

QUANTUM MECHANICS OF DISSIPATIVE SYSTEMS

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■ **Abstract** Quantum dissipation involves both energy relaxation and decoherence, leading toward quantum thermal equilibrium. There are several theoretical prescriptions of quantum dissipation but none of them is simple enough to be treated exactly in real applications. As a result, formulations in different prescriptions are practically used with different approximation schemes. This review examines both theoretical and application aspects on various perturbative formulations, especially those that are exact up to second-order but nonequivalent in high-order system-bath coupling contributions. Discrimination is made in favor of an unconventional formulation that in a sense combines the merits of both the conventional time-local and memory-kernel prescriptions, where the latter is least favorite in terms of the applicability range of parameters for system-bath coupling, non-Markovian, and temperature. Also highlighted is the importance of correlated driving and dissipation effects, not only on the dynamics under strong external field driving, but also in the calculation of field-free correlation and response functions.

1. INTRODUCTION

Quantum dissipation refers to the dynamics of a quantum system of primary interest in contact with a quantum bath of practically infinite degrees of freedom. The key theoretical quantity in quantum dissipation is the reduced density operator $\rho(t) \equiv \text{tr}_B \rho_T(t)$, i.e., the partial trace of the total system and bath composite $\rho_T(t)$ over all the bath degrees of freedom. For a system dynamical variable A , its expectation value, $\bar{A}(t) = \text{Tr}[A\rho_T(t)] = \text{tr}[A\rho(t)]$, can therefore be evaluated with the substantially reduced system degrees of freedom. Quantum dissipation theory governs the evolution of the reduced density operator $\rho(t)$, where the effects of bath are treated in a quantum statistical manner. It thus provides not just the aforementioned numerical advantage, but also the irreversibility of quantum statistical mechanics.

Because of its fundamental importance in almost all fields of modern science, quantum dissipation theory has remained as an active topic of research since about the middle of the past century. Its development involved scientists working in fields as diversified as nuclear magnetic resonance (1–4), quantum optics (5–13),

quantum information and quantum measurement (14, 15), solid-state physics and material science (16–18), mathematical physics (19–23), nonlinear spectroscopy (24–34), and statistical dynamics and chemical physics (35–68).

The challenge here arises from the combined effects of strong system-bath interaction, non-Markovian dissipation, and time-dependent external driving. These combined effects can be incorporated in a formally exact manner via the Feynman-Vernon influence functionals of path integral formalism (35–37). Numerically exact methods for the path integral formalism, such as quantum Monte Carlo techniques combined with iterative forward-backward propagation schemes (69–71), have set up the benchmark to investigate non-Markovian dynamics beyond the weak system-bath interaction regime. The integral formalism is, however, neither numerically practical for realistic systems nor theoretically tractable for further construction of such as nonlinear spectroscopy formulations.

This review considers the differential formalism of quantum dissipation theory, abbreviated hereafter as QDT, which is also often termed as quantum master equations in literature. The present review focuses on some second-order QDT formulations in terms of their constructions and applicabilities, and sheds light particularly on the implication of the correlation between non-Markovian dissipation and external time-dependent field drive. The high-order perturbative formulations are usually too complicated for general purpose (72–74). The canonical transformation methods can effectively reduce the system-bath coupling strength via considering the transformed reduced system such as polaron (17, 18) or including the solvation modes into explicit consideration. Readers may refer to, for example, Reference 52 on this topic. Semiclassical methods (75–87) where it would be practical to include the strong system-bath coupling effects are not included in this review.

We note that there has recently been an increasing interest in stochastic Hilbert- and/or Liouville-space dynamics that unravels the reduced description (11, 12, 88–92). Quantum stochastic description provides not only numerical methods on reduced dynamics propagations (89, 90) but also powerful tools for the construction of exact QDT formulations (91–94). Stochastic interpretation is also closely related to continuous quantum measurements (14, 15). Readers may refer to Reference 88, for example, for the details of stochastic quantum dissipation.

Throughout this review, $\hbar \equiv 1$ and $\beta \equiv 1/(k_B T)$ for simplicity, where k_B is the Boltzmann constant and T the temperature.

2. BACKGROUND OF NONEQUILIBRIUM QUANTUM STATISTICAL MECHANICS

In this section, we review some background on the (two-time) correlation function and response function that arise from linear response theory. We discuss the symmetry, detailed-balance, and fluctuation-dissipation relations implied there. The importance of correlation/response functions to nonequilibrium statistical mechanics is similar to that of partition functions to equilibrium statistical

mechanics. Correlation/response functions have been widely used in the study of physical problems such as spectroscopy (24–34), transport (38–43), and reaction rate (68, 95–97).

2.1. Correlation and Response Functions Versus Linear Response Theory

Consider the measurement on a dynamical variable A with a classical probe field $\epsilon(t)$ that couples with the system as $-B\epsilon(t)$. For simplicity, both operators A and B are assumed to be Hermite. The total composite material system was initially at the thermal equilibrium, $\rho_M^{\text{eq}} = e^{-\beta H_M} / \text{Tr} e^{-\beta H_M}$, before external field disturbance. The field-induced deviation in $\bar{A}(t)$ from its equilibrium expectation value is $\delta\bar{A}(t) = \text{Tr}\{A[\rho_T(t) - \rho_M^{\text{eq}}]\}$, where $\rho_T(t)$ is the total composite system density operator in the presence of external field. To the first order of the external disturbance, we have

$$\delta\bar{A}(t) = \int_{-\infty}^t d\tau \chi_{AB}(t - \tau)\epsilon(\tau), \quad 2.1.$$

with the material response function given by

$$\chi_{AB}(t - \tau) \equiv i \langle [A(t), B(\tau)] \rangle_M. \quad 2.2.$$

Here, $[\dots, \dots]$ denotes a commutator, $O(t) \equiv e^{iH_M t} O e^{-iH_M t}$, and $\langle \dots \rangle_M \equiv \text{Tr}(\dots \rho_M^{\text{eq}})$. Physically, the response function $\chi_{AB}(t)$ is needed only for $t \geq 0$ due to causality (cf. Equation 2.1). Its extension to $t < 0$ is formally made with Equation 2.2 as

$$\chi_{AB}(-t) = -\chi_{BA}(t). \quad 2.3.$$

Obviously, the response function $\chi_{AB}(t)$ for Hermitian operators is real.

We now turn to the correlation function, denoted as

$$\tilde{C}_{AB}(t - \tau) \equiv \langle A(t)B(\tau) \rangle_M. \quad 2.4.$$

Either $\chi_{AB}(t - \tau)$ of Equation 2.2 or $\tilde{C}_{AB}(t - \tau)$ of Equation 2.4 depends only on the duration $t - \tau$. This is a property of the stationary statistics as $[H_M, \rho_M^{\text{eq}}] = 0$. We have also that $\langle A(t) \rangle_M = \langle A \rangle_M$, which does not depend on time, and

$$\langle \dot{A}(t)B(0) \rangle_M = -\langle A(t)\dot{B}(0) \rangle_M. \quad 2.5.$$

Moreover, the correlation function satisfies the following symmetry and detailed-balance relations:

$$\tilde{C}_{AB}^*(t) = \tilde{C}_{BA}(-t) = \tilde{C}_{AB}(t - i\beta). \quad 2.6.$$

Note that $\chi_{AB}(t) = -2\text{Im}\tilde{C}_{AB}(t)$. The common phenomenon of statistical independence as $t \rightarrow \infty$ implies that $\tilde{C}_{AB}(t \rightarrow \infty) = \langle A \rangle_M \langle B \rangle_M$ in a general dissipative

system. One may remove this nonzero asymptotic value by setting $A - \langle A \rangle_M$ and $B - \langle B \rangle_M$ as new variables and consider only the shifted correlation functions that satisfy $\tilde{C}_{AB}(t \rightarrow \infty) = 0$.

2.2. Spectrum and Dispersion Functions Versus Kramers-Kronig Relations

Let us now introduce the so-called causality transformation via

$$\hat{C}_{AB}(\omega) \equiv \int_0^\infty dt e^{i\omega t} \tilde{C}_{AB}(t). \tag{2.7}$$

Here, $\tilde{C}_{AB}(t \rightarrow \infty) = 0$ is implied. The generalized spectrum $C_{AB}(\omega)$ and dispersion $D_{AB}(\omega)$ functions, by which

$$\hat{C}_{AB}(\omega) = C_{AB}(\omega) + iD_{AB}(\omega), \tag{2.8}$$

can then be defined, respectively, as

$$C_{AB}(\omega) \equiv \frac{1}{2}[\hat{C}_{AB}(\omega) + \hat{C}_{BA}^*(\omega)] = \frac{1}{2} \int_{-\infty}^\infty dt e^{i\omega t} \tilde{C}_{AB}(t) = C_{BA}^*(\omega) \text{ and} \tag{2.9a}$$

$$D_{AB}(\omega) \equiv \frac{1}{2i}[\hat{C}_{AB}(\omega) - \hat{C}_{BA}^*(\omega)] = D_{BA}^*(\omega). \tag{2.9b}$$

Thus, Equation 2.8 represents the separation of Hermite and anti-Hermite components rather than that of real and imaginary parts.

An important mathematical property implied in the causality transform Equation 2.7 is that $\hat{C}_{AB}(z)$ is an analytical function in the upper plane ($\text{Im}z > 0$). By using the contour integration formalism, together with the identity $1/(\omega' - \omega) = \mathcal{P}\{1/(\omega' - \omega)\} + i\pi\delta(\omega' - \omega)$, we have

$$\hat{C}_{AB}(\omega) = \frac{i}{\pi} \mathcal{P} \int_{-\infty}^\infty d\omega' \frac{\hat{C}_{AB}(\omega')}{\omega - \omega'}. \tag{2.10}$$

Here, \mathcal{P} denotes the principle part. This is the Kramers-Kronig relation, which can be recast as

$$C_{AB}(\omega) = -\frac{1}{\pi} \mathcal{P} \int_{-\infty}^\infty d\omega' \frac{D_{AB}(\omega')}{\omega - \omega'}, \quad D_{AB}(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^\infty d\omega' \frac{C_{AB}(\omega')}{\omega - \omega'}. \tag{2.11}$$

Similarly, the causality transform of the response function is

$$\hat{\chi}_{AB}(\omega) \equiv \int_0^\infty dt e^{i\omega t} \chi_{AB}(t) = \hat{\chi}_{AB}^{(+)}(\omega) + i\hat{\chi}_{AB}^{(-)}(\omega), \tag{2.12}$$

where $\hat{\chi}_{AB}^{(+)}(\omega) = [\hat{\chi}_{BA}^{(+)}(\omega)]^*$ and $\hat{\chi}_{AB}^{(-)}(\omega) = [\hat{\chi}_{BA}^{(-)}(\omega)]^*$ are the Hermite and anti-Hermite components, respectively. They also satisfy the aforementioned Kramers-Kronig relations.

Using Equations 2.2, 2.7, 2.8 and 2.12, we obtain

$$\hat{\chi}_{AB}^{(+)}(\omega) = -[D_{AB}(\omega) + D_{BA}(-\omega)], \quad 2.13a.$$

$$\hat{\chi}_{AB}^{(-)}(\omega) = C_{AB}(\omega) - C_{BA}(-\omega) = \frac{1}{2i} \int_{-\infty}^{\infty} dt e^{i\omega t} \chi_{AB}(t). \quad 2.13b.$$

It then follows that, $\hat{\chi}_{AB}^{(+)}(-\omega) = \hat{\chi}_{BA}^{(+)}(\omega)$ and $\hat{\chi}_{AB}^{(-)}(-\omega) = -\hat{\chi}_{BA}^{(-)}(\omega)$. As described in Equation 2.13b, $\{\hat{\chi}_{AB}^{(-)}(\omega)\}$ is also termed the spectral density function.

2.3. Fluctuation-Dissipation Theorem

The detailed-balance relation in terms of spectrum functions reads as

$$C_{BA}(-\omega) = e^{-\beta\omega} C_{AB}(\omega). \quad 2.14.$$

Together with the first identity of Equation 2.13b, we have

$$\hat{\chi}_{AB}^{(-)}(\omega) = (1 - e^{-\beta\omega}) C_{AB}(\omega). \quad 2.15.$$

This relation is called the fluctuation-dissipation theorem (FDT). It can be recast as

$$\tilde{C}_{AB}(t) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \frac{e^{-i\omega t} \hat{\chi}_{AB}^{(-)}(\omega)}{1 - e^{-\beta\omega}}. \quad 2.16.$$

It thus also establishes the relation between the correlation function $\tilde{C}_{AB}(t)$ and the response function $\chi_{AB}(t)$ (cf. Equation 2.13b). The FDT is a result of the detailed-balance relation.

In Appendix A, the FDT is applied to formulate the equilibrium phase-space variances, $\sigma_{qq}^{\text{eq}} \equiv \langle q^2 \rangle - \langle q \rangle^2$ and $\sigma_{pp}^{\text{eq}} \equiv \langle p^2 \rangle - \langle p \rangle^2$, in terms of the coordinate response function $\chi_{qq}(t)$ for arbitrary one-dimensional systems. Some additional properties in relation to the spectrum and/or spectral density functions are summarized as follows.

