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Dynamics of the two-spin spin-boson model with a common bath

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Dynamics of the two-spin spin-boson model in the presence of Ohmic and sub-Ohmic baths is investigated by employing a multitude of the Davydov D_1 trial states, also known as the multi- D_1 *Ansatz*. Its accuracy in dynamics simulations of the two-spin SBM is improved significantly over the single D_1 *Ansatz*, especially in the weak to moderately strong coupling regime. Validity of the multi- D_1 *Ansatz* for various coupling strengths is also systematically examined by making use of the deviation vector which quantifies how faithfully the trial state obeys the Schrödinger equation. The time evolution of population difference and entanglement has been studied for various initial conditions and coupling strengths. Careful comparisons are carried out between our approach and three other methods, i.e., the time-dependent numerical renormalization group (TD-NRG) approach, the Bloch-Redfield theory, and a method based on a variational master equation. For strong coupling, the multi- D_1 trial state yields consistent results as the TD-NRG approach in the Ohmic regime while the two disagree in the sub-Ohmic regime, where the multi- D_1 trial state is shown to be more accurate. For weak coupling, the multi- D_1 trial state agrees with the two master-equation methods in the presence of both Ohmic and sub-Ohmic baths, but shows considerable differences with the TD-NRG approach in the presence of a sub-Ohmic bath, calling into question the validity of the TD-NRG results at long times in the literature. © 2016 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4945390>]

I. INTRODUCTION

As an extension of the spin-boson model (SBM), the two-spin SBM describes a pair of two-level systems coupled to a common bosonic bath. A physical example of the model is two quantum dots interacting with a common dissipative reservoir.¹ Other applications of the model can be readily found in various physical and chemical processes. For instance, it allows to study bipartite entanglement generation induced by a common bath,^{2–8} decoherence and dissipation in a two-qubit system (e.g., a quantum XOR gate operation) in the presence of decoherence,⁹ coupled Josephson-junction qubits,¹⁰ and two inductively coupled flux qubits,¹¹ which are fundamental in quantum information processing. The two-spin SBM has added interactions including direct spin-spin coupling, spin-bath interactions, and indirect spin-spin coupling induced by the common bath. Intuitively, one would expect that new physics would come out of the competition among various interactions.

The Hamiltonian for the two-spin SBM reads ($\hbar = 1$)

$$H = -\frac{\Delta_1}{2}\sigma_{x1} - \frac{\Delta_2}{2}\sigma_{x2} + V\sigma_{z1} \otimes \sigma_{z2} + \sum_l \omega_l b_l^\dagger b_l + \frac{\sigma_{z1}}{2} \sum_l \lambda_l (b_l^\dagger + b_l) + \frac{\sigma_{z2}}{2} \sum_l \tilde{\lambda}_l (b_l^\dagger + b_l), \quad (1)$$

where $\sigma_{\mu i}$ ($\mu = x, y, z, i = 1, 2$) is the μ -component Pauli matrix describing the i th spin, b_l ($b_l^\dagger$) is the annihilation (creation) operator of l th bosonic mode of frequency ω_l , Δ_i

($i = 1, 2$) is the tunneling amplitude of the i th spin. λ_l ($\tilde{\lambda}_l$) is the coupling constant between spin 1 (2) and the bath. The dissipative effect of the bath is characterized by the spectral density $J(\omega)$ as follows:¹²

$$J(\omega) = \sum_l \lambda_l^2 \delta(\omega - \omega_l) = 2\alpha\omega_c^{1-s} \omega^s \theta(\omega_c - \omega), \quad (2)$$

where α is the dimensionless coupling constant, ω_c is the cutoff frequency, $\theta(\omega)$ is the usual step function, and s is the spectral exponent. The bath is sorted into three types: sub-Ohmic ($s < 1$), Ohmic ($s = 1$), and super-Ohmic ($s > 1$). In this work, we study the well-known Ohmic and the sub-Ohmic bath.¹³

It is a nontrivial task to elucidate the dynamics of the two-spin SBM because of the infinite degrees of freedom in the bosonic bath. The reduced spin dynamics has been studied both numerically and analytically by a number of methods, including the polaron transformation approach,¹ the time-dependent numerical renormalization group (TD-NRG),^{14–16} quasi-adiabatic path integral (QUAPI),¹⁷ and hierarchical equations of motion (HEOM).¹⁸ Each has its own regimes of validity depending on the coupling strength, temperature of the bath, and the spectral density. The QUAPI is a numerically exact approach for finite temperatures and in the limit of zero temperature. The HEOM approach prefers the Drude spectral density since the correlation function of the bath can be decomposed into exponential form under such spectral density. It is claimed that the TD-NRG is a nonperturbative non-Markovian approach, capable to yield reliable spin dynamics over the entire parameter regimes.

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Orth and co-workers used the NRG method to study the ground-state phase diagram and the TD-NRG to simulate dynamics of the two-spin SBM with an Ohmic and a sub-Ohmic bath in both the weak and the strong coupling regimes.¹⁶ Surprisingly, they claimed that the second-order perturbation theory based on the Born-Markov approximation (i.e., the Bloch-Redfield theory)¹⁹ is not applicable to the two-spin SBM in the weak coupling regime where the theory works well for the single-spin SBM. Their argument is that the inter-spin Ising coupling caused by the bath is not accounted for by the Bloch-Redfield theory. In addition, it is found by those authors that the two-spin SBM is not a trivial generalization of the one-spin SBM; as the coupling strength reaches a certain threshold, for example, the two-spin SBM still finds itself stranded in the delocalized state while the one-spin SBM enters the localized phase.¹⁶

Entanglement generation as well as disentanglement in many-body systems is an issue of recent interest *per se*, and the spin-bath interactions are known to give rise to decoherence and destruction of inter-spin quantum correlations. As the two-spin SBM allows us to address the competition between the spin-spin interaction and the spin-bath coupling, we can therefore examine real time evolution of the inter-spin entanglement and spin-bath entanglement influenced by the interplay among these interactions. One may expect that distinct entanglement dynamics emerges because of the competition of various interactions, deviating from that of the two spins individually coupled to the Ohmic baths.²⁰ Moreover, the D_1 state employed in Ref. 20 can be improved in accuracy without much increased computational cost.

In this work we propose the multi- D_1 trial state as an extension of the Davydov D_1 state^{20–30} to study the real-time evolution of the two-spin SBM at zero temperature. The approach is based on the Dirac-Frenkel time-dependent variational principle, and is applicable to any spectral density function and the entire parameter ranges. More importantly, this newly developed approach is considerably more efficient and accurate than that based on the single D_1 state. In Sec. II, we formulate the multi- D_1 approach for two-spin SBM. In Sec. III, we briefly review the Bloch-Redfield master equation (BRME) and generalize the variational master equation (VME) for a two-level system developed in Ref. 31 to study the two-spin SBM. In Sec. IV, we compare the dynamics of population difference calculated from the three methods. We also examine the relative deviation which characterizes how faithfully the multi- D_1 state follows the Schrödinger equation. Moreover, we study the population difference dynamics for the intermediate and strong coupling regimes. In Sec. V, our approach is applied to address the dynamics of the quantum entanglement between the two spins quantified by the entanglement of formation. In addition, the entanglement between the two-spin system and the bath is also studied. Finally, conclusions are drawn in Sec. VI.

