Hierarchical equations of motion method based on Fano spectrum decomposition for low temperature environments

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ABSTRACT

The hierarchical equations of motion (HEOM) method has become one of the most popular methods for the studies of the open quantum system. However, its applicability to systems at ultra-low temperatures is largely restrained by the enormous computational cost, which is caused by the numerous exponential functions required to accurately characterize the non-Markovian memory of the reservoir environment. To overcome this problem, a Fano spectrum decomposition (FSD) scheme has been proposed recently [Cui et al., J. Chem. Phys. 151, 024110 (2019)], which expands the reservoir correlation functions using polynomial-exponential functions and hence greatly reduces the size of the memory basis set. In this work, we explicitly establish the FSD-based HEOM formalisms for both bosonic and fermionic environments. The accuracy and efficiency of the FSD-based HEOM are exemplified by the calculated low-temperature dissipative dynamics of a spin-boson model and the dynamic and static properties of a single-orbital Anderson impurity model in the Kondo regime. The encouraging numerical results highlight the practicality and usefulness of the FSD-based HEOM method for general open systems at ultra-low temperatures.

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I. INTRODUCTION

Open quantum systems have found widespread applications in physical, chemical, and biological systems and processes.1 The dissipative interactions between a quantum system and its environment often have profound influence on the dynamics and thermodynamics of the system. The underlying physics becomes particularly intriguing when the system-environment coupling is strong and the environmental modulation possesses a long-time memory. For instance, when the temperature declines to near zero, many fascinating phenomena arise, such as the Kondo effect in strongly correlated electronic systems and Bose–Einstein condensation.2–4 A comprehensive understanding of such many-body effects calls for non-perturbative and non-Markovian treatments.

To date, a variety of formally exact methods have been developed. These include the numerical renormalization group (NRG) theory,9,10 the quantum Monte Carlo (QMC) method,11–14 the density matrix renormalization group (DMRG) method15 and its time-dependent extension (TD-DMRG),16–18 the hierarchical equations of motion (HEOM) approach,19–27 the multi-configuration time-dependent Hartree (MCTDH) method28–30 and its multi-layer extension (ML-MCTDH)31,32 and second-quantized version (ML-MCTDH-SQR),31,32 the stochastic equation of motion (SEOM) method,33–37 and the real-time path integral methods.38–40 Among them, the HEOM method is currently a standard and universal tool for the studies of open systems. It has been extensively applied to study coherent energy or heat transfer,41–43 quantum electron transport,44–50 nonlinear spectroscopy,51–55 Kondo physics,56–60 and thermoelectric and local heating effects in quantum dots.56–63 With the HEOM method, to obtain the reduced system dynamics, one needs to propagate the system reduced density matrix.
together with a hierarchical set of auxiliary density operators (ADOs). Even in the long-time regime, the ADOs are still non-negligible, as they account for the non-Markovian memory of the system-environment coupling, as well as for the multi-particle or high-order dissipation processes. Schematically, the hierarchy of reduced and auxiliary density matrices is often represented by a two-dimensional pyramid. The horizontal dimension of the pyramid resolves all the available dissipation modes between the system and environment, and the width of the hierarchy is determined by the number of memory basis functions $P$ used to unravel the environmental correlation functions; while the vertical dimension resolves all the possible dissipation events that could occur through the available modes, and the height of hierarchy is determined by the terminal tier $L$. 

Despite the increasing popularity of the HEOM method, its limitation is well-known: the cost of computer memory can be rather large if the environment is highly non-Markovian ($P$ is large) or if the many-particle correlation or system-environment entanglement is strong ($L$ is large). It is generally believed that the practicality of HEOM is restricted in the high or intermediate temperature regime. This is because as the temperature approaches zero, the particle distribution function, such as the Fermi/Bose function, exhibits an abrupt jump at the zero frequency, which gives rise to the $1/\omega$ decay in the long-time regime of the environmental correlation function. It is difficult to accurately reproduce the slow $1/\omega$ decay using conventional memory unraveling techniques, such as the Matsubara spectrum decomposition scheme, or the Padé spectrum decomposition (PSD) scheme. This has remained a serious challenge for the HEOM method.

To overcome this problem, a variety of memory unraveling techniques and different types of memory basis functions have been proposed. These include the use of discrete Fourier series, 

Hermite polynomials, 

Chebyshev polynomials, and re-summation over poles. Such developments have pushed the applicability of HEOM to low or even the zero temperature regime. Specifically, most of these developments are time-domain-based schemes, which explicitly treat the correlation function within a finite time window. However, the $1/\omega$ tail is neglected or poorly described by these schemes, and thus, the long-time dynamics and stationary-state properties of the system may not be well preserved. In fact, it has been found that the low-frequency region of the correlation spectrum governs the long-time relaxation toward equilibrium. Therefore, frequency-domain-based schemes are expected to be very useful for the accurate characterization of long-time dissipative dynamics at low temperatures.

