Excess entropy production in quantum system: Quantum master equation approach

Satoshi Nakajima* and Yasuhiro Tokura†
Graduate School of Pure and Applied Sciences, University of Tsukuba, 1 − 1 − 1 , Tennodai, Tsukuba, 305 − 8571, Japan

October 25, 2017

Abstract

For open systems described by the quantum master equation (QME), we investigate the excess entropy production under quasistatic operations between nonequilibrium steady states. The average entropy production is composed of the time integral of the instantaneous steady entropy production rate and the excess entropy production. We propose to define average entropy production rate using the average energy and particle currents, which are calculated by using the full counting statistics with QME. The excess entropy production is given by a line integral in the control parameter space and its integrand is called the Berry-Sinitsyn-Nemenman (BSN) vector. In the weakly nonequilibrium regime, we show that BSN vector is described by $\ln \breve{\rho}_0$ and ρ_0 where ρ_0 is the instantaneous steady state of the QME and $\breve{\rho}_0$ is that of the QME which is given by reversing the sign of the Lamb shift term. If the system Hamiltonian is non-degenerate or the Lamb shift term is negligible, the excess entropy production approximately reduces to the difference between the von Neumann entropies of the system. Additionally, we point out that the expression of the entropy production obtained in the classical Markov jump process is different from our result and show that these are approximately equivalent only in the weakly nonequilibrium regime.

1 Introduction

In equilibrium thermodynamics, the central quantity is the entropy S, which describes both the macroscopic properties of equilibrium systems and the fundamental limits on the possible transitions among equilibrium states. In equilibrium thermodynamics, the Clausius equality

$$\Delta S = \beta Q, \tag{1.1}$$

tells us how one can determine the entropy by measuring the heat. Here, ΔS is the change in the entropy of the system during the operation, β is the inverse temperature of the bath contacting with the system, and Q is the heat transferred from the bath to the system during the operation. This equality is universally valid for quasistatic transitions between equilibrium states. In the equilibrium classical (quantum) system, the entropy is given by the Shannon entropy of the probability distribution (von Neumann entropy of the density matrix) of states.

The investigation of thermodynamic structures of nonequilibrium steady states (NESSs) has been a topic of active research in nonequilibrium statistical mechanics [1, 2, 3, 4, 5, 6, 7, 8, 9]. For instance,

*Email: subarusatosi@gmail.com
†Email: tokura.yasuhiro.ft@u.tsukuba.ac.jp

the extension of the relations in equilibrium thermodynamics, such as the Clausius equality, to NESSs has been one of the central subjects. Recently there has been progress in the extension of the Clausius equality to NESSs [10, 11, 12] (see also Refs.[13, 14, 15, 16, 17, 18]). In these studies, the excess heat $Q_{b,\mathrm{ex}}$ (of the bath b) [2] has been introduced instead of the total heat Q_b from the bath b. The excess heat $Q_{b,\mathrm{ex}}$, which describes an additional heat induced by a transition between NESSs with time-dependent external control parameters, is defined by subtracting from Q_b the time integral of the instantaneous steady heat current from the bath b. In the weakly nonequilibrium regime, there exists a scalar potential $\mathcal{S}$ of the control parameter space which approximately satisfies the *extended Clausius equality*

$$\sum_b \beta_b Q_{b,\mathrm{ex}} \approx \Delta\mathcal{S}. \tag{1.2}$$

Here, β_b is the inverse temperature of the bath b, $\Delta\mathcal{S} = \mathcal{S}(\alpha_\tau) - \mathcal{S}(\alpha_0)$, $\alpha_t = (\alpha_t^1, \alpha_t^2, \ldots)$ is the value of the set of the control parameters at time t, and $t = 0$ and $t = \tau$ are the initial and final times of the operation. We assume multiple baths to maintain the system out of the equilibrium and the symbol $\sum_b$ means the summation over the baths. In classical systems, $\mathcal{S}$ is the symmetrized Shannon entropy [11]. In quantum systems with the time-reversal symmetry, $\mathcal{S}$ is the symmetrized von Neumann entropy [12]. However, the study of the excess entropy in the quantum system without the time-reversal symmetry seems still lacking. This is the main objective of this paper.

In general, the left-hand side (LHS) of (1.2) is replaced by the excess entropy

$$\sigma_{\mathrm{ex}} \stackrel{\mathrm{def}}{=} \sigma - \int_0^\tau dt\ J_{\mathrm{ss}}^\sigma(\alpha_t), \tag{1.3}$$

where σ is the average entropy production and $J_{\mathrm{ss}}^\sigma(\alpha_t)$ is the instantaneous steady entropy production rate at time t [19, 20, 21]. In the quasistatic operation, the excess entropy is given by

$$\sigma_{\mathrm{ex}} = \Delta\mathcal{S} + \mathcal{O}(\varepsilon^2\delta), \tag{1.4}$$

where ε is a measure of degree of nonequilibrium and δ describes the amplitude of the change of the control parameters. Sagawa and Hayakawa [19] studied the full counting statistics (FCS) of the entropy production for classical systems described by the Markov jump process and showed that the excess entropy is characterized by the Berry-Sinitsyn-Nemenman (BSN) phase [22].

The method of Ref. [19] was generalized to quantum systems and applied to studies of the quantum pump [23, 24, 25]. Here we briefly explain the studies of the quantum pump. At $t = 0$ and $t = \tau$, we perform projection measurements of a *time-independent* observable O of the baths and obtain the outcomes $o(0)$ and $o(\tau)$. The generating function of $\Delta o = o(\tau) - o(0)$ is

$$Z_\tau(\chi) = \int d\Delta o\ P_\tau(\Delta o) e^{i\chi\Delta o} \tag{1.5}$$

where $P_\tau(\Delta o)$ is the probability density distribution of Δo and χ is called the counting field. To calculate the generating function, the method using the quantum master equation (QME) with the counting field (FCS-QME) [26] had been proposed. The solution of the FCS-QME, $\rho^\chi(t)$, provides the generating function as $Z_\tau(\chi) = \mathrm{Tr}_S[\rho^\chi(\tau)]$, where Tr_S denotes the trace of the system. The Berry phase [27] of the FCS-QME is the BSN phase. The average of the difference of the outcomes is given by

$$\langle\Delta o\rangle = \int_0^\tau dt\ i^O(t), \tag{1.6}$$

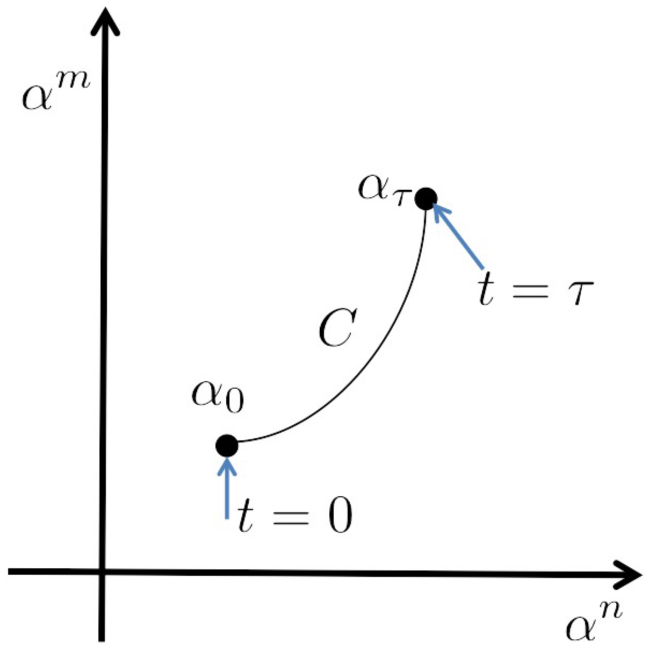

Figure 1: Schematic representation of the trajectory C in the phase space of the control parameters, α^n and α^m.

where $i^O(t)$ is the *current* of an operator O. If the state of the system at $t = 0$ is the instantaneous steady state and the modulation of the control parameters is slow, the following relation holds:

$$\langle \Delta o \rangle = \int_0^\tau dt\ i_{\rm ss}^O(\alpha_t) + \int_C d\alpha^n\ A_n^O(\alpha), \tag{1.7}$$

where $i_{\rm ss}^O(\alpha_t)$ is the instantaneous steady current of O. C is the trajectory from α_0 to α_τ as shown in Fig.1 and $A_n^O(\alpha)$ is the BSN vector derived from the BSN phase. α^n is n-th component of the control parameters and the summation symbol for n is omitted. The derived formula of the BSN vector depends on the approximations used for the QME. The Born-Markov approximation with or without the rotating wave approximation (RWA) [28] is frequently used. The QME in the Born-Markov approximation without RWA sometimes violates the non-negativity of the system reduced density operator. The QME of the RWA or the coarse-graining approximation (CGA) [29, 30] is the Lindblad type which guarantees the non-negativity [28]. If O is the total particle number of a bath b, there are several methods to calculate $A_n^O(\alpha)$ of (1.7) [1)].

In this paper, we propose to identify

$$\dot{\sigma}(t) \stackrel{\rm def}{=} \sum_b \beta_b(t) \left[-i^{H_b}(t) - \mu_b(t) \left\{ -i^{N_b}(t) \right\} \right] \tag{1.8}$$

[1)] For non-interacting system, $A_n^O(\alpha)$ is calculated from the Brouwer formula[31] using the scattering matrix. Recently, the quantum pump in interacting systems has been actively researched. There are three theoretical approaches. The first is the Green's function approach, [32] The second is the generalized master equation approach [34, 35]. The third is the FCS-QME approach. Reference[25] showed the equivalence between the second and the third approaches for all orders of pumping frequency (see also [33]).

with the *average entropy production rate*, where μ_b is the chemical potential of the bath b, and $i^{H_b}(t)$ and $i^{N_b}(t)$ are energy and particle currents from the system to the bath b, respectively. H_b and N_b represent the Hamiltonian and the total particle number of the bath b, respectively. This is a straightforward extention of the entropy production rate argued for an NESS [36] to a time-dependent system. Now, the excess entropy is obtained by

$$\sigma_{\rm ex} = \int_0^\tau dt\ \left[\dot{\sigma}(t) - J^\sigma_{\rm ss}(\alpha_t)\right] = \int_C d\alpha^n\ A^\sigma_n(\alpha), \tag{1.9}$$

where we used (1.7) in the second equation with

$$A^\sigma_n(\alpha) \ \stackrel{\rm def}{=} \ \sum_b \beta_b \left[-A^{H_b}_n(\alpha) - \mu_b\left\{-A^{N_b}_n(\alpha)\right\}\right]. \tag{1.10}$$

Here, $A^{H_b}_n(\alpha)$ and $A^{N_b}_n(\alpha)$ are the BSN vectors of H_b and N_b. It should be noted that β_b and μ_b could also be the elements of the set of the control parameters, α. The following expression is the main result of this manuscript,

$$A^\sigma_n(\alpha) = -{\rm Tr}_S\left[\ln\breve{\rho}_0(\alpha)\frac{\partial\rho_0(\alpha)}{\partial\alpha^n}\right] + \mathcal{O}(\varepsilon^2), \tag{1.11}$$

without any assumption on the time-reversal symmetry. $\rho_0(\alpha)$ is the instantaneous steady state of the QME and $\breve{\rho}_0(\alpha)$ is that of the QME which is given by reversing the sign of the Lamb shift term. If the system Hamiltonian is non-degenerate or the Lamb shift term is negligible, we obtain

$$\sigma_{\rm ex} = S_{\rm vN}\left(\rho_0(\alpha_\tau)\right) - S_{\rm vN}\left(\rho_0(\alpha_0)\right) + \mathcal{O}(\varepsilon^2\delta), \tag{1.12}$$

where $S_{\rm vN}(\rho) \stackrel{\rm def}{=} -{\rm Tr}_S[\rho\ln\rho]$ is the von Neumann entropy.

The structure of the paper is as follows. First, we explain the FCS-QME (§ 2.1) and the formula for the excess entropy. Then we introduce the generalized QME in § 2.2. In § 2.3, we explain the RWA and after this section we focus on the RWA except for § 3.3. In § 3.1, the BSN vector A^σ_n in the equilibrium is discussed. In § 3.2, the main result of this manuscript, (1.11), is derived. Next we mention the results in the Born-Markov approximation (§3.3). In §4, we compare the preceding study on the entropy production in the classical Markov jump process [21, 37] with ours. In § 5, we consider the time-reversal operation. At last (§ 6), we summarize this paper. In the Appendix A, we derive the formula of the derivative of the von Neumann entropy and in the Appendix B, we show the details of the derivation of the relation in a weakly nonequilibrium regime. In the Appendix C, we explain the definition of entropy production of the Markov jump process and the result of Ref. [21].

2 Quantum master equation and full counting statistics

2.1 Full counting statistics-quantum master equation

We consider system S weakly coupled to several baths (although we used the same symbol S as the entropy in § 1, S only means the 'system' in the following discussions). In order to maintain the system out of equilibrium and in NESS, the system needs to be coupled with more than one bath. The total Hamiltonian is given by

$$H_{\rm tot}(t) = H_S(\alpha_S(t)) + \sum_b \left[H_b + H_{Sb}(\alpha_{Sb}(t))\right]. \tag{2.1}$$

$H_S(\alpha_S)$ is the system Hamiltonian and α_S denotes a set of control parameters of the system. H_b is the Hamiltonian of the bath b. $H_{Sb}(\alpha_{Sb})$ is the coupling Hamiltonian between S and the bath b, and α_{Sb} is corresponding set of control parameters. We denote the inverse temperature and the chemical potential of the bath b by β_b and μ_b which can be the control parameters, and α_b denotes the set of β_b and $\beta_b\mu_b$. We symbolize the set of all control parameters (α_S, $\{\alpha_{Sb}\}_b$, $\alpha_B \stackrel{\text{def}}{=} \{\alpha_b\}_b$) by α. While α_S and α_{Sb} are dynamical parameters like energy levels, tunnel coupling strengths or a magnetic field, α_B are the thermodynamical parameters. We denote the set of all the linear operators of S by B.