It is easy to show that not just $C_{AA}(\omega) \geq 0$, but also $C_{AA}(\omega)C_{BB}(\omega) \geq |C_{AB}(\omega)|^2$, for all real ω . In fact, the Hermitian matrix $\{C_{AB}(\omega)\}$ of spectrum functions is of complete positivity (60, 98). It leads also to the positivity of spectral density $\{\hat{\chi}_{AB}^{(-)}(\omega)\}$ for $\omega \geq 0$ as inferred from Equation 2.15.

We have also the following expressions,

$$\hat{\chi}_{AB}(0) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \frac{\hat{\chi}_{AB}^{(-)}(\omega)}{\omega}, \quad \dot{\chi}_{AB}(0) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \omega \hat{\chi}_{AB}^{(-)}(\omega). \quad 2.17.$$

The first expression is obtained when the Kramer-Kronig relation is at zero frequency, together with $\hat{\chi}_{AB}^{(+)}(0) = \hat{\chi}_{AB}(0)$ as inferred from Equation 2.15 that $\hat{\chi}_{AB}^{(-)}(0) = 0$. Note that as $\chi_{AB}(t)$ is real for Hermite operators, $\hat{\chi}_{AB}(0)$ must be real (cf. Equation 2.12). The second expression in Equation 2.17 is obtained by taking the time derivative of the inverse Fourier transform of Equation 2.13b, followed by setting $t = 0$.

Later in this review, the bath correlation/response functions for a set of bath operators $\{F_a(t) \equiv e^{iH_B t} F_a e^{-iH_B t}\}$ will be exploited to describe the energy relaxation and decoherence processes in the reduced system of primary interest. The bath correlation functions will be denoted similarly as

$$\tilde{C}_{ab}(t) \equiv \text{tr}_B[F_a(t)F_b(0)\rho_B^{\text{eq}}] \equiv \langle F_a(t)F_b(0) \rangle_B. \quad 2.18.$$

However, the bath response functions $\chi_{ab}(t)$ will be renamed as

$$\phi_{ab}(t) \equiv i\langle [F_a(t), F_b(0)] \rangle_B, \quad 2.19.$$

to avoid possible confusions that may occur there. The bath spectral density functions $\hat{\phi}_{ab}^{(-)}(\omega)$ [or $\hat{\chi}_{ab}^{(-)}(\omega)$] will further be renamed as $J_{ab}(\omega)$. Clearly, all relations presented earlier in this section remain valid for the bath correlation/response functions. For example, the FDT of Equation 2.16 now reads as

$$\tilde{C}_{ab}(t) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \frac{e^{-i\omega t} J_{ab}(\omega)}{1 - e^{-\beta\omega}}. \quad 2.20.$$

The generalized frictional function $\gamma_{ab}(t)$ can also be introduced via

$$\phi_{ab}(t) \equiv -\dot{\gamma}_{ab}(t) \quad \text{or} \quad \hat{\phi}_{ab}(\omega) = \gamma_{ab}(0) + i\omega\hat{\gamma}_{ab}(\omega). \quad 2.21.$$

Note that $\gamma_{ab}(t) = \gamma_{ba}(-t)$. The first expression in Equation 2.17 is now

$$\gamma_{ab}(0) = \hat{\phi}_{ab}(0) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \frac{J_{ab}(\omega)}{\omega}. \quad 2.22.$$

3. EXACT DYNAMICS OF DRIVEN BROWNIAN OSCILLATORS

This section discusses an exactly solvable model, the driven Brownian oscillator (DBO) systems (36, 37, 60, 99, 100). It also highlights some issues on the general QDT formulation.

3.1. Model and Quantum Langevin Equations

Let us start with the total composite Hamiltonian in the presence of external classical field drive, $H_T(t) \equiv H_M + H_{\text{sf}}(t)$. In the DBO model, the system-field interaction assumes $H_{\text{sf}}(t) = -q\epsilon(t)$, and the total field-free system-plus-bath material Hamiltonian assumes the Caldeira-Leggett form (50, 51)

$$H_M = \left(\frac{p^2}{2M} + \frac{1}{2}M\Omega_0^2q^2 \right) + \sum_j \left[\frac{p_j^2}{2m_j} + \frac{1}{2}m_j\omega_j^2 \left(x_j - \frac{c_j}{m_j\omega_j^2}q \right)^2 \right]. \quad 3.1.$$

The last term here contains the free-bath $h_B = \sum_j [p_j^2/(2m_j) + m_j\omega_j^2x_j^2/2]$, the system-bath $\{q \cdot x_j\}$ -coupling, and the coupling-induced renormalization H_{ren} that depends only on the DBO's degree of freedom. Thus,

$$H_M = H_0 + H_{\text{ren}} + h_B - \sqrt{M}qF \equiv H_s + h_B - \sqrt{M}qF. \quad 3.2.$$

The mass-scaled Langevin force, $F = f/\sqrt{M} = \sum_j c_jx_j/\sqrt{M}$, is adopted to satisfy the relation in Equation 2.21, i.e.,

$$\phi(t) \equiv i\langle [F(t), F(0)] \rangle = -\dot{\gamma}(t) \quad \text{or} \quad \hat{\phi}(\omega) = \gamma(0) + i\omega\hat{\gamma}(\omega). \quad 3.3.$$

Here, $F(t) = e^{ih_{\text{B}}t} F e^{-ih_{\text{B}}t}$ and $\gamma(t)$ assumes the classical frictional function,

$$\gamma(t) = \frac{1}{M} \sum_j \frac{c_j^2}{m_j\omega_j^2} \cos(\omega_j t). \quad 3.4.$$

In Equation 3.2, H_0 is given by the first term in Equation 3.1 with frequency Ω_0 , and

$$H_s \equiv H_0 + H_{\text{ren}} = \frac{p^2}{2M} + \frac{1}{2}M\Omega_{\text{H}}^2q^2, \quad 3.5.$$

with

$$\Omega_{\text{H}}^2 \equiv \Omega_0^2 + \frac{1}{M} \sum_j \frac{c_j^2}{m_j\omega_j^2} = \Omega_0^2 + \gamma(0). \quad 3.6.$$

We shall later show that Ω_0 , rather than Ω_{H} , represents the DBO frequency in the Markovian white-noise limit. However, in general the DBO frequency appears as neither Ω_0 nor Ω_{H} .

The quantum Langevin equations for the DBO system can easily be derived via the Heisenberg equations of motion with $H_T(t) = H_M - q\epsilon(t)$. By using the formal solutions of bath degrees of freedom, we obtain

$$\dot{\hat{q}}(t) = \hat{p}(t)/M, \quad 3.7a$$

$$\begin{aligned} \dot{\hat{p}}(t) &= -M\Omega_H^2\hat{q}(t) + M \int_{t_0}^t d\tau \phi(t-\tau)\hat{q}(\tau) + \epsilon(t) + f_B(t) \\ &= -M\Omega_0^2\hat{q}(t) - \int_{t_0}^t d\tau \gamma(t-\tau)\hat{p}(\tau) + \epsilon(t) + f_B(t). \end{aligned} \quad 3.7b$$

Here $f_B(t) \equiv \sqrt{M}F(t)$ is the standard quantum Langevin force operator. The second identity of Equation 3.7b is the conventional form of the Langevin equation, obtained by performing the integration by part, together with Equations 3.3 and 3.6.

3.2. Quantum Master Equation

The formal solution to Equations 3.7 is given by (60)

$$\begin{bmatrix} \hat{q}(t) \\ \hat{p}(t) \end{bmatrix} = \mathbf{T}(t-t_0) \begin{bmatrix} \hat{q}(t_0) \\ \hat{p}(t_0) \end{bmatrix} + \int_{t_0}^t d\tau \mathbf{T}(t-\tau) \begin{bmatrix} 0 \\ \epsilon(\tau) + f_B(\tau) \end{bmatrix}, \quad 3.8$$

with

$$\mathbf{T}(t) = \begin{bmatrix} \chi_{pq}(t) & \chi_{qq}(t) \\ -\chi_{pp}(t) & \chi_{pq}(t) \end{bmatrix} \equiv \begin{bmatrix} M\dot{\chi}(t) & \chi(t) \\ M^2\ddot{\chi}(t) & M\dot{\chi}(t) \end{bmatrix}. \quad 3.9$$

Here, $\chi_{AB}(t)$ (Equation 2.2) denotes the conventional response function in the total composite material H_M -space. In the second identity of Equation 3.9, we made use of Equation 2.5 and $\chi(t) \equiv \chi_{qq}(t)$, which will be specified later in terms of bath response or frictional function (cf. Equations 3.13–3.15).

Equation 3.8 may be used to construct an exact quantum master equation via, for example, the Yan-Mukamel method based on the Gaussian wave packet dynamics in the Wigner phase space (26, 60). However, as pointed out by Karrlein & Grabert (100), there does not exist a generally exact QDT for arbitrary initial bath preparations.

In this review, we shall focus on the reduced dynamics induced by the external field. The natural initial condition to be adopted acquires the thermal equilibrium state for the total composite material system, $\rho_T(t_0) = \rho_M^{\text{eq}}$, before the external field interaction. The initial time can thus be set to $t_0 \rightarrow -\infty$.

The exact quantum master equation for the DBO system is summarized as follows (60):

$$\begin{aligned} \dot{\rho}(t) = & -i \left[\frac{p^2}{2M} + \frac{1}{2} M \tilde{\Omega}^2 q^2 - q \epsilon_{\text{eff}}(t), \rho(t) \right] - \frac{i}{2} \tilde{\gamma} [q, \{p, \rho(t)\}] \\ & - \tilde{\gamma} \sigma_{pp}^{\text{eq}} [q, [q, \rho(t)]] - (\sigma_{pp}^{\text{eq}}/M - M \tilde{\Omega}^2 \sigma_{qq}^{\text{eq}}) [q, [p, \rho(t)]]. \end{aligned} \quad 3.10.$$

Here, $([\dots, \dots])$ denotes a commutator and $\{\dots, \dots\}$ an anticommutator)

$$\tilde{\Omega} \equiv \lim_{t_0 \rightarrow -\infty} \tilde{\Omega}_{t-t_0}, \quad \tilde{\gamma} \equiv \lim_{t_0 \rightarrow -\infty} \tilde{\gamma}_{t-t_0}, \quad 3.11a.$$

with

$$\tilde{\Omega}_t^2 = \frac{\ddot{\chi}^2(t) - \ddot{\chi}(t)\dot{\chi}(t)}{\dot{\chi}^2(t) - \dot{\chi}(t)\chi(t)}, \quad \tilde{\gamma}_t = \frac{\ddot{\chi}(t)\chi(t) - \ddot{\chi}(t)\dot{\chi}(t)}{\dot{\chi}^2(t) - \dot{\chi}(t)\chi(t)}, \quad 3.11b.$$

and

$$\epsilon_{\text{eff}}(t) = \epsilon(t) + \int_{t_0}^t d\tau \chi_\epsilon(t, \tau) \epsilon(\tau), \quad 3.12a.$$

with

$$\chi_\epsilon(t, \tau) \equiv M[\tilde{\Omega}^2 \chi(t - \tau) + \tilde{\gamma} \dot{\chi}(t - \tau) + \ddot{\chi}(t - \tau)]. \quad 3.12b.$$

The thermal equilibrium phase-space variances, σ_{qq}^{eq} and σ_{pp}^{eq} , involved in Equation 3.10 are given by Equation A.1 or A.3 in terms of the causality transform of $\chi(t)$.

In deriving Equation 3.10, the determinant $|\mathbf{T}(t)|$ of the transfer matrix in Equation 3.9 is assumed nonzero; otherwise both $\tilde{\Omega}_t$ and $\tilde{\gamma}_t$ diverge (Equations 3.11). Equation 3.10 recovers the well-established result (100) by setting $\epsilon(t) = 0$ and replacing $\tilde{\Omega}$ and $\tilde{\gamma}$ with $\tilde{\Omega}_t$ and $\tilde{\gamma}_t$ as the initial time of $t_0 = 0$ was adopted in their work.

The key quantity here is the well-established Brownian response function $\chi(t) \equiv \chi_{qq}(t)$. With the linear response theory being applied for Equations 3.7, one can readily obtain that (cf. Equations 3.3 and 3.6)

$$\ddot{\chi}(t) + \Omega_{\text{H}}^2 \chi(t) - \int_0^t d\tau \phi(t - \tau) \chi(\tau) = 0, \quad 3.13a.$$

or equivalently

$$\ddot{\chi}(t) + \Omega_0^2 \chi(t) + \int_0^t d\tau \gamma(t - \tau) \dot{\chi}(\tau) = 0. \quad 3.13b.$$

From its definition, Equation 2.5, and Equation 3.13, we have

$$\chi(0) = \dot{\chi}(0) = 0, \quad \dot{\chi}(0) = -\ddot{\chi}(0)/\Omega_{\text{H}}^2 = 1/M. \quad 3.14.$$

Therefore (cf. Equation 3.3)

$$\hat{\chi}(\omega) = \frac{1/M}{\Omega_H^2 - \omega^2 - \hat{\phi}(\omega)} = \frac{1/M}{\Omega_0^2 - \omega^2 - i\omega\hat{\gamma}(\omega)}. \quad 3.15.$$

3.3. Comments

The correlated driving-dissipation effect on the reduced DBO dynamics is described by the second term in the right-hand side of Equation 3.12a as an effective field correction. This correlated effect is completely coherent in the present case. In a general anharmonic system, the correlated driving and dissipation effects are usually quite complicated. We shall come back to this point below.

