Stochastic Representation of Non-Markovian Fermionic Quantum Dissipation

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Quantum Brownian motion plays a fundamental role in many areas of modern physics. In the path-integral formulation, the environmental quantum fluctuations driving the system dynamics can be characterized by auxiliary stochastic fields. For fermion bath environment the stochastic fields are Grassmann-valued, and cannot be represented by conventional classical numbers. In this Letter, we propose a strategy to map the nonclassical Grassmann fields onto Gaussian white noises along with a set of quantized pseudo-states. This results in a numerically feasible stochastic equation of motion (SEOM) method for fermionic open systems. The SEOM yields exact physical observables for noninteracting systems, and accurate approximate results for interacting systems. The practicality and accuracy of the proposed SEOM are exemplified by direct stochastic simulations conducted on a single-impurity Anderson model.

Over a century ago, Einstein has explained the nature of Brownian motion by establishing a quantitative relation between the dissipative forces driving a classical particle and the environmental thermal fluctuations. Nowadays, quantum Brownian motion, i.e., the dissipative dynamics of a quantum system driven by quantum fluctuations in surrounding environments, plays a fundamental role in many subdisciplines of modern physics. The sources of quantum fluctuations are rather general, e.g., the excitations of various types of particles or quasiparticles (photons, phonons, electrons, excitons, spins, etc.) This makes quantum Brownian motion closely pertinent to a wide range of applications, including nanoelectronics, nanomotors, solar energy conversion, superconductors, quantum information, and quantum computation.

The main challenge in describing quantum Brownian motion or more general quantum dissipative dynamics is to elucidate the combined effects of system-environment coupling, many-body correlation, and non-Markovian memory. This requires a complete characterization of the influence of environment which hosts the quantum fluctuations and usually has infinite degrees of freedom. Remarkable progress has been made, much thanks to the path-integral formulation developed by Feynman and Vernon. Particularly, it has been proposed that the influence of environment can be captured by introducing a set of auxiliary stochastic fields.

If the environment is a boson bath, the auxiliary stochastic fields representing the environmental quantum fluctuations are classical, and can be realized via c-number noises. Such formal simplicity has greatly facilitated the development of stochastic theories. For instance, quantum state diffusion (QSD) theory and stochastic equation of motion (SEOM) theory have been established and applied to investigate quantum dissipative dynamics in realistic systems, such as the transfer of excitons in molecular aggregates and photo-induced electron transfer at interfaces of organic solar cells.

While a boson mode corresponds to a classical harmonic oscillator, there is no such classical counterpart for a fermion mode. Therefore, the situation is highly nontrivial for fermion bath environment, for which the auxiliary stochastic fields have to be Grassmann-valued, i.e., the fields mutually anticommute, to preserve the even parity of all physical observables. Unlike c-numbers, Grassmann numbers are nonclassical and cannot be represented by conventional means. Such difficulty has severely hindered the practical implementation of stochastic theories. From the early attempts on describing fermion Brownian motion to the recent extension of QSD and SEOM theories to fermionic open systems, all previous efforts were limited to formal derivations; whereas to the best of our knowledge, no stochastic simulation has been conducted on quantum dissipative dynamics of fermions.

To break the status quo, and to enable direct numerical simulation of fermion Brownian motion, we propose in this Letter a mapping strategy, as schematically illustrated in Fig. 1 and the elaborations below, with which the stochastic Grassmann fields are effectively mapped onto conventional c-number fields along with a set of quantized pseudo-states. This finally leads to the construction of a numerically feasible and accurate SEOM for fermionic open systems.

Without loss of generality, we illustrate our strategy with a single-level system coupled linearly to a fermion bath. The system-bath interaction is $H_{SB} = c^\dagger F + F^\dagger c$, where...
with \( \hat{F} = \sum_k \tau_k \hat{d}_k \). Here, \( \{ \tau_k \} \) are coupling strengths; \( \hat{c} (\hat{\bar{c}}) \) and \( \hat{d}_k (\hat{\bar{d}}_k) \) are the annihilation (creation) operators for system and bath levels, respectively. Set \( \hbar = 1 \) hereafter. Following the standard stochastic decoupling method [23], we factorize \( e^{-iH_B dt} \) by introducing time-dependent auxiliary Grassmann fields.

To fulfill the fermion statistics, we introduce four involutive Grassmann–Wiener processes \( \{ \hat{\psi}_{jt} = \eta_{jt}; j = 1, \ldots, 4 \} \), which satisfy \( \langle \eta_{jt} \rangle = \langle \bar{\eta}_{jt} \rangle = 0 \) and \( \langle \eta_{jt} \bar{\eta}_{j'} t \rangle = \delta_{jj'} \delta(t - \tau) \). The resultant decoupled system and bath density matrix dynamics under the time-dependent auxiliary Grassmann fields (AGFs) read [39]

\[
\dot{\rho}_s = -i[H_s, \rho_s] + e^{-i\pi/4} (\hat{c}^\dagger \eta_{1t} + \bar{\eta}_{2t} \hat{c}) \rho_s + e^{i\pi/4} \rho_s (\hat{c}^\dagger \bar{\eta}_{3t} + \eta_{4t} \bar{c}), \tag{1}
\]

\[
\dot{\rho}_B = -i[H_B, \rho_B] + e^{-i\pi/4} (\hat{\bar{c}}^\dagger \eta_t \hat{F} + \hat{F}^\dagger \eta_{t}) \rho_B + e^{i\pi/4} \rho_B (\hat{\bar{c}}^\dagger \bar{\eta}_t \hat{F} + \hat{F}^\dagger \eta_{t}). \tag{2}
\]

Here, \( H_s (H_B) \) is the system (bath) Hamiltonian. The total density matrix of the system-and-bath composite can be expressed as the stochastic average on the product of the solutions to Eqs. (1) and (2), i.e.,

\[
\rho_T = \langle \rho_s \rho_B \rangle = \int_{t_0}^t D\eta D\bar{\eta} e^{-\int_{t_0}^t \eta_{s, t} \bar{\eta}_{s, t} dt} \rho_s \rho_B. \tag{3}
\]

Here, \( D\eta D\bar{\eta} e^{-\int_{t_0}^t \eta_{s, t} \bar{\eta}_{s, t} dt} \) is the Grassmann–Wiener measure, with \( \eta = \{ \eta_{jt} \} \) and \( \bar{\eta} = \{ \bar{\eta}_{jt} \} \). By using the Itô’s formula [40], one can easily verify that Eqs. (1) and (2), the result of mapping (i) in Fig. 1 recover exactly the Schrödinger equation for \( \rho_T \).

