DIFFERENTIAL EQUATION BASED PATH INTEGRAL FOR SYSTEM-BATH DYNAMICS

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ABSTRACT. We propose the differential equation based path integral (DEBPI) method to simulate the realtime evolution of open quantum systems. In this method, a system of partial differential equations is derived based on the continuation of a classical numerical method called iterative quasi-adiabatic propagator path integral (i-QuAPI). While the resulting system has infinite equations, we introduce a reasonable closure to obtain a series of finite systems. New numerical schemes can be derived by discretizing these differential equations. It is numerically verified that in certain cases, by selecting appropriate systems and applying suitable numerical schemes, the memory cost required in the i-QuAPI method can be significantly reduced.

1. INTRODUCTION

In physics, an open quantum system refers to a quantum system coupled with an external environment (bath). In reality, no quantum system can be absolutely isolated, and therefore the study of open quantum system is of great importance and nowadays has wide applications in the fields including quantum optics [3], chemical physics [27, 9], quantum information [21] and even social sciences [8]. In general, for an open quantum system, the large number of degrees of freedom in the bath prevents us from directly computing the density matrix of the coupled system. To carry out simulations, the bath is usually assumed to be some analytically solvable models such as harmonic oscillators so that one can represent the quantum dynamics using path integrals, and the influence of the bath on the system of interest can be reduced to an influence functional [5].

Despite such simplification, the difficulty introduced by the system-bath coupling still exists because the influence functional couples the states of the system at any two time points, leading to a non-Markovian process. Such process is often formulated using an integro-differential equation (known as Nakajima-Zwanzig equation), where the effect of the bath is expressed as a memory kernel, represented by the Nakajima-Zwanzig operator [20, 30]. The numerical methods based on the computation of path integrals, such as quasiadiabatic propagator path integral (QuAPI) [12] and hierarchical equations of motion (HEOM) [25, 24] methods, require significant memory cost to store the contributions from different paths. To reduce the storage requirement, one idea is to make use of the decaying property of the influence functional to develop iterative schemes. One typical method is the iterative QuAPI (i-QuAPI) method [13, 14], which truncates the influence functional to achieve finite memory length, so that partial summation can be carried out in the path integral. Other approaches based on this idea include the blip-summed method [16, 17], small matrix decomposition of the path integral (SMatPI) method [18], and important path sampling [23] design algorithms, which explore extra the numerical sparsity of the problem to further reduce the memory cost. Some stochastic methods, such as diagrammatic quantum Monte Carlo (dQMC) method [28] and inchworm Monte Carlo method [4, 1, 29, 2], apply stochastic approaches to estimate path integrals. These methods no longer suffer from the curse of memory cost. However, numerical sign problem may appear for long-time simulations [11, 2]. There are also some modeling techniques to simplify the Nakajima-Zwanzig equation, among which the Lindblad equation [10, 6] considers the weak coupling limit, so that the memory effect can be ignored and the master equation becomes Markovian. Other approaches based on computable master equations include the post-Markovian master equation [22] and the generalized quantum master equation [7].

Despite the fast progress in the numerical computation of open quantum systems, the i-QuAPI method remains as one of the fundamental and reliable methods, and is often used to compute reference solutions in

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the development of other methods. Since the strongest limitation of i-QuAPI is its memory cost, which grows exponentially with $1/\Delta t$, the present paper follows the basic idea of the i-QuAPI method and tries to break such exponential relationship by developing the differential equation based path integral (DEBPI) method. The DEBPI method formulates a system of differential equations such that i-QuAPI can be considered as its numerical scheme. Afterwards, we can re-design the numerical scheme by discretizing the PDE system. It is numerically verified that in certain cases, the memory cost can be significantly reduced.

Throughout this paper, we will mainly focus on a simple open quantum system consisting of a flipping spin and a thermal bath. Despite the simple setting of the system, it contains all the difficulties related to the system-bath coupling in general open quantum systems. We refer the authors to [26] for some detailed discussion on this model.

For the rest of this paper, we present the derivation of DEBPI method and some numerical tests. Section 2 is a brief introduction to the spin-boson model and the i-QuAPI method. In order to derive the continuous formulation for i-QuAPI method, we give a representation of the continuation of path segments and introduce notations for the relative quantities in Section 3. With all the preparation in Sections 2 and 3, we derive a governing partial differential equation system and discuss the required boundary condition in Section 4. In addition, as the partial differential equation system includes infinite equations, we also provide the idea of dimensional truncation and the closure of the system in Section 4. In Section 5, we carry out some numerical experiments based on finite difference discretization method for DEBPI method and compare our results with i-QuAPI method. In Section 6, we outline the application of the idea in other finite state models and give a simple conclusion. The derivation of i-QuAPI method is given in the appendix for reference.

2. Spin-boson model and iterative QuAPI method

In a quantum system with Hamiltonian H, the density matrix $\rho(t)$ satisfies the von Neumann equation

$$i\hbar \frac{\mathrm{d}}{\mathrm{d}t}\rho(t) = H\rho(t) - \rho(t)H_{t}$$

where i is the imaginary unit, and \hbar is the reduced Planck's constant. For simplicity, \hbar is set to be 1 in this paper. The solution to the von Neumann equation can be formally written as $\rho(t) = e^{-iHt}\rho(0)e^{iHt}$.

For an open quantum system, the operators are defined on the tensor product space $\mathcal{H}_s \otimes \mathcal{H}_b$, where \mathcal{H}_s and \mathcal{H}_b represent the Hilbert spaces for the system and the bath, respectively. In general, the Hamiltonian H has the form

(1)
$$H = H_s \otimes \mathrm{id}_b + \mathrm{id}_s \otimes H_b + H_{sb}$$

where $\mathrm{id}_s, \mathrm{id}_b$ are the identity operators on \mathcal{H}_s and \mathcal{H}_b , respectively. The operators H_s and H_b are the Hamiltonian operators for the system and the bath without coupling, and the last term H_{sb} describes the system-bath interaction, which lead to entanglement between system and bath. For simplicity, below we focus on the study of a specific case of the open quantum system, where the system contains a single spin. The model and the iterative QuAPI method will be introduced in the following subsections.

2.1. **Spin-boson model.** A fundamental example of open quantum systems is the spin-boson model where the Hilbert spaces are

$$\mathcal{H}_s = \operatorname{span}\{|-1\rangle, |+1\rangle\}, \quad \mathcal{H}_b = \bigotimes_j (L^2(\mathbb{R}^3))$$

with $L^2(\mathbb{R}^3)$ being the L^2 space over \mathbb{R}^3 and j is the index of harmonic oscillators. In spin-boson system, there are two energy levels in the energy set $S = \{-1, +1\}$. In such a system, the Hamiltonians in (1) can be represented by

(2)
$$H_s = \epsilon \hat{\sigma}_z + \Delta \hat{\sigma}_x, \quad H_b = \sum_j \frac{1}{2} (\hat{p}_j^2 + \omega_j^2 \hat{q}_j^2), \quad H_{sb} = \hat{\sigma}_z \otimes \left(\sum_j c_j \hat{q}_j \right)$$

where $\hat{\sigma}_x, \hat{\sigma}_z$ are Pauli matrices, and \hat{p}_j and \hat{q}_j are momentum operator and position operator of the *j*-th harmonic oscillator, respectively. ϵ represents the energy difference between two spin states and Δ is the frequency of the spin flipping. The parameter ω_j is the frequency of the *j*-th harmonic oscillator while c_j is the coupling intensity between the spin and the *j*-th harmonic oscillator.

For convenience, it is supposed that the system and bath are not coupled initially, which can be expressed mathematically by

$$\rho(0) = \rho_s(0) \otimes \rho_b(0)$$

with ρ_s and ρ_b representing the density matrices of the system and the bath, respectively. The reduced density matrix, which is the density matrix of the system, can be represented by the partial trace of the whole density matrix:

$$\rho_s(t) = \operatorname{tr}_b \rho(t),$$

where tr_b is the partial trace operator with respect to \mathcal{H}_b . In the spin-boson model, this reduced density matrix ρ_s is essentially a 2 × 2 matrix. The algorithm discussed in this paper can be generalized to more complicated model. However, in this paper, only spin-boson model is involved and there is a simple discussion about other finite state models in Section 6.

2.2. Iterative QuAPI method. To compute the reduced density matrix in a spin-boson model, the iterative quasi-adiabatic propagator path integral (i-QuAPI) method is developed in [13, 14]. It is assumed that the density matrix has a finite memory length T although it is non-Markovian. Then we choose the time step Δt such that $\Delta k := T/\Delta t$ is an integer. The density matrix can be obtained through computing the maps $A_l : S^{2\Delta k} \to \mathbb{C}$ iteratively:

(3)
$$A_0(S_{\Delta k-1}, \cdots, S_0) := \prod_{k_1=0}^{\Delta k-1} \prod_{k_2=0}^{k_1} I(S_{k_1}, S_{k_2}) \left\langle s_0^+ \big| \rho_s(0) \big| s_0^- \right\rangle;$$

(4)
$$\Lambda(S_k, \cdots, S_{k-\Delta k}) := \prod_{m=0}^{\Delta k} I(S_k, S_{k-m})$$

(5)
$$A_{k-\Delta k+1}(S_k, \cdots, S_{k-\Delta k+1}) := \sum_{S_{k-\Delta k} \in S^2} \Lambda(S_k, \cdots, S_{k-\Delta k}) A_{k-\Delta k}(S_{k-1}, \cdots, S_{k-\Delta k})$$
for $k = \Delta k, \cdots, N-1$.

where $S_j = (s_j^+, s_j^-)$ represents the states on both branches of the path and $I(S_j, S_{j'})$ is defined to be

(6)
$$I(S_{j}, S_{j'}) = \begin{cases} e^{-(s_{j}^{+} - s_{j}^{-})(\eta_{j,j'}s_{j'}^{+} - \eta_{j,j'}^{*}s_{j'}^{-})}, \quad j - j' \neq 1 \\ \langle s_{j}^{+} | e^{-iH_{0}\Delta t} | s_{j-1}^{+} \rangle \langle s_{j-1}^{-} | e^{iH_{0}\Delta t} | s_{j}^{-} \rangle e^{-(s_{j}^{+} - s_{j}^{-})(\eta_{j,j'}s_{j'}^{+} - \eta_{j,j'}^{*}s_{j'}^{-})}, \quad j - j' = 1 \end{cases}$$

where η^* is the complex conjugate of η . The complex function η has different forms for different j and j'[19]. For example, if only positive frequencies are considered, when 0 < j' < j < N,

(7)
$$\eta_{j,j'} = \frac{4}{\pi} \int_0^\infty d\omega \frac{J(\omega)}{\omega^2} \sin^2\left(\frac{\omega\Delta t}{2}\right) \left(\coth\left(\frac{\beta\omega}{2}\right) \cos(\omega\Delta t(k-k')) - i\sin(\omega\Delta t(k-k')) \right).$$

In the expression of η ,

$$J(\omega) = \frac{\pi}{2} \sum_{j} \frac{c_j^2}{m_j \omega_j} \delta(\omega - \omega_j)$$

and δ is the Dirac delta function. The derivation of the i-QuAPI method can be found in the appendix. With the values of $A_{k-\Delta k}(S_k, \dots, S_{k-\Delta k})$, the entries in the reduced density matrix can be computed by

(8)
$$\rho_s(N\Delta t) = \sum_{S_{N-\Delta k+1},\cdots,S_N \in \mathcal{S}} A_{N-\Delta k}(S_N,\cdots,S_{N-\Delta k+1}) \left| s_N^+ \right\rangle \left\langle s_N^- \right|$$

The system-bath coupling leads to a non-Markovian process where the evolution of the system density matrix depends on the complete history, and the intensity of the dependence is determined by the discrete bath response function $\eta_{j,j'}$. The method of iterative quasi-adiabatic propagator path integral (i-QuAPI), however, makes use of the decaying property of the discrete bath response function $\eta_{j,j'}$ and truncate the memory length to a fixed and finite time T, so that the evolution is similar to the time-delay systems, making the simulation more feasible.

The major obstacle of this method is the memory constraint. In each step, there are in total $2^{2\Delta k}$ different values of A_l to be stored in the memory. As Δk is defined to be $T/\Delta t$, and T is fixed according to the problem itself, the restriction on Δk directly results in a lower bound of Δt . The contradiction is that

for more accurate results, smaller Δt should be chosen, which is prohibited by the memory. For example, if the truncation time T is 1.5 and the time step Δt is chosen to be 0.1, then $\Delta k = 15$ and the memory cost is approximately $2^{2\times15} \times (16 \text{ bytes}) = 16\text{GB}$, where "16 bytes" stands for the size of a double-precision complex number. However, if a smaller time step, for example, $\Delta t = 0.075$, is required, the memory cost will become $2^{2\times20} \times (16 \text{ bytes}) = 16\text{TB}$, which is beyond the capacity of most workstations.

3. Continuation of the Path Segments

In order to cut off the memory cost, the most straightforward idea is to reduce the number of path segments that need to be stored. Equation (6) shows that when the flipping rate Δ is small, the path segments with many spin flips may have little contribution to the density matrix, and therefore may be dropped in the path integrals without much loss of numerical accuracy. To achieve this, we will carry out the following:

- (1) Find the governing equations of i-QuAPI by taking the limit $\Delta t \rightarrow 0$;
- (2) Truncate the small terms and provide a reasonable closure;
- (3) Discretize the resulting system partial differential equations to get a scheme with less memory cost.

In this section, we will focus only on the representation of the unknown function which stands for a path segment with memory time T. The complete form of the governing equations will be derived in Section 4.

3.1. Representation of the paths. In the i-QuAPI method, each A_l has a set of parameters $S_{k-1}, \dots, S_{k-\Delta k}$, representing a path segment in the path integral. While the sequence $S_{k-1}, \dots, S_{k-\Delta k}$ represents the discrete path segment, we can regard it as a piecewise constant path segment of the continuous time $s_{\Delta t} : [0, T) \to S^2$, defined by

(9)
$$s_{\Delta t}(\tau) = S_{k-j}, \quad \forall \tau \in \left[(\Delta k - j) \Delta t, \ (\Delta k - j + 1) \Delta t \right], \quad j = 1, \cdots, \Delta k,$$

where $\Delta t = T/\Delta k$. More generally, we would like to consider any path segment $h: [0,T) \to S^2$ with finite discontinuities, where each discontinuity denotes a flip of the classical spin. We assume that there are D spin flips in the path segment h. To define h, we introduce a D-dimensional open simplex $\Delta_t^{(D)}$:

(10)
$$\Delta_t^{(D)} = \left\{ (\tau_1, \cdots, \tau_D) \, \middle| \, \tau_1 > 0, \cdots, \tau_D > 0; \, \sum_{k=1}^D \tau_k < t \right\}$$

so that h has the form

(11)
$$h(\tau) = \begin{cases} (r_0^+, r_0^-), & \text{if } \tau \in [0, \tau_1), \\ (r_d^+, r_d^-), & \text{if } \tau \in \left[\sum_{k=1}^d \tau_k, \sum_{k=1}^{d+1} \tau_k\right), & d = 1, \cdots, D-1, \\ (r_D^+, r_D^-), & \text{if } \tau \in \left[\sum_{k=1}^D \tau_k, T\right), \end{cases}$$

where $(\tau_1, \dots, \tau_D) \in \Delta_T^{(D)}$. In h, we allow the locations of the jumps to be anywhere in (0, T) instead of only being the multiples of Δt . For the paths $s_{\Delta t}$ and h, we use $s_{\Delta t}^+$ $(s_{\Delta t}^-)$ and h^+ (h^-) to denote their positive (negative) branches.

With these definitions, it is natural to express the quantity $A_{k-\Delta k}(S_{k-1}, \dots, S_{k-\Delta k})$ as $A((k-\Delta k)\Delta t, s_{\Delta t})$, where the first argument $(k - \Delta k)\Delta t$ plays the role of the subscript $(k - \Delta k)$, denoting the starting time of the path segment. This can then be further extended to continuous time by writing the quantity as A(t, h) for any $t \ge 0$ and h holding the form (11). However, such a quantity A(t, h) includes a non-classical argument h, which is inconvenient for the formulation of differential equations and the design of numerical scheme. We would therefore like to find a more explicit form of A(t, h) containing only real parameters. With the assumption that h contains only finite discontinuities, the path segment can be fully determined by the following set of parameters:

- (r^+, r^-) : the state of the path at time t, *i.e.* the value of $h(0) = (h^+(0), h^-(0)) = (r_0^+, r_0^-)$.
- D: the number of spin flips (discontinuous points) in h.
- sgn: a list of D signs representing the branch of each spin flip. Each sign is either "+" (representing the positive branch h^+) or "-" (representing the negative branch h^-). In addition, we introduce an augmenting operation by adding a sign at the beginning of sgn. For example, if sgn = [+, -, +, +], then [+, sgn] := [+, +, -, +, +].

