Dynamics of a small quantum system open to a bath with thermostat

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We investigate dynamics of a small quantum system open to a bath with thermostat. We introduce another bath, called super bath, weakly coupled with the bath to provide it with thermostat, which has either the Lindblad or Redfield type. We treat the interaction between the system and bath via a rigorous perturbation theory. Due to the thermostat, the bath behaves dissipative and stochastic, for which the usual Born-Markov assumption is not needed. We consider a specific example of a harmonic oscillator system, and a photonic bath in a large container, and a super bath of the Caldeira-Legget oscillators distributed on the inner surface of the container. We use the P-representation for the total harmonic system. We derive the reduced time-evolution equation for the system by explicitly finding the correlation between the system and bath beyond the product state, that was not obtainable in the previous theory for the system and bath isolated from environment, and marginalizing bath degrees of freedom. Remarkably, the associated dynamic equation for the system density matrix is of the same form as the Redfield master equation with different coefficients depending on thermostat used. We find steady state does not depend on thermostat, but time-dependent state does, that agrees with common expectation. We expect to apply our theory to general systems. Unlike the usual quantum master equations, our reduced dynamics allows investigation for time-dependent protocols and non-equilibrium quantum stochastic dynamics will be investigated in future.

Open quantum systems have usually been studied in the paradigm that a composite system (S) and bath (B), denoted by SB, is isolated from environment [1–3]. Through marginalization of infinite bath degrees of freedom, the Redfield (RF) and Lindbald (LB) quantum master equations have been derived via the Born-Markov assumption [4–8]. They have been applied to and many other fields such as quantum optics [9–13], transport problems in quantum circuits [14–17], quantum heat engines [18–21]. There have been fundamental issues arisen such as complete positivity [22–28], local versus global coupling [29, 30], non-equilibrium quantum fluctuations [31–37].

In many experimental situations, a composite SB is open to environment in such a way that B is coupled to a more extensive bath, called a super bath (SU), but S remains apart from SU. We need an extended theory for this new paradigm of tripartite systems, which has been considered in a recent study from the perspective of the relation to the previous paradigm [38].

In this study, we investigate such a tripartite system in detail by dealing with the interaction between B and SU via Born-Markov assumption. Through marginalization of SU degrees of freedom, B is provided with a thermostat, by which B can be thermalized by itself and S is in turn expected to be such via interaction between the two. There arises a fundamental issue on the local versus global coupling scheme for the interaction between SB and SU. From the previous theories, if the interaction between S and B is so weak as to be treated perturbatively, the two coupling schemes are approximately equivalent [29, 30]. We observe typical interactions between S and B intrinsically weak enough. We apply the local coupling scheme to marginalize SU degrees of freedom. The global coupling theory requires the knowledge of whole composite eigenstates of SB, which is complicated to find due to infinite B degrees of freedom but can be found for weak interaction limit. We will report the comparison study between the two coupling schemes in a near future.

We consider that B fills an extensive volume V in which S is isolated apart from the boundary ∂V and that B interacts with SU at ∂V which is kept in equilibrium at inverse temperature β. Let $\hat{H}_{\rm S}$, $\hat{H}_{\rm B}$, and $\hat{H}_{\rm SU}$ be respectively the Hamiltonians of the three systems. Let $\hat{H}_{\rm I}$ and $\hat{H}_{\text{I-BSU}}$ be respectively interaction Hamiltonians between S and B, and B and SU. Let $\hat{H}_{\rm B}$ and $\hat{H}_{\rm SU}$ be quadratic in their own observables. Let interaction Hamiltonians be bilinear such that $\hat{H}_{\rm I} = \hat{S}\otimes\hat{B}$ and $\hat{H}_{\text{I-BSU}} = \hat{B}\otimes\hat{U}$ where $\hat{S}$, $\hat{B}$, $\hat{U}$ denote observables for S, B, SU, respectively.

We follow the conventional weak coupling theory for the whole interaction $\hat{H}_{\rm I} + \hat{H}_{\text{I-BSU}}$ [7]. Taking the trace over SU states, the cross terms coming from $\hat{H}_{\rm I}(t)$ and $\hat{H}_{\text{I-BSU}}(t')$ vanishes. Then, we obtain the time-evolution equation for the density matrix of SB

$$\dot{\hat{\rho}}_{\rm SB} = -\frac{i}{\hbar}\left[\hat{H}_{\rm S} + \hat{H}_{\rm B} + \hat{H}_{\rm I}, \hat{\rho}_{\rm SB}\right] + \mathcal{D}_{\rm B}^{\rm X}[\hat{\rho}_{\rm SB}] \qquad (1)$$

where $\mathcal{D}_{\rm B}^{\rm X}$ is the dissipator (superoperator) of only B's operators. acting on $\hat{\rho}_{\rm SB}$. B is provided with a thermostat via dissipator of type X, either RF or LB. See the derivation in detail in Sec. I, Supplementary Material (Suppl.). In the absence of S, the equation leads to the quantum master equation for B, either the RF or LB equation. The appearance of $\mathcal{D}_{\mathrm{B}}^{\mathrm{X}}$ makes difference from the previous theory for isolated SB.