II. THE MULTI- D_1 ANSATZ

In general, the time evolution of the coupled spin-boson system, given by Eq. (1), can be fully captured by the

Schrödinger equation

$$i \frac{d}{dt} |\psi(t)\rangle = H |\psi(t)\rangle, \quad (3)$$

where $|\psi(t)\rangle$ is the combined state of the two spins and the bath of bosons. To solve the above Schrödinger equation, we use the time-dependent variational method with a trial state termed as the “Davydov multi- D_1 Ansatz.” The Ansatz is assumed to take the following form:

$$\begin{aligned} |D(t)\rangle = & \sum_{u=1}^N A_u(t) |\uparrow\uparrow\rangle \exp \left[\sum_l f_{ul}(t) b_l^\dagger - f_{ul}^*(t) b_l \right] |0\rangle_B \\ & + \sum_{u=1}^N B_u(t) |\uparrow\downarrow\rangle \exp \left[\sum_l g_{ul}(t) b_l^\dagger - g_{ul}^*(t) b_l \right] |0\rangle_B \\ & + \sum_{u=1}^N C_u(t) |\downarrow\uparrow\rangle \exp \left[\sum_l m_{ul}(t) b_l^\dagger - m_{ul}^*(t) b_l \right] |0\rangle_B \\ & + \sum_{u=1}^N D_u(t) |\downarrow\downarrow\rangle \exp \left[\sum_l w_{ul}(t) b_l^\dagger - w_{ul}^*(t) b_l \right] |0\rangle_B, \end{aligned} \quad (4)$$

where $|0\rangle_B$ is the vacuum state of the bosonic bath, $A_u(t)$, $B_u(t)$, $C_u(t)$, and $D_u(t)$ are variational parameters representing occupation amplitudes in the four spin states $|\uparrow\uparrow\rangle$, $|\uparrow\downarrow\rangle$, $|\downarrow\uparrow\rangle$, and $|\downarrow\downarrow\rangle$, respectively, and $f_{ul}(t)$, $g_{ul}(t)$, $m_{ul}(t)$, and $w_{ul}(t)$ label the corresponding phonon displacements of the l th mode. The parameter N refers to the number of single D_1 states we used in the Ansatz (for a specific N , the trial state is called N - D_1 Ansatz in this context). The equations of motion for these parameters, which can be derived from the Dirac-Frenkel time-dependent variational principle, are given explicitly in the Appendix.

To deal with the equations of motion for the variational parameters, we need to evaluate λ_l , $\tilde{\lambda}_l$, and ω_l for a given spectral density. In general, one needs to use a specific discretization form for the continuum spectral density given a certain type of the bath. For the Ohmic spectral density, it is proper to use a linear discretization procedure, namely, one divides the phonon frequency domain $[0, \omega_c]$ into equally spaced M intervals. For the sub-Ohmic spectral density, since the low frequency modes are of importance, it is reasonable to use a power discretization, i.e., one divides the domain $[0, \omega_c]$ into M intervals $\{[-(x+1)/M+1]^\Lambda, (-x/M+1)^\Lambda\} \omega_c$ ($x = 0, 1, \dots, M-1$). In this work, we set $\Lambda = 2.5$ for the sub-Ohmic $s = 0.5$ bath. Denoting the intervals in an unified expression by $[\nu_1, \nu_2]$ whichever discretization method we use, the parameters λ_l and ω_l can be obtained as follows:

$$\lambda_l^2 = \int_{\nu_1}^{\nu_2} J(x) dx, \quad \omega_l = \lambda_l^{-2} \int_{\nu_1}^{\nu_2} J(x) x dx. \quad (5)$$

Note that the linear discretization regards all frequency of phonons equally important while the power discretization includes more contributions from the low frequency phonons than the linear one. In this work, we use 90 (120) bosonic modes in the numerical simulation for the Ohmic (sub-Ohmic) bath and 5- D_1 Ansatz unless specified otherwise. Convergence

of our results has been carefully checked by adjusting the number of the bosonic modes for the parameter regimes considered.

To quantify how faithfully our result follows the Schrödinger equation, we use the relative deviation defined as

$$\sigma(t) = \frac{\sqrt{\langle \delta(t) | \delta(t) \rangle}}{\bar{E}_{\text{bath}}}, \quad (6)$$

where $\bar{E}_{\text{bath}}$ denotes the mean energy of bath within time interval considered, and $|\delta(t)\rangle$ is the deviation vector given by

$$|\delta(t)\rangle = (i\partial_t - H) |D(t)\rangle. \quad (7)$$

Generally, $\sigma(t)$ evolves from 0 to some finite values, and we choose the maximum value of $\sigma(t)$ during the simulation time as the referred relative error σ . Obviously, the smaller relative error is, the more accurate our trial state $|D(t)\rangle$ is.

III. MASTER EQUATION

As is well known, the BRME is applicable to the single-spin SBM in the weak coupling regime and at low temperature. However, it was claimed in Ref. 16 that the master equation is inapplicable to the two-spin SBM even in the weak coupling regime and at zero temperature since the approach fails to take into account the indirect spin-spin coupling mediated by the common bath. Here, we shall re-examine the validity of the BRME by comparing it with our multi- D_1 results and those obtained by a VME developed in Ref. 31. Below we consider the same parameter regime as studied in Ref. 16 in which the breakdown of the Bloch-Redfield theory was claimed to be found, i.e., $\Delta_1 = \Delta_2 = \Delta$ and $\lambda_l = \tilde{\lambda}_l$.

A. Bloch-Redfield master equation

We first derive the BRME for the two-spin SBM. In the weak coupling regime, we can divide the Hamiltonian into two parts

$$H = H_0 + H_1, \quad (8)$$

$$H_0 = -\frac{\Delta_1}{2}\sigma_{x1} - \frac{\Delta_2}{2}\sigma_{x2} + V\sigma_{z1}\sigma_{z2} + \sum_l \omega_l b_l^\dagger b_l, \quad (9)$$

$$H_1 = \frac{\sigma_{z1}}{2} \sum_l \lambda_l (b_l^\dagger + b_l) + \frac{\sigma_{z2}}{2} \sum_l \tilde{\lambda}_l (b_l^\dagger + b_l), \quad (10)$$

where H_0 is the free Hamiltonian and H_1 is the perturbation. In the interaction picture, the evolution for the total state of the two spins and the bath $\rho_{\text{SB}}^I(t)$ is given by

$$\frac{d}{dt} \rho_{\text{SB}}^I(t) = -i[H_I(t), \rho_{\text{SB}}^I(t)], \quad (11)$$

where $H_I(t) = \exp(iH_0 t) H_1 \exp(-iH_0 t)$. The equation of motion can be formally integrated, yielding

$$\rho_{\text{SB}}^I(t) = \rho_{\text{SB}}^I(0) - i \int_0^t [H_I(s), \rho_{\text{SB}}^I(s)] ds. \quad (12)$$

Substituting this solution into Eq. (11), we arrive at

$$\begin{aligned} \frac{d}{dt} \rho_{\text{SB}}^I(t) = & -i[H_I(t), \rho_{\text{SB}}^I(0)] \\ & - \int_0^t [H_I(t), [H_I(s), \rho_{\text{SB}}^I(s)]] ds. \end{aligned} \quad (13)$$

To proceed, we invoke the Born-Markovian approximation, i.e., by setting $\rho_{\text{SB}}^I(s) \approx \rho_{\text{S}}^I(t) \rho_{\text{B}}$ with ρ_{B} being the vacuum state of the bath in the integrand and use the factorized initial state, and take trace over the degrees of freedom of the bath, which leads to the BRME,³²

$$\frac{d}{dt} \rho_{\text{S}}^I(t) = - \int_0^t \text{Tr}_{\text{B}}[H_I(t), [H_I(s), \rho_{\text{S}}^I(t) \rho_{\text{B}}]] ds, \quad (14)$$

where $\rho_{\text{S}}^I(t) = \text{Tr}_{\text{B}}[\rho_{\text{SB}}^I(t)]$ is the reduced density matrix for the two spins. This master equation can be transformed into the Schrödinger picture and takes the form

$$\begin{aligned} \frac{d}{dt} \rho_{\text{S}}(t) = & -i[H_{\text{S}}, \rho_{\text{S}}(t)] \\ & - \int_0^t ds \text{Tr}_{\text{B}}[H_{\text{I}}, [H_{\text{I}}(s-t), \rho_{\text{S}}(t) \rho_{\text{B}}]], \end{aligned} \quad (15)$$

where $H_{\text{S}} = -\frac{\Delta_1}{2}\sigma_{x1} - \frac{\Delta_2}{2}\sigma_{x2} + V\sigma_{z1}\sigma_{z2}$. Using the explicit form of $H_{\text{I}}(\tau)$,

$$\begin{aligned} H_{\text{I}}(\tau) = & \sum_{n,m} |n\rangle\langle n| \exp(iH_0 \tau) H_1 \exp(-iH_0 \tau) |m\rangle\langle m| \\ = & \frac{1}{2} \sum_{n,m,l} \lambda_l |n\rangle\langle m| \langle n | (\sigma_{z1} + \sigma_{z2}) | m \rangle \\ & \times (b_l e^{-i\omega_l \tau} + b_l^\dagger e^{i\omega_l \tau}) \exp[i(E_n - E_m)\tau], \end{aligned} \quad (16)$$

where $H_{\text{S}}|n\rangle = E_n|n\rangle$, we can derive the BRME in the following form:

$$\begin{aligned} \frac{d}{dt} \rho_{\text{S}}(t) = & -i[H_{\text{S}}, \rho_{\text{S}}(t)] - \sum_{n,m} \{K(\omega_{mn}, t) \\ & \times [S_3, \langle S_3 \rangle_{nm} A_{nm} \rho_{\text{S}}(t)] + \text{h.c.}\}. \end{aligned} \quad (17)$$

