Abstract

For open systems described by the quantum Markovian master equation, we study a possible extension of the Clausius equality to quasistatic operations between nonequilibrium steady states (NESSs). We investigate the excess heat divided by temperature (i.e., excess entropy production) which is transferred into the system during the operations. We derive a geometrical expression for the excess entropy production, which is analogous to the Berry phase in unitary evolution. Our result implies that any scalar thermodynamic potential cannot be defined in terms of the excess heat for NESSs far from equilibrium, and that a vector potential plays a crucial role in the thermodynamics for NESSs. In the weakly nonequilibrium regime, we show that the geometrical expression reduces to the extended Clausius equality derived by Saito and Tasaki (J. Stat. Phys. 145, 1275 (2011)). As an example, we investigate a spinless electron system in quantum dots. We find that there exists a scalar potential for the operation on a single reservoir in noninteracting systems, but that this is not valid in interacting systems.

1 Introduction

Thermodynamics and statistical mechanics are universal and powerful frameworks to describe systems in equilibrium states. 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 the equilibrium states. Its operational definition relies on the Clausius equality:

$$\Delta S = \beta Q.$$  \hspace{1cm} (1.1)

This equality is valid for quasistatic operations between two equilibrium states. Here, $\Delta S$ is the change in the entropy of the system before and after the operation, $\beta$ is the inverse temperature of the reservoir that is in contact with the system, and $Q$ is the heat transferred from the reservoir to the system during the operation. Equilibrium statistical mechanics tells that the entropy $S$ is given by the Shannon entropy of the probability distribution (von Neumann entropy of the density matrix) of microscopic states in the equilibrium classical (quantum) system. This connects the microscopic physics to the macroscopic one, where we do not need to solve the equation of motion in the microscopic level.

The construction of analogous frameworks of thermodynamics and statistical mechanics for nonequilibrium systems has been one of the central subjects in statistical physics \cite{1, 9, 19, 21, 22, 26, 28, 34}. Recently there has been progress in the extension of the Clausius equality to nonequilibrium steady states (NESSs) \cite{16, 17, 23, 25} (see also Refs. \cite{5, 6, 10, 13, 20, 30}). In these studies, the excess heat $Q^{ex}$, proposed in Ref. \cite{21}, has been used instead of the total heat $Q$ in the equilibrium equality (1.1). The excess heat $Q^{ex}$ is defined by subtracting from $Q$ the contribution $Q_{hk}$ (called housekeeping heat)}
of steady heat dissipation in NESS. Then it has been shown that in the weakly nonequilibrium regime there exists a scalar potential $S_{\text{sym}}$ which satisfies the extended Clausius equality,

$$\Delta S_{\text{sym}} = \beta Q^\text{ex},$$

for quasistatic operations. We refer to $\beta Q^\text{ex}$ as excess entropy production during the operation. Moreover, $S_{\text{sym}}$ in Eq. (1.2) is given by a symmetrized version of the Shannon (von Neumann) entropy of the NESS.

More recently, a formula for the excess entropy production during quasistatic operations in the strongly nonequilibrium regime is derived in generic classical systems described by the Markov jump process [24]. This formula is expressed by a geometrical (Berry-phase-like) quantity [3, 27]; i.e., the excess entropy production is given by a line integral of a vector potential in the operation parameter space. Therefore, the excess entropy production depends on the whole of the operation path in the parameter space. This implies that, in general, the extended Clausius equality does not hold in the strongly nonequilibrium regime. Therefore the operational definition of the nonequilibrium entropy (scalar thermodynamic potential) through the excess entropy production is impossible, but the vector potential plays an important role in the thermodynamics for NESSs.

Since the extended Clausius equality holds for both the classical systems [16, 17] and quantum systems [25], it is expected that the geometrical expression for the excess entropy production in the strongly nonequilibrium regime [24] can also be generalized to quantum systems. In this paper we show that this expectation is true; we derive a quantum version of the geometrical expression in open systems described by the quantum Markovian master equation (QMME). This result suggests the universality of the geometrical expression and the importance of the vector potential in the thermodynamics for NESSs. It should be noted that in the field of adiabatic pumping the path-dependent physical variables play important roles thanks to the existence of the geometrical phase, where there exists a current between reservoirs without dc bias [8, 18, 27, 29, 31, 33]. We believe that the concept of path-dependent quantities is important in general nonequilibrium situations.

This paper is organized as follows. In Sec. 2.1 we explain the generic setup of the system which we consider in this paper. We give the QMME description of the system of interest in the setup. In Sec. 2.2 we define the entropy production during an operation. We use the technique of the full counting statistics incorporated into the QMME [11] to calculate the cumulant generating function and average of the entropy production. In Sec. 2.3 we derive an explicit form of the QMME by using the eigenoperators. We also discuss the rotating wave approximation (or secular approximation) in the QMME. We give the main result in Sec. 3.1 we derive the geometrical expression for the excess entropy production for an arbitrary quasistatic operation in the QMME system. We also show that the results within and without the rotating wave approximation are equivalent. In Secs. 3.2 and 3.3 we show that the geometrical expression reduces to the equilibrium and extended Clausius equalities (1.1) and (1.2), in the equilibrium states and in the weakly nonequilibrium regime, respectively. In Sec. 4 we investigate a spinless electron system in quantum dots as a simple example. In Sec. 5 we give a summary with a discussion.

## 2 Setup

### 2.1 Quantum Markovian Master Equation

We consider a quantum system $S$ in contact with reservoirs $R_b$ ($b = 1, 2, \ldots$). The system $S$ and each reservoir can exchange particles and energy. We assume that the dimension of the Hilbert space $\mathcal{H}_S$ associated with $S$ is finite. We also assume that the reservoirs are large enough compared to the system
S. The total system $S + \{R_b\}_b$ is closed except for external operations. Then the total system evolves according to the Liouville-von Neumann equation:

$$\frac{\partial \hat{\rho}_{\text{tot}}(t)}{\partial t} = \frac{1}{i\hbar}[\hat{H}_{\text{tot}}, \hat{\rho}_{\text{tot}}(t)], \quad (2.1)$$

where $\hat{\rho}_{\text{tot}}$ and $\hat{H}_{\text{tot}}$ denote respectively the density matrix and Hamiltonian of the total system. $\hat{H}_{\text{tot}}$ is written as

$$\hat{H}_{\text{tot}} = \hat{H}_S(\alpha_S) + \sum_b [\hat{H}_b + u\hat{H}_{\text{SB}}], \quad (2.2)$$

where $\hat{H}_S$ is the system Hamiltonian, $\alpha_S$ is the set of operation parameters in the system $S$, $\hat{H}_b$ is the Hamiltonian of the $b$th reservoir $R_b$, and $\hat{H}_{SB}$ is the coupling Hamiltonian between $S$ and $R_b$. We assume that $[\hat{H}_S, \hat{N}_S] = [\hat{H}_b, \hat{N}_b] = 0$ holds with $\hat{N}_S$ and $\hat{N}_b$ being the particle number operators of $S$ and $R_b$.

We denote the eigenvalue of $\hat{H}_S$ by $E_{\nu}$ and the corresponding eigenstate by $|E_{\nu}, n\rangle$, where $n$ is the index for distinguishing the degeneracy. We also assume that the coupling between the system and reservoirs is weak. To keep in mind this weak coupling assumption, we introduced the parameter $u$ in Eq. (2.2).