In the Markovian white-noise limit, $\gamma(t) = 2\gamma_{\text{mar}}\delta(t)$ or $\hat{\gamma}(\omega) = \gamma_{\text{mar}}$ is a constant. In this limit, Equations 3.15, 3.11 and 3.12 reduce, respectively, to

$$\chi(t) \rightarrow \frac{\sin[(\Omega_0^2 - \gamma_{\text{mar}}^2/4)^{\frac{1}{2}} t]}{M(\Omega_0^2 - \gamma_{\text{mar}}^2/4)^{\frac{1}{2}}} e^{-\gamma_{\text{mar}}t/2}, \quad 3.16a.$$

$$\tilde{\Omega}_t \rightarrow \Omega_0, \quad \tilde{\gamma}_t \rightarrow \gamma_{\text{mar}}, \quad 3.16b.$$

$$\chi_\epsilon(t, \tau) = 0, \quad \epsilon_{\text{eff}}(t) = \epsilon(t). \quad 3.16c.$$

The effects of the driving and dissipation correlation, due to $\delta\epsilon_{\text{eff}}(t) \equiv \epsilon_{\text{eff}}(t) - \epsilon(t)$ in the DBO system, vanish completely in the Markovian limit. The $\delta(t)$ -noise is ill-defined for short time; it causes σ_{pp}^{eq} to diverge (Equation A.3). However, analysis of the white-noise limit is instructive; it implies that H_0 would rather be the choice of the reduced system Hamiltonian if a phenomenological description of dissipation were to be adopted. For example, σ_{qq}^{eq} and σ_{pp}^{eq} may be replaced with their zeroth-order values σ_{qq}^0 and σ_{pp}^0 in the pure- H_0 system, and approximate Equation 3.10 as (55)

$$\dot{\rho}(t) \rightarrow -i[H_0 - q\epsilon(t), \rho(t)] - \frac{i}{2}\tilde{\gamma}[q, \{p, \rho(t)\}] - \tilde{\gamma}\sigma_{pp}^0[q, [q, \rho(t)]]. \quad 3.17.$$

The last term in Equation 3.10 becomes zero in this phenomenological description.

Note that $\tilde{\Omega}$ and $\tilde{\gamma}$ serve as the frequency and friction constants entering into Equation 3.10, and they are the long-time asymptotic values of $\tilde{\Omega}_t$ and $\tilde{\gamma}_t$ as described by Equations 3.11. Clearly, $\tilde{\Omega}_{t=0} = \Omega_H$ and $\tilde{\gamma}_{t=0} = 0$ as inferred from Equation 3.14. In strong non-Markovian interaction regime, $\tilde{\Omega}_t$ ($\tilde{\Omega}$) and $\tilde{\gamma}_t$ ($\tilde{\gamma}$) may diverge. One may argue that $\tilde{\Omega}$ and $\tilde{\gamma}$ could be treated phenomenologically as the Markovian parameters in Equation 3.10. However, the non-Markovian nature of Equation 3.10 still remains as $\epsilon_{\text{eff}}(t) \neq \epsilon(t)$. As mentioned earlier, this inequality characterizes the correlated driving-dissipation effects in the present case. We shall see later that for a general system the field-free dissipation could be characterized by a Markovian-like, time-independent dissipative superoperator, as the system was initially in the thermal equilibrium state before the external field

excitation. The dynamical non-Markovian effects enter only through the field-dressed dissipative superoperator component.

4. TWO PRESCRIPTIONS OF QUANTUM DISSIPATION THEORY

4.1. General Description of Total Composite Hamiltonian

The total composite Hamiltonian in the presence of classical external field can be written as

$$H_T = H_M + H_{\text{sf}}(t) \equiv H_s + H_{\text{sf}}(t) + h_B - \sum_a Q_a F_a. \quad 4.1.$$

The last term in Equation 4.1 describes the system-bath couplings, in which $\{Q_a\}$ are Hermite operators of the primary system and can be called the generalized dissipative modes. The generalized Langevin forces $\{F_a(t) \equiv e^{ih_B t} F_a e^{-ih_B t}\}$ are Hermite bath operators in the stochastic bath subspace assuming Gaussian statistics. Without loss of generality, their stochastic mean values are set to $\langle F_a(t) \rangle_B = 0$. The effects of Langevin forces on the reduced primary system are therefore completely characterized by their correlation functions $\tilde{C}_{ab}(t) = \langle F_a(t) F_b(0) \rangle_B$ (Equation 2.18), or other equivalent properties, such as the generalized frictional functions $\gamma_{ab}(t)$, described in Section 2. For the later construction of coupled differential equations of motion, we adopt the extended Meier-Tannor parameterization model that, as detailed in Appendix B, leads the bath correlation functions to the following form (57, 60, 61):

$$\tilde{C}_{ab}(t \geq 0) = \sum_{m=0}^{\tilde{m}} \nu_m^{ab} t^{\delta_{m0}} e^{-\zeta_m^{ab} t}; \quad \text{with } \zeta_0^{ab} \equiv \zeta_1^{ab}. \quad 4.2.$$

In Equation 4.1, the reduced system Hamiltonian in the presence of external classical field is

$$H(t) \equiv H_s + H_{\text{sf}}(t) \equiv H_0 + H_{\text{ren}} + H_{\text{sf}}(t). \quad 4.3.$$

Here, H_s is the time-independent, field-free Hamiltonian, whereas $H_{\text{sf}}(t)$ is the interaction between the system and the external classical field $\epsilon(t)$. The Caldeira-Leggett form of renormalization Hamiltonian assumes $H_{\text{ren}} = \frac{1}{2} \sum \gamma_{ab}(0) Q_a Q_b$ (cf. Equation 3.6). As discussed in Section 3, it is H_0 , rather than H_s , that resembles the effective system Hamiltonian to be observed in the high temperature or Markovian limit. The result is that H_0 is adopted as the reduced field-free system Hamiltonian in some phenomenological QDT formulations that also assume the thermal equilibrium reduced state of $\rho_{\text{eq}} \propto e^{-\beta H_0}$. Further discussion on the issue will be made in the last paragraph of Section 4.2.

The theory presented below goes beyond the phenomenological level, where H_s enters as the reduced field-free system Hamiltonian. Both the reduced dynamics

and the reduced thermal canonical state should be evaluated with dissipation. For later use, let us denote the following Liouvillians,

$$\mathcal{L}(t)O \equiv [H(t), O], \quad \mathcal{L}_s O \equiv [H_s, O], \quad \mathcal{L}_{\text{sf}}(t)O \equiv [H_{\text{sf}}(t), O]. \quad 4.4.$$

The coherent propagator $\mathcal{G}(t, \tau)$ associating with $\mathcal{L}(t)$ is defined via

$$\frac{\partial}{\partial t} \mathcal{G}(t, \tau) \equiv -i\mathcal{L}(t)\mathcal{G}(t, \tau). \quad 4.5.$$

It is equivalent to $\mathcal{G}(t, \tau)O = G(t, \tau)OG^\dagger(t, \tau)$, where $G(t, \tau)$ is the Hilbert-space propagator with $\partial_t G(t, \tau) = -iH(t)G(t, \tau)$. The field-free counterpart is denoted as $\mathcal{G}_s(t, \tau)$, which is given by

$$\mathcal{G}_s(t, \tau) = e^{-i\mathcal{L}_s(t-\tau)} \equiv \mathcal{G}_s(t - \tau). \quad 4.6.$$

4.2. Perturbative Formulations in Two Prescriptions

There are two commonly used prescriptions of QDT. One is characterized by a memory dissipation kernel $\Upsilon(t, \tau)$ and reads as follows (39–42):

$$\dot{\rho}(t) = -i\mathcal{L}(t)\rho(t) - \int_{-\infty}^t d\tau \Upsilon(t, \tau)\rho(\tau). \quad 4.7.$$

According to the temporal sequence of the involving actions in $\Upsilon(t, \tau)\rho(\tau)$, Equation 4.7 is also said to be in the chronological ordering prescription (COP). An alternative prescription of the QDT is characterized by a time-local dissipation kernel $\mathcal{R}(t)$ and reads (43)

$$\dot{\rho}(t) = -i\mathcal{L}(t)\rho(t) - \mathcal{R}(t)\rho(t). \quad 4.8.$$

According to the temporal sequence of the involving actions in $\mathcal{R}(t)\rho(t)$, Equation 4.8 is also said to be in a partial ordering prescription (POP) in contrast to Equation 4.7. In principle, both $\Upsilon(t, \tau)$ and $\mathcal{R}(t)$ can be formulated exactly by using, for example, the Nakajima-Zwanzig-Mori projection operator techniques (39–43). In this sense, Equation 4.7 and Equation 4.8 are equivalent. However, the exact QDT in the forms of differential equations of motion (EOM) are by far numerically tractable in very few systems, such as the DBO system (described in Section 3) and the spin-boson system (89, 101, 102). In most cases, certain approximation schemes are employed.

Let us consider the weak system-bath interaction regime and focus on a so-called complete second-order quantum dissipation theory (CS-QDT). Here, the system-bath couplings are rigorously accounted for to second order, not only for the dynamics of $\rho(t)$, but also for the initial reduced canonical state $\rho(t_0) = \text{tr}_B \rho_M^{\text{eq}}(T)$; this includes the nonfactorizable $\rho_M^{\text{eq}}(T)$, before external field excitation. Various forms of CS-QDT are the same at the second order system-bath interaction level, but differ at their partial resummation schemes in approximating higher order

contributions. In this review, we shall discuss three nonequivalent forms of CS-QDT. Two of them resemble Equations 4.7 and 4.8, and will be presented later in this section as the COP-CS-QDT and the POP-CS-QDT, respectively. The third one is nonconventional and will be discussed in detail in the next section.

A perturbative QDT can be formulated by some relatively simple methods without explicitly invoking the Nakajima-Zwanzig-Mori projection operator techniques (4–8, 38, 52–57, 59). By using explicitly the decompose form of system-bath couplings as shown in the last term of Equation 4.1, the COP-CS-QDT of Equation 4.7 can be readily obtained, where (57, 59)

$$\int_{-\infty}^t d\tau \Upsilon(t, \tau) \rho(\tau) = \sum_a \{ [Q_a, \tilde{Q}_a^{\text{cop}}(t)] + \text{H.c.} \}, \quad 4.9a.$$

with

$$\tilde{Q}_a^{\text{cop}}(t) = \sum_b \int_{-\infty}^t d\tau \tilde{C}_{ab}(t - \tau) \mathcal{G}(t, \tau) [Q_b \rho(\tau)], \quad 4.9b.$$

and the POP-CS-QDT of Equation 4.8, where (23, 59)

$$\mathcal{R}(t) \rho(t) = \sum_a \{ [Q_a, \tilde{Q}_a^{\text{pop}}(t) \rho(t)] + \text{H.c.} \}, \quad 4.10a.$$

with

$$\tilde{Q}_a^{\text{pop}}(t) = \sum_b \int_{-\infty}^t d\tau \tilde{C}_{ab}(t - \tau) \mathcal{G}(t, \tau) Q_b. \quad 4.10b.$$

To investigate the correlated driving-dissipation effects involved here, we make use of the following identity,

$$\mathcal{G}(t, \tau) = \mathcal{G}_s(t, \tau) - i \int_{\tau}^t d\tau' \mathcal{G}(t, \tau') \mathcal{L}_{\text{sf}}(\tau') \mathcal{G}_s(\tau', \tau). \quad 4.11.$$

In particular, we can recast Equation 4.10b as

$$\tilde{Q}_a^{\text{pop}}(t) = \tilde{Q}_a - i \sum_b \int_{-\infty}^t d\tau \int_{\tau}^t d\tau' \tilde{C}_{ab}(t - \tau) \mathcal{G}(t, \tau') \mathcal{L}_{\text{sf}}(\tau') \mathcal{G}_s(\tau' - \tau) Q_b, \quad 4.12.$$

with

$$\tilde{Q}_a \equiv \sum_b \int_{-\infty}^t d\tau \tilde{C}_{ab}(t - \tau) e^{-i\mathcal{L}_s(t-\tau)} Q_b = \sum_b \hat{C}_{ab}(-\mathcal{L}_s) Q_b. \quad 4.13.$$

Here, $\hat{C}_{ab}(-\mathcal{L}_s)$ is a reduced Liouville-space operator, defined by the field-free Liouvillian \mathcal{L}_s and the causality spectrum $\hat{C}_{ab}(\omega)$ of the bath correlation function. The field-free dissipation in the POP-CS-QDT is therefore characterized by the time-independent \mathcal{R}_s , with (cf. Equation 4.10a)

$$\mathcal{R}_s \rho(t) \equiv \sum_a [Q_a, \tilde{Q}_a \rho(t) - \rho(t) \tilde{Q}_a^\dagger]. \quad 4.14.$$

Similarly, the field-free dissipation in the COP-CS-QDT is characterized by the memory kernel of $\Upsilon_s(t - \tau)$, which is defined similarly as Equations 4.9 but with the $\mathcal{G}(t, \tau)$ in Equation 4.9b replaced by $\mathcal{G}_s(t, \tau) = e^{-i\mathcal{L}_s \cdot (t-\tau)}$.