Recently, we have proposed an accurate and efficient scheme, the Fano spectrum decomposition (FSD) scheme, for achieving a sum-over-poles (SOP) expansion of the Fermi/Bose function at very low temperatures. With the FSD, the Bose/Fermi function is decomposed into two parts: a high-temperature reference and a low-temperature correction. While the former is associated with a reference high temperature and expanded using the Padé scheme, the latter explicitly includes the abrupt jump at the zero frequency and is accurately approximated using a set of modified Fano functions. The high-order poles of the modified Fano functions result in polynomial-exponential memory basis functions, which decay more slowly than the conventional exponential functions. This makes the FSD particularly suitable for the studies of open quantum systems at low temperatures, as it accurately captures the long-time environmental memory. Since polynomial-exponential functions form a self-closed set, they have been adopted for the construction of HEOM in previous studies.

In this work, FSD-based fermionic/bosonic HEOM formalisms are established for low temperature environments. The accuracy and efficiency of the FSD-based HEOM are demonstrated by examining the real-time dynamics and response properties of a spin-boson model and a single impurity Anderson model.

The remainder of this paper is organized as follows: In Sec. II, we briefly introduce the FSD scheme and present the analytic formalism of FSD-based HEOM for both fermionic and bosonic environments. The numerical performance of the FSD-based HEOM is demonstrated in Sec. III through calculations carried out on a spin-boson model and a single impurity Anderson model at low temperatures. In Sec. IV, we summarize this work.

II. ANALYTIC FORMULATION OF FSD-BASED HEOM

A. Description of system and environment

Let us consider a typical open quantum system setup where a quantum system of primary interest is coupled to a bosonic/fermionic reservoir environment. The total Hamiltonian reads

$$H_{\text{tot}} = H_S + H_{\text{sys}} + H_{\text{env}} = H_S + H_{\text{sys}} + \sum_j \tilde{d}_j \tilde{d}_j^\dagger,$$

Here, $H_S$ is the system Hamiltonian and the environment is modeled by a non-interacting bosonic/fermionic reservoir, $H_{\text{env}} = \sum_j \epsilon_j \tilde{d}_j \tilde{d}_j^\dagger$, with $\tilde{d}_j^\dagger$ ($\tilde{d}_j$) being the creation (annihilation) operator for a particle on the $j$th reservoir state of energy $\epsilon_j$. The operators satisfy $[\tilde{d}_j, \tilde{d}_j^\dagger] = 1$ and $[\tilde{d}_j, \tilde{d}_k^\dagger] = \delta_{jk}$ for bosons/fermions, respectively. We specifically consider the case of a linear system-reservoir coupling through a single dissipation mode. For a boson reservoir, we consider $H_{\text{env}} = \tilde{d}^\dagger \tilde{d}$, where $\tilde{d}$ is a dimensionless system operator and $\tilde{d} = \sum_j \epsilon_j \tilde{d}_j^\dagger$. This is a linear combination of the reservoir coordinate. For a fermionic reservoir, we consider $H_{\text{env}} = \tilde{u}^\dagger \tilde{F} \tilde{F}^\dagger \tilde{u}$, where $\tilde{u}$ is a creation/annihilation operator of a system state and $\tilde{F} = (F^\dagger)^{-1} \sum_j \epsilon_j \tilde{d}_j^\dagger$. Here, $\epsilon_j$ is the coupling strength between the system state and the $j$th reservoir state.

A non-interacting reservoir that couples linearly to the system satisfies Gaussian statistics, and hence, its influence on the system dynamics is completely characterized by the two-time correlation functions $C_\text{B}(t, \tau) \equiv \langle \tilde{B}(t) \tilde{B}(\tau) \rangle_B$ and $C_\text{F}(t, \tau) \equiv \langle \tilde{F}(t) \tilde{F}(\tau) \rangle_F$ for bosonic and fermionic reservoirs, respectively. Here, $\delta \equiv \pm$, $\tilde{d} \equiv -\delta$, $O(t) \equiv e^{\delta \tilde{d} t} O e^{-\delta \tilde{d} t}$, and $\langle \tilde{O} \rangle_B = \langle \tilde{O} \rangle_B(t) = \langle \tilde{O} \rangle_B$, with $\rho_B$ being the density matrix of a bare reservoir. Atomic units are adopted throughout this paper, i.e., $\hbar = c = 1$, and we set $k_B = 1$.

For a bare reservoir at thermal equilibrium, the reservoir correlation functions have translational invariance, i.e., $C_\text{B}(t, \tau) = C_\text{B}(t - \tau)$. They are related to the reservoir spectral functions, $f_B(\omega) = \pi \sum |c|^2 |\delta(\omega - e_j)|$, and $f_F(\omega) = \pi \sum |c|^2 |\delta(\omega - e_j)|$, via the fluctuation–dissipation theorem as follows:

$$C_\text{B}(t - \tau) = \frac{1}{\pi} \int_{-\infty}^{\infty} \text{d} \omega \ e^{-i(\omega - t)} f_B(\omega) f_B(\omega, T),$$

(2a)
The accuracy of the expansion of Eq. (6) is quantified by an error defined by
\[
D_{B/F} = \int_0^\infty \omega \left[ f_{B/F}(\omega, T) - f_{B/F}^{\text{SD}}(\omega, T) \right]^2 d\omega,
\]
where the index \(\sigma\) of the Fermi function has been omitted for brevity. In Ref. 88, we have proposed two sets of PSD parameters \([a_d]\) and \([b_d]\) for the Bose/Fermi function with \(\chi = 100\) and \(\chi = 1000\). In Table 1, we provide a new set of parameters, which yield an even more accurate expansion of the Fermi function, but with a slightly larger \(P_1\). Like the two sets proposed in Ref. 88, the new set can also be adopted directly for any temperature \(T\) without refitting. Since \(\chi\) is fixed for a given parameter set, the reference temperature determined by \(T_0 = \chi T\) varies with \(T\). The user is free to choose a parameter set that suits his/her preference in accuracy or efficiency.