We set the initial states of the reservoirs to be equilibrium states with different temperatures and chemical potentials. Precisely, the initial state of the reservoirs is $\hat{\rho}_R = \bigotimes_b \hat{\rho}_b(\alpha_b)$, where $\hat{\rho}_b(\alpha_b) := e^{-\beta_b(\hat{H}_b - \mu_b \hat{N}_b)}/Z_b(\alpha_b)$ is the grand canonical state of the $b$th reservoir with the inverse temperature $\beta_b$ and chemical potential $\mu_b$. Here, $\alpha_b$ denotes the set of the $b$th reservoir parameters $(\beta_b, \mu_b)$, $Z_b(\alpha_b) := \text{Tr}_b e^{-\beta_b(\hat{H}_b - \mu_b \hat{N}_b)}$ is the grand partition function, and $\text{Tr}_b$ is the trace over the degrees of freedom of the $b$th reservoir. The initial state $\hat{\rho}(0)$ of the system $S$ is arbitrary, so that the initial state of the total system is the uncorrelated state $\hat{\rho}_{\text{tot}}(0) = \hat{\rho}(0) \otimes \hat{\rho}_R$. Because the reservoirs are much larger than the system $S$, we expect that there exists a certain long time range $\tau_\text{long}$ in which the state of the system $S$ can change considerably whereas the reservoirs approximately remain in their initial equilibrium states. We also expect that, in a more restricted but still long time range, the system settles down in a NESS which is uniquely determined by the reservoir parameters $\alpha_B := \{\alpha_b\}_b$ and system parameters $\alpha_S$.

We write the set of the control parameters $(\alpha_S, \alpha_B)$ as $\alpha$. An arbitrary external operation on the system $S$ is represented by a modulation of $\alpha$. Thus $\alpha$ may depend on time. Theoretically, we can treat the time-dependent $\alpha_B$ by refreshing the reservoirs to other values of $\alpha_B$ within the time range $\tau_\text{long}$.

To investigate the dynamics of the system $S$ in the above situation, we employ the quantum Markovian master equation (QMME) approach, where the dynamics is described by an equation of motion for the reduced density matrix $\hat{\rho} = \text{Tr}_R \hat{\rho}_{\text{tot}}$ of $S$, where $\text{Tr}_R$ is the trace over the reservoirs. In this approach we make the following assumption in addition to the weak coupling: the correlation time of the reservoirs is much shorter than the time scale of the system evolution. Starting from the Liouville-von Neumann equation (2.1), and after tracing out the reservoirs’ degrees of freedom, we perform the Born and Markov approximations on the basis of the above assumptions. For fixed $\alpha$, the result is written in the Schrödinger picture as [7,12]

$$\frac{\partial \hat{\rho}(t)}{\partial t} = \frac{1}{i\hbar}[\hat{H}_S(\alpha_S), \hat{\rho}(t)] - \frac{v}{\hbar^2} \sum_b \int_0^\infty dt' \text{Tr}_b \left[ \hat{H}_{SB}, \left[ \hat{H}_{SB}(-t'), \hat{\rho}(t) \otimes \hat{\rho}_b(\alpha_b) \right] \right], \quad (2.3)$$

with $v := u^2$. In this paper we refer to this equation as QMME. Here, the symbol $\cdot \cdot \cdot$ stands for the Heisenberg picture in terms of $\hat{H}_S + \sum_b \hat{H}_b$, i.e., $\hat{O}(t) := \hat{U}(t)\hat{O}\hat{U}(t)$, where $\hat{U}(t) = \exp\{-[\hat{H}_S(\alpha_S) + \sum_b \hat{H}_b]/\hbar\}$. We note that the Born approximation is in the second order with respect to the system-reservoir coupling, which is represented by $v = u^2$ in the second term of Eq. (2.3).
We denote by \( B \) the set of all the linear operators on \( \mathcal{H}_S \). Because the dimension of \( \mathcal{H}_S \) is finite, any \( \hat{Y} \) in \( B \) is a trace class operator. We can define the Hilbert-Schmidt inner product in \( B \) as \( \text{Tr}_S(\hat{Y}_1 \hat{Y}_2^\dagger) \) for any \( \hat{Y}_1, \hat{Y}_2 \in B \), where \( \text{Tr}_S \) is the trace in \( \mathcal{H}_S \). With this inner product, \( B \) is a separable Hilbert space. We refer to the linear operators on \( B \) as superoperators to distinguish with the operators on \( \mathcal{H}_S \). We define the adjoint \( O^\dagger \) of a superoperator \( O \) such that \( \text{Tr}_S([O^\dagger \hat{Y}_1] \hat{Y}_2) = \text{Tr}_S(\hat{Y}_1^\dagger O \hat{Y}_2) \) holds for any \( \hat{Y}_1, \hat{Y}_2 \in B \).

From the right-hand side (RHS) of the QMME (2.3), we can define the generator \( K \) of the QMME as \( \mathcal{K}\hat{Y} := [\text{RHS of Eq. (2.3)} \text{ with } \dot{\rho}(t) \rightarrow \hat{Y}] \) for any \( \hat{Y} \in B \). Since \( K \) depends on the control parameters \( \alpha \), we sometimes write them in the argument of the generator as \( \mathcal{K}(\alpha) \).

The right and left eigenvalue equations for \( K \) with fixed \( \alpha \) are respectively given by

\[
\mathcal{K}(\alpha) \hat{r}_m(\alpha) = \lambda_m(\alpha) \hat{r}_m(\alpha),
\]

(2.4) where the complex number \( \lambda_m(\alpha) \) is the eigenvalue labeled by \( m \) (we denote the complex conjugate of a complex number \( c \) by \( c^\ast \)), and \( \hat{r}_m(\alpha) \) and \( \hat{\ell}_m(\alpha) \in B \) are respectively the corresponding right and left eigenvectors. In the following, we assume that \( \mathcal{K}(\alpha) \) has the zero eigenvalue \( \lambda_0 = 0 \) (labeled by \( m = 0 \)) without degeneracy, so that \( \mathcal{K}(\alpha) \hat{r}_0(\alpha) = 0 \) and \( \mathcal{K}^\dagger(\alpha) \hat{\ell}_0(\alpha) = 0 \) hold. This assumption implies that the QMME has a unique steady solution \( \dot{\rho}_{\text{ss}}(\alpha) = \hat{r}_0(\alpha) \) for fixed \( \alpha \). It should be noted, however, that the uniqueness of the steady solution of the QMME is not trivial especially in the case where \( \hat{H}_S \) has degenerate eigenenergies. We note that \( \mathcal{K}(\alpha) \hat{r}_0(\alpha) = \hat{r}_0(\alpha) \) holds for any \( \alpha \) because of the trace-preserving property of the QMME.

When we modulate \( \alpha \) in time to perform an external operation onto the system \( S \), we can use the QMME with time-dependent \( \alpha \) for investigating the dynamics of \( S \): \( \partial \rho(t)/\partial t = \mathcal{K}(\alpha(t)) \rho(t) \). This is valid for the operations whose time scale is sufficiently slower than the correlation time of the reservoirs. This is a kind of Markov approximation other than the one used in deriving the QMME (2.3). There are four characteristic time scales in the present setup: the time scale \( \tau_\text{S} \) of the intrinsic evolution of the system \( S \), the relaxation time \( \tau_{\text{Rlx}} \) of \( S \) as an open system in contact with the reservoirs, the correlation time \( \tau_\text{R} \) of the reservoirs, and the time scale \( \tau_{\text{ctrl}} \) of the operation of changing the control parameters \( \alpha \). For the Markov approximation used here, \( \tau_\text{R} \ll \tau_{\text{ctrl}} \) is required, whereas \( \tau_\text{Rlx} \ll \tau_{\text{Rlx}} \) is required for the Markov approximation in deriving Eq. (2.3). For the rotating wave approximation (or secular approximation), which will be explained in Sec. 2.3, \( \tau_\text{S} \ll \tau_{\text{Rlx}} \) is required. For quasistatic operations, required is \( \tau_{\text{Rlx}} \ll \tau_{\text{ctrl}} \), which ensures the validity of an adiabatic approximation used in the next section.

