

# Hybrid hierarchical equation of motion for open quantum systems

Yuanran Zhu<sup>1,\*</sup>

<sup>a</sup>*Applied Mathematics & Computational Research Division, Lawrence Berkeley National Laboratory,  
Berkeley, CA 94720, USA*

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## Abstract

In this note, we briefly introduce the Hybrid hierarchical equation of motion (Hybrid HEOM) for open quantum systems developed by T. Fay and D. Limmer in [3]. Generally speaking, this approach can be viewed as a natural combination of two commonly used computational methods for open quantum systems. The first approach is the HEOM, which provides an accurate solver for the reduced system density matrix by solving a high-dimensional extended linear system. Another one is the Nakajima-Mori-Zwanzig (NMZ) approach which is a perturbation method and therefore works well for systems that are weakly coupling with the environment.

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## 1. Introduction

A general open quantum system is described by the Hamiltonian  $\mathcal{H} = \mathcal{H}_S + \mathcal{H}_B + \mathcal{H}_I$ , where  $\mathcal{H}_S$  is the system Hamiltonian,  $\mathcal{H}_B$  is the bath, and  $\mathcal{H}_I$  encodes the interaction between the system and bath Hamiltonian. For most applications, we are only concerned with the system part dynamics. Therefore the mainstream computational methods aim to get an effective equation of motion (EOM) for the system part reduced density matrix  $\rho_S(t) = \text{Tr}_B[\rho(t)]$ . Over the years, many computational approaches have been developed:

$$\begin{array}{ccc} \rho(0) & \xrightarrow{\text{Tr}_B\{\cdot\}} & \rho_S(0) \\ \downarrow e^{t\mathcal{L}} & & \downarrow e^{t\mathcal{L}_S} \\ \rho(t) & \xrightarrow{\text{Tr}_B\{\cdot\}} & \rho_S(t) \end{array}$$

1. Embedding methods: The embedding methods aim to find proper approximations to the open system Hamiltonian ( $\mathcal{H} \approx \tilde{\mathcal{H}}$ ) or the Liouville superoperator for the EOM of the density matrix ( $\mathcal{L}_S \approx \tilde{\mathcal{L}}_S$ ). For the Hamiltonian embedding, one often finds a discrete Hamilton  $\tilde{\mathcal{H}}$  that approximates  $\mathcal{H}$  and solves the Von-Neumann-Liouville equation:  $\rho_S(t) \approx \text{Tr}_B[e^{t\tilde{\mathcal{L}}}\rho(0)]$  to get the reduced density matrix. While the Liouvillian embedding operates at a lower level and aims to find an effective superoperator  $\tilde{\mathcal{L}}_S$  and solves for  $\rho_S(t)$  using  $\rho_S(t) \approx e^{t\tilde{\mathcal{L}}_S}\rho_S(0)$ . (See the above commutative diagram)
2. HEOM (See Section 2 for brief introduction)
3. Perturbation method: NMZ equation (See Section 3 for brief introduction)
4. Influence functional approach (This part will be discussed by Gunhee and Erika)
5. ...

As we briefly mentioned in the abstract, the hybrid EOM approach can be viewed as HEOM+NMZ. Fay and Limmer's original paper[3] introduced this method using the superfermion formulation and applied it to a rather complicated chemical system. For simplicity, in this note, we will avoid using the superfermion language and adopt a simple schematic model to explain the main idea of the approach. This note is organized using materials from [3, 4, 1, 2].

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\*Corresponding author  
Email address: yzhu4@lbl.gov (Yuanran Zhu)

## 2. Hierarchical equation of motion: an overview

The HEOM [5] provides a method to simulate the dynamics of the reduced density matrix for a system *linearly* coupled to *harmonic* (quadratic) baths. The linearity and harmonicity conditions are essential for the HEOM to work. Let us consider a schematic Hamiltonian:

$$\mathcal{H} = \mathcal{H}_S + \underbrace{\sum_{j=1}^L \mathcal{H}_{B,j}}_{\mathcal{H}_B} + \underbrace{\sum_{j=1}^L \mathcal{V}_j \mathcal{B}_j}_{\mathcal{H}_I}, \quad (1)$$