• $(\tau_1, \cdots, \tau_D) \in \triangle^{(D)}$: a sequence of times with the following meaning:

$$\tau_k = \begin{cases} \text{time difference of the first spin flip and the starting time } t, \text{ if } k = 1; \\ \text{time difference of the } k\text{-th flip and the } (k-1)\text{-th flip, if } k = 2, \cdots, D. \end{cases}$$

For example, suppose the truncation time is T = 4 and the graph of the path segment h is given in Figure 1. Then the corresponding parameters are

- $(r^+, r^-) = (h^+(0), h^-(0)) = (+1, -1);$
- D = 5 (total number of spin flips, 2 in positive branch and 3 in negative branch);
 sgn = [+, -, -, +, -] (branches of the spin flips);
- $[\tau_1, \tau_2, \tau_3, \tau_4, \tau_5] = [1.0, 0.5, 0.5, 0.5, 1.0]$ (indicating the times of the spin flips are 1.0, 1.5, 2.0, 2.5, 3.5 from t).



FIGURE 1. An example of function h.

Based on such notations, we write down the continuous version of A_l as

(12)
$$A^{D,\text{sgn}}_{(r^+,r^-)}(t,[\tau_1,\cdots,\tau_D]).$$

In the example given in Figure 1, A can be written as

(13)
$$A_{(+1,-1)}^{5,[+,-,-,+,-]}(t,[1.0,0.5,0.5,0.5,1.0]).$$

An advantage of the notation (12) is that we can define the value of (12) for $(\tau_1, \dots, \tau_D) \in \partial \triangle_T^{(D)}$ by taking the limit. In other words, the domain of definition for (τ_1, \dots, τ_D) can be extended from $\Delta_T^{(D)}$ to $\overline{\Delta_T^{(D)}}$, which is important for us to close the system and formulate the boundary conditions in future sections. Note that when (τ_1, \dots, τ_D) takes values on $\partial \triangle_T^{(D)}$, the path segment h may have one or more spin flips at the same time, or have spin flips at the initial time or the final time. Nevertheless, both notations A(t, h) and (12) will be used in our further discussions according to the context. The precise relation between A(t, h)and $A_{k-\Delta k}$ will be detailed later in Section 3.3.

In i-QuAPI, another quantity $\Lambda(S_k, \dots, S_{k-\Delta k})$ is also determined by a path segment, while it has one more argument than $A_{k-\Delta k}(S_{k-1}, \dots, S_{k-\Delta k})$. In the continuous form, the length of this extra argument becomes infinitesimal so that the associated continuous quantity is also expected to be determined by h. This will be discussed in detail in Section 3.2. The integral over the paths, which forms the density matrix, will be considered in Section 3.4.

In the following sections, our major task is to take the continuous limit $\Delta t \to 0$ to define the quantities we need to formulate differential equations. While taking the limit of the discrete path, we assume that the memory length $T = \Delta k \Delta t$ is fixed, and S_{k-i} takes the value of a fixed path segment $h(\tau)$ with $\tau \in [0,T)$ being the continuous time:

(14)
$$S_{k-j} = h((\Delta k - j)\Delta t), \qquad j = 1, \cdots, \Delta k.$$

With this assumption, it is clear that when Δt decreases (or when Δk increases), the function $s_{\Delta t}$ defined in (9) converges to the path segment $h(\tau)$. Thereby, we would like to provide proper definitions for the functions with continuous times.

3.2. Continuous limit of the propagator. According to (4) and (6), the discrete propagator Λ can be expressed by

(15)
$$\Lambda(S_k, \cdots, S_{k-\Delta k}) = \exp\left(-\sum_{m=0}^{\Delta k} (s_k^+ - s_k^-)(\eta_{k,k-m}s_{k-m}^+ - \eta_{k,k-m}^*s_{k-m}^-)\right) \\ \langle s_k^+ | e^{-iH_0\Delta t} | s_{k-1}^+ \rangle \langle s_{k-1}^- | e^{iH_0\Delta t} | s_k^- \rangle.$$

In the continuous limit, all the arguments of Λ become a path segment h, and $\eta_{k,k'}$ needs to be replaced by the bath response function [19] defined by¹

$$\tilde{\eta}(\tau) = \frac{1}{\pi} \int_0^\infty d\omega J(\omega) \left(\coth\left(\frac{\beta\omega}{2}\right) \cos(\omega\tau) - i\sin(\omega\tau) \right).$$

When $\tau = (k - k')\Delta t$, the relation between $\eta_{k,k'}$ and $\tilde{\eta}(\tau)$ is

(16)
$$\tilde{\eta}(\tau) = \lim_{\Delta t \to 0} \frac{\eta_{k,k'}}{\Delta t^2}$$

According to the correspondence between $h(\cdot)$ and S_j given by (14), the continuous limit of s_{k-1}^{\pm} appearing in the bra-ket notations in (15) should be the left limit of $h^{\pm}(t)$ at t = T. In the case where h is left continuous at time T, the following proposition holds.

Proposition 1. Given a path segment h, suppose that the discrete path is chosen according to (14) and $S_k = S_{k-1}$. It holds that

$$\lim_{\Delta t \to 0} \frac{\Lambda(S_k, \cdots, S_{k-\Delta k}) - 1}{\Delta t} = -W(h)$$

with

(17)
$$W(h) = \int_0^T \left((h^+(T) - h^-(T)) \left(\tilde{\eta}(\tau) h^+(T - \tau) - \tilde{\eta}^*(\tau) h^-(T - t) \right) d\tau + i \left(\left\langle h^+(T) \middle| H_0 \middle| h^+(T) \right\rangle - \left\langle h^-(T) \middle| H_0 \middle| h^-(T) \right\rangle \right).$$

where $(h^+(T), h^-(T)) = (s_k^+, s_k^-)$.

Proof. By Taylor expansion of the operators $e^{\pm iH_0\Delta t} = id \pm iH_0\Delta t + \mathcal{O}(\Delta t^2)$, the limit of the bra-kets in (15) can be given by

$$\lim_{\Delta t \to 0} \frac{\langle s_k^+ | e^{-iH_0 \Delta t} | s_{k-1}^+ \rangle \langle s_{k-1}^- | e^{iH_0 \Delta t} | s_k^- \rangle - 1}{\Delta t} = -i \left(\langle h^+(T) | H_0 | h^+(T) \rangle - \langle h^-(T) | H_0 | h^-(T) \rangle \right).$$

Here due to the left continuity of h, the limits of s_k^{\pm} and s_{k-1}^{\pm} are the same. According to the relation of discrete $\eta_{k,k'}$ and continuous $\tilde{\eta}$ in (16), the following limit holds:

$$\lim_{\Delta t \to 0} \frac{\sum_{m=0}^{\Delta k} \left(s_k^+ - s_k^- \right) \left(\eta_{k,k-m} s_{k-m}^+ - \eta_{k,k-m}^* s_{k-m}^- \right)}{\Delta t}$$
$$= \int_0^T (h^+(T) - h^-(T)) \left(\tilde{\eta}(\tau) h^+(T-\tau) - \tilde{\eta}^*(\tau) h^-(T-\tau) \right) \mathrm{d}\tau.$$

The proposition follows by combining the two limits and applying the Taylor expansion $e^x = 1 + x + \mathcal{O}(x^2)$. \Box

For a given path h with $(\tau_1, \dots, \tau_D) \in \triangle_T^{(D)}$, one can always increase Δk to satisfy the condition $S_k = S_{k-1}$. For the case when $S_k \neq S_{k-1}$, more details will be discussed in Section 4.2.

According to this proposition, the path-dependent function W(h) can be regarded as the continuous form of the propagator. As h can be fully determined by the set of parameters described in Section 3.1, we can mimic the notation in (12) to write W as

$$W^{D,\text{sgn}}_{(r^+,r^-)}([\tau_1,\cdots,\tau_D]).$$

¹Compared with the expression in [19], we have omitted the term with $(s^+)^2 - (s^-)^2$ since both s^+ and s^- only take values ±1. Also, the integral domain $[0, +\infty)$ comes from our assumption that all the harmonic oscillators have positive frequencies.

3.3. Continuous limit of A's. In this section, we would now like to explore the relation between the function A(t,h) and the quantities $A_{k-\Delta k+1}(S_k, \dots, S_{k-\Delta k+1})$. By the idea of the i-QuAPI method, the quantity $A_{k-\Delta k+1}(S_k, \dots, S_{k-\Delta k+1})$ denotes the ensemble of all the discrete paths whose last segments of length T are given by $S_{k-\Delta k+1}, \dots, S_k$. This can be observed by applying (5) recursively, resulting in

$$A_{k-\Delta k+1}(S_{k},\cdots,S_{k-\Delta k+1}) = \sum_{\substack{S_{k-\Delta k},\cdots,S_{0}\in\mathcal{S}^{2}\\ \langle s_{k}^{+}|e^{-iH_{0}\Delta t}|s_{k-1}^{+}\rangle\cdots\langle s_{1}^{+}|e^{-iH_{0}\Delta t}|s_{0}^{+}\rangle\langle s_{0}^{+}|\tilde{\rho}_{s}(0)|s_{0}^{-}\rangle}$$
(18)
$$\begin{pmatrix} s_{0}^{-}|e^{iH_{0}\Delta t}|s_{1}^{-}\rangle\cdots\langle s_{k-1}^{-}|e^{iH_{0}\Delta t}|s_{k}^{-}\rangle\\ \prod_{j_{1}=0}^{k}\prod_{j_{2}=\max\{0,k-\Delta k\}}^{j_{1}}\exp\left(-(s_{j_{1}}^{+}-s_{j_{1}}^{-})(\eta_{j_{1},j_{2}}s_{j_{2}}^{+}-\eta_{j_{1},j_{2}}^{*}s_{j_{2}}^{-})\right)\right],$$

where the summation symbol takes into account all the possible paths before the segment specified in the arguments.

We first take the limit of the system part, represented by the product of several bra-kets in (18). Since the path segment h is denoted by specifying the locations of spin flips, we choose to deal with the continuous segments of h and the discontinuities in h separately, requiring the following two lemmas:

Lemma 2. Suppose in the time interval $[\mathcal{T}_1, \mathcal{T}_2] = [k_1 \Delta t, k_2 \Delta t]$, the state of the system remains unchanged, *i.e.*

$$S_{k_1} = \dots = S_{k_2} = S := (r^+, r^-)$$

For fixed \mathcal{T}_1 and \mathcal{T}_2 , it holds that

$$\lim_{\Delta t \to 0} \left\langle s_{k_{2}}^{+} \middle| e^{-iH_{0}\Delta t} \middle| s_{k_{2}-1}^{+} \right\rangle \cdots \left\langle s_{k_{1}+1}^{+} \middle| e^{-iH_{0}\Delta t} \middle| s_{k_{1}}^{+} \right\rangle = e^{-i\left\langle r^{+} \middle| H_{0} \middle| r^{+} \right\rangle (\mathcal{T}_{2} - \mathcal{T}_{1})},$$
$$\lim_{\Delta t \to 0} \left\langle s_{k_{1}}^{-} \middle| e^{iH_{0}\Delta t} \middle| s_{k_{1}+1}^{-} \right\rangle \cdots \left\langle s_{k_{2}-1}^{-} \middle| e^{iH_{0}\Delta t} \middle| s_{k_{2}}^{-} \right\rangle = e^{i\left\langle r^{-} \middle| H_{0} \middle| r^{-} \right\rangle (\mathcal{T}_{2} - \mathcal{T}_{1})}.$$

Proof. By the limit $e = \lim_{x\to 0} (1+x)^{1/x}$ and Taylor expansion of the operator $e^{-iH_0\Delta t} = id - iH_0\Delta t + O(\Delta t^2)$,

$$\begin{split} &\lim_{\Delta t \to 0} \left\langle s_{k_{2}}^{+} \right| \mathrm{e}^{-\mathrm{i}H_{0}\Delta t} \left| s_{k_{2}-1}^{+} \right\rangle \cdots \left\langle s_{k_{1}+1}^{+} \right| \mathrm{e}^{-\mathrm{i}H_{0}\Delta t} \left| s_{k_{1}}^{+} \right\rangle \\ &= \lim_{\Delta t \to 0} \left\langle r^{+} \right| \mathrm{id} - \mathrm{i}H_{0}\Delta t + \mathcal{O}(\Delta t^{2}) \left| r^{+} \right\rangle^{(\mathcal{T}_{2} - \mathcal{T}_{1})/\Delta t} \\ &= \lim_{\Delta t \to 0} \left(1 - \mathrm{i}\Delta t \left\langle r^{+} \right| H_{0} \left| r^{+} \right\rangle + \mathcal{O}(\Delta t^{2}) \right)^{(\mathcal{T}_{2} - \mathcal{T}_{1})/\Delta t} \\ &= \mathrm{e}^{-\mathrm{i}\left\langle r^{+} \right| H_{0} \left| r^{+} \right\rangle (\mathcal{T}_{2} - \mathcal{T}_{1})}. \end{split}$$

The proof for the second limit is similar.

Lemma 3. When $s_k^+ \neq s_{k+1}^+$, the following limit holds.

(19)
$$\lim_{\Delta t \to 0} \frac{\left\langle s_{k+1}^+ \middle| e^{-iH_0\Delta t} \middle| s_k^+ \right\rangle}{\Delta t} = -i \left\langle s_{k+1}^+ \middle| H_0 \middle| s_k^+ \right\rangle.$$

Similarly, when $s_k^- \neq s_k^-$, we have

(20)
$$\lim_{\Delta t \to 0} \frac{\langle s_k^- | \mathrm{e}^{\mathrm{i}H_0 \Delta t} | s_{k+1}^- \rangle}{\Delta t} = \mathrm{i} \langle s_k^- | H_0 | s_{k+1}^- \rangle.$$

Proof. By Taylor expansion of $e^{-iH_0\Delta t}$,

$$\lim_{\Delta t \to 0} \frac{\left\langle s_{k+1}^{+} \middle| e^{-iH_{0}\Delta t} \middle| s_{k}^{+} \right\rangle}{\Delta t} = \lim_{\Delta t \to 0} \frac{\left\langle s_{k+1}^{+} \middle| id - iH_{0}\Delta t + \mathcal{O}(\Delta t^{2}) \middle| s_{k}^{+} \right\rangle}{\Delta t}$$
$$= \lim_{\Delta t \to 0} \frac{-i\Delta t \left\langle s_{k+1}^{+} \middle| H_{0} \middle| s_{k}^{+} \right\rangle + \mathcal{O}(\Delta t^{2})}{\Delta t} = -i \left\langle s_{k+1}^{+} \middle| H_{0} \middle| s_{k}^{+} \right\rangle.$$

The proof for (20) is similar.

This lemma shows that the quantity $A_{k-\Delta k+1}(S_k, \dots, S_{k-\Delta k+1})$ has magnitude Δt^D if s_k^{\pm} changes values D times. Thus, it is natural to let the function A(t,h) demonstrated in Section 3.1 be the limit of $\Delta t^{-D}A_{k-\Delta k+1}(S_k, \dots, S_{k-\Delta k+1})$. We write down the result of such a limit in the following definition:

Definition 4. For a path h given by (11), the quantity A(t,h) or equivalently, $A_{(r_0^+,r_0^-)}^{D,\text{sgn}}(t,[\tau_1,\cdots,\tau_D])$ is defined by

(21)
$$A(t,h) = \sum_{\tilde{D}=0}^{\infty} \sum_{(\tilde{r}_{0}^{+},\tilde{r}_{0}^{-})\in\mathcal{S}^{2}} \sum_{\widetilde{\operatorname{sgn}}\in\{+,-\}^{\tilde{D}}} \int_{\triangle_{t}^{(\tilde{D})}} \mathrm{d}\tilde{\tau} X(g) \delta_{\tilde{r}_{D}^{+}}^{r_{0}^{+}} \delta_{\tilde{r}_{D}^{-}}^{r_{0}^{-}} Y(h) \mathrm{e}^{Z(g,h)}$$

with

$$X(g) = \langle \tilde{r}_{0}^{+} | \rho_{s}(0) | \tilde{r}_{0}^{-} \rangle e^{-i(\langle \tilde{r}_{0}^{+} | H_{0} | \tilde{r}_{0}^{+} \rangle - \langle \tilde{r}_{0}^{-} | H_{0} | \tilde{r}_{0}^{-} \rangle) \tilde{r}_{1}}$$

$$(22) \qquad \prod_{j=1}^{\tilde{D}-1} \left(e^{-i(\langle \tilde{r}_{j}^{+} | H_{0} | \tilde{r}_{j}^{+} \rangle - \langle \tilde{r}_{j}^{-} | H_{0} | \tilde{r}_{j}^{-} \rangle) \tilde{r}_{j+1}} \left(-i \langle \tilde{r}_{j}^{+} | H_{0} | \tilde{r}_{j-1}^{+} \rangle \delta_{\tilde{sgn}_{j}}^{+} + i \langle \tilde{r}_{j-1}^{-} | H_{0} | \tilde{r}_{j}^{-} \rangle \delta_{\tilde{sgn}_{j}}^{-} \right) \right)$$

$$e^{-i(\langle \tilde{r}_{D}^{+} | H_{0} | \tilde{r}_{D}^{+} \rangle - \langle \tilde{r}_{D}^{-} | H_{0} | \tilde{r}_{D}^{-} \rangle) (t - \sum_{j=1}^{\tilde{D}} \tilde{r}_{j})} \left(-i \langle \tilde{r}_{D}^{+} | H_{0} | \tilde{r}_{D-1}^{+} \rangle \delta_{\tilde{sgn}_{D}}^{+} + i \langle \tilde{r}_{D-1}^{-} | H_{0} | \tilde{r}_{D}^{-} \rangle \delta_{\tilde{sgn}_{D}}^{-} \right);$$

$$Y(h) = e^{-i(\langle r_{0}^{+} | H_{0} | r_{0}^{+} \rangle - \langle r_{0}^{-} | H_{0} | r_{0}^{-} \rangle) \tau_{1}}$$

$$(23) \qquad \prod_{j=1}^{D-1} \left(e^{-i(\langle r_{j}^{+} | H_{0} | r_{j}^{+} \rangle - \langle r_{j}^{-} | H_{0} | r_{j}^{-} \rangle) \tau_{j+1}} \left(-i \langle r_{j}^{+} | H_{0} | r_{j-1}^{+} \rangle \delta_{sgn_{j}}^{+} + i \langle r_{j-1}^{-} | H_{0} | r_{j}^{-} \rangle \delta_{sgn_{j}}^{-} \right) \right)$$

$$e^{-i(\langle r_{D}^{+} | H_{0} | r_{D}^{-} \rangle - \langle r_{D}^{-} | H_{0} | r_{D}^{-} \rangle) (T - \sum_{j=1}^{D} \tau_{j})} \left(-i \langle r_{D}^{+} | H_{0} | r_{D-1}^{+} \rangle \delta_{sgn_{j}}^{+} + i \langle r_{D-1}^{-} | H_{0} | r_{D}^{-} \rangle \delta_{sgn_{j}}^{-} \right) \right);$$