Consider slow modulation of the control parameters during $0 \le t \le \tau$. At $t = 0$ and $t = \tau$, we perform projection measurements of *time-independent* observables $\{O_\mu\}$ of the baths which commute with each other. The index μ distinguishes the time-independent observables of the baths. $\{o_\mu^{(\tau)}\}$ ($\{o_\mu^{(0)}\}$) denotes the set of outcomes at $t = \tau$ ($t = 0$). The generating function

$$Z_\tau(\{\chi_{O_\mu}\}) = \int \prod_\mu d\Delta o_\mu \; P_\tau(\{\Delta o_\mu\}) e^{i\sum_\mu \chi_{O_\mu} \Delta o_\mu} \tag{2.2}$$

is the Fourier transform of the joint probability density distribution $P_\tau(\{\Delta o_\mu\})$ where $\Delta o_\mu \stackrel{\text{def}}{=} o_\mu^{(\tau)} - o_\mu^{(0)}$. Here, χ_{O_μ} is the counting field for O_μ. The generating function is given by

$$Z_\tau(\{\chi_{O_\mu}\}) = \mathrm{Tr}_{\mathrm{tot}}\left[\rho_{\mathrm{tot}}^\chi(t=\tau)\right] \tag{2.3}$$

using an operator of the total system $\rho_{\mathrm{tot}}^\chi(t)$ obeying the modified von Neumann equation [26]

$$\frac{d}{dt}\rho_{\mathrm{tot}}^\chi(t) = -i\left[H_{\mathrm{tot}}(t), \rho_{\mathrm{tot}}^\chi(t)\right]_\chi. \tag{2.4}$$

In this paper, we set $\hbar = 1$. Here, for two operators A and B, $[A,B]_\chi \stackrel{\text{def}}{=} A_\chi B - BA_{-\chi}$ and

$$A_\chi \stackrel{\text{def}}{=} e^{i\sum_\mu \chi_{O_\mu} O_\mu/2} A e^{-i\sum_\mu \chi_{O_\mu} O_\mu/2}. \tag{2.5}$$

χ denotes the set of the counting fields $\{\chi_{O_\mu}\}$. The initial condition of $\rho_{\mathrm{tot}}^\chi(t)$ is given by $\rho_{\mathrm{tot}}^\chi(0) = \sum_{\{o_\nu\}} P_{\{o_\nu\}}\rho_{\mathrm{tot}}(0)P_{\{o_\nu\}}$ [26]. Here, $\rho_{\mathrm{tot}}(0)$ is the initial state of the total system, $\{o_\nu\}$ denotes eigenvalues of $\{O_\nu\}$ and $P_{\{o_\nu\}}$ is a projection operator defined by $O_\mu P_{\{o_\nu\}} = o_\mu P_{\{o_\nu\}}$, $P_{\{o_\nu\}}P_{\{o'_\nu\}} = P_{\{o_\nu\}}\prod_\mu \delta_{o_\mu,o'_\mu}$, and $P_{\{o_\nu\}}^\dagger = P_{\{o_\nu\}}$. We suppose

$$\rho_{\mathrm{tot}}(0) = \rho(0) \otimes \rho_B(\alpha_B(0)), \tag{2.6}$$

where $\rho(0)$ is the initial state of the system and

$$\rho_B(\alpha_B(0)) \stackrel{\text{def}}{=} \bigotimes_b \frac{1}{\Xi_b(\alpha_b(0))} e^{-\beta_b(0)[H_b - \mu_b(0)N_b]} \tag{2.7}$$

with $\Xi_b(\alpha_b) \stackrel{\text{def}}{=} \mathrm{Tr}_b[e^{-\beta_b[H_b-\mu_b N_b]}]$. Tr_b denotes the trace of the bath b, and Tr_B denotes the trace over all baths' degrees of freedom. Then, we have

$$\rho_{\mathrm{tot}}^\chi(0) = \rho(0) \otimes \sum_{\{o_\nu\}} P_{\{o_\nu\}}\rho_B(\alpha_B(0))P_{\{o_\nu\}}. \tag{2.8}$$

We suppose $[H_b, N_b] = 0$. If all O_μ commute with H_b and N_b, $P_{\{o_\nu\}}$ commutes with $\rho_B(\alpha_B(0))$ and $\rho_{\mathrm{tot}}^\chi(0) = \rho(0) \otimes \rho_B(\alpha_B(0))$ holds because $\sum_{\{o_\nu\}} P_{\{o_\nu\}} = 1$.

We defined $\rho^\chi(t) \stackrel{\rm def}{=} {\rm Tr}_B[\rho^\chi_{\rm tot}(t)]$ and the generating function is calculated with

$$Z_\tau(\{\chi_{O_\mu}\}) = {\rm Tr}_S[\rho^\chi(t=\tau)]. \tag{2.9}$$

We assume

$$\rho^\chi_{\rm tot}(t) \approx \rho^\chi(t) \otimes \rho_B(\alpha_B(t)) \quad (0 < t \le \tau) \tag{2.10}$$

where

$$\rho_B(\alpha_B(t)) \stackrel{\rm def}{=} \bigotimes_b \frac{1}{\Xi_b(\alpha_b(t))} e^{-\beta_b(t)[H_b - \mu_b(t) N_b]}. \tag{2.11}$$

The FCS-QME [26] is

$$\frac{d\rho^\chi(t)}{dt} = \hat{K}^\chi(\alpha_t)\rho^\chi(t), \tag{2.12}$$

and the initial condition is $\rho^\chi(0) = \rho(0)$. Here $\hat{K}^\chi(\alpha_t)$ is the Liouvillian modified by χ. The Liouvillian is given by

$$\hat{K}^\chi(\alpha)\bullet = -i[H_S(\alpha_S), \bullet] + \sum_b \mathcal{L}^\chi_b(\alpha)\bullet. \tag{2.13}$$

Here and in the following, $\bullet \in$ B. $\mathcal{L}^\chi_b(\alpha)$ describes the coupling effects between S and the bath b and depends on used approximations, for instance, the Born-Markov approximation without or within the RWA and the CGA. Generally, $\mathcal{L}^\chi_b(\alpha)$ has the form:

$$\mathcal{L}^\chi_b(\alpha)\bullet = \sum_a c^\chi_{ba}(\alpha) A_a \bullet B_a, \tag{2.14}$$

where A_a and B_a belong to B and depend on α_S, and $c^\chi_{ba}(\alpha)$ is a complex number which depends on α_S, α_{Sb} and α_b. If and only if $A_a, B_a \neq 1$, $c^\chi_{ba}(\alpha)$ depends on χ. After § 2.3 we choose the Born-Markov approximation within RWA; however, in this subsection we assume only Markov property (i.e., $\hat{K}^\chi$ just depends on α_t). Explicit expression of (2.14) will be given in § 2.3 . At $\chi = 0$, the FCS-QME becomes the quantum master equation (QME)

$$\frac{d\rho(t)}{dt} = \hat{K}(\alpha_t)\rho(t). \tag{2.15}$$

$\hat{K}(\alpha_t)$ equals $\hat{K}^\chi(\alpha_t)$ at $\chi = 0$. In the following, a symbol X without χ denotes $X^\chi|_{\chi=0}$.

In the Liouville space [25, 26], the left and right eigenvalue equations of the Liouvillian are

$$\begin{aligned} \hat{K}^\chi(\alpha)|\rho^\chi_n(\alpha)\rangle\rangle &= \lambda^\chi_n(\alpha)|\rho^\chi_n(\alpha)\rangle\rangle, && (2.16)\\ \langle\langle l^\chi_n(\alpha)|\hat{K}^\chi(\alpha) &= \lambda^\chi_n(\alpha)\langle\langle l^\chi_n(\alpha)|. && (2.17) \end{aligned}$$

In the Liouville space, $A \in$ B is described by $|A\rangle\rangle$. The inner produce is defined by $\langle\langle A|B\rangle\rangle = {\rm Tr}_S(A^\dagger B)$ ($A, B \in$ B). In particular, $\langle\langle 1|A\rangle\rangle = {\rm Tr}_S A$ holds. A superoperator which operates to a liner operator of the system becomes an operator of the Liouville space. The left eigenvectors $l^\chi_n(\alpha)$ and the right eigenvectors $\rho^\chi_m(\alpha)$ satisfy $\langle\langle l^\chi_n(\alpha)|\rho^\chi_m(\alpha)\rangle\rangle = \delta_{nm}$. The mode which has the eigenvalue $\lambda^\chi_n(\alpha)$ with the maximum real part is assigned by the label $n = 0$. Because the conservation of the probability, $\langle\langle 1|\rho(t)\rangle\rangle = 1$, and using (2.15), the relation

$$\frac{d}{dt}\langle\langle 1|\rho(t)\rangle\rangle = \langle\langle 1|\hat{K}(\alpha_t)|\rho(t)\rangle\rangle = 0 \tag{2.18}$$

leads $\langle\langle 1|\hat{K}(\alpha) = 0$, in the limit $\chi \to 0$, $\lambda_0^\chi(\alpha)$ becomes 0 and $\langle\langle l_0^\chi(\alpha)|$ becomes $\langle\langle 1|$ (i.e., $l_0(\alpha)$ is an identity operator). In addition, the special state $|\rho_0(\alpha)\rangle\rangle$ determined by $\hat{K}(\alpha)|\rho_0(\alpha)\rangle\rangle = 0$ represents the *instantaneous steady state*.

The formal solution of the FCS-QME (2.12) is

$$|\rho^\chi(t)\rangle\rangle = \mathrm{T}\exp\left[\int_0^t ds\ \hat{K}^\chi(\alpha_s)\right]|\rho(0)\rangle\rangle, \tag{2.19}$$

where T denotes the time-ordering operation. Using this, we obtain the averages of Δo_μ at time t

$$\begin{aligned}\langle\Delta o_\mu\rangle_t &= \frac{\partial}{\partial(i\chi_{O_\mu})}\langle\langle 1|\rho^\chi(t)\rangle\rangle\Big|_{\chi=0} \\ &= \int_0^t du\ \langle\langle 1|\hat{K}^{O_\mu}(\alpha_u)|\rho(u)\rangle\rangle \equiv \int_0^t du\ i^{O_\mu}(u),\end{aligned} \tag{2.20}$$

where

$$X^{O_\mu}(\alpha) \stackrel{\mathrm{def}}{=} \frac{\partial X^\chi(\alpha)}{\partial(i\chi_{O_\mu})}\Big|_{\chi=0}, \tag{2.21}$$

when X is an (super)operator or a c-number and $i^{O_\mu}(u)$ is the *current* of O_μ at time u. Here, we used $\langle\langle 1|\hat{K}(\alpha) = 0$. Moreover, using $\langle\langle l_0(\alpha)| = \langle\langle 1|$, $\lambda_0(\alpha) = 0$ and (2.17), we obtain

$$\langle\langle 1|\hat{K}^{O_\mu}(\alpha) = \lambda_0^{O_\mu}(\alpha)\langle\langle 1| - \langle\langle l_0^{O_\mu}(\alpha)|\hat{K}(\alpha). \tag{2.22}$$

Here, $\langle\langle l_0^{O_\mu}(\alpha)|$ is defined by $\frac{\partial\langle\langle l_0^\chi(\alpha)|}{\partial(i\chi_{O_\mu})}\Big|_{\chi=0}$, then $l_0^{O_\mu} = -\frac{\partial l_0^\chi(\alpha)}{\partial(i\chi_{O_\mu})}\Big|_{\chi=0}$ holds. Now, the current $i^{O_\mu}(t)$ is given by [24]

$$\begin{aligned}i^{O_\mu}(t) &= \langle\langle 1|\hat{K}^{O_\mu}(\alpha_t)|\rho(t)\rangle\rangle \\ &= \lambda_0^{O_\mu}(\alpha_t)\langle\langle 1|\rho(t)\rangle\rangle - \langle\langle l_0^{O_\mu}(\alpha_t)|\hat{K}(\alpha_t)|\rho(t)\rangle\rangle \\ &= \lambda_0^{O_\mu}(\alpha_t) - \langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\rho(t)\rangle\rangle.\end{aligned} \tag{2.23}$$

The current can also be written as

$$i^{O_\mu}(t) = \langle\langle 1|W^{O_\mu}(\alpha_t)|\rho(t)\rangle\rangle, \tag{2.24}$$

where $W^{O_\mu}(\alpha)$ is the current operator defined by

$$\langle\langle 1|W^{O_\mu}(\alpha) = \langle\langle 1|\hat{K}^{O_\mu}(\alpha), \tag{2.25}$$

i.e., $\mathrm{Tr}_S[W^{O_\mu}(\alpha)\bullet] = \mathrm{Tr}_S[\hat{K}^{O_\mu}(\alpha)\bullet]$ for any $\bullet \in \mathrm{B}$. Therefore, using (2.14), the current operator is given by

$$W^{O_\mu}(\alpha) = \sum_{b,a} c_{ba}^{O_\mu}(\alpha)B_aA_a. \tag{2.26}$$

Using (2.22), the *instantaneous steady current* is given by

$$\langle\langle 1|W^{O_\mu}(\alpha)|\rho_0(\alpha)\rangle\rangle = \lambda_0^{O_\mu}(\alpha) \equiv i_{\rm ss}^{O_\mu}(\alpha). \tag{2.27}$$