In the following, we investigate Eq. (1) via the rigorous perturbation theory for small $\hat{H}_{\mathrm{I}}$ without using the Born-Markov assumption. B behaves dissipative due to inherent thermostat operated by $\mathcal{D}_{\mathrm{B}}^{\mathrm{X}}$, while it is assumed to remain in equilibrium in the previous theory for isolated SB. We focus ourselves on a particular case in which S is a single harmonic oscillator located at the center of a cavity of large volume V, B is composed of photons in the cavity, and SU composed of Caldeira-Legett (CL) oscillators [39] on the inner surface ∂V.

We have $\hat{H}_{\mathrm{B}} = \sum_{\mathbf{k}\lambda} \hbar\omega_k \hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda}$ for $\omega_k = |\mathbf{k}|c$ and $\lambda = 1,2$ denote two polarization directions given wave vector $\mathbf{k}$. $\hat{b}_{\mathbf{k}\lambda}$, $(\hat{b}^\dagger_{\mathbf{k}\lambda})$ is an annihilation (creation) operator for photons with mode given by $\mathbf{k}$ and λ. There are nearly infinitely many CL molecules, denoted by $m = 1, 2, \cdots, M$, distributed on large ∂V. m-th oscillator has angular frequency ω_m, mass m_m, charge q_m, electric dipole moment $\hat{\mathbf{p}}_m = \mathbf{e}_m q_m \hat{x}_m$ with fixed direction in unit vector $\mathbf{e}_m$ and for instantaneous displacement $\hat{x}_m$. In terms of ladder operators $\{\hat{c}_m, \hat{c}^\dagger_m\}$, we have $\hat{H}_{\mathrm{SU}} = \sum_m \hbar\omega_m(\hat{c}^\dagger_m\hat{c}_m + 1/2)$ and $\hat{\mathbf{p}}_m = \mathbf{e}_m\kappa_m(\hat{c}_m + \hat{c}^\dagger_m)$ with $\kappa_m = \sqrt{\hbar q_m^2/(2m_m\omega_m)}$.

We write the second-quantized electric field at m-th molecule's position $\mathbf{r}_m$

$$\hat{\mathbf{E}}_m = \sum_{\mathbf{k}\lambda} \mathbf{E}_{\mathbf{k}\lambda,m}\hat{b}_{\mathbf{k}\lambda} + \text{h.c.} \tag{2}$$

where $\mathbf{E}_{\mathbf{k}\lambda,m} = i\sqrt{2\pi\hbar\omega_k/V}\mathbf{e}_{\mathbf{k}\lambda}e^{i\mathbf{k}\cdot\mathbf{r}_m}$ and h.c. denotes the hermitian conjugate of the first term. $\mathbf{e}_{\mathbf{k}\lambda}$ is a unit vector of polarization. Then, we have interaction Hamiltonian by dipole interaction between photons and CL molecules

$$\hat{H}_{\text{I-BSU}} = -\sum_{\omega}\sum_{m,a} \hat{E}^a_m(\omega)\hat{p}^a_m \tag{3}$$

where $a = 1,2,3$ denotes a component of vector. $\hat{E}^a_m(\omega)$ is an eigen-operator of $\hat{E}^a_m$, defined as $\hat{E}^a_m(\omega) = \sum_{\epsilon'_B - \epsilon_B = \hbar\omega} |\epsilon_B\rangle\langle\epsilon_B|\hat{E}^a_m|\epsilon'_B\rangle\langle\epsilon'_B|$ where $|\epsilon_B\rangle$ is an eigenket of $\hat{H}_{\mathrm{B}}$. We find

$$\hat{E}^a_m(\omega) = {\sum_{\mathbf{k}\lambda}}' \left(\Theta(\omega)E^a_{\mathbf{k}\lambda,m}\hat{b}_{\mathbf{k}\lambda} + \Theta(-\omega)E^{a*}_{\mathbf{k}\lambda,m}\hat{b}^\dagger_{\mathbf{k}\lambda}\right) \tag{4}$$

where $\sum'_{\mathbf{k}\lambda}$ denotes the summation restricted for $\omega_k = |\omega|$ and $\Theta(\omega)$ is the heaviside step function.

