Exact homogeneous master equation for open quantum systems incorporating initial correlations

Pei-Yun Yang¹ and Wei-Min Zhang^{1,*}

¹Department of Physics and Center for Quantum Information Science, National Cheng Kung University, Tainan 70101, Taiwan

We show that the exact master equation incorporating initial correlations for open quantum systems, within the Nakajima-Zwanzig operator-projection method, is a homegenous master equation for the reduced density matrix. We also using the quantum Langevin equation to derive explicitly the exact master equation incorporating initial correlations for a large class of bosonic and fermionic open quantum systems described by the Fano-Anderson Hamiltonian, and the resulting master equation is homogenous. We find that the effects of the initial correlations can be fully embedded into the fluctuation dynamics through the exact homogenous master equation. Also a generalized nonequilibrium fluctuation-dissipation theorem incorporating various initial correlations is obtained.

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Realistic systems in nature have inevitable interactions with its surrounding environments. When such interactions are not negligible, the systems must be treated as an open system [1]. Many physical, chemical and biological systems, and all kinds of quantum devices developed in recent years for nano and quantum technologies. should be considered as open quantum systems. In contrast to an isolated quantum system, whose dynamics is governed by the Schrödinger equation, the dynamical evolution of an open quantum system can be described by the master equation [2, 3]. Formally, the exact master equation for arbitrary open system is obtainable through the operator-projection method which was initially proposed by Nakajima [4] and Zwanzig [5]. When there are initial correlations between the system and its environment, it was claimed [2] that, the Nakajima-Zwanzig (NZ) equation becomes inhomogeneous. However, if a maser equation for open systems is inhomogeneous, it may violate the trace-preserving property of the density matrix and thereby induce unphysical states to the open system dynamics. In this work, we prove that the exact master equation is always homogeneous for any open quantum system incorporating arbitrary initial correlations.

In fact, nonequilibrium dynamics of open systems incorporating initial correlations is a long-standing problem in mesoscopic physics and also in the foundation of statistical mechanics [6, 7]. In the past two decades, investigation of quantum nonequilibrium dynamics has mainly been focused on steady-state phenomena [8, 9], where initial correlations are usually considered to be not essential due to the memory loss and therefore are often ignored. Recent experimental developments allow one to measure transient quantum dynamics of open systems, in particular, for nano-scale quantum devices and various atomic and molecular structure-based biological systems [10–14]. In the transient nonequilibrium regime, initial correlations could provide important information on the formation and evolution of a large number of different physical objects. In this work, we will also derive explicitly the exact homogeneous master equation for a large class of open systems incorporating initial correlations.

1. NZ master equation is homogenous. The NZ equation was originally proposed to describe the dynamics of a subspace of interest, projected out from the whole Hilbert space $\mathcal E$ of a large system [4, 5]. It begins with a decomposition of the Hilbert space into a relevant part plus an irrelevant part,

$$\rho_{tot}(t) = \mathcal{P}\rho_{tot}(t) + \mathcal{Q}\rho_{tot}(t), \tag{1}$$

where \mathcal{P} and \mathcal{Q} are the two projection operators that satisfy the projection operator properties: $\mathcal{P} + \mathcal{Q} = I$, $\mathcal{P}^2 = \mathcal{P}$, $\mathcal{Q}^2 = \mathcal{Q}$, and $\mathcal{P}\mathcal{Q} = \mathcal{Q}\mathcal{P} = 0$. From the quantum Liouvillian equation of the system, $\frac{d}{dt}\rho_{tot}(t) = -i[H_{tot}, \rho_{tot}(t)] \equiv \mathcal{L}(t)\rho_{tot}(t)$, one can formally derive the following NZ equation for the relevant subspace of the system,