In the following, we suppose that the state of the system at $t = 0$ is the instantaneous steady state, $\rho(0) = \rho_0(\alpha_0)$. Then, $\rho(t) = \rho_0(\alpha_t) + \mathcal{O}(\omega/\Gamma)$ holds [25] where $\omega = 2\pi/\tau$ and $\Gamma = \min_{n\neq 0}\{-{\rm Re}(\lambda_n)\}$. In $\omega \ll \Gamma$ limit, we obtain

$$i^{O_\mu}(t) = i_{\rm ss}^{O_\mu}(\alpha_t) - \langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle + \mathcal{O}(\frac{\omega^2}{\Gamma}), \tag{2.28}$$

which leads to

$$\langle \Delta o_\mu\rangle_\tau = \int_0^\tau dt\ i_{\rm ss}^{O_\mu}(\alpha_t) + \int_C d\alpha^n\ A_n^{O_\mu}(\alpha) + \mathcal{O}(\frac{\omega}{\Gamma}), \tag{2.29}$$

where in the second term, the summation symbol $\sum_n$ is omitted. Here, α^n is the n-th component of the control parameters, C is the trajectory from α_0 to α_τ, and

$$A_n^{O_\mu}(\alpha) \stackrel{\rm def}{=} -\langle\langle l_0^{O_\mu}(\alpha)|\frac{\partial}{\partial\alpha^n}|\rho_0(\alpha)\rangle\rangle, \tag{2.30}$$

is the BSN vector. The BSN vector is also given by [25]

$$A_n^{O_\mu}(\alpha) = \langle\langle 1|W^{O_\mu}(\alpha)\mathcal{R}(\alpha)\frac{\partial}{\partial\alpha^n}|\rho_0(\alpha)\rangle\rangle, \tag{2.31}$$

where $\mathcal{R}(\alpha)$ is the pseudoinverse of the Liouvillian defined by

$$\mathcal{R}(\alpha)\hat{K}(\alpha) = 1 - |\rho_0(\alpha)\rangle\rangle\langle\langle 1|. \tag{2.32}$$

The expression of (2.29) was originally derived like the following. The formal solution of the FCS-QME is expanded as

$$|\rho^\chi(t)\rangle\rangle = \sum_n c_n^\chi(t) e^{\int_0^t ds\ \lambda_n^\chi(\alpha_s)}|\rho_n^\chi(\alpha_t)\rangle\rangle. \tag{2.33}$$

Because $e^{\int_0^t ds\ \lambda_n^\chi(\alpha_s)}$ $(n \neq 0)$ exponentially damps as a function of time, only $n = 0$ term remains if $\Gamma\tau \gg 1$. Solving the time evolution equation of $c_0^\chi(t)$ in $\omega \ll \Gamma$ limit, we obtain

$$c_0^\chi(\tau) = c_0^\chi(0)\exp\left[-\int_0^\tau dt\ \langle\langle l_0^\chi(\alpha_t)|\frac{d}{dt}|\rho_0^\chi(\alpha_t)\rangle\rangle\right]. \tag{2.34}$$

Here, the argument of the exponential function is called the BSN phase. Substituting this expression and $c_0^\chi(0) = \langle\langle l_0^\chi(\alpha_0)|\rho_0(\alpha_0)\rangle\rangle$ into (2.33), we obtain the expression of $\rho^\chi(\tau)$ which provides (2.29). However, when we consider only the average of Δo_μ, the BSN phase is not essential. All informations of the counting fields up to the first order are included in W^{O_μ} [2)]

[2)] In the research of adiabatic pumping, the expression of (2.29) is essential. In Refs.[23, 24, 25], (2.29) with (2.30) was used to study the quantum pump. On the other hand, in Ref. [35], (2.29) was derived using the generalized master equation [34] and without using the FCS. In Ref. [35], $A_n^{O_\mu}(\alpha)$ was described by the quantity corresponding to the current operator and the pseudoinverse of the Liouvillian, as shown in (2.31). Reference[25] showed the equivalence between the FCS-QME approach and the generalized master equation approach for all orders of pumping frequency.

As discussed in § 1, we propose to identify the average entropy production rate with

$$\dot{\sigma}(t) \stackrel{\text{def}}{=} \sum_b \beta_b(t) \left[-i^{H_b}(t) - \mu_b(t) \left\{-i^{N_b}(t)\right\}\right]. \tag{2.35}$$

This is given by $\dot{\sigma}(t) = \mathrm{Tr}_S[W^\sigma(\alpha_t)\rho(t)]$ with

$$W^\sigma(\alpha) \stackrel{\text{def}}{=} \sum_b \beta_b[-W^{H_b}(\alpha) - \mu_b\{-W^{N_b}(\alpha)\}]. \tag{2.36}$$

The average entropy production is given by

$$\begin{aligned} \sigma &\stackrel{\text{def}}{=} \int_0^\tau dt\ \dot{\sigma}(t) \\ &= \int_0^\tau dt\ J^\sigma_{\text{ss}}(\alpha_t) + \int_C d\alpha^n\ A^\sigma_n(\alpha) + \mathcal{O}(\frac{\omega}{\Gamma}), \end{aligned} \tag{2.37}$$

where

$$J^\sigma_{\text{ss}}(\alpha) \stackrel{\text{def}}{=} \sum_b \beta_b[-i^{H_b}_{\text{ss}}(\alpha) - \mu_b\{-i^{N_b}_{\text{ss}}(\alpha)\}] \tag{2.38}$$

and $A^\sigma_n(\alpha)$ is defined in (1.10). Here, we used (2.28) for $O_\mu = H_b, N_b$. The excess entropy production is defined as (1.9) by

$$\sigma_{\text{ex}} \stackrel{\text{def}}{=} \int_C d\alpha^n\ A^\sigma_n(\alpha) + \mathcal{O}(\frac{\omega}{\Gamma}). \tag{2.39}$$

2.2 Generalized quantum master equation for entropy production

We consider a kind of generalized quantum master equation (GQME)

$$\frac{d}{dt}\rho^\lambda(t) = \mathcal{K}^\lambda(\alpha_t)\rho^\lambda(t), \tag{2.40}$$

with the initial condition $\rho^\lambda(0) = \rho(0)$. Here, λ is a single real parameter. We suppose that the Liouvillian is given by

$$\mathcal{K}^\lambda(\alpha)\bullet = -i[H_S(\alpha_S), \bullet] + \sum_b \mathcal{L}^\lambda_b(\alpha)\bullet \tag{2.41}$$

with $\mathcal{L}^\lambda_b(\alpha)\bullet = \sum_a c^\lambda_{ba}(\alpha) A_a \bullet B_a$ and $c^\lambda_{ba}(\alpha)\big|_{\lambda=0} = c_{ba}$. While $c^\chi_{ba}(\alpha)$ of (2.14) depends on χ if and only if $A_a, B_a \neq 1$, $c^\lambda_{ba}(\alpha)$ can depend on λ for all a. We suppose that the solution of (2.40) satisfies

$$\mathrm{Tr}_S[\rho'(\tau)] = \sigma, \tag{2.42}$$

where $X' \stackrel{\text{def}}{=} \frac{\partial X^\lambda}{\partial(i\lambda)}\Big|_{\lambda=0}$. This condition is equivalent to

$$\langle\!\langle 1|\mathcal{K}'(\alpha) = \langle\!\langle 1|W^\sigma(\alpha). \tag{2.43}$$

Let's consider

$$\langle\!\langle l^\lambda_0(\alpha)|\mathcal{K}^\lambda(\alpha) = \lambda^\lambda_0(\alpha)\langle\!\langle l^\lambda_0(\alpha)|, \tag{2.44}$$

corresponding to (2.17) for $n = 0$. Similar to (2.27) and (2.30),

$$\lambda_0'(\alpha) = \langle\langle 1|W^\sigma(\alpha)|\rho_0(\alpha)\rangle\rangle = J_{\rm ss}^\sigma(\alpha), \tag{2.45}$$

and

$$A_n^\sigma(\alpha) = -\langle\langle l_0'(\alpha)|\frac{\partial}{\partial\alpha^n}|\rho_0(\alpha)\rangle\rangle = \langle\langle 1|W^\sigma(\alpha)\mathcal{R}(\alpha)\frac{\partial}{\partial\alpha^n}|\rho_0(\alpha)\rangle\rangle, \tag{2.46}$$

hold. Although $\lambda_0^\lambda(\alpha)$ and $l_0^\lambda(\alpha)$ depend on the choice of $\mathcal{K}^\lambda(\alpha)$, $\lambda_0'(\alpha)$ and $A_n^\sigma(\alpha)$ do not depend, as can be seen in the right-hand side (RHS) of the (2.45) and (2.46). The LHS of (2.43) is given by

$$\langle\langle 1|\mathcal{K}'(\alpha) = \langle\langle 1|\sum_{b,a} c_{ba}'(\alpha)B_aA_a. \tag{2.47}$$

Using this and (2.26), (2.43) becomes

$$\sum_{b,a} c_{ba}'(\alpha)B_aA_a = \sum_{b,a}\left[-\beta_b c_{ba}^{H_b}(\alpha) + \beta_b\mu_b c_{ba}^{N_b}(\alpha)\right]B_aA_a. \tag{2.48}$$

Infinite solutions of this equation exist. One choice of $\mathcal{K}^\lambda(\alpha)$ satisfying this relation is $\hat{K}^\chi(\alpha)$ in the limit of $\chi_{H_b} \to -\beta_b\lambda$ and $\chi_{N_b} \to \beta_b\mu_b\lambda$.

While we can calculate the average of the entropy production as shown in § 2.1 and in this subsection, our formalism is not compatible to discuss the higher moments of the entropy production. "Higher moments" $\frac{\partial^n}{\partial(i\lambda)^n}{\rm Tr}_S[\rho^\lambda(\tau)]\big|_{\lambda=0}$ $(n = 2, 3, \cdots)$ depend on the choice of $\mathcal{K}^\lambda(\alpha)$ and currently there seems no physical guiding principle to determine an adequate $\mathcal{K}^\lambda(\alpha)$. Although (2.29) is the average of the difference between outcomes at $t = \tau$ and $t = 0$ of O_μ, there is no bath's operator corresponding to σ if α_B are modulated. In contrast, the higher moments of the entropy production could be considered for the classical Markov jump process. In Appendix C, we review the entropy production of the Markov jump process [21, 37], and in § 4, we compare that and (2.37).

2.3 Rotating wave approximation

In this subsection, we introduce the FCS-QME within RWA. First, we introduce the CGA. An operator in the interaction picture corresponding to $A(t)$ is defined by $A^I(t) = U_0^\dagger(t)A(t)U_0(t)$ with

$$\frac{dU_0(t)}{dt} = -i\left[H_S(\alpha_S(t)) + \sum_b H_b\right]U_0(t) \tag{2.49}$$

and $U_0(0) = 1$. The system reduced density operator in the interaction picture is given by $\rho^{I,\chi}(t) = {\rm Tr}_B[\rho_{\rm tot}^{I,\chi}(t)]$ where $\rho_{\rm tot}^{I,\chi}(t) = U_0(t)\rho_{\rm tot}^\chi(t)U_0^\dagger(t)$. $\rho_{\rm tot}^{I,\chi}(t)$ is governed by

$$\frac{d\rho_{\rm tot}^{I,\chi}(t)}{dt} = -i[H_{\rm int}^I(t), \rho_{\rm tot}^{I,\chi}(t)]_\chi, \tag{2.50}$$

with $H_{\rm int} = \sum_b H_{Sb}$. Up to the second order perturbation in $H_{\rm int}$, we obtain

$$\begin{aligned}\rho^{I,\chi}(t+\tau_{\rm CG}) &= \rho^{I,\chi}(t) \\ &- \int_t^{t+\tau_{\rm CG}} du \int_t^u ds\ {\rm Tr}_B\left\{[H_{\rm int}^I(u), [H_{\rm int}^I(s), \rho^{I,\chi}(t)\otimes\rho_B(\alpha_B(t))]_\chi]_\chi\right\} \\ &\equiv \rho^{I,\chi}(t) + \tau_{\rm CG}\hat{L}_{\tau_{\rm CG}}^\chi(t)\rho^{I,\chi}(t),\end{aligned} \tag{2.51}$$

using the large-reservoir approximation:

$$\rho_{\rm tot}^{I,\chi}(t) \approx \rho^{I,\chi}(t) \otimes \rho_B(\alpha_B(t)) \tag{2.52}$$

and $\mathrm{Tr}_B[H_{\rm int}^I(u)\rho_B(\alpha_B(t))] = 0$. The arbitrary parameter $\tau_{\rm CG}$ (> 0) is called the coarse-graining time. The CGA [29, 30] is defined by

$$\frac{d}{dt}\rho^{I,\chi}(t) = \hat{L}_{\tau_{\rm CG}}^{\chi}(t)\rho^{I,\chi}(t). \tag{2.53}$$

At $\chi = 0$, this is Lindblad type. If $\tau \gg \tau_{\rm CG}$, the superoperator $\hat{L}_{\tau_{\rm CG}}^{\chi}(t)$ is described as a function of the set of control parameters at time t. In this paper, we suppose $\tau \gg \tau_{\rm CG}$. Moreover, $\tau_{\rm CG}$ should be much shorter than the relaxation time of the system, $\tau_S \sim \frac{1}{\Gamma}$. On the other hand, $\tau_S \ll \tau$ should hold for the adiabatic modulation. Hence $\tau_{\rm CG} \ll \frac{1}{\Gamma} \ll \tau$ should hold. By the way, the Born-Markov approximation is given by

$$\frac{d\rho^{I,\chi}(t)}{dt} = -\int_0^\infty ds\ \mathrm{Tr}_B\Big\{[H_{\rm int}^I(t), [H_{\rm int}^I(t-s), \rho^{I,\chi}(t) \otimes \rho_B(\alpha_B(t))]_\chi]_\chi\Big\}. \tag{2.54}$$

Now we suppose

$$H_{Sb}(\alpha_{Sb}) = \sum_{k,\alpha} V_{bk,\alpha}(\alpha_{Sb}) a_\alpha^\dagger c_{bk} + \text{h.c.}, \tag{2.55}$$

where a_α and c_{bk} are single-particle annihilation operators of the system and of the bath b. Although we have used indeces α or β to distinguish the system operators, this may not confuse the readers with the set of control parameters or the inverse temperature since they only appear as a subscript of the operator a (or $a^\dagger$) and the parameters like $V_{bk,\alpha}$, $\Phi^{\pm}_{b,\alpha\beta}$, $\Psi^{\pm}_{b,\alpha\beta}$ or under the summation symbol. The eigenoperator defined by

$$a_\alpha(\omega) = \sum_{n,r,m,s} \delta_{\omega_{mn},\omega} |E_n, r\rangle\langle E_n, r|a_\alpha|E_m, s\rangle\langle E_m, s|. \tag{2.56}$$

is useful to describe the FCS-QME. Here, $\omega_{mn} \overset{\rm def}{=} E_m - E_n$,

$$H_S|E_n, r\rangle = E_n|E_n, r\rangle \tag{2.57}$$

and r denotes the label of the degeneracy. ω is one of the elements of

$$\mathcal{W} = \{\omega_{mn}|\ \langle E_n, r|a_\alpha|E_m, s\rangle \neq 0\ \ ^\exists\alpha\}. \tag{2.58}$$

$a_\alpha(\omega)$ and ω depend on α_S. $\sum_\omega a_\alpha(\omega) = a_\alpha$,

$$[H_S, a_\alpha(\omega)] = -\omega a_\alpha(\omega) \ \text{ and } \ [N_S, a_\alpha(\omega)] = -a_\alpha(\omega) \tag{2.59}$$

hold. Here, N_S is total number operator of the system. We suppose $[N_S, H_S] = 0$. In the CGA or Born-Markov approximation, the FCS-QME is described by $a_\alpha(\omega)$ and $[a_\alpha(\omega')]^\dagger$ ($\omega, \omega' \in \mathcal{W}$). If H_S is time dependent, the generalization of usual RWA [28] with static H_S is unclear. In this paper, the RWA is defined as the limit $\tau_{\rm CG} \to \infty$ ($\tau_{\rm CG} \cdot \min_{\omega \neq \omega'} |\omega - \omega'| \gg 1$) of the CGA. If H_S is time independent, this RWA is equivalent to usual RWA.

