Exact master equation for generalized quantum Brownian motion with momentum-dependent system-environment couplings

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In this paper we generalize the quantum Brownian motion (QBM) to include the momentum-dependent system-environment couplings. The resulting Hamiltonian in the particle number representation is given by $H_{\rm tot} = \hbar\omega_{\rm S} a^\dagger a + \sum_k \hbar\omega_k b_k^\dagger b_k + \sum_k \hbar(V_k a^\dagger b_k + V_k^* b_k^\dagger a) + \sum_k \hbar(W_k a^\dagger b_k^\dagger + W_k^* b_k a)$. The conventional QBM model corresponds to the special case $W_k = V_k$. The generalized QBM can capture more physical applications because the single-particle transition and the two-particle pair production between the system and the environment represent two very different physical processes that usually do not have the same coupling strengths. We discuss the physical realizations of the generalized QBM in different physical systems, and derive its exact master equation for both the initial decoupled states and initial correlated states. The Hu-Paz-Zhang master equation of the conventional QBM model is reproduced as a special case. We find that the renormalized system Hamiltonian after tracing out all the environmental states induced naturally a momentum-dependent potential, which shows the consistency of including the momentum-dependent coupling in the QBM Hamiltonian. In the Hu-Paz-Zhang master equation, such a renormalized potential is misplaced so that the correct renormalization Hamiltonian has not been found. With the exact master equation for both the initial decoupled and initial correlated states, the issues about the initial jolt are also reexamined. We find that the so-called "initial jolt," which has been thought to be an artificial effect due to the use of the initial decoupled system-environment states, has nothing to do with the initial decoupled state. The exact master equation for the generalized QBM also has potential applications to photonics quantum computing.

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I. INTRODUCTION

As is well known, any realistic system inevitably interacts with its environment. For nanoscale quantum devices or more general mesoscopic and quantum systems, such interactions are usually not negligible, and thus these systems must be treated as open quantum systems. The essential physics of open quantum systems is the entanglement generation between the system and the environment so that the system cannot be retained in pure quantum states. This makes the system lose its quantum coherence, known as quantum decoherence which is the key obstacle in the development of quantum technology. To date, a theoretical description of open quantum systems is still a big challenge in both fundamental research and practical applications. The major difficulty for solving the dynamics of open quantum systems is how to find the physically reasonable system-environment coupling, which is usually unknown *a priori*, and how to derive unambiguously the equation of motion, called the master equation, for the dynamical evolution of open quantum systems in terms of the reduced density matrix, which is not always computable.

Historically, a prototype of open quantum systems is quantum Brownian motion (QBM) [1,2]. Quantum mechanically, the Brownian particle is modeled as a harmonic oscillator that linearly couples to the environment, which is made of a continuous distribution of an infinite number of harmonic oscillators, as originally proposed in the seminal works of Feynman and Vernon [3] and Caldeira and Leggett [4,5]. The system-environment coupling in QBM was assumed as a strict linear coupling $\sum_k C_k x q_k$, where x and q_k are the position coordinates of the system and environment oscillators, respectively, and C_k is the coupling amplitude. The exact master equation of the QBM for such a system-environment coupling has been derived during the 1980s–1990s with different approaches [4,6–8]. One of the derivations was given by Hu *et al.* utilizing the Feynman-Vernon influence functional approach [3], and the resulting master equation is called Hu-Paz-Zhang master equation in the literature. Furthermore, the dissipative dynamics of QBM has been extensively investigated in rather broad physical research areas [1,2]. In particular, the classical dissipative dynamics, the fluctuation-dissipation theorem, and the classical Langevin equation have been deduced from this QBM in the weak-coupling and high-temperature limits [1–5].

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On the other hand, in the 2000s we have developed a theory for open systems by extending the Feynman-Vernon influence functional approach [3] to the coherent state path integral representation [9]. We derived the exact master equation for both bosonic and fermionic open systems which exchange matters, energies, and information with environments through particle tunneling processes [10–12]. Recently we have further extended such approach to topological systems and derived the exact master equation to Majorana bound states [13,14]. We also unified the master equation approach and the nonequilibrium Green function technique to reformulate the quantum transport theory that can capture the transient quantum transport phenomena with initial correlations [10,11,15,16]. The nonequilibrium Green function technique was originally proposed by Schwinger for QBM [17] and later developed by Kadanoff and Baym and by Keldysh for many-body systems [18,19]. It is certain interesting to see how the Hu-Paz-Zhang master equation for QBM can be naturally reproduced within our framework. Note that the Hu-Paz-Zhang master equation was also derived using the Feynman-Vernon influence functional, but the framework they used is quite different from the coherent state approach, and it cannot be applied to fermionic systems. The connection of these two approaches has not been realized.

On the other hand, microscopically, the classically well-defined linear coupling $\sum_k C_k x q_k$ depicts the Brownian particle dissipation and fluctuations through energy exchange between the system and the environment. In the particle number representation, $\sum_k C_k x q_k = \sum_k \hbar V_k (a^\dagger b_k + b_k^\dagger a + a^\dagger b_k^\dagger + b_k a)$ with $V_k = C_k/2\sqrt{M m_k \omega_{\rm S} \omega_k}$, where $a^\dagger$ and a are the corresponding creation and annihilation operators obeying commutation relation $[a, a^\dagger] = 1$, and M, m_k are the masses of the system oscillator and the environmental oscillator mode k, respectively. It shows that the first two terms, proportional to $a^\dagger b_k + b_k^\dagger a$, depict the single-particle transition processes for energy exchange (energy transition) between the system and the environment, while the last two terms, proportional to $a^\dagger b_k^\dagger + b_k a$, describe the processes of generating and annihilating separately two quanta of energy. As one knows, the energy quanta of harmonic oscillator can be realized with photons or phonons. Physically one can generate or annihilate two photons (phonons) through nonlinear crystals or atomic virtual states. But these physical processes are very different from the single-photon transition processes of the first two terms in linear quantum optics. In other words, the coupling strength V_k for the single-particle transition processes usually cannot be the same as the processes of two-particle pair generation or annihilation. It is also interesting to see if we can generalize the system-environment coupling to the form $\sum_k \hbar V_k (a^\dagger b_k + b_k^\dagger a) + \sum_k \hbar W_k (a^\dagger b_k^\dagger + b_k a)$ with $V_k \neq W_k$ for the realistic photonic processes.

The rest of paper is organized as follows. We give first a detailed analysis of the system-environment couplings for QBM that were originally proposed by Feynman and Vernon [3] and later by Caldeira and Leggett [4,5]. Then we generalize the QBM by including the momentum-dependent coupling, and we also discuss practical realizations of such a generalization. In Sec. III we derive the exact master equation of the generalized QBM for both the initial decoupled and initial correlated states. We also explore the relation of dissipation and fluctuation dynamics in open quantum systems with the decay and diffusions of the Brownian particle. In Sec. IV we show how the Hu-Paz-Zhang master equation for the QBM without including momentum-dependent coupling [4,6–8] can be reproduced as a special case. We also reexamine some debated issues about QBM, such as the origin of the initial jolt phenomena and the initial decoupled state problem often questioned for the study of the QBM with the Hu-Paz-Zhang master equation. Finally, discussions and perspectives are given in Sec. V. In Appendix A the nonequilibrium Green functions for open quantum systems we previously introduced are generalized to open systems containing pairing couplings through the Heisenberg equation of motion. The generalized nonequilibrium fluctuation-dissipation theorem in the time domain is also derived there.

II. A GENERAL ANALYSIS OF MOMENTUM-DEPENDENT SYSTEM-ENVIRONMENT COUPLINGS

In early studies of quantum dissipative dynamics, system-environment couplings in open systems are mainly modeled based on classical mechanics. The prototype example is given in the pioneering work of Feynman and Vernon for developing their influence functional theory for QBM [3]. In the exploration of the dissipative and noise effects of other interaction systems on the system of interest, Feynman and Vernon proposed a general test system x interacting with linear systems $\{q_k\}$ (an oscillator bath) with the Lagrangian

$$L_{\rm FV} = L_{\rm S}(\dot{x}, x) + \frac{1}{2}\sum_k m_k\left(\dot{q}_k^2 - \omega_k^2 q_k^2\right) - U(x)\sum_k C_k q_k. \quad (1)$$

Without loss of generality, one usually takes a linear coupling $U(x) = x$. The oscillator bath is an environment consisting of a collection of harmonic oscillators with a continuous frequency distribution. Such an environment can be easily realized as radiative electromagnetic fields or vibrations of lattices in matter. The Lagrangian formulation in terms of the dynamical variables of positions and velocities is set up in order to conventionally use the Feynman path integrals for the trace over all the environmental states [20]. In such circumstances, the path integrals over environmental degrees of freedom can be done exactly. Thus the environment effect on the test system is given by an influence functional of coordinates of the system only. It is equivalent to external semiclassical dissipative and fluctuating (random) forces acting on the test system. As Feynman and Vernon pointed out [3], if a general nonlinear interaction system $\{q_k\}$ is weakly coupled to a test system x with a more general interaction potential $U(x)U'(\{q_k\})$ which is small, then the effect on the test system is that of a sum of oscillators whose frequencies correspond to the possible transitions of the interaction system. Thus, to the extent that second-order perturbation theory yields sufficient accuracy, the effect of an interaction system is that of a linear system [3].

In 1980s Caldeira and Leggett made further investigations into QBM and quantum tunneling of dissipative systems, in which similar linear couplings are made in order to derive dissipative (damping) equations of motion from quantum mechanics [4,5]. Specifically, to consistently produce the semiclassical damped equation of motion, $M\ddot{x} + \eta(x)\dot{x} + dV/dx = F_{\rm ext}(t)$ where $F_{\rm ext}(t)$ is an external force, Caldeira and Leggett also proposed the environment as a continuous frequency distribution of a set of harmonic oscillators interacting linearly with the system,

$$\begin{aligned} L_{\rm CL} = \frac{1}{2}M\dot{x}^2 - V(x) + \frac{1}{2}\sum_k m_k\big(\dot{q}_k^2 - \omega_k^2 q_k^2\big) \\ - \sum_k F_k(x)q_k - \sum_k F_k^2(x)/2m_k\omega_k^2. \end{aligned} \tag{2}$$

When the system is a tunneling system, Eq. (2) is often called the Caldeira-Leggett model in the literature. Except for the last term which is added as a counterterm for the stability of potential renormalization, Eq. (2) is equivalent to Eq. (1). For x-independent dissipation $\eta(x) = \eta$, the semiclassical damped equation of motion can be realized with the strict linear coupling between the system and the environment $F_k(x) = C_k x$. Other more complicated coupling mechanisms are analyzed extensively in Appendix C of [5], but these analyses are indeed superfluous from the microscopical level of interacting systems, as Caldeira and Leggett pointed out in their own work [5].

To have a clearer physical picture on the microscopical level of interacting systems in the Feynman-Vernon model or the Caldeira-Leggett model, it will be more convenient to reformulate the Hamiltonian of Eq. (2) in the standard second quantization framework:

$$\begin{aligned} H_{\rm CL} &= \frac{p^2}{2M} + V(x) + \sum_k\left(\frac{p_k^2}{2m_k} + \frac{1}{2}m_k\omega_k^2 q_k^2\right) \\ &\quad + \sum_k F_k(x)q_k + \sum_k F_k^2(x)/2m_k\omega_k^2 \\ &= \sum_i \varepsilon_i a_i^\dagger a_i + \sum_k \hbar\omega_k b_k^\dagger b_k + \sum_{ijk} V_{ijk} a_i^\dagger a_j (b_k + b_k^\dagger). \end{aligned} \tag{3}$$

Here ε_i is the spectrum of the system Hamiltonian $H_{\rm S} = \frac{1}{2M}p^2 + V_{\rm eff}(x)$ with $H_{\rm S}\psi_i(x) = \varepsilon_i\psi_i(x)$, where $V_{\rm eff}(x) = V(x) + \sum_k \frac{F_k^2(x)}{2m_k\omega_k^2}$. The operators $a_i^\dagger$ and a_i are, respectively, the creation and annihilation operators that create the system state $|\psi_i\rangle$ from the vacuum state $|0\rangle$ in the standard second quantization scheme: $|\psi_i\rangle = a_i^\dagger|0\rangle$ and $a_i|0\rangle = 0$ with $\psi_i(x) = \langle x|\psi_i\rangle$. These creation and annihilation operators obey the commutation or anticommutation relation, $[a_i, a_j^\dagger]_\mp = a_i a_j^\dagger \mp a_j^\dagger a_i = \delta_{ij}$, depending on the system being a bosonic or a fermionic system. The creation and annihilation operators $b_k^\dagger$, b_k of the harmonic bath can be simply defined by $b_k^\dagger = \sqrt{\frac{m_k\omega_k}{2\hbar}}(q_k - \frac{i}{m_k\omega_k}p_k)$, $b_k = \sqrt{\frac{m_k\omega_k}{2\hbar}}(q_k + \frac{i}{m_k\omega_k}p_k)$. The system-environment coupling $V_{ijk} = \sqrt{\frac{\hbar}{2m_k\omega_k}}\int dx\psi_i^*(x)F_k(x)\psi_j(x)$ describes the transition of the system from the state $|\psi_j\rangle$ to the state $|\psi_i\rangle$ due to the linear coupling to the environmental oscillating mode ω_k. Now one can see that the position-dependent system-environment coupling in Eq. (3) induces the same transition of the system from the state $|\psi_j\rangle$ to the state $|\psi_i\rangle$ regardless whether it emits or absorbs an energy quanta with the energy $\hbar\omega_k$. These two processes with the same transition amplitude (see Fig. 1) should barely happen together in quantum mechanics.