(24)

$$Z(g,h) = -\int_{0}^{t} dx_{1} \int_{\max\{0,x_{1}-T\}}^{x_{1}} dx_{2}(g^{+}(x_{1}) - g^{-}(x_{1}))(g^{+}(x_{2})\tilde{\eta}(x_{1} - x_{2}) - g^{-}(x_{2})\tilde{\eta}^{*}(x_{1} - x_{2}))$$

$$-\int_{t}^{t+T} dx_{1} \int_{\max\{0,x_{1}-T\}}^{t} dx_{2}(h^{+}(x_{1} - t) - h^{-}(x_{1} - t))(g^{+}(x_{2})\tilde{\eta}(x_{1} - x_{2}) - g^{-}(x_{2})\tilde{\eta}^{*}(x_{1} - x_{2}))$$

$$-\int_{0}^{T} dx_{1} \int_{0}^{x_{1}} dx_{2}(h^{+}(x_{1}) - h^{-}(x_{1}))\left(h^{+}(x_{2})\tilde{\eta}(x_{1} - x_{2}) - h^{-}(x_{2})\tilde{\eta}^{*}(x_{1} - x_{2})\right)$$
and $q: [0, t) \to S^{2}$ is defined by

(25)
$$g(\tau) = \begin{cases} (\tilde{r}_0^+, \tilde{r}_0^-), & \forall \tau \in [0, \tilde{\tau}_1) \\ (\tilde{r}_k^+, \tilde{r}_k^-), & \forall \tau \in [\sum_{j=1}^k \tilde{\tau}_j, \sum_{j=1}^{k+1} \tilde{\tau}_j), & k = 1, \cdots, \tilde{D-1} \\ (\tilde{r}_{\tilde{D}}^+, \tilde{r}_{\tilde{D}}^-), & \forall \tau \in [\sum_{j=1}^{\tilde{D}} \tilde{\tau}_j, t) \end{cases}$$

with
$$\boldsymbol{\tau} = (\tilde{\tau}_1, \cdots, \tilde{\tau}_D) \in \Delta_t^{(\tilde{D})}$$
. In (21), $\delta_{\tilde{r}_D^+}^{r_D^+} = \begin{cases} 1, & \text{if } \tilde{r}_{\tilde{D}}^+ = r_0^+ \\ 0, & \text{if } \tilde{r}_{\tilde{D}}^+ \neq r_0^+ \end{cases}$ and $\delta_{\tilde{r}_D^-}^{r_0^-} = \begin{cases} 1, & \text{if } \tilde{r}_{\tilde{D}}^- = r_0^- \\ 0, & \text{if } \tilde{r}_{\tilde{D}}^- \neq r_0^- \end{cases}$. In (22) and (23), sgn_k represents the k-th component of sgn , $\delta_{\operatorname{sgn}_j}^+ = \begin{cases} 1, & \text{if } \operatorname{sgn}_j \text{ is } + \\ 0, & \text{if } \operatorname{sgn}_j \text{ is } - \end{cases}$, $\operatorname{and} \delta_{\operatorname{sgn}_j}^- = \begin{cases} 1, & \text{if } \operatorname{sgn}_j \text{ is } - \\ 0, & \text{if } \operatorname{sgn}_j \text{ is } - \end{cases}$.

One can observe from the above definition that the path is separated into two segments. The first segment, denoted by g, covers the interval [0, t), and all possibilities of g have been considered and summed up on the right-hand side of (21). The second segment, denoted by h, is specified in the arguments of A. The two delta symbols in (21) indicate that the last state of g agrees with the first state of h, so that the two segments are connected without discontinuities. The contributions of g and h coming from the system evolution are summarized in X(g) and Y(h), respectively, and the non-Markovian contribution coming from the system the system-bath interaction is provided in the term $e^{Z(g,h)}$, which couples both segments of the path. Note that when t = 0, the value of A(0, h) is not necessarily zero. The only terms contributing to the final result are the terms with $\tilde{D} = 0$ while other terms vanish as the integral domain $\Delta_t^{(D)}$ has measure zero. The formulation of A(t, h) follows the same idea of its discrete counterpart $A_{k-\Delta k}(S_{k-1}, \dots, S_{k-\Delta k})$, and the relation between these two quantities are given in the following theorem:

Theorem 5. For any $t \ge 0$ and given h, the following limit holds:

$$A(t,h) = \lim_{\Delta t \to 0} \frac{A_{k-\Delta k}(S_{k-1},\cdots,S_{k-\Delta k})}{\Delta t^D}$$

where $S_{k-\Delta k}, \cdots, S_{k-1}$ is given by (14).

Proof. For any given \tilde{D} , $(r_0^+, r_0^-) \in S^2$ and $\widetilde{\text{sgn}} \in \{+, -\}^{\tilde{D}}$, we first estimate X(g), Y(h) and Z(g, h). Let $C_1 = \max_{r,s \in S} |\langle r|H_0|s \rangle|$. Then

$$|X(g)| \leq C_1^{\tilde{D}}, \qquad |Y(h)| \leq C_1^{D}.$$

In addition, since $\tilde{\eta}(\cdot)$ is a continuous function on [0, T], we have

$$|Z(g,h)| \leqslant \frac{T^2 + 2Tt}{2} \cdot 4 \max |\tilde{\eta}| := C_2(t).$$

Therefore, we have

T

$$R_{1} := \left| \sum_{\tilde{D}=\overline{D}}^{\infty} \sum_{\left(\tilde{r}_{0}^{+},\tilde{r}_{0}^{-}\right)\in\mathcal{S}^{2}} \sum_{\widetilde{\operatorname{sgn}}\in\{+,-\}^{\tilde{D}}} \int_{\Delta_{t}^{\tilde{D}}} \mathrm{d}\tilde{\tau} X(g) \delta_{\tilde{r}_{0}^{+}}^{r_{0}^{-}} \delta_{\tilde{r}_{D}^{-}}^{r_{0}^{-}} Y(h) \mathrm{e}^{Z(g,h)} \right|$$

$$\leq \sum_{\tilde{D}=\overline{D}}^{\infty} \sum_{\left(\tilde{r}_{0}^{+},\tilde{r}_{0}^{-}\right)\in\mathcal{S}^{2}} \sum_{\widetilde{\operatorname{sgn}}\in\{+,-\}^{\tilde{D}}} \int_{\Delta_{t}^{\tilde{D}}} \mathrm{d}\tilde{\tau} C_{1}^{\tilde{D}+D} C_{2}(t)$$

$$\leq \sum_{\tilde{D}=\overline{D}}^{\infty} 4 \cdot 2^{\tilde{D}} C_{1}^{\tilde{D}+D} C_{2}(t) \frac{t^{\tilde{D}}}{\tilde{D}!} \to 0 \quad \text{as} \quad \overline{D} \to \infty.$$

Similarly, in (18), for given $h, S_{k-\Delta k+1}, \dots, S_k$ are determined by h as in (14).

$$\begin{aligned} R_{2} := \left| \sum_{\substack{S_{k-\Delta k}, \cdots, S_{0} \in \mathcal{S}^{2} \\ \text{number of spin flips} \geqslant \overline{D}}} \langle s_{k}^{+} | e^{-iH_{0}\Delta t} | s_{k-1}^{+} \rangle \cdots \langle s_{1}^{+} | e^{-iH_{0}\Delta t} | s_{0}^{+} \rangle \langle s_{0}^{+} | \tilde{\rho}_{s}(0) | s_{0}^{-} \rangle \\ & \left\langle s_{0}^{-} | e^{iH_{0}\Delta t} | s_{1}^{-} \rangle \cdots \langle s_{k-1}^{-} | e^{iH_{0}\Delta t} | s_{k}^{-} \rangle \prod_{j_{1}=0}^{k} \prod_{j_{2}=\max\{0,k-\Delta k\}}^{j_{1}} \exp\left(-(s_{j_{1}}^{+} - s_{j_{1}}^{-})(\eta_{j_{1},j_{2}}s_{j_{2}}^{+} - \eta_{j_{1},j_{2}}^{*}s_{j_{2}}^{-})\right) \right| \\ & \leq \sum_{\bar{D}=\bar{D}}^{2\Delta k-2} \sum_{\substack{S_{k-\Delta k}, \cdots, S_{0} \in \mathcal{S}^{2} \\ \text{number of spin flips}=\bar{D}}} \left| \langle s_{k}^{+} | e^{-iH_{0}\Delta t} | s_{k-1}^{+} \rangle \cdots \langle s_{1}^{+} | e^{-iH_{0}\Delta t} | s_{0}^{+} \rangle \langle s_{0}^{+} | \tilde{\rho}_{s}(0) | s_{0}^{-} \rangle \\ & \left\langle s_{0}^{-} | e^{iH_{0}\Delta t} | s_{1}^{-} \rangle \cdots \langle s_{k-1}^{-} | e^{iH_{0}\Delta t} | s_{k}^{-} \rangle \prod_{j_{1}=0}^{k} \prod_{j_{2}=\max\{0,k-\Delta k\}}^{j_{1}} \exp\left(-(s_{j_{1}}^{+} - s_{j_{1}}^{-})(\eta_{j_{1},j_{2}}s_{j_{2}}^{+} - \eta_{j_{1},j_{2}}^{*}s_{j_{2}}^{-})\right) \right| \\ & \leq \sum_{\bar{D}=\bar{D}}^{2\Delta k-2} \left(\frac{2\Delta k-2}{\bar{D}}\right) \Delta t^{\bar{D}+D} \leqslant \sum_{\bar{D}=\bar{D}}^{\infty} \frac{(2\Delta k-2)^{\bar{D}}}{\bar{D}!} \Delta t^{\bar{D}+D} \leqslant \sum_{\bar{D}=\bar{D}}^{\infty} \frac{(2T)^{\bar{D}}}{\bar{D}!} \Delta t^{D}. \end{aligned}$$

Therefore, $\Delta t^{-D} R_2 \to 0$ as $\overline{D} \to \infty$. For any $\varepsilon > 0$, there exist a sufficiently large integer \overline{D} , such that

$$R_1 < \varepsilon$$
 and $\Delta t^{-D} R_2 < \varepsilon$.

For the integral over $\Delta_t^{\tilde{D}}$, according to definition of Riemann integral, for a large enough Δk ,

$$\left| \int_{\Delta_t^{\tilde{D}}} \mathrm{d}\tilde{\boldsymbol{\tau}} X(g) \delta_{\tilde{r}_{\tilde{D}}^+}^{r_0^+} \delta_{\tilde{r}_{\tilde{D}}^-}^{r_0^-} Y(h) \mathrm{e}^{Z(g,h)} - \sum_{g \in \mathcal{G}_{(\tilde{r}_0^+, \tilde{r}_0^-)}^{\tilde{D}}} X(g) \delta_{\tilde{r}_{\tilde{D}}^+}^{r_0^+} \delta_{\tilde{r}_{\tilde{D}}^-}^{r_0^-} Y(h) \mathrm{e}^{Z(g,h)} \Delta t^{\tilde{D}} \right| < \frac{\varepsilon}{\tilde{D}!}$$

with $\mathcal{G}_{(\tilde{r}_0^+, \tilde{r}_0^-)}^{\tilde{D}}$ being the set of paths that have initial states $(\tilde{r}_0^+, \tilde{r}_0^-)$ and \tilde{D} spin flips in the set of all possible time discrete paths $S_{k-\Delta k}, \cdots, S_{k-1}$.

In addition, for any given path $g \in \mathcal{G}_{(\tilde{r}_0^+, \tilde{r}_0^-)}^{\tilde{D}}$ with \tilde{D} spin flips, according to Lemma 2, Lemma 3, (16) and the definition of Riemann integral, there exists a large enough Δk , such that

$$\left| \Delta t^{\tilde{D}} X(g) \delta^{r_0^+}_{\tilde{r}_{\tilde{D}}^+} \delta^{r_0^-}_{\tilde{r}_{\tilde{D}}^-} Y(h) \mathrm{e}^{Z(g,h)} - \Delta t^{-D} \left\langle s_k^+ \left| \mathrm{e}^{-\mathrm{i}H_0 \Delta t} \right| s_{k-1}^+ \right\rangle \cdots \left\langle s_1^+ \left| \mathrm{e}^{-\mathrm{i}H_0 \Delta t} \right| s_0^+ \right\rangle \left\langle s_0^+ \left| \tilde{\rho}_s(0) \right| s_0^- \right\rangle \right. \\ \left. \left\langle s_0^- \left| \mathrm{e}^{\mathrm{i}H_0 \Delta t} \right| s_1^- \right\rangle \cdots \left\langle s_{k-1}^- \left| \mathrm{e}^{\mathrm{i}H_0 \Delta t} \right| s_k^- \right\rangle \prod_{j_1=0}^k \prod_{j_2=\max\{0,k-\Delta k\}}^{j_1} \exp\left(-(s_{j_1}^+ - s_{j_1}^-)(\eta_{j_1,j_2} s_{j_2}^+ - \eta_{j_1,j_2}^* s_{j_2}^-) \right) \right| < \varepsilon \Delta t^{\tilde{D}} \right|$$

where $S_j = g(j\Delta t)$ for $j = 0, \dots, k - \Delta k - 1$. Combine all the results, we have

 $\left| \begin{array}{l} A(t,h) - \frac{A_{k-\Delta k}(S_{k-1},\cdots,S_{k-\Delta k})}{\Delta t^{D}} \right| \\ \leqslant R_{1} + \Delta t^{-D}R_{2} + \sum_{\tilde{D}=0}^{\overline{D}-1} 4 \cdot 2^{\tilde{D}} \left(\frac{\varepsilon}{\tilde{D}!} + \binom{2\Delta k - 2}{\tilde{D}} \varepsilon \Delta t^{\tilde{D}} \right) \\ < \left(2 + 4e^{2} + 4e^{4T}\right) \varepsilon \end{array} \right.$

where $4 \cdot 2^{\tilde{D}}$ comes from the summation over $(\tilde{r}_0^+, \tilde{r}_0^-)$ and $\widetilde{\text{sgn}}$, the binomial coefficient is the cardinality of $\mathcal{G}_{(\tilde{r}_0^+, \tilde{r}_0^-)}^{\tilde{D}}$. The proof is completed as ε is arbitrary.

According to the definition of A(t, h) for $(\tau_1, \dots, \tau_D) \in \Delta_T^{(D)}$ in Definition 4, we can extend the definition of A(t, h) to $(\tau_1, \dots, \tau_D) \in \partial \Delta_T^{(D)}$ by taking the limit. The following theorem guarantees that such extension is well defined.

Theorem 6. For $\boldsymbol{\tau}' = (\tau'_1, \cdots, \tau'_D) \in \partial \triangle_T^{(D)}$, the limit

$$\lim_{(\tau_1,\cdots,\tau_D)\to(\tau_1',\cdots,\tau_D')} A^{D,\mathrm{sgn}}_{(r^+,r^-)}(t,[\tau_1,\cdots,\tau_D])$$

exists and therefore, $A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1',\cdots,\tau_D'])$ can be defined by this limit.