Applying the standard weak coupling theory, we find $\mathcal{D}_{\mathrm{B}}^{\mathrm{X}}$ expressed in complicated cross terms for different sets of indices, $\{\omega, \mathbf{k}\lambda, m, a\}$ and $\{\omega', \mathbf{k}'\lambda', l, b\}$. It can be simplified as follows. From independence of CL molecules, we have $m = l$. On the large ∂V, $e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_m}$ is fast oscillating, so we have $\mathbf{k} = \mathbf{k}'$. Requiring a cut-off frequency ω_c for infinitely many frequencies of CL molecules, $\{\omega_m\}$ forms a nearly continuous distribution. In small window $d\omega$, there are still many values of $\omega_m \in [\omega, \omega + d\omega]$ which are collected from the whole surface so that associated dipole moments are randomly directed. So, we replace $e^a_m e^b_m$ by $\delta_{ab}/3$. Choosing orthogonal polarizations of the electric field for given $\mathbf{k}$, we get $\sum_a e^a_{\mathbf{k}\lambda}e^a_{\mathbf{k}\lambda'} = \delta_{\lambda\lambda'}$. As a characteristics of CL model, the spectral density is defined as $J(\omega) = M^{-1}\sum_m q_m^2/(m_m\omega_m)\delta(\omega - \omega_m)$. We present the mathematical procedure in detail in Sec. II, Suppl.

We present the dissipators of both the RF and LB type in Sec. II, Suppl. For simplicity, we show the LB type dissipator

$$\mathcal{D}^{\mathrm{LB}}_{\mathrm{B}}[\rho_{\mathrm{SB}}] = \sum_{\mathbf{k}\lambda, \omega_k < \omega_c} \mu(\omega_k)\left[(N(\omega_k)+1)\left(\hat{b}_{\mathbf{k}\lambda}\rho_{\mathrm{SB}}\hat{b}^\dagger_{\mathbf{k}\lambda} - \frac{1}{2}\left\{\hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda}, \rho_{\mathrm{SB}}\right\}\right) + N(\omega_k)\left(\hat{b}^\dagger_{\mathbf{k}\lambda}\rho_{\mathrm{SB}}\hat{b}_{\mathbf{k}\lambda} - \frac{1}{2}\left\{\hat{b}_{\mathbf{k}\lambda}\hat{b}^\dagger_{\mathbf{k}\lambda}, \rho_{\mathrm{SB}}\right\}\right)\right], \tag{5}$$

where $\mu(\omega_k) = \mu_0\omega_k J(\omega_k)$ for $\mu_0 = (\pi^2/3)M/V$. Writing $M = V^{2/3}/a_0^2$ for mean intramolecular distance a_0 of CL molecules, $\mu_0 = \pi^2/(3a_0^2)V^{-1/3}$. $\mu(\omega_k) \sim V^{-1/3}$ is an important characteristics originating from the boundary interaction between B and SU.

We consider a single harmonic oscillator S of angular frequency ω_0, mass m, charge q, located at $\mathbf{r} = \mathbf{0}$ inside V and oscillates in x direction. In terms of $\hat{a}$ and $\hat{a}^\dagger$, lowering and raising operators, we have $\hat{H}_{\mathrm{S}} = \hbar\omega_0\left(\hat{a}\hat{a}^\dagger + 1/2\right)$ and dipole moment $\hat{p} = \sqrt{\hbar q^2/(2m\omega_0)}(\hat{a} + \hat{a}^\dagger)$. Then, the diople interaction between S and B gives

$$\hat{H}_{\mathrm{I}} = -\hat{p}\hat{E}_x(\mathbf{0}) = -i\hbar\sum_{\mathbf{k}\lambda}\gamma_{\mathbf{k}\lambda}(\hat{a} + \hat{a}^\dagger)(\hat{b}_{\mathbf{k}\lambda} - \hat{b}^\dagger_{\mathbf{k}\lambda}) \tag{6}$$

where $\gamma_{\mathbf{k}\lambda} = \sqrt{\pi q^2/(m\omega_0 V)}\omega_k^{1/2}e^x_{\mathbf{k}\lambda}$ for $\omega_k = c|\mathbf{k}|$.

We regard the composite SB as a collection of harmonic oscillators. Let $\hat{b}_i$ $(\hat{b}^\dagger_i)$ for $i = 0, 1, 2, \cdots$ be lowering (raising) operators where $\hat{b}_0 = \hat{a}$ and $\hat{b}_i$ for $i \neq 0$ denotes $\hat{b}_{\mathbf{k}\lambda}$. We introduce the coherent state $|z_i\rangle$, the eigenstate of $\hat{b}_i$, given by $e^{-|z_i|^2}/2\sum_n z^n/\sqrt{n!}$. Let $|u\rangle = |z, z_1, \cdots, z_i, \cdots\rangle$ be the composite coherent state where z represents eigenvalue of $\hat{b}_0$. Using the P-representation [40–42] in high-dimensional complex space, we write $\rho_{\mathrm{SB}} = \int d^2u P(u,u^*)|u\rangle\langle u|$.

