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Onsets of hierarchy truncation and self-consistent Born approximation with quantum mechanics prescriptions invariance

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The issue of efficient hierarchy truncation is related to many approximate theories. In this paper, we revisit this issue from both the numerical efficiency and quantum mechanics prescription invariance aspects. The latter requires that the truncation approximation made in Schrödinger picture, such as the quantum master equations and their self-consistent-Born-approximation improvements, should be transferable to their Heisenberg-picture correspondences, without further approximations. We address this issue with the dissipaton equation of motion (DEOM), which is a unique theory for the dynamics of not only reduced systems but also hybrid bath environments. We also highlight the DEOM theory is not only about how its dynamical variables evolve in time, but also the underlying dissipaton algebra. We demonstrate this unique feature of DEOM with model systems and report some intriguing nonlinear Fano interferences characteristics that are experimentally measurable. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4936831>]

I. PRELUDE

Interference between system and environment plays a pivotal role in many fields of science nowadays. It manifests in such as Fano interference,^{1,2} surface plasmonic enhancement spectroscopy,³ and coherent excitation energy transfer in nanostructured biological pigment-protein complexes.^{4–7} The dissipaton equation of motion (DEOM) approach^{8–10} is a universal theoretical tool in these studies. As an exact and non-perturbative hierarchy-type method, it is capable of investigating the central system and the coupled bath simultaneously.

This paper is mainly concerned with the issue of the hierarchy truncation, to be addressed from not just practical aspect, but more importantly the fundamental quantum mechanics principles. This issue is rather common, involving in various approximate theories, as long as they are rooted at an exact but infinity hierarchy-type of theory. Examples are such as quantum master equations and their self-consistent-Born-approximation improvements.^{11–15} These theories are widely adopted in scientific community.

To be general, we consider the DEOM formalism.^{8–10} This theory goes with a quasi-particle (dissipaton) picture for the participating bath environment and the underlying dissipaton algebra.⁸ These unique features enable DEOM a theory for not only reduced system but also hybrid bath dynamics explicitly (cf. Sec. V for the demonstrations). On the other hand, as the reduced system dynamics or the hierarchy level truncation is concerned, DEOM is identical to the hierarchy equations of motion (HEOMs) formalism.^{16–24} Their common generic

form reads

$$\dot{\rho}_{\mathbf{n}}^{(n)} = -(i\mathcal{L}_S + \gamma_{\mathbf{n}})\rho_{\mathbf{n}}^{(n)} - \{\rho_{\mathbf{n}^-}^{(n-1)}\} - \{\rho_{\mathbf{n}^+}^{(n+1)}\}. \quad (1)$$

It involves an infinity hierarchy that needs to be truncated at a certain $n_{\text{trun}} = L$ level. While $\{\rho_{\mathbf{n}}^{(n>0)}\}$ in HEOM are mathematical auxiliary operators, the DEOM theory identifies them the dissipaton density operators (DDOs),^{8–10}

$$\rho_{\mathbf{n}}^{(n)}(t) \equiv \text{tr}_B \left[\left(\prod_{ak} \hat{f}_{ak}^{n_{ak}} \right)^\circ \rho_T(t) \right]. \quad (2)$$

Here, $\{\hat{f}_{ak}\}$ arise from the so-called dissipatons decomposition of the hybridization bath operators $\{\hat{F}_a^B\}$, involving the system-bath coupling Hamiltonian. That is

$$H_{SB} = \sum_a \hat{Q}_a^S \hat{F}_a^B \equiv \sum_a \sum_{k=1}^K \hat{Q}_a^S \hat{f}_{ak}. \quad (3)$$

The dissipatons decomposition algorithm follows two rules:^{8–10} (i) It preserves the bare bath statistics on $\langle \hat{F}_a^B(t) \hat{F}_a^B(0) \rangle_B$; (ii) The resulting $\langle \hat{f}_{ak}(t) \hat{f}_{bj}(0) \rangle_B \propto \delta_{kj} e^{-\gamma_k t}$, and the reverse $\langle \hat{f}_{bj}(0) \hat{f}_{ak}(t) \rangle_B$ go with the same exponent γ_k . In Eq. (2), the dissipaton operators product within the circled parenthesis is *irreducible*. The notation here follows $(c\text{-number})^\circ = 0$, and all bosonic dissipatons are permutable, $(\hat{f}_{ak} \hat{f}_{bj})^\circ = (\hat{f}_{bj} \hat{f}_{ak})^\circ$. The DDO $\rho_{\mathbf{n}}^{(n)}$ of Eq. (2), where $n = \sum_{ak} n_{ak}$, specifies a configuration of n -body dissipatons participating in the DEOM dynamics. Apparently, $\rho^{(0)}(t) = \text{tr}_B \rho_T(t) = \rho_S(t)$, the reduced system operator. It is worth to point out that the irreducibility notation $(\cdots)^\circ$ in Eq. (2) highlights the non-perturbative nature of DEOM method. Moreover, it goes with the generalized Wick's theorem^{8–10} [cf. Eq. (29)] that plays crucial roles in the DEOM-based evaluation of hybrid bath dynamics, as demonstrated later in this paper.