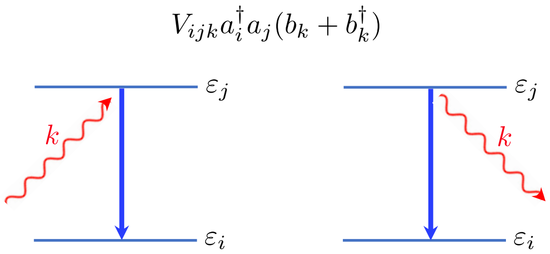


FIG. 1. The two physical processes in the conventional QBM with the position-dependent system-environment couplings only that are described by $\sum_{ijk} V_{ijk}a_i^\dagger a_j(b_k + b_k^\dagger)$ in Eq. (3). These two processes correspond to the same transit of the Brownian particle from the energy eigenstate $|\psi_j\rangle$ to the energy eigenstate $|\psi_i\rangle$ regardless whether it emits or absorbs an energy quanta, which barely occur in quantum mechanics.

In fact, a general Hamiltonian of the system coupled to the oscillator-bath environment should contain both the position-dependent and the momentum-dependent system-environment couplings, as Caldeira and Leggett first pointed out in their original paper [5,21],

$$\begin{aligned} H_{\text{S-E}} = \frac{p^2}{2M} + V(x) + \sum_k\left(\frac{p_k^2}{2m_k} + \frac{1}{2}m_k\omega_k^2 q_k^2\right) \\ + \sum_k [F_k(p,x)q_k + G_k(p,x)p_k] + \Phi(p,x), \end{aligned} \tag{4}$$

where $\Phi(p,x)$ is a counterterm for the renormalization of system Hamiltonian arisen from the system-environment couplings. Equation (4) contains various position and momentum dependent system-environment couplings. The second quantization of the above Hamiltonian leads to

$$\begin{aligned} H_{\text{S-E}} = \sum_i \varepsilon_i a_i^\dagger a_i + \sum_k \hbar\omega_k b_k^\dagger b_k \\ + \sum_{ijk}(V'_{ijk}a_i^\dagger a_j b_k^\dagger + V'^*_{ijk}a_j^\dagger a_i b_k). \end{aligned} \tag{5}$$

Similarly, ε_i is the spectrum of the system Hamiltonian $H_{\rm S} = \frac{p^2}{2M} + V(x) + \Phi(p,x)$ with $H_{\rm S}|\psi_i\rangle = \varepsilon_i|\psi_i\rangle$, the system-environment couplings V'_{ijk} are given by

$$\begin{aligned} V'_{ijk} = \sqrt{\frac{\hbar}{2m_k\omega_k}}\langle\psi_i|F_k(p,x)|\psi_j\rangle \\ + i\sqrt{\frac{\hbar m_k\omega_k}{2}}\langle\psi_i|G_k(p,x)|\psi_j\rangle. \end{aligned} \tag{6a}$$

We find that the system-environment coupling [the last two terms in Eq. (5)] contains the physical process of the system transiting from the state $|\psi_i\rangle$ to the state $|\psi_j\rangle$ by *absorbing* a photon, and the inverse process of transiting the state $|\psi_j\rangle$ back to the state $|\psi_i\rangle$ by *emitting* a photon with the same energy, as shown by Fig. 2. In other words, it shows that with the generalization of Brownian motion Hamiltonian of including the momentum-dependent system-environment coupling, the Hamiltonian (5) becomes indeed the most popular

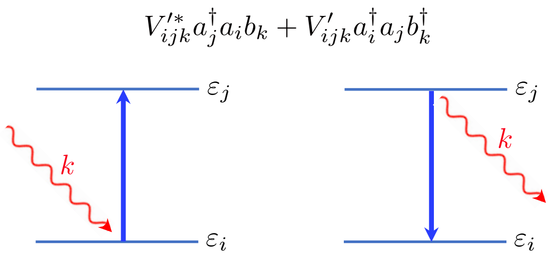


FIG. 2. The two physical processes in the generalized QBM including the momentum-dependent system-environmental couplings that are described by $V'^*_{ijk}a^\dagger_j a_i b_k + V'_{ijk}a^\dagger_i a_j b^\dagger_k$ in Eq. (5). These two processes correspond to the Brownian particle transition from the energy eigenstate $|\psi_i\rangle$ to the energy eigenstate $|\psi_j\rangle$ by absorbing an energy quanta, and its inverse process of the transition from the energy eigenstate $|\psi_j\rangle$ to the energy eigenstate $|\psi_i\rangle$ by emitting the same energy quanta, respectively, as the standard processes in quantum mechanics or more fundamentally in quantum field theory.

Hamiltonian described matter-photon interaction or matter-phonon interaction in physics. Such a general Hamiltonian can be derived rigorously from the nonrelativistic quantum electrodynamics in field theory.

In fact, for atoms interacting with radiative electromagnetic field, the momentum-dependent coupling is naturally manifested. Explicitly, the nonrelativistic atom-photon interaction Hamiltonian can be rigorously written as [22]

$$H_{\text{e-p}} = \sum_i \frac{1}{2m}[\boldsymbol{P}_i - e\boldsymbol{A}(\boldsymbol{r}_i)]^2 + \sum_{\boldsymbol{k},\lambda=1,2}\hbar\omega_{\boldsymbol{k}}b^\dagger_{\boldsymbol{k},\lambda}b_{\boldsymbol{k},\lambda} + V_{\text{coul}}, \tag{7}$$

where m and e are the electron mass and charge, $\boldsymbol{P}_i = -i\hbar\nabla_i$ is the ith electron momentum operator in the atom. The vector field $\boldsymbol{A}(\boldsymbol{r})$ is the canonical coordinate of the electromagnetic field and is quantized by

$$\boldsymbol{A}(\boldsymbol{r}) = \sum_{\boldsymbol{k},\lambda=1,2}\sqrt{\frac{\hbar}{2\epsilon_0\omega_{\boldsymbol{k}}V}}\,\hat{\boldsymbol{e}}_\lambda\left(b_{\boldsymbol{k},\lambda}e^{i\boldsymbol{k}\cdot\boldsymbol{r}} + b^\dagger_{\boldsymbol{k},\lambda}e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}\right) \tag{8}$$

with the Coulomb gauge $\boldsymbol{k}\cdot\hat{\boldsymbol{e}}_\lambda = 0$. The last term $V_{\text{coul}} = V^{\text{e-N}}_{\text{coul}} + V^{\text{e-e}}_{\text{coul}}$ in Eq. (7) contains all the electron-nuclei and electron-electron Coulomb interactions in the atom arisen from the charge-induced scalar potentials. The atom-light interaction in Eq. (7) is given by the term $-\frac{e}{m}\sum_i \boldsymbol{A}(\boldsymbol{r}_i)\cdot\boldsymbol{P}_i$, when the Coulomb gauge $\nabla\cdot\boldsymbol{A}(\boldsymbol{r}) = 0$ is used. Explicitly, Eq. (7) can be rewritten as

$$\begin{aligned} H_{\text{e-p}} &= \sum_i \frac{1}{2m}\boldsymbol{P}_i^2 + V_{\text{coul}} + \sum_{\boldsymbol{k},\lambda=1,2}\hbar\omega_{\boldsymbol{k}}b^\dagger_{\boldsymbol{k},\lambda}b_{\boldsymbol{k},\lambda} \\ &\quad - \frac{e}{m}\sum_i \boldsymbol{A}(\boldsymbol{r}_i)\cdot\boldsymbol{P}_i + \frac{e^2}{2m}\sum_i \boldsymbol{A}^2(\boldsymbol{r}_i). \end{aligned} \tag{9}$$

Here the term $\frac{e^2}{2m}\sum_i \boldsymbol{A}^2(\boldsymbol{r}_i)$ can be ignored because it gives shift only to the radiative frequencies for all the radiative field modes. Equation (9) describes a typical momentum-dependent coupling between the system and the environment given explicitly by $\frac{e}{m}\sum_i \boldsymbol{A}(\boldsymbol{r}_i)\cdot\boldsymbol{P}_i$ when the atom is considered as the system and the radiative field is treated as the environment.

Taking the second quantization to the electron energy eigenfunctions $\psi_\alpha(\boldsymbol{r})$ of the atom under the interaction with nuclei, we have $[\frac{1}{2m}\boldsymbol{P}^2 + V^{\text{e-N}}_{\text{coul}}(\boldsymbol{r})]\psi_\alpha(\boldsymbol{r}) = E_\alpha\psi_\alpha(\boldsymbol{r})$, where the nuclei is considered to be static. Then the atom-light interaction Hamiltonian of Eq. (7) can be rewritten as

$$\begin{aligned} H_{\text{e-p}} &= \sum_\alpha E_\alpha a^\dagger_\alpha a_\alpha + V^{\text{e-e}}_{\text{coul}} + \sum_{\boldsymbol{k},\lambda=1,2}\hbar\omega_{\boldsymbol{k}}b^\dagger_{\boldsymbol{k},\lambda}b_{\boldsymbol{k},\lambda} \\ &\quad + \sum_{\alpha\beta\boldsymbol{k},\lambda=1,2}\left(V_{\alpha\beta\boldsymbol{k}\lambda}a^\dagger_\alpha a_\beta b^\dagger_{\boldsymbol{k},\lambda} + V^*_{\alpha\beta\boldsymbol{k}\lambda}a^\dagger_\beta a_\alpha b_{\boldsymbol{k},\lambda}\right). \end{aligned} \tag{10}$$

The operator $a^\dagger_\alpha$ creates the single-particle state $\psi_\alpha(\boldsymbol{r})$ from the vacuum state. The transition amplitude $V_{\alpha\beta\boldsymbol{k}\lambda} = -\frac{e}{m}\sqrt{\frac{\hbar}{2\epsilon_0\omega_{\boldsymbol{k}}V}}\int e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}\psi^*_\alpha(\boldsymbol{r})\hat{\boldsymbol{e}}_\lambda\cdot\boldsymbol{P}\psi_\beta(\boldsymbol{r})\,d^3\boldsymbol{r}$ describes the photon-emitting transition from the atomic state ψ_β to the atomic state ψ_α under the constraints of energy, angular momentum, and parity conservations (the transition selection rules). The transition amplitude $V^*_{\alpha\beta\boldsymbol{k}\lambda}$ describes the photon-absorbing transition from the atomic state ψ_α to the atomic state ψ_β. It shows that the momentum-dependent interaction dominates all the electron-photon transition processes in quantum electrodynamics.

However, even for the Feynman-Vernon model (1) and the Caldeira-Leggett model (2), or the extension model of Eq. (5) with the momentum-dependent system-environment coupling, one cannot solve exactly the corresponding nonequilibrium decoherence dynamics in general. Only when one takes the system also as a simple harmonic oscillator, $H_{\text{S}} = \frac{1}{2M}p^2 + \frac{1}{2}M\Omega^2x^2$, and focuses on the strict linear coupling between the system and the environment, i.e., $U(x) = x$ in Eq. (1) and $F_k(x) = C_kx$ in Eq. (2), has the exact master equation for the reduced density matrix of the system been derived [6–8]. Explicitly, let us write the Hamiltonian of Eq. (1) or Eq. (2) for the system being a harmonic oscillator linearly coupled to the oscillator bath in the particle number representation,

$$\begin{aligned} H &= \frac{p^2}{2M} + \frac{1}{2}M\Omega^2x^2 + \sum_k\left(\frac{p_k^2}{2m_k} + \frac{1}{2}m_k\omega_k^2q_k^2\right) \\ &\quad + \sum_k C_kxq_k + \sum_k C_k^2x^2/2m_k\omega_k^2 \\ &= \hbar\omega_{\text{s}}a^\dagger a + \sum_k\hbar\omega_kb^\dagger_kb_k \\ &\quad + \sum_k\hbar V_k(a^\dagger b_k + b^\dagger_ka + a^\dagger b^\dagger_k + b_ka), \end{aligned} \tag{11}$$

where $\omega_{\text{s}}^2 = \Omega^2 + \Omega_{\text{c}}^2$ and $\Omega_{\text{c}}^2 = \sum_k C_k^2/Mm_k\omega_k^2$ arises from the counterterm to cancel the possible divergence of the renormalized frequency shift [5], and $V_k = C_k/2\sqrt{M\omega_{\text{s}}m_k\omega_k}$. In Eq. (11) the particle creation and annihilation operators $a^\dagger$, a are simply defined by $a^\dagger = \sqrt{\frac{M\omega_{\text{s}}}{2\hbar}}(x - \frac{i}{M\omega_{\text{s}}}p)$, $a = \sqrt{\frac{M\omega_{\text{s}}}{2\hbar}}(x + \frac{i}{M\omega_{\text{s}}}p)$ for a harmonic oscillator. However, it is also easy to deduce Eq. (11) directly from the second general quantization form of Eq. (3); see the detailed derivation given in Appendix B.

As we have pointed out in the introduction, physically, the terms proportional to $a^\dagger b^\dagger_k + b_ka$ correspond to the processes of two-particle (two energy quanta) pair generation and annihilation. Consider a practical realization of quantum harmonic oscillator by photons: these processes correspond to two-photon creation and annihilation in nonlinear optics. These two-photon processes could be physically realized, for example, by spontaneous parametric down-conversion, which is very different from the other two terms proportional to $a^\dagger b_k + b_k^\dagger a$ in Eq. (11). The latter is a linear particle process corresponding to the quantum energy tunneling (energy quanta exchange) between the system and the environment. Consequently, the system-environment coupling strengths for the linear particle exchange and the nonlinear particle pairing production and annihilation are usually different, which are two very different physical processes.