The primary goal of theoretical formulation is to acquire the reduced system density matrix \( \rho = tr_B (\rho_T) = \langle \rho_{s, t} (tr_B \rho_B) \rangle \), with which the expectation value of any system observable, \( O = tr_s (\bar{O} \rho) \), can be evaluated. Here, \( tr_B \) (\( tr_s \)) denotes the trace over the bath (system) subspace. For a noninteracting fermion bath being initially in thermal equilibrium, one can formally solve Eq. (4) via the Magnus expansion [41] and explicitly evaluate \( tr_B (\rho) \) [39]. The influence of bath on reduced system dynamics can be represented in a compact way, by introducing bath-induced Grassmann fields, \( g^+ \), which depend on the original AGFs in a linear yet time-nonlocal manner:

\[
g^+_t = \int_{t_0}^t \{ [C^+ (t - \tau)]^* \eta_{4t} - iC^- (t - \tau) \eta_{2t} \} d\tau, \tag{4}
\]

\[
g^-_t = \int_{t_0}^t \{ [C^- (t - \tau)]^* \eta_{3t} - iC^+ (t - \tau) \eta_{1t} \} d\tau. \tag{5}
\]

FIG. 1. Schematic illustration of the establishment of a stochastic method for fermionic open systems proposed in this Letter. After a series of mappings, labeled by (i), (ii) and (iii), the influence of fermion bath on the system dynamics is finally captured by conventional c-number fields and a set of ladders with each consisting of three pseudo-states.

Here, \( C^+ (t - \tau) = tr_B [\hat{F}(t) \hat{F}(\tau) \rho_B^{eq}] \) and \( C^- (t - \tau) = tr_B [\hat{F}(t) \hat{F}^\dagger (\tau) \rho_B^{eq}] \) are bath correlation functions, with \( \hat{F}(t) = e^{iH_B t} \hat{F} e^{-iH_B t} \). The reduced system density matrix is given by \( \rho = \langle \hat{\rho}_s \rangle \), with \( \hat{\rho}_s \) satisfying the following rigorous SEOM:

\[
\hat{\rho}_s = -i[H_s, \hat{\rho}_s] + e^{-i\pi/4} (\hat{c}^\dagger \eta_{1t} + \bar{\eta}_{2t} \hat{c}) \hat{\rho}_s + e^{i\pi/4} \hat{\rho}_s (\hat{c}^\dagger \bar{\eta}_{3t} + \eta_{4t} \bar{c}), \tag{6}
\]

This is the result of mapping (ii) in Fig. 1.

As will be shown later, the formally exact fermionic hierarchical equations of motion (HEOM) theory [42, 43], which has been employed to solve quantum impurity problems [44–48], can be established based on Eq. (5). The analogues of Eq. (5) have been obtained in the forms of stochastic quantum Liouville equation [33] and non-Markovian QSD equation [34]. Making practical use of Eq. (5) with conventional stochastic algorithms faces fundamental difficulties, which originate from the aforementioned nature of Grassmann variables. For instance, although Grassmann variables can be represented by mutually anticommutating matrices, it would require a huge number of matrices with huge dimensions to completely model all the time-dependent AGFs involved in Eq. (5). Such kind of difficulties has prohibited any direct numerical application of Eq. (5) or its analogues.

For practical purposes, the Grassmann–Wiener processes in Eq. (5) need to be replaced by some operable quantities. To show such replacement is possible, let us start with a prototypical equation of motion,

\[
y = y \left[ D(t) \eta_t + \int_{t_0}^t C(\tau) \eta_t d\tau \right]. \tag{6}
\]

While \( \eta_t \) and \( \bar{\eta}_t \) are time-dependent AGFs, \( C(t) \) and \( D(t) \) are conventional functions. The stochastic average, \( \langle y \rangle \), is defined similarly to Eq. (6). Like Eq. (6), Eq. (8) cannot be solved directly.

We now propose by intuition and will verify analytically later a mapping scheme, denoted by

\[
\eta_t \mapsto \nu_t X^- , \quad \bar{\eta}_t \mapsto \nu_t X^+. \tag{7}
\]
By Eq. (7), each pair of AGFs, $\eta_l$ and $\bar{\eta}_l$, is mapped to a Gaussian white noise, $\eta_l$, and a pair of time-independent pseudo-operators, $X^+$ and $X^-$, defined in the space of $S = \{-1, 0, 1\}$. Let $\tilde{y} = \sum_{l \in S} y_l$. The action of $X^\pm$ on $\tilde{y}$ gives $\tilde{y}_l X^\pm = \delta_l^\pm \tilde{y}_l$, with $\delta^+_l = \delta^-_{l+1} = -\delta^-_l = 1$ and $\delta^+_l = 0$. This thus transforms Eq. (6) into a normal stochastic differential equation (SDE) as

$$\dot{\tilde{y}} = \tilde{y} \left[ D(t) v_t X^- + \int_{t_0}^t C(\tau) v_\tau \, d\tau X^+ \right].$$

Equation (8) is illustrated as the result of mapping (iii) in Fig. 1. There, the space $S_j$ is represented by a ladder, whose three rungs, corresponding to the elements $1, 0$ and $-1$, can be interpreted as one-particle, vacuum, and one-hole pseudo-states, respectively. The actions of pseudo-operators $X^\pm$ result in transitions between these pseudo-states. Therefore, the mapping (iii) in Fig. 1 can be viewed as a single-configuration-interaction treatment for the bath, and the system-bath dissipation processes are modeled by stochastic exchanges of fermion particles between the system and the pseudo-states. Moreover, the quantized pseudo-states highlight the non-classical nature of the AGFs associated with the quantum fluctuations of fermions.

We now assess whether and how the mapping (iii) in Fig. 1 retains or compromises the exactness of Eq. (8). This is done by relating Eq. (11) to the HEOM formalism. Traditionally, the HEOM are constructed based on exponential unravelling of bath correlation functions,

$$C^\pm(t) = \sum_m C^\pm_m(t) = \sum_m A^m_m e^{\gamma^\pm_m t},$$

subject to the symmetry $\gamma^+_m = (\gamma^-_m)^*$ via the fermionic fluctuation-dissipation theorem [43]. In the HEOM theory, a general $(I + J)$th-tier auxiliary density operator (ADO) is defined in the path-integral form of $(I$ and $J$ are arbitrary non-negative integers) [43]:

$$\rho^{(\cdots,-,\cdots,+)} = \int D\psi D\psi^* e^{iS_f J_FY} e^{-iS_h} \times B^- \cdots B^- B^+ \cdots B^+ \rho(t_0).$$

Here, $\psi = \{\psi_\tau\}$ and $\psi^* = \{\psi^*_\tau\}$ are Grassmann variables associated with $\tilde{c}^\dagger$ and $\psi = \{\psi_\tau\}$ and $\psi^* = \{\psi^*_\tau\}$ are associated with $\tilde{c}$, respectively. $S_f (S_h)$ is the forward (backward) action functional associated with $H_f, J_FY$ is the Feynman–Vernon influence functional [10, 43], and

$$B^- = -i \int_{t_0}^t \left[ A^+_{\psi^*} (A^-_{\psi})^* \right] e^{\gamma^-_m (t-\tau)}.$$