Using the property: $\hat{b}_i|z_i\rangle = z_i|z_i\rangle\langle z_i|$, $\hat{b}_i^\dagger|z_i\rangle\langle z_i| = (\partial/\partial z_i + z_i^*)\,|z_i\rangle\langle z_i|$, we can derive the differential equation for $P(u,u^*)$ from Eq. (1) to have the form of the Fokker-Planck (FP) equation. Writing $z_i = x_i + iy_i$ for $i>0$ and $z = x+iy$, we find for the LB type thermostat

$$\begin{aligned}\dot{P} = \Bigg[&\omega_0\left(-\frac{\partial}{\partial x}y + \frac{\partial}{\partial y}x\right) + \sum_{i>0}\omega_i\left(-\frac{\partial}{\partial x_i}y_i + \frac{\partial}{\partial y_i}x_i\right)\\ &-\sum_{i>0}\gamma_i\left[\frac{\partial}{\partial x_i}x - \frac{\partial}{\partial y}y_i - \frac{1}{4}\left(\frac{\partial^2}{\partial x_i\partial x} - \frac{\partial^2}{\partial y_i \partial y}\right)\right] \\ &+\sum_{i>0}\frac{\mu_i}{2}\left[\left(\frac{\partial}{\partial x_i}x_i + \frac{\partial}{\partial y_i}y_i\right) + \frac{N_i}{2}\left(\frac{\partial^2}{\partial x_i^2} + \frac{\partial^2}{\partial y_i^2}\right)\right]\Bigg]P\ .\end{aligned} \tag{7}$$

For $i>0$, ω_i, γ_i, and μ_i stand for ω_k, $\gamma_{\mathbf{k}\lambda}$, and $\mu(\omega_k)$, respectively, and $N_i = (e^{\beta\hbar\omega_i}-1)^{-1}$. $\sum_{i>0}$ stands for $\sum_{\mathbf{k}\lambda}$, which leads to $V/(2\pi)^3\int d^3\mathbf{k}\sum_\lambda$ in continuum limit of $\mathbf{k}$ for large V. The expression for the RF type thermostat is given in Sec. III, Suppl.

Writing $\mathbf{q} = (x,y,x_1,y_1,x_2,y_2,\cdots)^{\mathrm{t}}$ with subscript t denoting transpose, we get a compact form of the differential equation

$$\dot{P}(\mathbf{q},t) = \partial_{\mathbf{q}}\cdot(\mathsf{F}\cdot\mathbf{q} + \mathsf{D}\cdot\partial_{\mathbf{q}})P(\mathbf{q},t) \tag{8}$$

where the drift matrix F and D are written as

$$\mathsf{F} = \begin{pmatrix}\mathsf{F}_{\mathrm{S}} & \mathsf{F}_{\mathrm{off}}\\ \widetilde{\mathsf{F}}_{\mathrm{off}} & \mathsf{F}_{\mathrm{B}}\end{pmatrix},\ \mathsf{D} = \begin{pmatrix}0 & \mathsf{D}_{\mathrm{off}}\\ \mathsf{D}_{\mathrm{off}}^{\mathrm{t}} & \mathsf{D}_{\mathrm{B}}\end{pmatrix}, \tag{9}$$

which is given in detail in Sec. III, Suppl. It is an Ornstein-Uhlenbeck (OU) process in infinite dimensions. However, D is not positive definite. Moreover, P can be negative though it is normalizable, so is called quasi-probability distribution [40, 41].

There are two small parameters, $\mu(\omega_k) \sim V^{-1/3}$ in Eq. (5) and $\gamma_{\mathbf{k}\lambda} \sim V^{-1/2}$ in Eq. (6). Both are of dimension of ω_0, so we define dimensionless parameters, $\bar{\mu}(\omega_k) = \omega_0^{-1}\mu(\omega_k)$ and $\bar{\gamma}_{\mathbf{k}\lambda} = \omega_0^{-1}\gamma_{\mathbf{k}\lambda}$. Intuitively, we expect that the former is much larger than the latter. We need a finer dimensional analysis for the two. We specifically consider the Ohmic spectral density, $\underline{J(\omega) = c\omega}$, and c is given from $\int_0^{\omega_c} d\omega J(\omega) = c\,\omega_c^2/2 = \overline{q_n^2/(m_n\omega_n)}$. Then, we have $\bar{\mu}(\omega_k) = \frac{2\pi(\omega_k/\omega_0)^2}{3\omega_c/\omega_0}\overline{\left(\frac{q_n^2}{m_n\omega_n}\right)}\frac{1}{\omega_c a_0^2 V^{1/3}}$ and $\bar{\gamma}_{\mathbf{k}\lambda} = e_{\mathbf{k}\lambda}^x\left[\pi(\nu/V)(\omega_k/\omega_0)\right]^{1/2}$ for $\nu = q^2/(m\omega_0^2)$. There are some typical experimental values: electron's charge $= 4.8\times10^{-10}$esu, reduced mass of $H_2 = 1.7\times10^{-24}$g, typical angular frequency of diatomic molecules $\sim 10^{13}$-$10^{14}\mathrm{s}^{-1}$, $V\sim 10^3$-$10^6\mathrm{cm}^3$, and mean intramolecular distance $a_0 \sim 10^{-8}$cm for $M \sim V^{2/3}/a_0^2$. Regarding all angular frequencies, ω_0, ω, ω_n as having the same order of magnitude, we find $\bar{\gamma}_{\mathbf{k}\lambda},\ \bar{\mu}(\omega_k) \ll 1$, $\bar{\gamma}_{\mathbf{k}\lambda}/\bar{\mu}(\omega) \sim \left[m\omega_0^2a_0^4/(q^2V^{1/3})\right]^{1/2} \ll 1$. Then, we have an important inequality

$$\bar{\gamma}_{\mathbf{k}\lambda} \ll \bar{\mu}(\omega) \ll 1\ . \tag{10}$$

According to this inequality, we can perform perturbation expansions in $\bar{\gamma}_{\mathbf{k}\lambda}$ while $\bar{\mu}(\omega_k)$ is treated rigorously until the end of expansions.