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The need for efficient hierarchy level truncation methods may be understood as follow. The number of n -body configurations $\{\rho_n^{(n)}\}$ is $\frac{(n+N_a K-1)!}{n!(N_a K-1)!}$, with N_a being the number of system-bath coupling modes in the first expression of Eq. (3). While the K -space is concerned with a minimum basis-set dissipatons for the DEOM construction,^{25,26} optimal formalism should also go with an efficient L -space truncation. The number of $\{\rho_n^{(L+1)}\}$ is often comparable to or even larger than that of $\{\rho_n^{(n \leq L)}\}$ in total. Consider, for example, the biological Fenna-Mathews-Olson exciton model of $N_a = 8$ coupling exciton modes.²⁷⁻²⁹ Assume one dissipaton ($K = 1$) for each exciton. The number of $\{\rho_n^{(n \leq 4)}\}$ is 495 in total, but that of $\{\rho_n^{(n=5)}\}$ alone is 792. Thus, it would be practically important to have a truly improved resum scheme to retrieve the influence of $\{\rho_n^{(L+1)}\}$ back to the final DEOM dynamics.

Existing truncation schemes by far are all proposed from the practical or numerical aspect. Often the standard (also the simplest) scheme that sets all $\{\rho_n^{(n > L)} = 0\}$ would be sufficient,^{20-24,30-32} if the system is not too big and/or the temperature is too low. This simplest scheme is also numerically reliable, due to the non-perturbative nature of the DEOM/HEOM formalism.^{20,24} As inferred from the DDOs expression, Eq. (2), the standard scheme treats all L -body dissipatons exactly. The higher level many-body effects would have been accounted for via a certain Wick's contraction type of meanfield approximation.⁸ This fact may explain why other seemingly sophisticated schemes^{21,22,33} show only little improvement over the simplest one. Currently the quest of efficient truncation scheme would just be an improvement by one higher tier level than the simplest/standard. Another aspect about truncation, which is largely missing in the literature, is about the prescription invariance requirement of quantum mechanics. It demands a proper truncation scheme in the Schrödinger picture be transferable to the equivalence in Heisenberg prescription,^{6,34-36} without further approximations.

In this work, we address the hierarchy level truncation from both aspects described above. We revisit the so-called the derivative-resum scheme,³⁷ and compare it closely to its classical version proposed originally³³ and used extensively by Tanimura and coworkers.³⁸⁻⁴¹ In Sec. II, we review the DEOM/HEOM formalism in Schrödinger picture, followed by its Heisenberg prescription equivalence.^{6,34,35} We then present the derivative-resum truncation, which preserves the prescription invariance. In Sec. III, we generalize the high-order self-consistent Born approximation, with the aid of the continued fraction Green's function approach.^{20,42,43} In Sec. IV, we compare the derivative-resum truncation with other existing schemes. In Sec. V, we exemplify the DEOM-based study in nonlinear Fano interference polarization in a model system. We summarize this paper in Sec. VI.

II. DEOM TRUNCATION WITH PRESCRIPTION INVARIANCE

A. DEOM formalism in Schrödinger and Heisenberg picture

To address the prescription invariance, we can focus on the single dissipaton case, i.e., $\hat{F}^B = \hat{f}$ or $H_{SB} = \hat{Q}^S \hat{f}$, without

lose of generality, with

$$\langle \hat{f}(t) \hat{f}(0) \rangle_B = \eta e^{-\gamma t}. \quad (4)$$

Its time-reverse counterpart is $\langle \hat{f}(0) \hat{f}(t) \rangle_B = \eta^* e^{-\gamma^* t}$. The DEOM formalism is readily constructed as^{8,16-21}

$$\dot{\rho}^{(n)} = -(i\mathcal{L}_S + n\gamma)\rho^{(n)} - i n C \rho^{(n-1)} - i \mathcal{A} \rho^{(n+1)}, \quad (5)$$

where $\mathcal{L}_S$ is the system Liouvillian,

$$C\hat{O} \equiv \eta \hat{Q}^S \hat{O} - \eta^* \hat{O} \hat{Q}^S \quad \text{and} \quad \mathcal{A}\hat{O} \equiv [\hat{Q}^S, \hat{O}]. \quad (6)$$

Equation (5) can be formally written as

$$\dot{\rho}(t) = -i\mathcal{L}\rho(t). \quad (7)$$

This is the Schrödinger picture, with the DEOM-space dynamics generator $\mathcal{L}$ governing the time evolution of $\rho(t) \equiv \{\rho(t), \rho^{(n>0)}(t)\}$. The Heisenberg picture uses the same generator but for the evolution of DEOM-space dynamical variables,^{6,34-36}

$$\dot{\hat{A}}(t) = -i\hat{A}(t)\mathcal{L}. \quad (8)$$

Here, $\hat{A} \equiv \{\hat{A}^{(n)}\}$ denotes the DEOM-space extension of a conventional dynamical variable $\hat{A}$. For the bare local system operator $\hat{A}_S$, we have $\hat{A}_S \equiv \{\hat{A}_S, \hat{A}_S^{(n>0)} = 0\}$. For nonlocal $\hat{A}$ involving also hybrid bath degree of freedom, the corresponding $\{\hat{A}^{(n)}\}$ can also be readily determined with the DEOM/HEOM-space algebra, so that^{6,34,35}

$$\text{Tr}(\hat{A}\rho_T) \equiv \langle \langle \hat{A} | \rho \rangle \rangle \equiv \sum_{n=0}^{\infty} \langle \langle \hat{A}^{(n)} | \rho^{(n)} \rangle \rangle \equiv \sum_{n=0}^{\infty} \text{tr}_S[\hat{A}^{(n)} \rho^{(n)}].$$