It can be proved that, based on unravelling of $\tilde{g}_l^\pm$ via Eq. (13) and the one-to-one correspondence between $B^\pm$ and $g^\pm_m$, the $(I + J)$th-tier ADO is retrieved exactly by the formal solution of Eq. (13) as in [39]

$$\rho^{(\cdots,-,\cdots,+)} = e^{i(I + J)\pi/4} \tilde{g}^+_m \cdots \tilde{g}^+_m \tilde{g}^-_m \cdots \tilde{g}^-_m \rho(t_0).$$

In parallel, the $(I + J)$th-tier ADO from Eq. (11) reads

$$\rho^{(\cdots,-,\cdots,+)} = e^{i(I + J)\pi/4} \tilde{g}^+_m \cdots \tilde{g}^+_m \tilde{g}^-_m \cdots \tilde{g}^-_m \rho(t_0).$$
Such a density variable is automatically zero if it involves two or more identical pseudo-operators $X^\pm_j$. This is because $\{(X^\pm_j)^p \hat{g}\}^{[00]} = \{\hat{g} (X^\pm_j)^p\}^{[00]} = 0$ holds for any vector $\hat{g}$ in the space $V$ and $p \geq 2$. In the context of HEOM, this amounts to setting all interference ADOs to zero. Here, interference means the right-hand side of Eq. (14) involves two or more $B^m_n$-terms that differ only in $m$. Consequently, Eq. (11) is not equivalent to the exact HEOM, but correspond to a simplified version of HEOM. It thus becomes clear that the substitution of Eq. (7) is an approximation, which leads to the missing of certain detailed information on the system dissipative dynamics.

Extension of Eqs. (5) and (11) to general multi-level systems is straightforward, and the above assessment remains true. In the case of $H_{\text{in}} = \sum_{\nu=1}^{N_{\text{e}}}(c^\dagger_{\nu} F^\nu c_{\nu} + F^\nu c^\dagger_{\nu} c_{\nu})$ with $N_{\text{e}}$ being the system’s degrees of freedom, the decoupling of system and bath is realized by introducing the AGFs $\{\eta_{\text{AGF},\nu}^j\}$ and $\{\eta_{\text{AGF},\nu}^j\}$, which are then represented by Gaussian white noises $\{\eta_{\text{AGF},\nu}^j\}$ and pseudo-operators $\{X^\pm_j\}$ in the same way as Eq. (7).

The simplified-HEOM (sim-HEOM) method has been established in Ref. [10] with the role of interference ADOs discussed extensively therein. Because of their formal equivalence, the SEOM of Eq. (11) and its multi-level extension share the same features as the sim-HEOM [10]: (a) They yield exact $\rho$ if $C^\pm(t)$ is a single exponential function. This is obvious because the resulting hierarchy does not involve any interference ADO. (b) For general noninteracting systems they preserve the exact reduced single-electron density matrix $\rho$ as well as any system property that can be evaluated from $\rho$. This is because the omitted interference ADOs have no influence on $\rho$. (c) For interacting systems they are in principle approximate, and the interference ADOs are important for the quantitative description of strong correlation effects such as Kondo phenomena. Nevertheless, as will be shown below, Eq. (11) can still provide reasonably accurate predictions for system dynamical properties.

We now demonstrate the practicality and accuracy of Eq. (11) with open electronic systems described by the single-impurity Anderson model. The impurity (system) Hamiltonian is $H_s = \sum_{s=\uparrow,\downarrow} \epsilon_s \hat{n}_s + U \hat{n}_\uparrow \hat{n}_\downarrow$, where $\epsilon_s$ is the energy of spin-$s$ level, $\hat{n}_s$ is the electron number operator, and $U$ is the electron-electron Coulomb interaction energy. The reservoir (bath) Hamiltonian is $H_b = \sum_{k} \epsilon_k \hat{n}_{k}$, and its influence on the impurity is characterized by the hybridization functions, which assume a Lorentzian form of $\Delta_s(\omega) \equiv \pi \sum_{k} |t_{ks}|^2 \delta(\omega - \epsilon_k) = \frac{\Gamma}{\pi} \frac{W^2}{(\omega - \epsilon_k)^2 + \Gamma^2}$. Here, $\Gamma$, $\Omega$, and $W$ are the effective impurity-reservoir coupling strength, and the reservoir band-center and bandwidth, respectively.

Suppose at initial time ($t_0 = 0$) the decoupled impurity is doubly occupied by spin-up and spin-down electrons. $H_{\text{in}}$ is turned on at $t > 0$, which triggers the electron transfer between the impurity and reservoir. The time evolution of $\rho(t)$ is obtained by solving a spin-resolved version of Eq. (11). The number of electrons on the impurity is computed by $n_{s}(t) = \text{tr}_{\text{traj}} [\hat{n}_s \rho(t)]$ and shown in Fig. 2 along with quantitatively accurate results obtained by the full HEOM (with all ADOs kept). For all the systems examined in Fig. 2 $U$ assumes an appreciable value, yet the results of our proposed SEOM agree remarkably with the HEOM counterparts.

Regarding numerical efficiency, the SEOM does not require an explicit unravelling of $C^\pm(t)$, and hence its memory cost is substantially smaller than the HEOM. This allows the SEOM to exploit the regime of extremely low temperatures which remains prohibitive for the present HEOM. Moreover, the trajectory-based algorithms for the SEOM could benefit from the massive parallel computing techniques.

Admittedly, there may exist some strongly correlated quantum impurity systems, for which the substitution of Eq. (7) for the AGFs and the resulting SEOM lead to less satisfactory numerical descriptions. Even for such systems, the proposed SEOM still lays a valuable foundation for future development of more sophisticated practical schemes. For instance, the two-electron interaction in $H_s$ can be equivalently characterized by interactions between each electron and auxiliary stochastic fields via a
Hubbard–Stratonovich transformation [50], and thus the system becomes effectively noninteracting. The resulting SEOM is expected to yield the exact $\rho$ for the effective noninteracting system, from which any physical observable of the original interacting dissipative system can be evaluated.

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Supplemental Materials for

Stochastic Equation of Motion for Dissipative Dynamics of Fermionic Open Systems

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References
I. DECOUPLING THE DYNAMICS OF SYSTEM AND BATH

Based on the Gaussian integral for Grassmann variables, we have the equality
\[ e^{A\bar{\psi}\theta} = \int d\bar{\eta}d\eta e^{-\bar{\eta}\eta + \sqrt{\lambda}\psi\eta + \sqrt{\lambda}\bar{\eta}\theta}. \] (S1)

Here, \( A \) is a c-number, and \{\bar{\eta}, \eta, \bar{\psi}, \psi, \theta\} are Grassmann variables which anticommute with each other.

