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Time-Local Quantum-State-Diffusion Equation for Multilevel Quantum Dynamics

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An open quantum system with multiple levels coupled to a bosonic environment at zero temperature is investigated systematically using the non-Markovian quantum state diffusion (QSD) method [W. T. Strunz, L. Diósi and N. Gisin, Phys. Rev. Lett. **82**, 1801 (1999)]. We have established exact time-local QSD equations for a set of interesting multilevel open systems including the high-spin systems, multiple transition atom models, and multilevel atom models driven by time-dependent external fields. These exact QSD equations have paved a way to evaluate the dynamics of the open multilevel atomic systems in the general non-Markovian regimes without any approximation.

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I. INTRODUCTION

Many quantum processes, including emission, absorption and quantum interference [1, 2], typically involve multilevel atomic systems coupled to a quantized bosonic field. For example, quantum multilevel models are used in studies of the high-spin model [3], super-radiance, quantum phase transition in Dicke model [4, 5], electromagnetically induced transparency (EIT) [6], molecular aggregate [7] and many other phenomena in quantum optics [8, 9] and quantum chemistry [10, 11]. More recently, multilevel atomic systems also play a crucial role in describing coherent state transfer [12] and entanglement dynamics and control [13–18]. All of those models concern the interactions between a multilevel atomic system and its surrounding environment and external driving fields leading to collective and competitive behaviors including dissipation, fluctuation, decoherence and revival. Approximate master equations and other alternatives such as the Langevin equations or quantum trajectories are used to describe quantum dynamics of the multilevel atomic system when the weak-coupling and Markov approximations are valid [19].

In this paper, the multilevel atomic systems will be treated in the framework of the non-Markovian quantum state diffusion equation [20–25] without any approximations. Unlike the methods of Markov quantum state diffusion or quantum jump simulations [26–30], where quantum trajectories are unravelings of the density operator of the system of interest, the non-Markovian QSD equation is derived from the first principle and is determined uniquely by the system Hamiltonian, the coupling operator (called the Lindblad operator) and the spectral density of the environment [21, 31]. It has been shown that the non-Markovian QSD equation can be useful in the following ways: (i) it is an exact description of quantum dynamics of the system under the influence of the environmental noise with finite memory time [32–36], in particular, it serves as a powerful tool in numerical simulations of quantum open systems; (ii) it could also be used to derive the exact or approximate master equations for the reduced density matrix [37–39].

Exact QSD equations have been found in several interesting models including a two-level atom in the dissipative environment [22], a harmonic oscillator in Brownian motion [38], a three-level atom in the dissipation model [25], and a two-qubit model [40]. However, it remains unknown how exact QSD equations can be derived for general multilevel systems coupled to a dissipative environment. The purpose of this paper is to establish a set of exact QSD equations for a large class of multilevel atomic models coupled to a dissipative environment with or without external driving fields.

This paper will be organized as follows. In Sec. II, we will begin by reviewing the basic concepts of the non-Markovian QSD approach that will be useful for the discussions presented in the subsequent sections including the formal QSD equation and the O-operators. In Sec. III, we will then proceed to discuss the construction of the time-local QSD equation by explicitly determining O-operators in various multilevel models. Afterwards, we present the numerical results from QSD simulations to illustrate the non-Markovian dynamics of multilevel atomic systems under environmental noises in Sec. IV. Finally we will conclude the paper in Sec. V.

II. NON-MARKOVIAN QSD EQUATION

In the framework of system-plus-environment, the total Hamiltonian is given by (setting $\hbar = 1$): $H_{\rm tot} = H_{\rm sys} + \sum_{\mathbf{k}}(g^*_{\mathbf{k}} L a^\dagger_{\mathbf{k}} + g_{\mathbf{k}} L^\dagger a_{\mathbf{k}}) + \sum_{\mathbf{k}} \omega_{\mathbf{k}} a^\dagger_{\mathbf{k}} a_{\mathbf{k}}$, where $H_{\rm sys}$ is the system Hamiltonian and L is said to be the Lindblad operator coupling the system to the environment. The environment is described by a set of harmonic oscillators