In the following, except for § 3.3, we consider the RWA. Then, $\mathcal{L}_b^\chi(\alpha)$ is generally given by (for the details of the derivation, please refer [25, 26])

$$\mathcal{L}_b^\chi(\alpha)\bullet = \Pi_b^\chi(\alpha)\bullet - i[h_b(\alpha), \bullet], \tag{2.60}$$

where $h_b(\alpha)$ is a Hermitian operator describing the *Lamb shift.* $h_b(\alpha)$ commutes with $H_S(\alpha_S)$ for general model and with N_S for the model (2.55). $H_\mathrm{L}(\alpha) \stackrel{\mathrm{def}}{=} \sum_b h_b(\alpha)$ is called *Lamb shift Hamiltonian.* The superoperator $\Pi_b^\chi(\alpha)$ represents the dissipation.

Here, we suppose the free Hamiltonian of the bath b:

$$H_b = \sum_k \varepsilon_{bk} c_{bk}^\dagger c_{bk}, \tag{2.61}$$

and $\{O_\mu\} = \{N_b, H_b\}_b$. $\Pi_b^\chi(\alpha)$ in (2.60) is given by

$$\begin{aligned} \Pi_b^\chi(\alpha)\bullet \;=\; & \sum_\omega \sum_{\alpha,\beta} \Big[\Phi_{b,\alpha\beta}^{-,\chi}(\omega) a_\beta(\omega) \bullet [a_\alpha(\omega)]^\dagger - \frac{1}{2}\Phi_{b,\alpha\beta}^{-}(\omega) \bullet [a_\alpha(\omega)]^\dagger a_\beta(\omega) \\ & - \frac{1}{2}\Phi_{b,\alpha\beta}^{-}(\omega)[a_\alpha(\omega)]^\dagger a_\beta(\omega)\bullet + \Phi_{b,\alpha\beta}^{+,\chi}(\omega)[a_\beta(\omega)]^\dagger \bullet a_\alpha(\omega) \\ & - \frac{1}{2}\Phi_{b,\alpha\beta}^{+}(\omega) \bullet a_\alpha(\omega)[a_\beta(\omega)]^\dagger - \frac{1}{2}\Phi_{b,\alpha\beta}^{+}(\omega) a_\alpha(\omega)[a_\beta(\omega)]^\dagger \bullet \Big], \end{aligned} \tag{2.62}$$

where

$$\begin{aligned} \Phi_{b,\alpha\beta}^{-,\chi}(\Omega) &= 2\pi \sum_k V_{bk,\alpha} V_{bk,\beta}^* F_b^-(\varepsilon_{bk}) e^{i\chi_{N_b}} e^{i\chi_{H_b}\varepsilon_{bk}} \delta(\varepsilon_{bk} - \Omega) \\ &= e^{i\chi_{N_b} + i\chi_{H_b}\Omega} \Phi_{b,\alpha\beta}^{-}(\Omega), \end{aligned} \tag{2.63}$$

$$\begin{aligned} \Phi_{b,\alpha\beta}^{+,\chi}(\Omega) &= 2\pi \sum_k V_{bk,\alpha}^* V_{bk,\beta} F_b^+(\varepsilon_{bk}) e^{-i\chi_{N_b}} e^{-i\chi_{H_b}\varepsilon_{bk}} \delta(\varepsilon_{bk} - \Omega) \\ &= e^{-i\chi_{N_b} - i\chi_{H_b}\Omega} \Phi_{b,\alpha\beta}^{+}(\Omega). \end{aligned} \tag{2.64}$$

Here, χ_{N_b} and χ_{H_b} are the counting fields for N_b and H_b. If the baths are fermions,

$$F_b^+(\varepsilon) = f_b(\varepsilon) \stackrel{\mathrm{def}}{=} \frac{1}{e^{\beta_b(\varepsilon - \mu_b)} + 1} \tag{2.65}$$

and $F_b^-(\varepsilon) = 1 - f_b(\varepsilon)$. If the baths are bosons,

$$F_b^+(\varepsilon) = n_b(\varepsilon) \stackrel{\mathrm{def}}{=} \frac{1}{e^{\beta_b(\varepsilon - \mu_b)} - 1} \tag{2.66}$$

and $F_b^-(\varepsilon) = 1 + n_b(\varepsilon)$. The Lamb shift is given by

$$h_b(\alpha) = \sum_\omega \sum_{\alpha,\beta} \Big(-\frac{1}{2}\Psi_{b,\alpha\beta}^{-}(\omega)[a_\alpha(\omega)]^\dagger a_\beta(\omega) + \frac{1}{2}\Psi_{b,\alpha\beta}^{+}(\omega) a_\alpha(\omega)[a_\beta(\omega)]^\dagger \Big), \tag{2.67}$$

where

$$\Psi_{b,\alpha\beta}^{-}(\Omega) = 2\sum_k V_{bk,\alpha} V_{bk,\beta}^* F_b^-(\varepsilon_{bk}) \mathrm{P}\frac{1}{\varepsilon_{bk} - \Omega}, \tag{2.68}$$

$$\Psi_{b,\alpha\beta}^{+}(\Omega) = 2\sum_k V_{bk,\alpha}^* V_{bk,\beta} F_b^+(\varepsilon_{bk}) \mathrm{P}\frac{1}{\varepsilon_{bk} - \Omega}. \tag{2.69}$$

Here, P denotes the Cauchy principal value. $\Phi^{\pm}_{b,\alpha\beta}(\Omega)$ satisfy

$$
\begin{aligned}
[\Phi^{\pm}_{b,\alpha\beta}(\Omega)]^* &= \Phi^{\pm}_{b,\beta\alpha}(\Omega), &(2.70)\\
\Phi^{+}_{b,\alpha\beta}(\Omega) &= e^{-\beta_b(\Omega-\mu_b)}\Phi^{-}_{b,\beta\alpha}(\Omega). &(2.71)
\end{aligned}
$$

The latter is the Kubo-Martin-Schwinger (KMS) condition.

We introduce projection superoperators $\mathcal{P}(\alpha_S)$ and $\mathcal{Q}(\alpha_S)$ by

$$
\mathcal{P}(\alpha_S)|E_n, r\rangle\langle E_m, s| = \delta_{E_n,E_m}|E_n, r\rangle\langle E_m, s|, \tag{2.72}
$$

and $\mathcal{Q}(\alpha_S) = 1 - \mathcal{P}(\alpha_S)$. We define sets of operators $\mathrm{B_P} \stackrel{\text{def}}{=} \{X \in \mathrm{B}|\mathcal{P}X = X\}$ and $\mathrm{B_Q} \stackrel{\text{def}}{=} \{X \in \mathrm{B}|\mathcal{Q}X = X\}$. $\hat{K}^\chi\mathcal{P}\bullet \in \mathrm{B_P}$ holds. Then, $\hat{K}^\chi\mathcal{Q}\bullet \in \mathrm{B_Q}$ and

$$
\mathcal{Q}\hat{K}^\chi\mathcal{P} = 0 = \mathcal{P}\hat{K}^\chi\mathcal{Q}, \tag{2.73}
$$

hold. This implies that the right eigenvalue equations (2.16) are decomposed into two closed systems of equations for $\mathcal{P}\rho_n^\chi$ and for $\mathcal{Q}\rho_n^\chi$. Thus, ρ_n^χ is an element of $\mathrm{B_P}$ or $\mathrm{B_Q}$. In particular, $\rho_0^\chi \in \mathrm{B_P}$. Then, the matrix representation of $\rho_0(\alpha)$ by $|E_n, r\rangle$ is block diagonalized. This implies

$$
[H_S(\alpha_S), \rho_0(\alpha)] = 0. \tag{2.74}
$$

The particle and energy current operators from the system into bath b, $w^{N_b}(\alpha)$ and $w^{H_b}(\alpha)$, are usually defined by

$$
w^{X_b}(\alpha) \stackrel{\text{def}}{=} -[\mathcal{L}_b^\dagger(\alpha)X_S]^\dagger = -\mathcal{L}_b^\dagger(\alpha)X_S \quad (X = N, H). \tag{2.75}
$$

For a superoperator $\mathcal{J}$, $\mathcal{J}^\dagger$ is defined by $\langle\langle \mathcal{J}^\dagger X|Y\rangle\rangle = \langle\langle X|\mathcal{J}Y\rangle\rangle$ $(X, Y \in \mathrm{B})$. $\mathcal{L}_b^\dagger(\alpha)\bullet = \sum_a c^*_{ba}(\alpha)A_a^\dagger \bullet B_a^\dagger$ holds. $w^{X_b}(\alpha)$ is a Hermitian operator and is given by

$$
w^{X_b}(\alpha) = -\sum_a c_{ba}(\alpha)B_a X_S A_a \quad (X = N, H). \tag{2.76}
$$

For the Born-Markov approximation and the CGA, $w^{N_b}(\alpha) = W^{N_b}$, while $w^{H_b}(\alpha) \neq W^{H_b}(\alpha)$. For RWA,

$$
\begin{aligned}
w^{N_b}(\alpha) &= W^{N_b}(\alpha)\\
&= \sum_\omega\sum_{\alpha,\beta}\Big\{\Phi^-_{b,\alpha\beta}(\omega)[a_\alpha(\omega)]^\dagger a_\beta(\omega) - \Phi^+_{b,\alpha\beta}(\omega)a_\alpha(\omega)[a_\beta(\omega)]^\dagger\Big\}, &(2.77)\\
w^{H_b}(\alpha) &= W^{H_b}(\alpha)\\
&= \sum_\omega\sum_{\alpha,\beta}\Big\{\omega\Phi^-_{b,\alpha\beta}(\omega)[a_\alpha(\omega)]^\dagger a_\beta(\omega) - \omega\Phi^+_{b,\alpha\beta}(\omega)a_\alpha(\omega)[a_\beta(\omega)]^\dagger\Big\}, &(2.78)
\end{aligned}
$$

hold. Therefore, (2.36) and (2.75) imply that $W^\sigma(\alpha)$ is given by

$$
W^\sigma(\alpha) = \sum_b \mathcal{L}_b^\dagger(\alpha)(\beta_b H_S - \beta_b\mu_b N_S) = \sum_b \Pi_b^\dagger(\alpha)(\beta_b H_S - \beta_b\mu_b N_S). \tag{2.79}
$$

3 Geometrical expression of excess entropy production

3.1 Equilibrium state

In this subsection, we consider equilibrium state $\beta_b = \beta$ and $\mu_b = \mu$, and α denotes the set of (α_S, $\{\alpha_{Sb}\}_b$, β, $\beta\mu$). We show that $A_n^\sigma(\alpha)$ is a total derivative of the von Neumann entropy of the instantaneous steady state. Differentiating (2.44) by $i\lambda$ and setting $\lambda = 0$, we obtain

$$\langle\langle l_0'(\alpha)|\hat{K}(\alpha) + \langle\langle 1|\mathcal{K}'(\alpha) = \lambda_0'(\alpha)\langle\langle 1|. \tag{3.1}$$

In the RHS, $\lambda_0'(\alpha) = J_{\rm ss}^\sigma(\alpha) = 0$ holds. The second term of the LHS is $\langle\langle 1|W^\sigma(\alpha)$. (2.79) leads

$$W^\sigma(\alpha) = \beta \sum_b \mathcal{L}_b^\dagger(\alpha)[H_S - \mu N_S] = \beta \hat{K}^\dagger(\alpha)[H_S - \mu N_S], \tag{3.2}$$

i.e.,

$$\langle\langle \beta[H_S - \mu N_S]|\hat{K}(\alpha) = \langle\langle 1|W^\sigma(\alpha). \tag{3.3}$$

Then, (3.1) leads

$$\big[\langle\langle l_0'(\alpha)| + \langle\langle \beta[H_S - \mu N_S]|\big]\hat{K}(\alpha) = 0. \tag{3.4}$$

This implies

$$\langle\langle l_0'(\alpha)| = -\langle\langle \beta[H_S - \mu N_S]| + c(\alpha)\langle\langle 1|, \tag{3.5}$$

i.e., $\{l_0'(\alpha)\}^\dagger = -\beta[H_S - \mu N_S] + c(\alpha)$ where $c(\alpha)$ is an unimportant complex number. The equilibrium state, $\rho_0(\alpha)$, is given by

$$\rho_0(\alpha) = \rho_{\rm gc}(\alpha_S; \beta, \beta\mu) \stackrel{\rm def}{=} \frac{e^{-\beta(H_S(\alpha_S) - \mu N_S)}}{\Xi(\alpha_S; \beta, \beta\mu)}, \tag{3.6}$$

with $\Xi(\alpha_S; \beta, \beta\mu) \stackrel{\rm def}{=} {\rm Tr}_S[e^{-\beta(H_S(\alpha_S) - \mu N_S)}]$. Then,

$$\{l_0'(\alpha)\}^\dagger = \ln \rho_{\rm gc}(\alpha_S; \beta, \beta\mu) + c'(\alpha)1 \tag{3.7}$$

with $c'(\alpha) = c(\alpha) + \ln \Xi(\alpha_S; \beta, \beta\mu)$, holds. Substituting this equation into (2.46), we obtain

$$A_n^\sigma(\alpha) = \frac{\partial}{\partial \alpha^n} S_{\rm vN}(\rho_{\rm gc}(\alpha_S; \beta, \beta\mu)), \tag{3.8}$$

where we used (A.1) in the Appendix A.