Consider a single-level system coupled to a fermion bath. In the fermionic coherent-state path-integral representation, the forward propagator of the system-bath interaction Hamiltonian is (we set \( \hbar = 1 \) hereafter)
\[
\mathcal{U}_{SB}(t, t_0) = \exp \left\{ -i \int_{t_0}^{t} d\tau H_{SB} \right\} = \int_{t_0}^{t} \mathcal{D}\bar{\psi} \mathcal{D}\psi \mathcal{D}\bar{\theta} \mathcal{D}\theta \ e^{-i \int_{t_0}^{t} (\bar{\psi}_{\tau} \theta_{\tau} + \bar{\theta}_{\tau} \psi_{\tau})} dr.
\]

Here, \( B = e^{-i\pi/4} \), and \{\bar{\eta}_j, \eta_j\} = \{\bar{\eta}_{j\tau}, \eta_{j\tau}\} for \( t_0 < \tau < t \). The time-dependent Grassmann variables \{\bar{\psi}_\tau, \psi_\tau, \bar{\theta}_\tau, \theta_\tau\} are associated with the operators \{\lambda \hat{\xi}^\dagger, \lambda \hat{\xi}^{\dagger}, \lambda^{\frac{1}{2}} \hat{F}^\dagger, \lambda^{\frac{1}{2}} \hat{\xi} \} in the path-integral formulation, with \( \lambda \) being a reference energy of any positive value. For simplicity, we choose \( \lambda = 1 \) in the main text and throughout this Supplemental Material.

The backward propagator \( \mathcal{U}_{SB}^4(t, t_0) \) can be expressed similarly by introducing the auxiliary Grassmann fields (AGFs) \{\bar{\eta}_{3\tau}, \eta_{3\tau}, \bar{\eta}_{4\tau}, \eta_{4\tau}\}. With the use of AGFs, the system and bath is formally decoupled from each other. Instead, they are coupled to the AGFs \{\bar{\eta}_{j\tau}, \eta_{j\tau}\} ( \( j = 1, \ldots, 4 \)). With the initial factorization condition of \( \rho_{\bar{\eta}}(t_0) = \rho_{\bar{\eta}}(t_0) \rho_{\bar{\eta}}(t_0) \), the equations of motion (EOM) for \( \rho_\bar{\eta} \) and \( \rho_\bar{\eta} \) are given by Eqs. (1) and (2) in the main text. The density matrix of the total system is obtained by \( \rho_T = \langle \rho_\bar{\eta} \rho_\bar{\eta} \rangle \). From the Itô’s formula, we have
\[
d\rho_T = \langle (d\rho_\eta) \rho_\bar{\eta} + \rho_\eta (d\rho_\bar{\eta}) + (d\rho_\bar{\eta}) (d\rho_\bar{\eta}) \rangle
\]
\[= -i[H_\eta + H_\bar{\eta}, \rho_T] dt - i \left( \langle \hat{c}^\dagger \eta_1 + \bar{\eta}_2 \hat{c}\rangle \rho_\eta (\bar{\eta}_1 \hat{F} + \hat{F}^\dagger \eta_2) \right) (dt)^2
\]
\[+ i \left( \rho_\eta (\hat{c}^\dagger \eta_3 + \bar{\eta}_4 \hat{c}) \rho_\bar{\eta} (\bar{\eta}_3 \hat{F} + \hat{F}^\dagger \eta_4) \right) (dt)^2
\]
\[= -i[H_\eta + H_\bar{\eta}, \rho_T] dt - i \left( \langle \hat{c}^\dagger \eta_1 + \bar{\eta}_2 \hat{c}\rangle (\bar{\eta}_1 \hat{F} + \hat{F}^\dagger \eta_2) \right) \langle \rho_\eta \rho_\bar{\eta} \rangle (dt)^2
\]
\[+ i \langle \rho_\eta \rho_\bar{\eta} \rangle \left( \langle \hat{c}^\dagger \eta_3 + \bar{\eta}_4 \hat{c}\rangle (\bar{\eta}_3 \hat{F} + \hat{F}^\dagger \eta_4) \right) (dt)^2
\]
\[= -i[H_\eta + H_\bar{\eta} + H_{SB}, \rho_T] dt
\]
\[= -i[H_T, \rho_T] dt. \] (S3)

Here, we have used the causality relation that \( \rho_\eta \) and \( \rho_\bar{\eta} \) at time \( t \) depend only on AGFs at time \( \tau \leq t \); the equalities \( \langle \eta_{j\tau} \rangle = \langle \eta_{j\tau} \rangle = 0 \) and \( \langle \eta_{j\tau} \eta_{j'\tau'} \rangle = \delta_{jj'} \delta(\tau - \tau') \); together with the equalities \( \rho_\eta (\bar{\eta}_1 \hat{F} + \hat{F}^\dagger \eta_2) = (\bar{\eta}_1 \hat{F} + \hat{F}^\dagger \eta_2) \rho_\eta \) and \( (\hat{c}^\dagger \eta_3 + \bar{\eta}_4 \hat{c}) \rho_\bar{\eta} = \rho_\bar{\eta} (\hat{c}^\dagger \eta_3 + \bar{\eta}_4 \hat{c}) \).
In the $H_B$–interaction picture, define the bath density matrix $\tilde{\rho}_B = e^{i \int_0^t H_B dt} \rho_b e^{-i \int_0^t H_B dt}$ and the operator $\tilde{F}(t) = e^{i \int_0^t H_B dt} \tilde{F} e^{-i \int_0^t H_B dt}$. From Eq. (2) of main text, we have

$$\dot{\tilde{\rho}}_B = B [\tilde{\eta}_t \tilde{F}(t) + \tilde{F}^\dagger(t) \eta_2 t] \tilde{\rho}_B + B^* \tilde{\rho}_B [\tilde{\eta}_t \tilde{F}(t) + \tilde{F}^\dagger(t) \eta_4 t],$$

(S4)

which can be solved formally by the Magnus expansion. If the bath is initially in thermal equilibrium, and $\rho_B(t_0) = \rho_B^{eq}$ satisfies Gaussian statistics, we have

$$\text{tr}_B (\rho_B) = \text{tr}_B (\tilde{\rho}_B) = e^{\int_0^t [\tilde{\eta}_r - i \tilde{\eta}_r \gamma_r \tilde{\rho}^\dagger + (\gamma_r - i \gamma_r) \tilde{\rho}] dr}.$$

(S5)