where  $\mathcal{H}_S$  is the system Hamiltonian,  $\mathcal{V}_j$  are system operators, and  $\mathcal{H}_{B,j} = \sum_{\alpha=1}^{N_j} \omega_{j\alpha} a_{j\alpha}^\dagger a_{j\alpha}$  is the Hamiltonian of harmonic bath  $j$ . The bath displacement operator  $\mathcal{B}_j$  is defined as  $\mathcal{B}_j = \sum_{\alpha}^{N_j} g_{j\alpha} (a_{j\alpha}^\dagger + a_{j\alpha})$ . It can be proved that (using Dyson's series and Wick's theorem) that the dynamics of the reduced density matrix  $\rho_S(t) = \text{Tr}_B[\rho(t)]$  for a system evolving from separable initial density  $\rho_0 = \rho_S(0) \otimes e^{-\beta \mathcal{H}_B}$  is entirely determined by the hybridization function (for bosonic bath):

$$C_j(t) = \frac{1}{2\pi} \int_0^{+\infty} J_j(\omega) [n(\omega) e^{i\omega t} + (n(\omega) + 1) e^{-i\omega t}],$$

where  $n(\omega) = \frac{1}{e^{\beta\omega} - 1}$  is the Bose-Einstein distribution, and

$$J_j(\omega) = 2\pi \sum_{\alpha}^{N_j} |g_{j\alpha}|^2 \delta(\omega - \omega_{j\alpha})$$

is a spectral function that encodes the excitation frequency  $\omega_{j\alpha}$  of the bath and its interaction strength  $g_{j\alpha}$  with the system. For a typical open quantum system,  $J_j(\omega)$  is normally chosen to be a continuous function of  $\omega$  by taking  $N_j \rightarrow \infty$ . Physically, this means that the bath Hamiltonian has continuous excitation/spectrum. Suppose the hybridization function can be approximated using summation of exponential terms:

$$C_j(t) \approx \sum_{k=1}^K a_{jk} e^{-v_{jk} t},$$

then the reduced density matrix  $\rho_S(t)$  can be obtained by solving a linear ODE for a hierarchy of auxiliary density operators (ADOs)  $\rho_{\mathbf{n}}(t)$ . This equation is known as the hierarchical equation of motion (HEOM) for open quantum systems and it admits an explicit form:

$$\begin{aligned} \frac{d}{dt} \rho_{\mathbf{n}}(t) = & -i[\mathcal{H}_S, \rho_{\mathbf{n}}(t)] - \gamma_{\mathbf{n}} \rho_{\mathbf{n}}(t) \\ & - i \sum_{jk} \sqrt{(n_{jk} + 1) |a_{jk}|} [\mathcal{V}_j, \rho_{\mathbf{n}_{jk}^+}] - i \sum_{jk} \sqrt{n_{jk} |a_{jk}|} (a_{jk} \mathcal{V}_j \rho_{\mathbf{n}_{jk}^-} - a_{jk}^* \rho_{\mathbf{n}_{jk}^-} \mathcal{V}_j), \end{aligned}$$

where  $\mathbf{n} = (n_{1,1}, n_{1,2}, \dots, n_{j,k}, \dots, n_{L,K})$  is a multi-index that specifies the excitation level of mode  $k$  for  $j$ -th bath for a given ADO  $\rho_{\mathbf{n}}(t)$ .  $\mathbf{n}_{jk}^{\pm} = (n_{1,1}, n_{1,2}, \dots, n_{j,k} \pm 1, \dots, n_{L,K})$ .  $\gamma_{\mathbf{n}} = \sum_{jk} n_{jk} v_{jk}$ . The reduced system density matrix  $\rho_S(t) = \rho_{\mathbf{0}}(t)$ . The initial condition for the ADOs are given by  $\rho_{\mathbf{n}}(0) = \mathbf{0}$ .

The HEOM is a high-dimensional linear ODE. When  $\mathcal{H}_S$  is the Hamiltonian for a  $M$ -level system, each ADO  $\rho_{\mathbf{n}}(t) \in \mathbb{C}^{M \times M}$ . For a quantum system with  $L$  baths and each bath-system hybridization function is represented by  $K$  exponentials, a HEOM with  $N$  layers will contain  $N_{ADO} = (LK + N)! / (LK! N!)$  number of ADOs since the multi-index  $\mathbf{n}$  will go over all non-negative partition of the integer up to  $N$ <sup>1</sup>. As an example,  $L = 8, K = 5, N = 5, N_{ADO} \sim 10^6$ . The computational cost of solving HEOM for a multi-level