$$\frac{d}{dt} \mathcal{P}\rho_{tot}(t) = \mathcal{P}\mathcal{L}(t)\mathcal{P}\rho_{tot}(t) + \mathcal{P}\mathcal{L}(t)\mathcal{G}(t, t_0)\mathcal{Q}\rho_{tot}(t_0)
+ \int_{t_0}^t d\tau \mathcal{P}\mathcal{L}(t)\mathcal{G}(t, \tau)\mathcal{Q}\mathcal{L}(\tau)\mathcal{P}\rho_{tot}(\tau), \quad (2)$$

where $\mathcal{G}(t,\tau)$ is an evolution operator given by

$$G(t,\tau) = T \exp \left\{ \int_{\tau}^{t} ds Q \mathcal{L}(s) \right\},$$
 (3)

and \mathcal{T} is the time-ordering operator. In Ref. [2], it is claimed that the second term in Eq. (2), which is obtained from the initial system-environment correlations, is an inhomogeneous term to the master equation of open systems. Only when there is no initial system-environment correlation, the second term vanishes, and the corresponding master equation becomes a homogeneous master equation [2]. We will first show that this claim is incorrect.

To be explicit, let us focus on the large system consisting of a subsystem of interest (refereed to the principal system [1], i.e. the open system hereafter) coupling to the rest as its environment, then by specifying the projection operator,

$$\mathcal{P}\rho_{tot} \equiv \text{Tr}_E[\rho_{tot}] \otimes \text{Tr}_S[\rho_{tot}] = \rho_S \otimes \rho_E, \tag{4}$$

one projects the entire Hilbert space of the system into the subspace written apparently as direct products of the density matrices of the open system with its environment. Notice that Tr_E and Tr_S are traces over separately the environmental states and the system states. Thus, $\rho_S = \text{Tr}_E[\rho_{tot}]$ and $\rho_E = \text{Tr}_S[\rho_{tot}]$ are the corresponding reduced density matrices describing respectively quantum states of the open system and the environment that mutually carry the entanglement information between the system and the environment. The left part of the Hilbert space is $\mathcal{Q}\rho_{tot} = \rho_{tot} - \rho_S \otimes \rho_E$. By taking a trace over all the environmental states on the both sides of Eq. (2), the NZ equation is reduced to the master equation describing the time evolution of the reduced density matrix $\rho_S(t)$ for the open system.

Now, we can show that the second term in Eq. (2) is not an inhomogeneous term to the master equation for the reduced density matrix $\rho_S(t)$. Note that the second term in Eq. (2), $\mathcal{PL}(t)\mathcal{G}(t,t_0)\mathcal{Q}\rho_{tot}(t_0)$ must be a density matrix belonging to the same subspace of $\mathcal{P}\rho_{tot}(t)$ due to the project operator \mathcal{P} . Then the trace over the environment states on the second term in Eq. (2) gives a reduced density matrix $\rho'_S(t)$ which could be (and usually is) different from $\rho_S(t)$:

$$\operatorname{Tr}_{E}[\mathcal{PL}(t)\mathcal{G}(t,t_{0})\mathcal{Q}\rho_{tot}(t_{0})] \equiv \rho_{S}'(t) \neq \rho_{S}(t). \tag{5}$$

However, $\rho_S'(t)$ and $\rho_S(t)$ are two different reduced density matrices of the open system, it is *always* possible to connect $\rho_S'(t)$ with $\rho_S(t)$ by superoperators, i.e.

$$\rho_S'(t) \longrightarrow \sum_{ij} \chi_{ij}(t) A_j \rho_S(t) B_i,$$
 (6)

where $\chi_{ij}(t)$ is in general a time-dependent coefficient that depends on the initial system-environment correlations, and A_i , B_j are operators of the open system. More specifically, these superoperators can be written with the following symmetric and trace-preserving form:

$$\rho_S'(t) \longrightarrow \sum_{ij} \chi_{ij}(t) \left[A_j \rho_S(t) B_i + B_i \rho_S(t) A_j - A_j B_i \rho_S(t) - \rho_S(t) B_i A_j \right], \quad (6')$$

Consequently, it shows that the second term in Eq. (2) can always be reduced to an homogeneous term to the master equation.