Here the notations are defined as $S_3 = \sigma_{z1} + \sigma_{z2}$, $\langle S_3 \rangle_{nm} = \langle n | (\sigma_{z1} + \sigma_{z2}) | m \rangle$, $\omega_{mn} = E_m - E_n$, and $A_{nm} = |n\rangle\langle m|$. The time dependent function $K(\omega, t)$ is simply given by

$$K(\omega, t) = \int_0^t e^{i\omega \tau} \Lambda(\tau) d\tau, \quad (18)$$

with the bath correlation function $\Lambda(\tau) = \frac{1}{4} \int_0^{\omega_c} J(\omega) e^{-i\omega \tau} d\omega$.

In fact, in the moderately weak coupling regime, we can further simplify the master equation by introducing the Markovian approximation, i.e., using the time-independent coefficients

$$K(\omega) = \int_0^\infty \Lambda(\tau) e^{i\omega \tau} d\tau \quad (19)$$

instead of the time-dependent coefficients in Eq. (17), yielding the usual Born-Markovian master equation. In this work, we shall examine validity of the time-independent BRME as discussed in Ref. 16, where the large deviation from the TD-NRG has been revealed in the weak coupling regime.

B. Variational master equation

To explicitly include the bath induced spin-spin coupling, we perform the unitary transformation on Hamiltonian (1) arriving at $H' = e^S H e^{-S}$ with

$$S = \sum_l \frac{f_l}{2\omega_l} (b_l^\dagger - b_l)(\sigma_{z1} + \sigma_{z2}), \quad (20)$$

where f_l is a variational parameter. The transformed Hamiltonian H' can be partitioned as follows:

$$H' = H'_0 + H'_1, \quad (21)$$

$$H'_0 = -\frac{1}{2}\eta\Delta(\sigma_{x1} + \sigma_{x2}) + (V - V_c)\sigma_{z1}\sigma_{z2} + \sum_l \omega_l b_l^\dagger b_l + V_c, \quad (22)$$

$$H'_1 = -\frac{\Delta}{2}(\sigma_{x1} + \sigma_{x2})(\cosh X - \eta) - i\frac{\Delta}{2}(\sigma_{y1} + \sigma_{y2})\sinh X + \sum_l \frac{1}{2}(\lambda_l - f_l)(\sigma_{z1} + \sigma_{z2})(b_l^\dagger + b_l), \quad (23)$$

where

$$\eta = \text{Tr}_B(\rho_B \cosh X) = \exp\left[-\frac{1}{2}\sum_l \left(\frac{f_l}{\omega_l}\right)^2\right], \quad (24)$$

$$X = \sum_l \frac{f_l}{\omega_l} (b_l^\dagger - b_l), \quad (25)$$

$$V_c = \sum_l \frac{f_l}{2\omega_l} (2\lambda_l - f_l). \quad (26)$$

To proceed, we should determine the parameter f_l according to the variational principle. It is clear that H'_0 can be exactly diagonalized since the spins and the bath are decoupled. The ground state energy can be found for H'_0 as follows:

$$E_0 = -\sqrt{(\eta\Delta)^2 + (V - V_c)^2} - V_c. \quad (27)$$

We determine the parameter f_l via $\frac{\partial E_0}{\partial f_l} = 0$ and find³³

$$f_l = \frac{\lambda_l \omega_l}{\omega_l + \Omega_+ + V - V_c}, \quad (28)$$

where $\Omega_+ = \sqrt{(\eta\Delta)^2 + (V - V_c)^2}$.

In the variational framework, we find that the tunneling becomes renormalized by the factor η , and the spin-spin coupling includes contributions from both the direct coupling and the indirect coupling induced by the bath with a strength of $-V_c$. Using Eq. (28), we readily derive the renormalized factor and the indirect coupling strength in the continuum limit as follows:

$$\eta = \exp\left[-\frac{1}{2}\int_0^{\omega_c} \frac{J(\omega)}{\omega^2} F(\omega)^2 d\omega\right], \quad (29)$$

$$V_c = \int_0^{\omega_c} \frac{J(\omega)}{2\omega} F(\omega)[2 - F(\omega)] d\omega, \quad (30)$$

where

$$F(\omega) = \frac{\omega}{\omega + \Omega_+ + V - V_c}.$$

Obviously, Eqs. (29) and (30) can be solved numerically.

One may derive a new master equation from the transformed Hamiltonian in the variational framework. The general procedure is similar as the Bloch-Redfield theory. Here we use H'_0 as the free Hamiltonian and H'_1 as the perturbation. At zero temperature, using the initial condition $\rho_{SB}(0) = e^{-S}\rho_S(0)\rho_B e^S$ and Eq. (15), we can derive the VME for the two-spin SBM as follows:

$$\frac{d}{dt}\rho'_S(t) = -i[H'_S, \rho'_S(t)] - \sum_{i,j} \sum_{n,m} \{K_{ij}(\omega_{mn}, t) \times [S_i, \langle S_j \rangle_{nm} A_{nm} \rho'_S(t)] + \text{h.c.}\}. \quad (31)$$

Here prime of $\rho'_S(t)$ denotes the variational framework. $H'_S = -\frac{1}{2}\eta\Delta(\sigma_{x1} + \sigma_{x2}) + (V - V_c)\sigma_{z1}\sigma_{z2}$ is the two-spin Hamiltonian. S_i ($i, j = 1, 2, 3$) denotes $(\sigma_{x1} + \sigma_{x2})$, $(\sigma_{y1} + \sigma_{y2})$, and $(\sigma_{z1} + \sigma_{z2})$, respectively. $\langle S_j \rangle_{nm} = \langle n | S_j | m \rangle$ is the matrix element of S_j in terms of eigenstates of H'_S , i.e., $\{|n\rangle\}$. $\omega_{mn} = E'_m - E'_n$ denotes the difference between two eigenenergies of H'_S . $A_{nm} = |n\rangle\langle m|$ is an operator consisting of ket and bra of eigenstates of H'_S . The time dependent function $K_{ij}(\omega, t)$ is defined as

$$K_{ij}(\omega, t) = \int_0^t e^{i\omega\tau} \Lambda_{ij}(\tau) d\tau, \quad (32)$$

where $\Lambda_{ij}(\tau)$ are the bath correlation functions given by³¹

$$\Lambda_{11}(\tau) = \frac{(\eta\Delta)^2}{8} [e^{\phi(\tau)} + e^{-\phi(\tau)} - 2], \quad (33)$$

$$\Lambda_{22}(\tau) = \frac{(\eta\Delta)^2}{8} [e^{\phi(\tau)} - e^{-\phi(\tau)}], \quad (34)$$

$$\Lambda_{33}(\tau) = \frac{1}{4} \int_0^{\omega_c} J(\omega) [1 - F(\omega)]^2 e^{-i\omega\tau} d\omega, \quad (35)$$

$$\Lambda_{23}(\tau) = -\Lambda_{32}(\tau) = \frac{i\eta\Delta}{4} \int_0^{\omega_c} \frac{J(\omega)}{\omega} F(\omega) [1 - F(\omega)] e^{-i\omega\tau} d\omega, \quad (36)$$

$$\Lambda_{12}(\tau) = \Lambda_{21}(\tau) = \Lambda_{13}(\tau) = \Lambda_{31}(\tau) = 0, \quad (37)$$

with $\phi(\tau) = \int_0^{\omega_c} \frac{J(\omega)}{\omega^2} F(\omega)^2 e^{-i\omega\tau} d\omega$.