3.2 Weakly nonequilibrium regime

In this subsection, we study the BSN vector and the excess entropy production in a weakly nonequilibrium condition. We introduce parameters characterizing the degree of nonequilibrium:

$$\varepsilon_{1,b} \stackrel{\rm def}{=} \beta_b - \overline{\beta},\ \varepsilon_{2,b} \stackrel{\rm def}{=} \beta_b\mu_b - \overline{\beta\mu},\ \varepsilon \stackrel{\rm def}{=} \max_b \Big\{\frac{|\varepsilon_{1,b}|}{\overline{\beta}}, \frac{|\varepsilon_{2,b}|}{|\overline{\beta\mu}|}\Big\}, \tag{3.9}$$

where $\overline{\beta}$ and $\overline{\beta\mu}$ are the reference values, which satisfy $\min_b \beta_b \leq \overline{\beta} \leq \max_b \beta_b$ and $\min_b \beta_b\mu_b \leq \overline{\beta\mu} \leq \max_b \mu_b\beta_b$. ε is a measure of degree of nonequilibrium. We consider $\varepsilon \ll 1$ regime. Now, we introduce

$$\hat{K}_\kappa(\alpha)\bullet \stackrel{\text{def}}{=} -i[H_S(\alpha_S) + \kappa H_{\rm L}(\alpha), \bullet] + \sum_b \Pi_b(\alpha)\bullet, \tag{3.10}$$

and corresponding instantaneous steady state $\rho_0^{(\kappa)}(\alpha)$:

$$\hat{K}_\kappa(\alpha)\rho_0^{(\kappa)}(\alpha) = 0. \tag{3.11}$$

Here, κ is a real parameter satisfying $-1 \leq \kappa \leq 1$ controlling the Lamb shift Hamiltonian. $\langle\!\langle 1|\hat{K}_\kappa(\alpha) = 0$ holds. We use the following notations:

$$\alpha_{1,b} \stackrel{\text{def}}{=} \beta_b,\ \alpha_{2,b} \stackrel{\text{def}}{=} \beta_b\mu_b,\ \overline{X} \stackrel{\text{def}}{=} X\big|_{\alpha_{i,b}=\overline{\alpha_i}}. \tag{3.12}$$

We expand $\rho_0^{(\kappa)}$ and l_0' (the derivative of $n=0$ left eigenavector for $\kappa = +1$)

$$\rho_0^{(\kappa)}(\alpha) = \overline{\rho_0^{(\kappa)}} + \sum_b \left(\varepsilon_{1,b}\rho_{1,b}^{(\kappa)} + \varepsilon_{2,b}\rho_{2,b}^{(\kappa)}\right) + \mathcal{O}(\varepsilon^2), \tag{3.13}$$

$$l_0'(\alpha) = \overline{l_0'(\alpha)} + \sum_b (\varepsilon_{1,b}k_{1,b} + \varepsilon_{2,b}k_{2,b}) + \mathcal{O}(\varepsilon^2), \tag{3.14}$$

with

$$\overline{\rho_0^{(\kappa)}} = \rho_{\rm gc}, \quad \overline{l_0'(\alpha)} = -\overline{\beta}H_S + \overline{\beta\mu}N_S + \overline{c}^*1 = \ln\rho_{\rm gc} + \overline{c'}^*1. \tag{3.15}$$

Here, $\rho_{\rm gc} \stackrel{\text{def}}{=} \rho_{\rm gc}(\alpha_S;\overline{\beta},\overline{\beta\mu})$, $\overline{c}$ and $\overline{c'}$ are the same with $c(\alpha)$ and $c'(\alpha)$ in § 3.1. After some calculations, we obtain following relation ($i = 1, 2$):

$$k_{i,b} = \rho_{i,b}^{(-1)}\rho_{\rm gc}^{-1} + \overline{c_{i,b}}1, \tag{3.16}$$

where $\overline{c_{i,b}}$ is an arbitrary complex number. The details of the derivation are explained in the Appendix B. Using this relation, (3.14) becomes

$$\begin{aligned} l_0'(\alpha) &= \ln\rho_{\rm gc}(\alpha_S;\overline{\beta},\overline{\beta\mu}) + C(\alpha)1 + \sum_b\sum_{i=1}^{2}\varepsilon_{i,b}\rho_{i,b}^{(-1)}\rho_{\rm gc}^{-1} + \mathcal{O}(\varepsilon^2) \\ &= \ln\rho_0^{(-1)}(\alpha) + C(\alpha)1 + \mathcal{O}(\varepsilon^2), \end{aligned} \tag{3.17}$$

where $C(\alpha) \stackrel{\text{def}}{=} \overline{c'}^* + \sum_{b,i}\overline{c_{i,b}}\varepsilon_{i,b}$. Substituting this equation into (2.46), we obtain

$$A_n^\sigma(\alpha) = -\mathrm{Tr}_S\Big[\ln\rho_0^{(-1)}(\alpha)\frac{\partial\rho_0^{(1)}(\alpha)}{\partial\alpha^n}\Big] + \mathcal{O}(\varepsilon^2), \tag{3.18}$$

where the notation $\rho_0(\alpha) = \rho_0^{(1)}(\alpha)$ and $\breve{\rho}_0(\alpha) = \rho_0^{(-1)}(\alpha)$ is used in § 1 for clarity. We supposed $[\rho_{\rm gc}, \rho_{i,b}^{(-1)}] = 0$, which leads

$$\ln\rho_0^{(-1)}(\alpha) = \ln\rho_{\rm gc} + \sum_{i,b}\varepsilon_{i,b}\rho_{i,b}^{(-1)}\rho_{\rm gc}^{-1} + \mathcal{O}(\varepsilon^2). \tag{3.19}$$

This supposition is satisfied if $[N_S, \rho_0^{(-1)}(\alpha)] = \mathcal{O}(\varepsilon^2)$ (which leads $[N_S, \rho_{i,b}^{(-1)}] = 0$) or $\overline{\beta\mu} = 0$ holds. If H_S is non-degenerate, $[N_S, \rho_0^{(-1)}(\alpha)] = 0$ holds, then $[N_S, \rho_{i,b}^{(-1)}] = 0$, $[\rho_{\rm gc}, \rho_{i,b}^{(-1)}] = 0$ and (3.17) hold. If the states of the baths are in the canonical distributions ($\mu_b \to 0$), $\rho_{\rm gc}$ is replaced by the canonical distribution and (3.17) holds without any assumption.

If

$$[H_{\rm L}(\alpha), \rho_0^{(\kappa)}(\alpha)] = 0, \tag{3.20}$$

holds, $\rho_0^{(\kappa)}(\alpha)$ is independent of κ ($\rho_0^{(\kappa)}(\alpha) = \rho_0(\alpha)$), then (3.18) becomes

$$A_n^\sigma(\alpha) = \frac{\partial}{\partial \alpha^n} S_{\rm vN}(\rho_0(\alpha)) + \mathcal{O}(\varepsilon^2), \tag{3.21}$$

using (A.1). (3.20) holds if H_S is non-degenerate. (3.21) can be shown from $[\overline{H_{\rm L}}, \rho_{i,b}^{(1)}] = 0$, which is weaker assumption than (3.20) and is derived from (3.20) for $\kappa = 1$. If we neglect the Lamb shift Hamiltonian, namely we consider the QME for $\hat{K}_0(\alpha)$, (3.21) holds (with a replacement $\rho_0 \to \rho_0^{(0)}$). From (3.21), we obtain

$$\sigma_{\rm ex} = S_{\rm vN}(\rho_0(\alpha_\tau)) - S_{\rm vN}(\rho_0(\alpha_0)) + \mathcal{O}(\varepsilon^2 \delta), \tag{3.22}$$

with

$$\delta = \max_{n, \alpha \in C} \frac{|\alpha^n - \alpha_0^n|}{|\bar{\alpha}^n|}, \tag{3.23}$$

where $\bar{\alpha}^n$ is typical value of the n-th control parameter.

Yuge *et al.* [20] applied the FCS-QME approach to the excess entropy production of the quantum system. They introduced a *time-dependent observable* $A(t) = -\sum_b \beta_b(t)[H_b - \mu_b(t) N_b]$ and considered the outputs at $t = 0$ and $t = \tau$ as $a(0)$ and $a(\tau)$. Then, they identified the average $\sigma' \stackrel{\rm def}{=} \langle a(\tau) - a(0) \rangle$ as the average entropy production. However, σ' seems not the average entropy production σ. The average σ' can be rewritten as

$$\begin{aligned} \sigma' &\approx \mathrm{Tr}_{\rm tot}[A(\tau)\rho_{\rm tot}(\tau)] - \mathrm{Tr}_{\rm tot}[A(0)\rho_{\rm tot}(0)] = \int_0^\tau dt \left\{ \frac{d}{dt} \mathrm{Tr}_{\rm tot}[A(t)\rho_{\rm tot}(t)] \right\} \\ &\approx -\int_0^\tau dt \sum_b \Big[\frac{d\beta_b(t)}{dt} \langle H_b \rangle_t - \frac{d[\beta_b(t)\mu_b(t)]}{dt} \langle N_b \rangle_t \Big] \\ &\quad + \int_0^\tau dt \sum_b \Big[\beta_b(t)\{-\frac{d}{dt}\langle H_b \rangle_t\} - \beta_b(t)\mu_b(t)\{-\frac{d}{dt}\langle N_b \rangle_t\} \Big]. \end{aligned} \tag{3.24}$$

Here, $\langle \bullet \rangle_t \stackrel{\rm def}{=} \mathrm{Tr}_{\rm tot}[\bullet \rho_{\rm tot}(t)]$, $\rho_{\rm tot}(t)$ is the total system state and $\mathrm{Tr}_{\rm tot}$ denotes the trace of the total system. The integrand of the second term of the last expression of (3.24) roughly equals to $\dot{\sigma}$ [3)] The first term, while its physical meaning is not clear, is nonzero in general. Moreover, it should be noted that the FCS-QME is applicable only for a *time-independent* observable although $A(t)$ is time-dependent. These two issues are the problems of Ref. [20]. Nevertheless, the obtained Liouvillian (of which the Lamb shift Hamiltonian is neglected) incidentally satisfies (2.43). Using that Liouvillian, for the system with time-reversal symmetry, Yuge *et al.* studied the relation between $A_n^\sigma(\alpha)$ and the symmetrized von Neumann entropy. In contrast, we do not suppose the time-reversal symmetry to derive (3.18). In § 5, we consider the time-reversal symmetric system.

[3)] Here, we supposed $\frac{d}{dt}\langle O \rangle_t \approx i^O(t)$ for $O = H_b, N_b$. However, because the thermodynamic parameters β_b and μ_b are modulated, $\frac{d}{dt}\langle H_b \rangle_t$ and $\frac{d}{dt}\langle N_b \rangle_t$ also include the currents from the outside of the total system to the bath b.

3.3 Born-Markov approximation

We denote the BSN vector for the entropy production and instantaneous steady state of the Born-Markov approximation by $A_n^{\sigma,\mathrm{BM}}(\alpha)$ and $\rho_0^{\mathrm{BM}}(\alpha)$. Then,

$$\begin{aligned} A_n^{\sigma,\mathrm{BM}}(\alpha) &= A_n^{\sigma}(\alpha) + \mathcal{O}(v^2), && (3.25)\\ S_{\mathrm{vN}}(\rho_0^{\mathrm{BM}}(\alpha)) &= S_{\mathrm{vN}}(\rho_0(\alpha)) + \mathcal{O}(v^2), && (3.26) \end{aligned}$$

hold [20]. Here, $v = u^2$ and $u(\ll 1)$ describes the order of H_{Sb}. The above two equations and (3.21) lead

$$A_n^{\sigma,\mathrm{BM}}(\alpha) = \frac{\partial}{\partial \alpha^n} S_{\mathrm{vN}}(\rho_0^{\mathrm{BM}}(\alpha)) + \mathcal{O}(\varepsilon^2) + \mathcal{O}(v^2). \tag{3.27}$$

4 Comparison of two definitions of entropy production

In this section, we compare the preceding study on the entropy production in the classical Markov jump process [21, 37] with ours. We consider the Markov jump process among the states $n = 1, 2, \cdots, \mathcal{N}$, where the definitions are explained in Appendix C. The probability to find the system in a state n is $p_n(t)$ and it obeys the master equation:

$$\frac{dp_n(t)}{dt} = \sum_{m=1}^{\mathcal{N}} K_{nm}(\alpha_t) p_m(t). \tag{4.1}$$

The Liouvillian is given by

$$K_{nm}(\alpha) = \sum_b K_{nm}^{(b)}(\alpha) \tag{4.2}$$

where $K_{nm}^{(b)}$ originates the couping between the system and the bath b. $\sum_n K_{nm}^{(b)}(\alpha) = 0$ holds. We suppose that $K_{mn}^{(b)}(\alpha) \neq 0 (= 0)$ holds if $K_{nm}^{(b)}(\alpha) \neq 0 (= 0)$ for all $n \neq m$. The definition of the entropy production for each Markov jump process (C.1) is (C.4). The average entropy production σ^{C} is given by (see (C.10))

$$\sigma^{\mathrm{C}} = \int_0^\tau dt \sum_{n,m} \sigma_{nm}^{\mathrm{C}}(\alpha_t) p_m(t), \tag{4.3}$$

where

$$\sigma_{nm}^{\mathrm{C}}(\alpha) = -K_{nm}(\alpha) \ln \frac{K_{nm}(\alpha)}{K_{mn}(\alpha)}. \tag{4.4}$$