In fact, the second and the third terms in the NZ equation (2) have the same operator structure. This can be seen explicitly by writing $\mathcal{L}(\tau)\mathcal{P}\rho_{tot}(\tau) \equiv \rho''_{tot}(\tau)$, as another state of the total system. Then the integration function of the non-local time integral in the third term of Eq. (2) can be rewritten as

$$\mathcal{PL}(t)\mathcal{G}(t,\tau)\mathcal{QL}(\tau)\mathcal{P}\rho_{tot}(\tau) = \mathcal{PL}(t)\mathcal{G}(t,\tau)\mathcal{Q}\rho_{tot}''(\tau), \tag{7}$$

which shows the same operator structure as the second term, $\mathcal{PL}(t)\mathcal{G}(t,t_0)\mathcal{Q}\rho_{tot}(t_0)$ in Eq. (2). Thus, after taken the trace over all the environment states, the second term in the NZ equation (2) must produce a homogenous term to the master equation as does the third

term. In other words, initial correlations *cannot* produce an inhomogeneous term to the master equation for open systems. The claim in Ref. [2] that initial correlations result in an inhomogeneous master equation cannot be correct.

2. Derivation of the exact homogenous master equation for the Fano-Anderson model. On the other hand, using cumulant expansions [15] rather than the NZ operator-projection methods, it was formally shown that the reduced density matrix $\rho_S(t)$ obeys an exact homogeneous evolution equation when the initial systemenvironment correlations are considered [16]. Recently, explicit exact homogeneous master equations are derived for the pure dephasing model of a two-level system [17], for the nano-cavity system in photonic crystals [18], and for the nano-structured electronic systems in mesoscopic physics [19], where initial correlations between the system and its reservoirs are taken into account. Here we shall derive further an exact homogeneous master equation incorporating initial system-environment correlations for a large class of open systems investigated widely in condensed mater physics and atomic physics.

These open systems we concerned here are described by the Fano-Anderson Hamiltonian,

$$H^{FA}(t) = H_S(t) + H_E(t) + H_{SE}(t)$$

$$= \sum_{ij} \varepsilon_{ij}(t) a_i^{\dagger} a_j + \sum_{\alpha k} \epsilon_{\alpha k}(t) b_{\alpha k}^{\dagger} b_{\alpha k}$$

$$+ \sum_{\alpha ik} \left(V_{i\alpha k}(t) a_i^{\dagger} b_{\alpha k} + V_{i\alpha k}^*(t) b_{\alpha k}^{\dagger} a_i \right), \quad (8)$$

as it was originally introduced by Anderson [20] and Fano [21] independently for investigating impurity electrons in solid-state physics and discrete states embedded in a continuum in atomic physics, respectively. In Eq. (8), we let all the energy levels and couplings be time-dependent because these parameters can be manipulated experimentally through the new development of nanotechnologies in the investigation of nonequilibrium dynamics. In the past two decades, the Fano-Anderson Hamiltonian has often been used as a starting point to investigate various nonequilibrium transport phenomena and decoherence dynamics in semiconductor nanoelectronics and spintronics [6, 9, 23]. The Fano-Anderson Hamiltonian was also referred as the Lee-Friedrichs Hamiltonian [24, 25] in atomic and molecular physics, nuclear physics and scalar field theory with the physcial relevance of Fano resonances, Dicke effect and superradiance, etc.