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$a_{\mathbf{k}}^\dagger, a_{\mathbf{k}}$ satisfying $[a_{\mathbf{k}}, a_{\mathbf{k}'}^\dagger] = \delta_{\mathbf{k},\mathbf{k}'}$. The non-Markovian QSD approach is designed to recover the reduced density matrix for the open system by the average of the pure states driven by a colored Gaussian process z_t^*:

$$\rho_t = M[|\psi_t(z^*)\rangle\langle\psi_t(z^*)|], \tag{1}$$

where M stands for the statistical average over the noise z_t^*. The dynamics of the stochastic unravelings $\psi_t(z^*)$ (quantum trajectories) is governed by the non-Markovian QSD equation [20, 21]:

$$\partial_t\psi_t(z^*) = \left[-iH_{\rm sys} + Lz_t^* - L^\dagger\int_0^t ds\alpha(t,s)\frac{\delta}{\delta z_s^*}\right]\psi_t(z^*), \tag{2}$$

where $\alpha(t,s) = \sum_{\mathbf{k}}|g_{\mathbf{k}}|^2 e^{-i\omega_{\mathbf{k}}(t-s)}$ is the environmental correlation function. By construction, the Gaussian noise z_t^* satisfies $M[z_t^*] = M[z_t^* z_s^*] = 0$ and $M[z_t z_s^*] = \alpha(t,s)$. Note that the functional derivative contained in Eq. (2) is a time non-local term depending on the entire evolution history from 0 to t. This time non-local term is a major obstacle in using Eq. (2) as a numerical tool or as an analytical approach to deriving the corresponding non-Markovian master equation. One way to transform the formal non-Markovian QSD equation (2) to a time-local equation is to introduce an O-operator satisfying $\frac{\delta\psi_t(z^*)}{\delta z_s^*} \equiv O(t,s,z^*)\psi_t(z^*)$, then the QSD equation could be recast into a convolutionless form:

$$\partial_t\psi_t(z^*) = \left[-iH_{\rm sys} + Lz_t^* - L^\dagger\bar{O}(t,z^*)\right]\psi_t(z^*), \tag{3}$$

where $\bar{O}(t,z^*) \equiv \int_0^t ds\alpha(t,s)O(t,s,z^*)$ and the O-operator may be determined from the following equation:

$$\frac{\partial O(t,s,z^*)}{\partial t} = [-iH_{\rm sys} + Lz_t^* - L^\dagger\bar{O}(t,z^*), O(t,s,z^*)] - L^\dagger\frac{\delta\bar{O}(t,z^*)}{\delta z_s^*}. \tag{4}$$

Hence, the key issue in establishing the exact time-local QSD equations is to explicitly construct the O-operator defined in (4). In what follows, we shall show how to determine the O-operator explicitly for a large class of multilevel atomic systems.

It is worthwhile to note that, from the density matrix reconstruction defined in Eq. (1) and Novikov theorem [37], we may obtain an exact master equation from Eq. (3):

$$\partial_t\rho_t = [-iH_{\rm sys},\rho_t] + [L, M[\hat{P}_t\bar{O}^\dagger]] + [M[\bar{O}\hat{P}_t], L^\dagger], \tag{5}$$

where $\hat{P}_t \equiv |\psi_t(z^*)\rangle\langle\psi_t(z^*)|$. The master equation takes a more concise form when the O-operator is noise-free: $O(t,s,z^*) = O(t,s)$,

$$\partial_t\rho_t = -i[H_{\rm sys},\rho_t] + [L,\rho_t\bar{O}^\dagger] + [\bar{O}\rho_t, L^\dagger]. \tag{6}$$

In particular, under the Born-Markov approximation with the delta function $\alpha(t,s) = \Gamma\delta(t-s)$, then $\bar{O}(t,s,z^*) = \Gamma L/2$. Eq. (6) recovers the Lindblad master equation:

$$\partial_t\rho_t = -i[H_{\rm sys},\rho_t] + \frac{\Gamma}{2}\left([L,\rho_t L^\dagger] + [L\rho_t, L^\dagger]\right). \tag{7}$$

III. TIME-LOCAL QSD EQUATIONS

A. High-Spin Model

Now we consider a collective angular momentum (Dicke) model or the high-spin model described by

$$H_{\rm sys} = \omega J_z, \quad L = J_-, \tag{8}$$

where J_z and J_- are the angular momentum operators with spin-l. It can be proved (see Appendix A) that the O-operator contains $l(2l+1)$ terms with the basis operators $O_j^{(k)} = |j\rangle\langle j+k+1|$, $j = 1,\cdots,2l-k$, for each $k = 0,\cdots,2l-1$. And the terms with $O_j^{(k)}$ contain kth order noises for $k = 0,\cdots,2l-1$.

Clearly, one may construct many different sets of basis operators, but they can always be realized by linear combinations of the operators $O_j^{(k)}$ selected here. The basis operators may also be given by $O_j^{(k)} = J_z^{j-1}J_-^{k+1}$. The simplest model is the spin-1/2 system ($l = 1/2$), where the O-operator contains only one term with the basis operator $O_1^{(0)} = \sigma_-$. Another model is spin-3/2. We can show the O-operator $O(t,s,z^*)$ totally contains 6 terms, amongst, only one term contains the second-order noise:

$$O = \sum_{j=1}^{3} f_j(t,s)O_j^{(0)} + \sum_{j=1}^{2}\int_0^t p_j^{(1)}(t,s,s_1)z_{s_1}^* ds_1 O_j^{(1)} + \int_0^t\int_0^t p_1^{(2)}(t,s,s_1,s_2)z_{s_1}^* z_{s_2}^* ds_1 ds_2 O_1^{(2)}. \tag{9}$$

In Table I, we list the numbers of the basis operators for the high-spin models from spin-1/2 to spin-7/2.