This validates the initial conditions to DEOM (8) in the Heisenberg picture. In analogy to $\text{Tr}[\hat{A}(t)\rho_T] = \text{Tr}[\hat{A}\dot{\rho}(t)]$, the prescription invariance reads in the DEOM-space as

$$\langle \langle \dot{\hat{A}}(t) | \rho \rangle \rangle = \langle \langle \hat{A} | \dot{\rho}(t) \rangle \rangle. \quad (9)$$

It together with Eq. (5) lead to

$$\begin{aligned} \langle \langle \dot{\hat{A}} | \rho \rangle \rangle &= \sum_n \langle \langle \dot{\hat{A}}^{(n)} | \rho^{(n)} \rangle \rangle \\ &= \sum_n \left\{ -\langle \langle \hat{A}^{(n)} | i\mathcal{L}_S + n\gamma | \rho^{(n)} \rangle \rangle \right. \\ &\quad \left. - i n \langle \langle \hat{A}^{(n)} | C | \rho^{(n-1)} \rangle \rangle \right. \\ &\quad \left. - i \langle \langle \hat{A}^{(n)} | \mathcal{A} | \rho^{(n+1)} \rangle \rangle \right\}. \end{aligned} \quad (10)$$

By fixing the ket $|\rho^{(n)}\rangle$ in the second expression, we immediately obtain the DEOM formalism in the Heisenberg picture,^{6,34,35}

$$\begin{aligned} \dot{\hat{A}}^{(n)} &= -\hat{A}^{(n)}(i\mathcal{L}_S + n\gamma) - i(n+1)\hat{A}^{(n+1)}C \\ &\quad - i\hat{A}^{(n-1)}\mathcal{A}. \end{aligned} \quad (11)$$

The superoperator actions in Heisenberg picture follow Eq. (6) but read

$$\hat{O}C \equiv \eta \hat{Q}^S \hat{O} - \eta^* \hat{O} \hat{Q}^S \quad \text{and} \quad \hat{O}\mathcal{A} \equiv [\hat{O}, \hat{Q}^S]. \quad (12)$$

B. Derivative-resum truncation with prescription invariance

DEOM in Schrödinger and Heisenberg picture, Eqs. (5) and (11), are equivalent. Both involve infinite hierarchy.

Efficient truncation with prescription invariance would be needed. To proceed, we rewrite here the equation of motion at the $(L+1)$ -dissipations level [cf. Eq. (5)]:

$$\dot{\rho}^{(L+1)} = -[i\mathcal{L}_S + (L+1)\gamma]\rho^{(L+1)} - i(L+1)C\rho^{(L)} - i\mathcal{A}\rho^{(L+2)}. \quad (13)$$

Proposed in this work is the so-called derivative-resum scheme,

$$\dot{\rho}^{(L+1)} \approx -i\mathcal{A}\rho^{(L+2)}. \quad (14)$$

The rest of Eq. (13) then leads to³⁷

$$\rho^{(L+1)}(t) = \frac{-i(L+1)}{i\mathcal{L}_S + (L+1)\gamma} C\rho^{(L)}(t). \quad (15)$$

It leads to Eq. (5) a closed set equations for $\{\rho^{(n \leq L)}\}$. In the Heisenberg picture, it is Eq. (14) used directly, by which Eq. (11) are closed with the additional one,

$$\dot{\hat{A}}^{(L+1)} = -i\hat{A}^{(L)}\mathcal{A}. \quad (16)$$

Both terminal Eqs. (15) and (16), in the Schrödinger picture and the Heisenberg picture, are equivalent to Eq. (14), without invoking further approximation. This concludes that the derivative-resum proposed here is a prescription invariance truncation scheme.

On the other hand, the standard scheme of level truncation is to set $\rho^{(L+1)} = 0$. It leads to Eq. (11), at $n = L$, having the $\hat{A}^{(L-1)}$ -term vanished, without affecting the $\hat{A}^{(L+1)}$ -term at all. This concludes that none of the $\{\rho^{(n)}\}$ -based truncation preserves the prescription invariance. Mixed Heisenberg-and-Schrödinger picture had been used in efficient evaluation of nonlinear response functions.^{6,34} There adopted $\rho^{(L+1)} = 0$ for Schrödinger picture and $\hat{A}^{(L+1)} = 0$ for Heisenberg picture, involving actually two approximations.

Note that a variation of Eq. (15), developed originally by Tanimura and Wolynes,³³ supports also a prescription invariant correspondence. However, the derivation there involved effectively a local classical treatment, by assuming $i\mathcal{L}_S + (L+1)\gamma \approx (L+1)\gamma$. This comprises the numerical efficiency to about the same as the standard $\{\rho^{(n \geq L+1)} = 0\}$ -based truncation. We will elaborate this in Sec. IV.

Actually, Eq. (15) had been proposed before, as an efficient (fermionic) HEOM truncation method.³⁷ The resulting reduced system dynamics is shown to be about the same as that of the standard $\{\rho^{(n \geq L+2)} = 0\}$ truncation scheme. It would suggest that $\rho^{(L+1)}(t)$ of Eq. (15) in terms of $\rho^{(L)}(t)$ be itself a useful part of the DEOM theory. It could be used directly in the DEOM evaluation of hybrid bath dynamics, in such as Fano interference spectroscopy and quantum transport current counting statistics.⁸⁻¹⁰ However, the previous work³⁷ addressed only the numerical aspect of truncation, with setting individual $\dot{\rho}^{(L+1)}$ and $\rho^{(L+2)}$ be zeroes. Apparently, this double setting is overkilled.