Proof. In order to prove the limit exists, we only need to show that for any $\varepsilon > 0$, there exist a small enough positive number δ , such that

$$\left|A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1^{(1)},\cdots,\tau_D^{(1)}]) - A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1^{(2)},\cdots,\tau_D^{(2)}])\right| < \varepsilon$$

for any $\boldsymbol{\tau}_1 = (\tau_1^{(1)}, \cdots, \tau_D^{(1)})$ and $\boldsymbol{\tau}_2 = (\tau_1^{(2)}, \cdots, \tau_D^{(2)})$ with $\boldsymbol{\tau}_1, \boldsymbol{\tau}_2 \in \Delta_T^{(D)}, \|\boldsymbol{\tau}_1 - \boldsymbol{\tau}'\|_1 < \delta/2$ and $\|\boldsymbol{\tau}_2 - \boldsymbol{\tau}'\|_1 < \delta/2$. Denote the path of $A_{(r^+, r^-)}^{D, \text{sgn}}(t, [\tau_1^{(1)}, \cdots, \tau_D^{(1)}])$ by h_1 and the path of $A_{(r^+, r^-)}^{D, \text{sgn}}(t, [\tau_1^{(2)}, \cdots, \tau_D^{(2)}])$ by h_2 . h_1 and h_2 differs only on intervals whose sum is smaller than δ . Therefore,

$$|Y(h_1) - Y(h_2)| \leq DL_1 C_1^D \delta$$

where L_1 is the Lipsichitz constant such that

$$\left| e^{-i(\langle r^+ | H_0 | r^+ \rangle - \langle r^- | H_0 | r^- \rangle)t_1} - e^{-i(\langle r^+ | H_0 | r^+ \rangle - \langle r^- | H_0 | r^- \rangle)t_2} \right| \leq L_1 | t_1 - t_2$$

for any $(r^+,r^-)\in \mathcal{S}^2$. In addition, for any path $g:[0,t)\to \mathcal{S}^2,$

$$|Z(g,h_1) - Z(g,h_2)| \leq 4\delta T \cdot 2 \max |\tilde{\eta}|.$$

Then

$$\left| \mathbf{e}^{Z(g,h_1)} - \mathbf{e}^{Z(g,h_2)} \right| \leqslant 8TL_2 \max |\tilde{\eta}| \delta$$

with L_2 is the Lipsichitz constant of function e^x in the interval $|x| \leq C_2(t)$ where $C_2(t)$ is defined in Theorem 5. Therefore,

$$|X(g)Y(h_1)Z(g,h_1) - X(g)Y(h_2)Z(g,h_2)| < C_1^{\bar{D}}C_1^{\bar{D}}C_2(t) \left(DL_1C_1^{\bar{D}} + 8TL_2\max|\tilde{\eta}|\right)\delta := C_1^{\bar{D}}C_3\delta$$

Consequentially we have

$$|A(t,h_1) - A(t,h_2)| < \sum_{\tilde{D}=0}^{\infty} \sum_{(\tilde{r}_0^+, \tilde{r}_0^-) \in \mathcal{S}^2} \sum_{\widetilde{\text{sgn}} \in \{+,-\}^{\tilde{D}}} \int_{\triangle_t^{(\tilde{D})}} \mathrm{d}\tilde{\tau} C_1^{\tilde{D}} C_3 \delta \leqslant \sum_{\tilde{D}=0}^{\infty} 4 \frac{(2tC_1)^{\tilde{D}}}{\tilde{D}!} C_3 \delta = 4 \mathrm{e}^{2tC_1} C_3 \delta.$$

The proof is completed by choosing $\delta = (4e^{2tC_1}C_3)^{-1}\varepsilon$.

3.4. **Density Matrix and Observable.** Based on the definition of A(t, h), we would like to represent the density matrix (8) using the form with continuous time. On the left-hand side of (8), the state $S_N = (s_N^+, s_N^-)$ denotes the "final state" of the path segment. Such a state corresponds to the left limit of $h(\tau)$ at $\tau = T$. For the spin-boson model, it is fully determined by the initial state (r^+, r^-) , the number of spin flips D and the sign vector sgn. We denote this final state by

(27)
$$r_f^{\pm} = r_f^{\pm}(r^+, r^-, D, \operatorname{sgn}).$$

For example, in Figure 1, the path segment is represented by the parameters in (13) which determines the final state to be $(r_f^+, r_f^-) = (+1, +1)$.

Similar to the discrete case, the values of continuous A's are closely related to the density matrix, which contains all information of a quantum system. Here is the method to construct the reduced density matrix $\tilde{\rho}(t)$ based on the values of A's.

Theorem 7. The density matrix of a spin-boson model is

(28)
$$\rho_s(t) = \sum_{D=0}^{\infty} \sum_{(r^+, r^-) \in \mathcal{S}^2} \sum_{\mathrm{sgn} \in \{+, -\}^D} \int_{\triangle_T^{(D)}} A^{D, \mathrm{sgn}}_{(r^+, r^-)} \left(t, [\tau_1, \cdots, \tau_D] \right) \left| r_f^+ \right\rangle \!\! \left\langle r_f^- \right| \mathrm{d}\boldsymbol{\tau}$$

with $\triangle_T^{(D)}$ is defined by (10), r_f^{\pm} is determined by (27) and $\boldsymbol{\tau} = (\tau_1, \cdots, \tau_D) \in \triangle_T^{(D)}$.

Proof. For any fixed t and a path h with D spin flips, by choosing $\overline{D} = 0$ in (26), we have

$$\left|A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1,\cdots,\tau_D])\right| \leqslant 4\mathrm{e}^{2C_1t}C_1^D C_2(t).$$

For any \overline{D} , we have

$$\begin{split} R_{1} &:= \left\| \sum_{D=\overline{D}}^{\infty} \sum_{(r^{+},r^{-})\in\mathcal{S}^{2}} \sum_{\mathrm{sgn}\in\{+,-\}^{D}} \int_{\bigtriangleup_{T}^{(D)}} A^{D,\mathrm{sgn}}_{(r^{+},r^{-})}(t,[\tau_{1},\cdots,\tau_{D}]) \left| r_{f}^{+} \right\rangle \! \left\langle r_{f}^{-} \right| \mathrm{d}\tau \right\| \\ &\leqslant \sum_{D=\overline{D}}^{\infty} \sum_{(r^{+},r^{-})\in\mathcal{S}^{2}} \sum_{\mathrm{sgn}\in\{+,-\}^{D}} \int_{\bigtriangleup_{T}^{(D)}} \left| A^{D,\mathrm{sgn}}_{(r^{+},r^{-})}(t,[\tau_{1},\cdots,\tau_{D}]) \right| \mathrm{d}\tau \\ &\leqslant \sum_{D=\overline{D}}^{\infty} \sum_{(r^{+},r^{-})\in\mathcal{S}^{2}} \sum_{\mathrm{sgn}\in\{+,-\}^{D}} \int_{\bigtriangleup_{T}^{(D)}} 4\mathrm{e}^{2C_{1}t}C_{1}^{D}C_{2}(t)\mathrm{d}\tau \\ &= \sum_{D=\overline{D}}^{\infty} 4\cdot 2^{D} \frac{T^{D}}{D!} 4\mathrm{e}^{2C_{1}t}C_{1}^{D}C_{2}(t) = \sum_{D=\overline{D}}^{\infty} 16\mathrm{e}^{2C_{1}t}C_{2}(t) \frac{(2C_{1}T)^{D}}{D!} \to 0 \quad \mathrm{as} \quad \overline{D} \to \infty \end{split}$$

Similarly, for the discrete expression of density matrix, we have the following estimation.

$$R_{2} := \left\| \sum_{\substack{S_{k-\Delta k+1,\cdots,S_{k}} \\ \text{number of spin flips} \geqslant \overline{D}}} A_{k-\Delta k+1}(S_{k},\cdots,S_{k-\Delta k+1}) \left| s_{k}^{+} \right\rangle \! \left\langle s_{k}^{-} \right| \right. \\ \left. \lesssim 4 \sum_{D=\overline{D}}^{2\Delta k-2} \binom{2\Delta k-2}{D} 4 e^{2C_{1}t} C_{1}^{D} C_{2}(t) \Delta t^{D} \right. \\ \left. \leqslant 4 \sum_{D=\overline{D}}^{2\Delta k-2} \frac{(2\Delta k-2)^{D}}{D!} 4 e^{2C_{1}t} C_{1}^{D} C_{2}(t) \Delta t^{D} \right. \\ \left. \leqslant \sum_{D=\overline{D}}^{\infty} 16 e^{2C_{1}t} C_{2}(t) \frac{(2C_{1}T)^{D}}{D!} \to 0 \quad \text{as} \quad \overline{D} \to \infty. \right.$$

Therefore, for any $\varepsilon > 0$, there exists a large enough integer \overline{D} , such that $R_1 < \varepsilon, R_2 < \varepsilon$. For each $D = 0, \dots, \overline{D}, (r^+, r^-) \in S^2$ and $\operatorname{sgn} \in \{+, -\}^D, |r_f^+\rangle \langle r_f^-|$ is fixed. Therefore, according to the definition of Riemann integral, there exists a large enough integer M_1 with $M_1 > \overline{D}$, such that for all and $\Delta k > M_1, D = 0, \dots, \overline{D}, (r^+, r^-) \in S^2$ and $\operatorname{sgn} \in \{+, -\}^D$,

$$\left| \int_{\Delta_T^{(D)}} A_{(r^+, r^-)}^{D, \operatorname{sgn}}(t, [\tau_1, \cdots, \tau_D]) \mathrm{d}\boldsymbol{\tau} - \sum_{s_{\Delta t} \in \mathcal{I}_D^{\Delta k}} A(t, s_{\Delta t}) \Delta t^D \right| < \frac{\varepsilon}{D!}$$

where $\mathcal{I}_D^{\Delta k}$ is the set of all piecewise constant path segment with D spin flips of continuous time determined by the sequence $S_k, \dots, S_{k-\Delta k+1}$. According to Theorem 5, there exists a large enough integer M_2 , such that for $\Delta k > M_2$

$$\left|A(t, s_{\Delta t})\Delta t^{D} - A_{k-\Delta k+1}(S_{k}, \cdots, S_{k-\Delta k+1})\right| < \varepsilon \Delta t^{D}$$

holds for all $D = 0, \dots, \overline{D}, (r^+, r^-) \in S^2$ and sgn $\in \{+, -\}^D$. For the paths with number of spin flips being D (not greater than \overline{D}), we have

$$\left| \int_{\Delta_T^{(D)}} A_{(r+,r-)}^{D,\mathrm{sgn}}(t, [\tau_1, \cdots, \tau_D]) \mathrm{d}\boldsymbol{\tau} - \sum_{\substack{S_{k-1}, \cdots, S_{k-\Delta k+1} \in \mathcal{S}^2 \\ \text{number of spin flips} = D}} A_{k-\Delta k+1}(S_k, \cdots, S_{k-\Delta k+1}) \right| \\ < \binom{2\Delta k - 4}{D} \varepsilon \Delta t^D + \frac{\varepsilon}{D!} \leqslant \left(\frac{(2\Delta k - 4)^D}{D!} \Delta t^D + \frac{1}{D!} \right) \varepsilon < \frac{(2T)^D + 1}{D!} \varepsilon.$$

Therefore, choose $\Delta k > \max\{M_1, M_2\}$, we finally have

$$\begin{split} \left\| \sum_{D=0}^{\infty} \sum_{(r^+,r^-)\in\mathcal{S}^2} \sum_{\mathrm{sgn}\in\{+,-\}^D} \int_{\Delta_T^{(D)}} A_{(r^+,r^-)}^{D,\mathrm{sgn}} \left(t, [\tau_1,\cdots,\tau_D]\right) \left| r_f^+ \right\rangle \! \left\langle r_f^- \right| \mathrm{d}\tau \\ &- \sum_{S_N-\Delta k+1,\cdots,S_N\in\mathcal{S}} A_{N-\Delta k}(S_N,\cdots,S_{N-\Delta k+1}) \left| s_N^+ \right\rangle \! \left\langle s_N^- \right| \right\| \\ \leqslant \left\| \sum_{D=0}^{\overline{D}} \sum_{(r^+,r^-)\in\mathcal{S}^2} \sum_{\mathrm{sgn}\in\{+,-\}^D} \int_{\Delta_T^{(D)}} A_{(r^+,r^-)}^{D,\mathrm{sgn}} \left(t, [\tau_1,\cdots,\tau_D]\right) \left| r_f^+ \right\rangle \! \left\langle r_f^- \right| \mathrm{d}\tau \\ &- \sum_{S_N-\Delta k+1,\cdots,S_N\in\mathcal{S}} A_{N-\Delta k}(S_N,\cdots,S_{N-\Delta k+1}) \left| s_N^+ \right\rangle \! \left\langle s_N^- \right| \right\| + \varepsilon \\ \leqslant \sum_{D=0}^{\overline{D}} \sum_{(r^+,r^-)\in\mathcal{S}^2} \sum_{\mathrm{sgn}\in\{+,-\}^D} \left| \int_{\Delta_T^{(D)}} A_{(r^+,r^-)}^{D,\mathrm{sgn}} \left(t, [\tau_1,\cdots,\tau_D]\right) \mathrm{d}\tau \\ &- \sum_{S_k=(r_f^+,r_f^-),\mathrm{number of spin flips}=D} A_{N-\Delta k}(S_N,\cdots,S_{N-\Delta k+1}) \right| + 2\varepsilon \\ \leqslant \sum_{D=0}^{\overline{D}} 2^{D+2} \frac{(2T)^D+1}{D!} \varepsilon + 2\varepsilon \leqslant (4e^{4T}+4e^2+2)\varepsilon. \end{split}$$

The proof is completed as ε is arbitrary.

With the reduced density matrix $\tilde{\rho}(t)$, any observable O can be computed by

$$\langle O(t) \rangle = \operatorname{tr}(\rho_s(t)O)$$

In the following discussion, we mainly focus on the case with $O = \sigma_z = \text{diag}(1, -1)$, and therefore

$$\langle \sigma_z(t) \rangle = \langle +1 | \rho_s(t) | +1 \rangle - \langle -1 | \rho_s(t) | -1 \rangle.$$

4. DIFFERENTIAL EQUATIONS FOR PATH INTEGRALS

We are now ready to formulate the equations for the functions $A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t, [\tau_1, \cdots, \tau_D])$ based on the i-QuAPI iteration (5). While the resulting equations hold for $(\tau_1, \cdots, \tau_D) \in \Delta_T^{(D)}$ with $\Delta_T^{(D)}$ being an open set, they have to be completed by supplementing with appropriate boundary conditions. Moreover, as D can take any positive integer, the form $A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t, [\tau_1, \cdots, \tau_D])$ actually includes infinite functions. In practice, one has to truncate the system and apply reasonable closure to make it numerically solvable. These topics will be discussed separately in the following subsections.

4.1. Formulation of the Differential Equations. With Proposition 1, QuAPI partial differential equation can be derived in the domain $\triangle_T^{(D)}$ for a spin-boson system. The boundary case will be discussed in Theorem 9 in the next section.

In this section, a PDE based on (5) for a two-state system is derived. As is discussed above, A's will be expressed as the form (12).

Theorem 8. In the spin-boson model, the following partial differential equation holds for A defined by (21) with the assumption that $A_{(r^+,r^-)}^{D,\text{sgn}}(t,[\tau_1,\cdots,\tau_D])$ is differentiable with respect to t and τ_1 .

(29)

$$\frac{\partial}{\partial t} A^{D,\text{sgn}}_{(r^+,r^-)} \left(t, [\tau_1, \cdots, \tau_D] \right) = -W^{D,\text{sgn}}_{(r^+,r^-)} \left([\tau_1, \cdots, \tau_D] \right) A^{D,\text{sgn}}_{(r^+,r^-)} \left(t, [\tau_1, \cdots, \tau_D] \right)
+ A^{D+1,[-,\text{sgn}]}_{(r^+,\hat{r}^-)} \left(t, [0,\tau_1, \cdots, \tau_D] \right) + A^{D+1,[+,\text{sgn}]}_{(\hat{r}^+,r^-)} \left(t, [0,\tau_1, \cdots, \tau_D] \right)
+ \frac{\partial}{\partial \tau_1} A^{D,\text{sgn}}_{(r^+,r^-)} \left(t, [\tau_1, \cdots, \tau_D] \right)$$

where $W_{(r^+,r^-)}^{D,\text{sgn}}([\tau_1,\cdots,\tau_D])$ is defined in (17) and \hat{r} means the state different from r, that is, $\widehat{+1} = -1$ and $\widehat{-1} = +1$. Note that when D = 0, the derivative of A with respect to τ_1 is zero.