Let us consider the first case for Eq. (8), in which the total system is initially in equilibrium, which is called the partition-free scheme in the literature [26, 27]:

$$\rho_{tot}(t_0) = \frac{1}{Z} e^{-\beta(H^{FA}(t_0) - \mu N)}.$$
 (9)

Obviously, this initial state contains initial systemenvironment correlations. In other words, $Q\rho_{tot}(t_0) \neq 0$ in the NZ project method so that the second term in Eq. (2), regarded as an inhomogeneous term in [2], does not vanish. Meantime, by experimentally tuning on the time-dependent parameters in Eq. (8) through external fields, the total system becomes nonequilibrium, and the time evolution of the total density matrix is determined by

$$\rho_{tot}(t) = U(t, t_0)\rho_{tot}(t_0)U^{\dagger}(t, t_0), \tag{10}$$

where $U(t,t_0) = \mathcal{T} \exp \left\{ -i \int_{t_0}^t d\tau H^{FA}(\tau) \right\}$ is the evolution operator of the total system. Because $H^{FA}(t)$ in Eq. (8) has a bilinear form of the particle creation and annihilation operators, the total density matrix $\rho_{tot}(t)$ will remain as a Gaussian function all the time. Taking the trace over the environment states (e.g. by path-integral method with the Gaussian integral), the reduced density matrix $\rho_S(t) = \text{Tr}_E[\rho_{tot}(t)]$ is still a Gaussian function of the system variables only, with a prefactor which is only a function of time. Then the time-derivative of the reduced density matrix must be proportional to the same Gaussian function. Consequently, the resulting master equation must be homogeneous.

Actually, using the Feynman-Vernon influence functional approach [1], we have previously derived the exact homogenous master equation of the reduced density matrix $\rho_S(t)$ from Eq. (10), for both boson and fermion systems in the absence of initial system-environment correlation [28–31], where the initial state of the system and the environment is assumed to be decoupled [32]. In this exact master equation formalism, the environmentinduced energy-level renormalization, the particle dissipations, and thermal fluctuations, due to the influence of the environment, are naturally incorporated into the time-dependent dissipation and fluctuations coefficients in the exact master equation. These damping and fluctuation coefficient are determined exactly and nonperturbatively by the Schwinger-Keldysh nonequilibrium Green's functions in many-body systems [33, 34]. By solving these nonequilibrium Green's functions, we investigated nonperturbatively various environment-induced non-Markovian dynamics through the time evolution of open quantum systems, in the absence of initial systemenvironment correlations [30].

Now, to derive the exact master equation incorporating the initial system-environment correlations, we utilize the exact quantum Langevin equation from Eq. (8) [18, 19, 36],

$$\frac{d}{dt}a_{i}(t) = -i\sum_{j} \boldsymbol{\varepsilon}_{ij}(t)a_{j}(t) - \sum_{j} \int_{t_{0}}^{t} d\tau \boldsymbol{g}_{ij}(t,\tau)a_{j}(\tau) + f_{i}(t),$$
(11)

where

$$\mathbf{g}_{ij}(t,\tau) = \sum_{\alpha k} V_{i\alpha k}(t) V_{j\alpha k}^*(\tau) e^{-i\int_{\tau}^{t} \epsilon_{\alpha k}(\tau_1) d\tau_1}, \quad (12a)$$

$$f_i(t) = -i \sum_{\alpha k} V_{i\alpha k}(t) b_{\alpha k}(t_0) e^{-i \int_{t_0}^t \epsilon_{\alpha k}(\tau_1) d\tau_1}, \quad (12b)$$

which is obtained by eliminating *exactly* all the environmental degrees of freedom through the Heisenberg equation of motion, and it is valid for arbitrary initial state of the system and the environment. Because of the

linearity, the general solution of Eq. (11) has the form [18, 19, 36]

$$a_i(t) = \sum_j \mathbf{u}_{ij}(t, t_0) a_j(t_0) + F_i(t),$$
 (13)

where $u_{ij}(t,t_0) = \langle [a_i(t), a_j^{\dagger}(t_0)] \rangle$ is the propagating Green function of the particles in the system, which carries all the information of the dissipation dynamics of the system induced by the tunneling coupligs between the system and the environment, and $F_i(t)$ is the fluctuation function resulting from the noise force $f_i(t)$ of the environment, see Eqs. (11) and (12b).