It is interesting to see that if the momentum-dependent couplings are included, the general strict linear coupling Hamiltonian can have the form

$$\begin{aligned}&\sum_k (C_{1k} x q_k + C_{2k} p p_k + C_{3k} x p_k + C_{4k} p q_k)\\&\quad = \sum_k \hbar (V_k a^\dagger b_k + V_k^* b_k^\dagger a + W_k a^\dagger b_k^\dagger + W_k^* b_k a). \qquad (12)\end{aligned}$$

In other words, the quantum mechanically feasible QBM Hamiltonian that includes momentum-dependent couplings should be

$$\begin{aligned}H_{\rm tot} = {}& \hbar\omega_{\rm s} a^\dagger a + \sum_k \hbar\omega_k b_k^\dagger b_k + \sum_k \hbar (V_k a^\dagger b_k + V_k^* b_k^\dagger a)\\&+ \sum_k \hbar (W_k a^\dagger b_k^\dagger + W_k^* b_k a), \qquad (13)\end{aligned}$$

where $W_k \neq V_k$ in general. Obviously, the conventional QBM Eq. (11) in the literature is a special case of Eq. (13) with $W_k = V_k$. Furthermore, Eq. (13) can be easily realized in many different quantum systems. A typical example is the cavity coupled to the environment with an external squeezed driving,

$$\begin{aligned}H = {}& \hbar\omega_0 c^\dagger c + f(t) c^{\dagger 2} + f^*(t) c^2\\&+ \sum_k \hbar\omega_k b_k^\dagger b_k + \sum_k (\mathcal{V}_k c^\dagger b_k + \mathcal{V}_k^* b_k^\dagger c). \qquad (14)\end{aligned}$$

Taking a Bogoliubov transformation $a = uc + vc^\dagger$, $a^\dagger = u^* c^\dagger + v^* c$ with $|u|^2 - |v|^2 = 1$, it is easy to show that this Hamiltonian becomes Eq. (13) with $\omega_{\rm s}^2 = \omega_0^2 - 4|f(t)|^2/\hbar^2$ and $V_k = u\mathcal{V}_k/\hbar$, $W_k = -v\mathcal{V}_k^*/\hbar$ such that $W_k \neq V_k$. This shows explicitly how the system-environment coupling strengths for one- and two-photon processes become different in the realistic photonic process. Thus, the generalization of the QBM with momentum-dependent coupling between the system and the environment is physically acceptable.

Moreover, investigations of quantum photonics have made great progress for solving the boson sampling problem very recently [23–26]. Quantum photonics is made of many squeezers and interferometers (beam splitters and phase shifters), which can be described by the following Hamiltonian including various photon loss:

$$\begin{aligned}H_{\rm Q.P.}(t) = {}& \sum_i \left[\hbar\omega_{{\rm s},i} c_i^\dagger c_i + f_i(t) c_i^{\dagger 2} + f_i^*(t) c_i^2\right]\\&+ \sum_{ij} (B_{ij} c_i^\dagger c_j + B_{ij}^* c_j^\dagger c_i) + \sum_{\alpha k} \hbar\omega_{\alpha k} b_{\alpha k}^\dagger b_{\alpha k}\\&+ \sum_{i\alpha k} (\mathcal{V}_{i,\alpha k} c_i^\dagger b_{\alpha k} + \mathcal{V}_{i,\alpha k}^* b_{\alpha k}^\dagger c_i), \qquad (15)\end{aligned}$$

where index α labels different photon loss environments. After diagonalizing the Hamiltonian of the squeezers and interferometers (the first two terms in the above Hamiltonian) through a Bogoliubov transformation, the total Hamiltonian can be reduced to

$$\begin{aligned}H_{\rm Q.P.}(t) = {}& \sum_i \hbar\omega_{{\rm s},i}(t) a_i^\dagger a_i + \sum_{\alpha k} \hbar\omega_{\alpha k} b_{\alpha k}^\dagger b_{\alpha k}\\&+ \sum_{i\alpha k} [V_{i,\alpha k}(t) a_i^\dagger b_{\alpha k} + V_{i,\alpha k}^*(t) b_{\alpha k}^\dagger a_i\\&+ W_{i,\alpha k}(t) a_i^\dagger b_{\alpha k}^\dagger + W_{i,\alpha k}^*(t) b_{\alpha k} a_i]. \qquad (16)\end{aligned}$$

This is an extension of the generalized QBM Eq. (13) to multimodes that is practically used in the integrated quantum photonics for the current investigation of photonics quantum computing [27–31].

Analogically, in the current investigation of topological phases of matter, many quantum devices are made by superconductor-semiconductor, superconductor-insulator, and superconductor-metal hybrid systems [32–35]. They are coupled to leads and thereby become open quantum systems. The corresponding Hamiltonian, after a Bogoliubov transformation, can be equivalently written as

$$\begin{aligned}H_{\rm T.S.} = {}& \sum_i \varepsilon_i(t) a_i^\dagger a_i + \sum_{\alpha k} \epsilon_{\alpha k}(t) b_{\alpha k}^\dagger b_{\alpha k}\\&+ \sum_{i\alpha k} [V_{i,\alpha k}(t) a_i^\dagger b_{\alpha k} + \Delta_{i,\alpha k}(t) a_i^\dagger b_{\alpha k}^\dagger + {\rm H.c.}], \qquad (17)\end{aligned}$$

where index α labels different leads. This Hamiltonian has the same structure as the one for photonic quantum processor of Eq. (16), except that here it describes electron dynamics rather than photon dynamics in open quantum systems. Physically, Eq. (17) can be considered as a fermionic realization of the generalized QBM. The general exact master equation of Eq. (17) has been derived recently by us for initially correlated system-environment states [13]. Applications to the study of decoherence dynamics on exotic states, such as anyons and Majorana quasiparticles for topological quantum computers, have also been explored [14,36–38].

The above analysis indicates that there has been too much emphasis on quantization in the literature (i.e. general methods of obtaining quantum mechanics from classical methods) as opposed to the converse problem of the classical limit of quantum mechanics. This is unfortunate because the latter is an important question for various areas of modern physics while the former is a chimera [39,40]. In the following, we shall focus on the more general and quantum mechanically self-consistent model with Eq. (13) as a generalization of the conventional QBM. We will derive its exact master equation for both initially decoupled and initially entangled system-environment states. For its applications in quantum photonics, such as the integrated quantum processor made of many squeezers and interferometers with 100 more photonic modes for photonics quantum computing that is described by Eq. (16), we will leave these for further investigation.

III. THE EXACT MASTER EQUATION OF THE GENERALIZED QBM

For an arbitrary initial state $\rho_{\text{tot}}(t_0)$, the time evolution of the total system (system plus environment) is determined by the Liouville–von Neumann equation of the quantum mechanics [41],

$$\frac{d}{dt}\rho_{\text{tot}}(t) = \frac{1}{i\hbar}[H_{\text{tot}}(t), \rho_{\text{tot}}(t)]. \tag{18}$$

Because the system and the environment together form a closed system, the Liouville–von Neumann equation is the same as the Schrödinger equation of quantum mechanics for the evolution of pure quantum states. But the Liouville–von Neumann equation is more general because it is also valid for statistically mixed states, such as Eqs. (23) and (24), where the Schrödinger equation is not applicable. The time evolution of the system is determined by the reduced density matrix which is defined as the partial trace of the total density matrix over all the environment states,

$$\rho_{\text{S}}(t) = \text{Tr}_{\text{E}}[\rho_{\text{tot}}(t)]. \tag{19}$$

Applying the partial trace over the environment states to Eq. (18) with the total Hamiltonian of Eq. (13), we obtain formally the exact master equation

$$\frac{d}{dt}\rho_{\text{S}}(t) = \frac{1}{i\hbar}[H_{\text{S}}, \rho_{\text{S}}(t)] + \mathcal{L}^{+}[\rho_{\text{S}}(t)] + \mathcal{L}^{-}[\rho_{\text{S}}(t)], \tag{20}$$

where

$$\mathcal{L}^{+}[\rho_{\text{S}}(t)] = a^{\dagger}A[\rho_{\text{S}}(t)] + A^{\dagger}[\rho_{\text{S}}(t)]a, \tag{21a}$$

$$\mathcal{L}^{-}[\rho_{\text{S}}(t)] = -aA^{\dagger}[\rho_{\text{S}}(t)] - A[\rho_{\text{S}}(t)]a^{\dagger}, \tag{21b}$$

and $A[\rho_{\text{S}}(t)]$ is a collective operator defined by

$$A[\rho_{\text{S}}(t)] \equiv -i\,\text{Tr}_{\text{E}}[(\boldsymbol{V}\cdot\boldsymbol{b}+\boldsymbol{W}\cdot\boldsymbol{b}^{\dagger})\rho_{\text{tot}}(t)]. \tag{22}$$

Here $\mathcal{L}^{\pm}[\rho_{\text{S}}(t)]$ is positive and negative supercurrent operators that describe particles flowing into and out of the system. It contains the usual current through particle tunnelings between the system and the environment, and also the current via particle pair production and annihilation. The latter is similar to the Andreev reflections through the interface between superconductors and normal metals. Particle dissipation and fluctuations arisen from the system-environment back-actions can be naturally manifested after completed the partial trace over all the environment states in Eq. (22).

Now the problem left is how to carry out explicitly the partial trace over all the environment states in Eq. (22). For the initial total density matrix $\rho_{\text{tot}}(t_0)$ being a partition-free thermal state (containing the initial system-environment correlation),

$$\rho_{\text{tot}}(t_0) = \frac{1}{Z_{\text{tot}}}\exp(-H_{\text{tot}}/k_{\text{B}}T), \tag{23}$$

because the total Hamiltonian is a bilinear function of particle creation and annihilation operators of the system and environment, the total density matrix and the reduced density matrix always evolve in Gaussian states after a dynamical quench. In this case, it is rather easy to trace over all the environment states with Gaussian integrals in the coherent state representation. Such a configuration has been presented in our previous derivation of the exact master equation for dissipative topological systems [13]. Below we will focus on cases with an initial decoupled state. At the end of the section, we will make a connection to the master equation with the above initial correlated state.

Thus, we now assume that the system and the environment are initially decoupled, and the environment is initially in a thermal state [3,4],

$$\rho_{\text{tot}}(t_0) = \rho_{\text{S}}(t_0)\otimes\rho_{\text{E}}^{\text{th}}, \quad \rho_{\text{E}}^{\text{th}} = \frac{1}{Z_{\text{E}}}e^{-H_{\text{E}}/k_{\text{B}}T}. \tag{24}$$

Such an initial state can be prepared by tuning coupling strength between the system and the environment to zero at initial time. Although the initial total state is a system-environment decoupled state, the system initial state $\rho_{\text{S}}(t_0)$ can be arbitrary (non-Gaussian states). Therefore, the partial trace over the environment states in Eq. (22) is rather difficult to carry out. To complete this partial trace, we shall use the coherent state path integral method [9]. In the coherent state representation, the matrix element of the collective operator Eq. (22) can be expressed as

$$\begin{aligned}\langle z_t^{*}|\begin{bmatrix}A[\rho_{\text{S}}(t)]\\ A^{\dagger}[\rho_{\text{S}}(t)]\end{bmatrix}|z_t'\rangle = \int d\mu(z_0^{*}, z_0)\,d\mu(z_0'^{*}, z_0')\langle z_0^{*}|\rho_{\text{S}}(t_0)|z_0'\rangle\\ \times\ \mathcal{J}^{A}(z_t^{*}, z_t', t; z_0, z_0'^{*}, t_0).\end{aligned} \tag{25}$$

In Eq. (25), $|z\rangle \equiv e^{a^{\dagger}z}|0\rangle$ is the unnormalized coherent state. Correspondingly, the integral measure over the complex space is given by $d\mu(z^{*}, z) \equiv \frac{dz^{*}dz}{2\pi i}e^{-|z|^2}$. The A-operator-associated propagating function $\mathcal{J}^{A}(z_t^{*}, z_t', t; z_0, z_0'^{*}, t_0)$ is defined in a similar way as the propagating function for the reduced density matrix in the coherent state representation [11],

$$\begin{aligned}\langle z_t^{*}|\rho_{\text{S}}(t)|z_t'\rangle = \int d\mu(z_0^{*}, z_0)\,d\mu(z_0'^{*}, z_0')\langle z_0^{*}|\rho_{\text{S}}(t_0)|z_0'\rangle\\ \times\ \mathcal{J}(z_t^{*}, z_t', t; z_0, z_0'^{*}, t_0).\end{aligned} \tag{26}$$

The propagating function $\mathcal{J}(z_t^{*}, z_t', t; z_0, z_0'^{*}, t_0)$ fully describes the time evolution of the reduced density matrix of Eq. (19). Similarly, $\mathcal{J}^{A}(z_t^{*}, z_t', t; z_0, z_0'^{*}, t_0)$ fully determines the evolution of the collective operator $A[\rho_{\text{S}}(t)]$ of Eq. (22).