<sup>1</sup>Suppose  $L = 1, K = 2, N = 2$ , then  $\mathbf{n} = (n_1, n_2)$  and  $n_1 + n_2 \leq 2$ , for all possible  $n_1, n_2 \geq 0$ . This gives rise to  $6 = 4! / (2! 2!)$  ADOs:  $\rho_{00}, \rho_{10}, \rho_{01}, \rho_{11}, \rho_{02}, \rho_{20}$ .

system is high, but normally it will give a rather accurate approximation for  $\rho_S(t)$ . For later convenience, we could rewrite the HEOM in an abstract form:

$$\frac{d}{dt}\rho_{\mathbf{n}}(t) = \mathcal{L}\rho_{\mathbf{n}}(t)$$

where  $\mathcal{L}$  can be called the HEOM-Liouville superoperator.

### 3. Nakajima-Mori-Zwanizg equation and its perturbative expansion

The Nakajima-Mori-Zwanizg equation (NMZ) uses perturbation theory to calculate  $\rho_S(t)$ . The computational cost is much lower than HEOM since the NMZ equation for  $\rho_S(t)$  is an integro-differential equation of the dimension  $M \times M$ . However, as a perturbation method, it is an efficient and accurate solver only when the coupling between the system and bath is weak (For Hamiltonian (1), this means  $g_{jk} \ll 1$ ). The most commonly used perturbation orders are  $N = 2, 4$ .

#### 3.1. A simple example: Dyson-Duhamel's principle

Consider a linear system:

$$\frac{d}{dt} \begin{bmatrix} \mathbf{x} \\ \mathbf{y} \end{bmatrix} = \begin{bmatrix} A_{11} & A_{12} \\ A_{21} & A_{22} \end{bmatrix} \begin{bmatrix} \mathbf{x} \\ \mathbf{y} \end{bmatrix},$$

where  $\mathbf{x} \in \mathbb{R}^n, \mathbf{y} \in \mathbb{R}^m$ ,  $A_{11} \in \mathbb{R}^{n \times n}, A_{12} \in \mathbb{R}^{n \times m}, A_{21} \in \mathbb{R}^{m \times n}, A_{22} \in \mathbb{R}^{m \times m}$ . The Dyson-Duhamel's principle implies that we could get a closed equation for  $\mathbf{x}(t)$ :

$$\frac{d}{dt}\mathbf{x} = A_{11}\mathbf{x} + \underbrace{\int_0^t A_{12}e^{(t-s)A_{22}}A_{21} \mathbf{x}(s)ds}_{K(t-s)} + \underbrace{A_{12}e^{tA_{22}}\mathbf{y}(0)}_{f(t)}, \quad (2)$$

and for time-dependent system, we have

$$\frac{d}{dt}\mathbf{x} = A_{11}(t)\mathbf{x} + \underbrace{\int_0^t A_{12}(t)e^{\int_s^t A_{22}(\tau)d\tau}A_{21}(s) \mathbf{x}(s)ds}_{K(t,s)} + \underbrace{A_{12}(t)e^{\int_0^t A_{22}(\tau)d\tau}\mathbf{y}(0)}_{f(t)}. \quad (3)$$

In (2)-(3),  $K(t, s), K(t - s) \in \mathbb{R}^{n \times n}$  are known as the memory kernel.  $f(t)$  is often called the "fluctuation force" for historical reasons. This EOM is a reduced-order model for  $\mathbf{x}(t)$ . The NMZ equation is an operator version of Dyson-Duhamel's principle. In most applications,  $e^{(t-s)A_{22}}$  and  $e^{\int_s^t A_{22}(\tau)d\tau}$  will be rather complicated evolution operators. Hence, one often chooses to use series expansion to approximate such operators, leading to a perturbative expansion for the NMZ equation. For instance, if  $e^{\int_s^t A_{22}(\tau)d\tau} \approx 1$  and  $\mathbf{y}(0) = 0$ , then we get the lowest order approximation for (3):

$$\frac{d}{dt}\mathbf{x} \approx A_{11}(t)\mathbf{x}(t) + \int_0^t A_{12}(t)A_{21}(s)\mathbf{x}(s)ds \quad (4)$$

If further using Markovian approximation, we arrive at the "Markovian Master equation" for  $\mathbf{x}(t)$ :

$$\frac{d}{dt}\mathbf{x} \approx A_{11}(t)\mathbf{x}(t) + \underbrace{\int_0^\infty A_{12}(t)A_{21}(s)ds}_{R_{11}(t)}\mathbf{x}(t) \quad (5)$$

This Markovian master equation is obtained using the zeroth order approximation to evolution operator  $e^{\int_s^t A_{22}(\tau)d\tau}$ . However, it is called the 2nd order perturbation theory in the literature since  $A_{12}(t)A_{21}(s)$  is normally of the order  $O(g^2)$  for  $g \ll 1$ .