Similarly, this VME can also be simplified as the BRME to yield the Markovian VME by replacing the time-dependent coefficients $K_{ij}(\omega, t)$ with the time-independent coefficients¹²

$$K_{ij}(\omega) = \int_0^\infty e^{i\omega\tau} \Lambda_{ij}(\tau) d\tau. \quad (38)$$

In this work, we use the time-dependent form of the VME to simulate the dynamics for two spins.

By solving Eq. (31), we obtain the reduced density matrix $\rho'_S(t)$ in the variational framework, which can be related to density matrix in the original framework by the relation

$$\begin{aligned} \rho_S(t) &= \text{Tr}_B[e^{-S}\rho'_S(t)\rho_B e^S] \\ &= \frac{1}{8}(3 + 4\eta + \eta^4)\rho'_S(t) - \frac{1}{8}(1 - \eta^4)\rho'_S(t)\sigma_{z1}\sigma_{z2} \\ &\quad - \frac{1}{8}(1 - \eta^4)\sigma_{z1}\sigma_{z2}\rho'_S(t) \\ &\quad + \frac{1}{8}(3 - 4\eta + \eta^4)\sigma_{z1}\sigma_{z2}\rho'_S(t)\sigma_{z1}\sigma_{z2} \\ &\quad + \frac{1}{8}(1 - \eta^4)(\sigma_{z1} + \sigma_{z2})\rho'_S(t)(\sigma_{z1} + \sigma_{z2}). \end{aligned} \quad (39)$$

IV. POPULATION DIFFERENCE

We first analyze the validity of our approach by calculating for various coupling strengths the relative deviation σ , which is defined as the maximum of $\sigma(t)$. In Table I, we display for various values of N the relative deviations in the presence of an Ohmic bath. It is found that for fixed α (N) the deviation decreases with the increase of N (α), and the multi- D_1 Ansatz is more accurate in the strong coupling regime than in the weak coupling regime. Nevertheless, its performance in the weak coupling regime is also satisfactory with an acceptable accuracy. In Table II, we show the relative deviations for the sub-Ohmic bath with fixed N and various coupling strengths. From weak to strong coupling, we find that the 5- D_1 state possesses similar accuracy as for the Ohmic bath. We can therefore conclude that the multi- D_1 approach provides a reliable description for the two-spin SBM over a wide range of parameters. On the other hand, it is found that the single Davydov D_1 Ansatz, corresponding to the case of $N = 1$ in the present formalism, provides also accurate solutions in the strong coupling regime with a larger error than the multi- D_1 trial state.

We now proceed to evaluate physical observables of interest for comparison with those from the master equation approaches in order to assess the validity of our formalism, the Bloch-Redfield theory, and the VME method. The spin population differences are the main observables to be examined, given by

$$\langle \sigma_{zi}(t) \rangle = \text{Tr}[\sigma_{zi} \rho_S(t)], \quad (40)$$

representing i th spin population difference. In this subsection, the initial spin state is set to be $|\downarrow\downarrow\rangle$.

A. Strong coupling

First of all, spin dynamics in the moderately strong coupling regime and for two uncoupled spins is computed by the three formalisms, namely, the multi- D_1 trial state, the BRME, and the VME method. In Fig. 1, we compare time-dependent population differences from the three methods. In the case of an Ohmic bath and $\alpha = 0.03$, it is found that the VME result is much closer to the 5- D_1 result than the BRME. If α is increased to 0.1, it is evident that the VME method breaks down as compared to the multi- D_1 results. Increasing the coupling strength further beyond 0.1, we find that the two spins become localized, which is consistent with the prediction of the TD-NRG.¹⁶ On the other hand, in

TABLE I. The relative deviation for various coupling strengths for $s = 1$, $V = 0$, and $\Delta_1 = \Delta_2 = 0.1\omega_c$.

$N \backslash \alpha$	10^{-4}	10^{-3}	5×10^{-3}	10^{-2}	0.03	0.1	0.2	0.3
1	64.803	6.069	1.289	0.936	0.3646	0.1113	0.0894	0.0589
2	18.833	0.320	0.521	0.352	0.1303	0.0944	0.0244	0.019
3	0.117	0.320	0.0754	0.105	0.0954	0.0558	0.0227	0.0164
4	0.117	0.104	0.0340	0.0650	0.0887	0.0495	0.0212	0.0151
5	0.124	0.0194	0.0339	0.0372	0.0883	0.0443	0.0215	0.0142

TABLE II. The relative deviation for various coupling strengths for $s = 0.5$, $V = 0$, $\Delta_1 = \Delta_2 = 0.1\omega_c$, and $N = 5$.

$N \backslash \alpha$	10^{-4}	10^{-3}	5×10^{-3}	10^{-2}	0.02	0.04	0.06	0.08	0.1
5	0.0997	0.0229	0.0484	0.0648	0.1002	0.0746	0.0451	0.0315	0.0238

the presence of a sub-Ohmic bath, one finds that the VME and BRME methods yield similar results for $\alpha = 0.02$. If $\alpha = 0.04$, correct time evolution is predicted by the VME method for short times as revealed by the multi- D_1 trial state. Once again the two spins are found localized as the coupling strength is increased. In addition, one readily observes that the beat behavior appears in the evolution of the population difference of the two identical spins even in the absence of the direct spin-spin coupling for all values of the spin-bath coupling strengths. This can be understood by recalling that the spin-bath coupling also induces the indirect coupling between the two spins. In particular, the beat is also predicted by BRME for $\alpha = 0.03$. However, there is no direct coupling between spins. It follows that the reasonable explanation can only be that the Bloch-Redfield theory is able to predict the bath induced spin-spin coupling.

We now consider the dynamics in the moderately strong coupling regime when the two spins have direct coupling and compare our results with those of TD-NRG. In Fig. 2, again via the population difference, we illustrate the spin-pair dynamics with direct coupling $V = 0.05\omega_c$ for various spin-bath coupling strengths calculated by the multi- D_1 approach for both Ohmic and sub-Ohmic baths. We also present the result of VME for the specific coupling strength. The results predicted by the TD-NRG under the same conditions are extracted from Fig. 18 in Ref. 16. Let us first focus on dynamics in an Ohmic bath. In Fig. 2(a), the TD-NRG and the multi- D_1 results are in great agreement with each other. For $\alpha = 0.05$, the VME approach also yields results

FIG. 1. Time evolution of the population difference for (a) $s = 1$ and (b) $s = 0.5$ and various coupling constants α . Other parameters used are $\Delta_1 = \Delta_2 = 0.1\omega_c$, $V = 0$. The perturbative results are presented only for the moderately weak coupling strengths. VME and BRME represent the variational and Bloch-Redfield master equation, respectively.

FIG. 2. Time evolution of the population difference for (a) $s=1$ and (b) $s=0.5$ and various coupling constants α . Other parameters are $\Delta_1 = \Delta_2 = 0.1\omega_c$, $V = 0.05\omega_c$. The perturbative results are presented only for the moderately weak coupling strengths. The TD-NRG results are extracted from Ref. 16.

similar to those of the multi-D₁ approach and TD-NRG. In addition, as compared to Fig. 1(a), the beat behavior is found to vanish in the presence of direct spin-spin coupling if $\alpha = 0.05$, an outcome that is anticipated as the effective coupling between the spins combines both the direct and indirect coupling. The effective spin-spin coupling can be easily evaluated in the VME formalism. For $\alpha = 0.05$ and $\Delta_1 = \Delta_2 = 0.1\omega_c = 2V$, the effective coupling strength $V_{\text{eff}} = V - V_c = 0.0044\omega_c$, which is much smaller than the direct coupling and the induced coupling $V_c = 0.0455\omega_c$. For $\alpha > 0.1$, the VME formalism loses its validity as judged by the multi-D₁ approach. For $\alpha > 0.2$, the spins become localized.