$2l \setminus k$	0	1	2	3	4	5	6	7	N
1	1	0	0	0	0	0	0	0	1
2	2	1	0	0	0	0	0	0	3
3	3	2	1	0	0	0	0	0	6
4	4	3	2	1	0	0	0	0	10
5	5	4	3	2	1	0	0	0	15
6	6	5	4	3	2	1	0	0	21
7	7	6	5	4	3	2	1	0	28

TABLE I. The number of basis operators with k-fold integration over noises in the O-operator for spin-l system in a dissipation model. N is total number of the terms in the O-operators.

The Hamiltonian and Lindblad operator considered in Eq. (8) imply that the energy levels of the multilevel atom are equidistant and all the exited levels dissipate to the lower neighboring levels. However, these two constraints can be relaxed. We start with the model Eq. (8) with the modified Hamiltonian and coupling operator:

$$H_{\rm sys} = \sum_j \omega_j|j\rangle\langle j|, \quad L = \sum_j \kappa_j|j\rangle\langle j+1|. \tag{10}$$

More specifically, we use a three-level system as an example for simplicity. It can be verified that the O-operator

can be constructed as follows (For details, see Appendix A or Eq. (6) of [25]):

$$O = f_1(t,s)J_- + f_2(t,s)J_zJ_- + \int_0^t p(t,s,s_1)z^*_{s_1}ds_1J_-^2. \tag{11}$$

Consequently, $\bar{O}(t,z^*) = F_1(t)J_- + F_2(t)J_zJ_- + \int_0^t P(t,s_1)z^*_{s_1}ds_1J_-^2$, where $F_j(t) \equiv \int_0^t \alpha(t,s)f_j(t,s)ds$, $j=1,2$ and $P(t,s_1) \equiv \int_0^t \alpha(t,s)p(t,s,s_1)ds$. The initial conditions for these coefficients are $f_1(s,s) = \kappa_2/\sqrt{2}$, $f_2(s,s) = (\kappa_2-\kappa_1)/\sqrt{2}$, $p(s,s,s_1) = 0$ and they satisfy:

$$\begin{aligned}
\partial_t f_1 &= i(\omega_3-\omega_2)f_1 - \sqrt{2}[(\kappa_1-\kappa_2)F_1 - \kappa_1F_2]f_1 \\
&\quad - \sqrt{2}i\kappa_1P(t,s), \\
\partial_t f_2 &= i(\omega_3+\omega_1-\omega_2)f_1 - \sqrt{2}i\kappa_1P(t,s) \\
&\quad + i(\omega_2-\omega_1)f_2 + \sqrt{2}\kappa_1(F_1-F_2)f_2 \\
&\quad - \sqrt{2}[(2\kappa_1-\kappa_2)F_1 - 2\kappa_1F_2]f_1, \\
\partial_t p &= i(\omega_3-\omega_1)p + \sqrt{2}\kappa_2F_1p + \sqrt{2}\kappa_1P(f_1-f_2), \\
p(t,s,t) &= i\frac{\kappa_2-\kappa_1}{\sqrt{2}}f_1(t,s) - i\frac{\kappa_2}{\sqrt{2}}f_2(t,s).
\end{aligned} \tag{12}$$

These equations will reduce to Eqs. (7), (8) and (9) in [25] by setting $\omega_3 = \omega$, $\omega_2 = 0$, $\omega_1 = -\omega$ and $\kappa_1 = \kappa_2 = \sqrt{2}$. As shown in the next subsection, the time-local QSD approach can be extended to the driven atomic models with the multiple transitions between the energy levels.

B. Multiple Transition and Driven Atomic Models

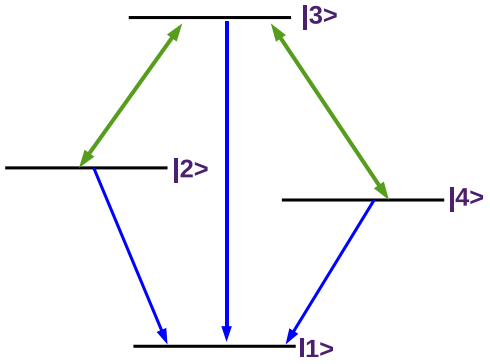

FIG. 1. (Color online) Schematic diagram of a general EIT model: A four-level atomic system with two driving fields (green double arrow lines) as $\Delta_2(t)T_{23} + h.c.$ and $\Delta_4(t)T_{34} + h.c.$. The admitted damping channels (blue single arrow lines) are T_{12}, T_{13} and T_{14}.