It should be pointed out that in all the existing FSD schemes, there is always an \(a_d\) that is conspicuously larger than the rest of the \([a_d]\) parameters. Numerically, such a large \(a_d\) is crucial for achieving a minimal error \(D_{B/F}\). Physically, it leads to an extraordinarily small \(y_d\), which is essentially important for describing the slowly decaying tail of \(C_{B/F}(t - \tau)\). Moreover, if the reference temperature \(T_0 = \chi T\) is not high enough, we may use \(N\) reference temperatures, \(\{T_i\; i = 0, \ldots, N - 1\}\), with \(T_{i+1} = \chi T_i\), and \(T_N = T\). The low-temperature correction in Eq. (6) can be applied recursively (see Sec. II B of Ref. 88 for details). This, in principle, allows the FSD scheme to access any ultra-low temperature.

In general, the coefficients in Eq. (7) are complex numbers. By definition, the spectral function of a boson reservoir is an antisymmetric function, i.e., \(f_B(\omega) = -f_B(-\omega)\), and hence, the coefficients \([\tilde{h}_d]\) associated with the high-order poles of \(f_B^{\text{SD}}(\omega, T)\) are all real. In contrast, for a fermion reservoir, in principle, \(f_F(\omega)\) can be any non-negative function. Nevertheless, in practice, \(f_F(\omega)\) is often taken as a symmetric function in the studies of quantum impurity systems because the density of states of many bulk metals (such as gold, silver, and copper) is roughly constant around the Fermi energy. If \(f_F(\omega) = f_F(-\omega)\) holds true, the coefficients

Here, \([m_d]\) are non-negative integers, \(P = P_1 + P_0 + P_D\), where \(P_1\), \(P_0\), and \(P_D = \sum_{\sigma = 1}^{D} J_d\) count the memory basis functions originating from \(f_{B/F}(\omega)\), \(f_{B/F}^{\text{SD}}(\omega, T_0)\), and the low-temperature correction of \(f_{B/F}^{\text{SD}}(\omega, T)\), respectively.

### Table 1

<table>
<thead>
<tr>
<th>(d)</th>
<th>(j_d)</th>
<th>(a_d)</th>
<th>(b_d)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1</td>
<td>269,600</td>
<td>-0.779 276</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>28,143 6</td>
<td>-0.643 308</td>
</tr>
<tr>
<td>3</td>
<td>3</td>
<td>2,610 76</td>
<td>-0.815 771</td>
</tr>
<tr>
<td>4</td>
<td>2</td>
<td>0,754 740</td>
<td>-0.766 179</td>
</tr>
<tr>
<td>5</td>
<td>3</td>
<td>6,376 50</td>
<td>-0.800 728</td>
</tr>
</tbody>
</table>

\(D_B = 1.941 \times 10^{-4}\)
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The expression as an integral of the Lagrangian and its time derivative reads

Here, the influence functional, constitutes a generic basis set for the system subspace such that 

Increasing the size of the hierarchy.

will have to further decompose resulting from the FSD scheme still form a closed set.

It is worth noting that if \( \eta_\rho \) is complex, such as when it arises from a high-order pole of \( J_B(\omega) \), the coefficients \( \eta_\rho \) and \( \eta_\rho^* \) cannot be taken out of the square bracket of Eq. (18). In such a case, we will have to further decompose \( B_p \) into two parts, which will greatly increase the size of the hierarchy.

Compared to the conventional way of constructing the HEOM, one only needs to pay attention to the polynomial-exponential term in Eq. (7a) and the related generating functionals. Based on the definition of Eq. (16) and by using Eqs. (11) and (19), we obtain a compact form of the FSD-based bosonic HEOM as follows:

Therefore, Eq. (18) can be recast into

Apparently, the generating functionals \( \{ B_p \} \) resulting from the FSD scheme still form a closed set.

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\[
\hat{\rho}^{(n)}_{\eta_\rho(\cdot)} = -i \sum_{p=1}^{P_B} \sum_{n_{\eta_\rho}} \hat{\rho}^{(n)}_{\eta_\rho(\cdot)} \hat{\rho}^{(n)}_{\eta_\rho(\cdot)} - i \sum_{p=1}^{P_B} \sum_{n_{\eta_\rho}} \hat{\rho}^{(n)}_{\eta_\rho(\cdot)} \hat{\rho}^{(n)}_{\eta_\rho(\cdot)} - i \sum_{p=1}^{P_B} \sum_{n_{\eta_\rho}} \hat{\rho}^{(n)}_{\eta_\rho(\cdot)} \hat{\rho}^{(n)}_{\eta_\rho(\cdot)}
\]

\[
\hat{\rho}^{(n)}_{\eta_\rho(\cdot)} = \left[ H_{\hat{\rho}}, \hat{O} \right].
\]