### 3.2. NMZ for reduced density matrix

The Von-Neumann-Liouville equation for the full density matrix of Hamiltonian system (1) is given by:

$$\frac{d}{dt}\rho(t) = -i[H, \rho] = \mathcal{L}\rho, \quad \mathcal{L}(\cdot) = -i[H, (\cdot)]$$

The NMZ can be derived from the following Dyson's identity:

$$\frac{d}{dt}\mathcal{P}e^{t\mathcal{L}} = \mathcal{P}\mathcal{L}\mathcal{P}e^{t\mathcal{L}} + \mathcal{P}\mathcal{L}e^{t\mathcal{Q}\mathcal{L}}\mathcal{Q} + \int_0^t ds \mathcal{P}\mathcal{L}e^{(t-s)\mathcal{Q}\mathcal{L}}\mathcal{Q}\mathcal{L}\mathcal{P}e^{s\mathcal{L}} \quad (6)$$

where  $\mathcal{P}, \mathcal{Q} = \mathcal{I} - \mathcal{P}$  are a pair of projection (super)operators satisfying  $\mathcal{P}^2 = \mathcal{P}, \mathcal{Q}^2 = \mathcal{Q}$ . Applying this operator identity into the initial density  $\rho(0) = \rho_S(0) \otimes \rho_B$  and define  $\mathcal{P} := \text{Tr}_B[\cdot] \otimes \rho_B$ , we could get the following *formally exact* EOM for  $\rho_S(t) = \text{Tr}_B[\rho(t)]$ :

$$\begin{aligned} \frac{d}{dt}\mathcal{P}\rho(t) &= \mathcal{P}\mathcal{L}\mathcal{P}\rho(t) + \mathcal{P}\mathcal{L}e^{t\mathcal{Q}\mathcal{L}}\mathcal{Q}\rho(0) + \int_0^t ds \mathcal{P}\mathcal{L}e^{(t-s)\mathcal{Q}\mathcal{L}}\mathcal{Q}\mathcal{L}\mathcal{P}\rho(s) \\ \Rightarrow \frac{d}{dt}\rho_S(t) &= \mathcal{P}\mathcal{L}\rho_S(t) + \int_0^t ds \mathcal{P}\mathcal{L}e^{(t-s)\mathcal{Q}\mathcal{L}}\mathcal{Q}\mathcal{L}\rho_S(s) \end{aligned} \quad (7)$$

Here we slightly abuse notation and use  $\mathcal{P}\mathcal{L}\mathcal{P}\rho(t) = \text{Tr}_B[\mathcal{L}(\rho_S(t) \otimes \rho_B)] \otimes \rho_B$  to represent  $\text{Tr}_B[\mathcal{L}(\rho_S(t) \otimes \rho_B)]$  since  $\rho_B$  in two sides of the equation will be canceled out. We can also derive the NMZ in the interaction picture. See the details in Appendix A. Here we consider a slightly different formulation which will be used later in the hybrid HEOM. We choose  $\mathcal{H}_0 = \mathcal{H}_S + \mathcal{H}_B$  and define  $\mathcal{L}_0 = -i[\mathcal{H}_0, \cdot]$ ,  $\mathcal{L}_I = \mathcal{L} - \mathcal{L}_0$ , it is easy to verify that

$$e^{t\mathcal{L}} = e^{t\mathcal{L}_0} e^{\int_0^t \mathcal{L}_I(\tau) d\tau}, \quad e^{t\mathcal{Q}\mathcal{L}} = e^{t\mathcal{Q}\mathcal{L}_0} e^{\int_0^t \mathcal{Q}\mathcal{L}_I(\tau) d\tau} \quad (8)$$

where

$$\mathcal{L}_I(t) = e^{-t\mathcal{L}_0} \mathcal{L}_I e^{t\mathcal{L}_0} = -i[\mathcal{H}_I(t), \cdot], \quad \mathcal{H}_I(t) = e^{i\mathcal{H}_0 t} \mathcal{H}_I e^{-i\mathcal{H}_0 t}$$