Derivative-resum, Eq. (14), with its equivalence Eq. (15) or (16), is preferred for the prescription invariance requirement, while remains numerically efficient. Actually, it performs the overall best among all existing schemes we have tested so far. Both Eqs. (15) and (16) contain the resumed $(L+1)$ -body effect. In other words, expression (15) for

$\rho^{(L+1)}(t)$ would also be useful, whenever the derivative-resum at the given L -body level is reasonable. Moreover, while the steady-state $\rho_{\text{eq}}^{(L+2)} = 0$, as implied in Eq. (14), dynamical $\rho^{(L+2)}(t)$ effect may have partially accounted for.

III. ADVANCEMENT IN SELF-CONSISTENT BORN APPROXIMATION

In this section, we revisit the quantum master equations and their self-consistent-Born-approximation improvements, on the basis of derivative-resum scheme. The conventional quantum master equation reads⁴⁴

$$\dot{\rho}_S(t) = -i\mathcal{L}_S\rho_S(t) - \int_{t_0}^t d\tau \Sigma(t-\tau)\rho_S(\tau), \quad (17)$$

characterized by the memory kernel $\Sigma(t-\tau)$. The widely used second order approach adopts $\Sigma(t-\tau) \approx \langle \mathcal{L}_{\text{SB}}(t)\mathcal{G}_S(t-\tau)\mathcal{L}_{\text{SB}}(\tau) \rangle_B$, with the free system propagator $\mathcal{G}_S(t-\tau) = e^{-i\mathcal{L}_S(t-\tau)}$. This formalism is equivalent to the truncated DEOM at the $\{\rho^{(n \leq 2)}(t) = 0\}$ level. As we discussed before, it violates the prescription invariance. Improvement can be readily achieved by involving the derivative-resum expression for $\rho^{(L=2)}$. The resultant formalism includes the coupled equations of motion of ρ_S and $\rho^{(1)}$. This is actually the simplest self-consistent Born approximation. Moreover, it includes also the $\rho^{(L=2)}$ expression that is also useful for hybrid bath dynamics. [cf. Eq. (2)].

In the following, we generalize the self-energy in terms of continued fraction Green's functions,^{16,20,42,43} in absence of external field. Consider the initial factorization of total density operator ansatz, $\rho_T(0) = \rho_S(0) \otimes \rho_B^{\text{eq}}$. It leads to Eq. (2) the initial conditions of $\rho^{(n)}(0) = \rho_S(0)\delta_{n0}$ for DDOs. Introduce the DEOM-space propagators $\{\mathcal{U}_{n \geq 0}(t)\}$ via^{16,20,42,43}

$$\rho^{(n)}(t) \equiv \mathcal{U}_n(t)\rho_S(0), \quad \text{with } \mathcal{U}_n(0) = \delta_{n0}, \quad (18)$$

and recast Eq. (5) as

$$\begin{aligned} \dot{\mathcal{U}}_n(t) = & -(i\mathcal{L} + n\gamma)\mathcal{U}_n(t) - i n C \mathcal{U}_{n-1}(t) \\ & - i \mathcal{A} \mathcal{U}_{n+1}(t), \end{aligned} \quad (19)$$

for $n = 0, \dots, L$, and

$$\mathcal{U}_{L+1} = \frac{-i(L+1)}{i\mathcal{L}_S + (L+1)\gamma} C \mathcal{U}_L. \quad (20)$$

In the frequency domain, $X(\omega) \equiv \int_0^\infty dt e^{i\omega t} X(t)$, Eq. (20) reads the same for $\mathcal{U}_{L+1}(\omega)$ and Eq. (19) becomes

$$\begin{aligned} \delta_{n0} = & [i(\mathcal{L}_S - \omega) + n\gamma]\mathcal{U}_n(\omega) + i n C \mathcal{U}_{n-1}(\omega) \\ & + i \mathcal{A} \mathcal{U}_{n+1}(\omega). \end{aligned} \quad (21)$$

Define the DEOM-space Green's functions, $\{\mathcal{G}^{(n)}(\omega)\}$, via^{42,43}

$$\mathcal{U}_0(\omega) \equiv \mathcal{G}^{(0)}(\omega), \quad (22a)$$

and

$$\mathcal{U}_n(\omega) \equiv -i n \mathcal{G}^{(n)}(\omega) C \mathcal{U}_{n-1}(\omega); \quad n > 0. \quad (22b)$$

It leads to Eq. (20) the terminal Green's function

$$\mathcal{G}^{(L+1)}(\omega) = \frac{i}{-\mathcal{L}_S + i(L+1)\gamma}. \quad (23)$$

The combination of Eqs. (21) and (22) results in the continued fraction Green's function^{42,43}

$$\mathcal{G}^{(n)}(\omega) = \frac{i}{\omega - \mathcal{L}_S + in\gamma + i\Sigma^{(n)}(\omega)}, \quad n \geq 0, \quad (24)$$

with the self-energy being defined as

$$\Sigma^{(n)}(\omega) = (n+1)\mathcal{A}\mathcal{G}^{(n+1)}(\omega)\mathcal{C}. \quad (25)$$

They are evaluated inversely starting from given $\mathcal{G}^{(L+1)}(\omega)$ in Eq. (23). Conventional quantum master equation (17) is of frequency dependent $\Sigma^{(L=0)}(\omega)$ but setting $\Sigma^{(1)}(\omega) = 0$. The self-consistent Born approximation can be carried out at $L = 1$ or 2 level, following the aforementioned algorithm.