Utilizing the coherent state path integrals, we have

$$\begin{aligned}&\mathcal{J}(z_t^{*}, z_t', t; z_0, z_0'^{*}, t_0)\\ &\quad= \int_{z_0}^{z_t^{*}}\mathfrak{D}[z^{*}, z]e^{\frac{i}{\hbar}S_{\text{S}}[z^{*},z]}\\ &\quad\times\ \int_{z_0'^{*}}^{z_t'}\mathfrak{D}[z'^{*}, z']e^{-\frac{i}{\hbar}S_{\text{S}}^{*}[z'^{*},z']}\mathcal{F}[z^{*}, z'^{*}, z, z'],\end{aligned} \tag{27}$$

and similarly,

$$\begin{aligned}\mathcal{J}^A&(z_t^*, z_t', t; z_0, z_0'^*, t_0)\\ &= \int_{z_0}^{z_t^*}\mathfrak{D}[z^*, z]e^{\frac{i}{\hbar}S_{\mathrm{s}}[z^*,z]}\\ &\quad\times \int_{z_0'^*}^{z_t'}\mathfrak{D}[z'^*, z']e^{-\frac{i}{\hbar}S_{\mathrm{s}}^*[z'^*,z']}\mathcal{F}^A[z^*, z'^*, z, z'].\end{aligned}\tag{28}$$

Here $S_{\mathrm{s}}[z^*, z]$ is the action of the system, given by

$$\begin{aligned}\frac{i}{\hbar}S_{\mathrm{s}}[z^*, z] &= \frac{1}{2}(z^*(t_0)z_0 + z_t^* z(t)) + \int_{t_0}^{t} d\tau\\ &\quad\times\left[\frac{\dot{z}^*(\tau)z(\tau) - z^*(\tau)\dot{z}(\tau)}{2} - i\omega_{\mathrm{s}}z^*(\tau)z(\tau)\right].\end{aligned}\tag{29}$$

The first two terms in the action are geometric terms arising from the boundary (the end points of the coherent state path integral). This is because the paths in the coherent state path integrals are fixed only on one side: $z(t_0) = z_0$, $z^*(t) = z_t^*$ and $z'^*(t_0) = z_0'^*$, $z'(t) = z_t'$; see Eqs. (25) and (26). While $(z(t), z^*(t))$ and $(z'(t), z'^*(t))$ are not the complex conjugate pairs of the above boundary conditions, they must be determined by solving the path integral explicitly [42].

On the other hand, $\mathcal{F}[z^*, z'^*, z, z']$ in Eq. (27) is the influence functional arising from the partial trace over all the environmental states [3]. It is obtained by rigorously integrating out all the environment degrees of freedom through the closed-loop path integrals [10],

$$\begin{aligned}\mathcal{F}&[z^*, z'^*, z, z']\\ &= \exp\Bigg(-\frac{1}{2}\int_{t_0}^{t}d\tau\Bigg\{\int_{t_0}^{\tau}d\tau'[z^{\mathrm{q}*}(\tau)\ z^{\mathrm{q}}(\tau)]\mathcal{G}(\tau,\tau')\begin{bmatrix}z(\tau')\\ z^*(\tau')\end{bmatrix}\\ &\quad - \int_{\tau}^{t}d\tau'[z'^*(\tau)\ z'(\tau)]\mathcal{G}(\tau,\tau')\begin{bmatrix}z^{\mathrm{q}}(\tau')\\ z^{\mathrm{q}*}(\tau')\end{bmatrix}\\ &\quad + \int_{t_0}^{t}d\tau'[z^{\mathrm{q}*}(\tau)\ z^{\mathrm{q}}(\tau)]\widetilde{\mathcal{G}}(\tau,\tau')\begin{bmatrix}z^{\mathrm{q}}(\tau')\\ z^{\mathrm{q}*}(\tau')\end{bmatrix}\Bigg\}\Bigg),\end{aligned}\tag{30}$$

where we have introduced the new variables $z^{\mathrm{q}} \equiv z - z'$, $z^{\mathrm{q}*} \equiv z^* - z'^*$. The integral kernels $\mathcal{G}(\tau, \tau')$ and $\widetilde{\mathcal{G}}(\tau, \tau')$ are the two-time system-environment correlation functions,

$$\begin{aligned}\mathcal{G}(\tau,\tau') &= \sum_k\begin{bmatrix}V_k(\tau-t_0) & W_k(\tau-t_0)\\ W_k^*(\tau-t_0) & V_k^*(\tau-t_0)\end{bmatrix}\\ &\quad\times\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}\begin{bmatrix}V_k^*(\tau'-t_0) & W_k(\tau'-t_0)\\ W_k^*(\tau'-t_0) & V_k(\tau'-t_0)\end{bmatrix},\end{aligned}\tag{31a}$$

$$\begin{aligned}\widetilde{\mathcal{G}}(\tau,\tau') &= \sum_k\begin{bmatrix}V_k(\tau-t_0) & W_k(\tau-t_0)\\ W_k^*(\tau-t_0) & V_k^*(\tau-t_0)\end{bmatrix}\begin{bmatrix}\overline{n}_k(t_0) & \overline{s}_k(t_0)\\ \overline{s}_k^*(t_0) & \overline{h}_k(t_0)\end{bmatrix}\\ &\quad\times\begin{bmatrix}V_k^*(\tau'-t_0) & W_k(\tau'-t_0)\\ W_k^*(\tau'-t_0) & V_k(\tau'-t_0)\end{bmatrix}.\end{aligned}\tag{31b}$$

Here we have also introduced the notations $V_k(\tau - t_0) \equiv V_k e^{-i\omega_k(\tau-t_0)}$ and $W_k(\tau - t_0) \equiv W_k e^{+i\omega_k(\tau-t_0)}$. The phase factors come from the particle evolution in the environment before it transits into the system through the tunneling or is created from the pairing coupling in Eq. (13). It shows that only the time correlation function $\widetilde{\mathcal{G}}(\tau, \tau')$ relates to the initial environment state through the initial particle occupations and particle-squeezed parameters in the environment, $\overline{n}_k(t_0) = \langle b_k^\dagger(t_0)b_k(t_0)\rangle$, $\overline{s}_k(t_0) = \langle b_k(t_0)b_k(t_0)\rangle$, $\overline{h}_k(t_0) = \langle b_k(t_0)b_k^\dagger(t_0)\rangle = 1 + \overline{n}_k(t_0)$. For the initial environment thermal state (24), the particle-squeezed parameters $\overline{s}_k(t_0)$ become zero, but here we formally keep it so that it can also apply to other initial environment states. These time correlations depict back-action processes between the system and the environment, while the operator A-associated influence functional, after taking the partial trace over the environment states, can be reduced as

$$\begin{aligned}\mathcal{F}^A[z^*, z'^*, z, z'] &= -\int_{t_0}^{t}d\tau\Bigg\{\mathcal{Z}\mathcal{G}(t,\tau)\begin{bmatrix}z(\tau)\\ z^*(\tau)\end{bmatrix}\\ &\quad + \mathcal{Z}\widetilde{\mathcal{G}}(t,\tau)\begin{bmatrix}z^{\mathrm{q}}(\tau)\\ z^{\mathrm{q}*}(\tau)\end{bmatrix}\Bigg\}\mathcal{F}[z^*, z'^*, z, z'],\end{aligned}\tag{32}$$

where $\mathcal{Z} = [\begin{smallmatrix}1 & 0\\ 0 & -1\end{smallmatrix}]$ is the z-component Pauli matrix.

Furthermore, the path integrals in the propagating function of Eq. (27) can be exactly carried out because of quadratic form of the effective action (including the influence functional). The result is

$$\begin{aligned}\mathcal{J}&(z_t^*, z_t', t; z_0, z_0'^*, t_0)\\ &= \frac{1}{Z(t,t_0)}\exp(z_0'^* z_0 + z_t^* z_t')\\ &\quad\times\exp\left(\frac{1}{2}\left\{[z_t^*\ z_t']\mathcal{Z}\begin{bmatrix}z^{\mathrm{q}}(t)\\ z^{\mathrm{q}*}(t)\end{bmatrix} - [z_0'^*\ z_0]\mathcal{Z}\begin{bmatrix}z^{\mathrm{q}}(t_0)\\ z^{\mathrm{q}*}(t_0)\end{bmatrix}\right\}\right).\end{aligned}\tag{33}$$

Here $z^{\mathrm{q}}(t)$, $z^{\mathrm{q}*}(t)$ and $z^{\mathrm{q}}(t_0)$, $z^{\mathrm{q}*}(t_0)$ are determined by the stationary path,

$$\frac{d}{d\tau}\begin{bmatrix}z^{\mathrm{q}}(\tau)\\ z^{\mathrm{q}*}(\tau)\end{bmatrix} + i\omega_{\mathrm{s}}\mathcal{Z}\begin{bmatrix}z^{\mathrm{q}}(\tau)\\ z^{\mathrm{q}*}(\tau)\end{bmatrix} - \int_{\tau}^{t}d\tau'\mathcal{Z}\mathcal{G}(\tau,\tau')\begin{bmatrix}z^{\mathrm{q}}(\tau')\\ z^{\mathrm{q}*}(\tau')\end{bmatrix} = 0,\tag{34a}$$

$$\begin{aligned}\frac{d}{d\tau}\begin{bmatrix}z(\tau)\\ z^*(\tau)\end{bmatrix} + i\omega_{\mathrm{s}}\mathcal{Z}\begin{bmatrix}z(\tau)\\ z^*(\tau)\end{bmatrix} + \int_{t_0}^{\tau}d\tau'\mathcal{Z}\mathcal{G}(\tau,\tau')\begin{bmatrix}z(\tau')\\ z^*(\tau')\end{bmatrix}\\ = -\int_{t_0}^{t}d\tau'\mathcal{Z}\widetilde{\mathcal{G}}(\tau,\tau')\begin{bmatrix}z^{\mathrm{q}}(\tau')\\ z^{\mathrm{q}*}(\tau')\end{bmatrix},\end{aligned}\tag{34b}$$

with $t_0 \leqslant \tau \leqslant t$. This is a generalization of our previous work [10] to the open systems with paring correlations. To solve the above equation, we introduce the transformation with different boundary conditions of the initial and final ends in the coherent state path integrals,

$$\begin{bmatrix}z^{\mathrm{q}}(\tau)\\ z^{\mathrm{q}*}(\tau)\end{bmatrix} = \mathcal{U}(\tau,t)\begin{bmatrix}z^{\mathrm{q}}(t)\\ z^{\mathrm{q}*}(t)\end{bmatrix},\tag{35a}$$

$$\begin{bmatrix}z(\tau)\\ z^*(\tau)\end{bmatrix} = \mathcal{U}(\tau,t_0)\begin{bmatrix}z(t_0)\\ z^*(t_0)\end{bmatrix} - \mathcal{V}(\tau,t)\mathcal{Z}\begin{bmatrix}z^{\mathrm{q}}(t)\\ z^{\mathrm{q}*}(t)\end{bmatrix}.\tag{35b}$$

Then Eq. (34) is reduced to

$$\frac{d}{d\tau}\mathcal{U}(\tau,t_0)+i\omega_s\mathcal{Z}\mathcal{U}(\tau,t_0)+\int_{t_0}^{\tau}d\tau'\mathcal{Z}\mathcal{G}(\tau,\tau')\mathcal{U}(\tau',t_0)=0, \tag{36a}$$

$$\begin{aligned}\frac{d}{d\tau}\mathcal{V}(\tau,t)+i\omega_s\mathcal{Z}\mathcal{V}(\tau,t)+\int_{t_0}^{\tau}d\tau'\mathcal{Z}\mathcal{G}(\tau,\tau')\mathcal{V}(\tau',t)\\ =\int_{t_0}^{t}d\tau'\mathcal{Z}\widetilde{\mathcal{G}}(\tau,\tau')\mathcal{U}(\tau',t)\mathcal{Z},\end{aligned} \tag{36b}$$

subjected to the boundary conditions $\mathcal{U}(t_0,t_0)=I$ and $\mathcal{V}(t_0,t)=0$. It is interesting to find that $\mathcal{U}(\tau,t_0)$ and $\mathcal{V}(\tau,t)$ are the generalization of the nonequilibrium Green functions for open quantum systems we previously introduced [10–12,43] to the case involving pairing processes. In terms of the usual definition of nonequilibrium Green functions, they can be expressed as

$$\mathcal{U}(t,t_0)\mathcal{Z}=\begin{bmatrix}\langle[a(t),a^\dagger(t_0)]\rangle & \langle[a(t),a(t_0)]\rangle\\ \langle[a^\dagger(t),a^\dagger(t_0)]\rangle & \langle[a^\dagger(t),a(t_0)]\rangle\end{bmatrix}, \tag{37a}$$

$$\begin{aligned}\mathcal{V}(\tau,t)=&\begin{bmatrix}\langle a^\dagger(t)a(\tau)\rangle & \langle a(t)a(\tau)\rangle\\ \langle a^\dagger(t)a^\dagger(\tau)\rangle & \langle a(t)a^\dagger(\tau)\rangle\end{bmatrix}\\ &-\mathcal{U}(\tau,t_0)\begin{bmatrix}\langle a^\dagger(t_0)a(t_0)\rangle & \langle a(t_0)a(t_0)\rangle\\ \langle a^\dagger(t_0)a^\dagger(t_0)\rangle & \langle a(t_0)a^\dagger(t_0)\rangle\end{bmatrix}\mathcal{U}^\dagger(t,t_0).\end{aligned} \tag{37b}$$

They fully describe the transient dissipation and fluctuation dynamics of open quantum system incorporating various back-actions from the environment, and obey the integro-differential equations of motion (36), which can also be derived from the Heisenberg equation of motion; see the detailed derivation given in Appendix A. The solution of Eq. (36b) for the nonequilibrium noise-induced correlation Green function $\mathcal{V}(\tau,t)$ can be obtained easily,

$$\mathcal{V}(\tau,t)=\int_{t_0}^{\tau}d\tau'\int_{t_0}^{t}dt'\mathcal{U}(\tau,\tau')\mathcal{Z}\widetilde{\mathcal{G}}(\tau',t')\mathcal{Z}\mathcal{U}^\dagger(t,t'), \tag{38}$$

which is indeed the generalization of the Keldysh's correlation Green function with pairing couplings [19]. This solution is the generalized nonequilibrium fluctuation-dissipation theorem [43]; also see the derivation given in Appendix A.