It is easy to show from Eqs. (11) and (13) that the propagating Green function obeys the Dyson equation,

$$\frac{d}{dt}\boldsymbol{u}(t,t_0) + i\boldsymbol{\varepsilon}(t)\boldsymbol{u}(t,t_0) + \int_{t_0}^{t} d\tau \boldsymbol{g}(t,\tau)\boldsymbol{u}(\tau,t_0) = 0, \quad (14)$$

subjected to the initial condition $u(t_0, t_0) = 1$, where the time non-local integral kernel, $g(t, \tau)$ given by Eq. (12a), describes the probability amplitude of a particle tunneling from the system into the environment at time τ , propagating in the environment within the time τ to t, and then tunneling back into the system at time t. Thus, the τ -integration from the very initial time t_0 to the present time t through this non-local time integral kernel in Eq. (14) records all the historical evolution of the particle dissipated from the system into the environment. This characterizes the microscopic picture of the non-Markovian memory processes [30]. The fluctuation function $F_i(t)$, a contribution from the inhomogeneous color-noise force $f_i(t)$ in the quantum Langevin equation (11), is given explicitly by

$$F_{i}(t) = -i \sum_{\alpha k j} \int_{t_{0}}^{t} d\tau \left[\mathbf{u}_{ij}(t,\tau) V_{j\alpha k}(\tau) e^{-i \int_{t_{0}}^{\tau} \epsilon_{\alpha k}(\tau_{1}) d\tau_{1}} \right] c_{\alpha k}(t_{0}), \tag{15}$$

which describes particles tunneling from the environment into the system at time τ , and then propagating to the level i during the time from τ to t. This provides the underlying fluctuation dynamics induced by particles initially in the environment. The initial operator-dependence in Eqs. (13) and (15), proportional to $a_i(t_0)$ and $c_{\alpha k}(t_0)$ respectively, fully incorporate all the initial correlations of the total system.

Combining the dissipation and fluctuation dynamics together through the above exact quantum Langevin equation, we obtain the exact homogenous master equation for the reduced density matrix of the system incorporating various initial correlations:

$$\frac{d\rho_S(t)}{dt} = -i \left[H_S'(t), \rho_S(t) \right] + \sum_{ij} \left\{ \gamma_{ij}(t) \mathcal{D}(a_j, a_i^{\dagger}) + \widetilde{\gamma}_{ij}(t) \mathcal{F}(a_j, a_i^{\dagger}) + \overline{\gamma}_{ij}(t) \mathcal{F}(a_j, a_i^{\dagger}) + \overline{\gamma}_{ij}^{\dagger}(t) \mathcal{F}(a_j, a_i) \right\},$$
(16)

The renormalized Hamiltonian $H'_S = \sum_{ij} \varepsilon'_{ij}(t) a_i^{\dagger} a_j$,

and the dissipation and fluctuation superoperators:

$$\mathcal{D}(a_{j}, a_{i}^{\dagger}) = 2a_{j}\rho_{S}(t)a_{i}^{\dagger} - a_{i}^{\dagger}a_{j}\rho_{S}(t) - \rho_{S}(t)a_{i}^{\dagger}a_{j}, \qquad (17a)$$

$$\mathcal{F}(a_{j}, a_{i}^{\dagger}) = a_{i}^{\dagger}\rho_{S}(t)a_{j} \pm a_{j}\rho_{S}(t)a_{i}^{\dagger} \mp a_{i}^{\dagger}a_{j}\rho_{S}(t) - \rho_{S}(t)a_{j}a_{i}^{\dagger}. \qquad (17b)$$

where the up and low signs of \pm correspond to both the system and reservoirs consisting of bosons or fermions. The renormalized energy $\varepsilon'_{ij}(t)$, the dissipation and fluctuation coefficients, $\gamma_{ij}(t)$, $\tilde{\gamma}_{ij}(t)$ and $\overline{\gamma}_{ij}(t)$ are fully determined by the following relations,