II. FORMAL EQUIVALENCE BETWEEN EQ. (5) OF MAIN TEXT AND THE FERMIONIC HEOM FORMULATION

The fermionic hierarchical equations of motion (HEOM) are constructed based on unravelling of two-time bath correlation functions by exponential functions: $C^\sigma(t) = \sum_m \sigma_m(t) = \sum_m A^\sigma_m e^{\gamma_m^\sigma t}$ with $\sigma = +$ or $-$. In the HEOM theory, the EOM for $\rho^{(-\cdots-\cdots+)}_{m_1 \cdots m_{1+n} \cdots n}$ can be recast into a compact form of

$$\dot{\rho}^{(-\cdots-\cdots+)}_{m_1 \cdots m_{1+n} \cdots n} = \left(-i \mathcal{L}_S + \sum_{i=1}^J \gamma_{m_i}^- + \sum_{j=1}^I \gamma_{n_j}^+ \right) \rho^{(-\cdots-\cdots+)}_{m_1 \cdots m_{1+n} \cdots n} + \sum_{i=1}^J \mathcal{C}_{m_i}^- \rho^{(-\cdots-\cdots+)}_{m_1 \cdots m_{i-1} m_{i+1} \cdots n}$$

$$+ \sum_{j=1}^I \mathcal{C}_{n_j}^+ \rho^{(-\cdots-\cdots+)}_{m_1 \cdots n_{j-1} n_{j+1} \cdots n} + \sum_{\sigma=+,-} \sum_{r} A^\sigma_r \rho^{(-\cdots-\cdots+)}_{m_1 \cdots m_{1+n} \cdots n},$$

(S6)

where $\mathcal{L}_S \equiv [H_s, \cdot]$. The detailed forms of the superoperators $\{\mathcal{C}_{m_i}^-, \mathcal{C}_{n_j}^+, A^\sigma_r\}$ have been given by Eqs. (26)–(29) in Ref. [1].

Regarding the Grassmann-valued SEOM for $\tilde{\rho}_S$ given by Eq. (5) in the main text, the bath-induced AGFs are decomposed as $g^\sigma_i = \sum_m g^\sigma_{m} i(t)$. The EOM for each component is self-closed:

$$\dot{g}^-_m = [-i A^-_m \eta_2 t + (A^+_m)^* \eta_4 t] + \gamma^-_m g^-_m,$$

$$\dot{g}^+_m = [-i A^+_m \eta_2 t + (A^-_m)^* \eta_4 t] + \gamma^+_m g^+_m.$$

(S7)

The same $(I + J)$th-tier ADO can be retrieved by $\tilde{\rho}_S$ of Eq. (5) in the main text as follows:

$$\rho^{(-\cdots-\cdots+)}_{m_1 \cdots m_{1+n} \cdots n} \equiv (B^*)^{I+J} \langle g^-_{m_1} \cdots g^-_{m_I} \tilde{\rho}_S g^+_{n_1} \cdots g^+_{n_J} \rangle.$$

(S8)

Based on Itô’s formula, its differential consists of three parts:

$$d\rho^{(-\cdots-\cdots+)}_{m_1 \cdots m_{1+n} \cdots n} = \Xi_1 + \Xi_2 + \Xi_3.$$

(S9)

Presuming the system creation and annihilation operators ($\hat{c}$ and $\hat{c}^\dagger$) commute with all the AGFs, and using the equality

$$\langle f(t) g^\sigma_m(t) \rangle = -\langle g^\sigma_m(t) f(t) \rangle,$$

(S10)
which holds for any analytic function \( f(t) \) of the AGFs \( \{ \eta_{\ell}, \eta_t \} \), we express the three parts of \( d\rho_{m_1\cdots m_j\cdots n_1\cdots n_j} \) respectively as follows.

\[
\Xi_1 = (B^*)^{I+J} \langle \tilde{g}_{m_1} \cdots \tilde{g}_{m_j} d\tilde{\rho}_s g_{n_1}^+ \cdots g_{n_j}^+ \rangle \\
= -i \int [H_S, \rho_{m_1\cdots m_j\cdots n_1\cdots n_j}] dt - i (B^*)^{I+J+1} \sum_r \left( \hat{c}^\dagger \langle \cdots \tilde{g}_{m_1} g_r^- \tilde{\rho}_s \cdots \rangle + \langle \cdots \tilde{\rho}_s g_r^- g_{n_1}^+ \cdots \rangle \hat{c}^\dagger \right) \\
- \hat{c} \langle \cdots \tilde{g}_{m_1} g_r^+ \tilde{\rho}_s \cdots \rangle - \langle \cdots \tilde{\rho}_s g_r^+ g_{n_1}^+ \cdots \rangle \hat{c} \right) dt \\
= -i \int [H_S, \rho_{m_1\cdots m_j\cdots n_1\cdots n_j}] dt - i \sum_r \left( \hat{c}^\dagger \rho_{m_1\cdots m_j \cdots n_1\cdots n_j} - (I+J)^{I+J+1} \rho_{m_1\cdots m_j \cdots n_1\cdots n_j} \hat{c}^\dagger \\
+ (-1)^{I+J} \hat{c} \rho_{m_1\cdots m_j \cdots n_1\cdots n_j} \right) dt. 
\]

(S11)

The causality relation ensures \( \tilde{\rho}_s(t) \) and \( \{ g^\rho_m(t) \} \) depend only on the AGFs prior to the time \( t \), and thus we have

\[
\Xi_2 = \sum_{i=1}^I \langle \cdots d\tilde{g}_{m_1} \cdots \rangle + \sum_{j=1}^J \langle \cdots \tilde{\rho}_s \cdots d\rho_{n_1}^+ \cdots \rangle = \left( \sum_{i=1}^I \gamma_{m_i}^- + \sum_{j=1}^J \gamma_{n_j}^+ \right) \rho_{m_1\cdots m_j\cdots n_1\cdots n_j} dt. 
\]

(S12)

\[
\Xi_3 = \sum_{i=1}^I \langle \cdots d\tilde{g}_{m_1} \cdots \rangle + \sum_{j=1}^J \langle \cdots d\tilde{\rho}_s \cdots d\rho_{n_1}^+ \cdots \rangle \\
= (B^*)^{I+J-1} \left\{ \sum_{i=1}^I -iA_{m_i}^- \hat{c} \langle \cdots \tilde{g}_{m_{i-1}} \eta_{2t} g_{m_{i+1}}^- \cdots \rangle + i(A_{m_i}^+)^* \langle \cdots \tilde{g}_{m_{i-1}} \eta_t g_{m_{i+1}}^- \cdots \tilde{\rho}_s \eta_{2t} \cdots \rangle \hat{c}^\dagger \right. \\
+ \left. \sum_{j=1}^J -iA_{n_j}^+ \hat{c}^\dagger \langle \cdots \eta_{t \cdots} \rho_{n_{j-1}}^- g_{n_{j+1}}^+ \cdots \rangle + i(A_{n_j}^-)^* \langle \cdots \rho_{n_{j-1}}^- g_{n_{j+1}}^+ \cdots \eta_{t \cdots} \cdots \rangle \hat{c} \right\} (dt)^2 \\
= -i \sum_{i=1}^I \left( A_{m_i}^- (-1)^{I-i} \hat{c} \rho_{m_1\cdots m_{i-1} m_{i+1} \cdots n_j} - (A_{m_i}^+)^* (-1)^{I-i} \rho_{m_1\cdots m_{i-1} m_{i+1} \cdots n_j} \hat{c} \right) dt \\
- i \sum_{j=1}^J \left( A_{n_j}^+ (-1)^{I+J-j} \hat{c} \rho_{m_1\cdots m_j \cdots n_{j-1} n_{j+1} \cdots n_j} - (A_{n_j}^-)^* (-1)^{I+J-j} \rho_{m_1\cdots m_j \cdots n_{j-1} n_{j+1} \cdots n_j} \hat{c} \right) dt. 
\]