Evidently, derivative-resum, Eq. (14), as inferred from Eqs. (23) and (24), amounts to $\Sigma^{(L+1)}(\omega) \approx i\omega$. This reads in the time domain $\Sigma^{(L+1)}(t) \approx 2[\Delta\delta(t) - \dot{\delta}(t)]$, with $(2\pi i)^{-1} \int d\omega \Sigma_{\text{exact}}^{(L+1)}(\omega)/\omega \approx \Delta$ being a constant. It concludes that the derivative-resum assumes an effective white noise to the self-energy associating with $\rho^{(L+1)}(t)$. It can then be formally integrated out, resulting in Eq. (15), the expression of $\rho^{(L+1)}(t)$ in terms of $\rho^{(L)}(t)$.

IV. COMPARISON WITH OTHER SCHEMES AND COMMENTS

The issue of efficient level truncation, with the focus on the resulting reduced system dynamics of $\rho_S(t) = \rho^{(0)}(t)$, had been extensively addressed before, from the numerical aspect of the DEOM/HEOM formalism, Eq. (5). This is a nonperturbative theory, as inferred from its continued fraction equivalence presented in Sec. III. In other words, DEOM/HEOM is basically a monotonically converged theory. In fact, the n -body DDO, $\rho^{(n)}$ of Eq. (2), is of the leading $(2n)$ th-order in system-bath coupling strength.^{21,22}

The simplest scheme,^{20-24,30-32} which sets all $\{\rho^{(n>L)}(t) = 0\}$, is used reference for the efficiencies of different truncation schemes. In this standard (L) truncation scheme, the resulting $\rho_S(t)$ is exact up to the $(2L)$ th-order system-bath coupling.^{21,22}

Derivative-resum scheme, Eq. (14) or its equivalent Eq. (15), termed as d-resum (L) below, does achieve the similar results of standard ($L+1$) on the reduced system $\rho_S(t)$ dynamics. Other existing level truncation schemes^{20-22,33} are practically about the same of the standard one.

For illustration, we examine closely the truncation scheme, proposed originally³³ and used extensively by Tanimura and co-workers,³⁸⁻⁴¹

$$\rho^{(L+1)}(t) \approx -\frac{i}{\gamma} \mathcal{C} \rho^{(L)}(t), \quad (\text{standard}'). \quad (26)$$

This scheme, to be called the standard' (L) below, is also of prescription invariance, but compromises the underlying quantum nature of reduced system at the $(L+1)$ -dissipations level. In other words, Eq. (26) is a classical Markov limit of Eq. (15), assuming the reduced system Liouvillian $\mathcal{L}_S$ can be effectively neglected in comparing with $(L+1)\gamma$. Green's function, Eq. (23), becomes a purely diffusive form of $\mathcal{G}^{(L+1)}(\omega) \approx 1/[(L+1)\gamma]$.

FIG. 1. The performances of the derivative-resum (solid-red), standard' (solid-blue), and standard (dashed) level-truncation schemes, comparing with the converged results (solid-black). Reported are the reduced system dynamics of a spin-boson model: $H_S = (\epsilon/2)\hat{\sigma}_z + V\hat{\sigma}_x$ and $\hat{Q} = \hat{\sigma}_z$, with $\beta\epsilon = \beta V = 1$, where $\beta = 1/(k_B T)$. The bath spectral density adopts Drude form [cf. Eq. (40)], with the reorganization energy $\beta\lambda = 1$ and cutoff frequency $\beta\gamma = 2$.

Figure 1 reports the performances of derivative-resum (d-resum) [Eq. (15)] and standard' [Eq. (26)] schemes, with respect to the simplest (standard) truncation, for a spin-boson model specified in the figure caption. The resulting reduced system $\rho_S(t)$ comparisons are reported in terms of the expectation values of Pauli spin operators. The exact results are also included for reference.

Apparently, the derivative-resum truncated at $L = 2$ performs numerically comparable to the standard scheme at $L = 3$ level results. This feature had been observed in our extensive fermionic HEOM evaluations.³⁷ As the reduced system dynamics is concerned (cf. Fig. 1),

$$\text{d-resum}(L-1) \simeq \text{standard}(L) \approx \text{standard}'(L). \quad (27)$$

This observation may be understood by considering the $(L+1)$ -level Green's function, which for the d-resum scheme is given by Eq. (23). However, the standard' scheme adopts the classical Markovian approximation, resulting in the purely diffusive $\mathcal{G}^{(L+1)}(\omega) \approx 1/[(L+1)\gamma]$ that is closer to zero, the result of the standard truncation, rather than to Eq. (23). The standard' scheme has practically no improvement over the simplest truncation method. To further elaborate this, we exploit Eq. (13) and recast Eq. (26) with its derivative equivalence,

$$\dot{\rho}^{(L+1)} = -i\mathcal{L}_S \rho^{(L+1)} - i\mathcal{A}\rho^{(L+2)}, \quad (\text{standard}'). \quad (28)$$