D. FSD-based fermionic HEOM

The FSD-based fermionic HEOM can be constructed in a similar manner. Let us start with the fermionic path integral formalism,
\[ \mathcal{U}(t, t_0) = \int D\psi D\psi' D\psi'' D\psi''' e^{i S_0[\psi, \psi']} F_{\mathcal{F}}[\psi, \psi'; \psi, \psi''; \psi', \psi'''], \]

Here, \( \psi \equiv \{ \psi_i \} \), \( \psi' \equiv \{ \psi_i' \} \), and \( \psi'' \equiv \{ \psi_i'' \} \) with 0 \( \leq \tau \leq t \) are the fermionic coherent states in the system subspace; and \( \psi \) and \( \psi' \) are Grassmann numbers (g-number) that are associated with \( \hat{a}^* \) and \( \hat{a} \), respectively, which satisfy the relation \( \psi \psi = -\psi \psi' \).

The path integral of Eq. (21) accounts for all the possible paths that connect the initial state at \( t_0 \), \( \psi(t_0) = \psi_0^* \) and \( \psi(t) = \psi_0 \), to the final state at \( t \), \( \psi(t) = \psi_i \) and \( \psi'(t) = \psi_i' \). \( S_f \) and \( S_b \) are the forward and backward action functionals of the bare system, which are expressed as

\[ S_f = \int_{t_0}^{t} dt \left[ i \dot{\psi}_{\tau} \psi_{\tau} - H_S(\psi_{\tau}, \dot{\psi}_{\tau}) \right] - i \psi_{\tau} \psi_{\tau}, \]

\[ S_b = \int_{t_0}^{t} dt \left[ -i \dot{\psi}'_{\tau} \psi'_{\tau} - H_S(\psi'_{\tau}, \dot{\psi}'_{\tau}) \right] + i \psi'_{\tau} \psi'_{\tau}, \]

and their time derivatives are

\[ \partial_\tau S_f = -H_S(\psi_{\tau}, \dot{\psi}_{\tau}), \]

\[ \partial_\tau S_b = -H_S(\psi'_{\tau}, \dot{\psi}'_{\tau}). \]

If the fermionic reservoir satisfies Gaussian statistics, its influence on the reduced system dynamics is fully characterized by the influence functional, \( F_{\mathcal{F}}[\psi, \psi'; \psi, \psi''] = e^{-\Phi_F[\psi, \psi', \psi]}, \) where the fermionic dissipation functional \( \Phi_F \) has the form of

\[ \Phi_F = \int_{t_0}^{t} dt \left[ \phi_{\tau} B_f^*(\tau) + \phi_{\tau} B^*(\tau) \right]. \]

Here, \( \phi_{\tau} = \psi_{\tau} + \psi_{\tau}' \), \( \phi_{\tau}' = \psi_{\tau} + \psi_{\tau}' \), and the generating functionals are expressed as

\[ \Phi_F^{(m, \cdots, m-1)} = \left\{ \left( i \mathcal{L}_S + \sum_{i=1}^{p} \sum_{j=1}^{i} \overline{y}_{i j} + \sum_{i=1}^{q} \overline{y}_i^* \right) \rho^{(m, \cdots, m-1)} \right\} \]

\[ - i \sum_{i=1}^{p} \delta_{m_0} \left[ \left( -1 \right)^{1+i} \rho^{(m, \cdots, m-1)} \hat{a} - \left( -1 \right)^{1+i} \rho^{(m, \cdots, m-1)} \hat{a}^* \right] \]

\[ - i \sum_{i=1}^{q} \delta_{m_0} \left[ \left( -1 \right)^{1+i} \rho^{(m, \cdots, m-1)} \hat{a} - \left( -1 \right)^{1+i} \rho^{(m, \cdots, m-1)} \hat{a}^* \right] \]

\[ + \sum_{i=1}^{m_0} \left( \rho^{(m, \cdots, m-1)} \rho^{(m, \cdots, m-1)} \right) \rho^{(m, \cdots, m-1)} \rho^{(m, \cdots, m-1)} \rho^{(m, \cdots, m-1)} \rho^{(m, \cdots, m-1)} \rho^{(m, \cdots, m-1)} \rho^{(m, \cdots, m-1)} \rho^{(m, \cdots, m-1)} \rho^{(m, \cdots, m-1)} . \]

If the fermionic reservoir spectral function is not an even function of \( \omega \), the equality \( \eta^*_p / \eta_{p-1} = (\eta^*_p / \eta_{p-1})^* \) no longer holds. In such cases, \( B_p^* \) must be further decomposed into two terms (forward and backward propagation terms) to form a closed set of generating functionals, and the resulting HEOM are more complex than Eq. (28). Extension of Eq. (28) to a multi-dissipation-mode and/or multi-reservoir model is straightforward.
III. NUMERICAL APPLICATIONS OF FSD-BASED HEOM TO OPEN QUANTUM SYSTEMS AT LOW TEMPERATURES

A. Quantum dynamics of a spin-boson model at low temperatures

To demonstrate the numerical applicability of the FSD-based HEOM approach, we first study the quantum dynamics of a spin-boson model under low temperatures. The total Hamiltonian of the spin-boson model is

$$ H = \Delta_x \sigma_z + \left( \frac{\alpha_z}{2} \right) \sum_j \epsilon_j n_j + \sum_j \left( \frac{\omega_j}{2} \right) \left( n_j^{\dagger} n_j + \sigma_z^{\dagger} \sigma_z \right), $$

where the operators $\sigma_z$ and $\sigma^z_+$ are represented by the Pauli matrices and $\Delta_x$ is the electronic coupling between the spin-up and spin-down states.