$$\boldsymbol{\varepsilon}'_{ij}(t) = \frac{i}{2} \left[\dot{\boldsymbol{u}}(t, t_0) \boldsymbol{u}^{-1}(t, t_0) - \text{H.c.} \right]_{ij}, \tag{18a}$$

$$\gamma_{ij}(t) = -\frac{1}{2} [\dot{\boldsymbol{u}}(t, t_0) \boldsymbol{u}^{-1}(t, t_0) + \text{H.c.}]_{ij},$$
 (18b)

$$\widetilde{\gamma}_{ij}(t) = \dot{\boldsymbol{v}}_{ij}(t,t) - [\dot{\boldsymbol{u}}(t,t_0)\boldsymbol{u}^{-1}(t,t_0)\boldsymbol{v}(t,t) + \text{H.c.}]_{ij},$$
(18c)

$$\overline{\gamma}_{ij}(t) = \mp \frac{1}{2} \dot{\boldsymbol{\nu}}_{ij}(t,t) \pm \frac{1}{2} [\dot{\boldsymbol{u}}(t,t_0) \boldsymbol{u}^{-1}(t,t_0) \boldsymbol{\nu}(t,t) + \mathbf{T}]_{ij}$$
(18d)

where $\boldsymbol{u}(t,t_0)$ is the propagating Green function of Eq. (14), and $\mathbf{v}(t,t)$ and $\mathbf{v}(t,t)$ are the particle-particle and particle-pair correlation Green functions in nonequilibrium many-body systems,

$$\boldsymbol{v}(\tau,t) = \int_{t_0}^{\tau} d\tau_1 \int_{t_0}^{t} d\tau_2 \boldsymbol{u}(\tau,\tau_1) \widetilde{\boldsymbol{g}}(\tau_1,\tau_2) \boldsymbol{u}^{\dagger}(t,\tau_2), \qquad (19a)$$

$$\boldsymbol{\nu}(\tau,t) = \int_{t_0}^{\tau} d\tau_1 \int_{t_0}^{t} d\tau_2 \boldsymbol{u}(\tau,\tau_1) \overline{\boldsymbol{g}}(\tau_1,\tau_2) \boldsymbol{u}^T(t,\tau_2), \quad (19b)$$

which are directly proportional to the initial correlations through the non-local time integral kernels, and $\widetilde{q}(\tau_1, \tau_2)$ and $\overline{\boldsymbol{g}}(\tau_1, \tau_2)$,

$$\widetilde{\mathbf{g}}_{ij}(\tau_1, \tau_2) = \widetilde{\mathbf{g}}_{ij}^{sb}(\tau_1, \tau_2) + \widetilde{\mathbf{g}}_{ij}^{bb}(\tau_1, \tau_2), \tag{20a}$$

$$\overline{\boldsymbol{g}}_{ij}(\tau_1, \tau_2) = \overline{\boldsymbol{g}}_{ij}^{sb}(\tau_1, \tau_2) + \overline{\boldsymbol{g}}_{ij}^{bb}(\tau_1, \tau_2), \tag{20b}$$

with

$$\widetilde{\mathbf{g}}_{ij}^{sb}(\tau_{1},\tau_{2}) = -2i\sum_{\alpha k} \left[V_{i\alpha k}(\tau_{1})e^{-i\int_{t_{0}}^{\tau_{1}}\epsilon_{\alpha k}(\tau)d\tau}\delta(\tau_{2}-t_{0}) \right] \times \langle a_{j}^{\dagger}(t_{0})b_{\alpha k}(t_{0})\rangle - \mathrm{H.c}(i\leftrightarrow j,\tau_{1}\leftrightarrow\tau_{2}) , \quad (21a)$$