From the property of the OU process, dissipation (relaxation) to steady state is described by $e^{-\mathsf{F}t}$ accompanied by $e^{-\mathsf{F}^{\mathrm{t}}t}$. We can find eigenvalues of F by series expansions in $\bar{\gamma}_{\mathbf{k}\lambda}$ and twice the real parts of eigenvalues give relaxation rates (inverse relaxation times); τ_{S}^{-1} for S and $\tau_B(\omega_k)^{-1}$ for photon modes of B. Up to the leading order, $\mathcal{O}(\bar{\gamma}_{\mathbf{k}\lambda}^2)$, we find that $\tau_{\mathrm{S}}^{-1} \simeq \omega_0\nu(\omega_0/c)^3/6$ and $\tau_{\mathrm{B}}(\omega_k)^{-1} \simeq \omega_0\bar{\mu}(\omega_k)$ depending on frequencies of photon modes; see Sec. III, Suppl. for derivation. The ratio of the two rates is given as $\tau_{\mathrm{S}}^{-1}/\tau_{\mathrm{B}}^{-1} \sim \omega_0^3a_0^2V^{1/3}/c^3 \ll 1$. In terms of relaxation times, we have another important inequality

$$\tau_{\mathrm{S}} \gg \tau_{\mathrm{B}}(\omega_k) \gg \omega_0^{-1}\ . \tag{11}$$

It implies that B relaxes much faster than S though both relaxations are still very slow compared to the system's internal time scale $\sim \omega_0^{-1}$. Note that this separation of *fast and slow* time scales are given intrinsically from the boundary interaction between B and SU. It serves an essential condition in marginalization to average out fast degrees of freedom of B in P-distribution.

The conditional P-distribution is defined as the distribution of $\mathbf{q}$ at time t for an initial delta distribution with peak at $\mathbf{q}_0$, formally written as $P(\mathbf{q},t|\mathbf{q}_0,0) \propto e^{-(1/2)(\mathbf{q}-e^{-\mathsf{F}t}\cdot\mathbf{q}_0)^{\mathrm{t}}\cdot\mathbf{A}(t)\cdot(\mathbf{q}-e^{-\mathsf{F}t}\cdot\mathbf{q}_0)}$. Writing $\mathbf{q} = (\mathbf{r},\mathbf{R})^{\mathrm{t}}$ where $\mathbf{r}^{\mathrm{t}} = (x,y)$ for S and $\mathbf{R}^{\mathrm{t}} = (x_1,y_1,x_2,y_2,\cdots)$ for B, we conjecture

$$P(\mathbf{r},\mathbf{R},t|\mathbf{r}_0,0) \propto e^{-(1/2)\widetilde{\mathbf{R}}^{\mathrm{t}}\cdot(\mathsf{A}_{\mathrm{B}}+\Gamma)\cdot\widetilde{\mathbf{R}}}P_{\mathrm{S}}(\mathbf{r},t|\mathbf{r}_0,0) \tag{12}$$

where $\widetilde{\mathbf{R}} = \mathbf{R}-\mathbf{u}$ and $\mathbf{u} = \mathbf{u}(\mathbf{r},\mathbf{r_0})$ is the local equilibrium state of B. Note that $\ln P(\mathbf{r},\mathbf{R},t|\mathbf{r}_0,0)$ should be quadratic in $\mathbf{r}$ and $\mathbf{r_0}$, as characteristic of the OU process. This conjecture reflects that initial memory of $\mathbf{R}_0$ is decayed fast as e^{-t/τ_B} in relaxation time scale of S. $\mathbf{u}$ is responsible for the correlation between S and B. This conjecture can be confirmed by finding $\mathbf{u}$ and Γ self-consistently. The classical probability density in momentum space in the overdamped limit was conjectured in a similar manner as $\propto e^{-\beta/(2m)(\mathbf{p}-m\mathbf{v})^2}$. It reflects that momentum variables fast reaches a local equilibrium $m\mathbf{v}$. $\mathbf{v}$ was found self-consistently to be the probability current of the derived FP-equation in position space [43].

In the absence of interaction, $\mathbf{u}$ and Γ vanish, so the bath P-distribution $\propto e^{-(1/2)\mathbf{R}^{\mathrm{t}}\cdot\mathsf{A}_{\mathrm{B}}\cdot\mathbf{R}}$ corresponds to equilibrium steady state density matrix $\propto e^{-\beta H_{\mathrm{B}}}$. Integrating Eq. (12) over $\widetilde{\mathbf{R}}$, one get $P_{\mathrm{S}}(\mathbf{r},t|\mathbf{r}_0,0)$, which is the conditional P-distribution for S given initial state $\mathbf{r}_0$.