The relaxation of the spin system is driven by the longitudinal fluctuations in the boson reservoir, which is described by an Ohmic spectral function,

$$ j_B(\omega) = \frac{2\pi \alpha \zeta(\omega/W)}. \tag{29} $$

Here, $\alpha$ is the Kondo parameter that determines the system-reservoir dissipation strength, $\zeta(\omega/W)$ is a function that effectively cuts off the high-frequency region, with $W$ being the cutoff frequency.

We consider two different types of window functions, which correspond to the Drude and super-Drude models, respectively,

$$ f_{B_{\text{Drude}}}^{\text{Drude}}(\omega) = \frac{2\pi \alpha}{1 + (\omega/W)^2}, \tag{30a} $$

$$ f_{B_{\text{Sup}}}^{\text{Sup}}(\omega) = \frac{2\pi \alpha}{1 + (\omega/W)^2}. \tag{30b} $$

Note that $z = -iW$ is a first-order pole of $f_{B_{\text{Drude}}}^{\text{Drude}}(z)$, but a second-order pole of $f_{B_{\text{Sup}}}^{\text{Sup}}(z)$. In the latter case, the second-order pole contributes a polynomial-exponential term, $e^{-\pi \alpha \omega} \pi \alpha \omega / \omega$ to $C_{\text{up}}(t)$.

Figure 1 compares $C_{\text{Drude}}(t)$ and $C_{\text{sup}}(t)$ calculated using the FSD and PSD schemes with the exact values. Apparently, for the ultra-low temperature examined, to achieve the same level of accuracy, the FSD scheme requests much fewer memory basis functions than the PSD scheme. It is also demonstrated that the reservoir correlation functions of the Drude and super-Drude models with the same parameters coincide with each other in the low-frequency or the long-time regime.

When the HEOM method is applied to study the spin-boson model, the cost of computer memory is normally dictated by the number of ADOs, which is largely determined by $P$. Therefore, by replacing the conventional PSD scheme with the FSD scheme, the saving in memory cost is estimated to be $1/(P_{\text{FSD}}/P_{\text{PSD}})^2$, with $L$ being the truncation tier of the hierarchy. For instance, for the Drude model examined in Fig. 1, $P_{\text{PSD}} = 12$ and $P_{\text{FSD}} = 81$ yield similar $C_{\text{Drude}}(t)$. Thus, the memory cost of the FSD-based HEOM truncated at $L = 10$ is only $(12/81)^{10} \approx 5 \times 10^{-9}$ of that of the PSD-based HEOM. Such a drastic reduction in memory cost makes it possible to access the ultra-low temperature regime that was prohibitive to the HEOM method.

Figure 2 depicts the quantum relaxation dynamics of a spin-boson system from a decoupled initial state to the final equilibrium state calculated using the FSD-based HEOM method. Both the Drude and super-Drude reservoir models examined in Fig. 1 are adopted, and a number of dissipation strengths ranging from $\alpha = 0.1$ to $\alpha = 0.8$ are scrutinized. The relaxation dynamics exhibits distinctly different behavior in the weak ($\alpha < 0.5$) and strong ($\alpha > 0.5$) dissipation regimes, where the reduced system dynamics is apparently coherent and incoherent, respectively. Moreover, despite the conspicuous difference in the high-frequency (or short-time) reservoir correlation function, the Drude and super-Drude models yield similar relaxation dynamics toward equilibrium. This highlights the significance of an accurate characterization of the low-frequency (or long-time) components of the reservoir memory.

All the calculations shown in Fig. 2 are performed on a desktop computer with an Intel Core i7-8700K processor and a GeForce GTX 1080 graphics card. For the most challenging case with a super-Drude model and $\alpha = 0.8$, we choose $P_{\text{PSD}} = 15$ and $L = 10$, and the total number of ADOs is 3 268 760. For stronger couplings ($\alpha > 0.8$), the truncation at $L = 10$ becomes inadequate, while a higher truncation tier would cost too much computer memory that is beyond the resources at our disposal. To ensure a high accuracy, the dynamical filtering algorithm is turned off. By using a standard fourth-order Runge–Kutta integrator with a time step of 0.01$\Delta_\tau$, the propagation of the FSD-based HEOM for $N_t = 3000$ time steps costs only about half an hour with the graphics processing unit (GPU)-accelerated implementation, vs about 2–3 days in the sequential computing mode, resulting in a speedup of more than 100 times.

Since the temperature adopted in the calculation ($T = 0.001\Delta_e$) is already quite low, further lowering of the temperature will not
lead to appreciable changes in the relaxation dynamics. Therefore, the numerical data presented in Fig. 2 can be compared directly with the previous studies for the zero temperature.  