Using (8),  $e^{\int_s^t \mathcal{Q}\mathcal{L}_I(\tau) d\tau} \approx \mathcal{I}$  in (7) and the Markovian approximation, we can get the quantum Master equation for  $\rho_S(t)$ :

$$\begin{aligned} \frac{d}{dt}\rho_S(t) &\approx \mathcal{P}\mathcal{L}\rho_S(t) + \int_0^t ds \mathcal{P}\mathcal{L}e^{(t-s)\mathcal{Q}\mathcal{L}_0}\mathcal{Q}\mathcal{L}\rho_S(s) \\ &\approx \mathcal{P}\mathcal{L}\rho_S(t) + \int_0^\infty \mathcal{P}\mathcal{L}e^{s\mathcal{Q}\mathcal{L}_0}\mathcal{Q}\mathcal{L}ds\rho_S(t) \end{aligned} \quad (9)$$

The perturbation theory works when  $\mathcal{H}_I \ll 1$ .

*Remark.* This quantum Master equation is *not* in the Lindblad form. However, further applying the rotating wave approximation, we can get the Lindblad equation. See details in [2], Page 127.

### 3.3. NMZ for quantum state population

We also use the NMZ framework to get the evolution equation for the quantum state population, this could further reduce the dimensionality of the NMZ equation to  $M$  since only the diagonal of the density matrix will be considered. Define projection operator  $\mathcal{P} = \sum_A \mathcal{P}_A$ , where  $\mathcal{P}_A = \mathbf{P}_A \text{Tr}_B[\cdot] \mathbf{P}_A \otimes \rho_B$ ,  $\mathbf{P}_A = |A\rangle\langle A|$ . Then the population of a system Hamiltonian eigenstate  $|A\rangle$  can be calculated as  $\sigma_A(t) = \langle A|\rho_S(t)|A\rangle = \text{Tr}_B[\langle A|\mathcal{P}_A\rho(t)|A\rangle]$ . Using this projection in the above NMZ equation, we can get the formally exact EOM for the state population:

$$\frac{d}{dt}\sigma_A(t) = L_A\sigma_A(t) + \sum_B \int_0^t K_{AB}(t-s)\sigma_B(s)ds \quad (10)$$

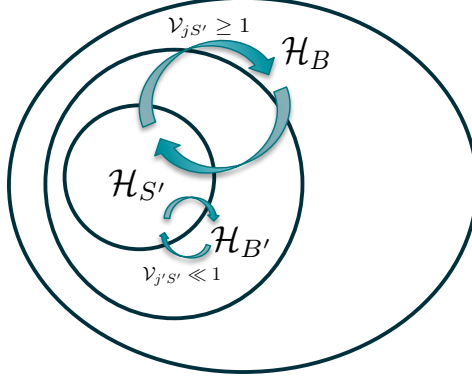


Figure 1: Schematic illustration for a typical open quantum system where the hybrid HEOM approach applies. The interaction between  $\mathcal{H}_B$  and  $\mathcal{H}_{S'}$  is strong ( $V_{jS'} \geq 1$ ) therefore this fold of interaction is handled by HEOM. On the other hand, the interaction between  $\mathcal{H}_{B'}$  and  $\mathcal{H}_{S'}$  is weak ( $V_{j'S'} \ll 1$ ) therefore this fold of interaction can be handled by perturbation method such as the NMZ.

where  $L_A, K_{AB}(t-s)$  are the matrix representation of the corresponding superoperators. Following the same procedure, we can get the quantum Master equation:

$$\frac{d}{dt}\sigma_A(t) = L_A\sigma_A(t) + \underbrace{\sum_B \int_0^\infty K_{AB}(s)ds}_{R_{AB}}\sigma_B(t) \quad (11)$$

Note that for this case, we also need a small tunneling amplitude between different quantum states in  $\mathcal{H}_S$  for the perturbation theory to work [3].