We plug Eq. (12) into Eq. (8). We perform perturbation expansion, treating $\mathbf{u} \sim \mathcal{O}(\bar{\gamma}_i)$ and $\Gamma \sim \mathcal{O}(\bar{\gamma}_i^2)$, which can indeed be confirmed self-consistently. Integrating the plugged in differential equation over $\widetilde{\mathbf{R}}$, we find the marginal differential equation up to $\mathcal{O}(\bar{\gamma}_i^2)$

$$\dot{P}_{\rm S}(\mathbf{r},t|\mathbf{r}_0) = \partial_{\mathbf{r}} \cdot (\mathsf{F}_{\rm S}\cdot\mathbf{r} + \mathsf{F}_{\rm off}\cdot\mathbf{u}) P_{\rm S}(\mathbf{r},t|\mathbf{r}_0) \ . \quad (13)$$

and $\mathbf{u}$ is shown to satisfy

$$\dot{\mathbf{u}} = -\mathsf{F}_{\rm B}\cdot\mathbf{u} + \mathbf{r}^{\rm t}\cdot\mathsf{F}_{\rm S}^{\rm t}\cdot\partial_{\mathbf{r}}\mathbf{u} - \widetilde{\mathsf{F}}_{\rm off}\cdot\mathbf{r} + \widetilde{\mathsf{D}}\cdot\partial_{\mathbf{r}}\ln P_{\rm S}(\mathbf{r},t|\mathbf{r}_0) \quad (14)$$

where $\widetilde{\mathsf{D}} = \mathsf{A}_{\rm B}^{-1}\mathsf{F}_{\rm off}^{\rm t} - 2\mathsf{D}_{\rm off}^{\rm t}$. The equation for $\mathbf{u}$ is kept up to $\mathcal{O}(\bar{\gamma}_i)$. Γ is not needed up to this order. The mathematical steps to derive the marginal equation are given in detail in Sec. IV, Suppl.

We further conjecture for the conditional P-distribution for S as

$$P_{\rm S}(\mathbf{r},t|\mathbf{r}_0,0) = \frac{[\det\mathsf{A}_{\rm S}(t)]^{1/2}}{2\pi} e^{-(1/2)\widetilde{\mathbf{r}}^{\rm t}\cdot\mathsf{A}_{\rm S}(t)\cdot\widetilde{\mathbf{r}}} \quad (15)$$

where $\widetilde{\mathbf{r}} = \mathbf{r} - e^{-\mathsf{F}_{\rm eff}t}\cdot\mathbf{r}_0$. It is a formal expression for the OU process [44, 45]. Using the property that $\mathbf{u}$ is linear in $\mathbf{r}$ and $\mathbf{r}_0$, we write $\mathbf{u} = \mathsf{A}_{\rm off}\cdot\mathbf{r} + \mathsf{B}_{\rm off}e^{-\mathsf{F}_{\rm eff}t}\cdot\mathbf{r}_0$. Through some mathematical steps, we can find $\mathsf{A}_{\rm off}\cdot\mathbf{r}$, $\mathsf{B}_{\rm off}$ from which $\mathsf{F}_{\rm eff}$ and $\mathsf{A}_{\rm S}$ can be determined. We finally derive the marginal differential equation as

$$\dot{P}_S = \partial_{\mathbf{r}} \cdot (\mathsf{F}_{\rm eff}\cdot\mathbf{r} + \mathsf{D}_{\rm eff}\cdot\partial_{\mathbf{r}}]\, P_{\rm S} \ . \quad (16)$$

where $\mathsf{F}_{\rm eff}$ and $\mathsf{D}_{\rm eff}$ are 2×2 matrices. The derivation is given in detail in Sec. V, Suppl.

We show

$$\omega_0^{-1}\mathsf{F}_{\rm eff} = \begin{pmatrix} 0 & -1 \\ 1+\bar{\nu}f_2 & \bar{\nu}f_1 \end{pmatrix} \ , \ \ \omega_0^{-1}\mathsf{D}_{\rm eff} = \begin{pmatrix} 0 & \bar{\nu}d_2 \\ \bar{\nu}d_2 & \bar{\nu}d_1 \end{pmatrix} \ . \quad (17)$$

where $\bar{\nu} = \nu(\omega_0/c)^3$ for $\nu = q^2/(m\omega_0^2)$ is small and dimensionless, coming from the perturbation parameter $\bar{\gamma}_{\mathbf{k}\lambda}$. The kernel $\mathsf{A}_{\rm S}$ for the solution can be found from

$$\dot{\mathsf{A}}_{\rm S}^{-1} = -\mathsf{F}_{\rm eff}\mathsf{A}_{\rm S}^{-1} - \mathsf{A}_{\rm S}^{-1}\mathsf{F}_{\rm eff}^{\rm t} + 2\mathsf{D}_{\rm eff}. \quad (18)$$

.