We here introduce the following projection superoperator \( \mathcal{P} \):

\[
\mathcal{P}|E_\kappa,k\rangle\langle E_\nu,n| = \begin{cases} |E_\kappa,k\rangle\langle E_\nu,n| & \text{(if } E_\kappa = E_\nu) \\ 0 & \text{(if } E_\kappa \neq E_\nu). \end{cases}
\]

(2.6)

In the matrix representation of any operator \( \hat{Y} \in B \) in the basis of the eigenstates of \( \hat{H}_S \), \( \mathcal{P} \) leaves unchanged only the matrix elements constructed from the eigenstates with the same energy eigenvalues. By using \( \mathcal{P} \), we define a subspace \( \mathcal{P} \) of \( B \) as \( \mathcal{P} := \{ \hat{Y} \in B \mid \mathcal{P}\hat{Y} = \hat{Y} \} \). We denote the orthogonal complement of \( \mathcal{P} \) by \( \mathcal{Q} \) and the projection superoperator onto \( \mathcal{Q} \) by \( \mathcal{Q} \).

We also define the time-reversal operation. We denote the time-reversal operator on \( \mathcal{H}_S \) by \( \theta \). In this paper, we assume that the system Hamiltonian is time-reversal invariant: \( \theta\hat{H}_S\theta^{-1} = \hat{H}_S \). We also define the tilde superoperation on \( B \) by

\[
\tilde{\hat{Y}} := \theta\hat{Y}^\dagger\theta^{-1},
\]

(2.7)
for any \( \hat{Y} \in B \). We note that \( \hat{Y} = \hat{\theta}^{(-1)} \hat{Y} \hat{\theta}^{-1} \) if \( \hat{Y} \) is self-adjoint. Therefore the time reversal of a state \( \hat{\rho} \) is given by \( \hat{\theta} \hat{\rho} \hat{\theta}^{-1} = \hat{\rho} \). Using the superoperation (2.7), we define the tilde \( \hat{O} \) of a superoperator \( O \) by

\[
\hat{O} \hat{Y} := \hat{O} \hat{\theta}^{(-1)} \hat{Y} \hat{\theta}^{-1} = \hat{O} \hat{Y} = \hat{O} \hat{Y},
\]

for any \( \hat{Y} \in B \).

### 2.2 Full Counting Statistics of Entropy Production

We next introduce the entropy production \( \sigma \) generated during an external operation with a time interval \( \tau \) as follows. At the initial time \( t = 0 \), we perform a projection measurement of reservoir observables \( \{ \hat{A}_b(0) := \beta_b(0)(\hat{H}_b - \mu_b(0)\hat{N}_b) \}_b \) to obtain measurement outcomes \( \{ a_b(0) \}_b \). Because we assume \( [\hat{H}_b, \hat{N}_b] = 0 \), the simultaneous projection measurement of \( \hat{H}_b \) and \( \hat{N}_b \) is possible. Note that we can construct the outcome \( a_b(0) \) from the measurement outcomes of \( \hat{H}_b \) and \( \hat{N}_b \) because \( \beta_b(0) \) and \( \mu_b(0) \) are the initial (known) values of the control parameters. For \( t > 0 \), we make an external operation by changing the control parameters \( \alpha \). During the operation the system evolves with interacting with the reservoirs. At \( t = \tau \), we again perform a projection measurement of \( \{ \hat{A}_b(\tau) := \beta_b(\tau)(\hat{H}_b - \mu_b(\tau)\hat{N}_b) \}_b \) to obtain measurement outcomes \( \{ a_b(\tau) \}_b \). Since the reservoirs are assumed to remain in the equilibrium states during the operation, the difference of the outcomes gives the energy change \( \sum_b \Delta E_b \) minus work \( \sum_b \mu_b \Delta N_b \) associated with the particle exchange (divided by the temperature). Therefore we can regard the difference of the outcomes as the heat (divided by the temperature) that is transferred from the reservoirs into the system S. We thus define the entropy production during the operation as

\[
\sigma := \sum_b [a_b(0) - a_b(\tau)].
\]  

By repeating this measurement scheme many times, we obtain a probability distribution \( p_\tau(\sigma) \) of \( \sigma \). We are interested in the average of \( \sigma \), which is defined as \( \langle \sigma \rangle_\tau := \int d\sigma p_\tau(\sigma) \). Note that, for large \( \tau \) with \( \alpha \) being fixed, \( \langle \sigma \rangle_\tau / \tau \) approaches a steady value \( J_\sigma(\alpha) \) of the entropy flux in the NESS. In this paper, instead of directly calculating the average, we investigate it from the cumulant generating function \( G_\tau(\chi) \), which is given by

\[
G_\tau(\chi) := \ln \int d\sigma p_\tau(\sigma) e^{i\chi \sigma}.
\]  

Here \( \chi \) is the counting field. The derivatives of \( G_\tau(\chi) \) give the cumulants; in particular, \( \langle \sigma \rangle_\tau = \partial G_\tau(\chi)/\partial(i\chi)|_{\chi=0} \).

To calculate \( G_\tau(\chi) \), we use a technique of the full counting statistics \[11\]. This technique provides us with the formula for the cumulant generating function: \( G_\tau(\chi) = \ln \text{Tr} \hat{\rho}_\text{tot}^\chi(\tau) \). Here, \( \text{Tr} \) is the trace over the total system and \( \hat{\rho}_\text{tot}^\chi \) is the solution of the generalized Liouville-von Neumann equation:

\[
\frac{\partial \hat{\rho}_\text{tot}^\chi(t)}{\partial t} = \frac{i}{\hbar} [\hat{H}_\text{tot}^\chi \hat{\rho}_\text{tot}^\chi(t) - \hat{\rho}_\text{tot}^\chi(t) \hat{H}_\text{tot}^\chi].
\]  

Here, the \( \chi \)-modified Hamiltonian \( \hat{H}_\text{tot}^\chi \) is given by

\[
\hat{H}_\text{tot}^\chi := e^{-i\chi \hat{A}^2/2} \hat{H}_\text{tot} e^{i\chi \hat{A}^2/2},
\]  

where \( \hat{A} := \sum_b \hat{A}_b \) with \( \hat{A}_b := \beta_b(\hat{H}_b - \mu_b\hat{N}_b) \).
In the QMME approach, starting from the generalized Liouville-von Neumann equation (2.11), and taking the same procedure as in the previous subsection, we obtain the generalized quantum Markovian master equation (GQMME) for the reduced ($\chi$-modified) density matrix $\hat{\rho}^\chi = \text{Tr}_R \hat{\rho}_{\text{tot}}^\chi$ as

$$\frac{\partial \hat{\rho}^\chi(t)}{\partial t} = -\frac{i}{\hbar} \{ \hat{H}_S(\alpha_S), \hat{\rho}^\chi(t) \} - \frac{C}{\hbar^2} \sum_b \int_0^\infty dt' \text{Tr}_b \left[ \hat{H}_{SB}(t'), \hat{\rho}^\chi(t) \otimes \hat{\rho}_b(\alpha_b) \right] \chi.$$  

(2.13)

Here, $[\hat{O}_1, \hat{O}_2]_\chi := \hat{O}_1^\chi \hat{O}_2 - \hat{O}_2 \hat{O}_1^{-\chi}$ and $\hat{O}^\chi := e^{-i\chi\hat{A}/2} \hat{O} e^{i\chi\hat{A}/2}$. Thanks to the above-mentioned formula, we can calculate the generating function from the solution of the GQMME as

$$G_\tau(\chi) = \ln \text{Tr}_S \hat{\rho}^\chi(\tau).$$  

(2.14)