We denote the solution of the QME with RWA by $\rho(t)$. We suppose $p_n(t) \overset{\mathrm{def}}{=} \langle n|\rho(t)|n\rangle$ is governed by (4.1) with $K_{nm}^{(b)}(\alpha) = (\Pi_b(\alpha))_{nn,mm}$. Here, $|n\rangle$ is the energy eigenstate of $H_S(\alpha_S)$,

$$(\Pi_b \bullet)_{nm} = \sum_{k,l} (\Pi_b(\alpha))_{nm,kl} (\bullet)_{kl} \tag{4.5}$$

and $(\bullet)_{kl} \overset{\mathrm{def}}{=} \langle k| \bullet |n\rangle$. This supposition implies (3.20). A sufficient condition by which $p_n(t)$ obeys (4.1) is below: (1) $H_S(\alpha_S)$ is non-degenerate and (2) $\{\alpha^n \in \alpha_S |\ \frac{\partial}{\partial \alpha^n}|n\rangle \neq 0\}$ are fixed. The eigenenergy can depend on $\{\alpha^n \in \alpha_S | \frac{\partial}{\partial \alpha^n}|n\rangle = 0\}$. We show that our average entropy production (2.37) is given by a similar expression of (4.3):

$$\sigma = \int_0^\tau dt \sum_{n,m} \sigma_{nm}(\alpha_t) p_m(t). \tag{4.6}$$

Here,

$$\sigma_{nm}(\alpha) \stackrel{\text{def}}{=} \sum_b K_{nm}^{(b)}(\alpha)\theta_{nm}^{(b)}(\alpha) = -\sum_b K_{nm}^{(b)}(\alpha) \ln \frac{K_{nm}^{(b)}(\alpha)}{K_{mn}^{(b)}(\alpha)}, \tag{4.7}$$

with

$$\theta_{nm}^{(b)}(\alpha) \stackrel{\text{def}}{=} \begin{cases} -\ln \frac{K_{nm}^{(b)}(\alpha)}{K_{mn}^{(b)}(\alpha)} & K_{nm}^{(b)}(\alpha) \neq 0 \\ 0 & K_{nm}^{(b)}(\alpha) = 0 \end{cases}. \tag{4.8}$$

Because of (2.75), (2.77) and (2.78), the particle and energy currents are given by $i^{X_b} = \mathrm{Tr}_S[W^{X_b}\rho(t)]$ with $W^{X_b} = -(\Pi_b^\dagger X_S)^\dagger$ $(X = H, N)$. (2.76) leads

$$(W^{X_b})_{nm} = -\sum_{k,l} (\Pi_b)_{lk,mn}(X_S)_{kl}. \tag{4.9}$$

We suppose $(X_S)_{nm} = (X_S)_{nn}\delta_{nm}$ for $X = N, H$. Since $(X_S)_{kl}$ is a diagonal matrix, $(W^{X_b})_{nm}$ is also a diagonal matrix. Then,

$$i^{X_b} = \sum_m (W^{X_b})_{mm} p_m(t), \tag{4.10}$$

holds. Substituting $(W^{X_b})_{mm} = -\sum_n K_{nm}^{(b)}(X_S)_{nn}$ into (4.10), we obtain

$$\begin{aligned} i^{X_b} &= -\sum_{n,m} K_{nm}^{(b)}(X_S)_{nn} p_m(t) \\ &= \sum_{n,m} K_{nm}^{(b)} \left[(X_S)_{mm} - (X_S)_{nn}\right] p_m(t). \end{aligned} \tag{4.11}$$

This equation leads

$$\dot{\sigma}(t) = -\sum_{n,m}\sum_b K_{nm}^{(b)}\beta_b(t)\{[(H_S)_{mm} - (H_S)_{nn}] - \mu_b(t)[(N_S)_{mm} - (N_S)_{nn}]\}p_m(t). \tag{4.12}$$

Using the local detailed balance condition

$$\ln \frac{K_{nm}^{(b)}(\alpha)}{K_{mn}^{(b)}(\alpha)} = \beta_b\{[(H_S)_{mm} - (H_S)_{nn}] - \mu_b[(N_S)_{mm} - (N_S)_{nn}]\}, \tag{4.13}$$

we obtain (4.6).

Now we introduce a matrix $\mathcal{K}^\lambda(\alpha)$ by

$$[\mathcal{K}^\lambda(\alpha)]_{nm} \stackrel{\text{def}}{=} \sum_b K_{nm}^{(b)}(\alpha) e^{i\lambda\theta_{nm}^{(b)}(\alpha)}. \tag{4.14}$$

Then, we obtain

$$\frac{\partial}{\partial(i\lambda)}\Big|_{\lambda=0}\sum_{n,m}\Big[\mathrm{T}\exp\big[\int_0^\tau dt\ \mathcal{K}^\lambda(\alpha_t)\big]\Big]_{nm}p_m(0)=\int_0^\tau dt\ \sum_{n,m}\sigma_{nm}(\alpha_t)p_m(t)=\sigma. \tag{4.15}$$

$\mathcal{K}^\lambda$ was originally introduced by Sagawa and Hayakawa [19]. About averages, our entropy production is the same with Sagawa and Hayakawa.

We show that the difference between $\sigma^{\mathrm{C}}_{nm}(\alpha)$ and $\sigma_{nm}(\alpha)$ is $\mathcal{O}(\varepsilon^2)$:

$$\sigma^{\mathrm{C}}_{nm}(\alpha)=\sigma_{nm}(\alpha)+\mathcal{O}(\varepsilon^2). \tag{4.16}$$

In fact, $K^{(b)}_{nm}$ can be expanded as

$$K^{(b)}_{nm}=\gamma_b\bar{K}_{nm}+\sum_{i=1,2}\varepsilon_{i,b}K^{i,b}_{nm}+\mathcal{O}(\varepsilon^2),\quad \sum_b\gamma_b=1, \tag{4.17}$$

then we obtain

$$\sigma^{\mathrm{C}}_{nm}(\alpha) = \sigma^{(0,1)}_{nm}+\sigma^{\mathrm{C}(2)}_{nm}(\alpha)+\mathcal{O}(\varepsilon^3), \tag{4.18}$$
$$\sigma_{nm}(\alpha) = \sigma^{(0,1)}_{nm}+\sigma^{(2)}_{nm}(\alpha)+\mathcal{O}(\varepsilon^3), \tag{4.19}$$

with

$$\sigma^{(0,1)}_{nm}\stackrel{\mathrm{def}}{=}-\bar{K}_{nm}\ln\frac{\bar{K}_{nm}}{\bar{K}_{mn}}+\sum_{i,b}\varepsilon_{i,b}\Big[K^{i,b}_{nm}\ln\frac{\bar{K}_{nm}}{\bar{K}_{mn}}+K^{i,b}_{nm}-K^{i,b}_{mn}\frac{\bar{K}_{nm}}{\bar{K}_{mn}}\Big]. \tag{4.20}$$

$\sigma^{\mathrm{C}(2)}_{nm}(\alpha)$ and $\sigma^{(2)}_{nm}(\alpha)$ are quadratic orders of $\varepsilon_{i,b}$. While the former includes $\varepsilon_{i,b}\varepsilon_{i',b'}$ $(b\neq b')$ terms, the latter dose not. (4.16) leads

$$\sigma^{\mathrm{C}}_{\mathrm{ex}} = \sigma_{\mathrm{ex}}+\mathcal{O}(\varepsilon^2\delta). \tag{4.21}$$

Here, $\sigma^{\mathrm{C}}_{\mathrm{ex}}$ is given by (C.13). Then, (C.12), the result of Ref. [21], coincides with (3.22) when $p_n(t)=\langle n|\rho(t)|n\rangle$ is governed by the master equation (4.1).

5 Time-reversal operations

In this section, we define the time-reversal operation and examine the dependence of the excess entropy production on the time-reversal symmetry. We denote the time-reversal operator of the system by θ. We then define

$$\tilde{Y}\stackrel{\mathrm{def}}{=}\theta Y\theta^{-1}, \tag{5.1}$$

for all $Y\in\mathrm{B}$ and

$$\tilde{\mathcal{J}}\tilde{Y}\stackrel{\mathrm{def}}{=}\theta(\mathcal{J}Y)\theta^{-1}, \tag{5.2}$$

for a superoperator $\mathcal{J}$ of the system. The time-reversal of $\hat{K}(\alpha)\rho_0(\alpha)=0$ is given by

$$i[\tilde{H}_{\mathrm{L}}(\alpha),\tilde{\rho}_0(\alpha)]+\sum_b\tilde{\Pi}_b(\alpha)\tilde{\rho}_0(\alpha)=0, \tag{5.3}$$

using (2.74). If

$$\tilde{H}_{\rm L}(\alpha) = H_{\rm L}(\alpha), \quad \tilde{\Pi}_b(\alpha) = \Pi_b(\alpha), \tag{5.4}$$

hold, the above equation coincides with the equation of $\rho_0^{(-1)}(\alpha)$ since $[H_S, \rho_0^{(\kappa)}] = 0$, then

$$\tilde{\rho}_0(\alpha) = \rho_0^{(-1)}(\alpha) \stackrel{\rm def}{=} \breve{\rho}_0(\alpha), \tag{5.5}$$

holds. If the total Hamiltonian is time-reversal invariant, (5.4) holds [38]. If (5.4) holds and we neglect the Lamb shift Hamiltonian, the instantaneous steady state is time-reversal invariant: $\tilde{\rho}_0^{(0)} = \rho_0^{(0)}$.

For time-reversal symmetric system,

$$\frac{\partial}{\partial \alpha^n} S_{\rm sym}(\rho_0(\alpha)) = -{\rm Tr}_S\Big[\ln \tilde{\rho}_0(\alpha) \frac{\partial \rho_0(\alpha)}{\partial \alpha^n}\Big] + \mathcal{O}(\varepsilon^2), \tag{5.6}$$

holds. Here,

$$S_{\rm sym}(\rho) \stackrel{\rm def}{=} -{\rm Tr}_S\big[\rho \frac{1}{2}(\ln \rho + \ln \tilde{\rho})\big], \tag{5.7}$$

is the symmetrized von Neumann entropy. Combining (3.18) with (5.5), we obtain

$$A_n^\sigma(\alpha) = \frac{\partial}{\partial \alpha^n} S_{\rm sym}(\rho_0(\alpha)) + \mathcal{O}(\varepsilon^2), \tag{5.8}$$

then, the equation (3.22) with $S_{\rm vN} \to S_{\rm sym}$ holds. As analogy, we consider

$$S'(\alpha) \stackrel{\rm def}{=} -{\rm Tr}_S\big[\rho_0(\alpha) \frac{1}{2}(\ln \rho_0(\alpha) + \ln \breve{\rho}_0(\alpha))\big], \tag{5.9}$$

for generally non-time-reversal symmetric system. The difference between $\partial S'(\alpha)/\partial \alpha^n$ and the first term of the RHS of (3.18) is

$$\begin{aligned} &\frac{\partial S'(\alpha)}{\partial \alpha^n} - \Big(-{\rm Tr}_S\Big[\ln \breve{\rho}_0(\alpha) \frac{\partial \rho_0(\alpha)}{\partial \alpha^n}\Big]\Big) \\ =& -\frac{1}{2}{\rm Tr}_S\big[\frac{\partial \rho_0}{\partial \alpha^n}(\ln \rho_0 - \ln \breve{\rho}_0)\big] - \frac{1}{2}{\rm Tr}_S\big[\rho_0 \frac{\partial}{\partial \alpha^n} \ln \breve{\rho}_0\big]. \end{aligned} \tag{5.10}$$

To calculate the RHS of this equation, we use formulas

$$\ln(A + \eta B) = \ln A + \int_0^\infty ds\, \Big(\eta \frac{1}{A+s} B \frac{1}{A+s} - \eta^2 \frac{1}{A+s} B \frac{1}{A+s} B \frac{1}{A+s} + \mathcal{O}(\eta^3)\Big), \tag{5.11}$$

$$\frac{\partial}{\partial \alpha^n} \ln A(\alpha) = \int_0^\infty ds\, \frac{1}{A(\alpha)+s} \frac{\partial A(\alpha)}{\partial \alpha^n} \frac{1}{A(\alpha)+s}, \tag{5.12}$$

where $A, B, A(\alpha) \in {\rm B}$ and η is small real number. $\rho_0 - \breve{\rho}_0 = \varepsilon \psi + \mathcal{O}(\varepsilon^2)$ holds because $\overline{\rho_0^{(\kappa)}} = \rho_{\rm gc}(\alpha_S; \overline{\beta}, \overline{\beta\nu})$. Then, the first term of the RHS of (5.10) is given by

$$-\frac{1}{2}{\rm Tr}_S\big[\frac{\partial \rho_0}{\partial \alpha^n}(\ln \rho_0 - \ln \breve{\rho}_0)\big] = -\frac{\varepsilon}{2}\int_0^\infty ds\, {\rm Tr}_S\Big[\frac{\partial \rho_0}{\partial \alpha^n} \frac{1}{\breve{\rho}_0+s} \psi \frac{1}{\breve{\rho}_0+s}\Big] + \mathcal{O}(\varepsilon^2). \tag{5.13}$$

The second term of the RHS of (5.10) is given by

$$
\begin{aligned}
-\frac{1}{2}\mathrm{Tr}_S\big[\rho_0\frac{\partial}{\partial\alpha^n}\ln\breve{\rho}_0\big] &= -\frac{1}{2}\int_0^\infty ds\ \mathrm{Tr}_S\Big[\frac{\partial\breve{\rho}_0}{\partial\alpha^n}\frac{1}{\breve{\rho}_0+s}(\breve{\rho}_0+\varepsilon\psi)\frac{1}{\breve{\rho}_0+s}\Big]+\mathcal{O}(\varepsilon^2)\\
&= -\frac{1}{2}\mathrm{Tr}_S\big[\frac{\partial\breve{\rho}_0}{\partial\alpha^n}\big]\\
&\quad -\frac{\varepsilon}{2}\int_0^\infty ds\ \mathrm{Tr}_S\Big[\frac{\partial\breve{\rho}_0}{\partial\alpha^n}\frac{1}{\breve{\rho}_0+s}\psi\frac{1}{\breve{\rho}_0+s}\Big]+\mathcal{O}(\varepsilon^2)\\
&= -\frac{\varepsilon}{2}\int_0^\infty ds\ \mathrm{Tr}_S\Big[\frac{\partial\breve{\rho}_0}{\partial\alpha^n}\frac{1}{\rho_0+s}\psi\frac{1}{\rho_0+s}\Big]+\mathcal{O}(\varepsilon^2)\\
&= -\frac{\varepsilon}{2}\int_0^\infty ds\ \mathrm{Tr}_S\Big[\frac{\partial(\theta\breve{\rho}_0\theta^{-1})}{\partial\alpha^n}\frac{1}{\tilde{\rho}_0+s}\tilde{\psi}\frac{1}{\tilde{\rho}_0+s}\Big]+\mathcal{O}(\varepsilon^2). \qquad (5.14)
\end{aligned}
$$

Here, we used $\varepsilon(\breve{\rho}_0+s)^{-1}=\varepsilon(\rho_0+s)^{-1}+\mathcal{O}(\varepsilon^2)$ and $\mathrm{Tr}_S\bullet=\mathrm{Tr}_S\tilde{\bullet}$ if $\mathrm{Tr}_S\bullet$ is real. In general, the RHS of (5.10) is not $\mathcal{O}(\varepsilon^2)$. However, if $\tilde{\rho}_0=\breve{\rho}_0$ holds, the RHS of (5.10) becomes $\mathcal{O}(\varepsilon^2)$ since $\tilde{\psi}=-\psi$, then (5.6) holds. In the proof of (5.6), Yuge *et al.* [20] used incorrect equations $\frac{\partial}{\partial\alpha^n}\ln\tilde{\rho}_0=\tilde{\rho}_0^{-1}\frac{\partial\tilde{\rho}_0}{\partial\alpha^n}$ and $\ln\rho_0-\ln\tilde{\rho}_0=\varepsilon\psi\tilde{\rho}_0^{-1}+\mathcal{O}(\varepsilon^2)$.