As shown clearly in the lower panel of Fig. 2, the numerical results of the FSD-based HEOM method agree closely with those obtained previously by a mixed random-deterministic approach in the weak dissipation regime ($\alpha < 0.5$) and by an extended HEOM approach in the strong dissipation regime ($\alpha > 0.5$).  

As shown in Fig. 2, some existing approaches such as those proposed in Refs. 75 and 83 are capable of addressing the zero-temperature dissipative dynamics of the spin-boson model. However, if the system includes dissipation channels of ultra-low energies, such as Kondo states in fermionic quantum impurity systems (see Subsections III B and III C), the existing approaches are expected to be less accurate because the ultra-low-energy components of the relaxation dynamics may not have been explicitly accounted for in the existing approaches. Instead, the FSD-based HEOM approach proposed in this work is expected to be more accurate.

**B. Voltage-driven dynamics of a single-orbital Anderson impurity model at low temperatures**

While the relaxation dynamics of a spin-boson model does not exhibit an obvious temperature dependence in the ultra-low temperature regime, the quantum dynamics of a fermionic quantum impurity model involving low-energy excitations can be significantly affected by the temperature.

We consider a quantum impurity system described by the single-orbital Anderson impurity model (AIM). The quantum impurity of primary interest is described by $H_S = \epsilon_S (\hat{n}_1 + \hat{n}_{\uparrow 1}) + U \hat{n}_1 \hat{n}_{\uparrow 1}$ with $\hat{n}_1 = \hat{a}_{\downarrow 1}^\dagger \hat{a}_{\downarrow 1}$ ($s = \uparrow, \downarrow$). Here, $\hat{a}_{\uparrow 1}$ (or $\hat{a}_{\downarrow 1}$) creates (annihilates) a spin-$s$ electron on the impurity orbital of energy $\epsilon_S$, and $U$ is the electron–electron Coulomb interaction energy. The impurity is coupled to two non-interacting electron reservoirs ($\alpha = L, R$), which are described by $H_E^\alpha = \sum_{\alpha k s} (\epsilon_{\alpha k} - V_0(t)) \hat{a}_{\alpha k s}^\dagger \hat{a}_{\alpha k s}$. The applied voltage causes a time-dependent shift in the reservoir chemical potential, i.e., $\mu_\alpha(t) = \mu_\alpha^{eq} - V_0(t)$. The impurity-reservoir coupling is $H_{SR}^\alpha = \sum_{\alpha k s} \hat{a}_{\alpha k s}^\dagger F_{\alpha s} + F_{\alpha s} \hat{a}_{\alpha k s}^\dagger$, with $F_{\alpha s} = (\hat{F}_{\alpha s})^\dagger = \sum_k c_{\alpha k s}^\dagger c_{\alpha k s}$.

FIG. 2. Quantum relaxation dynamics of a spin-boson system subject to various dissipation strengths (ranging from $\alpha = 0.1$ to 0.8) at a low temperature $T = 0.001$. The vertical axis is $\langle \hat{S}_\uparrow (t) \rangle = \text{tr}_\alpha \{ \rho (t) \hat{S}_\uparrow \}$. Both the Drude (top) and super-Drude (bottom) reservoirs with a large cutoff frequency $W = 10$ are examined. The system initial reduced density matrix is set to be $\rho(0) = (\hat{S}_\uparrow + 1)/2$. All energies are in units of $\Delta_0$. The solid lines are the results of the FSD-based HEOM method for $T = 0.001$, while the scattered data of $\alpha < 0.5$ are the zero-temperature results of a mixed random-deterministic approach extracted manually from Ref. 75; the scattered data of $\alpha > 0.5$ are the zero-temperature results of an extended HEOM approach extracted manually from Ref. 83.

It has been revealed in our previous work that when the strongly correlated Kondo states emerge in the AIM at a low temperature, the dynamic $I$–$V$ characteristics exhibit prominent memory effects, which are manifested through the hysteresis line shapes and self-crossing features. We now re-examine such a dynamic process at an even lower temperature. All the calculations are performed by using the HEOM-QUICK program, which fully utilizes the sparsity feature of the ADOs.

Initially, the total AIM system is in a global equilibrium state with $\mu_\alpha = \mu_\alpha^{eq} = 0$. An ac voltage is applied to the left and right reservoirs from the time $t_0 = 0$, $V_0(t) = -V_0 \sin(\omega_0 t)$, with $V_0$ and $\omega_0$ being the amplitude and frequency of the voltage, respectively. Driven by the ac voltage, the impurity undergoes a non-equilibrium dynamics, and the time-dependent electric current flowing into the $a$th reservoir, $I_a(t)$, is computed by propagating the FSD-based HEOM and analyzing the first-tier ADOs.