(S13)

Here, we have used the equalities \( BB^* = 1 \) and \( (B^*)^2 = i \), and

\[
\langle \cdots \tilde{g}_{m_{i-1}} \eta_{2t} g_{m_{i+1}}^- \cdots \rangle dt = (-1)^{I-i} \langle \eta_{2t} \eta_{2t} \cdots \tilde{g}_{m_{i-1}} g_{m_{i+1}}^- \cdots \tilde{\rho}_s \cdots \rangle dt \\
= (-1)^{I-i} \rho_{m_1\cdots m_{i-1} m_{i+1} \cdots n_j}. 
\]

(S14)

Apparently, the EOM for \( \rho_{m_1\cdots m_j\cdots n_1\cdots n_j} \) defined by Eq. (S8) is formally identical to Eq. (S6).
III. EQUIVALENCE BETWEEN THE SOLUTIONS OF EQ. (6) AND EQ. (8) OF MAIN TEXT

We discretize the time domain by setting $t_0 = 0$ and $t = N_t \ dt$, with $dt$ being the infinitesimal increment time step and $N_t$ the number of steps. At $t_i = i dt$ and $t_{i+1} = (i+1) dt$, Eq. (6) of the main text leads to

$$
d y_i = y_i D_i \eta_i + y_i (C_0 \bar{\eta}_0 + C_1 \bar{\eta}_1 + \cdots + C_{i-1} \bar{\eta}_{i-1} + C_i \bar{\eta}_i), \quad \text{(S15)}$$

$$
d y_{i+1} = y_{i+1} D_{i+1} \eta_{i+1} + y_{i+1} (C_0 \bar{\eta}_0 + C_1 \bar{\eta}_1 + \cdots + C_i \bar{\eta}_i + C_{i+1} \bar{\eta}_{i+1}). \quad \text{(S16)}$$

By causality, $d y_i$ and $y_{i+1}$ depend on $\{\eta_0, \cdots, \eta_i; \bar{\eta}_0, \cdots, \bar{\eta}_i\}$. Taking the average over Grassmann fields $\{\eta_j, \eta_j\}$ $(0 \leq j \leq i + 1)$ for both sides of Eq. (S16), we have

$$\langle d y_{i+1} \rangle = \langle y_{i+1} D_{i+1} \eta_{i+1} \rangle + \langle (y_i + d y_i) (C_0 \bar{\eta}_0 + \cdots + C_{i-1} \bar{\eta}_{i-1} + C_i \bar{\eta}_i + C_{i+1} \bar{\eta}_{i+1}) \rangle$$

$$= \langle y_i (C_0 \bar{\eta}_0 + \cdots + C_{i-1} \bar{\eta}_{i-1} + C_i \bar{\eta}_i) \rangle + \langle y_i (C_0 \bar{\eta}_0 + \cdots + C_{i-1} \bar{\eta}_{i-1} + C_i \bar{\eta}_i + C_{i+1} \bar{\eta}_{i+1}) \rangle$$

$$= \langle y_i D_i \eta_i \rangle + \langle (y_i + d y_i) (C_0 \bar{\eta}_0 + \cdots + C_{i-1} \bar{\eta}_{i-1} + C_i \bar{\eta}_i + C_{i+1} \bar{\eta}_{i+1}) \rangle$$

$$= \langle d y_i \rangle + D_i C_i \langle y_i \rangle. \quad \text{(S17)}$$

Here, the last equality makes use of the causality relation that $y_i$ is independent of $\eta_i$ and $\bar{\eta}_i$. Equation (S17) thus gives the formal solution of $\langle y \rangle$ in the form of recursive relation for $\langle d y_i \rangle$ at discretized time steps.

Now, with the mapping $\eta_i \mapsto v_i X^-$ and $\bar{\eta}_i \mapsto v_i X^+$, $\tilde{y} = \sum_{t \in \{-1,0,1\}} \tilde{y}^0$. Eqs. (S15) and (S16) are replaced by

$$d \tilde{y}_i = \tilde{y}_i D_i v_i X^- + \tilde{y}_i (C_0 \bar{v}_0 + \cdots + C_{i-1} \bar{v}_{i-1} + C_i \bar{v}_i) X^+, \quad \text{(S18)}$$

$$d \tilde{y}_{i+1} = \tilde{y}_{i+1} D_{i+1} v_{i+1} X^- + \tilde{y}_{i+1} (C_0 \bar{v}_0 + \cdots + C_{i-1} \bar{v}_{i-1} + C_i \bar{v}_i + C_{i+1} \bar{v}_{i+1}) X^+. \quad \text{(S19)}$$

We have

$$\langle d \tilde{y}_{i+1} \rangle = \langle \tilde{y}_{i+1} D_{i+1} v_{i+1} X^- \rangle + \langle (\tilde{y}_i + d \tilde{y}_i) (C_0 \bar{v}_0 + \cdots + C_{i-1} \bar{v}_{i-1} + C_i \bar{v}_i + C_{i+1} \bar{v}_{i+1}) X^+ \rangle$$

$$= \langle d \tilde{y}_i \rangle + \langle \tilde{y}_i D_i v_i X^- \rangle + \langle \tilde{y}_i (C_0 \bar{v}_0 + \cdots + C_{i-1} \bar{v}_{i-1} + C_i \bar{v}_i + C_{i+1} \bar{v}_{i+1}) X^+ \rangle$$

$$= \langle \tilde{y}_i (C_0 \bar{v}_0 + \cdots + C_{i-1} \bar{v}_{i-1} + C_i \bar{v}_i + C_{i+1} \bar{v}_{i+1}) X^+ \rangle$$

$$= \langle d \tilde{y}_i \rangle + D_i C_i \langle \tilde{y}_i \rangle. \quad \text{(S20)}$$

Here, $\langle \tilde{y} \rangle = M(\tilde{y}^0)$. To achieve the last equality of Eq. (S20), we need to have $\langle \tilde{y} X^- X^+ \rangle = \langle \tilde{y} \rangle$ and $\langle \tilde{y} (X^+)^2 \rangle = 0$. These can be easily satisfied, e.g., by setting

$$\tilde{y}_i^{[-1]} X^- = 0, \quad \tilde{y}_i^{[0]} X^- = \tilde{y}_i^{[-1]}, \quad \tilde{y}_i^{[1]} X^- = -\tilde{y}_i^{[0]}, \quad \tilde{y}_i^{[0]} X^+ = \tilde{y}_i^{[1]}, \quad \tilde{y}_i^{[1]} X^+ = 0. \quad \text{(S21)}$$
Apparently, Eq. (S20) is identical to Eq. (S17). Therefore, the solution of Eq. (8) in the main text exactly retrieves that of Eq. (6).