6 Summary

In this paper, for open systems described by the quantum master equation (QME), we investigated the excess entropy production under quasistatic operations between nonequilibrium steady states (NESSs). We propose a new definition of the average entropy production rate $\dot{\sigma}(t)$ using the average energy and particle currents, which are calculated by using the full counting statistics (FCS) with QME (FCS-QME). Then, we introduced the generalized QMEs (GQMEs) providing $\dot{\sigma}(t)$. The GQMEs do not relate the higher moments (thus and the FCS) of the entropy production, but we can calculate only the average of the entropy production. Using the GQME, in weakly nonequilibrium regime, we analyzed the Berry-Sinitsyn-Nemenman (BSN) vector for the entropy production, $A_n^\sigma(\alpha)$, which provides the excess entropy production σ_{ex} under quasistatic operations between NESSs as the line integral of $A_n^\sigma(\alpha)$ in the parameter space. We have shown that the BSN vector $A_n^\sigma(\alpha)$ for the entropy production is given by $\rho_0(\alpha)$, the instantaneous steady state of the QME and $\breve{\rho}_0(\alpha)$, that of the QME which is given by reversing the sign of the Lamb shift term. If the system Hamiltonian is non-degenerate or the Lamb shift term is negligible, we obtain that the excess entropy production is given by the difference of the von Neumann entropies at the initial and final times of the operation. In general, the potential $\mathcal{S}(\alpha)$ such that $A_n^\sigma(\alpha)=\frac{\partial\mathcal{S}(\alpha)}{\partial\alpha^n}+\mathcal{O}(\varepsilon^2)$ dose not exist, but for time-reversal symmetric system, we showed that $\mathcal{S}(\alpha)$ is the symmetrized von Neumann entropy. Additionally, we pointed out that preceding expression of the entropy production in the classical Markov jump process [21, 37] is different from ours and showed that these approximately equivalent in the weakly nonequilibrium regime. We also checked that the definition of the average entropy production in the classical Markov jump process by Ref. [19] is equivalent to ours.

Acknowledgements

We acknowledge helpful discussions with S. Okada. Part of this work is supported by JSPS KAKENHI (26247051).

A Derivative of the von Neumann entropy

We show that

$$\frac{\partial S_{\mathrm{vN}}(\rho_0(\alpha))}{\partial \alpha^n} = -\mathrm{Tr}_S\Big[\ln\rho_0(\alpha)\frac{\partial\rho_0(\alpha)}{\partial\alpha^n}\Big]. \tag{A.1}$$

From the definition of the von Neumann entropy, the LHS of the above equation is given by

$$\frac{\partial S_{\mathrm{vN}}(\rho_0(\alpha))}{\partial \alpha^n} = -\mathrm{Tr}_S\Big[\ln\rho_0(\alpha)\frac{\partial\rho_0(\alpha)}{\partial\alpha^n}\Big] - \mathrm{Tr}_S\Big[\frac{\partial\ln\rho_0(\alpha)}{\partial\alpha^n}\rho_0(\alpha)\Big]. \tag{A.2}$$

Using (5.12), the second term of the RHS of the above equation becomes

$$\begin{aligned}
-\mathrm{Tr}_S\Big[\frac{\partial\ln\rho_0(\alpha)}{\partial\alpha^n}\rho_0(\alpha)\Big] &= -\mathrm{Tr}_S\Big[\int_0^\infty ds\ \frac{1}{\rho_0(\alpha)+s}\frac{\partial\rho_0(\alpha)}{\partial\alpha^n}\frac{1}{\rho_0(\alpha)+s}\rho_0(\alpha)\Big]\\
&= -\mathrm{Tr}_S\Big[\int_0^\infty ds\ \frac{\rho_0(\alpha)}{(\rho_0(\alpha)+s)^2}\frac{\partial\rho_0(\alpha)}{\partial\alpha^n}\Big]\\
&= -\mathrm{Tr}_S\Big[\frac{\partial\rho_0(\alpha)}{\partial\alpha^n}\Big] = 0.
\end{aligned} \tag{A.3}$$

Then, we obtain (A.1).

B Derivation of the relation between $k_{i,b}$ and $\rho^\kappa_{i,b}$

In this section, we examin the relation of the coefficients of the expansion of $\rho_0^\kappa(\alpha)$ and $l'_0(\alpha)$ in (3.14) of § 3.2.

First, we investigate $k_{i,b}$ in (3.14). (3.1) can be rewritten as

$$\hat{K}^\dagger(\alpha)l'_0(\alpha) + [\mathcal{K}'(\alpha)]^\dagger 1 = J^\sigma_{\mathrm{ss}}(\alpha). \tag{B.1}$$

Here, $J^\sigma_{\mathrm{ss}}(\alpha) = \mathcal{O}(\varepsilon^2)$ holds because $i^{H_b}_{\mathrm{ss}}(\alpha), i^{N_b}_{\mathrm{ss}}(\alpha) = \mathcal{O}(\varepsilon)$ and

$$J^\sigma_{\mathrm{ss}}(\alpha) = \sum_b \big(-i^{H_b}_{\mathrm{ss}}(\alpha)\varepsilon_{1,b} + i^{N_b}_{\mathrm{ss}}(\alpha)\varepsilon_{2,b}\big) \tag{B.2}$$

since

$$\sum_b i^{X_b}_{\mathrm{ss}}(\alpha) = -\mathrm{Tr}_S[X_S\sum_b \mathcal{L}_b(\alpha)\rho_0(\alpha)] = 0, \quad (X = N, H). \tag{B.3}$$

Then we obtain

$$\overline{\partial_{i,b}\mathcal{K}'}^\dagger 1 + \overline{K}^\dagger k_{i,b} + \overline{\partial_{i,b}\mathcal{L}_b}^\dagger \overline{l'_0} \ = \ 0, \tag{B.4}$$

in $\mathcal{O}(\varepsilon_{i,b})$. Here, $\partial_{i,b}X \overset{\mathrm{def}}{=} \partial X/\partial\alpha_{i,b}$ and $\overline{K} \overset{\mathrm{def}}{=} \overline{\hat{K}}$. The first term of the LHS is

$$\begin{aligned}
\overline{\partial_{i,b}\mathcal{K}'}^\dagger 1 &= \frac{\partial[\mathcal{K}']^\dagger 1}{\partial\alpha_{i,b}}\Big|_{\alpha_{i,b}=\overline{\alpha_i}}\\
&= \frac{\partial\mathcal{L}_b^\dagger[\alpha_{1,b}H_S - \alpha_{2,b}N_S]}{\partial\alpha_{i,b}}\Big|_{\alpha_{i,b}=\overline{\alpha_i}}\\
&= \overline{\partial_{i,b}\mathcal{L}_b}^\dagger[\overline{\beta}H_S - \overline{\beta\mu}N_S] + \overline{\Pi_b}^\dagger\frac{\partial[\alpha_{1,b}H_S - \alpha_{2,b}N_S]}{\partial\alpha_{i,b}}.
\end{aligned} \tag{B.5}$$

The third term of the LHS becomes

$$\begin{aligned}\overline{\partial_{i,b}\mathcal{L}_b}^\dagger\overline{l'_0} &= \overline{\partial_{i,b}\mathcal{L}_b}^\dagger(-\overline{\beta}H_S + \overline{\beta\mu}N_S + \overline{c'}^*1) \\ &= -\overline{\partial_{i,b}\mathcal{L}_b}^\dagger(\overline{\beta}H_S - \overline{\beta\mu}N_S). \end{aligned} \tag{B.6}$$

Here, we used $\overline{\partial_{i,b}\mathcal{L}_b}^\dagger 1 = 0$ derived from $\hat{K}^\dagger 1 = 0$. Then, (B.4) becomes

$$\overline{K}^\dagger k_{1,b} + \overline{\Pi_b}^\dagger H_S = 0, \tag{B.7}$$

$$\overline{K}^\dagger k_{2,b} - \overline{\Pi_b}^\dagger N_S = 0. \tag{B.8}$$

Next, we show the relation between $k_{i,b}$ and $\rho_{i,b}^{(-1)}$. (3.11) leads

$$\overline{K_\kappa}\rho_{i,b}^{(\kappa)} + \overline{\partial_{i,b}\mathcal{L}_b}\rho_{\text{gc}} = 0, \tag{B.9}$$

in $\mathcal{O}(\varepsilon_{i,b})$. Here, $\overline{K_\kappa} \overset{\text{def}}{=} \overline{\hat{K}_\kappa}$. By the way,

$$\mathcal{L}_b\rho_{\text{gc}}(\alpha_S;\beta_b,\beta_b\mu_b) = 0, \tag{B.10}$$

holds. Differentiating this equation by $\alpha_{i,b}$, we obtain

$$\overline{\partial_{i,b}\mathcal{L}_b}\rho_{\text{gc}} = -\overline{\mathcal{L}_b}\overline{\frac{\rho_{\text{gc}}(\alpha_S;\beta_b,\beta_b\mu_b)}{\partial\alpha_{i,b}}} = \overline{\mathcal{L}_b}\frac{\partial[\alpha_{1,b}H_S - \alpha_{2,b}N_S]}{\partial\alpha_{i,b}}\rho_{\text{gc}}(\alpha_S;\overline{\beta},\overline{\beta\mu}). \tag{B.11}$$

Substituting these equations into (B.9), we obtain

$$\overline{K}_\kappa\rho_{1,b}^{(\kappa)} + \overline{\Pi_b}(H_S\rho_{\text{gc}}) = 0, \tag{B.12}$$

$$\overline{K}_\kappa\rho_{2,b}^{(\kappa)} - \overline{\Pi_b}(N_S\rho_{\text{gc}}) = 0. \tag{B.13}$$

Now, we use

$$\overline{\Pi_b}(\bullet\rho_{\text{gc}}) = (\overline{\Pi_b}^\dagger\bullet)\rho_{\text{gc}}, \tag{B.14}$$

which is derived from KMS condition (2.71). Using this relation, we rewire (B.12) and (B.13) as

$$\overline{K}_\kappa\rho_{1,b}^{(\kappa)} + (\overline{\Pi_b}^\dagger H_S)\rho_{\text{gc}} = 0, \tag{B.15}$$

$$\overline{K}_\kappa\rho_{2,b}^{(\kappa)} - (\overline{\Pi_b}^\dagger N_S)\rho_{\text{gc}} = 0. \tag{B.16}$$

Multiplying ρ_{gc}^{-1} from the right, we obtain

$$(\overline{K}_\kappa\rho_{1,b}^{(\kappa)})\rho_{\text{gc}}^{-1} + \overline{\Pi_b}^\dagger H_S = 0, \tag{B.17}$$

$$(\overline{K}_\kappa\rho_{2,b}^{(\kappa)})\rho_{\text{gc}}^{-1} - \overline{\Pi_b}^\dagger N_S = 0. \tag{B.18}$$

(B.14) can be rewritten as

$$(\overline{\Pi_b}Y)\rho_{\text{gc}}^{-1} = \overline{\Pi_b}^\dagger(Y\rho_{\text{gc}}^{-1}), \tag{B.19}$$

for any $Y = \bullet\rho_{\rm gc} \in {\rm B}$ by multiplying $\rho_{\rm gc}^{-1}$ from the right. (B.19) leads

$$(\overline{\Pi}\rho_{i,b}^{(\kappa)})\rho_{\rm gc}^{-1} = \overline{\Pi}^{\dagger}(\rho_{i,b}^{(\kappa)}\rho_{\rm gc}^{-1}), \tag{B.20}$$

where $\overline{\Pi} \stackrel{\rm def}{=} \sum_b \overline{\Pi_b}$. By the way, $[H_S(\alpha_S), \rho_0^{(\kappa)}(\alpha)] = 0$ holds similarly to (2.74). Differentiating this equation by $\alpha_{i,b}$, we obtain

$$[H_S(\alpha_S), \rho_{i,b}^{(\kappa)}] = 0. \tag{B.21}$$

This relation leads

$$(\overline{H_\kappa^{\times}}\rho_{i,b}^{(\kappa)})\rho_{\rm gc}^{-1} = \overline{H_\kappa^{\times}}(\rho_{i,b}^{(\kappa)}\rho_{\rm gc}^{-1}) = \overline{H_{-\kappa}^{\times}}^{\dagger}(\rho_{i,b}^{(\kappa)}\rho_{\rm gc}^{-1}), \tag{B.22}$$

where $H_\kappa^{\times}\bullet \stackrel{\rm def}{=} -i[H_S(\alpha_S) + \kappa H_{\rm L}(\alpha), \bullet]$. We used $(H_\kappa^{\times})^{\dagger} = -H_\kappa^{\times}$. In the first equality, we used that $\rho_{\rm gc}$ commutes with H_S and $H_{\rm L}$. (B.20) and (B.22) lead

$$(\overline{K}_\kappa\rho_{i,b}^{(\kappa)})\rho_{\rm gc}^{-1} = \overline{K}_{-\kappa}^{\dagger}(\rho_{i,b}^{(\kappa)}\rho_{\rm gc}^{-1}). \tag{B.23}$$

Substituting this into (B.17) and (B.18), we obtain

$$\overline{K}_{-\kappa}^{\dagger}(\rho_{1,b}^{(\kappa)}\rho_{\rm gc}^{-1}) + \overline{\Pi_b}^{\dagger}H_S \quad = \quad 0, \tag{B.24}$$

$$\overline{K}_{-\kappa}^{\dagger}(\rho_{2,b}^{(\kappa)}\rho_{\rm gc}^{-1}) - \overline{\Pi_b}^{\dagger}N_S \quad = \quad 0. \tag{B.25}$$

Subtracting (B.24) ((B.25)) for $\kappa = -1$ from (B.7) ((B.8)), we obtain

$$\overline{K}^{\dagger}(k_{i,b} - \rho_{i,b}^{(-1)}\rho_{\rm gc}^{-1}) = 0. \tag{B.26}$$

This means

$$k_{i,b} = \rho_{i,b}^{(-1)}\rho_{\rm gc}^{-1} + \overline{c_{i,b}}1, \tag{B.27}$$

where $\overline{c_{i,b}}$ is an arbitrary complex number.