To conclude this subsection, let us make some comments on the non-Markovian nature of a QDT. Traditionally, a QDT characterized by a time-independent \mathcal{R}_s would be classified as Markovian, whereas a QDT characterized by a memory kernel $\Upsilon_s(t - \tau)$ would be classified as non-Markovian. However, \mathcal{R}_s and $\Upsilon_s(t - \tau)$ are equivalent at the field-free CS-QDT level and both of them can describe colored noises. In the white-noise limit, the field-free COP assumes

$$\int_{-\infty}^t d\tau \Upsilon_s(t - \tau) \rho(\tau) \rightarrow \mathcal{R}_{\text{mar}} \rho(t), \quad 4.15.$$

where

$$\mathcal{R}_{\text{mar}} \rho(t) \equiv \sum_{a,b} [Q_a, \hat{C}_{ab}(-\mathcal{L}_s) \{Q_b \rho(t)\}] + \text{H.c.} \quad 4.16.$$

Clearly, $\mathcal{R}_s \neq \mathcal{R}_{\text{mar}}$. This implies that classifying the \mathcal{R}_s -based CS-QDT as Markovian is due to the lack of driving-dissipation cooperativity rather than to the nature of bath. In fact, non-Markovian nature enters into the reduced equilibrium density operator $\rho_{\text{eq}}^{\text{pop}}$ of the \mathcal{R}_s -based CS-QDT, but does not enter into its $\Upsilon_s(t - \tau)$ -based counterpart $\rho_{\text{eq}}^{\text{cop}}$ (cf. Section 4.4).

It is worth mentioning here that in some phenomenological quantum master equations, the reduced equilibrium state is set to be independent of dissipation, together with the neglect of correlated driving and dissipation effects. In this case, the phenomenological QDT would read (55)

$$\dot{\rho}(t) \rightarrow -i[H_0 + H_{\text{st}}(t), \rho(t)] - \sum_{a,b} \{[Q_a, \{C_{ab}(-\mathcal{L}_0) Q_b\} \rho(t)] + \text{H.c.}\}. \quad 4.17.$$

Here, $H_0 = H_s - H_{\text{ren}}$ was given in Equation 4.3, and only the bath spectrum functions $\{C_{ab}(\omega)\}$ are involved. The effects of bath dispersions $\{D_{ab}(\omega)\}$ are phenomenologically incorporated into the coupling-induced renormalization contribution to the reduced system Hamiltonian. It can easily show that Equation 4.17 assumes the equilibrium state of $\rho_{\text{eq}}^0 \propto e^{-\beta H_0}$ (55). Equation 4.17 recovers Equation 3.17 in the DBO system. As discussed in Section 3.3, the phenomenological QDT (Equation 4.17) does amount to a Markovian formulation (cf. Appendix D of Reference 59).

4.3. Differential Equations of Motion

With the model bath of Equation 4.2, the COP-CS-QDT and the POP-CS-QDT presented in Section 4.2 can further be expressed in terms of closed sets of differential EOM. Let us start with the COP-CS-QDT (Equations 4.7 and 4.9). We have

$$\tilde{Q}_a^{\text{cop}}(t) \equiv \sum_{m=0}^{\tilde{m}} \sum_b v_m^{ab} K_{m,ab}^{\text{cop}}(t), \quad 4.18.$$

where

$$K_{m,ab}^{\text{cop}}(t) \equiv \int_{-\infty}^t d\tau (t - \tau)^{\delta_{m0}} e^{-\zeta_m^{ab}(t-\tau)} \mathcal{G}(t, \tau) [Q_b \rho(\tau)]. \quad 4.19.$$

Note that $\zeta_0^{ab} = \zeta_1^{ab}$. The final EOM for COP-CS-QDT read (60)

$$\dot{\rho}(t) = -i\mathcal{L}(t)\rho(t) - \sum_{m,a,b} \{v_m^{ab} [Q_a, K_{m,ab}^{\text{cop}}(t)] + \text{H.c.}\}, \quad 4.20a.$$

$$\dot{K}_{m,ab}^{\text{cop}}(t) = \delta_{m0} K_{1,ab}^{\text{cop}}(t) + (1 - \delta_{m0}) Q_b \rho(t) - [i\mathcal{L}(t) + \zeta_m^{ab}] K_{m,ab}^{\text{cop}}(t). \quad 4.20b.$$

The natural initial conditions to the above coupled set of EOM are

$$\rho(t_0) = \rho_{\text{eq}}^{\text{cop}} \text{ and } K_{m,ab}^{\text{cop}}(t_0) = (i\mathcal{L}_s + \zeta_m^{ab})^{-(\delta_{m0}+1)} (Q_b \rho_{\text{eq}}^{\text{cop}}). \quad 4.21.$$

Here, t_0 can be any time before the external field acts. The COP reduced thermal equilibrium density operator $\rho_{\text{eq}}^{\text{cop}}$ will be discussed in Section 4.4.

We now turn to the POP-CS-QDT (Equations 4.8 and 4.10). Let Equation 4.12 be recast as

$$\tilde{Q}_a^{\text{pop}}(t) \equiv \tilde{Q}_a + \delta \tilde{Q}_a^{\text{pop}}(t). \quad 4.22.$$

Here, $\delta \tilde{Q}_a^{\text{pop}}(t)$ is given by the second term of Equation 4.12, which together with Equation 4.2 can be expressed as

$$\delta \tilde{Q}_a^{\text{pop}}(t) = \sum_{m=0}^{\tilde{m}} \sum_b v_m^{ab} K_{m,ab}^{\text{pop}}(t), \quad 4.23.$$

with (noting that $\zeta_0^{ab} = \zeta_1^{ab}$)

$$\begin{aligned} K_{m,ab}^{\text{pop}}(t) &= -i \int_{-\infty}^t d\tau \int_{\tau}^t d\tau' (t - \tau)^{\delta_{m0}} e^{-\zeta_m^{ab}(t-\tau)} \\ &\quad \times \mathcal{G}(t, \tau') \mathcal{L}_{\text{sf}}(\tau') \mathcal{G}_s(\tau' - \tau) Q_b. \end{aligned} \quad 4.24.$$

The final EOM for POP-CS-QDT therefore read (60)

$$\dot{\rho}(t) = -i\mathcal{L}(t)\rho(t) - \mathcal{R}_s\rho(t) - \sum_{m,a,b} \{v_m^{ab} [Q_a, K_{m,ab}^{\text{POP}}(t)\rho(t)] + \text{H.c.}\}, \quad 4.25a.$$

$$\dot{K}_{m,ab}^{\text{POP}}(t) = \delta_{m0}K_{1,ab}^{\text{POP}}(t) - [i\mathcal{L}(t) + \zeta_m^{ab}]K_{m,ab}^{\text{POP}}(t) - i[H_{\text{st}}(t), \hat{Q}_m^{ab}]. \quad 4.25b.$$

Here \mathcal{R}_s was given by Equation 4.14, and the ordinary operator $\hat{Q}_m^{ab}(t)$ in the inhomogeneous term in Equation 4.25b is

$$\hat{Q}_m^{ab} \equiv (i\mathcal{L}_s + \zeta_m^{ab})^{-(\delta_{m0}+1)} Q_b. \quad 4.26.$$

Note that the EOM for $K_{m,ab}^{\text{POP}}(t)$ is independent, except for $K_{0,ab}^{\text{POP}}(t)$ that couples also to $K_{1,ab}^{\text{POP}}(t)$. The initial conditions to Equations 4.25 are $\rho(t_0) = \rho_{\text{eq}}^{\text{POP}}$, which will be discussed later, and $K_{m,ab}^{\text{POP}}(t_0) = 0$ for $m \geq 0$.

4.4. Evaluation of Reduced Canonical States

The reduced canonical equilibrium density operator applies for both the initial and the asymptotic states before and after the external field excitation. It can be evaluated via the field-free propagation for long time as $\rho_{\text{eq}} = \rho(t \rightarrow \pm\infty)$. Alternatively, ρ_{eq} can be evaluated via the nondynamical approach by considering the stationary condition for the field-free version of QDT, which in the POP-CS-QDT assumes

$$\dot{\rho}_{\text{eq}}^{\text{POP}} = -(i\mathcal{L}_s + \mathcal{R}_s)\rho_{\text{eq}}^{\text{POP}} = 0. \quad 4.27.$$

The details of evaluation of $\rho_{\text{eq}}^{\text{POP}}$ are presented in Appendix C. Similarly, we have (cf. Equations 4.7 and 4.9)

$$\dot{\rho}_{\text{eq}}^{\text{COP}} = -(i\mathcal{L}_s + \mathcal{R}_{\text{mar}})\rho_{\text{eq}}^{\text{COP}} = 0. \quad 4.28.$$

Here, \mathcal{R}_{mar} describes the white-noise Markovian limit of dissipation (Equation 4.16). Therefore, $\rho_{\text{eq}}^{\text{COP}}$ is identical to its Markovian counterpart! It is in contrast with $\rho_{\text{eq}}^{\text{POP}}$ in Equation 4.27, and implies that the time-independent \mathcal{R}_s does physically describe certain non-Markovian effects at least on the equilibrium properties.

To demonstrate the points raised above, let us consider the DBO system and compare $\rho_{\text{eq}}^{\text{POP}}$ and $\rho_{\text{eq}}^{\text{COP}}$ with respect to the exact $\rho_{\text{eq}}^{\text{ex}}$. Depicted in Figure 1 are the results of the reduced phase-space variances σ_{qq} and σ_{pp} as functions of temperature. Note that both $\rho_{\text{eq}}^{\text{ex}}$ and $\rho_{\text{eq}}^{\text{POP}}$ are Gaussian wave packets in the reduced phase space (cf. Appendix D); thus they are completely described by the variances shown in Figure 1 as their $\langle q \rangle_{\text{eq}}$, $\langle p \rangle_{\text{eq}}$, and $\langle pq + qp \rangle_{\text{eq}}$ are all zero values. Appendix D also explicitly shows that both the exact DBO propagator and its POP-CS-QDT counterpart are Gaussian and preserve positivity. The COP-CS-QDT does not have these properties. The non-Gaussian $\rho_{\text{eq}}^{\text{COP}}(T)$ starts to violate the positivity even at a moderately low temperature, which is $k_B T / \Omega_0 \approx 0.7$ (checked against the uncertainty principle) in the case under study in Figure 1.

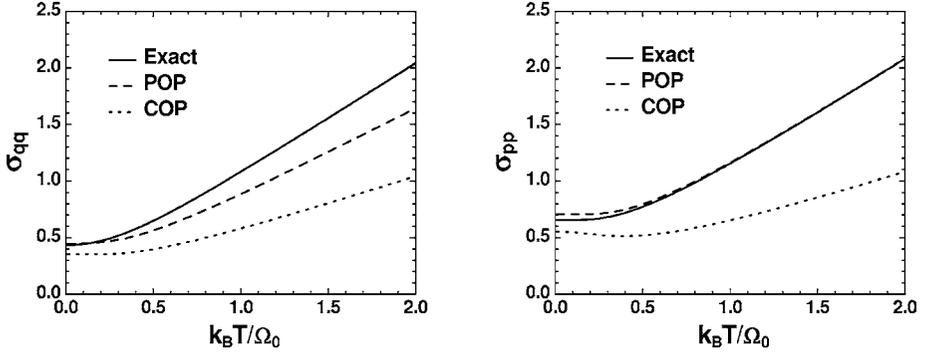


Figure 1 Equilibrium phase-space variances, σ_{qq} and σ_{pp} , depicted in the dimensionless unit for the Brownian oscillator of mass M and frequency Ω_0 , as functions of temperature. Shown are results from the exact formulation (Equation A.1 or Equation A.3), the POP-CS-QDT (Equations D.4), and the COP-CS-QDT (Equations D.5), with the Drude model of frictional function $\gamma(t) = \gamma_0 \omega_D e^{-\omega_D t}$, where $\gamma_0 = \omega_D = \Omega_0$ is used. Note that the exact results are of $(M\Omega_0) \sigma_{qq}^{\text{ex}}(T \rightarrow \infty) = \sigma_{pp}^{\text{ex}}(T \rightarrow \infty)/(M\Omega_0) = \bar{n}(\Omega_0) + \frac{1}{2}$, with $\bar{n}(\omega) = 1/(e^{\beta\hbar\omega} - 1)$, approaching their friction-free counterparts (not shown) in the high temperature regime.