As inferred from Sec. II B, this also belongs to the class of prescription invariance truncation schemes. However, the right-hand-side of Eq. (28) involves two terms, with different orders in the system-bath coupling strength. Numerically, this $\rho^{(L+2)}$ -term be an order-of-magnitude less important than the $\rho^{(L+1)}$ -term in Eq. (28). The underlying inconsistency is removed in derivative-resum scheme, Eq. (14) and its

FIG. 2. The $\rho^{(3)}(t)$ via derivative–resum [Eq. (15)] and standard' [Eq. (26)] schemes, both at $L=2$, compared with the converged dynamics. The comparisons on $\text{Re } \rho_{12}^{(3)}(t)$ (not shown here) are similar to that of $\text{Im } \rho_{12}^{(3)}(t)$, with much smaller amplitude.

equivalence Eq. (15). As inferred from the continued fraction Green's function analysis in Sec. III, the derivative–resum becomes valid when the local self-energy $\Sigma^{(L+1)}(\omega)$ can be approximated by white noise, i.e., $\Sigma^{(L+1)}(\omega) \approx i\omega$. The corresponding local Green's function $\mathcal{G}^{(L+1)}(\omega)$ of Eq. (23) is frequency independent, but its quantum nature via the $\mathcal{L}_S$ is retained. It is anticipated that at the vicinity of converged tier level, $\rho^{(L+1)}(t)$ has the order similar to $\rho^{(L+2)}(t)$. This is the numerical basis for the derivative–resum scheme, Eq. (14) and its equivalent Eq. (15). In contrast, the standard' scheme $\rho^{(L+1)}(t)$ of Eq. (28) is of the leading order similar to $\rho^{(L+1)}(t)$. The above order–based analysis concludes the numerical insights for the observed behaviours of Eq. (27).

Figure 2 depicts the resulting $\rho^{(L+1=3)}(t)$ from the d–resum(L) and the standard' (L) truncation schemes, comparing to the exact results for the test model of Fig. 1. Note that the scaled $\rho^{(3)}(t)/\sqrt{3!|\eta|^3}$, which is the normalized three–body DDO,⁴⁵ is about 50% of the reduced system density matrix, $\rho_S(t) \equiv \rho^{(0)}(t)$, whereas standard ($L=2$) scheme assumes $\rho^{(3)}(t) \approx 0$. The derivative–resum ($L=2$) does semi-quantitatively recovers the dynamics of various $\rho^{(3)}(t)$ matrix elements. On the other hand, standard'–resum [Eq. (26)], which is the classical version of derivative–resum [Eq. (15)], significantly compromises the off–diagonal $\rho^{(L+1)}(t)$ density matrix elements, and also the overall long–time behavior. The above analysis elaborates further the observed behaviours of Eq. (27), via the underlying quantum versus classical nature, at the truncation level. In contrast to standard' Eq. (26), derivative–resum Eq. (14) does capture the $(L+1)$ –body effect.

V. HYBRID SYSTEM–AND–BATH DYNAMICS

A. Generalized Wick's theorem for dissipatons

The DEOM theory is not just how the DDOs evolve in time, as governed by Eq. (1), but also the underlying statistical quasi-particle picture and the novel dissipaton algebra.⁸ In particular, the notion of irreducibility, $(\cdots)^\circ$ in Eq. (2) is

closely related to the generalized Wick's theorem,^{8–10}

$$\begin{aligned} \text{tr}_B[(\prod_{ak} \hat{f}_{ak}^{n_{ak}})^\circ \hat{f}_{bj} \rho_T] &= \rho_{n_{bj}}^{(n+1)} + \sum_{ak} n_{ak} \langle \hat{f}_{ak} \hat{f}_{bj} \rangle_B^\circ \rho_{n_{ak}}^{(n-1)} \\ &= \rho_{n_{bj}}^{(n+1)} + n_{bj} \eta_{bj} \rho_{n_{bj}}^{(n-1)} \end{aligned} \quad (29a)$$

and

$$\begin{aligned} \text{tr}_B[(\prod_{ak} \hat{f}_{ak}^{n_{ak}})^\circ \rho_T \hat{f}_{bj}] &= \rho_{n_{bj}}^{(n+1)} + \sum_{ak} n_{ak} \langle \hat{f}_{bj} \hat{f}_{ak} \rangle_B^\circ \rho_{n_{ak}}^{(n-1)} \\ &= \rho_{n_{bj}}^{(n+1)} + n_{bj} \eta_{bj}^* \rho_{n_{bj}}^{(n-1)}. \end{aligned} \quad (29b)$$

The above two equations are used in deriving the DEOM formalism.^{8–10} They are also crucially significant in evaluating the hybrid bath dynamics. This unique feature of the DEOM framework will be exemplified below.