Similarly to the case of the QMME, we can define the generator $\mathcal{K}^\chi$ of the GQMME as $\mathcal{K}^\chi(\alpha)\hat{Y} := [\text{RHS of Eq. (2.13)}$ with $\hat{\rho}^\chi(t) \to \hat{Y}]$ for any $\hat{Y} \in \mathbb{B}$. For fixed $\alpha$, we can also define the right and left eigenvectors of the GQMME generator $\mathcal{K}^\chi(\alpha)$ corresponding to the eigenvalue $\lambda^\chi_{m}(\alpha)$, which are respectively denoted by $\hat{r}^\chi_m(\alpha)$ and $\hat{\ell}^\chi_m(\alpha)$. They are normalized as $\text{Tr}_S(\hat{r}^\chi_m(\alpha) \hat{\ell}^\chi_m(\alpha)) = \delta_{m_1 m_2}$. We assign the label for the eigenvalue with the maximum real part to $m = 0$. Then $\hat{\rho}^\chi(\tau) \sim e^{\lambda^\chi_{0}}$ holds for large $\tau$, which results in $\lim_{\tau \to \infty} G_\tau(\chi)/\tau = \lambda^\chi_{0}$. Thus $\lambda^\chi_{0}(\alpha)$ is equal to the unit-time cumulant generating function $g(\chi)$ of the NESS for fixed $\alpha$ [11]. Therefore, the average entropy flux $J_\sigma(\alpha)$ in the NESS can be calculated by

$$J_\sigma(\alpha) = \frac{\partial \lambda^\chi_{0}(\alpha)}{\partial (i\chi)} \bigg|_{\chi=0}.$$  

(2.15)

If we set $\chi = 0$, the GQMME reduces to the original QMME, and $\mathcal{K}^\chi$, $\hat{r}^\chi_0$, and $\hat{\ell}^\chi_0$ also reduce to $\mathcal{K}$, $\hat{1}$, and $\hat{\rho}_{\text{ss}}$, respectively. We also note that, for the quasistatic operations ($\tau_R \ll \tau_{\text{ctrl}}$), we can use Eq. (2.13) with time-dependent $\alpha$:

$$\frac{\partial \hat{\rho}^\chi(t)}{\partial t} = \mathcal{K}^\chi(\alpha(t)) \hat{\rho}^\chi(t).$$  

(2.16)

### 2.3 Explicit Form of GQMME

We here consider the case where the system-reservoir coupling Hamiltonian is given by

$$\hat{H}_{SB} = \sum_l (\hat{X}_{b,l} \otimes \hat{B}_{b,l} + \hat{X}_{b,l}^\dagger \otimes \hat{B}_{b,l}^\dagger).$$  

(2.17)

Here, $\hat{X}_{b,l}$ and $\hat{B}_{b,l}$ ($\hat{X}_{b,l}^\dagger$ and $\hat{B}_{b,l}^\dagger$) are single-particle annihilation (creation) operators of the system S and of the bth reservoir $R_b$, respectively, so that $[\hat{N}_S, \hat{X}_{b,l}] = -\hat{X}_{b,l}$, $[\hat{N}_S, \hat{X}_{b,l}^\dagger] = \hat{X}_{b,l}^\dagger$, $[\hat{N}_b, \hat{B}_{b,l}] = -\hat{B}_{b,l}$, and $[\hat{N}_b, \hat{B}_{b,l}^\dagger] = \hat{B}_{b,l}^\dagger$ hold. The index $l$ is a label for distinguishing the types of the coupling. In this subsection, we derive an explicit form of the GQMME by introducing eigenoperators [7][12].

**Eigenoperator**

Let $\hat{P}_b(\mathcal{E}_b)$ be the projection operator in $R_b$ which projects onto the eigenspace belonging to the eigenvalue $\mathcal{E}_b$ of $H_b$. Then we introduce the eigenoperators of $R_b$ as

$$\hat{B}^{(\Omega_b)}_{b,l} := \sum_{\mathcal{E}_b} \hat{P}_b(\mathcal{E}_b - \hbar \Omega_b) \hat{B}_{b,l}(\mathcal{E}_b),$$  

(2.18)

$$\hat{B}^\dagger_{b,l}^{(\Omega_b)} := \sum_{\mathcal{E}_b} \hat{P}_b(\mathcal{E}_b + \hbar \Omega_b) \hat{B}_{b,l}^\dagger(\mathcal{E}_b),$$  

(2.19)
where \( h\Omega_b \) is a difference of the reservoir eigenenergies. \( \hat{B}_{b,l}^{\Omega_b} \) (\( \hat{B}^{\dagger \Omega_b}_{b,l} \)) decreases (increases) the energy and particle number of the reservoir \( R_b \) by \( h\Omega_b \) and 1, respectively. We note that \( \hat{B}_{b,l} \) and \( \hat{B}^{\dagger}_{b,l} \) can be decomposed into the eigenoperators:

\[
\hat{B}_{b,l} = \sum_{\Omega_b} \hat{B}^{\Omega_b}_{b,l}, \tag{2.20}
\]

\[
\hat{B}^{\dagger}_{b,l} = \sum_{\Omega_b} \hat{B}^{\dagger \Omega_b}_{b,l}. \tag{2.21}
\]

Similarly, we introduce the eigenoperators of the system \( S \). Let \( \hat{P}_S(E_\nu) \) be the projection operator in \( S \) which projects onto the eigenspace belonging to the eigenvalue \( E_\nu \) of \( \hat{H}_S \). Then we define the eigenoperators of \( S \) as

\[
\hat{X}^{(\omega_S)}_{b,l} := \sum_\nu \hat{P}_S(E_\nu - h\omega_S) \hat{X}_{b,l} \hat{P}_S(E_\nu), \tag{2.22}
\]

\[
\hat{X}^{\dagger (\omega_S)}_{b,l} := \sum_\nu \hat{P}_S(E_\nu + h\omega_S) \hat{X}^{\dagger}_{b,l} \hat{P}_S(E_\nu), \tag{2.23}
\]

where \( h\omega_S \) is a difference of the system eigenenergies. \( \hat{X}^{(\omega_S)}_{b,l} \) (\( \hat{X}^{\dagger (\omega_S)}_{b,l} \)) decreases (increases) the energy and particle number of the system \( S \) by \( h\omega_S \) and 1, respectively. We note that \( \hat{X}_{b,l} \) and \( \hat{X}^{\dagger}_{b,l} \) are reconstructed from the eigenoperators:

\[
\hat{X}_{b,l} = \sum_{\omega_S} \hat{X}^{(\omega_S)}_{b,l}, \tag{2.24}
\]

\[
\hat{X}^{\dagger}_{b,l} = \sum_{\omega_S} \hat{X}^{\dagger (\omega_S)}_{b,l}. \tag{2.25}
\]

**GQMME Generator with Eigenoperators**

By substituting Eqs. (2.17), (2.20), (2.21), (2.24), and (2.25) into Eq. (2.13), and using the properties of the eigenoperators, we obtain an explicit form of the GQMME generator as

\[
K^X = K_0 + \nu \sum_b L^X_b, \tag{2.26}
\]

where

\[
K_0 \dot{Y} = \frac{1}{i\hbar} [\hat{H}_S, \dot{Y}], \tag{2.27}
\]

\[
L^X_b \dot{Y} = -\frac{1}{2\hbar^2} \sum_{l,l', \omega_S, \omega'_S} \left[ \Phi^{+}_{b,l' l}(\omega'_S) \left\{ \hat{X}^{(\omega_S)}_{b,l} \hat{X}^{\dagger (\omega'_S)}_{b,l'} \dot{Y} + \dot{Y} \hat{X}^{(\omega'_S)}_{b,l} \hat{X}^{\dagger (\omega_S)}_{b,l'} \right\} 
- e^{i\chi(b_\omega - \omega_S - \mu_b)} \left( \hat{X}^{\dagger (\omega_S)}_{b,l'} \hat{X}^{(\omega'_S)}_{b,l} \hat{X}^{\dagger (\omega'_S)}_{b,l'} \right) \right] 
+ \Phi^{-}_{b,l' l}(\omega'_S) \left\{ \hat{X}^{\dagger (\omega'_S)}_{b,l} \hat{X}^{(\omega'_S)}_{b,l'} \dot{Y} + \dot{Y} \hat{X}^{(\omega'_S)}_{b,l} \hat{X}^{\dagger (\omega'_S)}_{b,l'} \right\} 
- e^{-i\chi(b_\omega - \omega_S - \mu_b)} \left( \hat{X}^{(\omega'_S)}_{b,l} \hat{X}^{\dagger (\omega'_S)}_{b,l'} \hat{X}^{\dagger (\omega'_S)}_{b,l'} \right) \right], \tag{2.28}
\]