To conclude, let us make some remarks on the two forms of CS-QDT presented in this section. Usually, POP-CS-QDT is superior to COP-CS-QDT owing to its significantly wider applicability range of parameters for system-bath interactions, non-Markovian nature, and temperature in all the systems we have studied, either harmonic or anharmonic (60, 61). For the DBO system where the exact solution is available, the POP-CS-QDT is always physically well behaved, whereas the COP-CS-QDT often leads to unphysical results, especially for its long-time dynamics, including the equilibrium properties. On the other hand, the POP-CS-QDT is composed of semicoupled nonlinear EOM (Equations 4.25), whereas the COP-CS-QDT is composed of a set of coupled linear EOM (Equations 4.20). In contrast with the $\{K_{m,ab}^{\text{cop}}(t)\}$ (Equation 4.20b), which are auxiliary state operators, the POP-CS-QDT auxiliary operators $\{K_{m,ab}^{\text{pop}}(t)\}$ (Equations 4.25) are dynamical variables. As a result, the evaluation of $\{K_{m,ab}^{\text{pop}}(t)\}$, which are often unbounded, with finite numerical grid may cause certain numerical problems. The main drawback of POP-CS-QDT is its nonlinearity [cf. the $K_{m,ab}^{\text{pop}}(t)\rho(t)$ terms in Equations 4.25], which makes it complicated to use, for example, in optical response and optimal control problems.

5. AN ALTERNATIVE FORMULATION—THE CODDE

We are now in a position to describe an alternative CS-QDT formulation (59–61), which will be termed correlated driving-dissipation equations (CODDE). Although it retains the aforementioned merits of POP-CS-QDT, the CODDE are composed

of a set of coupled linear equations of motion, which is convenient and versatile for applications (cf. Section 7). Thus, the CODDE constitutes the formulation of choice among the three nonequivalent CS-QDT.

The CODDE formulation results as a variation of POP-CS-QDT, but it fixes the drawback of nonlinearity that arises from the field-dressed dissipation and terms it as (59)

$$\begin{aligned}
 & \delta \tilde{Q}_a^{\text{POP}}(t)\rho(t) \\
 &= -i \sum_b \int_{-\infty}^t d\tau \int_{\tau}^t d\tau' \tilde{C}_{ab}(t-\tau) [\mathcal{G}(t, \tau') \mathcal{L}_{\text{sf}}(\tau') \mathcal{G}_s(\tau' - \tau) Q_b] \rho(t) \\
 &\approx -i \sum_b \int_{-\infty}^t d\tau \int_{\tau}^t d\tau' \tilde{C}_{ab}(t-\tau) [\mathcal{G}(t, \tau') \mathcal{L}_{\text{sf}}(\tau') \mathcal{G}_s(\tau' - \tau) Q_b] [\mathcal{G}(t, \tau') \rho(\tau')] \\
 &= -i \sum_b \int_{-\infty}^t d\tau' \int_{-\infty}^{\tau'} d\tau \tilde{C}_{ab}(t-\tau) \mathcal{G}(t, \tau') \{[\mathcal{L}_{\text{sf}}(\tau') \mathcal{G}_s(\tau' - \tau) Q_b] \rho(\tau')\} \\
 &\equiv \tilde{Q}_a^{\text{codde}}(t). \tag{5.1}
 \end{aligned}$$

The approximation above leads the POP-CS-QDT to a new form of CS-QDT, i.e.,

$$\dot{\rho}(t) = -i \mathcal{L}(t)\rho(t) - \mathcal{R}_s \rho(t) - \sum_a \{ [Q_a, \tilde{Q}_a^{\text{codde}}(t)] + \text{H.c.} \}. \tag{5.2}$$

This constitutes the integro-differential form of CODDE, which is of the same field-free dissipation \mathcal{R}_s as the POP-CS-QDT, but now the field-dressed dissipation is effectively described by a partially ordered memory kernel. Upon substituting the parameterized $\tilde{C}_{ab}(t)$ (Equation 4.2) and

$$\tilde{Q}_a^{\text{codde}}(t) \equiv \sum_{m=0}^{\bar{m}} \sum_b v_m^{ab} \rho_m^{ab}(t), \tag{5.3}$$

we obtain (59–61)

$$\dot{\rho}(t) = -i \mathcal{L}(t)\rho(t) - \mathcal{R}_s \rho(t) - \sum_{m,a,b} \{ v_m^{ab} [Q_a, \rho_m^{ab}(t)] + \text{H.c.} \}, \tag{5.4a}$$

$$\dot{\rho}_m^{ab}(t) = \delta_{m0} \rho_1^{ab}(t) - [i \mathcal{L}(t) + \zeta_m^{ab}] \rho_m^{ab}(t) - i [H_{\text{sf}}(t), \hat{Q}_m^{ab}] \rho(t). \tag{5.4b}$$

Here, \mathcal{R}_s and \hat{Q}_m^{ab} were given by Equations 4.14 and 4.26, respectively.

The CODDE formulation of CS-QDT (Equations 5.4) couples between $\rho(t)$ and a set of auxiliary state operators $\{\rho_m^{ab}(t); 0 \leq m \leq \bar{m}\}$ that describe the correlated driving and dissipation. The action of field-free dissipation \mathcal{R}_s (Equation

4.14) can be evaluated in terms of the causality spectral function $\hat{C}_{ab}(\omega)$ without invoking the parameterization form of Equation 4.2, which is required only for the correlated driving-dissipation effects described by the auxiliary operators.

The natural initial conditions for the CODDE (Equations 5.4) are

$$\rho(t_0) = \rho_{\text{eq}}(T) = \rho_{\text{eq}}^{\text{pop}} \quad \text{and} \quad \rho_m^{ab}(t_0) = 0; \quad m = 0, 1, \dots, \bar{m}. \quad 5.5.$$

Here, t_0 is chosen at any moment before the external field excitation. The above initial conditions also serve as the stationary state solution to Equations 5.4 if the external field contains no continuing-wave component. The CODDE and POP-CS-QDT share the same thermal equilibrium reduced density operator (Equation 4.27), i.e., $(i\mathcal{L}_s + \mathcal{R}_s)\rho_{\text{eq}}(T) = 0$, together with the normalization condition as described in Appendix C. The CODDE (Equations 5.4) is also applicable to other initial conditions that will be illustrated in the coming sections.

6. QUANTUM MECHANICS BASED ON THE CODDE FORMULATION

To illustrate the Liouville-space algebra in relation to the CODDE dynamics (Equations 5.4), it is sufficient to consider the single-dissipative-mode case in which the system-bath coupling contains only one term, $-QF(t)$. The bath correlation function is parameterized in the form of $\tilde{C}(t \geq 0) = \sum v_m t^{\delta_{m0}} \exp(-\zeta_m t)$ (cf. Equation 4.2) for its field-dressed dissipation dynamics. The multiple-dissipation-mode indexes a and b are omitted hereafter.

6.1. Schrödinger Picture

Let $\sigma(t)$ be an arbitrary reduced state operator, which can be non-Hermite, and $\{\sigma_m^{(\pm)}; m = 0, \dots, \bar{m}\}$ be the auxiliary operators for correlated driving-dissipation effects. The CODDE (Equations 5.4) now reads

$$\dot{\sigma}(t) = -[i\mathcal{L}(t) + \mathcal{R}_s]\sigma(t) - \sum_{m=0}^{\bar{m}} [Q, v_m \sigma_m^{(-)}(t) - v_m^* \sigma_m^{(+)}(t)], \quad 6.1a.$$

$$\dot{\sigma}_m^{(-)}(t) = \delta_{m0} \sigma_1^{(-)}(t) - [i\mathcal{L}(t) + \zeta_m] \sigma_m^{(-)}(t) - i[H_{\text{sf}}(t), \hat{Q}_m] \sigma(t), \quad 6.1b.$$

$$\dot{\sigma}_m^{(+)}(t) = \delta_{m0} \sigma_1^{(+)}(t) - [i\mathcal{L}(t) + \zeta_m^*] \sigma_m^{(+)}(t) - i\sigma(t)[H_{\text{sf}}(t), \hat{Q}_m^\dagger]. \quad 6.1c.$$

For the normal case where $H_{\text{sf}}(t)$ and $\sigma(t)$ are Hermite, and $[\sigma_m^{(+)}(t)]^\dagger = \sigma_m^{(-)}(t)$, Equations 6.1 become equivalent to Equations 5.4. In fact, they share the same CODDE dynamic generator and propagator. In general, the initial time t_0 and initial values for Equations 6.1 are to be specified depending on applications.

In Equations 6.1, $\hat{Q}_m = (i\mathcal{L}_s + \zeta_m)^{-(\delta_{m0} + 1)} Q$ was defined in Equation 4.26, while the Liouville-space operators, $\mathcal{L}(t)$ and \mathcal{R}_s were given respectively by

Equations 4.4 and 4.14 for their left-actions on an arbitrary operator, i.e.,

$$\mathcal{L}(t)A \equiv [H(t), A], \quad \mathcal{R}_s A \equiv [Q, \tilde{Q}A - A\tilde{Q}^\dagger]. \quad 6.2$$

For later use, we shall also define the right-action of a superoperator \mathcal{O} via the identity of $\text{Tr}[(A\mathcal{O})B] = \text{Tr}[A(\mathcal{O}B)]$. The right-actions equivalent to Equation 6.2 are therefore

$$A\mathcal{L}(t) = [A, H(t)], \quad A\mathcal{R}_s = [A, Q]\tilde{Q} - \tilde{Q}^\dagger[A, Q]. \quad 6.3$$

Clearly, the CODDE dynamics (Equations 6.1) can be numerically implemented at matrix level without invoking tensor manipulation. Particularly in the H_s -eigenstate representation, $\tilde{Q}_{uv} = \hat{C}(-\omega_{uv})Q_{uv}$, and $\hat{Q}_{uv}^{uv} = Q_{uv}/(i\omega_{uv} + \zeta_m)^{\delta_{m0}+1}$ (cf. Equations 4.13 and 4.26), where $\omega_{uv} \equiv \varepsilon_u - \varepsilon_v$ are the transition frequencies between the H_s -eigenstates. The tensor-free implementation of the CODDE thus follows immediately.

6.2. Related Linear-Space Algebra

For the algebraic construction, let us denote ($\{m = 0, 1, \dots, \bar{m}\}$ be implied hereafter)

$$\sigma(t) \equiv \{\sigma(t), \sigma_m^{(-)}(t), \sigma_m^{(+)}(t)\} \quad 6.4$$

as a vector of $1 + 2(\bar{m} + 1)$ elements, and recast Equations 6.1 as

$$\dot{\sigma}(t) = -\hat{\Lambda}(t)\sigma(t) \equiv -[\hat{\Lambda}_s + \hat{\Lambda}_{st}(t)]\sigma(t). \quad 6.5$$

The CODDE-space propagator $\hat{\mathcal{G}}(t, \tau)$ is then defined via the formal solution to Equation 6.5,

$$\sigma(t) \equiv \hat{\mathcal{G}}(t, \tau)\sigma(\tau); \quad \text{with } t \geq \tau. \quad 6.6$$

It is easy to show that

$$\partial \hat{\mathcal{G}}(t, \tau) / \partial t = -\hat{\Lambda}(t)\hat{\mathcal{G}}(t, \tau), \quad \partial \hat{\mathcal{G}}(t, \tau) / \partial \tau = \hat{\mathcal{G}}(t, \tau)\hat{\Lambda}(\tau), \quad 6.7a$$

$$\hat{\mathcal{G}}(\tau_2, \tau_0) = \hat{\mathcal{G}}(\tau_2, \tau_1)\hat{\mathcal{G}}(\tau_1, \tau_0), \quad \text{with } \tau_2 \geq \tau_1 \geq \tau_0. \quad 6.7b$$

The field-free propagator is given by $\hat{\mathcal{G}}_s(t, \tau) = \hat{\mathcal{G}}_s(t - \tau)$,

$$\hat{\mathcal{G}}_s(t) \equiv \exp(-\hat{\Lambda}_s t), \quad 6.8$$

with the time-independent field-free generator $\hat{\Lambda}_s$.

We shall hereafter refer to the linear space defined by Equation 6.5 (or Equations 6.1) as the CODDE space. Its element can be time-dependent and is defined as

$$A \equiv \{A, A_m^{(-)}, A_m^{(+)}\}, \quad 6.9$$

where A relates to an ordinary dynamical or state variable, while $\{A_m^{(-)}, A_m^{(+)}\}$ are a set of auxiliary components (cf. Equation 6.4). The CODDE-space scalar product

can then be defined in the tetradic notation as (103)

$$\langle\langle \mathbf{A} | \mathbf{B} \rangle\rangle \equiv \langle\langle A | B \rangle\rangle + \sum_m \langle\langle A_m^{(-)} | B_m^{(-)} \rangle\rangle + \sum_m \langle\langle A_m^{(+)} | B_m^{(+)} \rangle\rangle. \quad 6.10.$$

Here, $\langle\langle A | B \rangle\rangle \equiv \text{Tr}(A^\dagger B)$, and so on. The propagator $\hat{\mathcal{G}}(t, \tau)$ and its generator $\hat{\Lambda}(t)$ are examples of the CODDE-space operators.