Figure 3(a) depicts the voltage-driven electric current calculated by using three sets of parameters for the FSD. Among them, the two parameter sets with $(\chi = 100, P_D = 6)$ and $(\chi = 1000, P_D = 9)$ were proposed in Ref. 88, and a more accurate set of parameters with $(\chi = 1000, P_D = 10)$ is given in Table I of this work. The time-dependent currents resulted from the three schemes agree closely with each other, as long as the numerical results are convergent. On the other hand, it is noted that the calculated result starts to diverge after a certain time ($t > t_i$). This is the notorious asymmetric instability problem that has been discovered recently by Dunn et al. In the case of Fig. 3(a), such an instability clearly originates from the residual error associated with the SOP expansion of Eq. (6b) for the Fermi function (especially for the low-frequency region). This is affirmed by the fact that the numerical divergence occurs at a later time ($a$ larger $t_i$) with a more accurate FSD scheme (a smaller $D_o$).

The dynamic $I$–$V$ characteristics displayed in Fig. 3(b) exhibit a hysteresis behavior, with more self-crossing loops showing up than those observed previously. The emergence of the multiple loops originates from the Kondo resonance enhanced at a lower temperature. The spin-flip excitation energy associated with the Kondo resonance is much smaller than the driving frequency $\omega_0$, and thus, the spin-flip spin current shows a larger self-crossing features. This is affirmed by the fact that the numerical divergence occurs at a later time ($a$ larger $t_i$) with a more accurate FSD scheme (a smaller $D_o$).
FIG. 3. (a) Time-dependent electric current from the impurity into the left reservoir, $I_L(t)$, driven by the ac voltage $V_L(t) = -V_R(t) = V_0 \sin(\omega_0 t)$ and (b) the corresponding dynamic $I$–$V$ characteristics (only the converged data are displayed). The inset of (b) magnifies the self-crossing loops near $V_L = -V_0$. The arrows in (a) mark the time $t_l$ from which the calculations start to diverge. The parameters of the AIM and the applied voltage are (in units of $\Delta$) $U = 12$, $\Delta_L = \Delta_R = 0.5$, $T = 0.05$, $W = 20$, $eV_0 = 1.5$, and $\omega_0 = 0.3$. Three sets of parameters are adopted for the FSD, among which the newly proposed set (marked by the star in the legend) with $\chi = 1000$ and $P_D = 10$ is most accurate (see Table I). The FSD-based HEOM are truncated at the tier $L = 4$.

In Figs. 4(a) and 4(b), we explore the dynamic $I$–$V$ characteristics of a single-orbital impurity with its single- and double-occupation states in resonance with the reservoir chemical potential, respectively. In these two scenarios, the Kondo resonance is greatly suppressed by charge fluctuation and has little influence. The excitation energy for changing the occupation number of impurity is as large as $2\omega_0$. Such an energy is too high to be accessed by the external driving source, and thus, it is hardly manifested in the response current. Consequently, the time-dependent current is almost synchronized with the driving voltage, without prominent signatures of the overtones. Moreover, by comparing Figs. 4(c) and 4(d), it is found that for a Kondo impurity, the number of self-crossing loops decreases with the increase in $\omega_0$. This is because the ac voltage suppresses the Kondo resonance through decoherence of the impurity spin, and the decoherence rate increases with the increasing driving frequency.\(^{20}\)

Figure 5 examines the convergence of the calculated time-dependent current with respect to the truncation tier $L$. For the voltage-driven AIM studied in Fig. 3, the calculation with $L = 5$ diverges shortly after the initial time because of the aforementioned numerical instability problem. Nevertheless, the $L = 5$ data available in the transient regime agree closely with the $L = 4$ counterpart. This
suggests that the \( L = 4 \) truncation is sufficient to yield quantitatively accurate dissipative dynamics for the investigated AIM.

Regarding the saving of computer memory by employing the FSD, the standard PSD scheme would require \( P^{\text{PSD}} = 70 \) to achieve the same level of accuracy for unraveling the reservoir memory. Therefore, the memory cost of the FSD-based HEOM is only \( (P^{\text{PSD}}/P^{\text{FSD}})^L = (12/71)^4 \approx 8 \times 10^{-6} \) of that of the PSD-based HEOM.

C. Kondo characteristics in stationary-state single-orbital AIM at low temperatures

We now apply the FSD-based HEOM method to quantitatively characterize the strength of the Kondo correlation in stationary-state single-orbital AIMs at low temperatures.

The strength of the Kondo correlation is usually characterized by the height of the Kondo peak residing in the low-frequency region of the impurity spectral function, \( A_i(\omega) \equiv \frac{1}{N} \int_0^\infty d\epsilon \rho_{\epsilon i}(\epsilon, t) \delta(\epsilon - \omega) \), which is evaluated by employing the HEOM-space linear response theory. \(^{101,102}\) Here, to access the unprecedentedly low temperature of \( T = 0.01\Delta \), the FSD scheme of Ref. 88 with \( \chi = 1000 \) and \( P_D = 9 \) is adopted. It is found that while the quasi-particle peaks at around \( \omega = \epsilon_d + \omega = U + \epsilon_d \) hardly change with temperature, the Kondo peak centered at \( \omega = 0 \) is drastically enhanced with the lowering of temperature. This clearly indicates the strengthened Kondo correlation at an ultra-low temperature. From the inset of Fig. 6, it is clear that the calculated \( A_i(\omega) \) converge quantitatively at \( L = 5 \). Moreover, the normalization condition \( \int_0^\infty d\omega A_i(\omega) = 1 \) is always satisfied with a high precision, which also validates the numerical results of the FSD-based HEOM method.

\[ A_i(0) = \frac{\sin^2(\pi k)}{\pi \Delta}. \]  

Such a relation should hold exactly at zero temperature.