#### 4. Hybrid HEOM

The NMZ equation applies to any evolution operator generated by the Liouville (super)operator  $\mathcal{L}$ . Therefore it is quite easy to combine the NMZ equation with HEOM to get the hybrid HEOM [3] and one only needs to replace the Von-Neumann Liouville superoperator used in the previous section with the HEOM-Liouville superoperator introduced in Section 2. The hybrid HEOM is particularly useful for a hierarchical quantum system  $\mathcal{H} = \mathcal{H}_S + \mathcal{H}_B + \mathcal{H}_I$ , where  $\mathcal{H}_S$  is also an open quantum system Hamiltonian  $\mathcal{H}_S = \mathcal{H}_{S'} + \mathcal{H}_{B'} + \mathcal{H}_{I'}$  (See Figure 1). If one aims to get  $\rho_{S'}(t) = \text{Tr}_{B'}[\rho_S(t)] = \text{Tr}_{B'}[\text{Tr}_B[\rho(t)]]$ , directly applying HEOM to get  $\rho_S(t)$  will be computationally expensive since the eigenspace for  $\mathcal{H}_S$  is high-dimensional. However, if we know  $\mathcal{H}_{S'}$  has a weak interaction with the bath  $\mathcal{H}_{B'}$ , we could apply the perturbative NMZ equation to HEOM and get the evolution equation for  $\rho_{S'}(t)$ . This is the essence of the hybrid HEOM approach.

For the hierarchical open quantum system evolving from the initial density  $\rho_{S'}(0) \otimes \rho_{B'} \otimes \rho_B$ , suppose we use HEOM to get the evolution equation for ADOs  $\rho_{S,\mathbf{n}}(t)$ . Using the Liouville superoperator format, this EOM can be written as :

$$\frac{d}{dt}\rho_{S,\mathbf{n}}(t) = \mathcal{L}\rho_{S,\mathbf{n}}(t).$$

Define a projection superoperator  $\mathcal{P} := \text{Tr}_{B'}[\cdot] \otimes \rho_{B'}$  and then apply the Dyson's identity (6) into  $\rho_{S,\mathbf{n}}(0)$ , we could get the formally exact NMZ equation for  $\rho_{S',\mathbf{n}}(t) = \text{Tr}_{B'}[\rho_{S,\mathbf{n}}(t)]$ :

$$\frac{d}{dt}\rho_{S',\mathbf{n}}(t) = \mathcal{P}\mathcal{L}\rho_{S',\mathbf{n}}(t) + \int_0^t ds \mathcal{P}\mathcal{L}e^{(t-s)\mathcal{Q}\mathcal{L}}\mathcal{Q}\mathcal{L}\rho_{S',\mathbf{n}}(s). \quad (12)$$

Its Markovian approximation leads to

$$\frac{d}{dt}\rho_{S',\mathbf{n}}(t) = \mathcal{P}\mathcal{L}\rho_{S',\mathbf{n}}(t) + \underbrace{\int_0^\infty \mathcal{P}\mathcal{L}e^{s\mathcal{Q}\mathcal{L}_0}\mathcal{Q}\mathcal{L}ds}_{\mathcal{R}_{S',\mathbf{n}}}\rho_{S',\mathbf{n}}(t). \quad (13)$$

Eqn (13) (more precisely, its quantum state population version) is called the hybrid HEOM in [3]. This is a linear ODE with the dimensionality  $M'^2(LK + N)!/(LK!N!)$  (or  $M'(LK + N)!/(LK!N!)$  for the quantum state population version), where  $M'$  is the dimension of the eigenspace for Hamiltonian  $\mathcal{H}_{S'}$ . In comparison, if directly applying the HEOM to the  $\mathcal{H}_S$ , the resulting HEOM has dimensionality  $(M'D')^2(LK + N)!/(LK!N!)$ , where  $D'$  is the dimension of the eigenspace for  $\mathcal{H}_{B'}$ . It takes several more steps to further simplify/approximate the superoperator  $\mathcal{R}_{S',\mathbf{n}}$ . To get a basic understanding of what Eqn (13) looks like. We explicitly calculate the first term on the RHS. Suppose  $\mathcal{V}_j = \mathcal{V}_{jS'}$ , this means that  $\mathcal{H}_B$  only interact with  $\mathcal{H}_{S'}$ . Then  $\mathcal{P}\mathcal{L}\rho_{S',\mathbf{n}}(t)$  is given by:

$$\begin{aligned} \mathcal{P}\mathcal{L}\rho_{S',\mathbf{n}}(t) = & -i[\mathcal{H}_{S'}, \rho_{S',\mathbf{n}}(t)] - \gamma_{\mathbf{n}}\rho_{S',\mathbf{n}}(t) \\ & - i \sum_{jk} \sqrt{(n_{jk} + 1)} |a_{jk}| [\mathcal{V}_{jS'}, \rho_{S',\mathbf{n}_{jk}^+}] - i \sum_{jk} \sqrt{n_{jk}} |a_{jk}| (a_{jk} \mathcal{V}_{jS'} \rho_{S',\mathbf{n}_{jk}^-} - a_{jk}^* \rho_{S',\mathbf{n}_{jk}^-} \mathcal{V}_{jS'}). \end{aligned}$$