The left-actions of $\hat{\Lambda}(t)$ and its field-free $\hat{\Lambda}_s$ and field-dressed $\hat{\Lambda}_{\text{sf}}(t)$ counterparts are all specified via Equation 6.5 with Equations 6.1. Their right-actions can then be equivalently defined following the derivations presented in Appendix E. In particular, we have

$$\hat{\Lambda}_{\text{sf}}(t) \mathbf{A} = i[H_{\text{sf}}(t), \mathbf{A}] + \{0, i[H_{\text{sf}}(t), \hat{Q}_m] \mathbf{A}, i \mathbf{A}[H_{\text{sf}}(t), \hat{Q}_m^\dagger]\}, \quad 6.11a$$

$$\mathbf{A} \hat{\Lambda}_{\text{sf}}(t) = i[\mathbf{A}, H_{\text{sf}}(t)] + \left\{ i \sum_m ([H_{\text{sf}}, \hat{Q}_m^\dagger] A_m^{(-)} + A_m^{(+)} [H_{\text{sf}}, \hat{Q}_m]), 0, 0 \right\}. \quad 6.11b$$

6.3. Heisenberg Picture

We are now in a position to define the Heisenberg picture, for example, via the field-free generator $\hat{\Lambda}_s$,

$$\langle\langle \mathbf{A}(t) | \equiv \langle\langle \mathbf{A} | \exp(-\hat{\Lambda}_s t), \quad 6.12.$$

or equivalently

$$\mathbf{A}(t) \equiv \mathbf{A}(0) \exp(-\hat{\Lambda}_s t). \quad 6.13.$$

Here

$$\mathbf{A}(0) \equiv \{\mathbf{A}, 0, 0\}, \quad 6.14.$$

where A is an ordinary dynamic variable that can be non-Hermitian. The Heisenberg equation of motion in the CODDE space is then

$$\dot{\mathbf{A}}(t) = -\mathbf{A}(t) \hat{\Lambda}_s, \quad 6.15.$$

which is equivalent to (cf. Appendix E)

$$\dot{\mathbf{A}}(t) = -\mathbf{A}(t)(i\mathcal{L}_s + \mathcal{R}_s), \quad 6.16a.$$

$$\dot{A}_m^{(-)}(t) = \delta_{m1} A_0^{(-)}(t) - A_m^{(-)}(t)(i\mathcal{L}_s + \zeta_m^*) + \nu_m^* [A(t), Q], \quad 6.16b.$$

$$\dot{A}_m^{(+)}(t) = \delta_{m1} A_0^{(+)}(t) - A_m^{(+)}(t)(i\mathcal{L}_s + \zeta_m) - \nu_m [A(t), Q]. \quad 6.16c.$$

The right-actions of \mathcal{L}_s and \mathcal{R}_s were given by Equation 6.3. Clearly, $A(t) = A^\dagger(t)$ and $A_m^{(\pm)}(t) = A_m^{(\mp)\dagger}(t)$ if they were Hermitian conjugate initially. Note that the established Heisenberg picture here is closely related, but not identical, to the

backward propagation (cf. Equations 7.14 or 7.15) in the CODDE space. The latter will be discussed in relation to optimal control (cf. Section 7.2).

It is interesting to compare the Heisenberg dynamics of $A(t) = \{A(t), A_m^{(-)}(t), A_m^{(+)}(t)\}$, to its corresponding Schrödinger dynamics of $\sigma(t) = \{\sigma(t), \sigma_m^{(-)}(t), \sigma_m^{(+)}(t)\}$. The former is described by Equations 6.16 and the latter by Equations 6.1 by setting $H_{sf}(t) = 0$. The field-free propagation of $\sigma(t)$ is characterized by $\dot{\sigma}(t)$, which depends on $\sigma_m^{(\pm)}(t)$, but $\dot{\sigma}_m^{(\pm)}(t)$ does not depend on $\sigma(t)$. The Heisenberg dynamics of $A(t)$ in Equations 6.16 are opposite.

7. APPLICATIONS

7.1. Reduced Linear Response Theory

Consider the measurement on a dynamical variable A via a classical weak probe field $\epsilon_{pr}(t)$ that couples with the system by $H_{pr}(t) = -B\epsilon_{pr}(t)$. Both A and B are Hermite operators in the reduced system subspace. The weak probe-induced variation of the expectation value $\bar{A}(t)$ is then given by

$$\delta\bar{A}(t) = \text{tr}[A \delta\rho(t)] = \langle\langle A(0) | \delta\rho(t) \rangle\rangle. \tag{7.1}$$

Here, $A(0)$ and $\delta\rho(t)$ denote the CODDE-space extensions of the dynamical variable A and the probe-induced reduced density operator change $\delta\rho(t)$ (cf. Equations 6.4 and 6.14). By applying the standard first-order perturbation theory to Equation 6.5, we obtain

$$\delta\bar{A}(t) = i \int_{-\infty}^t d\tau \langle\langle A(0) | \hat{\mathcal{G}}(t, \tau) \hat{\mathcal{B}} | \rho(\tau) \rangle\rangle \epsilon_{pr}(\tau). \tag{7.2}$$

Here, $\hat{\mathcal{B}} \equiv i\hat{\Lambda}_{pr}(t)/\epsilon_{pr}(t)$, i.e., (cf. Equation 6.11a)

$$\hat{\mathcal{B}}\rho = [B, \rho] + \{0, [B, \hat{Q}_m]\rho, \rho[B, \hat{Q}_m^\dagger]\}. \tag{7.3}$$

The above formulation is valid whether there is a pump field or not. In the absence of pump excitation, $\rho(\tau)$ and $\hat{\mathcal{G}}(t, \tau)$ in Equation 7.2 assume the thermal equilibrium state $\rho_{eq}(T) \equiv \{\rho_{eq}(T), 0, 0\}$ and the field-free propagator $\hat{\mathcal{G}}_s(t - \tau)$ of Equation 6.8. In this case, Equation 7.2 assumes the conventional linear response theory (Equation 2.1), where the response function is now evaluated as

$$\chi_{AB}(t) = i \langle\langle A(0) | \exp(-\hat{\Lambda}_s t) \hat{\mathcal{B}} | \rho_{eq}(T) \rangle\rangle. \tag{7.4}$$

The nonlinear response formulations can also be readily constructed in terms of CODDE dynamics.

In the Schrödinger picture, the above equation reads as $\chi_{AB}(t) = \langle\langle A(0) | \sigma(t) \rangle\rangle = \text{tr}[A\sigma(t)]$ (cf. Equation 6.14). Here $\sigma(t) = \exp(-\hat{\Lambda}_s t)\sigma(0) = \{\sigma(t), \sigma_m^{(-)}(t), \sigma_m^{(+)}(t)\}$ is governed by Equations 6.1 with $H_{sf}(t) = 0$. The initially prepared

reduced state is $\sigma(0) = i\hat{\mathcal{B}}\rho_{\text{eq}}(T)$ (cf. Equation 7.3), which contains the non-vanished $\sigma_m^{(\pm)}(0)$ to incorporate the correlated driving-dissipation effects on $\sigma(t \geq 0)$. Note that for a Hermitian B , we have $\sigma(t) = \sigma^\dagger(t)$ and $\sigma_m^{(+)}(t) = [\sigma_m^{(-)}(t)]^\dagger$.

In the Heisenberg picture, Equation 7.4 reads as $\chi_{AB}(t) = i\langle\langle A(t)|\hat{\mathcal{B}}|\rho_{\text{eq}}(T)\rangle\rangle$, in which $A(t)$, with the initial condition of Equation 6.14, is governed by Equations 6.16. For a Hermitian A , we have $A(t) = A^\dagger(t)$, $A_m^{(-)}(t) = [A_m^{(+)}(t)]^\dagger$, and thus

$$\chi_{AB}(t) = i\langle[A(t), B(0)]\rangle + i\sum_m \langle A_m^\dagger(t)[B(0), \hat{Q}_m] + [B(0), \hat{Q}_m^\dagger]A_m(t)\rangle. \quad 7.5$$

Here, $A_m \equiv A_m^{(-)}$ and $\langle O \rangle \equiv \text{tr}[O\rho_{\text{eq}}(T)]$. Clearly, $\chi_{AB}(t)$ of Equation 7.5 is real. The first term in the right-hand-side of Equation 7.5 resembles the definition of the response function (Equation 2.2), but is now evaluated in the reduced system subspace, rather than the total space, of composite material. The second term in Equation 7.5 makes up the discrepancy, up to the second order in the system-bath coupling, via the correlated driving and dissipation contribution.

The correlation function in terms of the CODDE-space dynamics can be obtained as

$$\tilde{C}_{AB}(t) = \langle\langle A(0)|\exp(-\hat{\Lambda}_s t)\vec{\mathcal{B}}|\rho_{\text{eq}}(T)\rangle\rangle, \quad 7.6$$

with

$$\vec{\mathcal{B}}\rho_{\text{eq}}(T) = \{B\rho_{\text{eq}}, [B, \hat{Q}_m]\rho_{\text{eq}}, 0\}. \quad 7.7$$

Again, Equation 7.6 can be implemented in either the Schrödinger or the Heisenberg picture. In the latter case, Equation 7.6 assumes

$$\tilde{C}_{AB}(t) = \langle A(t)B(0)\rangle + \sum_m \langle A_m^\dagger(t)[B(0), \hat{Q}_m]\rangle. \quad 7.8$$

Clearly, we have $\chi_{AB}(t) = i[\tilde{C}_{AB}(t) - \tilde{C}_{AB}^*(t)]$ as required. Again, the second term in the right-hand-side of Equation 7.8, arising from the correlated driving and dissipation, makes up the difference, up to second order, between the reduced and the complete descriptions.

Note that the above formulations for correlation and response functions are valid for $t \geq 0$. Their values at $t < 0$ can be obtained via the symmetry relations of Equations 2.6 and 2.3, respectively. Where the CS-QDT theory is concerned, the response and correlation functions presented here satisfy the FDT (Equation 2.16) up to the second order of system-bath interaction.

7.2. Optimal Control Theory

In a control problem, an optimal field $\epsilon(t)$ is needed to drive the reduced system to have minimal deviation from a desired target ρ_{tar} at a specified time t_f . To formulate the control problem, let us start with the control discrepancy operator (61),

$$A \equiv \rho(t_f) - \rho_{\text{tar}}. \quad 7.9$$

The control objective is set to minimize the discrepancy,

$$\text{tr}A^2 = \text{tr}\rho^2(t_f) + \text{tr}\rho_{\text{tar}}^2 - 2\text{tr}[\rho_{\text{tar}}\rho(t_f)], \quad 7.10.$$

under certain penalties or constraints (104, 105). Consider here the simplest penalty might be that the incident energy of the control field is minimal in balance with meeting the control objective. In this case, we arrive at the following control equation for the optimal field (61),

$$\langle\langle A(t; t_f) | i\hat{\mathcal{D}} | \rho(t) \rangle\rangle = -\lambda\epsilon(t), \quad \text{with } t_0 \leq t \leq t_f. \quad 7.11.$$

Here, $\lambda > 0$ is a weight factor to enforce the energy constraint, and $\hat{\mathcal{D}}$ is the CODDE extension of the dipole commutator (cf. Equation 6.11a),

$$\hat{\mathcal{D}}\rho(t) = [\mu, \rho(t)] + \{0, [\mu, \hat{Q}_m]\rho(t), \rho(t)[\mu, \hat{Q}_m^\dagger]\}. \quad 7.12.$$

In Equation 7.11, $A(t; t_f)$ is the backward-propagated CODDE-space target,

$$A(t; t_f) \equiv A\hat{\mathcal{G}}(t_f, t), \quad \text{with } A(t_f; t_f) \equiv A \equiv \{A, 0, 0\}. \quad 7.13.$$

Using the second identity in Equation 6.7a, we have

$$\dot{A}(t; t_f) = A(t; t_f)\hat{\Lambda}(t), \quad 7.14.$$

which is equivalent to (cf. Appendix E)

$$\begin{aligned} \dot{A}(t; t_f) = i \sum_m \{ [H_{\text{sf}}(t), \hat{Q}_m^\dagger] A_m^{(-)}(t; t_f) + A_m^{(+)}(t; t_f) [H_{\text{sf}}(t), \hat{Q}_m] \\ + A(t; t_f) [i\mathcal{L}(t) + R_s], \end{aligned} \quad 7.15a.$$

$$\dot{A}_m^{(-)}(t; t_f) = A_m^{(-)}(t; t_f) [i\mathcal{L}(t) + \zeta_m^*] - v_m^* [A(t; t_f), Q] - \delta_{m1} A_0^{(-)}(t; t_f), \quad 7.15b.$$

$$\dot{A}_m^{(+)}(t; t_f) = A_m^{(+)}(t; t_f) [i\mathcal{L}(t) + \zeta_m] + v_m [A(t; t_f), Q] - \delta_{m1} A_0^{(+)}(t; t_f). \quad 7.15c.$$

In other words, Equations 7.15 define the right-action of the CODDE generator $\hat{\Lambda}(t)$ in the backward propagation of Equation 7.14.

We note that the Heisenberg equation of motion (Equation 6.15) is also defined via the right-action with the time-independent field-free generator $\hat{\Lambda}_s$. However, the Heisenberg equation of motion is intrinsically a forward-propagation, arising from the field-free variation of the first identity of Equation 6.7a, i.e., $\partial\hat{\mathcal{G}}_s(t, \tau)/\partial t = -\hat{\Lambda}_s\hat{\mathcal{G}}_s(t, \tau)$. As $\hat{\mathcal{G}}_s(t, \tau) \equiv \hat{\mathcal{G}}_s(t - \tau)$ and $\hat{\Lambda}_s\hat{\mathcal{G}}_s(t) = \hat{\mathcal{G}}_s(t)\hat{\Lambda}_s$, we have thus $\partial\hat{\mathcal{G}}_s(t)/\partial t = -\hat{\mathcal{G}}_s(t)\hat{\Lambda}_s$. It is this variation of forward-propagation that constitutes the Heisenberg equation of motion in Equation 6.15.