In this subsection, we consider a driven multi-level atom with multiple dissipative channels. The system Hamiltonian is given by,

$$H_{\rm sys} = \sum_{j=1}^N \omega_j|j\rangle\langle j| + \sum_{k_1\neq k_2}[\Delta_{k_1k_2}(t)|k_1\rangle\langle k_2| + h.c.], \tag{13}$$

where $\Delta_{k_1k_2}(t)$'s are the time-dependent functions and $1 \leqslant k_1, k_2 \leqslant N$. Also, we consider a general Lindblad operator, which is given by

$$L = \sum_{n_1\neq n_2}\kappa_{n_1n_2}|n_1\rangle\langle n_2|. \tag{14}$$

We remark that one may always determine the exact time-local QSD equation for the above generic Hamiltonian and the Lindblad operator. However, it can be verified that a particularly simple noise-free O-operator exists if the following two conditions are satisfied: (i) any operator $|k_1\rangle\langle k_2|$ appearing in the driving term is not present in the Lindblad operator in Eq. (14); (ii) the cycle transition terms, for example, $|n_1\rangle\langle n_2|$, $|n_2\rangle\langle n_3|$ and $|n_1\rangle\langle n_3|$, cannot be simultaneously contained in the Lindblad operator. As an illustration, we consider an interesting model consisting of a four-level atom shown in Fig. (1), the system is described by

$$\begin{aligned}
H_{\rm sys} &= \sum_{j=1}^4 \omega_j|j\rangle\langle j| + [\Delta_2(t)|2\rangle\langle 3| + \Delta_4(t)|3\rangle\langle 4| + h.c.], \\
L &= \kappa_2|1\rangle\langle 2| + \kappa_3|1\rangle\langle 3| + \kappa_4|1\rangle\langle 4|,
\end{aligned} \tag{15}$$

where $\Delta_2(t)$ and $\Delta_4(t)$ are independent driving external fields. Thus the O-operator can be constructed as $O(t,s,z^*) = \sum_{j=2}^4 f_j(t,s)|1\rangle\langle j|$ with $f_j(s,s) = \kappa_j$. And the differential equations for the coefficients are

$$\begin{aligned}
\partial_t f_2(t,s) &= -i\omega_{12}f_2 + i\Delta_2^*f_3 + F_2\sum_{j=2}^4\kappa_j^*f_j, \\
\partial_t f_3(t,s) &= -i\omega_{13}f_3 + i\Delta_2f_2 + i\Delta_4^*f_4 + F_3\sum_{j=2}^4\kappa_j^*f_j \\
\partial_t f_4(t,s) &= -i\omega_{14}f_4 + i\Delta_4f_3 + F_4\sum_{j=2}^4\kappa_j^*f_j,
\end{aligned} \tag{16}$$

where $\omega_{1j} \equiv \omega_1 - \omega_j$ and $F_j \equiv F_j(t) = \int_0^t ds\alpha(t,s)f_j(t,s)$, $j=2,3,4$.

When the driving terms in $H_{\rm sys}$ are omitted, the model in Eq. (13) reduces to an N-level atom with multiple transition channels [41] where the transition takes place between the highest level and all the lower energy levels, and transitions between any other levels are forbidden. Explicitly, the Hamiltonian and Lindblad operator in this case are given by

$$H_{\rm sys} = \sum_{j=1}^N \omega_j|j\rangle\langle j|, \quad L = \sum_{j=1}^{N-1}\kappa_j|j\rangle\langle N|, \tag{17}$$

respectively. For this model, we can show that the O-operator can be explicitly constructed as $O(t,s,z^*) = \sum_{j=1}^{N-1} f_j(t,s)|j\rangle\langle N|$. Hence $\bar{O}(t,z^*) = \sum_{j=1}^{N-1} F_j(t)|j\rangle\langle N|$ with the initial conditions, $f_j(s,s) = \kappa_j$, $j = 1,\cdots,N-1$. By the consistency condition (4),

one gets

$$\partial_t f_j(t,s) = i(\omega_N - \omega_j) f_j + f_j \sum_{k=1}^{N-1} \kappa_k^* F_k(t). \quad (18)$$