Proof. We take the derivatives directly based on the expression (21). As the integral domain $\triangle_t^{(\tilde{D})}$ is dependent on t, we first consider the derivative over the integral domain for general function f:

$$(30) \qquad \frac{\partial}{\partial t} \int_{\Delta_{t}^{(\tilde{D})}} f(t,\tilde{\tau}_{1},\cdots,\tilde{\tau}_{\tilde{D}}) \mathrm{d}\tilde{\boldsymbol{\tau}} = \int_{\Delta_{t}^{(\tilde{D})}} \frac{\partial}{\partial t} f(t,\tilde{\tau}_{1},\cdots,\tilde{\tau}_{\tilde{D}}) \mathrm{d}\tilde{\boldsymbol{\tau}} \\ + \int_{\Delta_{t}^{(\tilde{D}-1)}} f(t,\tilde{\tau}_{1},\cdots,\tilde{\tau}_{\tilde{D}-1},t-\tilde{\tau}_{1}-\cdots-\tilde{\tau}_{\tilde{D}-1}) \mathrm{d}\tilde{\tau}_{1}\cdots\mathrm{d}\tilde{\tau}_{\tilde{D}-1}.$$

Especially, when $\tilde{D} = 0$, the second term in (30) vanishes. Therefore,

$$(31) \qquad \begin{aligned} \frac{\partial}{\partial t} A^{D,\mathrm{sgn}}_{(r^+,r^-)}(t,[\tau_1,\cdots,\tau_D]) \\ &= \sum_{\tilde{D}=1}^{\infty} \sum_{\left(\tilde{r}^+_0,\tilde{r}^-_0\right)\in\mathcal{S}^2} \sum_{\widetilde{\mathrm{sgn}}\in\{+,-\}^{\tilde{D}}} \int_{\Delta^{(\tilde{D}-1)}_t} \mathrm{d}\tilde{\tau}_1\cdots\tilde{\tau}_{\tilde{D}-1} \left(X(g)\delta^{r^+_0}_{\tilde{r}^+_D}\delta^{r^-_0}_{\tilde{r}^-_D}Y(h)\mathrm{e}^{Z(g,h)}\right)\Big|_{\tilde{\tau}_{\tilde{D}}=t-\tilde{\tau}_1-\cdots-\tilde{\tau}_{\tilde{D}-1}} \\ &+ \sum_{\tilde{D}=0}^{\infty} \sum_{\left(\tilde{r}^+_0,\tilde{r}^-_0\right)\in\mathcal{S}^2} \sum_{\widetilde{\mathrm{sgn}}\in\{+,-\}^{\tilde{D}}} \int_{\Delta^{(\tilde{D})}_t} \mathrm{d}\tilde{\tau} \frac{\partial}{\partial t}X(g)\delta^{r^+_0}_{\tilde{r}^+_D}\delta^{r^-_0}_{\tilde{r}^-_D}Y(h)\mathrm{e}^{Z(g,h)} \\ &+ \sum_{\tilde{D}=0}^{\infty} \sum_{\left(\tilde{r}^+_0,\tilde{r}^-_0\right)\in\mathcal{S}^2} \sum_{\widetilde{\mathrm{sgn}}\in\{+,-\}^{\tilde{D}}} \int_{\Delta^{(\tilde{D})}_t} \mathrm{d}\tilde{\tau} X(g)\delta^{r^+_0}_{\tilde{r}^+_D}\delta^{r^-_0}_{\tilde{r}^-_D}Y(h)\frac{\partial}{\partial t}Z(g,h)\mathrm{e}^{Z(g,h)} \end{aligned}$$

In addition, we take the partial derivative with respect to τ_1 .

$$\begin{split} &\frac{\partial}{\partial \tau_1} A^{D,\mathrm{sgn}}_{(r^+,r^-)}(t,[\tau_1,\cdots,\tau_D]) \\ &= \sum_{\tilde{D}=0}^{\infty} \sum_{(\tilde{r}^+_0,\tilde{r}^-_0)\in\mathcal{S}^2} \sum_{\widetilde{\mathrm{sgn}}\in\{+,-\}^{\tilde{D}}} \int_{\triangle_t^{(\tilde{D})}} \mathrm{d}\tilde{\tau} X(g) \delta^{r^+_0}_{\tilde{r}^+_D} \delta^{r^-_0}_{\tilde{r}^-_D} \frac{\partial}{\partial \tau_1} Y(h) \mathrm{e}^{Z(g,h)} \\ &+ \sum_{\tilde{D}=0}^{\infty} \sum_{(\tilde{r}^+_0,\tilde{r}^-_0)\in\mathcal{S}^2} \sum_{\widetilde{\mathrm{sgn}}\in\{+,-\}^{\tilde{D}}} \int_{\triangle_t^{(\tilde{D})}} \mathrm{d}\tilde{\tau} X(g) \delta^{r^+_0}_{\tilde{r}^+_D} \delta^{r^-_0}_{\tilde{r}^-_D} Y(h) \frac{\partial}{\partial \tau_1} Z(g,h) \mathrm{e}^{Z(g,h)}. \end{split}$$

For any given path g,

$$\begin{split} &\frac{\partial}{\partial t}X(g)\delta_{\tilde{r}_{\bar{D}}^{+}}^{r_{\bar{D}}^{+}}Y(h)\mathrm{e}^{Z(g,h)}-X(g)\delta_{\tilde{r}_{\bar{D}}^{+}}^{r_{\bar{D}}^{+}}\delta_{\tilde{r}_{\bar{D}}^{-}}^{r_{\bar{D}}^{-}}\frac{\partial}{\partial\tau_{1}}Y(h)\mathrm{e}^{Z(g,h)}\\ &=-\mathrm{i}\left(\left<\tilde{r}_{\bar{D}}^{+}\Big|H_{0}\Big|\tilde{r}_{\bar{D}}^{+}\right>-\left<\tilde{r}_{\bar{D}}^{-}\Big|H_{0}\Big|\tilde{r}_{\bar{D}}^{-}\right>\right)X(g)\delta_{\tilde{r}_{\bar{D}}^{+}}^{r_{\bar{D}}^{+}}\delta_{\tilde{r}_{\bar{D}}^{-}}^{r_{\bar{D}}^{-}}Y(h)\mathrm{e}^{Z(g,h)}\\ &-\left(-\mathrm{i}\left(\left< r_{0}^{+}\Big|H_{0}\Big|r_{0}^{+}\right>-\left< r_{0}^{-}\Big|H_{0}\Big|r_{0}^{-}\right>\right)+\mathrm{i}\left(\left< r_{D}^{+}\Big|H_{0}\Big|r_{D}^{-}\right>-\left< r_{D}^{-}\Big|H_{0}\Big|r_{D}^{-}\right>\right)\right)X(g)\delta_{\tilde{r}_{\bar{D}}^{+}}^{r_{\bar{D}}^{+}}\delta_{\tilde{r}_{\bar{D}}^{-}}^{r_{0}^{-}}Y(h)\mathrm{e}^{Z(g,h)}\\ &=-\mathrm{i}\left(\left< r_{D}^{+}\Big|H_{0}\Big|r_{D}^{-}\right>-\left< r_{D}^{-}\Big|H_{0}\Big|r_{D}^{-}\right>\right)X(g)\delta_{\tilde{r}_{\bar{D}}^{+}}^{r_{0}^{+}}\delta_{\tilde{r}_{\bar{D}}^{-}}^{r_{0}^{-}}Y(h)\mathrm{e}^{Z(g,h)}. \end{split}$$

$$\begin{split} \text{If } \delta_{\vec{r}_{D}}^{\vec{r}_{D}^{+}} \delta_{\vec{r}_{D}^{-}}^{\vec{r}_{D}^{-}} &= 0, \text{ both } X(g) \delta_{\vec{r}_{D}^{+}}^{\vec{r}_{D}^{+}} \delta_{\vec{r}_{D}^{-}}^{\vec{r}_{D}^{-}} Y(h) \frac{\partial}{\partial t} Z(g,h) e^{Z(g,h)} \text{ and } X(g) \delta_{\vec{r}_{D}^{+}}^{\vec{r}_{D}^{+}} \delta_{\vec{r}_{D}^{-}}^{\vec{r}_{D}^{-}} Y(h) \frac{\partial}{\partial \tau_{1}} Z(g,h) e^{Z(g,h)} \text{ are zero.} \\ \delta_{\vec{r}_{D}^{+}}^{\vec{r}_{D}^{+}} \delta_{\vec{r}_{D}^{-}}^{\vec{r}_{D}^{-}} &= 1, \text{ we have } (\vec{r}_{D}^{+}, \vec{r}_{D}^{-}) = (r_{0}^{+}, r_{0}^{-}), \text{ we have} \\ & \frac{\partial}{\partial t} Z(g,h) - \frac{\partial}{\partial \tau_{1}} Z(g,h) \\ &= \lim_{\Delta t \to 0} \frac{1}{\Delta t} \left(-\int_{t+T}^{t+T+\Delta t} dx_{1} \int_{x_{1}-T}^{t+\Delta t} dx_{2}(h^{+}(x_{1}-t-\Delta t) - h^{-}(x_{1}-t-\Delta t)) \right) \\ & (\vec{r}_{D}^{+} \tilde{\eta}(x_{1}-x_{2}) - \vec{r}_{D}^{-} \tilde{\eta}^{*}(x_{1}-x_{2})) \\ & -\int_{t+T}^{t+T+\Delta t} dx_{1} \int_{t+\Delta t}^{x_{1}} dx_{2}(h^{+}(x_{1}-t-\Delta t) - h^{-}(x_{1}-t-\Delta t)) \\ & (h^{+}(x_{2}-t-\Delta t)\tilde{\eta}(x_{1}-x_{2}) - h^{-}(x_{2}-t-\Delta t)\tilde{\eta}^{*}(x_{1}-x_{2})) \right) \\ &= -\int_{0}^{T} dx_{2} \left(h^{+}(T) - h^{-}(T) \right) (h^{+}(x_{2})\tilde{\eta}(T-x_{2}) - h^{-}(x_{2})\tilde{\eta}^{*}(T-x_{2})) \right). \end{split}$$

For the first term in (31), consider the following two quantities:

$$(32) \qquad \sum_{\tilde{D}=1}^{\infty} \sum_{(\tilde{r}_{0}^{+}, \tilde{r}_{0}^{-}) \in \mathcal{S}^{2}} \sum_{\widetilde{\text{sgn}} \in \{+, -\}^{\tilde{D}}} \int_{\Delta_{t}^{(\tilde{D}-1)}} d\tilde{\tau}_{1} \cdots \tilde{\tau}_{\tilde{D}-1} \left(X(g) \delta_{\tilde{r}_{\tilde{D}}^{+}}^{r_{0}^{+}} \delta_{\tilde{r}_{\tilde{D}}^{-}}^{r_{0}^{-}} Y(h) e^{Z(g,h)} \right) \Big|_{\tilde{\tau}_{\tilde{D}}=t-\tilde{\tau}_{1}-\cdots-\tilde{\tau}_{\tilde{D}-1}};$$

$$(33) \qquad A_{(r^{+},\hat{r}^{-})}^{D+1,[-,\text{sgn}]} \left(t, [0,\tau_{1},\cdots,\tau_{D}] \right) + A_{(\hat{r}^{+},r^{-})}^{D+1,[+,\text{sgn}]} \left(t, [0,\tau_{1},\cdots,\tau_{D}] \right).$$

The first term in (31) involves letting $\tilde{\tau}_{\tilde{D}} = t - \tilde{\tau}_1 - \dots - \tilde{\tau}_{\tilde{D}-1}$, which means $(\tilde{\tau}_1, \dots, \tilde{\tau}_{\tilde{D}}) \in \partial \triangle_t^{(D)}$ and therefore, we need to regard it as a limit case. For any $\tilde{D} \ge 1$, $(\tilde{r}_0^+, \tilde{r}_0^-) \in \mathcal{S}^2$, $\widetilde{\text{sgn}} \in \{+, -\}^{\tilde{D}}$ with $\widetilde{\text{sgn}}_{\tilde{D}}$ being +, define the path $g: [0, t) \to \mathcal{S}^2$ by (25). In addition, define $g': [0, t) \to \mathcal{S}^2$ by

$$g'(\tau) = \begin{cases} g(\tau), & \text{if } \tau \in [0, \tilde{\tau}_1 + \dots + \tilde{\tau}_{\tilde{D}}) \\ \lim_{s \to (\tilde{\tau}_1 + \dots, \tilde{\tau}_{\tilde{D}})^-} g(\tau), & \text{if } \tau \in [\tilde{\tau}_1 + \dots + \tilde{\tau}_{\tilde{D}}, t) \end{cases}.$$

Compare with path g, path g' has one fewer spin flip on the positive branch at time $\tilde{\tau}_1 + \cdots + \tilde{\tau}_{\tilde{D}}$. In addition, defined a path $h_{\Delta t} : [0,T) \to S^2$ by

$$h_{\Delta t}(\tau) = \begin{cases} (\hat{r}_0^+, r_0^-), & \text{if } \tau \in [0, \Delta t) \\ h(\tau), & \text{if } \tau \in [\Delta t, T) \end{cases}.$$

If $\delta_{\tilde{r}_{\bar{D}}^+}^{r_0^+} \delta_{\tilde{r}_{\bar{D}}^-}^{r_0^-} = 0$, then we naturally have

(34)
$$\lim_{\tilde{\tau}_{\tilde{D}} \to t - \tilde{\tau}_{1} - \dots - \tilde{\tau}_{\tilde{D}-1}} X(g) \delta_{\tilde{r}_{\tilde{D}}^{+}}^{r_{0}^{-}} \delta_{\tilde{r}_{\tilde{D}}^{-}}^{r_{0}^{-}} Y(h) e^{Z(g,h)} = \lim_{\Delta t \to 0} X(g') \delta_{\tilde{r}_{\tilde{D}-1}^{+}}^{\hat{r}_{0}^{-}} \delta_{\tilde{r}_{\tilde{D}-1}^{-}}^{r_{0}^{-}} Y(h_{\Delta t}) e^{Z(g',h_{\Delta t})}$$

as they are both zero. If $\delta_{\tilde{r}_{\bar{D}}^+}^{r_0^+} \delta_{\tilde{r}_{\bar{D}}^-}^{r_0^-} = 1$, both $\delta_{\tilde{r}_{\bar{D}}^+}^{r_0^+} \delta_{\tilde{r}_{\bar{D}}^-}^{r_0^-} = \delta_{\tilde{r}_{\bar{D}-1}^+}^{\tilde{r}_0^+} \delta_{\tilde{r}_{\bar{D}-1}^-}^{r_0^-} = 1$ and we just omit the δ symbols in the following statements. We consider the quotient of $X(g)Y(h)e^{Z(g,h)}$ and $X(g')Y(h_{\Delta t})e^{Z(g,h)}$. By definition,

$$\frac{X(g)}{X(g')} = \frac{\mathrm{e}^{-\mathrm{i}\left(\left\langle \tilde{r}_{\tilde{D}}^{+} | H_{0} | \tilde{r}_{\tilde{D}}^{+} \rangle - \left\langle \tilde{r}_{\tilde{D}}^{-} | H_{0} | \tilde{r}_{\tilde{D}}^{-} \right\rangle\right)\left(t - \sum_{j=1}^{\tilde{D}} \tilde{\tau}_{j}\right)\left(-\mathrm{i}\left\langle \tilde{r}_{\tilde{D}}^{+} | H_{0} | \tilde{r}_{\tilde{D}-1}^{+} \right\rangle\right)}{\mathrm{e}^{-\mathrm{i}\left(\left\langle \tilde{r}_{\tilde{D}-1}^{+} | H_{0} | \tilde{r}_{\tilde{D}-1}^{+} \right\rangle - \left\langle \tilde{r}_{\tilde{D}-1}^{-} | H_{0} | \tilde{r}_{\tilde{D}-1}^{-} \right\rangle\left(t - \sum_{j=1}^{\tilde{D}} \tilde{\tau}_{j}\right)\right)}$$

and therefore

$$\lim_{\tilde{\tau}_D \to t - \tilde{\tau}_1 - \dots - \tilde{\tau}_{\tilde{D}-1}} \frac{X(g)}{X(g')} = -i \left\langle \tilde{r}_{\tilde{D}}^+ \middle| H_0 \middle| \tilde{r}_{\tilde{D}-1}^+ \right\rangle.$$

Similarly

$$\frac{Y(h)}{Y(h_{\Delta t})} = \frac{\mathrm{e}^{-\mathrm{i}\left(\left\langle r_{0}^{+} | H_{0} | r_{0}^{+} \right\rangle - \left\langle r_{0}^{-} | H_{0} | r_{0}^{-} \right\rangle\right) \Delta t}}{\mathrm{e}^{\left(\left\langle \hat{r}_{0}^{+} | H_{0} | \hat{r}_{0}^{+} \right\rangle - \left\langle r_{0}^{-} | H_{0} | r_{0}^{-} \right\rangle\right) \Delta t} \left(-\mathrm{i}\left\langle \hat{r}_{0}^{+} | H_{0} | r_{0}^{+} \right\rangle\right)}$$

If

and therefore,

$$\lim_{\Delta t \to 0} \frac{Y(h)}{Y(h_{\Delta t})} = \frac{1}{-\mathrm{i}\left\langle \hat{r}_0^+ \big| H_0 \big| r_0^+ \right\rangle}$$

As for the term Z, it is clear that

$$\lim_{\substack{\tilde{\tau}_D \to t - \tilde{\tau}_1 - \dots - \tilde{\tau}_{\tilde{D}-1} \\ \Delta t \to 0}} \frac{Z(g,h)}{Z(g',h_{\Delta t})} = 1$$

Therefore, (34) also holds for the case $\delta_{\tilde{r}_{D}^{+}}^{r_{0}^{+}} \delta_{\tilde{r}_{D}^{-}}^{r_{0}^{-}} = 1$. The proof is similar for the case when $\widetilde{\operatorname{sgn}}_{\tilde{D}}$ is '-'. Take the sum over \tilde{D} , $(\tilde{r}_{0}^{+}, \tilde{r}_{0}^{-})$ and $\widetilde{\operatorname{sgn}}$, we conclude that (32) equals to (33). Combine all the results and (29) holds with W defined in Proposition 1. Note that it is easy to check when D = 0, (29) holds by regarding the derivative of A with respect to τ_{1} being 0.