In Fig. 2(b), to our surprise, a large deviation is found between results calculated by the TD-NRG and the multi-D₁ approach in the presence of a sub-Ohmic bath. However, the multi-D₁ results are believed to be more reliable in the sub-Ohmic regime from several considerations. Firstly, from comparison with the VME results for a relatively weak coupling strength of $\alpha = 0.03$, good agreement is found between the VME and the multi-D₁ approach. Secondly, as shown in Table III, relative deviations of the multi-D₁ states, which decrease with increasing coupling strength, are extremely small in this regime giving unambiguous indications

TABLE III. The relative deviation for $V = 0.05\omega_c$, $\Delta_1 = \Delta_2 = 0.1\omega_c$, and $s = 0.5$.

α	10^{-4}	10^{-3}	5×10^{-3}	10^{-2}	0.02	0.03	0.04	0.05	0.06
N									
5	0.1633	0.1937	0.1103	0.0698	0.0621	0.0667	0.0703	0.0695	0.0519

that our results are reliable. Last but not least, the application of the NRG to the sub-Ohmic SBM has led to inappropriate conclusions regarding quantum critical behavior,³⁴ which calls into question validity of the TD-NRG results in the sub-Ohmic regime. It is therefore concluded that despite its superior numerical efficiency, the multi-D₁ approach is valid for a wide range of coupling strengths.

B. Weak coupling

We proceed to consider the weak coupling regime where the Bloch-Redfield theory was claimed to break down in comparison with the TD-NRG.¹⁶ In Fig. 3, we compare the multi-D₁ results ($N = 5$) for $\langle \sigma_{zi}(t) \rangle$ with those from the TD-NRG, BRME, and VME in the weak-coupling regime in the presence of an Ohmic bath for $V = 0$ and $V = 0.05\omega_c$. When $V = 0$, good agreement is found between the multi-D₁ trial state, the TD-NRG approach, and the two master equations methods. In particular, the VME approach predicts nearly the same results as the multi-D₁ approach. More interestingly, it is found that the deviation in $\langle \sigma_{zi}(t) \rangle$ of the BRME method from the multi-D₁ approach is negligible, indicating that the BRME method remains valid in the weak coupling regime. When $V = 0.05\omega_c$, results from the TD-NRG and the multi-D₁ methods start to differ after $t = 200\omega_c^{-1}$, suggesting a likely TD-NRG divergence [see Fig. 3(b)].

In Fig. 4, we further compare the multi-D₁ result with those of BRME, VME, and TD-NRG in the weak coupling

FIG. 3. Comparison of the 5-D₁ (solid), VME (dashed-dotted), BRME (dashed), and TD-NRG (dotted) for $\langle \sigma_{z1,2}(t) \rangle$ in weak dissipation regime of the Ohmic bath for $\Delta_1 = \Delta_2 = 0.1\omega_c$ and $\alpha = 0.01$. The initial state is chosen as $|\downarrow\downarrow\rangle$. (a) $V = 0$ and (b) $V = 0.05\omega_c$.

FIG. 4. Comparison of the 5-D₁ (solid), VME (dashed-dotted), BRME (dashed), and TD-NRG (dotted) for $\langle \sigma_{z1,2}(t) \rangle$ in weak dissipation regime of the sub-Ohmic- $s=0.5$ bath for $\Delta_1 = \Delta_2 = 0.1\omega_c$ and $\alpha = 0.01$. The initial state is chosen as $|\downarrow\downarrow\rangle$. (a) $V=0$ and (b) $V=0.05\omega_c$.

regime for $V=0$ and $V=0.05\omega_c$ in the presence of a sub-Ohmic bath. Once again one finds that the BRME result has an acceptable deviation from those of the multi-D₁ and VME methods, while the VME predicts nearly the same result as the multi-D₁ approach. However, the TD-NRG results are quantitatively different from those of the multi-D₁ approach and the master equation methods. We can therefore conclude that the multi-D₁ approach and the perturbative approaches give the reliable results in the weak coupling regime in the presence of both Ohmic and sub-Ohmic baths. However, the TD-NRG approach yields quantitatively different dynamics as compared to the other three methods at long times with an Ohmic bath and at short times with a sub-Ohmic bath, pointing to the unreliability of the TD-NRG results, particularly in the sub-Ohmic regime.

The non-negligible deviation between the TD-NRG and the BRME was first pointed out by Orth and co-workers in Ref. 16 in the same parameter regimes as given in Figs. 3 and 4. In addition, it was found that the deviation, attributed by the authors to the breakdown of the Bloch-Redfield theory, is larger with the sub-Ohmic bath than with the Ohmic bath. Specifically, they claimed that the Bloch-Redfield theory becomes invalid in the weak coupling regime ($\alpha \ln(\omega_c/\Delta) \ll 1$), where the theory is expected to be workable. Their argument is that the Bloch-Redfield theory cannot correctly take into account the bath-induced spin-spin coupling. Moreover, they had concluded that the synchronization of two spins cannot be described by the Bloch-Redfield theory. However, our findings in this work seem to call into doubts the validity of their conclusions. To our surprise, it is found in this work that the Bloch-Redfield theory properly takes into account the bath-induced coupling, up to second order in λ_I , and remains valid in the weak coupling regime as in the single-spin SBM.

Apart from benchmarking the BRME against the multi-D₁ approach, we can also benchmark the multi-D₁ approach against the BRME in the weak coupling regime if we can

FIG. 5. Two-spin synchronization dynamics calculated by the BRME for (a) $s=1$ and (b) $s=0.5$. Other parameters are $\Delta_2 = 0.5\Delta_1 = 0.001\omega_c$, $\alpha = 8 \times 10^{-4}$, and $V=0$.

prove that the BRME method is able to account for the indirect spin-spin coupling mediated by the bath. To directly prove that the Bloch-Redfield theory takes into account the indirect spin-spin coupling, we carry out the numerical simulation of the dynamics of the two spins by using the BRME method with the same parameters given in Fig. 11 of Ref. 16. In Fig. 5, we demonstrate that the BRME method can indeed predict spin synchronization in the absence of direct coupling ($V=0$) for $\alpha = 8 \times 10^{-4}$ and $\Delta_2 = 0.5\Delta_1 = 0.001\omega_c$ in both the Ohmic and sub-Ohmic regimes. Obviously, the parameters chosen satisfy the relation $\alpha \ln(\omega_c/\Delta_2) = 0.005526 \ll 1$ and the BRME method gives the similar results as the TD-NRG in Fig. 11 of Ref. 16, which proves that the Bloch-Redfield theory accounts for the bath-induced spin-spin coupling. Combined with the results in Figs. 3 and 4, we have to conclude that the Bloch-Redfield theory remains valid for the two-spin SBM as for the single-spin SBM in the weak coupling regime, and the indirect spin-spin coupling mediated by the common bath can be properly taken into account in the Bloch-Redfield formalism. Consequently, it is inappropriate to fault the BRME method for the divergence between TD-NRG and BRME. Our findings here also lend strong support to the reliability of the multi-D₁ approach, which is able to yield reliable dynamics of the two-spin SBM in the weak coupling regime.

V. ENTANGLEMENT

Quantum entanglement is an important resource that does not have a classical counterpart. It is believed to be fragile to environmental disturbance as interactions between a quantum system and its environment often lead to quick disentanglement in the system. Our current model allows us to address the influence of the direct spin-spin coupling, the indirect coupling mediated by the bath, and the spin-bath coupling on the disentanglement dynamics. The direct spin-spin coupling is expected to entangle two spins, whereas the spin-bath coupling generally induces dissipation and

destroys the spin coherence, leading to disentanglement. In the simultaneous presence of these two different types of coupling, one may expect a competition between the generation and the destruction of the entanglement. More interestingly, without the direct spin-spin coupling, the indirect coupling mediated by the common bath can also be responsible for the generation of the entanglement.¹ Therefore, we have another way to verify whether the BRME takes into account the bath-mediated spin-spin coupling by examining whether the entanglement generation from a non-entangled initial state can be predicted by the BRME without the direct spin-spin coupling. In what follows, we move to apply the formalisms to study the entanglement dynamics at the zero temperature and in the presence of an Ohmic bath. The degree of entanglement of the bipartite quantum system can be characterized by the entanglement of formation defined as follows:³⁵

$$Q = h \left(\frac{1}{2} + \frac{1}{2} \sqrt{1 - C^2} \right), \quad (41)$$

where

$$C = \max\{0, \sqrt{e_1} - \sqrt{e_2} - \sqrt{e_3} - \sqrt{e_4}\}. \quad (42)$$

Here, e_j ($j = 1$ to 4) are the eigenvalues of the matrix $\rho_S(t)(\sigma_{y1} \otimes \sigma_{y2})\rho_S^*(t)(\sigma_{y1} \otimes \sigma_{y2})$ arranged in a descending order. On the other hand, due to the interactions between spins and the bath, the two may be entangled. We use the von Neumann entropy to quantify the spin-bath entanglement,³⁶

$$\epsilon = - \sum_{k=1}^d \lambda_k \log_2 \lambda_k, \quad (43)$$

where λ_k are the eigenvalues of $\rho_S(t)$.