Here \( \Phi^{+}_{b,l' l}(\omega) \) is referred to as spectral function of the \( b \)th reservoir, which is given by

\[
\Phi^{+}_{b,l' l}(\omega) := 2\pi \sum_{\Omega_b} \delta(\Omega_b - \omega) \text{Tr}_b \left\{ \hat{P}_b(\alpha_b) \hat{B}^{\dagger \Omega_b}_{b,l} \hat{B}^{\Omega_b}_{b,l'} \right\}, \tag{2.29}
\]

\[
\Phi^{-}_{b,l' l}(\omega) := 2\pi \sum_{\Omega_b} \delta(\Omega_b - \omega) \text{Tr}_b \left\{ \hat{P}_b(\alpha_b) \hat{B}^{\Omega_b}_{b,l} \hat{B}^{\dagger \Omega_b}_{b,l'} \right\}. \tag{2.30}
\]
In Eq. (2.26), we neglected the terms which are proportional to the Hilbert transform of $\Phi_{b,l'}(\omega)$ because these terms are known to give negligible contribution to the dynamics [14]. The spectral function has the following properties:

\[
\begin{align*}
\left[\Phi_{b,l'}^+(\omega)\right]^* &= \Phi_{b,l'}^+(\omega), \\
\Phi_{b,l'}^+(\omega) &= e^{-\beta_b(h\omega - \mu_b)}\Phi_{b,l'}^-(\omega).
\end{align*}
\] (2.31) (2.32)

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

For later use, we introduce the $i\chi$-derivative of the generator: $K' := \partial K' / \partial (i\chi)|_{\chi=0}$. From Eq. (2.26), we can write $K'$ as $K' = v \sum_b L_b$ with $L_b := \partial L_b^\chi / \partial (i\chi)|_{\chi=0}$. Moreover, we can show that

\[
\mathcal{P} L_b' P \check{Y} = \beta_b(\check{H}_S - \mu_b \check{N}_S) \mathcal{P} L_b P \check{Y} - \mathcal{P} L_b P \beta_b(\check{H}_S - \mu_b \check{N}_S) \check{Y}
\] (2.33)

holds for any $\check{Y} \in B$, where $L_b = L_b^\chi = 0$.

**Rotating Wave Approximation**

We here consider the situation where the time scale $\tau_S$ of the intrinsic evolution of the system $S$ is much smaller than the relaxation time $\tau_{\text{rlx}}$ of $S$, where $\tau_S$ is given by a typical value of $|\omega_S - \omega'_S|^{-1}$ and $\tau_{\text{rlx}}$ is the time scale over which $S$ varies appreciably. In this case, the terms with $\omega_S \neq \omega'_S$ in Eq. (2.28) rapidly oscillate within the time scale $\tau_{\text{rlx}}$ if they are written in the interaction picture. Therefore we may neglect these terms and leave only the terms with $\omega_S = \omega'_S$. This approximation is known as the rotating wave approximation (RWA) or secular approximation. We express the quantities and (super)operators within the RWA by the subscript ‘r’; for example, we write the GQMME generator within the RWA as $K^\chi_r$.

Similarly to the case without the RWA, we can decompose the generator $K^\chi_r$ as

\[
K^\chi_r = K_0 + v \sum_b L^\chi_{b,r},
\] (2.34)

where we define $L^\chi_{b,r}$ by leaving only the $\omega'_S = \omega_S$ terms in the $\omega'_S$-sum in Eqs. (2.28). We can show the following equation

\[
\mathcal{Q} K^\chi_r \mathcal{P} = \mathcal{P} K^\chi_r \mathcal{Q} = 0.
\] (2.35)

This implies that the GQMME is decomposed into two closed systems of equations: one is for $\mathcal{P} \rho^\chi_r$ and the other is for $\mathcal{Q} \rho^\chi_r$. Furthermore, we can show that $\mathcal{P} K^\chi_r \mathcal{P} = \mathcal{P} K^\chi \mathcal{P}$ holds.

We write the right and left eigenvectors of $K^\chi_r$ corresponding to the eigenvalue $\lambda^\chi_{m,r}$ as $\hat{r}^\chi_{m,r}$ and $\hat{\ell}^\chi_{m,r}$. They are normalized as $\text{Tr}_S(\hat{r}^\chi_{m,r} \hat{r}^\chi_{n,r}) = \delta_{mn}$. We assign the label for the eigenvalue with the maximum real part to $m = 0$. For the reason mentioned below Eq. (2.35), we can classify the eigenvectors into two groups: one group is in $P$ and the other is in $Q$. In particular, the eigenvectors for $m = 0$ belong to the former group. We note that $\hat{\ell}^\chi_{0,r} = 0$ holds, and that $\hat{r}^\chi_{0,r} = 0$ is the steady solution of the QMME within the RWA. We assume that the steady solution is uniquely determined for fixed $\alpha$ also in the RWA.

**Remark on the fluctuation theorem**

Before closing this section, we make a remark on the fluctuation theorem (FT) in the present setup. First, we consider in the RWA. Due to the KMS condition (2.32), $L^\chi_{b,r}$ in Eq. (2.34) satisfies $L^\chi_{b,r} = -L^\chi_{b,r}$. Moreover, because $\hat{\ell}^\chi_{0,r}$ is in $P$, $K_0 \hat{\ell}^\chi_{0,r} = K^\chi_0 \hat{\ell}^\chi_{0,r} = 0$ holds. Therefore, we have $K^\chi_r \hat{\ell}^\chi_{0,r} = K^\chi_r \hat{\ell}^\chi_{0,r} = 0$. 8
\( \lambda_0^\chi \ell_0^\chi \); i.e., the eigenvalue \( \lambda_0^\chi \ell_0^\chi \) of \( K_0^\chi \) with the maximum real part is equal to \( \lambda_0^\chi \). Next, we consider without the RWA. In Sec. 3.1 we will show that \( \lambda_0^\chi = \lambda_0^\chi + O(v^2) \) holds. Therefore we obtain \( \lambda_0^\chi = \lambda_0^\chi + O(v^2) \). Note that \( O(v) \) is the same precision as that in the QMME.

This equality leads to the relation \( g(-\chi - i) = g^*(\chi) + O(v^2) \), because \( \lambda_0^\chi \) is equal to the unit-time cumulant generating function \( g(\chi) \) of the entropy production in the NESS for fixed \( \alpha \). Noting that the generating function satisfies \( g(-\chi) = g^*(\chi) \), we obtain the symmetry relation \( g(-\chi - i) = g(\chi) + O(v^2) \). This is one of the expressions of the steady-state FT for the entropy production [11]. This result supports that our definition (2.9) of the entropy production is reasonable. Note that the fluctuation theorem shown here is the steady state FT, where the control parameters \( \alpha \) are fixed. On the other hand, if a modulation of \( \alpha \) is considered as in the next section, the transient FT should hold (although we do not show in this paper).