With the equation of motion (34), the A-associated propagating functions become

$$\begin{aligned}\mathcal{J}^A(z_t^*,z_t',t;z_0,z_0'^*,t_0)=&\left\{\begin{bmatrix}\dot z(t)\\ \dot z^*(t)\end{bmatrix}+i\omega_s\mathcal{Z}\begin{bmatrix}z(t)\\ z^*(t)\end{bmatrix}\right\}\\ &\times\mathcal{J}(z_t^*,z_t',t;z_0,z_0'^*,t_0),\end{aligned} \tag{39}$$

and using Eq. (35), the additional term can be fully determined in terms of the fixed end points of the path integrals. From the above result, we obtain the current superoperators Eq. (21) in the coherent state representation

$$\begin{aligned}\langle z^*|\mathcal{L}^+[\rho_s(t)]|z'\rangle =& -\bigg\{[z^*\quad z']\mathcal{K}(t)\begin{bmatrix}\partial_{z^*}\\ z^*\end{bmatrix}\\ &+[z^*\quad z']\Lambda(t)\begin{bmatrix}\partial_{z^*}-z'\\ \partial_{z'}-z^*\end{bmatrix}\bigg\}\langle z^*|\rho_s(t)|z'\rangle,\end{aligned} \tag{40a}$$

$$\begin{aligned}\langle z^*|\mathcal{L}^-[\rho_s(t)]|z'\rangle =& \bigg\{[\partial_{z'}\quad \partial_{z^*}]\mathcal{K}(t)\begin{bmatrix}\partial_{z^*}\\ z^*\end{bmatrix}\\ &+[\partial_{z'}\quad \partial_{z^*}]\Lambda(t)\begin{bmatrix}\partial_{z^*}-z'\\ \partial_{z'}-z^*\end{bmatrix}\bigg\}\langle z^*|\rho_s(t)|z'\rangle,\end{aligned} \tag{40b}$$

where $\mathcal{K}(t)$ and $\Lambda(t)$ are defined as

$$\mathcal{K}(t)\equiv -i\omega_s\mathcal{Z}-\dot{\mathcal{U}}(t,t_0)\mathcal{U}(t,t_0)^{-1}, \tag{41a}$$

$$\Lambda(t)\equiv\dot{\mathcal{V}}(t,t)-\dot{\mathcal{U}}(t,t_0)\mathcal{U}(t,t_0)^{-1}\mathcal{V}(t,t). \tag{41b}$$

Note that here $\dot{\mathcal{V}}(t,t)$ represents only the time derivative with respect to the first parameter.

After some rearrangement, one can rewrite the exact master equation as the standard form as we did before:

$$\begin{aligned}\frac{d}{dt}\rho_s(t)=&\frac{1}{i\hbar}[H_s'(t),\rho_s(t)]\\ &+\gamma(t)\big[a\rho_s(t)a^\dagger-\tfrac{1}{2}a^\dagger a\rho_s(t)-\tfrac{1}{2}\rho_s(t)a^\dagger a\big]\\ &+\widetilde{\gamma}(t)\big[a^\dagger\rho_s(t)a-\tfrac{1}{2}aa^\dagger\rho_s(t)-\tfrac{1}{2}\rho_s(t)aa^\dagger\\ &+a\rho_s(t)a^\dagger-\tfrac{1}{2}a^\dagger a\rho_s(t)-\tfrac{1}{2}\rho_s(t)a^\dagger a\big]\\ &-\overline{\gamma}(t)\big[a^\dagger\rho_s(t)a^\dagger-\tfrac{1}{2}a^{\dagger 2}\rho_s(t)-\tfrac{1}{2}\rho_s(t)a^{\dagger 2}\big]\\ &-\overline{\gamma}^*(t)\big[a\rho_s(t)a-\tfrac{1}{2}a^2\rho_s(t)-\tfrac{1}{2}\rho_s(t)a^2\big],\end{aligned} \tag{42}$$

where $H_s'(t)=\hbar\omega_s'(t)a^\dagger a+\frac{1}{2}\hbar\overline{\omega}_s'(t)a^{\dagger 2}+\frac{1}{2}\hbar\overline{\omega}_s'^*(t)a^2$. Other coefficients are given by

$$\omega_s'(t)=\omega_s-\frac{i}{2}[\mathcal{K}_{11}(t)-\mathcal{K}_{11}^*(t)], \tag{43a}$$

$$\overline{\omega}_s'(t)=-\frac{i}{2}[\mathcal{K}_{12}(t)+\mathcal{K}_{21}^*(t)], \tag{43b}$$

$$\gamma(t)=\mathcal{K}_{11}(t)+\mathcal{K}_{11}^*(t), \tag{43c}$$

$$\widetilde{\gamma}(t)=\Lambda_{11}(t)+\Lambda_{11}^*(t), \tag{43d}$$

$$\overline{\gamma}(t)=\Lambda_{12}(t)+\Lambda_{21}^*(t). \tag{43e}$$

It shows that the coefficients $\omega_s'(t)$ and $\overline{\omega}_s'(t)$ are the renormalized frequency and renormalized pairing strength modified by the environment. By diagonalizing the renormalized Hamiltonian with a Bogoliubov transformation, the renormalized eigenfrequency is $\omega_r(t)=\sqrt{|\omega_s'(t)|^2-|\overline{\omega}_s'(t)|^2}$. The term proportional to $\gamma(t)$ represents a nonunitary evolution that describes the dissipation dynamics induced by the environment. Notice that the definition of $\gamma(t)$ here is twice of that in our previous paper [13] because we have rewritten the superoperator multiplied by a factor 1/2 so that all the superoperators have formally the same standard Lindblad form [44]. As a result, $\gamma(t)$ corresponds precisely to the decay coefficient of the system. Also, there is a term proportional to $\gamma'(t)$ in our previous paper [13] which vanishes here for the single

Brownian particle system. The other two terms proportional to $\widetilde{\gamma}(t)$ and $\overline{\gamma}(t)$ in the master equation describe fluctuation (diffusion) dynamics. They depend on the initial state of the environment. Note that the $\overline{\gamma}(t)$-associated fluctuations are induced by pair production and annihilation between the system and the environment.

To understand further the physical picture of the renormalized energy, the dissipation, and fluctuation dynamics induced by the environment, let us calculate the mean values and their deviations (equivalent to the quadrature covariance in terms of the position and momentum) of the system variables from the exact master equation. From the exact master equation (42), it is easy to find that

$$\frac{d}{dt}\begin{bmatrix}\langle a(t)\rangle\\ \langle a^\dagger(t)\rangle\end{bmatrix} = \begin{bmatrix}-i\omega'_{\mathrm{S}}(t)-\gamma(t)/2 & -i\overline{\omega}'_{\mathrm{S}}(t)\\ i\overline{\omega}'^{*}_{\mathrm{S}}(t) & i\omega'_{\mathrm{S}}(t)-\gamma(t)/2\end{bmatrix}\begin{bmatrix}\langle a(t)\rangle\\ \langle a^\dagger(t)\rangle\end{bmatrix}, \tag{44}$$

where $\langle a(t)\rangle = \mathrm{Tr}_{\mathrm{S}}[a\rho_{\mathrm{S}}(t)]$ and $\langle a^\dagger(t)\rangle = \mathrm{Tr}_{\mathrm{S}}[a^\dagger\rho_{\mathrm{S}}(t)]$. The above equation shows that the Brownian particle undergoes a damping oscillation with the renormalized frequencies $\omega'_{\mathrm{S}}(t)$ and $\overline{\omega}'_{\mathrm{S}}(t)$ and the decay coefficient $\gamma(t)$ induced from the environment. The mean value $\langle a(t)\rangle$ is mixed with its complex conjugation, which is related to the squeezed term $\frac{1}{2}\hbar\overline{\omega}'_{\mathrm{S}}(t)a^{\dagger 2}$ in the renormalized Hamiltonian, due to the pairing coupling between the system and the environment.

On the other hand, the mean-value deviations obey the following equations:

$$\frac{d}{dt}\begin{bmatrix}\Delta n(t) & \Delta s(t)\\ \Delta s^*(t) & \Delta h(t)\end{bmatrix} = \left\{\begin{bmatrix}-i\omega'_{\mathrm{S}}(t)-\gamma(t)/2 & -i\overline{\omega}'_{\mathrm{S}}(t)\\ i\overline{\omega}'^{*}_{\mathrm{S}}(t) & i\omega'_{\mathrm{S}}(t)-\gamma(t)/2\end{bmatrix} \times\begin{bmatrix}\Delta n(t) & \Delta s(t)\\ \Delta s^*(t) & \Delta h(t)\end{bmatrix} + \mathrm{H.c.}\right\} + \begin{bmatrix}\widetilde{\gamma}(t) & \overline{\gamma}(t)\\ \overline{\gamma}^*(t) & \gamma(t)+\widetilde{\gamma}(t)\end{bmatrix}, \tag{45}$$

where $\Delta n(t) = \mathrm{Tr}_{\mathrm{S}}[\delta a^\dagger\delta a\rho_{\mathrm{S}}(t)]$, $\Delta s(t) = \mathrm{Tr}_{\mathrm{S}}[\delta a\delta a\rho_{\mathrm{S}}(t)]$, $\Delta h(t) = \mathrm{Tr}_{\mathrm{S}}[\delta a\delta a^\dagger\rho_{\mathrm{S}}(t)] = 1+\Delta n(t)$, and $\delta a = a - \langle a\rangle$. The first term and its Hermitian conjugate in the right-hand side of the above equation describe a damping oscillation of the quadrature variables, which follows the same dynamics as given by Eq. (44). The last term in Eq. (45) is the noise sources coming from the particle transitions and pair productions between the system associated with the initial state of the environment. Thus, $\widetilde{\gamma}(t)$ and $\overline{\gamma}(t)$ can be interpreted as the diffusion rates.

We also find that the exact master equation (42) derived with the initial decoupled state indeed has the same form as the exact master equation for the initial correlated density matrix $\rho_{\mathrm{tot}}(t_0)$ of Eq. (23) that we have derived in our previous work [i.e., Eq. (12) in Ref. [13]]. In Eq. (23), H_{tot} is the total Hamiltonian of the system and the environment plus their interaction, as given by Eq. (13). Thus, the initial state cannot be decoupled into a direct product of the system state with the environment state. Of course, to let the system and the environment evolve into a nonequilibrium evolution with such an initial thermal state, one needs to quench the system. A simple quench can be made by replacing the system frequency ω_{S} with time-dependent one $\omega_{\mathrm{S}}(t)$, which is easy to be realized in experiments. All the formulas derived in this section remain the same forms with such a quenching modulation. As one can find from [13], with the initial correlated state (23), the only modification in the exact master equation is the fluctuation coefficients $\widetilde{\gamma}(t)$ and $\overline{\gamma}(t)$, which are still determined by the nonequilibrium Green function $\mathcal{V}(t,t)$, but $\mathcal{V}(t,t)$ of Eq. (41) is modified by

$$\mathcal{V}(t,t) = \int_{t_0}^{t} d\tau \int_{t_0}^{t} d\tau' \mathcal{U}(t,\tau)\mathcal{Z}[\widetilde{\mathcal{G}}(\tau,\tau') + \widetilde{\mathcal{G}}^{(\mathrm{ES})}(\tau,\tau') + \widetilde{\mathcal{G}}^{(\mathrm{ES})\dagger}(\tau',\tau)]\mathcal{Z}\mathcal{U}^\dagger(t,\tau'). \tag{46}$$

Here $\widetilde{\mathcal{G}}^{(\mathrm{ES})}(\tau,\tau')$ is given by

$$\widetilde{\mathcal{G}}^{(\mathrm{ES})}(\tau,\tau') = -2i\sum_k \begin{bmatrix}V_k(\tau-t_0) & W_k(\tau-t_0)\\ W_k^*(\tau-t_0) & V_k^*(\tau-t_0)\end{bmatrix} \times\begin{bmatrix}\overline{n}'_k(t_0) & \overline{s}'_k(t_0)\\ \overline{s}'^{*}_k(t_0) & \overline{n}'^{*}_k(t_0)\end{bmatrix}\mathcal{Z}\delta(\tau'-t_0), \tag{47}$$

with $\overline{n}'_k(t_0) = \langle a^\dagger(t_0)b_k(t_0)\rangle$ and $\overline{s}'_k(t_0) = \langle a(t_0)b_k(t_0)\rangle$ being initial correlations between the system and the environment. The factor 2 comes from the integral $\int_{t_0}^{t} d\tau\delta(\tau-t_0) = 1/2$.