IV. REMARKS ON THE LADDER PSEUDO-OPERATORS

In the context of Eq. (10) of the main text, the ladder pseudo-operators $X_1^\pm$ and $X_2^\pm$ can act to the left and right of $\tilde{\rho}_S$, which yield

$$
X_1^+ \tilde{\rho}_S^{[l_1,l_2]} = (-1)^{l_1+l_2} \tilde{\rho}_S^{[l_1,l_2]} X_1^+ = \lambda_{(-1,0)}^{l_1,} (-1)^{l_1} \tilde{\rho}_S^{[l_1+1,l_2]}, \\
X_1^- \tilde{\rho}_S^{[l_1,l_2]} = (-1)^{l_1+l_2} \tilde{\rho}_S^{[l_1,l_2]} X_1^- = \lambda_{(0,1)}^{l_1} \tilde{\rho}_S^{[l_1-1,l_2]}, \\
X_2^+ \tilde{\rho}_S^{[l_1,l_2]} = (-1)^{l_1+l_2} \tilde{\rho}_S^{[l_1,l_2]} X_2^+ = \lambda_{(-1,0)}^{l_2} (-1)^{l_1+l_2} \tilde{\rho}_S^{[l_1,l_2+1]}, \\
X_2^- \tilde{\rho}_S^{[l_1,l_2]} = (-1)^{l_1+l_2} \tilde{\rho}_S^{[l_1,l_2]} X_2^- = \lambda_{(0,1)}^{l_2} (-1)^{l_1} \tilde{\rho}_S^{[l_1,l_2-1]}.
$$

(S22)

Here, $\lambda_A^l = 1$ (if $l \in A$) or 0 (if $l \notin A$) is a step function, which ensures the action of $X_j^\sigma$ does not exceed the boundary of ladder space $S_j$. In the following, we elaborate more on the construction of these pseudo-operators.

Each pseudo-operator can be associated with a time-independent Grassmann variable, i.e.,

$$
X_j^+ \mapsto \bar{\xi}_j, \quad X_j^- \mapsto \xi_j.
$$

(S23)

A one-to-one mapping can be established between the ladder pseudo-states $[l_1, l_2]$ and the normal-ordered monomials of Grassmann variables $\{\xi_1, \xi_1, \bar{\xi}_2, \xi_2\}$. Here, a monomial is considered to be in normal order if its constituent Grassmann variables follow the sequence of $\xi_1 \bar{\xi}_1 \xi_2 \bar{\xi}_2$. For instance, $\xi_1 \xi_2$ and $\bar{\xi}_1 \bar{\xi}_2$ are in normal order, while $\bar{\xi}_2 \xi_1$ and $\xi_2 \bar{\xi}_1$ are not. This means that any vector $f$ in the space $V = V_S \otimes S_1 \otimes S_2$ can be represented uniquely by a polynomial of $\{\xi_1, \xi_1, \bar{\xi}_2, \xi_2\}$ as follows,

$$
f = \sum_{l_1 \in S_1} \sum_{l_2 \in S_2} f^{[l_1,l_2]} \mapsto \sum_{p_1, p_2, p_3, p_4 \in \{0,1\}} B_{p_1 p_2 p_3 p_4} \xi_1^{p_1} \bar{\xi}_1^{p_2} \xi_2^{p_3} \bar{\xi}_2^{p_4}.
$$

(S24)

The ladder space $S_j$ is spanned by only three pseudo-states. Specifically, $l_j = -1, 0$, and 1 correspond to $\xi_j, 1$, and $\bar{\xi}_j$, respectively; whereas there is no pseudo-state representing the dual variables $\xi_j \bar{\xi}_j$. Consequently, all monomials involving dual variables are suppressed in the polynomial of Eq. (S24), i.e., $B_{p_1 p_2 p_3 p_4} = 0$ if $p_1 = p_2 = 1$ or $p_3 = p_4 = 1$.

Take the first line of Eq. (S22) as an example — the action of $X_1^+$ to the left of $f^{[l_1,l_2]}$ gives

$$
X_1^+ f^{[l_1,l_2]} = \lambda_{(-1,0)}^{l_1} (-1)^{l_1} f^{[l_1+1,l_2]}.
$$

(S25)

Here, the step function $\lambda_{(-1,0)}^{l_1}$ enforces the action returns zero if $(l_1 + 1)$ exceeds the upper bound of the ladder. Suppose the pseudo-states $[l_1, l_2]$ and $[l_1 + 1, l_2]$ correspond to the normal-ordered monomials 1 and 2, respectively. The left action of $X_1^+$ amounts to multiplying $\bar{\xi}_1$ to the left of the monomial 1, which results in the monomial 1' (with dual variables suppressed). The prefactor $(-1)^{l_1} = 1$ (or −1) indicates that it requires an even (or odd) number of swaps of Grassmann variables to rearrange the monomial 1’ into the normal-ordered monomial 2.
For instance, the pseudo-state \([l_1, l_2] = [-1, 1]\) corresponds to the normal-ordered monomial \(\xi_1 \xi_2\), and \([l_1 + 1, l_2] = [0, 1]\) corresponds to the monomial \(\bar{\xi}_2\). Multiplying \(\bar{\xi}_1\) to the left of \(\xi_1 \bar{\xi}_2\) yields \(\xi_1 \bar{\xi}_1\xi_2 = -\xi_1 \bar{\xi}_1 \bar{\xi}_2 \mapsto -\bar{\xi}_2\). In the last step, the dual term \(\xi_1 \bar{\xi}_1\) is suppressed (reduced to 1), and the resulting minus sign recovers the prefactor \((-1)^{l_1} = -1\) in Eq. (S25).

V. FORMAL EQUIVALENCE BETWEEN EQ. (11) OF MAIN TEXT AND THE SIMPLIFIED-HEOM FORMULATION

In relation to Eq. (S8), with the AGFs represented by Gaussian white noises and ladder pseudo-operators, the \((I + J)\)th-tier ADO is constructed by

\[ ρ^{(−⋯−±⋯+)}_{m_1...m_J n_1...n_J} = (B^*)^{I+J} \langle \tilde{g}_{m_1} X_2^− \cdots \tilde{g}_{m_J} X_2^− \tilde{\rho} g_{n_1} X_1^+ \cdots \tilde{g}_{n_J} X_1^+ \rangle, \]  

(S26)

where \(\{\tilde{g}_m\}\) and \(\{\tilde{g}^+_n\}\) are bath-induced stochastic fields

\[ \tilde{g}_m(t) = \int_{t_0}^t [-iA_m^− v_{2r} + (A^+ m)_* v_{1r}] e^{\gamma_m(t-\tau)} d\tau, \]

\[ \tilde{g}^+_n(t) = \int_{t_0}^t [-iA^+_n v_{1r} + (A_n)_* v_{3r}] e^{\gamma_n^+(t-\tau)} d\tau. \]  

(S27)

For each ladder space \(S_j\), only one pseudo-operator \((X^+_j \text{ or } X^−_j)\) is involved in the construction of ADOs, and pseudo-operators belonging to different ladder spaces anticommute with each other.