In a more general case, we may consider an $(N+M)$-level atom with an upper energy band consisting of N levels and a lower band consisting of the other M levels. We assume that transitions between the upper band and the lower band are allowed, but those between the energy levels inside the upper band or the lower band are strictly forbidden. Such a model may be described by

$$H_{\rm sys} = \sum_{j=1}^{N+M} \omega_j |j\rangle\langle j|, \quad L = \sum_{j=1}^{M} \sum_{k=M+1}^{N} \kappa_{jk} |j\rangle\langle k|. \quad (19)$$

The O-operator is explicitly constructed as a noise-free formation by $O(t,s) = \sum_{j=1}^{M} \sum_{k=M+1}^{N} f_{jk}(t,s)|j\rangle\langle k|$ and $\bar{O}(t) = \sum_{j=1}^{M} \sum_{k=M+1}^{N} F_{jk}(t)|j\rangle\langle k|$ with $f_{jk}(s,s) = \kappa_{jk}$. And we have

$$\partial_t f_{jk}(t,s) = i(\omega_k - \omega_j) f_{jk} + \sum_{j'=1}^{M} \sum_{k'=M+1}^{N} f_{jk'} \kappa_{j'k'}^* F_{j'k}. \quad (20)$$

Therefore, for the cases with the noise-free O-operators, the exact master equations can be derived directly from Eq. (6). It is noted that such master equations may not be of a standard Lindblad form, but their positivity is automatically guaranteed by the derivation.

IV. NUMERICAL RESULTS AND DISCUSSIONS

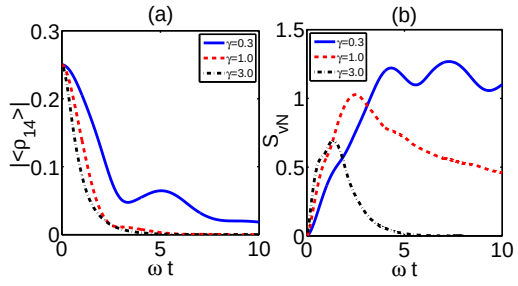

FIG. 2. (Color online) Coherence and von Neumann entropy [42] of a dissipative four-level model: $H_{\rm sys} = \omega J_z$, and $L = J_-$, and $l = 3/2$. Initially, $\psi_0 = (1/2)(|1\rangle + |2\rangle + |3\rangle + |4\rangle)$. The time evolution of coherence $|\langle\rho_{14}\rangle|$ and entropy $S_{\rm vN} = -\rho \log_2 \rho$ is averaged over 1000 trajectories for different values of γ ($\Gamma = \omega$) with the first-order noise term.

It is known that the stochastic wave-function $\psi_t(z^*)$ in Eq. (3) does not conserve the norm of the wave-function. To efficiently simulate quantum open systems, one usually uses the nonlinear QSD equation for the normalized state $\tilde{\psi}_t = \frac{\psi_t}{||\psi_t||}$:

$$\frac{d}{dt}\tilde{\psi}_t = \big[-iH_{sys} + (L - \langle L\rangle_t)\tilde{z}_t^* + \langle L^\dagger\rangle_t \big(\bar{O}(t,\tilde{z}^*) - \langle \bar{O}(t,\tilde{z}^*)\rangle_t\big) - \big(L^\dagger \bar{O}(t,\tilde{z}^*) - \langle L^\dagger \bar{O}(t,\tilde{z}^*)\rangle_t\big)\big]\tilde{\psi}_t, \quad (21)$$

where $\langle L\rangle_t = \langle\tilde{\psi}_t|L^\dagger|\tilde{\psi}_t\rangle$ and $\tilde{z}_t^* = z_t^* + \int_0^t \alpha^*(t,s)\langle L^\dagger\rangle_s ds$ is the shift complex Gaussian process.

The non-Markovian QSD approach is valid for an arbitrary correlation function. For simplicity and considering Markov limit, the non-Markovian effect is modeled by the Lorentz spectral density: $S(\omega) = \frac{1}{2\pi}\frac{\Gamma\gamma^2}{\gamma^2+\omega^2}$. Then the correlation function obtained from the Fourier transformation is given by $\alpha(t,s) = \int_0^\infty d\omega S(\omega) e^{-i\omega(t-s)} = \frac{\Gamma\gamma}{2} e^{-\gamma|t-s|}$, where $1/\gamma$ is an important non-Markovian parameter representing the memory time of the environment. When $\gamma \to \infty$, the correlation function approaches the Markov limit with $\alpha(t,s) \to \Gamma\delta(t-s)$.

