_0 / \gamma_0'. \quad (71) \]

As it formally satisfies \( \theta_0 = \theta_0', \) Eqs. (66) and (67) in the diffusive limit read, respectively,

\[ \begin{align*}
\alpha_0 &= \lambda \theta_0, \\
\alpha_1 &= -(2\lambda \gamma_0)^{\frac{1}{2}} \theta_0, \\
\alpha_2 &= \frac{\gamma_0}{2} (\theta_0 - 1)
\end{align*} \quad (72) \]

and

\[ \begin{align*}
\alpha'_0 &= \lambda, \\
\alpha'_1 &= (2\lambda \gamma_0)^{\frac{1}{2}}, \\
\alpha'_2 &= \frac{\gamma_0}{2} (1 - \theta_0) = -\alpha_2.
\end{align*} \quad (73) \]

The DEOM evaluations, with either the Brownian oscillator or the diffusive environment, all agree well with the aforementioned reference-bath invariance requirement. Further elaboration of this aspect will be given in Sec. IV.

**IV. ILLUSTRATIONS**

Consider first the DEOM evaluations on absorption and emission lineshapes of a two-level system, described by

\[ H_T = h_g |g\rangle\langle g| + (h_e + \omega_{eg}) |e\rangle\langle e| \]

\[ = \omega_{eg} |e\rangle\langle e| + h_e (|e\rangle\langle e| + |g\rangle\langle g|) \]

\[ = \omega_{eg} |e\rangle\langle e| + h_e (|g\rangle\langle g| + |e\rangle\langle e|), \quad (74) \]

The last two expressions represent the bath \( h_e \) and \( h_g \) based descriptions, associating with the system in the ground and excited states, respectively. The above total composite Hamiltonian does assume the form of Eq. (68), with \( H_0 = \omega_{eg} |e\rangle\langle e| \) and \( \hat{Q}_s = |e\rangle\langle e| \) that gives \( 1 - \hat{Q}_s = |g\rangle\langle g| \), whereas

\[ \delta h_b = h_e - h_g = \alpha_0 + \alpha_1 x_{\theta_b} + \alpha_2 z_{\theta_b}^2, \]

\[ \delta h'_b = h_g - h_e = \alpha'_0 + \alpha'_1 x_{\theta'_b} + \alpha'_2 z_{\theta'_b}^2. \quad (75) \]

The \( \alpha \)- and \( \alpha' \)-parameters were summarized in Sec. III D. For numerical demonstrations, we adopt

\[ \zeta(\omega) = \frac{i\tilde{\gamma}_\theta \omega_\theta}{\omega + i\tilde{\gamma}_\theta}. \quad (76) \]

All frequency variables are reported in units of \( \omega_\theta \). We set \( \tilde{\gamma} = 10, \tilde{\gamma}_\theta = 15, \) and \( k_B T = 1 \). Set also \( \lambda = 1 \), except for the linear-bath-coupling-free case where \( \lambda = 0 \).

The DEOM evaluations can be carried out with either the \( h_e \)-based or the \( h_g \)-based prescription. For an adiabatic two-level system, Eq. (74), the DEOM in each prescription has two steady-state solutions. They are the thermal equilibrium ground and excited states and used as the initial states for the DEOM evaluations of the absorption and emission spectrums, respectively. In particular, the DEOM (13) in the \( h_g \)-based prescription has the equilibrium ground state of \( \rho_0^{(a)} |g\rangle_{eq} = \rho_0^{(0)} \delta_{00} \), whereas the excited state correspondences, \( \{\rho_0^{(n)}: n \geq 0\}_{eq} \), need to be evaluated.\(^{32}\) The steady-state solutions to the \( h_g \)-based DEOM are just the opposite. The reference-bath invariance requires the DEOM evaluations on optical spectrums be identical, with either the \( h_g \)-based or the \( h_e \)-based prescription. The present DEOM formalism numerically well preserves the prescription invariance property, without viable deviations in the following demonstrations.

**Figure 1** reports the absorption (solid) and emission (dashed) line shapes for the non-Markovian Brownian oscillator bath. Spectrums are scaled individually to have the same peak height in each panel. In the absence of quadratic bath coupling (i.e., \( \alpha_2 = 0 \), the emission spectrum is the mirror image of the absorption, and each line shape profile appears to be symmetric around its peak position; see Fig. 1(c). Both the symmetric lineshape and the mirror image relations break down whenever the quadratic bath coupling exists; see Figs. 1(a) and 1(b). The observed skews in individual lineshape profiles are all in qualitative agreements with the secondary-bath-free but analytical results.\(^{13}\) The demonstrations here go with \( \theta_0 = 0.75 \). In the pure quadratic case, the absorption (solid, with either red or black) and the emission (dashed, with different color) are mirror images to each other, due to the inverted frequency arrangement here; see Fig. 1(b). In Fig. 1(a), the
We have also critically examined the recently proposed extended DEOM formalism, 25 which is exact for the pure-linear coupling bath limit. 31,32 In view of its construction, as discussed at the end of Sec. II, this extended approach resembles an Ehrenfest mean-field-type treatment on quadratic bath couplings. As a result, it would only be applicable in a limited range, estimated to be about 0.7 < ω′ 2 /ωη < 1.4 due to our numerical test so far. Nevertheless, those applicable DEOM results, covering from the non-Markovian Brownian oscillator to the diffusive bath environments, are found to be in very good agreement with the reference-bath invariance requirement. Efforts toward an exact theory with nonlinear coupling baths and extensions from two-level to multi-level systems are under way.

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