Because of the finite dimension of \(S_j\), \(\langle (X^\sigma_j)^p f \rangle = \langle f (X^\sigma_j)^p \rangle = 0\) holds for any \(f = \sum_{l_1 \in S_1} \sum_{l_2 \in S_2} f[l_1,l_2] \text{ and } p \geq 2\). Regarding Eq. (S26), it is immediately recognized that the ADO is zero if the right-hand side involves two or more identical \(X^\sigma_j\). In the context of original HEOM, this amounts to setting any ADO that involves two or more \(B^\sigma_m\)–terms that differ only in the index \(m\) to zero; see Eq. (15) of main text. Such ADOs are referred to as interference ADOs, which are important for the accurate description of strongly correlated states in fermionic dissipative systems.

By using the property that \(X_j^\pm\) commute with \(\hat{c}\) and \(\hat{c}^\dagger\), as well as the equality

\[ \langle X^\sigma_j f \rangle = -\langle f X^\sigma_j \rangle, \]  

(S28)

which is in parallel with Eq. (S10), the time differential of any nonzero \(ρ^{(−⋯−±⋯+)}_{m_1...m_J n_1...n_J}\) is also given by Eqs. (S11), (S12) and (S13). Therefore, Eq. (11) of main text is formally equivalent to the simplified-HEOM (sim-HEOM) formulation in which all interference ADOs are omitted from the original HEOM. The detailed derivation and important properties of the sim-HEOM method have been elaborated in Ref. [1].

In general cases where a multi-level system is coupled to more than one fermion baths, the \(\tilde{g}_m^\sigma X^\sigma_j\) in Eq. (S26) is replaced by \(\tilde{g}^\sigma_{\nu \alpha} X^\sigma_j\), where \(\nu\) labels the system levels and \(\alpha\) labels the baths. Correspondingly, \(B^\sigma_m\) is replaced by \(B^\sigma_{\nu \alpha}\). Again, interference means the ADO involves two or more \(B^\sigma_{\nu \alpha}\)–terms that differ only in \(m\). The HEOM formulation developed by omitting such interference ADOs is termed as the sim-HEOM–\(\alpha\); see Ref. [1]. Therefore, the multi-level-and-multi-bath extension of Eq. (11) is formally equivalent to the sim-HEOM–\(\alpha\) method.
VI. EXTENSION OF EQ. (11) OF MAIN TEXT TO SPIN-RESOLVED CASES

For the single-impurity Anderson model studied in the main text, the system involves explicitly the spin degree of freedom (labeled by \( s = \uparrow, \downarrow \)). The Grassmann-valued SEOM for \( \hat{\rho}_s \) is

\[
\dot{\hat{\rho}}_s = -i[H_s, \hat{\rho}_s] + \sum_s \left[ B\left( \hat{c}_s^\dagger \eta_{1st} + \bar{\eta}_{2st} \hat{c}_s \right) \hat{\rho}_s + B^* \hat{\rho}_s \left( \hat{c}_s^\dagger \eta_{3st} + \bar{\eta}_{4st} \hat{c}_s \right) \right] + \sum_s B \left\{ \hat{c}_s^\dagger g_s^-(t) - g_s^+(t) \hat{c}_s, \hat{\rho}_s \right\},
\]

where \( \{ g_s^\sigma(t) \} \) are given by

\[
g_s^-(t) = \int_{t_0}^t \left\{ [C_s^+(t - \tau)]^* \eta_{4st} - iC_s^-(t - \tau) \eta_{2st} \right\} d\tau, \\
g_s^+(t) = \int_{t_0}^t \left\{ [C_s^-(t - \tau)]^* \eta_{3st} - iC_s^+(t - \tau) \eta_{1st} \right\} d\tau.
\]

Here, \( C_s^+(t - \tau) = \text{tr}_B [\hat{F}_s(t) \hat{F}_s(\tau) \rho_B^{\text{eq}}] \) and \( C_s^-(t - \tau) = \text{tr}_B [\hat{F}_s(t) \hat{F}_s(\tau) \rho_B^{\text{eq}}] \), with \( \hat{F}_s(t) \equiv e^{i\int_{t_0}^t H_B d\tau} \hat{F}_s e^{-i\int_{t_0}^t H_B d\tau} \) and \( \hat{F}_s = \sum_k t_{ks} \hat{d}_{ks} \). By substituting the AGFs \( \{ \eta_{jst}, \bar{\eta}_{jst} \} \) with

\[
\eta_{jst} \mapsto v_{jst} X_j^{-}, \quad \bar{\eta}_{jst} \mapsto v_{jst} X_j^{+},
\]

Eq. (S29) is recast into the following numerically feasible form of

\[
\dot{\hat{\rho}}_s = -i[H_s, \hat{\rho}_s] + \sum_s \left( B \hat{c}_s^\dagger Y_{1s} \hat{\rho}_s + B Y_{2s} \hat{c}_s \hat{\rho}_s + B^* \hat{\rho}_s \hat{c}_s^\dagger Y_{3s} + B^* \hat{\rho}_s Y_{4s} \hat{c}_s \right),
\]

where \( \hat{\rho}_s = \sum_{l_1 \in S_1} \sum_{l_2 \in S_2} \sum_{l_3 \in S_3} \sum_{l_4 \in S_4} \hat{\rho}_s^{[l_1 l_2 l_3 l_4]} \), and

\[
Y_{1s} = v_{1st} X_{1s}^- + \bar{g}_{st} X_{2s}^-, \quad Y_{2s} = v_{2st} X_{2s}^+ - \bar{g}_{st} X_{1s}^+, \\
Y_{3s} = v_{3st} X_{3s}^- - i\bar{g}_{st} X_{2s}^-, \quad Y_{4s} = v_{4st} X_{2s}^+ + i\bar{g}_{st} X_{1s}^+.
\]

Here, the spin-resolved bath-induced stochastic fields \( \{ \tilde{g}_s^{\sigma} \} \) are

\[
\tilde{g}_s^- = \int_{t_0}^t \left\{ [C_s^+(t - \tau)]^* v_{4st} - iC_s^-(t - \tau) v_{2st} \right\} d\tau, \\
\tilde{g}_s^+ = \int_{t_0}^t \left\{ [C_s^-(t - \tau)]^* v_{3st} - iC_s^+(t - \tau) v_{1st} \right\} d\tau.
\]

The physical reduced system density matrix is finally obtained by

\[
\rho = \langle \hat{\rho}_s \rangle = M \left( \hat{\rho}_s^{[0000]} \right).
\]
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1 L. Han, H.-D. Zhang, X. Zheng, and Y. J. Yan, J. Chem. Phys. 148 (2018).