As our first example, we consider the numerical simulation of the dissipative dynamics of a four-level system with the O-operator given by Eq. (9). In the case of dissipative bath at the zero temperature, the quantum coherence of the four-level system will decay and the populations of the excited levels will be transferred to the ground state with time. When the bath is in a non-Markovian regime with $\gamma = 0.3$, it is shown that the decoherence dynamics deviates from the exponential decay of the Markov case (See the bump of ρ_{14} in Fig. 2(a)). Consequently, the decoherence is delayed as the entanglement between system and bath builds up. We also plot the von Neumann entropy [$S_{\rm vN} = -{\rm Tr}(\rho \ln \rho)$] (setting the Boltzmann constant $k_B = 1$) in Fig. 2(b). When the parameter γ increases to 1.0 (moderate non-Markovian regime) and 3.0 (near-Markov regime), the coherence quickly decays to zero at the time point ($\omega t \simeq 5$). However, we see that their entropy curves are different. For example, when $\gamma = 3.0$, $S_{\rm vN}$ approaches zero at $\omega t \simeq 5$, which means that the decoherence time between the ground state and the highest energy level and purification time (That is, $S_{\rm vN} \simeq 0$) coincide. Interestingly, we can see the $S_{\rm vN}$ is significantly modified by the non-Markovian effect with $\gamma \leqslant 1$.

In Fig. 3, we show the dynamics of a driven four-level atomic system coupled to a dissipative environment with different γ's. When $t = 0$, the atom is totally populated at the highest level $|3\rangle$. With the modulation of the driving fields $\Delta_2(t)$ and $\Delta_4(t)$, Level 2 and Level 4 will be coupled to Level 3, which helps to establish the coherence terms ρ_{23} and ρ_{34}. Those coherence terms will decay due to the dissipative channels to Level 1. From Fig. 3(a) and Fig. 3(b), when $\gamma = 0.3$ the non-Markovian environment with long memory time clearly increases the magnitude of the coherence terms. It should be noted that the differences between $\gamma = 1.0$ and $\gamma = 3.0$ are not significant since the system is relatively close to the Markov regimes. We can also see from Fig. 3(c) and Fig. 3(d),

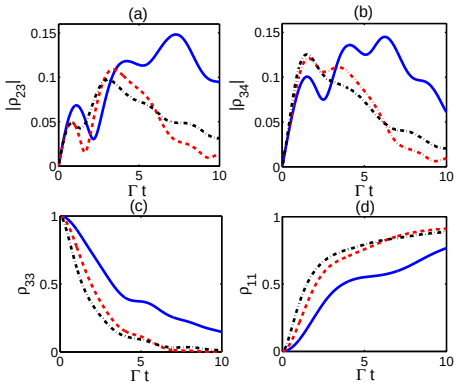

FIG. 3. (Color online) Coherence and population dynamics of a dissipative four-level model driven by two external fields. The model is described by Fig. 1 and Eq. (15). The parameters are chosen as follows: $\omega_1 = 0.1\Gamma$, $\omega_2 = 0.3\Gamma$, $\omega_3 = 0.6\Gamma$, $\omega_4 = 0.2\Gamma$, $\Delta_2(t) = 0.1\Gamma e^{2it}T_{23} + h.c.$, $\Delta_4(t) = 0.1\Gamma e^{2it}T_{34} + h.c.$, $\kappa_2 = 0.4$, $\kappa_3 = 0.8$ and $\kappa_4 = 0.3$. Initially, we choose $\psi_0 = |3\rangle$. The time evolution of the reduced density matrix is obtained by averaging over 1000 trajectories for each γ obtained from the QSD equation with the first-order noise term: (i) $\gamma = 0.3$, the blue solid lines; (ii) $\gamma = 1.0$, the red dashed lines; (iii) $\gamma = 3.0$, the black dot-dashed lines.

that when $\gamma = 0.3$, the non-Markovian effect causes population fluctuation of Level 3 and Level 1. Typically, we see that the population transfer rates to the ground state are increased by the shorter environment memory time.

V. CONCLUSION

In summary, we have studied the non-Markovian dynamics of multilevel atomic systems using the non-Markovian quantum trajectory method. The time-local QSD equations are obtained by the explicit O-operator construction for several physically interesting models. As shown in this paper, the time-local O-operators allow numerical simulations to be implemented efficiently for multilevel open systems. For the multilevel atoms without driving fields, we show explicitly how to construct the exact O-operators containing a finite-order noise terms. For the atomic models with multi-transition channels including the models with driving external fields, we verify that the O-operator can take a noise-free form, so the exact QSD equations can be easily established. In the case that the O-operator contains no noise, the exact convolutionless master equation can be derived. The results of this paper will open am avenue to exploring novel and fascinating phenomena in multilevel atom-field interaction problems where the non-Markovian features are important.