## 3 Results for Generic System

### 3.1 Geometrical Expression of Excess Entropy Production

We now consider an arbitrary quasistatic operation that connects two steady states. At the initial time \( t = 0 \), the system \( S \) is set to be in a steady state that is uniquely specified by \( \alpha(0) = \alpha^f \). Then the system \( S \) is subjected to an external operation that is characterized by a modulation of the parameters \( \alpha \) along a curve \( C \) in the parameter space. At \( t = \tau \), \( S \) settles in another steady state with \( \alpha(\tau) = \alpha^f \). For the quasistatic operation, the time interval \( \tau \) of the operation is sufficiently larger than the relaxation time scale of the system. Since there exist steady particle and energy currents in the NESS at each \( \alpha \) in \( C \), the average entropy production \( \langle \sigma \rangle_\tau \) includes a component that linearly increases with \( \tau \). This component is referred to as housekeeping part of the entropy production [21] and is given by

\[
\langle \sigma \rangle_{\tau}^{hk} := \int_0^\tau dt J_\sigma(\alpha(t)),
\]

where \( J_\sigma(\alpha) \) is the steady entropy flux given in Eq. (2.15) for fixed \( \alpha \). By subtracting this component from \( \langle \sigma \rangle_\tau \), we define the excess entropy production:

\[
\langle \sigma \rangle^{ex} := \langle \sigma \rangle_\tau - \langle \sigma \rangle_{\tau}^{hk}.
\]

As we shall show in the below, \( \langle \sigma \rangle^{ex} \) is independent of \( \tau \) for the quasistatic operation.

The main result of the present paper is the geometrical expression for \( \langle \sigma \rangle^{ex} \):

\[
\langle \sigma \rangle^{ex} = -\int_C Tr_S \left[ \ell_0^\dagger(\alpha) \frac{\partial \hat{p}_{ss}(\alpha)}{\partial \alpha} \right] d\alpha,
\]

where \( \ell_0 := \partial \ell_0^\chi / \partial (i\chi) \chi=0 \). We will give the derivation later in this subsection. This expression holds for any quasistatic operations between arbitrary NESSs if the system is described by the QMME. The RHS of Eq. (3.3) is analogous to the Berry phase in quantum mechanics [3]. It is geometrical because it depends only on the line integral along the curve \( C \) but not on \( \tau \). This implies that the excess entropy production is not given by the difference of a scalar potential but given by the integral of the vector potential \(-Tr_S \left[ \ell_0^\dagger(\partial \hat{p}_{ss}/\partial \alpha) \right] \).

Furthermore, as we will show later in this subsection, the analyses within and without the RWA give the equivalent result for the geometrical expression (3.3). That is, the following relation holds for the vector potential:

\[
-Tr_S \left[ \ell_0^\dagger(\alpha) \frac{\partial \hat{p}_{ss}(\alpha)}{\partial \alpha} \right] = -Tr_S \left[ \ell_0^\dagger(\alpha) \frac{\partial \hat{p}_{ss}(\alpha)}{\partial \alpha} \right] + O(v^2),
\]
where $\hat{\rho}_0, t := \partial \hat{\rho}_0, t / \partial (i \chi)|_{\chi = 0}$. Because the QMME is valid up to $O(v)$, this equation implies the equivalence between the vector potentials within and without the RWA. Therefore we can safely use the RWA to calculate the excess entropy production $\langle \sigma \rangle^{ex}$, whereas it is known that the internal current in the system $S$ vanishes in NESSs under the RWA \[32\].

**Derivation of Eq. (3.3)**

We first note that the excess entropy production can be written as $\langle \sigma \rangle^{ex} = \partial G^{ex}(\chi) / \partial (i \chi)|_{\chi = 0}$, where

$$G^{ex}(\chi) := G_\tau(\chi) - \int_0^\tau dt \lambda^\chi_0(\alpha(t))$$  \hspace{1cm} (3.5)

is the excess part of the cumulant generating function of the entropy production. This is because $\langle \sigma \rangle^{ex} = \partial G_\tau(\chi) / \partial (i \chi)|_{\chi = 0}$ and $J_{\sigma}(\alpha) = \partial \lambda^\chi_0(\alpha) / \partial (i \chi)|_{\chi = 0}$ as is mentioned in the previous section.

We derive the geometrical expression for $G^{ex}(\chi)$ by using the method similar to those in Refs [24, 27, 33]. To solve the GQMME for a given curve $C$ of $\alpha$ in the parameter space, we expand $\hat{\rho}^\chi(t)$ as

$$\hat{\rho}^\chi(t) := \sum_m c_m(t)e^{\lambda^\chi_m(t)}|m\rangle \langle m|$$  \hspace{1cm} (3.6)

where $\lambda^\chi_m(t) := \int_0^t dt' \lambda^\chi_m(\alpha(t'))$. Substituting this expansion into Eq. (2.16), we rewrite the left-hand side (LHS) and the RHS of Eq. (2.16) respectively as

[LHS of Eq. (2.16)] = $\sum_m e^{\lambda^\chi_m(t)}\left[\hat{c}_m(t)\hat{r}_m^\chi(\alpha(t)) + \lambda^\chi_m(\alpha(t))c_m(t)\hat{r}_m^\chi(\alpha(t)) + c_m(t)\hat{r}_m^\chi(\alpha(t))\right]$,

[RHS of Eq. (2.16)] = $\sum_m e^{\lambda^\chi_m(t)}\lambda^\chi_m(\alpha(t))c_m(t)\hat{r}_m^\chi(\alpha(t))$,

where $\cdot \cdot$ stands for the time derivative. Equating these equations and taking the Hilbert-Schmidt inner product with $\hat{\lambda}_0^\chi(\alpha(t))$, we obtain

$$\hat{c}_0(t) = -\sum_m c_m(t)e^{\lambda^\chi_m(t)-\lambda_0^\chi(t)}\text{Tr}_S \left[\hat{\lambda}_0^\chi(\alpha(t))\hat{r}_m^\chi(\alpha(t))\right].$$  \hspace{1cm} (3.7)

If the time scale of the modulation of $\alpha$ is much slower than that of the relaxation of the system, we can approximate the sum on the RHS of Eq. (3.7) by the contribution only from the term with $m = 0$ (note that $\text{Re}[\lambda^\chi_m(0) - \lambda_0^\chi(0)] < 0$ for $m \neq 0$):

$$\hat{c}_0(t) \simeq -c_0(t)\text{Tr}_S \left[\hat{\lambda}_0^\chi(\alpha(t))\hat{r}_0^\chi(\alpha(t))\right].$$  \hspace{1cm} (3.8)