C Definition of entropy production of the Markov jump process

Except (C.9), this section is based on Ref. [21]. We consider the Markov jump process on the states $n = 1, 2, \cdots, \mathcal{N}$:

$$n(t) = n_k \quad (t_k \le t < t_{k+1}),\ t_0 = 0 < t_1 < t_2 \cdots < t_n < t_{N+1} = \tau. \tag{C.1}$$

where $N = 0, 1, 2, \cdots$ is the total number of jumps. We denote the above path by

$$\hat{n} = (N, (n_0, n_1, \cdots, n_N), (t_1, t_2, \cdots, t_N)). \tag{C.2}$$

The probability to find the system in a state n is $p_n(t)$ and it obeys the master equation (4.1). We suppose the trajectory of the control $\hat{\alpha} = \big(\alpha(t)\big)_{t=0}^{\tau}$ is smooth. Now we introduce

$$\theta_{nm}(\alpha) \stackrel{\rm def}{=} \begin{cases} -\ln\frac{K_{nm}(\alpha)}{K_{mn}(\alpha)} & K_{nm}(\alpha) \neq 0 \\ 0 & K_{nm}(\alpha) = 0 \end{cases}. \tag{C.3}$$

If $n \neq m$, this is entropy production of process $m \to n$. The entropy production of process (C.2) is defined by

$$\Theta^{\hat{\alpha}}[\hat{n}] = \sum_{k=1}^{N} \theta_{n_k n_{k-1}}(\alpha_{t_k}). \tag{C.4}$$

Then the weight (the transition probability density) associated with a path $\hat{n}$ is

$$\mathcal{T}^{\hat{\alpha}}[\hat{n}] = \prod_{k=1}^{N} K_{n_k n_{k-1}}(\alpha_{t_k}) \exp\Big[\sum_{k=0}^{N} \int_{t_k}^{t_{k+1}} dt\ K_{n_k n_k}(\alpha_t)\Big]. \tag{C.5}$$

The integral over all the paths is defined by

$$\int \mathcal{D}\hat{n}\ Y[\hat{n}] \stackrel{\text{def}}{=} \sum_{N=0}^{\infty} \sum_{n_0, n_1, \cdots, n_N}^{n_{k-1} \neq n_k} \int_0^{\tau} dt_1 \int_{t_1}^{\tau} dt_2 \int_{t_3}^{\tau} dt_3 \cdots \int_{t_{N-1}}^{\tau} dt_N\ Y[\hat{n}], \tag{C.6}$$

and the expectation value of $X[\hat{n}]$ is defined by

$$\langle X \rangle^{\hat{\alpha}} \stackrel{\text{def}}{=} \int \mathcal{D}\hat{n}\ X[\hat{n}] p^{\text{ss}}_{n_0}(\alpha_0) \mathcal{T}^{\hat{\alpha}}[\hat{n}]. \tag{C.7}$$

Here, $p^{\text{ss}}_n(\alpha)$ is the instantaneous stationary probability distribution characterized by $\sum_m K_{nm}(\alpha) p^{\text{ss}}_m(\alpha) = 0$. We introduce a matrix $K^{\lambda}(\alpha)$ by

$$[K^{\lambda}(\alpha)]_{nm} \stackrel{\text{def}}{=} K_{nm}(\alpha) e^{i\lambda\theta_{nm}(\alpha)}. \tag{C.8}$$

Then, the k-th order moment of the entropy production is given by

$$\langle (\Theta^{\hat{\alpha}}[\hat{n}])^k \rangle^{\hat{\alpha}} = \frac{\partial^k}{\partial (i\lambda)^k}\Big|_{\lambda=0} \sum_{n,m} \Big[\mathrm{T} \exp\Big[\int_0^{\tau} dt\ K^{\lambda}(\alpha_t)\Big]\Big]_{nm} p^{\text{ss}}_m(\alpha_0). \tag{C.9}$$

In particular, the average is given by

$$\sigma^{\text{C}} \stackrel{\text{def}}{=} \langle \Theta^{\hat{\alpha}}[\hat{n}] \rangle^{\hat{\alpha}} = \int_0^{\tau} dt\ \sum_{n,m} \sigma^{\text{C}}_{nm}(\alpha_t) p_m(t), \tag{C.10}$$

where

$$\sigma^{\text{C}}_{nm}(\alpha) \stackrel{\text{def}}{=} K_{nm}(\alpha)\theta_{nm}(\alpha) = -K_{nm}(\alpha) \ln \frac{K_{nm}(\alpha)}{K_{mn}(\alpha)}. \tag{C.11}$$

According to Ref. [21], for a quasi-static operation,

$$\sigma^{\text{C}}_{\text{ex}} = S_{\text{Sh}}[p^{\text{ss}}(\alpha_{\tau})] - S_{\text{Sh}}[p^{\text{ss}}(\alpha_0)] + \mathcal{O}(\varepsilon^2\delta), \tag{C.12}$$

holds where

$$\sigma^{\text{C}}_{\text{ex}} \stackrel{\text{def}}{=} \sigma^{\text{C}} - \int_0^{\tau} dt\ \sum_{n,m} \sigma^{\text{C}}_{nm}(\alpha_t) p^{\text{ss}}_m(\alpha_t), \tag{C.13}$$

and $S_{\text{Sh}}[p] \stackrel{\text{def}}{=} -\sum_n p_n \ln p_n$.

References

[1] Landauer, R.: $dQ = TdS$ far from equilibrium. Phys. Rev. A **18**, 255 (1978).

[2] Oono, Y., Paniconi, M.: Steady State Thermodynamics. Prog. Theor. Phys. Suppl. **130**, 29-44 (1998).

[3] Sasa, S., Tasaki, H.: Steady State Thermodynamics. J. Stat. Phys. **125**, 125-227 (2006).

[4] Speck, T., Seifert, U.: Integral fluctuation theorem for the housekeeping heat. J. Phys. A: Math. Gen. **38**, L581-L588 (2005).

[5] Esposito, M., Harbola, U., Mukamel, S.: Entropy fluctuation theorems in driven open systems: Application to electron counting statistics. Phys. Rev. E **76**, 031132 (2007).

[6] Esposito, M., Van den Broeck, C.: Three Detailed Fluctuation Theorems Phys.Rev. Lett. **104**, 090601(2010).

[7] Pradhan, P., Ramsperger, R., Seifert, U.: Approximate thermodynamic structure for driven lattice gases in contact. Phys. Rev. E **84**, 041104 (2011).

[8] Crooks, G. E.: Entropy production fluctuation theorem and the nonequilibrium work relation for free energy differences. Phys. Rev. E **60**, 2721 (1999).

[9] Seifert, U.: Entropy Production along a Stochastic Trajectory and an Integral Fluctuation Theorem. Phys. Rev. Lett. **95**, 040602 (2005).

[10] Komatsu, T. S., Nakagawa, N., Sasa, S. Tasaki, H.: Steady-State Thermodynamics for Heat Conduction: Microscopic Derivation. Phys. Rev. Lett. **100**, 230602 (2008).

[11] Komatsu, T. S., Nakagawa, N., Sasa, S., Tasaki, H.: Entropy and Nonlinear Nonequilibrium Thermodynamic Relation for Heat Conducting Steady States. J. Stat. Phys. **142**, 127-153 (2011).

[12] Saito, K., Tasaki, H.: Extended Clausius Relation and Entropy for Nonequilibrium Steady States in Heat Conducting Quantum Systems. J. Stat. Phys. **145**, 1275-1290 (2011).

[13] Ruelle, D. P.: Extending the definition of entropy to nonequilibrium steady states. Proc. Natl. Acad. Sci. USA **100**, 3054-3058 (2003).

[14] Bertini, L., Gabrielli, D., Jona-Lasinio, G., Landim, C.: Thermodynamic Transformations of Nonequilibrium States. J. Stat. Phys. **149**, 773-802 (2012).

[15] L. Bertini, D. Gabrielli, G. Jona-Lasinio and C. Landim, Phys. Rev. Lett. **110**, 020601 (2013).

[16] Hatano, T., Sasa, S.: Steady-State Thermodynamics of Langevin Systems. Phys. Rev. Lett. **86**, 3463 (2001).

[17] Sasa, S.: Possible extended forms of thermodynamic entropy. J. Stat. Mech. P01004 (2014).

[18] Maes, C., Netočný, K.: A Nonequilibrium Extension of the Clausius Heat Theorem. J. Stat. Phys. **154**, 188-203 (2014).

[19] Sagawa, T., Hayakawa, H.: Geometrical expression of excess entropy production. Phys. Rev. E **84**, 051110 (2011).

[20] Yuge, T., Sagawa, T., Sugita, A., Hayakawa, H.: Geometrical Excess Entropy Production in Nonequilibrium Quantum Systems. J. Stat. Phys. **153**, 412-441 (2013).

[21] Komatsu, T. S., Nakagawa, N., Sasa, S., Tasaki, H.: Exact Equalities and Thermodynamic Relations for Nonequilibrium Steady States. J. Stat. Phys. **159**, 1237-1285 (2015).

[22] Sinitsyn, N. A., Nemenman, I.: The Berry phase and the pump flux in stochastic chemical kinetics. Europhys. Lett. **77**, 58001 (2007).

[23] Yuge, T., Sagawa, T., Sugita, A., Hayakawa, H.: Geometrical pumping in quantum transport: Quantum master equation approach. Phys. Rev. B **86**, 235308 (2012).

[24] Watanabe, K., Hayakawa, H.: Non-adiabatic effect in quantum pumping for a spin-boson system. Prog. Theor. Exp. Phys. 2014, 113A01 (2014).

[25] Nakajima, S., Taguchi, M., Kubo, T., Tokura, Y.: Interaction effect on adiabatic pump of charge and spin in quantum dot. Phys. Rev. B **92**, 195420 (2015).

[26] Esposito, M., Harbola, U., Mukamel, S.: Nonequilibrium fluctuations, fluctuation theorems, and counting statistics in quantum systems. Rev. Mod. Phys. **81**, 1665-1702 (2009).

[27] Berry, M. V.: Quantal phase factors accompanying adiabatic change. Proc. R Soc. A **392**, 45-57 (1984).

[28] Breuer, H. P., Petruccione, F.: *The Theory of Open Quantum Systems*. (Oxford University Press, Oxford, 2002).

[29] Schaller, G., Brandes, T.: Preservation of positivity by dynamical coarse graining. Phys. Rev. A **78**, 022106 (2008).

[30] Majenz, C., Albash, T., Breuer, H. P., Lidar, D. A.: Coarse graining can beat the rotating-wave approximation in quantum Markovian master equations. Phys. Rev. A **88**, 012103 (2013).

[31] Brouwer, P. W.: Scattering approach to parametric pumping. Phys. Rev. B **58**, 10135(R) (1998).

[32] Splettstoesser, J., Governale, M., König, J., Fazio, R.: Adiabatic Pumping through Interacting Quantum Dots. Phys. Rev. Lett. **95**, 246803 (2005).

[33] Pluecker, T., Wegewijs, M. R., Splettstoesser, J.: Gauge freedom in observables and Landsberg's nonadiabatic geometric phase: Pumping spectroscopy of interacting open quantum systems. Phys. Rev. B **95**, 155431 (2017)

[34] Cavaliere, F., Governale, M., König, J.: Nonadiabatic Pumping through Interacting Quantum Dots. Phys. Rev. L **103**, 136801 (2009).

[35] Calvo, H. L., Classen, L., Splettstoesser, J., Wegewijs, M. R.: Interaction-induced charge and spin pumping through a quantum dot at finite bias. Phys. Rev. B **86**, 245308 (2012).

[36] Utsumi, Y., Entin-Wohlman, O., Aharony, A., Kubo, T., Tokura, Y.: Fluctuation theorem for heat transport probed by a thermal probe electrode. Phys. Rev. B **89**, 205314 (2014).

[37] Mandal, D., Jarzynski, C.: Analysis of slow transitions between nonequilibrium steady states. J. Stat. Mech. 063204 (2016).

[38] Gorini, V., Frigerio, A., Verri, M., Kossakowski, A., Sudarshan, E. C. G.: Properties of quantum Markovian master equations. Rep. Math. Phys. **13**, 149 (1978).