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Appendix A: Proof for the O-operator in Dissipative High-Spin Models

This appendix provides the details of deriving the O-operator for a multilevel atom dissipative model in Eq. (8). The techniques with some necessary yet not complicated modifications can be used to derive the O-operators for a general multilevel models including the models with the external driving fields. For the high-spin model, the system Hamiltonian and the Lindblad operator may be generally rewritten as

$$H_{\rm sys} = \sum_{m=1}^{2l+1} C_m H_{\rm sys}^{(m)}, \quad H_{\rm sys}^{(m)} = |m\rangle\langle m|,$$
$$L = \sum_{n=2}^{2l+1} G_n L_n, \quad L_n = |n-1\rangle\langle n|. \tag{A1}$$

When C_m's and G_n's are chosen as the Clebsch-Gordan coefficients, these operators are referred to the angular momentum operators of spin-l. In practice, Eq. (A1) can be generalized to a general dissipative model for a multilevel atom with an arbitrary energy distribution. The model can also relaxed to the case containing time-dependent damping coefficients.

In what follows, we will show that there are $(2l-k)$ terms containing kth-order noise ($k = 0, \cdots, 2l-1$) in the O-operator. Explicitly, $O(t,s,z^*)$ may be expanded as

$$O = \sum_{j=1}^{2l} f_j(t,s) O_j^{(0)} + \sum_{j=1}^{2l-1} \int_0^t p_j^{(1)}(t,s,s_1) z_{s_1}^* ds_1 O_j^{(1)}$$
$$+ \cdots + \int_0^t \cdots \int_0^t p_1^{(2l-1)}(t,s,s_1,\cdots,s_{2l-1}) z_{s_1}^* \cdots$$
$$z_{s_{2l-1}}^* ds_1 ds_2 \cdots ds_{2l-1} O_1^{(2l-1)}, \tag{A2}$$

where the coefficients of $O_j^{(k)}$ $(1 \leqslant j \leqslant 2l-k)$ are symmetric functions of $s_1, \ldots s_k$. Subsequently,

$$\bar{O} = \sum_{j=1}^{2l} F_j(t) O_j^{(0)} + \sum_{j=1}^{2l-1} \int_0^t P_j^{(1)}(t,s_1) z_{s_1}^* ds_1 O_j^{(1)}$$
$$+ \cdots + \int_0^t \cdots \int_0^t P_1^{(2l-1)}(t,s_1,\cdots,s_{2l-1}) z_{s_1}^* \cdots$$
$$z_{s_{2l-1}}^* ds_1 ds_2 \cdots ds_{2l-1} O_1^{(2l-1)}. \tag{A3}$$

In accord with definition, $F_j(t) \equiv \int_0^t ds\alpha(t,s)f_j(t,s)$ and $P_j^{(k)}(t,s_1,\cdots,s_k) \equiv \int_0^t ds\alpha(t,s)p_j^{(k)}(t,s,s_1,\cdots,s_k)$, $k = 1,\cdots,2l-1$.

Clearly, the operators $O_j^{(k)}$ form a set of basis operators:

$$O_j^{(k)} = |j\rangle\langle j+k+1|. \tag{A4}$$

Hence the total number of the basis operators $O_j^{(k)}$ is $(2l+1)l$. In fact, any linear combinations of operators in Eq. (A4) may be used to construct the O-operator as long as they are linear-independent and satisfy Eq. (4). Here we use Eq. (A4) for the simplicity in the following proof. More often the notation $O_j^{(k)} = J_z^{j-1}J_-^{k+1}$ has a more transparent meaning for the angular momentum model.

By substituting Eq. (A1) into Eq. (4), the left hand side of Eq. (4) becomes

$$\begin{aligned}
&\sum_{j=1}^{2l}\frac{\partial}{\partial t}f_j(t,s)O_j^{(0)} + \sum_{j=1}^{2l-1}\Big[\int_0^t \frac{\partial}{\partial t}p_j^{(1)}(t,s,s_1)z_{s_1}^* ds_1 \\
&+ z_t^* p_j^{(1)}(t,s,t)\Big]O_j^{(1)} + \cdots + \Big[\int_0^t\cdots\int_0^t \frac{\partial}{\partial t} \\
&\quad p_1^{(2l-1)}(t,s,s_1,\cdots,s_{2l-1})z_{s_1}^*\cdots z_{s_{2l-1}}^* ds_1\cdots ds_{2l-1} \\
&+ (2l-1)z_t^*\int_0^t\cdots\int_0^t p_1^{(2l-1)}(t,s,s_1,\cdots,s_{2l-2},t) \\
&\times z_{s_1}^*\cdots z_{s_{2l-2}}^* ds_1\cdots ds_{2l-2}\Big]O_1^{(2l-1)}.
\end{aligned} \tag{A5}$$

The right hand side of the equation consists of four terms. It can be easily seen that each of them can be expressed as a linear combination of the operators given in Eq. (A4). The construction of the O-operator can be summarized in the following four crucial observations:

(i) $[-iH_{\rm sys}, O(t,s,z^*)]$ consisting of $[H_{\rm sys}^{(m)}, O_j^{(k)}]$ can be decomposed as

$$|j\rangle\langle j+k+1|(\delta_{j,m}-\delta_{j+k+1,m}) = O_j^{(k)}(\delta_{j,m}-\delta_{j+k+1,m}). \tag{A6}$$

(ii) $[Lz_t^*, O(t,s,z^*)]$ consisting of the noise term $z_t^*[L_n, O_j^{(k)}]$ turns out to be

$$\begin{aligned}
&z_t^*(|j-1\rangle\langle j+k+1|\delta_{j,n} - |j\rangle\langle j+k+2|\delta_{j+k+1,n-1}) \\
&= z_t^*(O_{j-1}^{(k+1)}\delta_{j,n} - O_j^{(k+1)}\delta_{j+k,n-2}).
\end{aligned} \tag{A7}$$

These terms correspond to those with z_t^* in Eq. (A5), which will appear in the boundary conditions between f_j's and $p_j^{(k)}$'s. When n in L_n runs from 2 to $2l+1$, $O_j^{(k+1)}$'s will present themselves successively with j in $O_j^{(k)}$ running from 1 to $2l-k$. It is also a necessary requirement for the initial condition of O-operator: $O(s,s,z^*) = L = \sum_{n=2}^{2l+1} G_n L_n$.

(iii) $[-L^\dagger\bar{O}, O]$ is consisted by $[O_j^{(k)}, |n\rangle\langle n-1|O_{j'}^{(k')}]$, which turns out to be

$$O_j^{(k+k')}\delta_{j+k,j'} - O_{j'+1}^{(k+k')}\delta_{j'+k'+1,j}. \tag{A8}$$

(iv) For $-L^\dagger\frac{\delta\bar{O}(t,z^*)}{\delta z_s^*}$, we only need to consider the terms with $k \geqslant 1$. The differential functional leaves the operator unchanged. Then one typical component is

$$-L_n^\dagger O_j^{(k)} = -|j+1\rangle\langle j+k+1|\delta_{n,j+1} = -O_{j+1}^{(k-1)}. \tag{A9}$$

It contributes to the presence of $O_j^{(0)}$'s.

To summarize the last four steps, we have proved the existence and constitutions of the O-operator for the angular momentum model of the multi-level atom. By introducing Eq. (A4), the consistency condition equation (4) is shown to be consistent and complete.

Now we consider a general four-level atom dissipative model with $l = 3/2$ in Eq. (A1). The group of closed differential equations for the coefficients can be derived through the above analysis. They are given by,

$$\begin{aligned}
\partial_t f_1 &= -i(C_1-C_2)f_1 + G_2^* f_1 F_1, \\
\partial_t f_2 &= -i(C_2-C_3)f_2 + G_3^* f_2 F_2 - G_2^* f_2 F_1 - G_2^* P_1^{(1)}, \\
\partial_t f_3 &= -i(C_3-C_4)f_3 + G_4^* f_3 F_3 - G_3^* f_3 F_2 - G_3^* P_2^{(1)}, \\
\partial_t p_1^{(1)} &= -i(C_1-C_3)p_1^{(1)} + G_2^* f_1 P_1^{(1)} + G_3^* F_2 p_1^{(1)}, \\
\partial_t p_2^{(1)} &= -i(C_2-C_4)p_2^{(1)} + G_3^* f_2 P_2^{(1)} + G_4^* F_3 p_2^{(1)} \\
&\quad - G_2^* F_1 p_2^{(1)} - G_2^* f_3 P_1^{(1)} - 2G_2^* P_1^{(2)}, \\
\partial_t p_1^{(2)} &= -i(C_1-C_4)p_1^{(2)} + G_2^* f_1 P_1^{(2)} + G_3^* P_2^{(1)} p_1^{(1)} \\
&\quad + G_4^* F_3 p_1^{(2)}.
\end{aligned} \tag{A10}$$

Here the boundary conditions are,

$$\begin{aligned}
2p_1^{(2)}(t,s,s_1,t) &= G_2 p_2^{(1)}(t,s,s_1) - G_4 p_1^{(1)}(t,s,s_1), \\
p_1^{(1)}(t,s,t) &= G_2 f_2(t,s) - G_3 f_1(t,s), \\
p_2^{(1)}(t,s,t) &= G_3 f_3(t,s) - G_4 f_2(t,s).
\end{aligned} \tag{A11}$$

The spin-3/2 dissipative model [See the O-operator in Eq. (9)] can be solved by setting $C_m = -l-1+m$, $m = 1,2,3,4$ and $G_2 = G_4 = \sqrt{3}$, $G_3 = 2$.

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