In contrast, the backward-propagation in Equation 7.14 arises from the second identity of Equation 6.7a, where the derivative is taken with respect to the early time of the two in the propagator. Therefore, the evaluation of the control kernel in the left-hand-side of Equation 7.11 involves the forward propagation of $\rho(t)$ from the initial time t_0 with $\rho(t_0) = \rho_{\text{eq}}(T) \equiv \{\rho_{\text{eq}}(T), 0, 0\}$, and backward propagation

of $A(t; t_f)$ from the final time t_f with the $A(t_f; t_f)$ in Equation 7.13. Both propagations are governed by the control field-dressed generator $\hat{\Lambda}(t)$. In principle, the forward and the backward propagations can be performed independently. However, the control equation (Equation 7.11) shall be solved in an iterative manner for the control field. To improve the numerical convergency, these two propagations may need to be carried out alternatively in iteration (61, 106, 107).

8. CONCLUDING REMARKS

An exact QDT may be constructed for arbitrary systems via, for example, stochastic system-bath decoupling methods (93, 94). However, the resulting hierarchical formulation is rather complicated, even when the bath correlation function is set to be of a single complex exponential term (62, 63, 93). If $C(t) = \nu e^{-\zeta t}$ were real, the resulting hierarchical QDT would be simplified as $\dot{\rho}^{(n)} = -in\nu[Q, \rho^{(n-1)}] - (i\mathcal{L} + n\zeta)\rho^{(n)} - i[Q, \rho^{(n+1)}]$, where the auxiliary operator $\rho^{(n)}$; $n \geq 1$ accounts for the effects of the $2n$ -order system-bath coupling on $\rho \equiv \rho^{(0)}$. The COP-CS-QDT (Equations 4.20) sets all $\rho^{(n \geq 2)} = 0$ in this type of hierarchical construction with general forms of bath correlation function. However, a satisfactory CS-QDT should go beyond this simple truncation scheme to meet some basic physical requirements at least in the simplest system as demonstrated in Section 4.4.

The CODDE theory presented in the final three sections is the choice of CS-QDT formulation by far. The CODDE (Equations 5.4 or 6.1) is a variation of the cumulant resum scheme to partially account for higher order contributions on the reduced dynamics including the reduced canonical states. The involving auxiliary operators $\{\rho_m\}$ or $\{\sigma_m^{(\pm)}\}$ are of the same number for the terms in the exponential series for bath correlation function (Equation 4.2). They account for the correlated driving-dissipation effects, which are usually nonnegligible for a non-Markovian bath, even in the calculation of field-free correlation/response functions.

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APPENDIX

A. EQUILIBRIUM PHASE-SPACE VARIANCES

The FDT of Equation 2.16 can be directly exploited to establish the general expressions for the thermal equilibrium values of $\sigma_{qq}^{\text{eq}} \equiv \langle q^2 \rangle - \langle q \rangle^2$ and $\sigma_{pp}^{\text{eq}} \equiv \langle p^2 \rangle - \langle p \rangle^2$ in terms of the response function $\chi_{qq}(t)$ or its causality Fourier

transform $\hat{\chi}_{qq}(\omega)$. Here, q and $p = M\dot{q}$ denote the Cartesian coordinate and momentum of an arbitrary reduced degree of freedom, respectively. Thus, $\sigma_{qq}^{\text{eq}} = \tilde{C}_{qq}(0)$ and $\sigma_{pp}^{\text{eq}} = -M^2\ddot{C}_{qq}(0)$. The latter is obtained by using Equation 2.5, which gives also $\sigma_{pq}^{\text{eq}} = \langle qp + pq \rangle / 2 = 0$. The FDT (Equation 2.16) leads immediately to the expressions of the phase-space variances

$$\sigma_{qq}^{\text{eq}} = \frac{1}{\pi} \text{Im} \int_{-\infty}^{\infty} d\omega \frac{\hat{\chi}_{qq}(\omega)}{1 - e^{-\beta\omega}}, \quad \sigma_{pp}^{\text{eq}} = \frac{M^2}{\pi} \text{Im} \int_{-\infty}^{\infty} d\omega \frac{\omega^2 \hat{\chi}_{qq}(\omega)}{1 - e^{-\beta\omega}}. \quad \text{A.1.}$$

Note that $\hat{\chi}_{qq}(z)$ is an analytical function in the upper plane ($\text{Im}z > 0$). Equation A.1 can then be recast in terms of contour integrations. The involving poles can be readily identified via the following Laurent expansion expression,

$$\frac{1}{1 - e^{-\beta\omega}} = \frac{1}{2} + \frac{1}{\beta\omega} + \frac{2}{\beta} \sum_{n=1}^{\infty} \frac{\omega}{\omega^2 + \varpi_n^2}. \quad \text{A.2.}$$

Here, $\varpi_n \equiv 2\pi n/\beta$ is the Matsubara frequency. In derivation, we also make use of the properties where $\chi_{qq}(t)$ is real with $\chi_{qq}(0) = 0$ and $\dot{\chi}_{qq}(0) = 1/M$, while real functions $\hat{\chi}_{qq}^{(+)}(\omega)$ and $\hat{\chi}_{qq}^{(-)}(\omega)$ are symmetric and antisymmetric, respectively (cf. Section 2.2). After some elementary algebra, we finally arrive at the following alternative expressions equivalent to Equation A.1 for the phase-space variances:

$$\sigma_{qq}^{\text{eq}} = \frac{1}{\beta} \sum_{n=-\infty}^{\infty} \hat{\chi}_{qq}(i|\varpi_n|), \quad \sigma_{pp}^{\text{eq}} = \frac{M}{\beta} \sum_{n=-\infty}^{\infty} [1 - M\varpi_n^2 \hat{\chi}_{qq}(i|\varpi_n|)]. \quad \text{A.3.}$$

Note that the above expressions were traditionally presented only for the harmonic Brownian oscillator systems (37). The general equivalence between Equation A.1 and Equation A.3 including anharmonic systems is proved via the principles in quantum mechanics (60).

B. PARAMETERIZATION OF BATH CORRELATION FUNCTIONS

The extended Meier-Tannor parameterization scheme to the exponential series of $\tilde{C}_{ab}(t)$ (Equation 4.2) starts with the following form of interaction bath spectral density functions (57, 60, 61):

$$J_{ab}(\omega) = \sum_{k=0}^{\bar{k}} \frac{\eta_k^{ab} \omega + i\bar{\eta}_k^{ab} \omega^2}{|\omega^2 - (\omega_k^{ab} + i\gamma_k^{ab})^2|}, \quad \text{with} \quad \omega_0^{ab} \equiv 0. \quad \text{B.1.}$$

The parameters here are all real ($\omega_{k \neq 0}^{ab}$ and γ_k^{ab} are positive as well) and satisfy the symmetry relations of $(\omega_k^{ba}, \gamma_k^{ba}, \eta_k^{ba}, \bar{\eta}_k^{ba}) = (\omega_k^{ab}, \gamma_k^{ab}, \eta_k^{ab}, -\bar{\eta}_k^{ab})$, along with $\bar{\eta}_k^{aa} = 0$. Thus, Equation B.1 meets the required symmetry relations, $J_{ab}(\omega) = -J_{ba}(-\omega) = J_{ba}^*(\omega)$, with $J_{ab}(0) = 0$, for the interaction bath spectral density

functions. In comparison with the original Meier-Tannor formulation (57), Equation B.1 extends to $a \neq b$ cases and also includes $\omega_0^{ab} = 0$ to improve the quality of parameterization. Note that Equation B.1 results in the frictional functions of (cf. Equation 2.21)

$$\gamma_{ab}(t \geq 0) = \left[\frac{\eta_0^{ab}}{(\gamma_0^{ab})^2} + \left(\frac{\eta_0^{ab}}{\gamma_0^{ab}} + \bar{\eta}_0^{ab} \right) t \right] \frac{e^{-\gamma_0^{ab} t}}{2\gamma_0^{ab}} + 2 \operatorname{Re} \sum_{k=1}^{\bar{k}} \frac{\alpha_k^{ab} e^{i z_k^{ab} t}}{z_k^{ab}}, \quad \text{B.2.}$$

where

$$z_k^{ab} \equiv \omega_k^{ab} + i\gamma_k^{ab} \quad \text{and} \quad \alpha_k^{ab} \equiv \frac{\eta_k^{ab} - i\bar{\eta}_k^{ab} z_k^{ab}}{4\omega_k^{ab} \gamma_k^{ab}}, \quad \text{for } k > 0. \quad \text{B.3.}$$

The bath correlation function can now be obtained by using the FDT in Equation 2.20 and the contour integration algorithm, which can be easily carried out via Equation A.2. We have

$$\begin{aligned} \tilde{C}_{ab}(t \geq 0) &= v_0^{ab} t e^{-\gamma_0^{ab} t} + v_1^{ab} e^{-\gamma_0^{ab} t} \\ &+ \sum_{k=1}^{\bar{k}} \left[\frac{\alpha_k^{ab}}{e^{\beta z_k^{ab}} - 1} e^{i z_k^{ab} t} + \frac{(\alpha_k^{ab})^*}{1 - e^{-\beta (z_k^{ab})^*}} e^{-i (z_k^{ab})^* t} \right] \\ &- \frac{2}{\beta} \sum_{n=1}^{\infty} \tilde{J}_{ab}(\varpi_n) e^{-\varpi_n t}, \end{aligned} \quad \text{B.4.}$$

with

$$v_0^{ab} \equiv \frac{\eta_0^{ab}/\gamma_0^{ab} + \bar{\eta}_0^{ab}}{2i(1 - e^{i\beta\gamma_0^{ab}})}, \quad \text{B.5a.}$$

$$v_1^{ab} \equiv \frac{\beta(\eta_0^{ab}/\gamma_0^{ab} + \bar{\eta}_0^{ab})}{2|1 - e^{i\beta\gamma_0^{ab}}|^2} + \frac{i\bar{\eta}_0^{ab}/\gamma_0^{ab}}{2(1 - e^{i\beta\gamma_0^{ab}})}, \quad \text{B.5b.}$$

and

$$\tilde{J}_{ab}(\omega) \equiv i J_{ab}(-i\omega) = \sum_{k=0}^{\bar{k}} \frac{\eta_k^{ab} \omega + \bar{\eta}_k^{ab} \omega^2}{|\omega^2 + (\omega_k^{ab} + i\gamma_k^{ab})^2|^2}. \quad \text{B.6.}$$

Note that $\tilde{J}_{ab}(\omega)$ is a real function, which is involved in Equation B.4 because of its values at the Matsubara frequencies $\varpi_n \equiv 2\pi n/\beta$. Equation B.4 assumes the form of Equation 4.2. The involving parameters, $\{v_m^{ab}, \zeta_m^{ab}; m = 0, \dots, \bar{m}\}$, with \bar{m} being determined via numerical convergence, are now all specified. The first two terms (with coefficients v_0^{ab} and v_1^{ab} and decaying exponents $\zeta_0^{ab} \equiv \zeta_1^{ab} = \gamma_0^{ab}$) in Equations B.4 or 4.2 arise from the $\omega_{k=0}^{ab} \equiv 0$ term in Equation B.1.