B. DEOM evaluation on system and bath dynamics

Consider a Fano interference setup where both discrete system and the continuum bath are optically active. The total composite Hamiltonian with field-driving assumes

$$H_T(t) = H_T - \hat{\mu}_T \varepsilon(t), \quad \text{with } \hat{\mu}_T = \hat{\mu}_S + \hat{\mu}_B. \quad (30)$$

The bath dipole assumes the form of $\hat{\mu}_B = \nu_B \hat{F}_B$, with $\hat{F}_B = \hat{f}$, where $\hat{f}$ standards for the dissipaton operator described in Eq. (3). The DEOM formalism can be obtained as^{8,9}

$$\begin{aligned} \dot{\rho}^{(n)}(t) &= -[i\mathcal{L}(t) + n\gamma] \rho^{(n)}(t) - in C_{\text{eff}}(t) \rho^{(n-1)}(t) \\ &\quad - i\mathcal{A} \rho^{(n+1)}(t). \end{aligned} \quad (31)$$

Here, $\mathcal{L}(t)\hat{O} \equiv [H_S - \hat{\mu}_S \varepsilon(t), \hat{O}]$ and [cf. Eq. (6)]

$$C_{\text{eff}}(t) = C - \nu_B \varepsilon(t)(\eta - \eta^*). \quad (32)$$

Terminal Eq. (15) reads now

$$\rho^{(L+1)}(t) = \frac{-i(L+1)}{i\mathcal{L}(t) + (L+1)\gamma} C_{\text{eff}}(t) \rho^{(L)}(t). \quad (33)$$

Turn to the evaluation of the optical response function,

$$\begin{aligned} \langle [\hat{\mu}_T(t), \hat{\mu}_T(0)] \rangle &= \text{Tr}[\hat{\mu}_T e^{-i\mathcal{L}t} [\hat{\mu}_T, \rho_T^{\text{eq}}]] \\ &\equiv \text{Tr}[\hat{\mu}_T \rho_T(t; \hat{\mu}_T)]. \end{aligned} \quad (34)$$

The correlation function is similar, with $\langle \hat{\mu}_T(t) \hat{\mu}_T(0) \rangle = \text{Tr}[\hat{\mu}_T e^{-i\mathcal{L}t} (\hat{\mu}_T \rho_T^{\text{eq}})]$. All response/correlation functions are defined in the total system–plus–bath composite matter space. In comparison, the DEOM describes the dynamics in the system–and–hybrid–bath space. Under this support, the response/correlation functions all have well defined DEOM–space correspondences. For example, the DEOM evaluation of Eq. (34) reads^{8,9}

$$\langle [\hat{\mu}_T(t), \hat{\mu}_T(0)] \rangle = \text{tr}_S[\hat{\mu}_S \rho^{(0)}(t; \hat{\mu}_T)] + \nu_B \text{tr}_S[\rho^{(1)}(t; \hat{\mu}_T)], \quad (35)$$

with the initial DDOs being determined as

$$\rho^{(n)}(t=0; \hat{\mu}_T) = [\hat{\mu}_S, \rho_{\text{eq}}^{(n)}] + n \nu_B (\eta - \eta^*) \rho_{\text{eq}}^{(n-1)}. \quad (36)$$

They are obtained via applying $\rho_T(t=0; \hat{\mu}_T) = [\hat{\mu}_T, \rho_T^{\text{eq}}]$, as implied in Eq. (34) to Eq. (2), followed by the generalized Wick's theorem, Eq. (29). The thermal equilibrium DDOs, $\{\rho_{\text{eq}}^{(n)}\}$ involving in Eq. (36), are the steady–state solutions

to the field-free DEOM, at given temperature. They can be obtained by propagating DEOM to equilibrium; also by setting $\{\dot{\rho}^{(n)} = 0\}$ and solving the resulting linear equations for $\{\rho_{\text{eq}}^{(n)}\}$, under the normalization condition $\text{tr}_S \rho_{\text{eq}}^{(0)} = 1$. Then the initial states $\{\rho^{(n)}(0; \hat{\mu}_T)\}$ are prepared as Eq. (36), and propagated using the same DEOM. Finally, Eq. (35) is evaluated.

The evaluation of nonlinear polarization, $P(t) \equiv \text{Tr}[\hat{\mu}_T \rho_T(t)]$, is similar and via [cf. Eq. (34)]

$$P(t) = \text{tr}_S[\hat{\mu}_S \rho^{(0)}(t)] + \nu_B \text{tr}_S[\rho^{(1)}(t)]. \quad (37)$$

It starts with the same equilibrium $\{\rho_{\text{eq}}^{(n)}\}$ as before, which also determined P_{eq} as Eq. (37). Then, field-driving DEOM (31) is used to propagate $\{\rho^{(n)}(t)\}$, and Eq. (37) is evaluated accordingly, upon the strong driving field $\varepsilon(t)$ switched on. Nonlinear polarization spectroscopy is related to the imaginary part of

$$\tilde{P}(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} [P(t) - P_{\text{eq}}]. \quad (38)$$

Linear spectrum can be obtained with the linear response theory,⁴⁶ which is identical to the Fourier transform of Eq. (34).

C. Nonlinear Fano interferences

We illustrate the DEOM evaluation of the linear/nonlinear Fano interference with a two-level model system as follows. The reduced system Hamiltonian H_S , the dissipative mode $\hat{Q}^S$ and the system dipole operator $\hat{\mu}_S$ are given in terms of Pauli

matrix as

$$H_S = \frac{1}{2} \Omega \hat{\sigma}_z, \quad \hat{Q}^S = \hat{\sigma}_x, \quad \text{and} \quad \hat{\mu}_S = \mu_S \hat{\sigma}_x, \quad (39)$$

respectively. The interacting bath adopts the Drude spectral density,

$$J(\omega) = \frac{2\lambda\gamma\omega}{\omega^2 + \gamma^2}, \quad (40)$$

where λ and γ are the system–bath coupling strength and the cutoff frequency. The bath dipole operator had assumed the form of $\hat{\mu}_B = \nu_B \hat{F}_B = \nu_B \hat{f}$ [see the sentence below Eq. (30)], which would suggest $\mu_B = 2\lambda\nu_B$ for the bath dipole strength. The external field is a Gaussian pulse, having

$$\mu_T \varepsilon(t) = \frac{\theta}{\sqrt{2\pi}\sigma} \exp\left(-\frac{t^2}{2\sigma^2}\right) \cos(\omega_c t), \quad (41)$$

with total dipole strength $\mu_T = \mu_S + \mu_B$, temporal variance σ and carrier frequency ω_c . Here, θ is the flipping angle, where a $\theta = \pi$ resonant field leads to complete conversion of population for dissipation-free system.