For the special case of $W_k = 0$, Eq. (13) is reduced to the Fano Hamiltonian that describes a discrete state coupling to a continuous spectral bath. In this case, the pairing-production or pairing-annihilation related coefficients vanish: $\overline{\gamma}(t) = 0$ and $\overline{\omega}'_{\mathrm{S}}(t) = 0$. Correspondingly, the exact master equation (42) is reduced to the one given in our earlier works [10,13,45–49]. For another special case of $W_k = V_k$ which corresponds to conventional QBM model [4,7], we will discuss the corresponding reduction in detail in the next section.

IV. REPRODUCTION OF THE CONVENTIONAL QBM AND THE RESOLUTION OF THE INITIAL JOLT PROBLEM

In this section we will show that the Hu-Paz-Zhang master equation of the QBM model [7] is a special case of our master equation (42) for the generalized QBM. We will also show that the initial jolt problem in the conventional QBM is artificial, but it has nothing to do with the initial decoupled state.

Without including momentum-dependent couplings, the strength of the coupling and the pairing are the same $W_k = V_k$, as we shown in Eq. (11). This condition leads to the following relations between the renormalized frequency, dissipation (decay), and fluctuation (diffusion) coefficients:

$$\mathrm{Re}[\overline{\omega}'_{\mathrm{S}}(t)] = \omega'_{\mathrm{S}}(t) - \omega_{\mathrm{S}}, \tag{48a}$$

$$\mathrm{Im}[\overline{\omega}'_{\mathrm{S}}(t)] = \gamma(t)/2, \tag{48b}$$

$$\mathrm{Re}[\overline{\gamma}(t)] = -\gamma(t)/2 - \widetilde{\gamma}(t). \tag{48c}$$

Through these relations, the Hu-Paz-Zhang master equation of the conventional QBM can be reproduced easily from our general exact master equation (42) as a special limit. It can be expressed alternatively as

$$
\begin{aligned}
i\hbar\frac{d}{dt}\rho_{\mathrm{S}}(t) = {} & [H_{\mathrm{R}}(t), \rho_{\mathrm{S}}(t)] \\
& + \Gamma(t)\big[x\rho_{\mathrm{S}}(t)p - \tfrac{1}{2}px\rho_{\mathrm{S}}(t) - \tfrac{1}{2}\rho_{\mathrm{S}}(t)px \\
& - p\rho_{\mathrm{S}}(t)x + \tfrac{1}{2}xp\rho_{\mathrm{S}}(t) + \tfrac{1}{2}\rho_{\mathrm{S}}(t)xp\big] \\
& + iM\Gamma(t)h(t)\big[2x\rho_{\mathrm{S}}(t)x - x^2\rho_{\mathrm{S}}(t) - \rho_{\mathrm{S}}(t)x^2\big] \\
& - i\Gamma(t)f(t)\big[x\rho_{\mathrm{S}}(t)p - \tfrac{1}{2}px\rho_{\mathrm{S}}(t) - \tfrac{1}{2}\rho_{\mathrm{S}}(t)px \\
& + p\rho_{\mathrm{S}}(t)x - \tfrac{1}{2}xp\rho_{\mathrm{S}}(t) - \tfrac{1}{2}\rho_{\mathrm{S}}(t)xp\big], \qquad (49)
\end{aligned}
$$

where $H_{\mathrm{R}}(t)$ is the complete renormalized system Hamiltonian,

$$
\begin{aligned}
H_{\mathrm{R}}(t) \equiv {} & \tfrac{1}{2M}p^2 + \tfrac{1}{2}M\omega_{\mathrm{S}}^2x^2 \\
& + \tfrac{1}{2}M\delta\omega_{\mathrm{S}}^2(t)x^2 + \tfrac{1}{2}\,\mathrm{Im}[\overline{\omega}'_{\mathrm{S}}(t)](xp+px). \qquad (50)
\end{aligned}
$$

All the coefficients in the above master equation are deduced from the coefficients of the general master equation (42) under the condition $W_k = V_k$. The results are given by

$$
\delta\omega_{\mathrm{S}}^2(t) = 2\omega_{\mathrm{S}}\,\mathrm{Re}[\overline{\omega}'_{\mathrm{S}}(t)], \quad \Gamma(t) = \gamma(t)/2, \qquad (51a)
$$

$$
\Gamma(t)h(t) = \omega_{\mathrm{S}}\,\mathrm{Re}[\overline{\gamma}(t)], \quad \Gamma(t)f(t) = -\,\mathrm{Im}[\overline{\gamma}(t)]. \qquad (51b)
$$

Notice that in the original Hu-Paz-Zhang master equation, the renormalized system Hamiltonian is incomplete, because not all commutator terms (associated with the unitary evolution) have been separated from the anticommutator terms (nonunitary evolution) in the master equation. This is how our complete renormalized system Hamiltonian is defined, as given by Eq. (50). It shows that the renormalized system Hamiltonian contains a potential of the position-momentum coupling. The reason that this momentum-dependent potential is misplaced into the dissipation (decay) term in the Hu-Paz-Zhang equation is because at the special case $W_k = V_k$, we have $\mathrm{Im}[\overline{\omega}'_{\mathrm{S}}(t)] = \gamma(t)/2$ [see Eq. (48)], so that this renormalization-induced potential has the same coefficient as the dissipation term in the master equation. On the other hand, this renormalization-induced momentum-dependent potential also indicates that including the momentum-dependent couplings between the system and the environment is a natural consequence, because even if only the position-position coupling is considered as in the conventional QBM, the renormalization not only changes the frequency of the system, but also induces a position-momentum coupling potential in the system Hamiltonian. As a consequence of this position-momentum coupling potential, the physical frequency of the Brownian particle is $\omega_{\mathrm{p}}^2(t) = \omega_{\mathrm{S}}^2 + \delta\omega_{\mathrm{S}}^2(t) - \Gamma^2(t)$, which is consistent with the renormalized eigenfrequency $\omega_{\mathrm{r}}(t) = \sqrt{|\omega'_{\mathrm{S}}(t)|^2 - |\overline{\omega}'_{\mathrm{S}}(t)|^2}$ in our general formalism; see the discussion after Eq. (43).

The decay and diffusion of the QBM can be more clearly manifested from the equation of motion for the quadrature covariance variables, which can be found from the master equation (49) directly:

$$
\begin{aligned}
\frac{d}{dt}\begin{bmatrix} \langle\Delta x^2(t)\rangle & \langle\Delta\{xp\}(t)\rangle \\ \langle\Delta\{px\}(t)\rangle & \langle\Delta p^2(t)\rangle \end{bmatrix}
&= \left\{\begin{bmatrix} 0 & 1/M \\ -M\big(\omega_{\mathrm{S}}^2 + \delta\omega_{\mathrm{S}}^2(t)\big) & -2\Gamma(t) \end{bmatrix}\begin{bmatrix} \langle\Delta x^2(t)\rangle & \langle\Delta\{xp\}(t)\rangle \\ \langle\Delta\{px\}(t)\rangle & \langle\Delta p^2(t)\rangle \end{bmatrix}\right. \\
&\quad \left. + \,\mathrm{H.c}\right\} + \begin{bmatrix} 0 & \Gamma(t)f(t) \\ \Gamma(t)f(t) & 2M\Gamma(t)h(t) \end{bmatrix}, \qquad (52)
\end{aligned}
$$

where $\langle\Delta\{xp\}(t)\rangle = \frac{1}{2}\langle\{\Delta x(t), \Delta p(t)\}\rangle$. Thus, the coefficients $\Gamma(t)f(t)$ and $\Gamma(t)h(t)$ can be interpreted as the diffusion rates of the quadrature covariance variables $\langle\Delta\{xp\}(t)\rangle$ and $\langle\Delta p^2(t)\rangle$, respectively. The missing diffusion rate for $\langle\Delta x^2(t)\rangle$ is due to the lack of position-momentum coupling in the conventional QBM Hamiltonian.

Based on the above reproduction of the Hu-Paz-Zhang master equation from our generalized QBM, we would also like to address the issue of "initial jolt" in the conventional QBM [7,50,51]. The initial jolt is associated with a peak at the beginning of the diffusion coefficient $\Gamma(t)h(t)$ at the low-temperature limit in the Hu-Paz-Zhang master equation, which is sensitive to the cutoff frequency Λ for the Ohmic-type spectral density. The inverse of this cutoff frequency, $1/\Lambda$, is the typical timescale of the environment. The amplitude of the initial jolt is proportional to Λ, and it diverges when one takes a constant damping coefficient for the conventional QBM model. This corresponds to an Ohmic bath with an infinite cutoff frequency limit $\Lambda \to \infty$. Such divergence causes covariances $(\Delta p, \Delta x)$ of the Brownian particle to increase immediately at the very beginning of the system evolution. In fact, all of the Ohmic, sub-Ohmic, and supra-Ohmic spectral baths suffer from the same divergence [7,50,51].

In the literature [7,51], it has been argued that such an initial jolt phenomenon is artificial and most likely comes from the assumption of the initial decoupled system-environment state one used in deriving the master equation. Here we prove that it is artificial but is not because of the use of the initial decoupled state. We find that not only the diffusion coefficients in the generalized QBM, but also the decay rate has similar behavior, while the decay rate is independent of the initial system-environment state. This can be seen from the combination of Eqs. (36), (41), and (43). It shows that the decay rate is determined purely by the same dissipation dynamics described by the retarded nonequilibrium Green function (36a), which is independent of the initial system-environment state. This indicates that the problem of "initial jolt" has nothing to do with the initial decoupled state that one often questioned [7,51]. Below we will show how does the serious divergence of the so-called "initial jolt" occurs and why it is not related to the choice of initial system-environment states.

If we plug in the Ohmic spectral density with an infinite cutoff into our exact master equation, the serious divergence occurs in the decay coefficient $\gamma(t)$ and the fluctuation (diffusion) coefficients $\widetilde{\gamma}(t)$, $\overline{\gamma}(t)$. To simplify the formulation, we take $J_v(\omega) \equiv 2\pi\sum_k |V_k|^2\delta(\omega - \omega_k) = \sqrt{\frac{\pi\gamma_0}{2\Lambda}}\omega e^{-\omega/\Lambda}$ and $\omega_{\mathrm{S}} = \sqrt{2\gamma_0\Lambda/\pi}$. This setup corresponds to Ohmic spectral density with an exponential cutoff and zero renormalized frequency in the conventional QBM model. The Fourier transformation of the Ohmic spectral density is given by

$$g_v(t) \equiv \int \frac{d\omega}{2\pi} J_v(\omega) e^{-i\omega t} = \sqrt{\frac{\gamma_0}{8\pi\Lambda}}(it + 1/\Lambda)^{-2}, \tag{53}$$

which has a peak in the timescale $1/\Lambda$. This function is related to the integral kernel in the time-convolution equation for the generalized nonequilibrium Green function (36), which depicts back-action processes between the system and the environment. The strength and the timescale of memory effect of the system are directly related to the cutoff frequency Λ, which is the key to making the decay and diffusion coefficients diverge.

To show how this divergence is purely associated with the cutoff frequency introduced in the Ohmic spectral densities, we evaluate the decay and diffusion coefficients $\gamma(t)$ and $\widetilde{\gamma}(t)$ by varying the ratio of transition coupling strength and pairing strength $|W_k/V_k| = \alpha$. Using the Heisenberg equation of motion, one can easily find the equation of motion for the average occupation and the squeezed parameter in an arbitrary Brownian particle state,

$$\frac{d}{dt}\begin{bmatrix} n(t) & s(t) \\ s^*(t) & h(t) \end{bmatrix} = \Bigg\{ -i\omega_S \mathcal{Z} \begin{bmatrix} n(t) & s(t) \\ s^*(t) & h(t) \end{bmatrix} - i\mathcal{Z}\begin{bmatrix} V_k & W_k \\ W_k^* & V_k^* \end{bmatrix}\begin{bmatrix} n'_k(t) & s'_k(t) \\ s'^*_k(t) & n'^*_k(t) \end{bmatrix} + \text{H.c.} \Bigg\}, \tag{54}$$

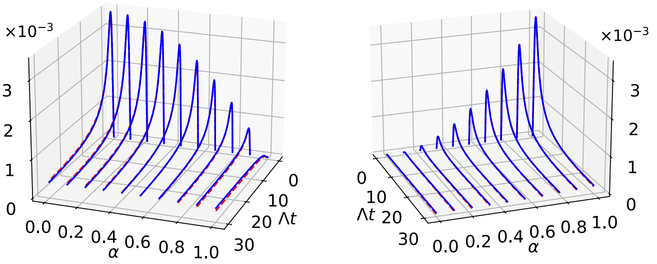
where $n'_k(t) = \langle a^\dagger(t) b_k(t)\rangle$ and $s'_k(t) = \langle a(t) b_k(t)\rangle$ are correlations between the system and the environment. The first term represents the free evolution of the system, and the last term represents the generalized information current flowing into the system. The later can be expressed explicitly as

$$\begin{aligned} \mathcal{I}(t) &\equiv \frac{d}{dt}\mathcal{N}(t,t) + [i\omega_S \mathcal{Z}\mathcal{N}(t,t) + \text{H.c.}] \\ &= \int_{t_0}^{t} d\tau [\mathcal{Z}\widetilde{\mathcal{G}}(t,\tau)\mathcal{Z}\mathcal{U}^\dagger(t,\tau) - \mathcal{Z}\mathcal{G}(t,\tau)\mathcal{N}(\tau,t)] + \text{H.c.} \end{aligned} \tag{55}$$

after solving the environmental equation of motion. Here $\mathcal{N}(\tau,t) \equiv \left[\begin{smallmatrix} \langle a^\dagger(t)a(\tau)\rangle & \langle a(t)a(\tau)\rangle \\ \langle a^\dagger(t)a^\dagger(\tau)\rangle & \langle a(t)a^\dagger(\tau)\rangle \end{smallmatrix}\right]$ is the quadrature correlation matrix of the average occupation and the squeezed parameter. The first part in the integration is the diffusion current due to the initial distribution of the environment. The second part is the dissipation current due to the initial state of the system, and the backflow from the system. Comparing Eq. (45) with (55), the diffusion and decay coefficients $\widetilde{\gamma}(t)$, $\overline{\gamma}(t)$ and $\gamma(t)$ can be also expressed as

$$\begin{aligned} &\begin{bmatrix} \widetilde{\gamma}(t) & \overline{\gamma}(t) \\ \overline{\gamma}^*(t) & \gamma(t) + \widetilde{\gamma}(t) \end{bmatrix} \\ &\quad = \int_{t_0}^{t} d\tau [\mathcal{Z}\widetilde{\mathcal{G}}(t,\tau)\mathcal{U}(\tau,t)\mathcal{Z} - \mathcal{Z}\mathcal{G}(t,\tau)\mathcal{V}(\tau,t)] \\ &\quad + \mathcal{K}(t)\mathcal{V}(t,t) + \text{H.c.} \end{aligned} \tag{56}$$