$$\widetilde{\mathbf{g}}_{ij}^{bb}(\tau_{1},\tau_{2}) = \sum_{\alpha k\alpha'k'} V_{i\alpha k}(\tau_{1})e^{-i\int_{t_{0}}^{\tau_{1}}\epsilon_{\alpha k}(\tau)d\tau}V_{j\alpha'k'}^{*}(\tau_{2}) \times e^{i\int_{t_{0}}^{\tau_{2}}\epsilon_{\alpha'k'}(\tau)d\tau}\langle b_{\alpha'k'}^{\dagger}(t_{0})b_{\alpha k}(t_{0})\rangle, \quad (21b)$$

$$\overline{\mathbf{g}}_{ij}^{sb}(\tau_{1},\tau_{2}) = -2i\sum_{\alpha k} \left[V_{i\alpha k}(\tau_{1})e^{-i\int_{t_{0}}^{\tau_{1}}\epsilon_{\alpha k}(\tau)d\tau}\delta(\tau_{2}-t_{0}) \times \langle a_{j}(t_{0})b_{\alpha k}(t_{0})\rangle \pm (i\leftrightarrow j,\tau_{1}\leftrightarrow\tau_{2}) \right], \quad (21c)$$

$$\overline{\mathbf{g}}_{ij}^{bb}(\tau_{1},\tau_{2}) = -\sum_{\alpha k\alpha'k'} V_{i\alpha k}(\tau_{1})e^{-i\int_{t_{0}}^{\tau_{1}}\epsilon_{\alpha k}(\tau)d\tau}V_{j\alpha'k'}(\tau_{2}) \times e^{-i\int_{t_{0}}^{\tau_{2}}\epsilon_{\alpha'k'}(\tau)d\tau}\langle b_{\alpha'k'}(t_{0})b_{\alpha k}(t_{0})\rangle. \quad (21d)$$

The integral kernels $\widetilde{g}^{sb}(\tau_1, \tau_2)$ and $\overline{g}^{sb}(\tau_1, \tau_2)$ are proportional to initial particle-particle and particle-pair correlations between the system and the environment,

(21d)

and $\tilde{g}^{bb}(\tau_1, \tau_2)$ and $\bar{g}^{bb}(\tau_1, \tau_2)$ are associated respectively with initial particle-particle correlations and particlepair correlations in the environment. The particle-pair correlations can exist when the system and the environment are initially in, for examples, a superconducting state (for electron systems) [37] or a squeezed state (for photon systems) [18]. In fact, because the Hamiltonian (8) is bilinear, other higher-order correlations can be uniquely determined in terms of these basic nonequilibrium two-time Green functions, $u(t,\tau)$, $v(\tau,t)$ and $\boldsymbol{\nu}(\tau,t)$.

3. Unification with the influence functional approach and the nonequilibrium Green function technique. When there are no initial particle-pair correlations, namely $\langle a_i(t_0)b_{\alpha k}(t_0)\rangle = 0$ and $\langle b_{\alpha'k'}(t_0)b_{\alpha k}(t_0)\rangle = 0$, we will have $\overline{g}(\tau_1, \tau_2) = 0$. As a result, $\nu(t, t) = 0$ and also $\overline{\gamma}(t) = 0$ so that the exact master equation (16) is reduced to the one incorporating initial particle-particle correlations only that we derived very recently [19]. If the initial system-environment correlations are also negligible, $\langle a_j^{\dagger}(t_0)b_{\alpha k}(t_0)\rangle \simeq 0$ and $\langle b_{\alpha k}^{\dagger}(t_0)a_i(t_0)\rangle \simeq 0$, namely, the system is initially decoupled from the environment [32], and meanwhile the environment is initially in the thermal equilibrium, i.e. $\langle b_{\alpha'k'}^{\dagger}(t_0)b_{\alpha k}(t_0)\rangle =$ $f(\varepsilon_{\alpha k}, T)\delta_{\alpha \alpha'}\delta_{kk'}$, then

$$\widetilde{\boldsymbol{g}}_{ij}(\tau,\tau') = \sum_{\alpha k} V_{i\alpha k}(\tau) V_{j\alpha k}^*(\tau') e^{-i\int_{\tau'}^{\tau} \epsilon_{\alpha k}(\tau_1) d\tau_1} f(\varepsilon_{\alpha k}, T)$$
(22)