Figure 3 reports the field-dressed polarization spectrum, in term of $-\text{Im} \tilde{P}(\omega)/(\beta\mu_T)$. Comparisons are made between the optically dark bath (top panel) and active bath (lower panels) cases. In the fundamental frequency region, from top to bottom panel, the resonant system peak declines, while the bath polarization signal emerges and intensifies. Observed in this region are also the interplay between the effective system resonance around $\Omega_{\text{eff}} = \sqrt{\Omega^2 + 4\lambda/\beta}$ and laser field carrier frequency $\omega_c = \Omega$. The bump feature of bare bath $J(\omega)$ peaked around $\Omega/2$ may also play roles. The third-harmonic-generation appears around $3\omega_c$. Observed in this region are multiple turnovers: from absorptive peaks at $\mu_B = 0$, to emissive dips at $\mu_B/\mu_S = 1$ and 2, to absorptive at $\mu_B/\mu_S = 5$, and to emissive at $\mu_S = 0$, respectively.

In the linear response regime, the polarization spectrum is related to the dipole response function $S(\omega)$, via $S(\omega) = -\text{Im}[\tilde{P}(\omega)/\tilde{\varepsilon}(\omega)]$. Here, we evaluate it directly via the linear

FIG. 3. Nonlinear Fano interference under strong external field manifested by system and hybrid bath polarization. The energy difference between two levels is set to be $\beta\Omega = 2$, and the bath parameters are $\beta\lambda = 0.1$ and $\beta\gamma = 1$. The Gaussian pulse has carrier frequency $\omega_c = \Omega$, temporal variance $\sigma = 12/(\sqrt{2}\ln 2\Omega)$, and intensity in terms of flipping angle θ that is proportional to total dipole of $\mu_T = \mu_S + \mu_B$. The values of μ_B/μ_S are reported in individual panels.

FIG. 4. The linear Fano interference turnovers, with the same system and bath parameters as Fig. 3 but plotted with respect to the effective resonant frequency, $\Omega_{\text{eff}} = \sqrt{\Omega^2 + 4\lambda/\beta}$.

response theory,⁴⁶

$$S(\omega) = \text{Re} \int_0^\infty dt e^{i\omega t} \langle [\hat{\mu}_T(t), \hat{\mu}_T(0)] \rangle. \quad (42)$$

Figure 4 depicts the simulated Fano interference profiles, with the same parameters as Fig. 3. It clearly shows the turnover of Fano lineshape, due to the competition of optical response between bath and system. These features appear around the effective system frequency Ω_{eff} . The driving field does not play roles in the linear response regime. It is worth to note that the DEOM can also treat individual correlation functions of hybrid bath operators.^{8,9} The fermionic DEOM formalism has also been developed and applied to evaluate transport shot-noise spectra.^{8,10}

VI. CONCLUDING REMARKS

The hierarchy truncation issue involves in many approximate methods. In this work, we have addressed this issue from not just the numerical efficiency aspect, but also the fundamental requirement of quantum mechanics prescriptions invariance. The latter demands the truncation approximation made in Schrödinger picture should be transferable to their equivalence in the Heisenberg picture without further approximations. This fundamental physical requirement rules out the $\{\rho_n^{(n>L)}\}$ -based truncation, including the widely used quantum master equations and their self-consistent-Born-approximation improvements. The proposed derivative-resum truncation scheme preserves the prescription invariance, while practically retrieve the influence of $\{\rho_n^{(L+1)}\}$ back to the final DEOM dynamics.

In relation to the truncation issue, we have also made contact with the continued fraction Green's function approach. We have shown that the derivative-resum truncation scheme is a kind of generalized high-order self-consistent Born approximation. It resembles a certain dynamical meanfield treatment of the high-order many-body DDO effect. Comments on other existing truncation schemes, especially the one proposed originally^{33,38} and used extensively by Tanimura and coworkers,^{20,30,31,41} are also presented in depth; see Sec. IV.

To highlight the capability of DEOM for system and hybrid bath dynamics, we carried out simulations on the Fano interference in model systems, and the well-known asymmetric Fano lineshape is observed in the linear response regime. Furthermore, the non-perturbative nature of DEOM enables us to study the strong field nonlinear Fano interference. The intriguing competition between system and bath polarization and driving field reported in Fig. 3 can be captured in experiments.

In the end, we emphasize that the DEOM is a novel type of reduced dynamics theory. It projects the total system-and-bath composite ρ_T to a set of physically well-defined DDOs in the system-and-hybrid-bath space. Moreover, the DEOM theory is not only about how the DDOs evolve in time but also the underlying dissipaton algebra. These unique features enable DEOM to describe both the system and the hybrid bath dynamics and their correlated interference.

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