At the beginning of the evolution, due to the initial condition $\mathcal{V}(t_0,t_0) = 0$ and $\mathcal{U}(t_0,t_0) = I$, we need to consider only the first part in the integration, and the off-diagonal terms of $\mathcal{U}(\tau,t)$ can be ignored. Then $\gamma(t)$ and $\widetilde{\gamma}(t)$ in the timescale $1/\Lambda$ can be approximated as



(a) (b)

FIG. 3. (a) The decay coefficient $\gamma(t)/\Lambda$ and (b) the fluctuation coefficient $\widetilde{\gamma}(t)/\Lambda$ in the time domain for different values of $\alpha = |W_k/V_k|$, the rate of the pairing strength with the single-particle coupling between the system and the environment in the generalized QBM. The exact solution (blue solid line) from Eq. (43) and the estimated solution (red dashed line) based on Eq. (57) agree with each other. Here we set $\gamma_0 = 0.0003\Lambda$, $k_B T/\hbar = 0.01\Lambda$. They are plotted against timescale $1/\Lambda$ to emphasize the initial jolt. The results show that the initial jolt exists in both the decay coefficient $\gamma(t)$ and the diffusion coefficient $\widetilde{\gamma}(t)$.

$$\widetilde{\gamma}(t) \approx 2\,\text{Re}\left[\int_{t_0}^{t} d\tau g_w(t,\tau) u_S(t,\tau)\right], \tag{57a}$$

$$\gamma(t) \approx 2\,\text{Re}\left\{\int_{t_0}^{t} d\tau [g_v^*(t,\tau) - g_w(t,\tau)] u_S(t,\tau)\right\}, \tag{57b}$$

where $u_S(t,\tau) \equiv \langle [a(t), a^\dagger(\tau)]\rangle = \mathcal{U}_{11}(t,\tau)$ and $g_w(t,\tau) \equiv \sum_k |W_k|^2 e^{-i\omega_k(t-\tau)} = \alpha^2 g_v(t,\tau)$. Here we discuss only the low-temperature case $k_B T/\hbar \ll \Lambda$, so the terms proportional to $\tilde{g}_v(t,\tau) \equiv \sum_k |V_k|^2 \overline{n}_k(t_0) e^{-i\omega_k(t-\tau)}$ and $\tilde{g}_w(t,\tau) \equiv \sum_k |W_k|^2 \overline{n}_k(t_0) e^{-i\omega_k(t-\tau)}$ have also been dropped.

We show numerically that the estimated results of the diffusion and decay coefficients $\widetilde{\gamma}(t)$ and $\gamma(t)$ calculated from Eq. (57) are almost the same as the exact solutions of Eq. (43) for all values of α: $0 \leqslant \alpha \leqslant 1$ at low temperature with a high cutoff frequency; see Fig. 3. When $\alpha = 1$, the amplitudes of $g_v(t,\tau)$ and $g_w(t,\tau)$ become the same, then the divergences in the decay coefficient are canceled, as analytically shown by Eq. (57b). Figure 3(a) clearly shows that for $\alpha = 1$, the decay coefficient $\gamma(t)$ suddenly increases only slightly within the timescale $1/\Lambda$, but then it approaches γ_0 instead of decreasing. In other words, no "initial jolt" occurs in the dissipation $\gamma(t)$ for the conventional QBM because of the accident cancellation of the divergences. For any other α values (which is more realistic in quantum optics), the divergence cannot be fully canceled so that the decay coefficient $\gamma(t)$ also shows the "initial jolt," as given in Fig. 3(a), because the decay coefficient $\gamma(t)$ is independent of the initial system-environment state. We conclude that the "initial jolt" has nothing to do with the initial decoupled state one used in deriving the exact master equation.

With the finding of this accident cancellation of the divergences in the decay coefficient $\gamma(t)$ due to the special choice of $\alpha = 1$ in the QBM, the origin of the initial jolt becomes clear. It comes purely from the Ohmic-type spectral densities in the absence of high-frequency cutoff $\Lambda \to \infty$. In fact, it has been known since the 1980s that logarithmic divergence of the momentum dispersion of a free Brownian particle always occurs in the strictly Ohmic limit [1,2]. For a fundamental renormalizable theory, one requires that any renormalized quantity must be independent of regularization scheme, so that the high-energy cutoff scale can be taken to the infinite limit in the calculations of energy. However, both the conventional and the generalized QBM are built on phenomenological model Hamiltonians, no such renormalizability is required *a priori*. Furthermore, the cutoff frequency in the Ohmic-type spectral densities is introduced to count the effective transition processes between the system and the environment. Quantum mechanically, the most active environmental modes ω_k that effectively couple to the system are dominated by these not far away from the thermal frequency $k_B T/\hbar$ so that physical transition between the system and the environment can occur (with large probabilities). Physically, the spectral density is a summation of the probabilities over the physical processes between the system and the environment. For the Ohmic-type spectral densities, taking a very large cutoff frequency implies that the environmental modes with higher energy will have the larger probability amplitude for coupling to the system mode. This is obviously unphysical. The physical reliable cutoff frequency should be estimated by the condition that the mean frequency of a given spectral density should be the same order of the system-characterized frequency, if the system-environment coupling cannot be derived from a more fundamental theory or no experimental data can be used to fix it. Under such a physical requirement, the problem of the "initial jolt" will never occur.

V. DISCUSSION AND PERSPECTIVES

In this paper we provide a detailed analysis of system-environmental couplings which are usually unknown *a priori* but are the key to the determination of dissipation and fluctuation dynamics in open quantum systems. In particular, we show that the quantum dissipative dynamics described by Feynman-Vernon model and Caldeira-Leggett model based on classical mechanics involve some processes that should barely occur in quantum mechanics. We further show that one can get rid of such processes by including momentum-dependent system-environment couplings. In particular, we generalize the QBM to include such momentum-dependent couplings, which are indeed easier to be realized in quantum optics. We derive the exact master equation of such generalized QBM for both the initially decoupled and initially correlated system-environment states.

With the generalized QBM and its exact master equation for both the initially decoupled and the initially correlated state, we reproduce the Hu-Paz-Zhang master equation as a special case. We then find that in the Hu-Paz-Zhang equation for the conventional QBM with the position-dependent coupling only, the renormalized Brownian particle Hamiltonian actually contains a renormalization-induced momentum-dependent potential. This momentum-dependent potential is misplaced into the dissipation term in the Hu-Paz-Zhang equation, so that the correct renormalized Brownian particle Hamiltonian was not found before. The environment induced momentum-dependent potential in the Hu-Paz-Zhang equation also indicates that including the momentum-dependent system-environment coupling is a natural consequence for the QBM.

Furthermore, we reexamine the problem of the initial jolt in the conventional QBM and Hu-Paz-Zhang master equation. The initial jolt is related to the divergence arising from the Ohmic-type spectral density with an infinite cutoff frequency. It has been thought that the initial jolt is a result of using the initial decoupled state, which is physically unacceptable, but we find that this is indeed a misunderstanding. The misunderstanding comes from the accident cancellation of the divergences in the decay coefficient in the Hu-Paz-Zhang equation, due to the same coupling strengths for the single-particle transition and the pairing processes. With the generalized QBM including the momentum-dependent coupling, we show that the initial jolt exists in both the decay and diffusion coefficients, while the decay coefficient is independent of the initial system-environment state. As a conclusion, the so-called "initial jolt" has nothing to do with the initial decoupled state. It is an artificial effect when one takes an extremely large cutoff frequency in comparison with the thermal energy of the environment, which is unphysical as we explained at the end of the last section.

With the generalized QBM, we have resolved these interesting and important problems for quantum dissipative dynamics, as we discussed in this work. Our exact master equation for the generalized QBM also has potential applications to photonics quantum computing. With an extension of the generalized QBM to multimodes, one can apply it to the integrated quantum processor made of many squeezers and interferometers with 100 more photonic modes that has currently attracted great attention in the development of quantum technology. We will leave this problem for further investigation.

ACKNOWLEDGMENTS

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APPENDIX A: THE GENERALIZED NONEQUILIBRIUM GREEN FUNCTIONS FOR OPEN QUANTUM SYSTEMS WITH PAIRING COUPLINGS AND FLUCTUATION-DISSIPATION THEOREM

The nonequilibrium Green functions we introduced for open quantum systems [10–12,43] are generalized to open quantum systems involving pairing couplings in this work, and are denoted by $\mathcal{U}(\tau, t_0)$ and $\mathcal{V}(\tau, t)$ [13]. They are defined as

$$\mathcal{U}(t, t_0)\mathcal{Z} = \begin{bmatrix} \langle [a(t), a^\dagger(t_0)] \rangle & \langle [a(t), a(t_0)] \rangle \\ \langle [a^\dagger(t), a^\dagger(t_0)] \rangle & \langle [a^\dagger(t), a(t_0)] \rangle \end{bmatrix}, \tag{A1}$$

$$\mathcal{V}(\tau, t) = \begin{bmatrix} \langle a^\dagger(t)a(\tau) \rangle & \langle a(t)a(\tau) \rangle \\ \langle a^\dagger(t)a^\dagger(\tau) \rangle & \langle a(t)a^\dagger(\tau) \rangle \end{bmatrix} - \text{(initial-dependent part)}, \tag{A2}$$

and can be easily determined directly from Heisenberg equation of motion for the generalized QBM Hamiltonian Eq. (13),

$$\frac{d}{dt}\begin{bmatrix} a(t) \\ a^\dagger(t) \end{bmatrix} = -i\omega_{\rm S}\mathcal{Z}\begin{bmatrix} a(t) \\ a^\dagger(t) \end{bmatrix} - i\sum_k \mathcal{Z}\begin{bmatrix} V_k & W_k \\ W_k^* & V_k^* \end{bmatrix}\begin{bmatrix} b_k(t) \\ b_k^\dagger(t) \end{bmatrix}, \tag{A3a}$$

$$\frac{d}{dt}\begin{bmatrix} b_k(t) \\ b_k^\dagger(t) \end{bmatrix} = -i\omega_k\mathcal{Z}\begin{bmatrix} b_k(t) \\ b_k^\dagger(t) \end{bmatrix} - i\mathcal{Z}\begin{bmatrix} V_k^* & W_k \\ W_k^* & V_k \end{bmatrix}\begin{bmatrix} a(t) \\ a^\dagger(t) \end{bmatrix}. \tag{A3b}$$

These equations of motion determine all the dissipation and fluctuation dynamics of the Brownian particle.

Formally, solving Eq. (A3b), we obtain

$$\begin{bmatrix} b_k(t) \\ b_k^\dagger(t) \end{bmatrix} = \begin{bmatrix} e^{-i\omega_k(t-t_0)} & 0 \\ 0 & e^{+i\omega_k(t-t_0)} \end{bmatrix}\begin{bmatrix} b_k(t_0) \\ b_k^\dagger(t_0) \end{bmatrix} - i\int_{t_0}^{t} d\tau\,\mathcal{Z}\begin{bmatrix} V_k^*(\tau-t) & W_k(\tau-t) \\ W_k^*(\tau-t) & V_k(\tau-t) \end{bmatrix}\begin{bmatrix} a(\tau) \\ a^\dagger(\tau) \end{bmatrix}. \tag{A4}$$

This formal solution allows us to eliminate exactly and completely all the environmental degrees of freedom (equivalent to the trace over all the environment states). Substituting this solution into Eq. (A3a), we have

$$\frac{d}{dt}\begin{bmatrix} a(t) \\ a^\dagger(t) \end{bmatrix} + i\omega_{\rm S}\mathcal{Z}\begin{bmatrix} a(t) \\ a^\dagger(t) \end{bmatrix} + \int_{t_0}^{t} d\tau\,\mathcal{Z}\mathcal{G}(t,\tau)\begin{bmatrix} a(\tau) \\ a^\dagger(\tau) \end{bmatrix} = -i\sum_k \mathcal{Z}\begin{bmatrix} V_k(t-t_0) & W_k(t-t_0) \\ W_k^*(t-t_0) & V_k^*(t-t_0) \end{bmatrix}\begin{bmatrix} b_k(t_0) \\ b_k^\dagger(t_0) \end{bmatrix}, \tag{A5}$$

where

$$\mathcal{G}(\tau,\tau') = \sum_k \begin{bmatrix} V_k(\tau-t_0) & W_k(\tau-t_0) \\ W_k^*(\tau-t_0) & V_k^*(\tau-t_0) \end{bmatrix} \times \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}\begin{bmatrix} V_k^*(\tau'-t_0) & W_k(\tau'-t_0) \\ W_k^*(\tau'-t_0) & V_k(\tau'-t_0) \end{bmatrix} \tag{A6}$$

is Eq. (31a). One can clearly see that Eq. (A5) is the generalized quantum Langevin equation, in which the third term in the left-hand side is the dissipation (decay) term due to the system-environment coupling, and the term in the right-hand side is the fundamental noise force generated by the environment through the system-environment coupling. It is obvious that the average of the noise force is zero if the environment is initially in a thermal state.