To study effects of the indirect coupling, we set the initial state the product state $|\downarrow\downarrow\rangle$ without any quantum correlation. Figure 6(a) shows the dynamics of entanglement for the two spins in an Ohmic bath in the absence of the direct spin-spin coupling ($V = 0$). The three approaches are in agreement on that the entanglement is found to increase

from zero and oscillate as the qubits become entangled. In fact, the small amplitude oscillation is similar to the beat in the population dynamics, which results from the indirect spin-spin coupling. In principle, one can verify that when choosing a proper direct coupling strength V such that $V_{\text{eff}} = V - V_c = 0$, one could expect that the generation of entanglement for two spins ceases to occur. As a rough verification, we set $V = 0.0091\omega_c$, leading to $V_{\text{eff}} = V - V_c = -3 \times 10^{-6}\omega_c$, where $V_c = 0.009103\omega_c$ evaluated from the VME method. In Fig. 6(b), we show the entanglement dynamics from the master equation formalisms for $V = 0.0091\omega_c$ with the other parameters same as in Fig. 6(a). Indeed, the two formalisms appear in qualitative agreement with each other with the prediction that the generation of the entanglement for two spins is significantly suppressed. This confirms that the indirect coupling induced by the common bath is responsible for the generation of the entanglement when the direct coupling is absent. We also present the entanglement entropy that quantifies the spin-bath entanglement. The result shows that the entropy increases, indicating the spin and bath become entangled. This is a natural phenomenon because the spin-bath coupling plays the role of mediating the indirect spin-spin coupling.

We now move to study how the entanglement of the two spins evolves if the two spins are initially in Bell states using the multi- D_1 approach. The Bell states are the fully entangled states,

$$|\Phi_{\pm}\rangle = (|\uparrow\uparrow\rangle \pm |\downarrow\downarrow\rangle)/\sqrt{2}, \quad (44)$$

$$|\Psi_{\pm}\rangle = (|\uparrow\downarrow\rangle \pm |\downarrow\uparrow\rangle)/\sqrt{2}, \quad (45)$$

with the maximum bipartite entanglement. Since the Bell state $|\Psi_{-}\rangle$ does not evolve when $V = 0$, we display in Fig. 7 the entanglement dynamics when the system is in the other three Bell states. It is not surprising to find that the entanglement decays more slowly in the weak coupling regime than in the strong coupling regime. When $\alpha = 0.05$ and 0.1, one finds that the entanglement vanishes in a short period of time, a phenomenon known as entanglement sudden death (ESD).²⁻⁵

FIG. 6. Time evolution of entanglement of formation (Q) and the entanglement entropy calculated by the 6- D_1 trial state (solid), the VME (dashed-dotted), and the BRME (dotted) for (a) $V = 0$ and (b) $V = 0.0091\omega_c$. Other parameters used are $s = 1$, $\Delta_1 = \Delta_2 = 0.1\omega_c$, and $\alpha = 0.01$.

FIG. 7. Entanglement dynamics calculated by the 6- D_1 trial state with initial Bell states: (a) $|\Phi_{+}\rangle$, (b) $|\Phi_{-}\rangle$, and (c) $|\Psi_{+}\rangle$. Other parameters used are $V = 0$, $\Delta_1 = \Delta_2 = 0.1\omega_c$, and $s = 1$.

FIG. 8. Entanglement dynamics calculated by the 6-D₁ trial state for (a) $\theta = \pi/12$ and (b) $\theta = \pi/6$, and various values of the coupling strength α . Other parameters are $\Delta_1 = \Delta_2 = 0.1\omega_c$, $V = 0$, and $s = 1$.

When the ESD occurs depends on the initial states. It takes longer for the two spins evolving from $|\Psi_+\rangle$ to undergo the ESD than from the other two Bell states. After the ESD, the two spins can quickly become entangled again due to the bath-mediated spin-spin coupling before the system reaches an equilibrium state.

To study the entanglement stability we present calculations starting from $\cos \theta |\uparrow\uparrow\rangle + \sin \theta |\downarrow\downarrow\rangle$ for various values of coupling strengths in Fig. 8. The results show that in both the weak and strong spin-bath coupling regimes, the dynamical behavior of the entanglement strongly depends on the initial preparation. Interestingly, if $\theta = \pi/12$ and $\alpha = 0.01$, the entanglement is able to survive for a relatively long time, without obvious decay. However, if $\theta = \pi/6$ and $\alpha = 0.01$, the entanglement slowly decays. In the strong coupling regime, the entanglement is found to vanish in a short time followed by a rapid revival. These findings show that the bath-induced indirect coupling plays an important role in entanglement generation.

VI. CONCLUSION

An extension to the single Davydov D₁ Ansatz has been proposed to treat successfully the dynamics of a spin pair coupled to a common reservoir of harmonic oscillators. In principle, this method can be generalized straightforwardly to deal with spin dynamics in the presence of a super-Ohmic bath and off-diagonal spin-bath coupling. The accuracy of our variational method is carefully checked by examining the relative deviation, which is defined as the norm maximum of the deviation vector during the time evolution of the multi-D₁ state. In particular, when the coupling increases, the relative deviation decreases significantly, which indicates that our method performs better in the strong coupling regime. Furthermore, it is found that even in the weak coupling regime our approach remains valid and agrees well with two master equation methods based on perturbation theory,

i.e., the Bloch-Redfield theory and the VME used with a unitary transformation. The multi-D₁ results are compared with those of the TD-NRG approach. The difference between the two methods is found to be negligible in the presence of the Ohmic bath, but become apparent for the sub-Ohmic regime, where the multi-D₁ method is known to be more accurate and is found to be in good agreement with the Bloch-Redfield theory and the VME approach. Only for very short times, the TD-NRG method is consistent with the multi-D₁ approach and the two master equations methods in the presence of a sub-Ohmic bath.

Our method is also used to study the time evolution of the population difference and the spin-spin and spin-bath entanglement dynamics. For population dynamics of the two-spin SBM, it is found that the Bloch-Redfield theory is valid in the weak coupling regime as shown by comparisons with the multi-D₁ trial state and the VME approach. The Bloch-Redfield theory is shown to yield bath-induced spin-spin coupling, indicating that its difference with TD-NRG cannot be attributed to a breakdown of the Bloch-Redfield theory, instead, may be caused by inaccuracy of the TD-NRG method at long times. For entanglement dynamics, it is found that the bath can destroy as well as generate spin-spin entanglement, and the entanglement dynamics also strongly depends on the initial preparation of the two spins.