This approximation corresponds to the adiabatic approximation in quantum mechanics. By solving this approximate equation we obtain

$$c_0(\tau) = c_0(0)\exp\left\{-\int_C \text{Tr}_S \left[\hat{\lambda}_0^\chi(\alpha)\hat{r}_0^\chi(\alpha)\right]\right\},$$  \hspace{1cm} (3.9)

where $d\hat{r}_0^\chi(\alpha) := (\partial \hat{r}_0^\chi(\alpha) / \partial \alpha) \cdot d\alpha$. If the initial state of the system $S$ is $\hat{\rho}^\chi(0) = \hat{\rho}_{ss}(\alpha^i)$, then $c_0(0) = \text{Tr}_S \left[\hat{\lambda}_0^\chi(\alpha^i)\hat{\rho}_{ss}(\alpha^i)\right]$. We substitute Eq. (3.9) into the $m = 0$ term in Eq. (3.6). At long time only the $m = 0$ term remains and $m \neq 0$ terms vanish in Eq. (3.6) since $\lambda^\chi_m(t)$ has the maximum real part. Therefore we obtain

$$\hat{\rho}^\chi(\tau) \simeq c_0(\tau)e^{\lambda_0^\chi(\tau)}\hat{r}_0^\chi(\alpha^f)$$

$$= e^{\lambda_0^\chi(\tau)}\hat{r}_0^\chi(\alpha^f)\text{Tr}_S \left[\hat{\lambda}_0^\chi(\alpha^i)\hat{\rho}_{ss}(\alpha^i)\right]\exp\left\{-\int_C \text{Tr}_S \left[\hat{\lambda}_0^\chi(\alpha)\hat{r}_0^\chi(\alpha)\right]\right\}.$$  \hspace{1cm} (3.10)
We thus obtain the excess cumulant generating function $G^\text{ex} (\chi) = \ln \text{Tr}_S \hat{\rho}^\chi (\tau) - \Lambda_0^\chi (\tau)$ for the slow modulation:

$$G^\text{ex} (\chi) = - \int_C \text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha) d\hat{\rho}_0^\chi (\alpha) \right] + \ln \text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha^i) \hat{\rho}_\text{ss} (\alpha^i) \right] + \ln \text{Tr}_S \hat{r}_0^\chi (\alpha^i). \quad (3.11)$$

Equation (3.11) is analogous to the Berry phase, and $\Lambda_0^\chi (\tau)$ corresponds to the dynamical phase.

By differentiating Eq. (3.11) with respect to $i\chi$ and setting $\chi = 0$, we obtain the expression for the excess entropy production:

$$\langle \sigma \rangle^\text{ex} = - \int_C \text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha) d\hat{\rho}_0^\chi (\alpha) \right] - \int_C \text{Tr}_S \left[ d\hat{\rho}_0^\chi (\alpha) \right] + \text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha^i) \hat{\rho}_\text{ss} (\alpha^i) \right] + \text{Tr}_S \hat{r}_0^\chi (\alpha^i)$$

$$= - \int_C \text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha) d\hat{\rho}_0^\chi (\alpha) \right] + \text{Tr}_S \left[ \hat{\rho}_0^\chi (\alpha^i) \right] + \text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha^i) \hat{\rho}_\text{ss} (\alpha^i) \right]$$

$$= - \int_C \text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha) d\hat{\rho}_0^\chi (\alpha) \right]. \quad (3.12)$$

We thus obtain Eq. (3.13). In Eq. (3.12), we introduced $\hat{r}_0^\chi := \partial \hat{\rho}_0^\chi / \partial (i\chi) |_{\chi = 0}$, and used $\hat{\ell}_0 = \hat{1}$ in the first line. In the third line, the surface terms vanish because they are rewritten as

$$\text{Tr}_S \left[ \hat{\rho}_0^\chi (\alpha^i) \right] + \text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha^i) \hat{\rho}_\text{ss} (\alpha^i) \right] = \frac{\partial}{\partial (i\chi)} \text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha^i) \hat{\rho}_0^\chi (\alpha^i) \right] \bigg|_{\chi = 0}, \quad (3.13)$$

and because of the normalization condition $\text{Tr}_S \left[ \hat{\ell}_0^\dagger (\alpha^i) \hat{\rho}_0^\chi (\alpha^i) \right] = 1$.

**Derivation of Eq. (3.4)**

To show the equivalence between the results within and without the RWA, we derive the relations between the eigenvalues and eigenvectors of the GQMME generators within and without the RWA. For this purpose, we decompose the generator as $K^\chi = K_r^\chi + vR^\chi$. Then $vR^\chi$ is $O(v)$ since $K_r^\chi$ contains all of the $O(v^0)$ terms (as well as a part of the $O(v)$ terms) in $K^\chi$. Motivated by this decomposition, we here make the ansatz that we can expand the eigenvalue $\lambda_0^\chi$ and eigenvectors $\hat{\ell}_0^\chi$ and $\hat{r}_0^\chi$ of $K^\chi$ around those of $K_r^\chi$ with respect to $v$:

$$\lambda_0^\chi = \lambda_{0,r}^\chi + v \Delta^\chi + O(v^2), \quad (3.14)$$

$$\hat{r}_0^\chi = \hat{r}_{0,r}^\chi + v \hat{\eta}^\chi + O(v^2), \quad (3.15)$$

$$\hat{\ell}_0^\chi = \hat{\ell}_{0,r}^\chi + v \hat{\zeta}^\chi + O(v^2). \quad (3.16)$$

We note that $\lambda_{0,r}^\chi$ is non-degenerate because the steady state is assumed to be uniquely determined for fixed $\alpha$ in the RWA. We also note that $\hat{\ell}_{0,r}^\chi$ and $\hat{r}_{0,r}^\chi$ may have both the $O(v^0)$ and $O(v)$ terms, because $K_r^\chi$ has $O(v)$ terms as well as $O(v^0)$ ones. Then, regarding $K_r^\chi$ as the unperturbed part and $vR^\chi$ as the perturbation, we apply the formal perturbation theory for the non-degenerate case to obtain

$$\Delta^\chi = \text{Tr}_S [\hat{\ell}_{0,r}^\dagger R \hat{r}_{0,r}^\chi], \quad (3.17)$$

$$\hat{\eta}^\chi = \sum_{m \neq 0} \frac{\text{Tr}_S [\hat{\lambda}_{m}^\chi R \hat{r}_{0,r}^\chi]}{\lambda_{0,r}^\chi - \lambda_{m,r}^\chi} \hat{r}_{m,r}^\chi, \quad (3.18)$$

$$\hat{\zeta}^\chi = \sum_{m \neq 0} \left( \frac{\text{Tr}_S [\hat{\lambda}_{m}^\chi R \hat{r}_{0,r}^\chi]}{\lambda_{0,r}^\chi - \lambda_{m,r}^\chi} \right)^* \hat{\ell}_{m,r}^\chi. \quad (3.19)$$
The terms with \( m = 0 \) vanish in Eqs. (3.18) and (3.19) because of the normalization condition \( \text{Tr}_S[\hat{\chi}^{\dagger}_0 \hat{\chi}_0] = 1 \). Here, \( \zeta^x \) and \( \eta^x \) must be \( O(v^0) \) for the ansatz and the formal perturbation theory to be consistent. As we will show later, the denominators \( \lambda^x_{0,r} - \lambda^x_{m,r} \) in Eqs. (3.18) and (3.19) are \( O(v) \) for \( m \)'s where the corresponding eigenvectors \( \hat{\imath}^x_{m,r} \) and \( \hat{r}^x_{m,r} \) are in \( \mathcal{P} \). Therefore it is necessary to show that the corresponding numerators vanish.

We can show this as follows. We first note that \( \mathcal{PR} \mathcal{X} \mathcal{P} = 0 \) since \( \mathcal{PK}^x \mathcal{P} = \mathcal{PK}^x \mathcal{P} \) holds as is mentioned below Eq. (2.35). This leads to \( \text{Tr}_S[\hat{Y}_1 \mathcal{R} \mathcal{X} \hat{Y}_2] = 0 \) if \( \hat{Y}_1, \hat{Y}_2 \in \mathcal{P} \). Because \( \hat{r}^x_{0,r} \in \mathcal{P} \), we have \( \text{Tr}_S[\hat{\imath}^{x\dagger}_0 \mathcal{R} \hat{\chi}^x_{0,r}] = 0 \) if \( \hat{\imath}^{x\dagger}_0, \hat{r}^x_{0,r} \in \mathcal{P} \). Similarly, we can show \( \text{Tr}_S[\mathcal{R} (\hat{\chi}^{x\dagger}_0 \hat{\imath}^{x\dagger}_0 + \hat{r}^{x\dagger}_0) \hat{r}^x_{m,r}] = 0 \) if \( \hat{r}^x_{m,r} \in \mathcal{P} \). These indicate that the ansatz (3.14)–(3.16) and the formal perturbation theory do work.