We examine the relation of Eq. (31) for an AIM with the electron-hole symmetry \( (\epsilon_d = -U/2) \). The calculated \( A_i(0) \) vs the scaled temperature \( T/T_K \) is shown in Fig. 7, where the Kondo temperature \( T_K \) is a characteristic energy scale of the Kondo correlation. For a symmetric AIM, \( T_K = \sqrt{\frac{U^2}{2}} \exp \left( \frac{-2\epsilon_d/\Delta}{\ln(1 + \frac{\Delta}{U})} \right) \) and the impurity is exactly half filled \( (n_i = 1/2) \). As demonstrated in Fig. 7, \( A_i(0) \) correctly approaches the value of \( 1/(\pi \Delta) \) with the decreasing \( T \). This thus verifies that the Friedel sum rule of Eq. (31) is accurately preserved by the FSD-based HEOM method.

For an AIM in the Kondo regime \( (T < T_K) \), the relation between the zero-bias conductance \( G = (dI/dV)|_{V=0} \) and the scaled temperature \( T/T_K \) is generally described by the empirical Goldhaber–Gordon formula \(^{105,106}\) as follows:

\[ \left( \frac{G}{G_0} \right)^{-1/\nu_c} = 1 + \left( 2^{1/\nu_c} - 1 \right) \left( \frac{T}{T_K} \right)^2, \]  

where \( G_0 = 2 \times 10^7 \text{h} \) is the conductance quantum and \( \nu_c = 0.22 \) is a universal scaling constant. \(^{105-106}\) It is important to note that Eq. (32) predicts \( G(T = 0) = G_0 \) in the zero-temperature limit.

We calculate \( G \) vs \( T \) for a series of symmetric AIMs with the FSD-based HEOM method. The differential conductance \( dI/dV \) is evaluated by using the finite difference method. The parameters for \( \chi = 1000 \) and \( P_D = 9 \) are adopted, and the resulting HEOM are truncated at the tier \( L = 5 \). The calculated data are fit to Eq. (32), and the value of \( \nu_c \) is optimized by scanning the region \((0.15, 0.25)\) until the best fit is achieved. \(^{106}\)

Figure 8 depicts the \( (G/G_0)^{-1/\nu_c} vs (T/T_K)^2 \) for three symmetric AIMs. Apparently, all the calculated data are distributed tightly around a straight line, which validates the universal scaling relation of Eq. (32). Besides, \( \nu_c \) is determined to be 0.221, agreeing
well with the value reported in the literature. Although it is still impractical to compute $G(T = 0)$ directly with the FSD-based HEOM, the zero-temperature limit can be accessed by extrapolating the calculated data of finite temperatures. The extrapolation in Fig. 8 yields $G(T = 0)/G_0 = 0.991$, which is very close to the exact value of 1. Such an extrapolation is much improved as compared to our previous work, which uses the data calculated using the FSD-based HEOM. The substantial improvement is certainly attributed to the FSD scheme, which allows a much lower temperature to be accessed explicitly by the HEOM calculation, and hence enables a more accurate extrapolation.

**IV. CONCLUDING REMARKS**

In summary, we have constructed the FSD-based HEOM formalisms for both bosonic and fermionic environments, and their final forms are given by Eqs. (20) and (28), respectively. With the FSD scheme, the reservoir correlation functions are accurately expanded by a small number of polynomial-exponential functions, and thus, the size of the memory basis set is substantially reduced, especially in the ultra-low temperature regime. In particular, in this paper, we present a new set of FSD parameters (see Table I), which reproduces the exact Fermi function more accurately than those given in our previous work. Such a new parameter set can be adopted directly for any temperature without refitting.

As demonstrated in Sec. III, the FSD scheme indeed allows the HEOM method to access unprecedentedly low temperatures and yield highly accurate results. In particular, the FSD-based HEOM method promises a high accuracy for the long-time dissipative dynamics as well as the stationary-state properties, provided that the preset truncation tier is sufficiently high.

Recall that the hierarchy extends in both horizontal and vertical directions. Given that the FSD scheme drastically reduces the width of the hierarchy, it is then the height of the hierarchy (the minimal truncation tier $L$) that presents the main bottleneck for the applicability of HEOM. For instance, as shown in Fig. 8, the calculated data (scattered points) start to deviate from the universal scaling relation (solid line) at $(T/T_K)^2 < 0.01$. This is because the preset truncation tier $L = 5$ is probably not enough for such low temperatures. Therefore, one of the next challenges of HEOM is to access the higher-tier in a more efficient manner.

In this work, we have also encountered the asymptotic instability problem, which has come to the attention of many researchers (see Fig. 3). In our case, the instability is alleviated by employing a more accurate FSD scheme. Therefore, it is always desirable to further improve the parameters for FSD. Work along this direction is underway.

**AUTHOR’S CONTRIBUTIONS**

H.-D.Z. and L.C. contributed equally to this work.

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**REFERENCES**