Matrix elements, $f_{1,2}$ and $d_{1,2}$, result from sums over all photon modes, hence the integrals over $\mathbf{k}$ in the continuum limit for large V. The integral forms can be found in Sec. VI, Suppl. Finally, changing variable $\xi = \omega_k/\omega_0$ and taking the limit $\bar{\mu} \to 0$, we find $\omega_c > \omega_0$

$$f_1 = \frac{1}{6}R_{\rm B} \ , \quad (19)$$

$$f_2 = -\frac{1}{3\pi}\,{\rm P}\int_0^{\xi_c} d\xi\, \frac{\xi^4}{\xi^2-1} \ , \quad (20)$$

$$d_1 = \frac{N_0}{12}R_{\rm B} \ , \quad (21)$$

$$d_2 = \frac{1}{24\pi}\,{\rm P}\int_0^{\xi_c} d\xi\, \xi^3\left[\frac{1}{\xi+1} - \frac{2N(\xi)}{\xi^2-1}\right] \ , \quad (22)$$

where N_0 is the average number of quanta for S and P denotes the principal value of an integral. We have

$$R_{\rm B} = \begin{cases} 1 \ ; & \text{LB-type thermostat} \\ \frac{J_0}{\sqrt{P_0^2+J_0^2}} ; & \text{LF-type thermostat} \end{cases} \quad (23)$$

where $J_0 = J(\omega_0)$ and $P_0 = \frac{1}{\pi}{\rm P}\int_0^{\omega_c}{\rm d}\Omega J(\Omega)\frac{2\Omega}{\omega_0^2-\Omega^2}$. Note that Ω is the variable continued from $\{\omega_m\}$ for the CL oscillators. See the detail derivation in Sec. VI, Suppl.

It is remarkable that the conventional RF equation for reduced density matrix for isolated SB leads to the same form in P-representation as in Eq. (16). However, matrix elements, $f_{1,2}^{\rm RF}$ and $d_{1,2}^{\rm RF}$, are similar but different from those in Eqs. (19)-(22) obtained for the limit $\bar{\mu} \to 0$. In fact, $f_{1,2}^{\rm RF}$ and $d_{1,2}^{\rm RF}$ are four times larger than $f_{1,2}$ and $d_{1,2}$ for $R_{\rm B} = 1$. See the comparison in detail in Sec. VII, Suppl.

The steady state kernel can be found from $\mathsf{F}_{\rm eff}\mathsf{A}_{\rm S}^{-1} + \mathsf{AS}^{-1}\mathsf{F}_{\rm eff}^{\rm t} - 2\mathsf{D}_{\rm eff} = 0$, found from Sec. V, Suppl. We get

$$\mathsf{A}_{\rm S}(\infty) = \frac{2}{N_0}\begin{pmatrix} 1+C\bar{\nu} & 0 \\ 0 & 1 \end{pmatrix} \ \ \text{for } \omega_c > \omega_0. \quad (24)$$

where $C = f_2 - (4/N_0)d_2$. The factor $2/N_0$ comes from f_1/d_1 in Eqs. (19) and (21) found for $\omega_c > \omega_0$. In the leading order, it gives the steady state density matrix $\propto e^{-\beta\hat{H}_{\rm S}}$. The condition $\omega_c > \omega_0$ implies that the frequency range of photons should include the system frequency ω_0 for S to be thermalized close to equilibrium. C specifies the correction of $\mathcal{O}(\bar{\nu})$, originating from the second order contribution of $\hat{H}_{\rm I}$. It is noteworthy that the steady state kernel be independent of the type of thermostat, shown up to $\mathcal{O}(\bar{\nu})$. The steady state density matrix $\hat{\rho}_{\rm S}^{\rm ss}$ for S can be found inversely from the steady state P distribution $\propto e^{-1/2\mathbf{r}^{\rm t}\cdot\mathsf{A}_{\rm S}(\infty)\cdot\mathbf{r}}$. We expand $\hat{\rho}_{\rm S}^{\rm SS} = \hat{\rho}^{(0)} + \hat{\rho}^{(1)}$, where the first and second terms are of the zeroth and first order in $\bar{\nu}$. We find $\hat{\rho}^{(0)} = e^{-\beta\hbar\omega_0(\hat{a}^\dagger\hat{a}+1/2)}/Z_0$ for the partition function Z_0, as expected. We also find

$$\begin{aligned} \hat{\rho}^{(1)} &= -\hat{\rho}^{(0)}\frac{C\bar{\nu}}{2N_0}e^{-2\beta\hbar\omega_0}\left(\hat{a}\hat{a} + e^{2\beta\hbar\omega_0}\hat{a}^\dagger\hat{a}^\dagger\right. \\ &\quad \left. + 2e^{\beta\hbar\omega_0}\hat{a}^\dagger\hat{a} - 2N_0e^{2\beta\hbar\omega_0}\right) \ . \end{aligned} \quad (25)$$

The derivation is given in Sec. VIII, Suppl. It can be shown not to be equal to the mean force Gibbs (MFG) density matrix $\propto {\rm Tr}_{\rm B}e^{-\beta(\hat{H}_{\rm S}+\hat{H}_{\rm B}+\hat{H}_{\rm I}}$, which is supposed to the steady state of an isolated SB, hence that of the conventional RF equation [22, 28].