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APPENDIX: THE EQUATION OF MOTION FOR THE VARIATIONAL PARAMETERS

In this work, the Lagrangian formalism of the Dirac-Frenkel time-dependent variational principle is employed to obtain the equations of motion for the variational parameters. The Lagrangian associated with the trial state $|\mathcal{D}(t)\rangle$ is given by

$$\begin{aligned} L &= \langle \mathcal{D}(t) | \frac{i}{2} \frac{\overleftrightarrow{\partial}}{\partial t} - H | \mathcal{D}(t) \rangle \\ &= \frac{i}{2} \left[\langle \mathcal{D}(t) | \frac{\overrightarrow{\partial}}{\partial t} | \mathcal{D}(t) \rangle - \langle \mathcal{D}(t) | \frac{\overleftarrow{\partial}}{\partial t} | \mathcal{D}(t) \rangle \right] \\ &\quad - \langle \mathcal{D}(t) | H | \mathcal{D}(t) \rangle. \end{aligned} \quad (\text{A1})$$

The Dirac-Frenkel time-dependent variational principle yields the equations of motion for variational parameter x ,

$$\frac{d}{dt} \left(\frac{\partial L}{\partial \dot{x}^*} \right) - \frac{\partial L}{\partial x^*} = 0, \quad (\text{A2})$$

where x^* denotes the complex conjugate of x , which can be $A_u(t)$, $B_u(t)$, $C_u(t)$, $D_u(t)$, $f_{ul}(t)$, $g_{ul}(t)$, $m_{ul}(t)$, or $w_{ul}(t)$. It is straightforward to derive the equations of motion for $A_u(t)$, $B_u(t)$, $C_u(t)$, $D_u(t)$ from Eq. (A2),

$$0 = \sum_{u=1}^N \left[i\dot{A}_u + \frac{i}{2} A_u \sum_k (2f_{nk}^* \dot{f}_{uk} - f_{uk}^* \dot{f}_{nk} - f_{uk} \dot{f}_{nk}^*) \right] R(f_n, f_u) - \sum_{u=1}^N \frac{A_u}{2} \sum_k (\lambda_k + \tilde{\lambda}_k) (f_{nk}^* + f_{uk}) R(f_n, f_u) \\ - \sum_{u=1}^N \sum_k \omega_k A_u f_{nk}^* f_{uk} R(f_n, f_u) + \sum_{u=1}^N \frac{\Delta_1}{2} C_u R(f_n, m_u) + \sum_{u=1}^N \frac{\Delta_2}{2} B_u R(f_n, g_u) - \sum_{u=1}^N V A_u R(f_n, f_u), \quad (\text{A3})$$

$$0 = \sum_{u=1}^N \left[i\dot{B}_u + \frac{i}{2} B_u \sum_k (2g_{nk}^* \dot{g}_{uk} - g_{uk}^* \dot{g}_{nk} - g_{uk} \dot{g}_{nk}^*) \right] R(g_n, g_u) - \sum_{u=1}^N \frac{B_u}{2} \sum_k (\lambda_k - \tilde{\lambda}_k) (g_{nk}^* + g_{uk}) R(g_n, g_u) \\ - \sum_{u=1}^N \sum_k \omega_k B_u g_{nk}^* g_{uk} R(g_n, g_u) + \sum_{u=1}^N \frac{\Delta_1}{2} D_u R(f_n, w_u) + \sum_{u=1}^N \frac{\Delta_2}{2} A_u R(g_n, f_u) + \sum_{u=1}^N V B_u R(g_n, g_u), \quad (\text{A4})$$

$$0 = \sum_u^N \left[i\dot{C}_u + \frac{i}{2} C_u \sum_k (2m_{nk}^* \dot{m}_{uk} - m_{uk}^* \dot{m}_{nk} - m_{uk} \dot{m}_{nk}^*) \right] R(m_n, m_u) + \sum_{u=1}^N \frac{C_u}{2} \sum_k (\lambda_k - \tilde{\lambda}_k) (m_{nk}^* + m_{uk}) R(m_n, m_u) \\ - \sum_{u=1}^N \sum_k \omega_k C_u m_{nk}^* m_{uk} R(m_n, m_u) + \sum_{u=1}^N \frac{\Delta_1}{2} A_u R(m_n, f_u) + \sum_{u=1}^N \frac{\Delta_2}{2} D_u R(m_n, w_u) + \sum_{u=1}^N V C_u R(m_n, m_u), \quad (\text{A5})$$

$$0 = \sum_{u=1}^N \left[i\dot{D}_u + \frac{i}{2} D_u \sum_k (2w_{nk}^* \dot{w}_{uk} - w_{uk}^* \dot{w}_{nk} - w_{uk} \dot{w}_{nk}^*) \right] R(w_n, w_u) + \sum_{u=1}^N \frac{D_u}{2} \sum_k (\lambda_k + \tilde{\lambda}_k) (w_{nk}^* + w_{uk}) R(w_n, w_u) \\ - \sum_{u=1}^N \sum_k \omega_k D_u w_{nk}^* w_{uk} R(w_n, w_u) + \sum_{u=1}^N \frac{\Delta_1}{2} B_u R(w_n, g_u) + \sum_{u=1}^N \frac{\Delta_2}{2} C_u R(w_n, m_u) - \sum_{u=1}^N V D_u R(w_n, w_u), \quad (\text{A6})$$

where

$$R(f_n, g_u) = \exp \left[\sum_k \left(f_{nk}^* g_{uk} - \frac{1}{2} |f_{nk}|^2 - \frac{1}{2} |g_{uk}|^2 \right) \right]. \quad (\text{A7})$$

Similarly, equations of motion for $f_{ul}(t)$, $g_{ul}(t)$,

$$\frac{i}{2} \sum_{u=1}^N \left[2A_n^* A_u \dot{f}_{ul} + 2A_n^* \dot{A}_u f_{ul} + A_n^* A_u f_{ul} \sum_k (2f_{nk}^* \dot{f}_{uk} - f_{uk}^* \dot{f}_{nk} - f_{uk} \dot{f}_{nk}^*) \right] R(f_n, f_u) \\ - \sum_{u=1}^N A_n^* A_u \left[\frac{\lambda_l + \tilde{\lambda}_l}{2} + \sum_k \frac{\lambda_k + \tilde{\lambda}_k}{2} f_{ul} (f_{nk}^* + f_{uk}) \right] R(f_n, f_u) - \sum_{u=1}^N A_n^* A_u f_{ul} \left[\omega_l + \sum_k \omega_k f_{nk}^* f_{uk} \right] R(f_n, f_u) \\ = - \sum_{u=1}^N \frac{\Delta_1}{2} C_u A_n^* m_{ul} R(f_n, m_u) - \sum_{u=1}^N \frac{\Delta_2}{2} B_u A_n^* g_{ul} R(f_n, g_u) + \sum_{u=1}^N V A_n^* A_u f_{ul} R(f_n, f_u), \quad (\text{A8})$$

$$\frac{i}{2} \sum_{u=1}^N \left[2B_n^* B_u \dot{g}_{ul} + 2B_n^* \dot{B}_u g_{ul} + B_n^* B_u g_{ul} \sum_k (2g_{nk}^* \dot{g}_{uk} - g_{uk}^* \dot{g}_{nk} - g_{uk} \dot{g}_{nk}^*) \right] R(g_n, g_u) \\ - \sum_{u=1}^N B_n^* B_u \left[\frac{\lambda_l - \tilde{\lambda}_l}{2} + \sum_k \frac{\lambda_k - \tilde{\lambda}_k}{2} g_{ul} (g_{nk}^* + g_{uk}) \right] R(g_n, g_u) - \sum_{u=1}^N B_n^* B_u g_{ul} \left[\omega_l + \sum_k \omega_k g_{nk}^* g_{uk} \right] R(g_n, g_u) \\ = - \sum_{u=1}^N \frac{\Delta_1}{2} D_u B_n^* w_{ul} R(g_n, w_u) - \sum_{u=1}^N \frac{\Delta_2}{2} A_u B_n^* f_{ul} R(g_n, f_u) - \sum_{u=1}^N V B_n^* B_u g_{ul} R(g_n, g_u), \quad (\text{A9})$$

$$\frac{i}{2} \sum_{u=1}^N \left[2C_n^* C_u \dot{m}_{ul} + 2C_n^* \dot{C}_u m_{ul} + C_n^* C_u m_{ul} \sum_k (2m_{nk}^* \dot{m}_{uk} - m_{uk}^* \dot{m}_{nk} - m_{uk} \dot{m}_{nk}^*) \right] R(m_n, m_u) \\ + \sum_{u=1}^N C_n^* C_u \left[\frac{\lambda_l - \tilde{\lambda}_l}{2} + \sum_k \frac{\lambda_k - \tilde{\lambda}_k}{2} m_{ul} (m_{nk}^* + m_{uk}) \right] R(m_n, m_u) - \sum_{u=1}^N C_n^* C_u m_{ul} \left[\omega_l + \sum_k \omega_k m_{nk}^* m_{uk} \right] R(m_n, m_u) \\ = - \sum_{u=1}^N \frac{\Delta_1}{2} A_u C_n^* f_{ul} R(m_n, f_u) - \sum_{u=1}^N \frac{\Delta_2}{2} D_u C_n^* w_{ul} R(m_n, w_u) - \sum_{u=1}^N V C_n^* C_u m_{ul} R(m_n, m_u), \quad (\text{A10})$$