Although only spin-boson model is considered in the derivation of the PDE, for more complicated cases, the derivation keeps the same but more terms should be taken into consideration.

4.2. Boundary Conditions. The equation (29) turns out to be a hyperbolic equation with linear advection in the τ_1 direction, and therefore requires initial and boundary conditions to complete the problem statement. While the initial condition has been provided in (21) by choosing t = 0, the boundary conditions are still to be formulated. The advection in (29) shows that the boundary condition should be provided at largest τ_1 for all possible choices of other arguments. Due to the constraint (10), the boundary condition should specify the function values at $\tau_1 = T - \tau_2 - \cdots - \tau_D$. Note that in this case, the corresponding path is not defined in the formula (11) since (τ_1, \dots, τ_D) locates on the boundary of $\Delta_T^{(D)}$. In fact, this corresponds to the path segment h with one or more spin flips occurring exactly at time T. In this special situation, the value of $A_{(r^+, r^-)}^{D, \text{spin}}(t, \boldsymbol{\tau})$ can be expressed by the value of A(t, h') with only D - 1 spin flips in h'. The details are given in the following theorem:

Theorem 9. For $(\tau_1, \cdots, \tau_{D-1}) \in \Delta_T^{(D-1)}$, sgn $\in \{+, -\}^D$ and $(r^+, r^-) \in S^2$, the following equality holds:

(35)
$$A^{D,\operatorname{sgn}}_{(r^+,r^-)}(t,[\tau_1,\cdots,\tau_{D-1},T-\tau_1-\cdots-\tau_{D-1}]) = A^{D-1,\operatorname{sgn}_{1:D-1}}_{(r^+,r^-)}(t,[\tau_1,\cdots,\tau_{D-1}]) \left(-\mathrm{i}\left\langle r_f^+ \left| H_0 \right| \hat{r}_f^+ \right\rangle \delta^+_{\operatorname{sgn}_D} + \mathrm{i}\left\langle \hat{r}_f^- \left| H_0 \right| r_f^- \right\rangle \delta^-_{\operatorname{sgn}_D} \right).$$

where $r_f^{\pm} = r_f^{\pm}(r^+, r^-, D, \text{sgn})$ is defined by (27). In this equation, $\text{sgn}_{k_1:k_2}$ is the k_1 -th to k_2 -th components of sgn.

Proof. It is clear that $(\tau_1, \dots, \tau_{D-1}, T - \tau_1 - \dots - \tau_{D-1}) \in \partial \triangle_T^{(D)}$. According to Theorem 6, we have the following limit:

$$A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1,\cdots,\tau_{D-1},T-\tau_1-\cdots-\tau_{D-1}]) = \lim_{\tau_D \to (T-\tau_1-\cdots-\tau_{D-1})^-} A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1,\cdots,\tau_D]).$$

For simplicity, given $(\tau_1, \dots, \tau_D) \in \Delta_T^{(D)}$, below we use h to denote the path segment defined by (11) and use h' to denote the path segment defined by removing the last spin flip in h:

$$h'(\tau) = \begin{cases} h(\tau), & \text{if } \tau \in [0, \tau_1 + \dots + \tau_D), \\ \lim_{s \to (\tau_1 + \dots + \tau_D)^-} h(s), & \text{if } \tau \in [\tau_1 + \dots + \tau_D, T). \end{cases}$$

Then we have $A_{(r^+,r^-)}^{D,\text{sgn}}(t,[\tau_1,\cdots,\tau_D]) = A(t,h)$ and $A_{(r^+,r^-)}^{D-1,\text{sgn}_{1:D-1}}(t,[\tau_1,\cdots,\tau_{D-1}]) = A(t,h')$. Since the only difference between this two paths is the last segment with length $T - \tau_1 - \cdots - \tau_D$, which vanishes as $\tau_D \to (T - \tau_1 - \cdots - \tau_{D-1})^-$, we conclude that

$$\lim_{\tau_D \to (T - \tau_1 - \dots - \tau_{D-1})^-} Z(g, h) = Z(g, h')$$

for any path $q: [0,t) \to S^2$. In addition,

$$\lim_{\tau_{D} \to (T - \tau_{1} - \dots - \tau_{D-1})^{-}} \frac{Y(h)}{Y(h')} = \lim_{\tau_{D} \to (T - \tau_{1} - \dots - \tau_{D-1})^{-}} \frac{e^{-i(\langle r_{D}^{+} | H_{0} | r_{D}^{+} \rangle - \langle r_{D}^{-} | H_{0} | r_{D}^{-} \rangle)(T - \sum_{j=1}^{D} \tau_{j})} \left(-i \langle r_{D}^{+} | H_{0} | r_{D-1}^{+} \rangle \delta_{\operatorname{sgn}_{D}}^{+} + i \langle r_{D-1}^{-} | H_{0} | r_{D}^{-} \rangle \delta_{\operatorname{sgn}_{D}}^{-}\right)}{e^{-i(\langle r_{D-1}^{+} | H_{0} | r_{D-1}^{+} \rangle - \langle r_{D-1}^{-} | H_{0} | r_{D-1}^{-} \rangle)(T - \sum_{j=1}^{D} \tau_{j})}} = -i \langle r_{D}^{+} | H_{0} | r_{D-1}^{+} \rangle \delta_{\operatorname{sgn}_{D}}^{+} + i \langle r_{D-1}^{-} | H_{0} | r_{D}^{-} \rangle \delta_{\operatorname{sgn}_{D}}^{-}.$$

Therefore, for any path g,

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$$\lim_{\tau_D \to (T-\tau_1-\dots-\tau_{D-1})^-} \frac{X(g)\delta_{\tilde{r}_D^+}^{r_0^-}Y(h)\mathrm{e}^{Z(g,h)}}{X(g)\delta_{\tilde{r}_D^+}^{r_0^+}\delta_{\tilde{r}_D^-}^{r_0^-}Y(h')\mathrm{e}^{Z(g,h')}} = \left(-\mathrm{i}\left\langle r_D^+\right|H_0\big|r_{D-1}^+\right\rangle\delta_{\mathrm{sgn}_D}^+ + \mathrm{i}\left\langle r_{D-1}^-\right|H_0\big|r_D^-\right\rangle\delta_{\mathrm{sgn}_D}^-\right)$$
consequently, the conclusion holds as $r_{\tilde{r}_D^+}^\pm = r_D^\pm$.

and consequently, the conclusion holds as $r_f^{\pm} = r_D^{\pm}$.

In conclusion, the boundary conditions for the functions with D spin flips are determined by the case with one less spin flip. Note that when we allow (τ_1, \dots, τ_D) to take any values on $\partial \Delta_T^{(D)}$, there may be more than one spin flips at time T, which happens if $\tau_1 + \cdots + \tau_D = T$ and $\tau_D = 0$. In this case, the boundary condition can be determined by applying (35) repeatedly. In general, for $k \leq D$, if $\tau_1 + \cdots + \tau_k = T$, we have

$$A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1,\cdots,\tau_k,0,\cdots,0]) = A_{(r^+,r^-)}^{k-1,\mathrm{sgn}_{1:k-1}}(t,[\tau_1,\cdots,\tau_{k-1}]) \prod_{j=k}^{D} \left(-\mathrm{i} \left\langle r_j^+ \big| H_0 \big| \hat{r}_j^+ \right\rangle \delta_{\mathrm{sgn}_j}^+ + \mathrm{i} \left\langle \hat{r}_j^- \big| H_0 \big| r_j^- \right\rangle \delta_{\mathrm{sgn}_j}^- \right),$$

where r_j is the *j*-th state in the path segment:

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$$r_j = r_f(r^+, r^-, j, \operatorname{sgn}_{1:j}).$$

By now, with the equation (29) and the boundary conditions (35), a complete system of infinite partial differential equations has been formulated.

4.3. Truncation and Closure of the System. The system (29) includes infinite equations since D can take the value of any non-negative integer. However, for practical numerical computation, we can only solve finite equations, which requires a truncation of the system. Physically, due to the finite spin flipping rate, the probability that the system undergoes D spin flips generally follows the Poisson distribution. Therefore when D is large, the contribution from $A_{(r^+,r^-)}^{D,\text{sgn}}$ can be regarded as small, so that it is reasonable to discard evolution equations for $A_{(r^+,r^-)}^{D,\text{sgn}}$ with large D. This can also be justified mathematically since

$$\left\|\sum_{D=\overline{D}+1}^{\infty}\sum_{(r^+,r^-)\in\mathcal{S}^2}\sum_{\mathrm{sgn}\in\{+,-\}^D}\int_{\triangle_T^{(D)}}A^{D,\mathrm{sgn}}_{(r^+,r^-)}(t,[\tau_1,\cdots,\tau_D])\left|r_f^+\right\rangle\!\!\left\langle r_f^-\right|\mathrm{d}\boldsymbol{\tau}\right\|\to 0,\quad\mathrm{as}\quad\overline{D}\to\infty,$$

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which has been shown in the proof of Theorem 7. Below in our numerical method, we will only focus on the equations of $A_{(r^+,r^-)}^{D,\text{sgn}}$ with $D \in \{0, 1, \dots, D_{\text{max}}\}$ for some D_{max} chosen suitable for the problem.

However, the equation (29) shows that the evolution of $A^{D_{\max},\text{sgn}}_{(r^+,r^-)}$ depends on the values of $A^{D_{\max}+1,\text{sgn}}_{(r^+,r^-)}$, meaning that our truncation does not form a closed system, which requires us to impose an appropriate closure. Precisely speaking, we need to provide some predictions of $A^{D_{\max}+1,\text{sgn}}_{(r^+,r^-)}$ based on the values of $A^{D,\text{sgn}}_{(r^+,r^-)}$ with $D \leq D_{\max}$. Our approach is to estimate the values of $A^{D_{\max}+1,\text{sgn}}_{(r^+,r^-)}$ via interpolations based on Theorem 9 and the following proposition Theorem 9 and the following proposition.

Proposition 10. For any (r^+, r^-) , $D \ge 2$ and sgn with $\operatorname{sgn}_k = \operatorname{sgn}_{k+1}$, if $\tau_k = 0$,

$$A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1,\cdots,\tau_D]) = -A_{(r^+,r^-)}^{D-2,[\mathrm{sgn}_{1:k-1},\mathrm{sgn}_{k+2:D}]}(t,[\tau_1,\cdots,\tau_{k-2},\tau_{k-1}+\tau_{k+1},\tau_{k+2},\cdots,\tau_D]) \langle +1|H_0|-1\rangle \langle -1|H_0|+1\rangle.$$

Proof. It is clear that $(\tau_1, \dots, \tau_D) \in \partial \triangle_T^{(D)}$. By Theorem 6, the following limit holds:

$$A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1,\cdots,\tau_D]) = \lim_{\Delta t \to 0^-} A_{(r^+,r^-)}^{D,\mathrm{sgn}}(t,[\tau_1,\cdots,\tau_{k-1},\Delta t,\tau_{k+1}-\Delta t,\tau_{k+2},\cdots,\tau_D]) := \lim_{\Delta t \to 0^-} A(t,h_{\Delta t}),$$

and for simplicity, we denote the path of $A_{(r^+,r^-)}^{D-2,[\operatorname{sgn}_{1:k-1},\operatorname{sgn}_{k+2:D}]}(t,[\tau_1,\cdots,\tau_{k-2},\tau_{k-1}+\tau_{k+1},\tau_{k+2},\cdots,\tau_D])$ as h'. The only difference between the path $h_{\Delta t}$ and h is in the interval $\left[\sum_{j=1}^k \tau_j, \sum_{j=1}^k \tau_j + \Delta t\right)$. Therefore, similar to the proof of Theorem 9, we have $\lim_{\Delta t \to 0^-} Z(g,h_{\Delta t}) = Z(g,h')$ for any path $g:[0,t) \to S^2$. In addition, similar to the proof of Theorem 9, we have

$$\lim_{\Delta t \to 0} \frac{Y(h_{\Delta t})}{Y(h')} = -\langle +1|H_0|-1\rangle \langle -1|H_0|+1\rangle$$

Consequently, we have

$$\lim_{\Delta t \to 0} \frac{X(g) \delta_{\vec{r}_{D}^{+}}^{r_{0}^{+}} \delta_{\vec{r}_{D}^{-}}^{r_{0}^{-}} Y(h_{\Delta t}) Z(g, h_{\Delta t})}{X(g) \delta_{\vec{r}_{D}^{+}}^{r_{0}^{+}} \delta_{\vec{r}_{D}^{-}}^{r_{0}^{-}} Y(h') Z(g, h')} = -\langle +1|H_{0}|-1\rangle \langle -1|H_{0}|+1\rangle$$

for all path g and the result holds.

The proposition above indicates that if two spin flips occur at the same time on the same branch, the function value of this path segment can be represented using the function value of the path segment with both spin flips removed. Therefore, this proposition establishes the relationship between the function values for path segments with different values of D. To demonstrate how this is applied to derive the closure of the system, we start with an example with $D_{\text{max}} = 5$, and we try to estimate $A_{(-1,-1)}^{6,[+,+,-,-,+,-]}(t, [0, 1.0, 0.5, 0.5, 0.5, 1.0])$ (see Figure 2). To this end, we consider the time point of the



last spin flip at t = 3.5 on the negative branch as a variable σ while fixing all other spin flips. According to the structure of h^- , the value of σ can take any number between t = 2 and t = 4. When $\sigma = 2$, we obtain a path with two spin flips at the same time point, which is illustrated in Figure 3, and the corresponding value of A(t, h) is represented by $A_{(-1,-1)}^{6,[+,+,-,-,-,+]}(t, [0, 1.0, 0.5, 0.5, 0.5, 0.5])$. Similarly, when $\sigma = 4$, we get $A_{(-1,-1)}^{6,[+,+,-,-,+,-]}(t, [0, 1.0, 0.5, 0.5, 0.5, 1.5])$ (see Figure 4). The idea of our closure is to use the following linear interpolation as an approximation:

$$\begin{split} &A^{6,[+,+,-,-,+,-]}_{(-1,-1)}(t,[0,1.0,0.5,0.5,0.5,1.0])\\ &\approx \frac{1}{4}A^{6,[+,+,-,-,-,+]}_{(-1,-1)}(t,[0,1.0,0.5,0.5,0.5,0.5]) + \frac{3}{4}A^{6,[+,+,-,-,+,-]}_{(-1,-1)}(t,[0,1.0,0.5,0.5,0.5,1.5]) \end{split}$$

By Proposition 10, the value of $A_{(-1,-1)}^{6,[+,+,-,-,-,+]}(t, [0, 1.0, 0.5, 0.5, 0, 0.5])$ can be directly obtained from the value of $A_{(-1,1)}^{4,[+,+,-,+]}(t, [0, 1.0, 0.5, 1.0])$. In addition, the boundary condition (35) in Theorem 9 allows us to compute the value of $A_{(-1,-1)}^{6,[+,+,-,-,+]}(t, [0, 1.0, 0.5, 0.5, 0.5, 1.5])$ by the value of $A_{(-1,-1)}^{5,[+,+,-,-,+]}(t, [0, 1.0, 0.5, 0.5, 0.5])$.



FIGURE 3. The illustration of the path segment in $A_{(-1,-1)}^{6,[+,+,-,-,-,+]}(t, [0, 1.0, 0.5, 0.5, 0, 0.5])$.



In summary, our final estimation is

$$\begin{split} &A_{(-1,-1)}^{6,[+,+,-,-,+,-]}(t,[0,1.0,0.5,0.5,0.5,1.0])\\ \approx &-\frac{1}{4}\left<+1|H_0|-1\right>\left<-1|H_0|+1\right>A_{(-1,-1)}^{4,[+,+,-,+]}(t,[0,1.0,0.5,1.0])\\ &+\frac{3}{4}\mathrm{i}\left<-1|H_0|+1\right>A_{(-1,-1)}^{5,[+,+,-,-,+]}(t,[0,1.0,0.5,0.5,0.5]). \end{split}$$

Therefore, we estimate the value of $A_{(-1,-1)}^{6,[+,+,-,-,+,-]}(t, [0, 1.0, 0.5, 0.5, 0.5, 1.0])$ from those with $D \leq D_{\max}$. The idea can be generalized to any paths with $D_{\max} + 1$ spin flips in a straightforward way. In the example

The idea can be generalized to any paths with $D_{\max} + 1$ spin flips in a straightforward way. In the example above, we choose to shift the last spin flip of negative branch. Clearly, the same approach also works for the positive branch. When the negative branch is chosen, σ can take values in the interval [2, 4], which has a length of 2. If the positive branch is chosen, the corresponding interval will have a length of 3. In this case, we prefer to choose the negative branch since the interpolation on a shorter interval is more likely to get a better approximation. Technically, to determine which branch to choose, we just need to look at the last three signs of sgn. If at least two of them are positive/negative, we choose the positive/negative branch to formulate the closure. This approach applies to any case with $D_{\max} \ge 2$.