The time-dependent state is found to depend on thermostat, as in realistic situations. In particular, the relaxation rate τ_S^{-1} estimated from $\mathsf{F}_{\rm eff}$ is equal to $\omega_0\bar{\nu}f_1$; see Eq. (19). We consider S to be initially in a pure state with $\rho_S(0) = \sum_{n,m}\rho_{nm}|n\rangle\langle m|$. It corresponds to $P_{\rm S}(z_0,0) = \sum_{n,m}\rho_{nm}\frac{e^{|z_0|^2}}{\sqrt{n!m!}}\frac{\partial^{n+m}}{\partial(-z_0)^n\partial(-\bar{z}_0)^m}\delta^{(2)}(z_0)$, which is not positive and highly singular, as well known. Then, we can get the marginal P distribution at time t by using Eq. (15.

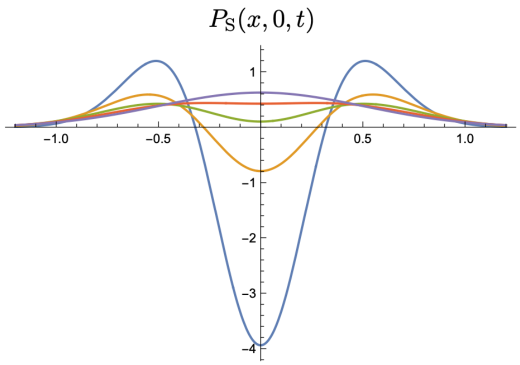

FIG. 1. $P_{\rm S}(x, y = 0, t)$ versus x is plotted for $\bar{\nu} = 0.1$, $\omega_c = 2\omega_0$, and $\beta\hbar\omega_0 = 1$. $\tau_{\rm S} \simeq 592\omega_0^{-1}$. The various curves are presented for $\omega_0 t = 200,\ 300,\ 400,\ 500,\ 1000$, represented by blue, yellow, green, red, black curves, respectively. As time increases, negative distribution decreases to reach a Gaussian distribution.

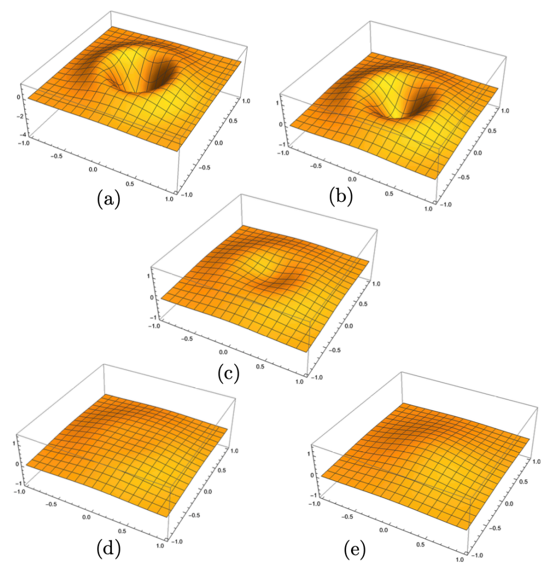

FIG. 2. $P_{\rm S}(x, y, t)$ is plotted for the same parameters as in Fig. 1. The panel (a) for $\omega_0 t = 200$, (b) for $\omega_0 t = 300$ shows negative distributions inside a valley around the origin. The panel (c) for $\omega_0 t = 400$ and (d) for $\omega_0 t = 500$ shows positive distribution everywhere with shallow valleys. The panel (e) $\omega_0 t = 1000$, corresponding to about $2 \times \tau_{\rm S}$, shows distribution nearly Gaussian with no valley.

As a simple case, we can find the time-dependent P-distribution for $\rho_{\rm S}(0) = |1\rangle\langle 1|$, given in Sec. IX, Suppl. We demonstrate it in Figs. 1 and 2.

In conclusion, we derive the marginal differential equation for P-distribution for S coupled with B equipped with thermostat. The corresponding equation for reduced density matrix is found to have the same form with different coefficients as the conventional RF equation for isolated SB. Thus, we conclude that the steady state of S in our study is not the MFG state, expected for isolated SB. We find the steady state does not depend on thermostat, while dynamical process does, which is reasonable in comparison with classical cases where steady state is independent of friction and diffusion coefficients. It is interesting whether or not the MFG state might be recovered by the global coupling scheme to replace $\mathcal{D}_B^{\rm X}$. We will generalize our theory to non-harmonic systems such as a two-level system. Since our theory does not require eigen-operators based on eigenstates of the static system, we expect to extend our theory to systems with time-dependent protocols and to investigate non-equilibrium quantum stochastic thermodynamics.

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