where $f(\varepsilon_{\alpha k}, T) = \frac{1}{e^{\beta(\varepsilon_{\alpha k} - \mu_{\alpha})} \mp 1}$ is the initial Bose-Einstein (Fermi-Dirac) distribution function at temperature T for bosonic (fermionic) environments. The resulting exact master equation goes to the master equation of Eq. (2) in Ref. [30] for the case of the decoupled initial system-environment states that can be derived directly [28–31] using the Feynman-Vernon influence functional approach [1]. However, in all the cases, the exact master equation for open systems are always homogenous, no matter if there are initial correlations or not.

As a self-consistent check, we show further that both the inhomogenous quantum Langevin equation (11) and the homogenous master equation (16) give the same nonequilibrium lesser Green functions:

$$G_{ij}^{\leq}(\tau,t) = i\langle a_j^{\dagger}(t)a_i(\tau)\rangle$$

$$= \left[\boldsymbol{u}(\tau,t_0)\boldsymbol{G}^{\leq}(t_0,t_0)\boldsymbol{u}^{\dagger}(t,t_0) + i\boldsymbol{v}(\tau,t)\right]_{ij},$$
(23a)

$$\overline{G}_{ij}^{\leq}(\tau,t) = i\langle a_j(t)a_i(\tau)\rangle
= \left[\boldsymbol{u}(\tau,t_0)\overline{G}^{\leq}(t_0,t_0)\boldsymbol{u}^T(t,t_0) + i\boldsymbol{\nu}(\tau,t) \right]_{ij},$$
(23b)

This shows that the inhomogeneity in physical observables induced by initial correlations can be fully taken into account in the exact homogeneous master equation. In fact, the Keldysh's correlation Green function v(t,t)and the generalized particle-pair correlation $\nu(t,t)$ in Eq. (23), given explicitly by Eq. (19), provide indeed the generalized nonequilibrium quantum fluctuationdissipation theorem in the time domain. In the weak system-environment coupling regime, the nonequilibrium quantum fluctuation-dissipation theorem will be reduced to the equilibrium fluctuation-dissipation theorem in the steady-state limit [38] (a simple proof can be found in the Supplemental Material of Ref. 30). Furthermore, in the high temperature limit, the equilibrium fluctuation-dissipation theorem is simply reduced to the Einstein's fluctuation-dissipation theorem [39] which can also be derived from the classical Langevin equation [40]. This provides a consistent check of the theory.

In conclusion, we show that the Nakajima-Zwanzig master equation in terms of the reduced density matrix for open quantum systems incorporating initial correlations is always a homogenous equation. We also derived explicitly, through the exact quantum Langevin equation, the exact homogenous master equation incorporating initial correlations. This master equation can describe the noneqilibrium dynamics for a large class of open quantum systems based on the Fano-Anderson Hamiltonian which has widely been used in atomic physics, condensed matter physics and various nano-scale systems in the past decades. The resulting exact homogenous master equation recovers the master equation derived from Feynman-Vernon influencefunctional in the absence of the initial correlations as a special limit. The dissipation and fluctuation dynamics, embedded in the time-dependent coefficients of the exact homogenous master equation, which take into account all initial correlation effects, are fully determined by the generalized Schwinger-Keldysh nonequlibrium Green functions. Thus, for the very first time, by connecting the Nakajima-Zwanzig method, the quantum Langevin equation method, the Feynman-Vernon influence functional approach and the Schwinger-Keldysh nonequlibrium Green function technique together, we develop the most general exact homogenous master equation for a large class of open quantum systems. We expect that this underlying master equation incorporating initial correlations would find more applications for the investigations of nonequilibrium dynamics not only in physics, but also in chemical and biological systems through the exploration of general dissipation and fluctuation dynamics.

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- * Electronic address: wzhang@mail.ncku.edu.tw
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