In general, when a path segment h has D spin flips with $D = D_{\max} + 1$, after selecting the branch by the aforementioned method, we approximate A(t, h) by linear interpolation. In the following discussion, we assume that the positive branch is selected. The procedure is the same if the negative branch is selected. According to the branch selection, there must be at least two spin flips on the chosen branch $(h^+ \text{ according}$ to our assumption). Let σ_0 and σ_* be the locations of the last two spin flips on h^+ , and $\sigma_* < \sigma_0$. If we move the last spin flip on h^+ to a different location $\sigma \in (\sigma_*, T)$, we obtain a new path $h_{\sigma} = (h_{\sigma}^+, h_{\sigma}^-) : [0, T) \to S^2$ defined by

$$h_{\sigma}^{+}(\tau) = \begin{cases} h^{+}(\tau), & \text{if } \tau < \sigma^{*} \\ h^{+}(\sigma_{*}), & \text{if } \sigma_{*} \leqslant \tau < \sigma \\ h^{+}(T^{-}), & \text{if } \sigma \leqslant \tau < T \end{cases}$$
$$h_{\sigma}^{-}(\tau) = h^{-}(\tau).$$

where $h^+(T^-) = \lim_{s \to T^-} h^+(s)$. Clearly, we have $h = h_{\sigma_0}$ and thus we can approximate A(t,h) by the following linear interpolation:

(36)
$$A(t,h) = A(t,h_{\sigma_0}) \approx \frac{T - \sigma_0}{T - \sigma_*} A(t,h_{\sigma_*}) + \frac{\sigma_0 - \sigma_*}{T - \sigma_*} A(t,h_T).$$

According to Theorem 9 and Proposition 10, we have

(37)
$$A(t,h_T) = -iA(t,h') \left\langle \hat{h}^+(T^-) \middle| H_0 \middle| h^+(T^-) \right\rangle, \qquad A(t,h_{\sigma_*}) = -A(t,h'') \left\langle +1 \middle| H_0 \middle| -1 \right\rangle \left\langle -1 \middle| H_0 \middle| +1 \right\rangle.$$

where the paths $h' = (h'^+, h'^-)$ and $h'' = (h''^+, h''^-)$ are given by

$$h'^{+}(\tau) = \begin{cases} h^{+}(\tau), & \text{if } \tau < \sigma^{*}, \\ h^{+}(\sigma_{*}), & \text{if } \sigma_{*} \leqslant \tau < T, \end{cases} \qquad h''^{+}(\tau) = \begin{cases} h^{+}(\tau), & \text{if } \tau < \sigma^{*}, \\ \lim_{s \to \sigma_{*}^{-}} h^{+}(s), & \text{if } \sigma_{*} \leqslant \tau < T, \end{cases}$$
$$h''^{-}(\tau) = h^{-}(\tau), \qquad h''^{-}(\tau) = h^{-}(\tau).$$

It can be seen that h' has one less spin flip than h, and h'' has two less spin flips than h. Therefore both A(t, h') and A(t, h'') are included in the truncated system. Plugging (37) into (36) yields an estimated value of A(t, h), which forms the closure of the PDE system (29).

In the next section, we will find the observable of the system by solve the PDE system numerically. We call this approach differential equation based path integral (DEBPI) method.

Remark. Here we would like to comment on the relationship between the i-QuAPI scheme and the PDE system (29). According to the the iteration (5), i-QuAPI considers $2^{2\Delta k}$ equations in the system. However, the i-QuAPI method does not perform the truncation by simply choosing a maximum value of D. Instead, it considers path segments with at most Δk spin flips on each branch, so that the maximum spin flips in the path segment can reach $2\Delta k$, but not all path segments with $2\Delta k$ spin flips are taken into account. Besides, the treatment of both boundary condition and closure of the system can be observed from (5). The boundary condition is needed when updating the value of A for path segments with a spin flip near the end of the path, which corresponds to the case $S_k \neq S_{k-1}$. In this case, such a spin flip is simply dropped on the right-hand side of (5), which is similar to the right-hand side of our boundary condition (35). The extra bra-ket terms in (35) appear in the i-QuAPI method as the bra-ket terms in $I(S_k, S_{k-1})$ defined in (6). As for the closure, according to the discretization of the i-QuAPI method, the closure is required only when there are Δk spin flips on either of the branches, and in this case, we have $\tau_1 = \Delta t$ and $\tau_1 + \cdots + \tau_D = T - \Delta t$. According to (29), one needs the values of $A_{(r^+, r^-)}^{D+1,[\pm, \text{sgn}]}(t, [0, \tau_1, \cdots, \tau_D])$ to close the system. From this point of view, the approach of i-QuAPI is to assume that

$$\begin{aligned} A_{(r^+,r^-)}^{D+1,[\pm,\mathrm{sgn}]}(t,[0,\tau_1,\cdots,\tau_D]) &= A_{(r^+,r^-)}^{D+1,[\pm,\mathrm{sgn}]}(t,[\Delta t,\tau_1,\cdots,\tau_D]) \\ &= A_{(r^+,r^-)}^{D,[\pm,\mathrm{sgn}_{1:D-1}]}(t,[\Delta t,\tau_1,\cdots,\tau_{D-1}]) \left(-\mathrm{i}\left\langle r_f^+ \left| H_0 \right| \hat{r}_f^+ \right\rangle \delta_{\mathrm{sgn}_D}^+ + \mathrm{i}\left\langle \hat{r}_f^- \left| H_0 \right| r_f^- \right\rangle \delta_{\mathrm{sgn}_D}^- \right), \end{aligned}$$

where the boundary condition (35) has been applied. Based on such a system closure, the i-QuAPI method can be considered as a numerical scheme of (29).

5. Numerical Experiments

To verify the correctness of the DEBPI method and show the possible memory savings by solving (29) directly, we present several numerical experiments in this section. The numerical method to solve (29) generally follows the standard numerical solvers of hyperbolic systems, which will be briefly introduced in Section 5.1. The numerical results will be exhibited and discussed in Section 5.2.

5.1. Numerical Method. The general structure of our numerical scheme is based on the Strang splitting method to deal with the zeroth- and first-order terms on the right-hand side of (29) separately. In general, for a differential equation having the form

$$\frac{\partial}{\partial t}u = L_1 u + L_2 u$$

with L_1 and L_2 being two linear operators, Strang splitting approximates the solution by

$$u(t + \Delta t) \approx e^{L_1 \Delta t/2} e^{L_2 \Delta t} e^{L_1 \Delta t/2} u(t).$$

In our case, we need to focus on the numerical solver of the following two equations:

$$(38) \qquad \frac{\partial}{\partial t} A^{D,\mathrm{sgn}}_{(s^+,s^-)}(t, [\tau_1, \cdots, \tau_D]) = \frac{\partial}{\partial \tau_1} A^{D,\mathrm{sgn}}_{(s^+,s^-)}(t, [\tau_1, \cdots, \tau_D]); (39) \qquad \frac{\partial}{\partial t} A^{D,\mathrm{sgn}}_{(s^+,s^-)}(t, [\tau_1, \cdots, \tau_D]) = -W^{D,\mathrm{sgn}}_{(s^+,s^-)}([\tau_1, \cdots, \tau_D]) A^{D,\mathrm{sgn}}_{(s^+,s^-)}(t, [\tau_1, \cdots, \tau_D]) + A^{D+1, [-,\mathrm{sgn}]}_{(s^+,s^-)}(t, [0, \tau_1, \cdots, \tau_D]) + A^{D+1, [+,\mathrm{sgn}]}_{(\hat{s}^+,s^-)}(t, [0, \tau_1, \cdots, \tau_D]).$$

Thus, evolving (29) by one time step Δt consists of solving (38) by $\Delta t/2$, and then solving (39) by Δt , followed by solving (38) again by $\Delta t/2$.

For both equations, a uniform mesh is used to discretize the domain $\overline{\Delta_T^{(D)}}$. Note that this is not needed for D = 0 since the path segment h is fully determined by its initial state. When D > 0, the sets of grid points are

$$\mathcal{P}_D = \left\{ \left(\frac{m_1 h_s}{N}, \cdots, \frac{m_D h_s}{N}\right); m_k \in \mathbb{Z}^+, \sum_{k=1}^D m_k < N \right\}, \quad D = 1, \cdots, D_{\max}.$$

where $h_s = T/N$ is the grid size. For example, Figure 5 shows the grid points in the case of D = 2, N = 5 and D = 3, N = 4. We do not place grid points on the hypotenuse since their values are determined by the boundary condition as discussed in Theorem 9.



FIGURE 5. The grid points on the two-dimensional simplex (red points) with N = 5 and three-dimensional simplex (points of all colors) with N = 4.

The τ_1 -derivative in (38) is discretized by the finite difference method. As (38) is a hyperbolic equation with linear advection, we apply second-order upwind scheme and Lax-Wendroff method for the τ_1 -derivative. Although there are no grid points on the hypotenuse, we still can calculate the values of A on the hypotenuse with Theorem 9. Therefore, the τ_1 -derivative of all points with $\tau_1 + \cdots + \tau_D \leq N - 2$ can be estimated by second-order upwind scheme. As for the grid points with $\tau_1 + \cdots + \tau_D = N - 1$, we apply Lax-Wendroff method for the estimation of τ_1 -derivative. (39) can be regarded as an ODE with respect to t. Therefore, we simply employ Runge-Kutta method for the numerical solution of (39). Note that the estimation for the terms with $D_{\text{max}} + 1$ spin flips is discussed in Section 4.3.

As a summary, we provide the outline of our algorithm below:

- For each set of parameters $D = 0, \dots, D_{\max}, (s^+, s^-) = (\pm 1, \pm 1)$ and $p \in \mathcal{P}_D$ compute $A^{D, \text{sgn}}_{(s^+, s^-)}(0, p)$ according to (21).
- Assume that we already have all values of $A_{(s^+,s^-)}^{D,\text{sgn}}(t,p)$ for $D = 0, \dots, D_{\text{max}}$ all possible choices of (s^+,s^-) and $p \in \mathcal{P}_D$ at specific time t. We evolve the system to obtain corresponding values of at time $t + \Delta t$ by the following steps.

- Evolve the system (38) by a half time step $\Delta t/2$ using the second-order upwind scheme (for the points with $\tau_1 + \cdots + \tau_D \leq N-2$) or Lax-Wendroff method (for the points with $\tau_1 + \cdots + \tau_D = N-1$). If points on hypotenuse (the sum of all components are T) are involved, boundary condition (35) is applied.
- Evolve the system (39) by a full time step Δt . If points on the $(D_{\text{max}} + 1)$ -dimensional simplex is involved, the method to close the system in Section 4.3 is applied.
- Evolve the system (38) by another half time step $\Delta t/2$ using the second-order upwind scheme or Lax-Wendroff method again.
- Compute the density matrix at time $t + \Delta t$ according to (28) and calculate the observable by $\langle O(t) \rangle = \operatorname{tr}(O\rho_s(t)).$
- Repeat the previous step to evolve the system iteratively.

Here the first step prepares the initial data, which requires numerical integration on $\overline{\Delta_T^{(D)}}$. For the evolution of numerical solution, we fix the time step Δt so that the solvers of both equations (38)(39) can be written as matrix-vector multiplications, and the matrices are the same for all steps.

At the end of this subsection, we would like to discuss the memory cost of our method. For a *D*-dimensional simplex with *N* nodes on each edge, there are $\binom{N+D}{D}$ grid points in one simplex. Besides, the initial state of *h*, *i.e.* the subscript (r^+, r^-) of *A*, has four different possibilities for the spin-boson model; the branches of the spin flips, *i.e.* sgn in the superscript of *A*, can take 2^D choices. Therefore, in total, we need

(40)
$$N_{\rm dof} = 4 \sum_{D=0}^{D_{\rm max}} 2^D \begin{pmatrix} N+D\\D \end{pmatrix}$$

degrees of freedom to store the numerical solution. To compare this with the i-QuAPI method, we first recall that i-QuAPI needs to store all values of the map $A_l : S^{2\Delta k} \to \mathbb{C}$ in each iteration, so the memory cost is $2^{2\Delta k}$. Assume that both methods have same truncation time and grid size, we then have $\Delta k = N$. Thus we need to compare N_{dof} and 2^{2N} . To this end, we estimate N_{dof} using Stirling's approximation $k! \sim \sqrt{2\pi} e^{-k} k^{k+1/2}$:

$$\begin{split} N_{\rm dof} &< 4(D_{\rm max}+1)2^{D_{\rm max}} \begin{pmatrix} N+D_{\rm max} \\ D_{\rm max} \end{pmatrix} \\ &\lesssim \frac{4(D_{\rm max}+1)2^{D_{\rm max}}}{D_{\rm max}!} \frac{(N+D_{\rm max})^{N+D_{\rm max}+1/2}}{{\rm e}^{D_{\rm max}}N^{N+1/2}} \\ &\lesssim \frac{4(D_{\rm max}+1)2^{D_{\rm max}}}{D_{\rm max}!} (N+D_{\rm max})^{D_{\rm max}}. \end{split}$$

It is now clear that if $D_{\text{max}} \ll N$, the value of N_{dof} can be significantly less than 2^{2N} so that our DEPBI method will require less memory than the i-QuAPI method.

5.2. Numerical Results. In all our experiments, the frequencies ω_j are distributed in $[0, \omega_{\text{max}}]$ following the Poisson distribution:

$$\omega_j = -\omega_c \ln\left(1 - \frac{j}{L}(1 - \exp(-\omega_{\max}/\omega_c))\right), \quad j = 1, \cdots, L$$

where L is the number of harmonic oscillators. The coupling intensity c_i is

$$c_j = \omega_j \sqrt{\frac{\xi \omega_c}{L} (1 - \exp(-\omega_{\max}/\omega_c))}, \quad j = 1, \cdots, L.$$

These parameters correspond to the bath with Ohmic spectral density [15]. The number of harmonic oscillators L is chosen to be 200, and the maximum frequency is chosen as $\omega_{\text{max}} = 4\omega_c$. For the DEPBI method, the time step Δt is fixed as 1/80 in all cases. Other parameters will be specified for each experiment.

5.2.1. Experiments with different coupling intensities. In order to check the validity of our method, we first study the following parameters, which have been considered in [7, 1]:

$$\Delta = 1, \quad \omega_c = 2.5\Delta, \quad \beta = 5/\Delta, \quad \epsilon = 0.$$

The numerical results for $\xi = 0.2$ and 0.4 are given in Figure 6. The results of our method is compared with the i-QuAPI results, and the parameters used for both methods are given in the caption. The results correctly show that when the bath-system coupling is stronger, the fluctuation of the observable damps faster. In both cases, the evolution of the observable matches well with each other, showing reliability of our approach.



FIGURE 6. Both methods use truncation time T = 1.5. For i-QuAPI method, Δk is chosen to be 10. For c-QuAPI, $D_{\text{max}} = 8$ and N = 8.

According to (40), it can be calculated that the DEBPI method needs to store $N_{dof} = 17444860$ different values of A(t, h) for each time step. In comparison, the i-QuAPI method, which has a smaller grid size, only requires to store $2^{2\times10} = 1048576$ different values. To understand such a difference, we note that each discrete path segment in the i-QuAPI method can also be represented using the initial state (r^+, r^-) , the number of spin flips D, the branches of the spin flips sgn, and the time difference between each pair of spin flips (τ_1, \dots, τ_D) . However, for the i-QuAPI method, there are never more than one spin flip occurring at the same time on the same branch. In other words, if $\operatorname{sgn}_k = \operatorname{sgn}_{k+1}$, then τ_{k+1} must be nonzero. For example, when D = 2 and $\operatorname{sgn}_1 = \operatorname{sgn}_2$, the five points located on the vertical axis of Figure 5a are not considered in the i-QuAPI method, leading to a lower memory cost than that in our discretization. For larger D, this will cause more significant difference in the memory usage even if the grid sizes for both methods are the same. This difference can be eliminated by using a smarter PDE solver, which will be studied to our future work.

Based on our current approach, the memory cost grows exponentially with D_{max} . Therefore, if the problem setting allows a lower value of D_{max} , the DEBPI method will show its advantage. Moreover, if D_{max} can be chosen small, we can use a small grid size in the discretization of $\Delta_T^{(D)}$ to achieve better accuracy, while for the i-QuAPI method, the memory cost grows exponentially with Δk , so that it is prohibitive to choose small grid sizes. In the following two subsections, we will show several cases with small spin-flipping frequency such that our method can achieve a lower memory cost.

5.2.2. Experiments with different biases. We now consider the following set of parameters:

$$\xi = 0.2, \quad \Delta = 0.2, \quad \omega_c = 5\Delta, \quad \beta = 5/\Delta,$$

where the spin flipping frequency Δ is smaller so we expect that a smaller D_{max} can be adopted for our DEBPI approach. Three different biases ϵ are chosen, and the results are given in Figure 7. For $D_{\text{max}} = 5$, the curves for the DEPBI method almost coincide with the i-QuAPI results. Since the parameter ϵ denotes the difference between the energies of the two states, the three figures correctly show that when ϵ is larger, the state of spin is more likely to be found as $|-1\rangle$, leading to the downshift of the curve in the second and third subplots in Figure 7.



FIGURE 7. Both methods use truncation time T = 4. For i-QuAPI method, Δk is chosen to be 10 and for c-QuAPI, $D_{\text{max}} = 5$ and N = 10.



FIGURE 8. $\epsilon = 0$ and $D_{\text{max}} = 3, 4, 5$.