C. EVALUATION OF REDUCED CANONICAL DENSITY MATRIX

In a given Hilbert-space representation $\{|u\rangle; u = 0, \dots, N - 1\}$, Equation 4.27 consists of N^2 equations,

$$\dot{\rho}_{uv}^{\text{eq}} = - \sum_{u'v'} (i\mathcal{L}_{uv,u'v'}^s + \mathcal{R}_{uv,u'v'}^s) \rho_{u'v'}^{\text{eq}} = 0. \tag{C.1}$$

Here, $\rho_{uv}^{\text{eq}} \equiv \langle u | \rho_{\text{eq}}^{\text{POP}} | v \rangle$. The tensor elements in Equation C.1 are given by (cf. Equations 4.4 and 4.14)

$$\mathcal{L}_{uv,u'v'}^s = H_{uu'}^s \delta_{v'v} - H_{v'v}^s \delta_{uu'}, \tag{C.2a}$$

$$\mathcal{R}_{uv,u'v'}^s = \sum_a [(Q_a \tilde{Q}_a)_{uu'} \delta_{v'v} + (Q_a \tilde{Q}_a)_{v'v}^* \delta_{uu'} - Q_{v'v}^a \tilde{Q}_{uu'}^a - Q_{uu'}^a \tilde{Q}_{v'v'}^{a*}]. \tag{C.2b}$$

Here, $\tilde{Q}_{uv}^a \equiv \langle u | \tilde{Q}_a | v \rangle$, which can be evaluated as follows (cf. Equation 4.13). Let $H_s |\bar{u}\rangle \equiv \varepsilon_{\bar{u}} |\bar{u}\rangle$ and $\omega_{\bar{u}\bar{v}} \equiv (\varepsilon_{\bar{u}} - \varepsilon_{\bar{v}})$. We then have $\mathcal{L}_{\bar{u}\bar{v},\bar{u}'\bar{v}'}^s = \omega_{\bar{u}\bar{v}} \delta_{\bar{u}\bar{u}'} \delta_{\bar{v}\bar{v}'}$ (cf. Equation C.2a), and (where $S_{\bar{u}\bar{u}} \equiv \langle \bar{u} | u \rangle$)

$$\tilde{Q}_{uv}^a = \sum_b \sum_{\bar{u}\bar{v}} S_{\bar{u}\bar{u}}^* \hat{C}_{ab} (-\omega_{\bar{u}\bar{v}}) Q_{\bar{u}\bar{v}}^b S_{\bar{v}\bar{v}}. \tag{C.3}$$

It is easy to show that $\sum_u \mathcal{L}_{uu,u'v'}^s = \sum_u \mathcal{R}_{uu,u'v'}^s = 0$. Therefore, the N^2 equations in Equation C.1 are not independent; they are subject to $\sum_u \rho_{uu}^{\text{eq}} = 1$. By using this normalization condition to replace a diagonal one in Equation C.1 (for example, $\dot{\rho}_{00}^{\text{eq}} = 0$), we obtain a set of N^2 independent linear equations (cf. Equation C.4) that uniquely determine ρ_{eq} in the specified Hilbert-space representation (60, 61). For systems involving degenerate states, Equation C.1 can further incorporate relevant conditions for evaluating ρ_{eq} unambiguously. To write the normalized Equation C.1 explicitly in terms of ordinary linear equations, we may rearrange the tensor $\Lambda_{uv,u'v'}^s \equiv i\mathcal{L}_{uv,u'v'}^s + \mathcal{R}_{uv,u'v'}^s$ and matrix ρ_{uv}^{eq} into the matrix $\Lambda_{\alpha\alpha'}^s$ and vector $\rho_{\alpha'}^{\text{eq}}$, respectively. Here, $\alpha = uN + v$. We then have

$$\sum_{\alpha'=0}^{N^2-1} \Lambda_{\alpha\alpha'}^s \rho_{\alpha'}^{\text{eq}} = 0, \quad \text{for } \alpha \neq 0, \tag{C.4a}$$

$$\sum_{u=0}^{N-1} \rho_{uN+u}^{\text{eq}} = 1. \tag{C.4b}$$

These N^2 equations uniquely determine the $\rho_{\text{eq}}^{\text{POP}}$ of POP-CS-QDT.

D. RESULTS OF PERTURBATIVE THEORIES ON DRIVEN BROWNIAN OSCILLATOR SYSTEMS

Let us start with the exact QDT for the DBO system described in Section 3.1, i.e, Equation 3.10, which can be recast as

$$\dot{\rho}(t) = -i[H(t) - q\delta\epsilon(t), \rho(t)] - \mathcal{R}_s^{\text{ex}}\rho(t), \quad \text{D.1}$$

where $\delta\epsilon(t)$ is given by the last term in Equation 3.12a, and

$$\begin{aligned} \mathcal{R}_s^{\text{ex}}\rho &= \frac{i}{2}M(\tilde{\Omega}^2 - \Omega_{\text{H}}^2)[q^2, \rho] + \frac{i}{2}\tilde{\gamma}[q, \{p, \rho(t)\}] + \tilde{\gamma}\sigma_{pp}^{\text{eq}}[q, [q, \rho(t)]] \\ &+ (\sigma_{pp}^{\text{eq}}/M - M\tilde{\Omega}^2\sigma_{qq}^{\text{eq}})[q, [p, \rho(t)]]. \end{aligned} \quad \text{D.2}$$

By using the boson algebra, one can also easily obtain the POP-CS-QDT (Equation 4.8 with Equations 4.10a, 4.12, and 4.14) for the DBO system, which is of the same expression as Equation D.1, but replacing $\mathcal{R}_s^{\text{ex}}$, we have (59, 60)

$$\begin{aligned} \mathcal{R}_s\rho(t) &= i\kappa_+''[q^2, \rho(t)] + i\frac{\kappa_+'}{M\Omega_{\text{H}}}[q, \{p, \rho(t)\}] + \kappa_+'[q, [q, \rho(t)]] \\ &- \frac{\kappa_+''}{M\Omega_{\text{H}}}[q, [p, \rho(t)]]. \end{aligned} \quad \text{D.3}$$

Here, $\kappa_{\pm} \equiv \frac{M}{2}[\hat{C}(\Omega_{\text{H}}) \pm \hat{C}(-\Omega_{\text{H}})] \equiv \kappa_{\pm}' + i\kappa_{\pm}''$. Also note that the local field correction $\delta\epsilon(t)$ is now characterized by $\chi_{\epsilon}(t) \equiv \int_0^{\infty} d\tau \cos(\Omega_{\text{H}}\tau)\gamma(t + \tau)$, in comparison with the exact $\chi_{\epsilon}^{\text{ex}}(t)$, which was given by Equation 3.12b in terms of $\chi_{\epsilon}^{\text{ex}}(t, \tau) = \chi_{\epsilon}^{\text{ex}}(t - \tau)$. The comparison between each individual term in Equation D.2 and their counterparts in Equation D.3 results therefore in four relations for the POP-CS-QDT counterparts of $\tilde{\Omega}$, $\tilde{\gamma}$, σ_{qq}^{eq} , and σ_{pp}^{eq} . In particular, the latter two are

$$\sigma_{qq}^{\text{pop}}(T) = \frac{\Omega_{\text{H}}^2 \coth(\beta\Omega_{\text{H}}/2) + D_-(\Omega_{\text{H}})}{2M\Omega_{\text{H}}[\Omega_{\text{H}}^2 + D_+(\Omega_{\text{H}})]}, \quad \text{D.4a}$$

$$\sigma_{pp}^{\text{pop}}(T) = \frac{1}{2}M\Omega_{\text{H}} \coth(\beta\Omega_{\text{H}}/2). \quad \text{D.4b}$$

Here, $D_{\pm}(\Omega_{\text{H}}) \equiv D(\Omega_{\text{H}}) \pm D(-\Omega_{\text{H}})$. Note that σ_{pp}^{pop} is the same as its dissipation-free value of the H_s -system. Obviously, the POP-CS-QDT preserves the Gaussian property.

In contrast, the COP-CS-QDT propagator results in a non-Gaussian $\rho^{\text{cop}}(t)$, despite the fact that it has the exact first moments. The non-Gaussian $\rho_{\text{eq}}^{\text{cop}}$ (Equation 4.28) can be found to have

$$\sigma_{qq}^{\text{cop}}(T) = \frac{\coth(\beta\Omega_{\text{H}}/2)}{2M\Omega_{\text{H}}}, \quad \text{D.5a}$$

$$\sigma_{pp}^{\text{cop}}(T) = \left[\frac{1}{2} M \Omega_H + \frac{M}{2\Omega_H} D_+(\Omega_H) \right] \coth(\beta\Omega_H/2) - \frac{M}{2\Omega_H} D_-(\Omega_H). \quad \text{D.5b.}$$

E. ABOUT THE RIGHT ACTION OF CODDE GENERATOR

Let $\langle\langle \tilde{A} | \equiv \langle\langle A | \hat{\Lambda}, | \tilde{B} \rangle\rangle \equiv \hat{\Lambda} | B \rangle\rangle$ and $\langle\langle \tilde{A} | B \rangle\rangle \equiv \langle\langle A | \tilde{B} \rangle\rangle$ define the relationship between the left and right actions of $\hat{\Lambda}$. Given that

$$\zeta_m^{(+)*} \equiv \zeta_m^{(-)} \equiv \zeta_m \quad \text{and} \quad \nu_m^{(+)*} \equiv \nu_m^{(-)} \equiv \nu_m, \quad \text{E.1.}$$

the action of $\hat{\Lambda}$ to the right, where $\tilde{B} = \hat{\Lambda} B$, is obtained by Equations 6.1 as

$$\tilde{B} = (i\mathcal{L} + \mathcal{R}_s)B + \sum_m [Q, \nu_m^{(-)} B_m^{(-)} - \nu_m^{(+)} B_m^{(+)}], \quad \text{E.2a.}$$

$$\tilde{B}_m^{(-)} = -\delta_{m0} B_1^{(-)} + [i\mathcal{L} + \zeta_m^{(-)}] B_m^{(-)} + i[H_{\text{sf}}, \hat{Q}_m] B, \quad \text{E.2b.}$$

$$\tilde{B}_m^{(+)} = -\delta_{m0} B_1^{(+)} + [i\mathcal{L} + \zeta_m^{(+)}] B_m^{(+)} + iB[H_{\text{sf}}, \hat{Q}_m^\dagger]. \quad \text{E.2c.}$$

The involving terms in $\langle\langle A | \tilde{B} \rangle\rangle$ are therefore (cf. Equation 6.10)

$$\langle\langle A | \tilde{B} \rangle\rangle = \langle\langle A | i\mathcal{L} + \mathcal{R}_s | B \rangle\rangle + \sum_m \langle\langle A | [Q, \nu_m^{(-)} B_m^{(-)} - \nu_m^{(+)} B_m^{(+)}] \rangle\rangle, \quad \text{E.3a.}$$

$$\begin{aligned} \langle\langle A_m^{(-)} | \tilde{B}_m^{(-)} \rangle\rangle &= -\delta_{m0} \langle\langle A_m^{(-)} | B_1^{(-)} \rangle\rangle + \langle\langle A_m^{(-)} | i\mathcal{L} + \zeta_m^{(-)} | B_m^{(-)} \rangle\rangle \\ &\quad + i \langle\langle A_m^{(-)} | [H_{\text{sf}}, \hat{Q}_m] B \rangle\rangle, \end{aligned} \quad \text{E.3b.}$$

$$\begin{aligned} \langle\langle A_m^{(+)} | \tilde{B}_m^{(+)} \rangle\rangle &= -\delta_{m0} \langle\langle A_m^{(+)} | B_1^{(+)} \rangle\rangle + \langle\langle A_m^{(+)} | i\mathcal{L} + \zeta_m^{(+)} | B_m^{(+)} \rangle\rangle \\ &\quad + i \langle\langle A_m^{(+)} | B [H_{\text{sf}}, \hat{Q}_m^\dagger] \rangle\rangle. \end{aligned} \quad \text{E.3c.}$$

Now using the identity $\langle\langle \tilde{A} | B \rangle\rangle = \langle\langle A | \tilde{B} \rangle\rangle$, we have

$$\begin{aligned} \langle\langle \tilde{A} | B \rangle\rangle &= \langle\langle A | i\mathcal{L} + \mathcal{R}_s | B \rangle\rangle \\ &\quad - i \sum_m \{ \langle\langle [H_{\text{sf}}, \hat{Q}_m^\dagger] A_m^{(-)} | B \rangle\rangle + \langle\langle A_m^{(+)} [H_{\text{sf}}, \hat{Q}_m] | B \rangle\rangle \}, \end{aligned} \quad \text{E.4a.}$$

$$\begin{aligned} \langle\langle \tilde{A}_m^{(\pm)} | B_m^{(\pm)} \rangle\rangle &= -\delta_{m1} \langle\langle A_0^{(\pm)} | B_m^{(\pm)} \rangle\rangle + \langle\langle A_m^{(\pm)} | i\mathcal{L} + \zeta_m^{(\pm)} | B_m^{(\pm)} \rangle\rangle \\ &\quad \mp \nu_m^{(\pm)} \langle\langle A | [Q, B_m^{(\pm)}] \rangle\rangle. \end{aligned} \quad \text{E.4b.}$$

Let us first consider Equation E.4a. By using the identity,

$$\langle\langle A | \mathcal{O} | B \rangle\rangle \equiv \text{Tr}[A^\dagger(\mathcal{O}B)] = \text{Tr}[(A^\dagger \mathcal{O})B], \quad \text{E.5.}$$

together with Equation 6.3, we have

$$\begin{aligned}
\{A^\dagger(i\mathcal{L} + \mathcal{R}_s)\}^\dagger &= \{i[A^\dagger, H] + [A^\dagger, Q]\tilde{Q} - \tilde{Q}^\dagger[A^\dagger, Q]\}^\dagger \\
&= i[A, H] - \tilde{Q}^\dagger[A, Q] + [A, Q]\tilde{Q} \\
&= A(i\mathcal{L} + \mathcal{R}_s).
\end{aligned}
\tag{E.6}$$

We thus obtain

$$\tilde{A} = A(i\mathcal{L} + \mathcal{R}_s) + i \sum_m \{[H_{\text{sf}}, \hat{Q}_m^\dagger]A_m^{(-)} + A_m^{(+)}[H_{\text{sf}}, \hat{Q}_m]\}.
\tag{E.7a}$$

Similarly, Equation E.4b gives us

$$\tilde{A}_m^{(\pm)} = -\delta_{m1}A_0^{(\pm)} + A_m^{(\pm)}(i\mathcal{L} + \zeta_m^{(\mp)}) \pm v_m^{(\mp)}[A, Q].
\tag{E.7b}$$

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