$$\begin{aligned}
& \frac{i}{2} \sum_{u=1}^N \left[2D_n^* D_u w_{ul} + 2D_n^* \dot{D}_u w_{ul} + D_n^* D_u w_{ul} \sum_k (2w_{nk}^* w_{uk} - w_{uk}^* w_{uk} - w_{uk} w_{uk}^*) \right] R(w_n, w_u) \\
& + \sum_{u=1}^N D_n^* D_u \left[\frac{\lambda_l + \tilde{\lambda}_l}{2} + \sum_k \frac{\lambda_k + \tilde{\lambda}_k}{2} w_{ul} (w_{nk}^* + w_{uk}) \right] R(w_n, w_u) - \sum_{u=1}^N D_n^* D_u w_{ul} \left[\omega_l + \sum_k \omega_k w_{nk}^* w_{uk} \right] R(w_n, w_u) \\
& = - \sum_{u=1}^N \frac{\Delta_1}{2} B_u D_n^* g_{ul} R(w_n, g_u) - \sum_{u=1}^N \frac{\Delta_2}{2} C_u D_n^* m_{ul} R(w_n, m_u) + \sum_{u=1}^N V D_n^* D_u w_{ul} R(w_n, w_u). \quad (\text{A11})
\end{aligned}$$

After solving the equations of motion, we can get the reduced density matrix of the two-spin system

$$\begin{aligned}
\rho_S(t) &= \text{Tr}_B [|D(t)\rangle\langle D(t)|] \\
&= \begin{pmatrix} \sum_{n,u} A_u A_n^* R(f_n, f_u) & \sum_{n,u} A_u B_n^* R(g_n, f_u) & \sum_{n,u} A_u C_n^* R(m_n, f_u) & \sum_{n,u} A_u D_n^* R(w_n, f_u) \\ \sum_{n,u} A_u^* B_n R(g_n, f_u)^* & \sum_{n,u} B_u B_n^* R(g_n, g_u) & \sum_{n,u} B_u C_n^* R(m_n, g_u) & \sum_{n,u} B_u D_n^* R(w_n, g_u) \\ \sum_{n,u} A_u^* C_n R(m_n, f_u)^* & \sum_{n,u} B_u^* C_n R(m_n, g_u)^* & \sum_{n,u} C_u C_n^* R(m_n, m_u) & \sum_{n,u} C_u D_n^* R(w_n, m_u) \\ \sum_{n,u} A_u^* D_n R(w_n, f_u)^* & \sum_{n,u} B_u^* D_n R(w_n, g_u)^* & \sum_{n,u} C_u^* D_n R(w_n, m_u)^* & \sum_{n,u} D_u D_n^* R(w_n, w_u) \end{pmatrix}. \quad (\text{A12})
\end{aligned}$$

- ¹L. D. Contreras-Pulido and R. Aguado, *Phys. Rev. B* **77**, 155420 (2008).
- ²D. Braun, *Phys. Rev. Lett.* **89**, 277901 (2002).
- ³Y. Zhao and G. Chen, *Physica A* **317**, 13 (2003); Y. Zhao, F. L. Zheng, J. Liu, and Y. Yao, *ibid.* **423**, 80 (2015).
- ⁴T. Yu and J. H. Eberly, *Phys. Rev. Lett.* **93**, 140404 (2004).
- ⁵T. Yu and J. H. Eberly, *Phys. Rev. Lett.* **97**, 140403 (2006).
- ⁶F. Mintert, A. R. R. Carvalho, M. Kuś, and A. Buchleitner, *Phys. Rep.* **415**, 207 (2005).
- ⁷M. Ikram, F.-l. Li, and M. S. Zubairy, *Phys. Rev. A* **75**, 062336 (2007).
- ⁸K.-L. Liu and H.-S. Goan, *Phys. Rev. A* **76**, 022312 (2007).
- ⁹M. Thorwart and P. Hänggi, *Phys. Rev. A* **65**, 012309 (2001).
- ¹⁰M. Governale, M. Grifoni, and G. Schön, *Chem. Phys.* **268**, 273 (2001).
- ¹¹F. Yoshihara, Y. Nakamura, and J. S. Tsai, *Phys. Rev. B* **81**, 132502 (2010).
- ¹²U. Weiss, *Quantum Dissipative Systems* (World Scientific, 1999).
- ¹³A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. P. A. Fisher, A. Garg, and W. Zwerger, *Rev. Mod. Phys.* **59**, 1 (1987).
- ¹⁴F. B. Anders and A. Schiller, *Phys. Rev. Lett.* **95**, 196801 (2005).
- ¹⁵F. B. Anders and A. Schiller, *Phys. Rev. B* **74**, 245113 (2006).
- ¹⁶P. P. Orth, D. Roosen, W. Hofstetter, and K. Le Hur, *Phys. Rev. B* **82**, 144423 (2010).
- ¹⁷M. M. Sahrpour and N. Makri, *J. Chem. Phys.* **138**, 114109 (2013).
- ¹⁸J. Ma, Z. Sun, X. Wang, and F. Nori, *Phys. Rev. A* **85**, 062323 (2012).
- ¹⁹M. J. Storcz and F. K. Wilhelm, *Phys. Rev. A* **67**, 042319 (2003).
- ²⁰L. W. Duan, H. Wang, Q. H. Chen, and Y. Zhao, *J. Chem. Phys.* **139**, 044115 (2013).
- ²¹J. Sun, B. Luo, and Y. Zhao, *Phys. Rev. B* **82**, 014305 (2010).
- ²²B. Luo, J. Ye, C. B. Guan, and Y. Zhao, *Phys. Chem. Chem. Phys.* **12**, 15073 (2010).
- ²³J. Ye, K. W. Sun, Y. Zhao, Y. J. Yu, C. K. Lee, and J. S. Cao, *J. Chem. Phys.* **136**, 245104 (2012).
- ²⁴Z. G. Lü, L. W. Duan, X. Li, P. Shenai, and Y. Zhao, *J. Chem. Phys.* **139**, 164103 (2013).
- ²⁵N. Wu, L. Duan, X. Li, and Y. Zhao, *J. Chem. Phys.* **138**, 084111 (2013).
- ²⁶S. J. Xiong, L. P. Chen, and Y. Zhao, *ChemPhysChem* **15**(13), 2859 (2014).
- ²⁷K. W. Sun, J. Ye, and Y. Zhao, *J. Chem. Phys.* **141**, 124103 (2014).
- ²⁸N. Zhou, L. Chen, Y. Zhao, D. Mozyrsky, V. Chernyak, and Y. Zhao, *Phys. Rev. B* **90**, 155135 (2014).
- ²⁹N. Zhou, L. Chen, D. Xu, V. Chernyak, and Y. Zhao, *Phys. Rev. B* **91**, 195129 (2015).
- ³⁰L. Wang, L. Chen, N. Zhou, and Y. Zhao, *J. Chem. Phys.* **144**, 024101 (2016).
- ³¹D. P. S. McCutcheon, N. S. Dattani, E. M. Gauger, B. W. Lovett, and A. Nazir, *Phys. Rev. B* **84**, 081305(R) (2011).
- ³²H. P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University Press, New York, 2006).
- ³³H. Zheng, Z. Lü, and Y. Zhao, *Phys. Rev. E* **91**, 062115 (2015).
- ³⁴M. Vojta, *Phys. Rev. B* **85**, 115113 (2012).
- ³⁵W. K. Wootters, *Phys. Rev. Lett.* **80**, 2245 (1998).
- ³⁶C. H. Bennett, D. P. DiVincenzo, J. A. Smolin, and W. K. Wootters, *Phys. Rev. A* **54**, 3824 (1996).