Because Eq. (A5) is a linear differential equation, its general solution has the form

$$\begin{bmatrix} a(t) \\ a^\dagger(t) \end{bmatrix} = \mathcal{U}(t,t_0)\begin{bmatrix} a(t_0) \\ a^\dagger(t_0) \end{bmatrix} + \begin{bmatrix} F(t) \\ F^\dagger(t) \end{bmatrix}. \tag{A7}$$

Substituting this general solution into Eq. (A5), one can find that the retarded Green function $\mathcal{U}(t,t_0)$ describes the dissipation dynamics of the Brownian particle and obeys the Dyson equation of motion,

$$\frac{d}{d\tau}\mathcal{U}(\tau,t_0) + i\omega_{\rm S}\mathcal{Z}\mathcal{U}(\tau,t_0) + \int_{t_0}^{\tau} d\tau'\,\mathcal{Z}\mathcal{G}(\tau,\tau')\mathcal{U}(\tau',t_0) = 0, \tag{A8}$$

which is just Eq. (36a). The integral kernel in the above equation characterizes the non-Markovian dissipation processes. It is easy to show that the definition of the retarded Green function $\mathcal{U}(t,t_0)$ in Eq. (A7) is the same as the definition of Eq. (A1),

$$\begin{aligned} \begin{bmatrix} \langle [a(t),a^\dagger(t_0)]\rangle & \langle [a(t),a(t_0)]\rangle \\ \langle [a^\dagger(t),a^\dagger(t_0)]\rangle & \langle [a^\dagger(t),a(t_0)]\rangle \end{bmatrix} &= \mathcal{U}(t,t_0)\begin{bmatrix} \langle [a(t_0),a^\dagger(t_0)]\rangle & \langle [a(t_0),a(t_0)]\rangle \\ \langle [a^\dagger(t_0),a^\dagger(t_0)]\rangle & \langle [a^\dagger(t_0),a(t_0)]\rangle \end{bmatrix} \\ &= \mathcal{U}(t,t_0)\mathcal{Z}. \end{aligned} \tag{A9}$$

The last term in Eq. (A7) describes the environment-induced fluctuation (noise) dynamics that obeys the following equation of motion:

$$\frac{d}{dt}\begin{bmatrix} F(t) \\ F^\dagger(t) \end{bmatrix} + i\omega_{\rm S}\mathcal{Z}\begin{bmatrix} F(t) \\ F^\dagger(t) \end{bmatrix} + \int_{t_0}^{t} d\tau\,\mathcal{Z}\mathcal{G}(t,\tau)\begin{bmatrix} F(\tau) \\ F^\dagger(\tau) \end{bmatrix} = -i\sum_k \mathcal{Z}\begin{bmatrix} V_k(t-t_0) & W_k(t-t_0) \\ W_k^*(t-t_0) & V_k^*(t-t_0) \end{bmatrix}\begin{bmatrix} b_k(t_0) \\ b_k^\dagger(t_0) \end{bmatrix}, \tag{A10}$$

subjected to the initial condition $[{F(t_0) \atop F^\dagger(t_0)}] = 0$. From this initial condition, the general solution of the equation for the noise dynamics is given explicitly,

$$\begin{bmatrix} F(t) \\ F^\dagger(t) \end{bmatrix} = -i\sum_k\int_{t_0}^{t} d\tau\,\mathcal{U}(t,\tau)\mathcal{Z} \times \begin{bmatrix} V_k(\tau-t_0) & W_k(\tau-t_0) \\ W_k^*(\tau-t_0) & V_k^*(\tau-t_0) \end{bmatrix}\begin{bmatrix} b_k(t_0) \\ b_k^\dagger(t_0) \end{bmatrix}. \tag{A11}$$

The nonequilibrium correlation functions are defined by

$$\begin{aligned} \begin{bmatrix} \langle a^\dagger(t)a(\tau)\rangle & \langle a(t)a(\tau)\rangle \\ \langle a^\dagger(t)a^\dagger(\tau)\rangle & \langle a(t)a^\dagger(\tau)\rangle \end{bmatrix} &= \mathcal{U}(\tau,t_0)\begin{bmatrix} \langle a^\dagger(t_0)a(t_0)\rangle & \langle a(t_0)a(t_0)\rangle \\ \langle a^\dagger(t_0)a^\dagger(t_0)\rangle & \langle a(t_0)a^\dagger(t_0)\rangle \end{bmatrix}\mathcal{U}^\dagger(t,t_0) \\ &\quad + \begin{bmatrix} \langle F^\dagger(t)F(\tau)\rangle & \langle F(t)F(\tau)\rangle \\ \langle F^\dagger(t)F^\dagger(\tau)\rangle & \langle F(t)F^\dagger(\tau)\rangle \end{bmatrix}. \end{aligned} \tag{A12}$$

The last term is the noise-induced correlation Green function $\mathcal{V}(\tau,t)$, which has the solution

$$\mathcal{V}(\tau,t) = \begin{bmatrix} \langle F^\dagger(t)F(\tau)\rangle & \langle F(t)F(\tau)\rangle \\ \langle F^\dagger(t)F^\dagger(\tau)\rangle & \langle F(t)F^\dagger(\tau)\rangle \end{bmatrix} = \int_{t_0}^{\tau} d\tau'\int_{t_0}^{t} dt'\,\mathcal{U}(\tau,\tau')\mathcal{Z}\widetilde{\mathcal{G}}(\tau',t')\mathcal{Z}\mathcal{U}^\dagger(t,t'). \tag{A13}$$

This gives the same solution Eq. (38) obtained in our exact master equation derivation. The initial-state-dependent system-environment correlation function $\widetilde{\mathcal{G}}(t,t')$ is given by

$$\widetilde{\mathcal{G}}(\tau',t') = \sum_k \begin{bmatrix} V_k(\tau'-t_0) & W_k(\tau'-t_0) \\ W_k^*(\tau'-t_0) & V_k^*(\tau'-t_0) \end{bmatrix} \times \begin{bmatrix} \langle b_k^\dagger(t_0)b_k(t_0)\rangle & \langle b_k(t_0)b_k(t_0)\rangle \\ \langle b_k^\dagger(t_0)b_k^\dagger(t_0)\rangle & \langle b_k(t_0)b_k^\dagger(t_0)\rangle \end{bmatrix} \times \begin{bmatrix} V_k^*(t'-t_0) & W_k(t'-t_0) \\ W_k^*(t'-t_0) & V_k(t'-t_0) \end{bmatrix}, \tag{A14}$$

which is Eq. (31b). The solution Eq. (A13) is also the generalization of the Keldysh's correlation function in the nonequilibrium technique [19]. It is indeed the generalized nonequilibrium fluctuation-dissipation theorem, as a consequence of the unitarity principle of quantum mechanics for the whole system (the system plus the environment) [43]. The usual equilibrium fluctuation-dissipation theorem can be easily obtained from this solution as a steady-state limit.

APPENDIX B: DERIVATION OF THE FIRST QUANTIZATION HAMILTONIAN (11) FROM THE SECOND QUANTIZATION HAMILTONIAN (3) FOR QUANTUM BROWNIAN MODEL

When the Brownian particle is specified as a simple harmonic oscillator, namely, setting $V(x) = \frac{1}{2}m\omega_S^2x^2$ in Eqs. (1) and (2), the system Hamiltonian in the general second quantization scheme is given by

$$H_S = \sum_n \varepsilon_n a_n^\dagger a_n \quad \text{with} \quad \varepsilon_n = \hbar\omega_S(n+1/2), \tag{B1}$$

as shown in Eq. (3). Here $a_n^\dagger$ and a_n are defined as the creation and annihilation operators of the single-particle energy eigenstate,

$$|n\rangle = a_n^\dagger|0\rangle, \quad a_n|0\rangle = 0, \tag{B2}$$

and they obey the commutation relations,

$$[a_n, a_{n'}^\dagger] = \delta_{nn'}, \quad [a_n^\dagger, a_{n'}^\dagger] = [a_n, a_{n'}] = 0. \tag{B3}$$

On the other hand, for the harmonic oscillator, one usually uses the following particle number representation:

$$a^\dagger = \sqrt{\frac{M\omega_S}{2\hbar}}\left(x - \frac{i}{M\omega_S}p\right), \quad a = \sqrt{\frac{M\omega_S}{2\hbar}}\left(x + \frac{i}{M\omega_S}p\right), \tag{B4}$$

and the energy eigenstate is given by

$$|n\rangle = \frac{1}{\sqrt{n!}}(a^\dagger)^n|0\rangle, \quad a|0\rangle = 0. \tag{B5}$$

Obviously, the operators $a_n^\dagger$ and $a^\dagger$ are very different but must be related to each other. In fact, from Eq. (B2) and Eq. (B5), which are true for arbitrary integer n, one can find the relation

$$a_n^\dagger = \frac{1}{\sqrt{n!}}(a^\dagger)^n. \tag{B6}$$

It shows that the creation operator $a_n^\dagger$ (creating the energy eigenstate $|n\rangle$ from the vacuum) is equivalent to the product of the n creation operator $a^\dagger$, and each creates a energy quanta $\hbar\omega_S$. Thus, the harmonic oscillator Hamiltonian can also be written as

$$H_S = \sum_n \varepsilon_n \frac{1}{n!}(a^\dagger)^n(a)^n. \tag{B7}$$

Using the relation $a|n\rangle = \sqrt{n}|n-1\rangle$, we have

$$(a)^n|n\rangle = \sqrt{n!}|0\rangle \tag{B8}$$

so that

$$\begin{aligned}\varepsilon_n\frac{1}{n!}(a^\dagger)^n(a)^n|n\rangle &= \varepsilon_n\frac{1}{\sqrt{n!}}(a^\dagger)^n|0\rangle = \varepsilon_n|n\rangle \\ &= \hbar\omega_S(n+1/2)|n\rangle \\ &= \hbar\omega_S(a^\dagger a + 1/2)|n\rangle.\end{aligned} \tag{B9}$$

This relation is true for arbitrary integer n. This indicates that for the harmonic oscillator,

$$H_S = \sum_n \varepsilon_n a_n^\dagger a_n = \hbar\omega_S(a^\dagger a + 1/2). \tag{B10}$$

They give the same energy spectrum with the corresponding eigenstates so that they must be equivalence.

Furthermore, for the system part in the system-environment coupling, $F_k(x) = C_k x$, its second quantization is given in Eq. (3) as

$$F_k(x) = C_k x = \sum_{nm} V_{mnk} a_m^\dagger a_n. \tag{B11}$$

For a harmonic oscillator,

$$\begin{aligned}V_{mnk} &= C_k\langle m|x|n\rangle \\ &= \frac{\hbar}{\sqrt{2M\omega_S}}(\sqrt{n+1}\delta_{m,n+1} + \sqrt{n}\delta_{m,n-1})\end{aligned} \tag{B12}$$

so that

$$F_k(x) = \sum_n \frac{\hbar}{\sqrt{2M\omega_S}}(\sqrt{n+1}a_{n+1}^\dagger a_n + \sqrt{n}a_{n-1}^\dagger a_n). \tag{B13}$$

Using the relation (B6) and the identity $\frac{1}{n!}(a^\dagger)^n(a)^n|n\rangle = |n\rangle$ given in Eq. (B9), we have

$$\begin{aligned}&(\sqrt{n+1}a_{n+1}^\dagger a_n + \sqrt{n}a_{n-1}^\dagger a_n)|n\rangle \\ &= \left[\sqrt{n+1}\frac{(a^\dagger)^{n+1}}{\sqrt{(n+1)!}}\frac{(a)^n}{\sqrt{n!}} + \sqrt{n}\frac{(a^\dagger)^{n-1}}{\sqrt{(n-1)!}}\frac{(a)^n}{\sqrt{n!}}\right]|n\rangle \\ &= (a^\dagger + a)|n\rangle\end{aligned} \tag{B14}$$

for arbitrary integer n. This leads to the following relation between the different quantization forms for the operator $F_k(x) = C_k x$:

$$\begin{aligned}F_k(x) &= \sum_n \frac{\hbar}{\sqrt{2M\omega_S}}(\sqrt{n+1}a_{n+1}^\dagger a_n + \sqrt{n}a_{n-1}^\dagger a_n) \\ &= \frac{\hbar}{\sqrt{2M\omega_S}}(a^\dagger + a).\end{aligned} \tag{B15}$$

Moreover, any arbitrary state $|\psi\rangle$ can always be expressed as a superposition of the energy eigenstate: $|\psi\rangle = \sum_n c_n|n\rangle$, it is easy to prove that the two quantization forms of all operators are always equivalent for a harmonic oscillator. This provides the general derivation of Eq. (11) from Eq. (3). If the system is not a harmonic oscillator, the definition (B4) cannot simplify the problem, so one can use only the general second quantization form of Eq. (3).

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