## Appendix A. NMZ in the interaction picture

Choose  $\mathcal{H}_0 = \mathcal{H}_S + \mathcal{H}_B$  and define the interaction picture density matrix as  $\rho_I(t) = e^{t\mathcal{L}_0}\rho(t) = e^{it\mathcal{H}_0}\rho(t)e^{-it\mathcal{H}_0}$ , and  $\mathcal{H}_I(t) = e^{it\mathcal{H}_0}\mathcal{H}_Ie^{-it\mathcal{H}_0}$ , then we have:

$$\frac{d}{dt}\rho_I(t) = -i[\mathcal{H}_I(t), \rho_I(t)] = \mathcal{L}_I(t)\rho_I(t), \quad \mathcal{L}_I(t)(\cdot) = -i[\mathcal{H}_I(t)(\cdot), \cdot] = e^{-t\mathcal{L}_0}\mathcal{L}_Ie^{t\mathcal{L}_0}.$$

The NMZ can be derived from the following Dyson's identity:

$$\frac{d}{dt}\mathcal{P}\mathcal{U}(t, 0) = \mathcal{P}\mathcal{L}_I(t)\mathcal{P}\mathcal{U}(t, 0) + \mathcal{P}\mathcal{L}_I(t)U_Q(t, 0)\mathcal{Q} + \int_0^t ds \mathcal{P}\mathcal{L}_I(t)U_Q(t, s)\mathcal{Q}\mathcal{L}_I(s)\mathcal{P}\mathcal{U}(s, 0),$$

where  $\mathcal{P}, \mathcal{Q} = \mathcal{I} - \mathcal{P}$  are a pair of projection (super)operators satisfying  $\mathcal{P}^2 = \mathcal{P}, \mathcal{Q}^2 = \mathcal{Q}, \mathcal{U}(t, s) = e^{\int_s^t \mathcal{L}_I(\tau)d\tau}$  and  $U_Q(t, s) = e^{\int_s^t \mathcal{Q}\mathcal{L}_I(\tau)d\tau}$ . Applying this operator identity into the initial density  $\rho(0) = \rho_S(0) \otimes \rho_B$  and define  $\mathcal{P} = \rho_B \text{Tr}_B[\cdot]$ , we could get the EOM for  $\rho_{SI}(t) = \text{Tr}_B[\rho_I(t)]$ :

$$\begin{aligned} \frac{d}{dt}\mathcal{P}\rho_I(t) &= \mathcal{P}\mathcal{L}_I(t)\mathcal{P}\rho_I(t) + \mathcal{P}\mathcal{L}_I(t)U_Q(t, 0)\mathcal{Q}\rho(0) + \int_0^t ds \mathcal{P}\mathcal{L}_I(t)U_Q(t, s)\mathcal{Q}\mathcal{L}_I(s)\mathcal{P}\rho_I(s) \\ \Rightarrow \frac{d}{dt}\rho_{SI}(t) &= \mathcal{P}\mathcal{L}_I(t)\mathcal{P}\rho_{SI}(t) + \int_0^t ds \mathcal{P}\mathcal{L}_I(t)U_Q(t, s)\mathcal{Q}\mathcal{L}_I(s)\mathcal{P}\rho_{SI}(s). \end{aligned}$$

Using the lowest order approximation  $U_Q(t, s) \approx \mathcal{I}$  and Markovian approximation, we arrive at the Markovian quantum Master equation for the interaction picture density matrix:

$$\frac{d}{dt}\rho_{SI}(t) = \mathcal{P}\mathcal{L}_I(t)\mathcal{P}\rho_{SI}(t) + \int_0^\infty \mathcal{P}\mathcal{L}_I(t)\mathcal{Q}\mathcal{L}_I(s)ds\mathcal{P}\rho_{SI}(t).$$

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