In these tests, DEBPI needs $\sum_{D=0}^{5} 4 \times 2^{D} {D+10 \choose D} = 458748$ numbers to store the numerical solution, while the i-QuAPI scheme needs $2^{2\times 10} = 1048576$. Thus the memory cost of our method is about a half of

while the i-QuAPI scheme needs $2^{2\times 10} = 1048576$. Thus the memory cost of our method is about a half of the cost of i-QuAPI. Note that further reducing D_{max} may cause significant error in the numerical solution. Figure 8 shows the numerical results for $D_{\text{max}} = 3, 4, 5$ when $\epsilon = 0$, indicating that $D_{\text{max}} = 5$ is a proper choice to guarantee the quality of the solution.

In general, the parameter D_{max} can be considered as a factor controlling the trade-off between the memory cost and the numerical accuracy. In order that D_{max} can be chosen small, we need both the memory time and the spin flipping frequency to be relatively small, meaning that within the path segment T, the spin is unlikely to flip many times. Under such circumstances, our method is more likely to outperform the method of i-QuAPI in terms of the memory cost.

5.2.3. Experiments with different temperatures. For the last set of tests, we choose the parameters

$$\xi = 0.2, \quad \Delta = 0.1, \quad \omega_c = 2.5\Delta, \quad \epsilon = 0,$$

and we let the inverse temperature of the bath β vary from $0.2/\Delta$ to $5/\Delta$. The results and the numerical parameters are given in Figure 9. As reflected in the numerical results, in the case of higher temperature, stronger quantum dissipation leads to faster reduction to the state with equal probabilities on both spin states.



FIGURE 9. Both methods use truncation time T = 4. For i-QuAPI method, Δk is chosen to be 10 and for c-QuAPI, $D_{\text{max}} = 3$ and N = 15.

Compared with the examples in Section 5.2.2, we use the same memory length T = 4, while the spin flipping frequency Δ is reduced by a half. Thus it can be expected that the value of D_{max} can also be reduced by a half. Therefore, choosing $D_{\text{max}} = 3$ is sufficient to capture the behavior of evolution processes here. In these examples, due to the small value of D_{max} , the memory cost of the DEBPI approach is much lower even if N is chosen to be greater than Δk . In detail, the DEBPI approach only requires to store $\sum_{D=0}^{3} 4 \times 2^{D} {D \choose D} = 28420$ values, while the i-QuAPI scheme requires $2^{2 \times 10} = 1048576$.

6. CONCLUSION AND DISCUSSION

We have formulated a PDE system for open quantum systems with truncated memory length in the bathinfluence functional. The PDE system is supplemented with proper initial and boundary conditions, and truncated with a reasonable closure. Such a formulation allows us to solve the density matrix using classical PDE-solvers for hyperbolic systems. By this approach, the memory cost can be saved in two possible ways. First, higher-order schemes can be applied to reduce the number of grid points. Second, in some situations, we can choose D_{max} to be smaller than N to avoid discretization of high-dimensional simplicies. In our numerical tests, we considered the second-order numerical schemes (the same as i-QuAPI), and illustrated several cases where D_{max} can be chosen small. The higher-order schemes are being studied in our ongoing work.

While this paper focuses mainly on the spin-boson model, the idea of deriving the governing differential equations can be generalized to any quantum system with finite states without much difficulty. When S contains more elements, the representation of the path segments will be slightly more complicated. In detail, the path segment h cannot be fully determined by the initial state and the locations of state hops. Instead, it is necessary to specify which state the system hops to at each τ_k . Nevertheless, the idea to derive the PDE system and the expression of density matrix stays the same. Roughly speaking, the equation of A(t, h) holds the form

(41)
$$\frac{\partial}{\partial t}A(t,h) = -W(h)A(t,h) + \sum_{\hat{h}}A(t,\hat{h}) + \frac{\partial}{\partial \tau_1}A(t,h),$$

where τ_1 is the location of the first state hop in h, and the path segment \hat{h} takes all possible paths satisfying $\hat{h}(\tau) = h(\tau)$ for all $\tau \in (0, T)$ and

$$\hat{h}^+(0) = \hat{h}^+(0) \text{ and } \hat{h}^-(0) \neq \hat{h}^-(0) \text{ or } \hat{h}^+(0) \neq \hat{h}^+(0) \text{ and } \hat{h}^-(0) = \hat{h}^-(0).$$

The equation (41) is essentially the same as (29) in the spin-boson case. However, when the cardinality of S is larger, the memory cost also grows faster with D_{max} and N, requiring more efficient numerical methods for the simulation. This will be considered in our future works.

APPENDIX. DERIVATION OF I-QUAPI METHOD

A component of the reduced density matrix ρ_s at time $t = N\Delta t$, denoted by $\langle s'' | \rho_s(N\Delta t) | s' \rangle$, can be represented by

$$\langle s'' | \rho_s(N\Delta t) | s' \rangle = \operatorname{tr}_b \langle s'' | \mathrm{e}^{-\mathrm{i}HN\Delta t} \rho(0) \mathrm{e}^{\mathrm{i}HN\Delta t} | s' \rangle.$$

With the completeness relation in a quantum system

$$\int_{\mathcal{S}} |s\rangle \langle s| \, \mathrm{d}s = \mathrm{id}_s$$

with \mathcal{S} being the set of an orthonormal basis, the reduced density matrix can be written as a path integral form

$$\begin{split} \langle s^{\prime\prime} | \rho_s(N\Delta t) | s^\prime \rangle = \mathrm{tr}_b \int_{\mathcal{S}} \mathrm{d} s_0^+ \cdots \int_{\mathcal{S}} \mathrm{d} s_{N-1}^+ \int_{\mathcal{S}} \mathrm{d} s_0^- \cdots \int_{\mathcal{S}} \mathrm{d} s_{N-1}^+ \left\langle s^{\prime\prime} | \mathrm{e}^{-\mathrm{i}H\Delta t} | s_{N-1}^+ \right\rangle \\ & \left\langle s_{N-1}^+ | \mathrm{e}^{-\mathrm{i}H\Delta t} | s_{N-2}^+ \right\rangle \cdots \left\langle s_1^+ | \mathrm{e}^{-\mathrm{i}H\Delta t} | s_0^- \right\rangle \left\langle s_0^- | \rho(0) | s_0^- \right\rangle \\ & \left\langle s_0^- | \mathrm{e}^{\mathrm{i}H\Delta t} | s_1^- \right\rangle \cdots \left\langle s_{N-2}^+ | \mathrm{e}^{\mathrm{i}H\Delta t} | s_{N-1}^- \right\rangle \left\langle s_{N-1}^- | \mathrm{e}^{\mathrm{i}H\Delta t} | s^\prime \right\rangle. \end{split}$$

(42) is called path integral as the states s_0^+, \dots, s_N^+ and s_0^-, \dots, s_N^- can be regarded as a path, an integral over all possible paths are taken here. It is clear that the path has two branches (positive branch and negative branch).

For the spin-boson model, $S = \{-1, +1\}$, therefore, Equation (42) becomes

$$\langle s'' | \rho_s(N\Delta t) | s' \rangle = \operatorname{tr}_b \sum_{s_0^+ = \pm 1} \cdots \sum_{s_{N-1}^+ = \pm 1} \sum_{s_0^- = \pm 1} \cdots \sum_{s_{N-1}^- = \pm 1} \langle s'' | e^{-iH\Delta t} | s_{N-1}^+ \rangle$$

$$\langle s_{N-1}^+ | e^{-iH\Delta t} | s_{N-2}^+ \rangle \cdots \langle s_1^+ | e^{-iH\Delta t} | s_0^+ \rangle \langle s_0^+ | \rho(0) | s_0^- \rangle$$

$$\langle s_0^- | e^{iH\Delta t} | s_1^- \rangle \cdots \langle s_{N-2}^+ | e^{iH\Delta t} | s_{N-1}^- \rangle \langle s_{N-1}^- | e^{iH\Delta t} | s' \rangle .$$

As for the special Hamiltonian given by (2), Trotter splitting can be used by introducing the following reference Hamiltonian

(42)
$$H_0 = \left(\epsilon \hat{\sigma}_z + \Delta \hat{\sigma}_x - \sum_j \frac{(c_j \hat{\sigma}_z)^2}{2\omega_j^2}\right) \otimes \mathrm{id}_b$$

Therefore, the remaining part is

$$H - H_0 = \sum_j \left(\mathrm{id}_s \otimes \frac{\hat{p}_j^2}{2} + \frac{1}{2} \omega_j^2 \left(\mathrm{id}_s \otimes \hat{q}_j - \frac{c_j}{\omega_j^2} \hat{\sigma}_z \otimes \mathrm{id}_b \right)^2 \right).$$

When the splitting is applied, the reduced density matrix can be written as

$$\begin{split} \langle s'' | \rho_s(N\Delta t) | s' \rangle &= \sum_{s_0^+ = \pm 1} \cdots \sum_{s_{N-1}^+ = \pm 1} \sum_{s_0^- = \pm 1} \cdots \sum_{s_{N-1}^- = \pm 1} \left\langle s'' | e^{-iH\Delta t} | s_{N-1}^+ \right\rangle \\ &\left\langle s_{N-1}^+ | e^{-iH\Delta t} | s_{N-2}^+ \right\rangle \cdots \left\langle s_1^+ | e^{-iH\Delta t} | s_0^+ \right\rangle \left\langle s_0^+ | \rho_s(0) | s_0^- \right\rangle \\ &\left\langle s_0^- | e^{iH\Delta t} | s_1^- \right\rangle \cdots \left\langle s_{N-2}^+ | e^{iH\Delta t} | s_{N-1}^- \right\rangle \left\langle s_{N-1}^- | e^{iH\Delta t} | s' \right\rangle \\ &F(s_0^+, s_1^+, \cdots, s_N^+, s_0^-, s_1^-, \cdots, s_N^-) \end{split}$$

with $s_N^+ = s'', s_N^- = s'$ and

$$\begin{split} F(s_0^+, s_1^+, \cdots, s_N^+, s_0^-, s_1^-, \cdots, s_N^-) \\ &= \operatorname{tr}_b \left[e^{-\mathrm{i}\frac{\Delta t}{2}(H - H_0(s''))} e^{-\mathrm{i}\Delta t(H - H_0(s_{N-1}^+))} \cdots e^{-\mathrm{i}\Delta t(H - H_0(s_1^+))} \right. \\ & e^{-\mathrm{i}\frac{\Delta t}{2}(H - H_0(s_0^+))} \rho_b(0) e^{\mathrm{i}\frac{\Delta t}{2}(H - H_0(s_0^-))} e^{\mathrm{i}\Delta t(H - H_0(s_1^-))} \cdots \\ & e^{\mathrm{i}\Delta t(H - H_0(s_{N-1}^-))} e^{\mathrm{i}\frac{\Delta t}{2}(H - H_0(s'))} \right]. \end{split}$$

The function F is called influence functional, calculating the influence of the environment on the system of interest. With (42) as reference Hamiltonian, the influence functional can be computed analytically [5][14] by

(43)
$$F(s_0^+, \cdots, s_N^+, s_0^-, \cdots, s_N^-) = \exp\left(-\sum_{j_1=0}^N \sum_{j_2=0}^{j_1} (s_{j_1}^+ - s_{j_1}^-)(\eta_{j_1, j_2} s_{j_2}^+ - \eta_{j_1, j_2}^* s_{j_2}^-)\right).$$

In the influence functional (43), η is a complex function and η^* is its complex conjugate. For different j_1, j_2 , η has different forms [19]. For example, when $0 < j_2 < j_1 < N$, we have

$$\eta_{j_1,j_2} = \frac{2}{\pi} \int_{-\infty}^{\infty} \mathrm{d}\omega \frac{J(\omega)}{\omega^2} \frac{\exp(\omega\beta/2)}{\sinh(\omega\beta/2)} \sin^2(\omega\Delta t/2) \exp\left(-\mathrm{i}(j_1 - j_2)\omega\Delta t\right)$$

where

$$J(\omega) = \frac{\pi}{2} \sum_{j} \frac{c_j^2}{m_j \omega_j} \delta(\omega - \omega_j)$$

and δ is the Dirac delta function.

The absolute value of η decreases to zero as $j_1 - j_2$ increases. Thus, when the difference of j_1 and j_2 is large, the contribution of η_{j_1,j_2} is small. From this point of view, the influence functional can be approximated by

$$F(s_0^+, \cdots, s_N^+, s_0^-, \cdots, s_N^-) \approx \exp\left(-\sum_{j_1=0}^N \sum_{j_2=\max\{0, k-\Delta k\}}^{j_1} (s_{j_1}^+ - s_{j_1}^-)(\eta_{j_1-j_2}s_{j_2}^+ - \eta_{j_1-j_2}^*s_{j_2}^-)\right).$$

For simplicity, the notation $S_j = (s_j^+, s_j^-)$ is introduced and the notation $I_{(S_j, S_{j'})}$ is defined by

$$I(S_{j}, S_{j'}) = \begin{cases} e^{-(s_{j}^{+} - s_{j}^{-})(\eta_{j,j'} s_{j'}^{+} - \eta_{j,j'}^{*} s_{j'}^{-})}, & j - j' \neq 1 \\ \langle s_{j}^{+} | e^{-iH_{0}\Delta t} | s_{j-1}^{+} \rangle \langle s_{j-1}^{-} | e^{iH_{0}\Delta t} | s_{j}^{-} \rangle e^{-(s_{j}^{+} - s_{j}^{-})(\eta_{j,j'} s_{j'}^{+} - \eta_{j,j'}^{*} s_{j'}^{-})}, & j - j' = 1 \end{cases}$$

With these notations,

$$\tilde{\rho}(S_{N}; N\Delta t) = \sum_{S_{N-1}} \cdots \sum_{S_{0}} \prod_{j_{1}=0}^{N} \prod_{j_{2}=0}^{j_{1}} I(S_{j_{1}}, S_{j_{2}}) \langle s_{0}^{+} | \rho_{s}(0) | s_{0}^{-} \rangle$$

$$= \sum_{S_{N-1}} \cdots \sum_{S_{0}} \left(\prod_{j=\Delta k}^{N} \prod_{m=0}^{\Delta k} I(S_{j}, S_{j-m}) \right) \underbrace{ \left(\prod_{k_{1}=0}^{\Delta k-1} \prod_{k_{2}=0}^{k_{1}} I(S_{j_{1}}, S_{j_{2}}) \langle s_{0}^{+} | \rho_{s}(0) | s_{0}^{-} \rangle \right)}_{:=A_{0}(S_{\Delta k-1}, \cdots, S_{0})}$$

$$(44) = \sum_{S_{N-1}} \cdots \sum_{S_{1}} \left(\sum_{j=\Delta k+1}^{N} \sum_{m=0}^{k} I(S_{j}, S_{j-m}) \right) \underbrace{ \sum_{s_{0}} \underbrace{ \left(\prod_{m=0}^{\Delta k} I(S_{\Delta k}, S_{\Delta k-m}) \right)}_{:=A_{1}(S_{\Delta k}, \cdots, S_{1})} A_{0}(S_{\Delta k-1}, \cdots, S_{0})}_{:=A_{1}(S_{\Delta k}, \cdots, S_{1})}$$

$$= \sum_{S_{N-1}} \cdots \sum_{S_{2}} \left(\sum_{j=\Delta k+2}^{N} \sum_{m=0}^{k} I(S_{j}, S_{j-m}) \right) \underbrace{ \sum_{s_{1}} \prod_{m=0}^{\Delta k} I(S_{\Delta k+1}, S_{\Delta k+1-m}) A_{1}(S_{\Delta k}, \cdots, S_{1})}_{:=A_{2}(S_{\Delta k+1}, \cdots, S_{2})}$$

.

An iterative method (i-QuAPI) is designed [13][14] according to (44) as follows.

$$\begin{aligned} A_0(S_{\Delta k-1}, \cdots, S_0) &:= \prod_{k_1=0}^{\Delta k-1} \prod_{k_2=0}^k I(S_{k_1}, S_{k_2}) \left\langle s_0^+ \big| \rho_s(0) \big| s_0^- \right\rangle; \\ \Lambda(S_k, \cdots, S_{k-\Delta k}) &:= \prod_{m=0}^{\Delta k} I(S_k, S_{k-m}); \\ A_{k+\Delta k+1}(S_k, \cdots, S_{k-\Delta k+1}) &:= \sum_{S_{k-\Delta k}} \Lambda(S_k, \cdots, S_{k-\Delta k}) A_{k-\Delta k}(S_{k-1}, \cdots, S_{k-\Delta k}). \end{aligned}$$

The density matrix then can be obtained component-wise by

$$\langle s_N^+ | \rho_s(N\Delta t) | s_N^- \rangle = \sum_{S_N - \Delta k+1, \cdots, S_{N-1}} A_{N-\Delta k}(S_N, \cdots, S_{N-\Delta k+1}).$$

With a density matrix of the system, the expected value of any observable O of the system can be computed by

$$\langle O(t) \rangle = \operatorname{tr}(\rho_s(t)O).$$

In spin-boson model, the density matrix $\rho_s(t)$ is a two-by-two matrix and the observable O is also a two-by-two matrix.

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