From this argument and Eq. (3.17), we have \( \Delta^x = 0 \). Therefore from Eq. (3.14) we obtain \( \lambda^x_0 = \lambda^x_{0,r} + O(v^2) \). This implies that the analyses within and without the RWA give the equivalent result for the unit-time cumulant generating function for the entropy production in the steady state for fixed \( \alpha \).

The above argument also implies that the nonzero contributions to \( \hat{\zeta}^x \) and \( \hat{\eta}^x \) come only from \( m \)'s which are in \( \mathcal{Q} \). Therefore we have

\[
\text{Tr}_S[\hat{\chi}^{x\dagger}_0 \hat{\imath}^{x\dagger}_0] = \text{Tr}_S[\hat{\imath}^{x\dagger}_0 \hat{\imath}^{x}_0] = 0.
\]

(3.20)

Substituting Eqs. (3.15) and (3.16) into the vector potential \( -\text{Tr}_S[\hat{\rho}^{x\dagger}_0 (\partial \hat{\rho}_{ss} / \partial \alpha)] \), we have

\[
-\text{Tr}_S[\hat{\rho}^{x\dagger}_0 (\partial \hat{\rho}_{ss} / \partial \alpha)] = -\text{Tr}_S[\hat{\rho}^{x\dagger}_{0,r}(\partial \hat{\rho}_{ss,r} / \partial \alpha)] + vD + O(v^2),
\]

(3.21)

where

\[
D := \text{Tr}_S[\hat{\zeta}'(\alpha) (\partial \hat{\rho}_{ss} / \partial \alpha)] + \text{Tr}_S[\hat{\chi}'(\alpha) (\partial \eta / \partial \alpha)].
\]

(3.22)

Finally, we show that \( \lambda^x_{0,r} - \lambda^x_{m,r} = O(v) \) if and only if \( \hat{\imath}^{x\dagger}_{m,r}, \hat{r}^x_{m,r} \in \mathcal{P} \). Thanks to the decomposition of \( \mathcal{K}^x_0 \) shown in Eq. (2.34), we can expand its eigenvalue as \( \lambda^x_{m,r} = \lambda^x_{m,r} + v \lambda^{(1)x}_{m,r} + O(v^2) \), and the eigenvectors as \( \hat{r}^x_{m,r} = \hat{r}^{(0)x}_{m,r} + v \hat{r}^{(1)x}_{m,r} + O(v^2) \). For \( m \)'s where \( \hat{r}^x_{m,r} \in \mathcal{P} \), we have \( \mathcal{K}^x_0 \hat{r}^{(0)x}_{m,r} = 0 \), which implies \( \lambda^{(0)x}_{m,r} = 0 \). Therefore, for the eigenvalues whose eigenvectors are in \( \mathcal{P} \), the difference \( \lambda^x_{0,r} - \lambda^x_{m,r} \) is \( O(v) \).

On the other hand, for \( m \)'s where \( \hat{r}^x_{m,r} \in \mathcal{Q} \), \( \mathcal{K}^x_0 \hat{r}^{(0)x}_{m,r} \neq 0 \) holds, so that \( \lambda^x_{0,r} - \lambda^x_{m,r} = O(v^0) \).

### 3.2 Clausius Equality in Equilibrium State

We next show that Eq. (3.3) reduces to the Clausius equality in the equilibrium setup. In this setup, all the temperatures and chemical potentials of the reservoirs are equal: \( \beta_1 = \beta_2 = \cdots =: \beta \) and \( \mu_1 = \mu_2 = \cdots =: \mu \), where \( \beta \) and \( \mu \) may be time-dependent. This situation is equivalent to the case where the system \( S \) is in contact with a single reservoir with the inverse temperature \( \beta \) and chemical potential \( \mu \). Therefore we can omit the index \( b \); for example, \( \mathcal{K}_f^x = \mathcal{K}_0 + vL_f^x \) and \( \mathcal{K}_r^x = vL_r^x \).

In this case, we can show that the grand-canonical state is the steady solution of the QMME \((\chi = 0)\) within the RWA. That is, \( \mathcal{K}_r \hat{\rho}_{ge}(\beta, \beta \mu) = 0 \), where \( \hat{\rho}_{ge}(\beta, \beta \mu) := e^{-\beta H_S + \beta \mu N_S} / Z_{ge}(\beta, \beta \mu) \) with \( Z_{ge}(\beta, \beta \mu) = \text{Tr}_S e^{-\beta H_S + \beta \mu N_S} \).

In order to have the explicit form of \( \hat{r}^{0}_0 \) in the equilibrium setup, we differentiate the left eigenvalue equation \( \mathcal{K}_r \hat{r}^{0}_0 = \lambda^x \hat{r}^{0}_0 \) with respect to \( i\chi \) and set \( \chi = 0 \):

\[
\mathcal{K}_r^{\dagger} \hat{r}^{0}_0 + \mathcal{K}_r^{\dagger} \hat{r}^{0}_0 = 0,
\]

(3.23)
where we used \( \lambda_0^x |_{x=0} = 0 \), \( \partial \lambda_0^x / \partial (i \chi)|_{x=0} = 0 \) and \( \hat{\chi}_{0,r}^+ = \hat{1} \). To rewrite Eq. (3.23), we note that the following equation holds for any \( \hat{Y} \in \mathcal{B} \):

\[
\text{Tr}_S[(\mathcal{L}_r^c \hat{1})^\dagger \hat{Y}] = \text{Tr}_S[(\mathcal{P} \mathcal{L}_r^c \mathcal{P})^\dagger \hat{Y}]
\]

\[
= \text{Tr}_S[\mathcal{P} \mathcal{L}_r^c \mathcal{P} \hat{Y}]
\]

\[
= \text{Tr}_S[\beta(\hat{H}_S - \mu \hat{N}_S) \mathcal{P} \mathcal{L}_r \mathcal{P} \hat{Y} - \mathcal{P} \mathcal{L}_r \mathcal{P} \beta(\hat{H}_S - \mu \hat{N}_S) \hat{Y}]
\]

\[
= \text{Tr}_S[\{\mathcal{P} \mathcal{L}_r^c \mathcal{P} \beta(\hat{H}_S - \mu \hat{N}_S)\}^\dagger \hat{Y} - \{\mathcal{P} \mathcal{L}_r^c \mathcal{P} \}^\dagger \beta(\hat{H}_S - \mu \hat{N}_S) \hat{Y}]
\]

\[
= \text{Tr}_S[\{\mathcal{L}_r^c \beta(\hat{H}_S - \mu \hat{N}_S)\}^\dagger \hat{Y}],
\]

(3.24)

where we used \( \mathcal{P} \hat{1} = \hat{1} \) and \( Q \mathcal{L}_r^c \mathcal{P} = 0 \) in the first line, Eq. (2.33) in the third line, \( \mathcal{L}_r^c \hat{1} = 0 \) in the fourth line, and \( \mathcal{P}(\hat{H}_S - \mu \hat{N}_S) = \hat{H}_S - \mu \hat{N}_S \) in the last line. This equation implies

\[
\mathcal{L}_r^c \hat{1} = \mathcal{L}_r^c \beta(\hat{H}_S - \mu \hat{N}_S).
\]

Therefore, we can rewrite Eq. (3.23) as

\[
\kappa_r^+ \{\hat{\rho}_{0,r} + \beta(\hat{H}_S - \mu \hat{N}_S)\} = 0.
\]

(3.26)

Since the left eigenvector of \( \kappa_r \) corresponding to the zero eigenvalue is proportional to the identity operator \( \hat{1} \), we have

\[
\hat{\rho}_{0,r} = -\beta(\hat{H}_S - \mu \hat{N}_S) + c \hat{1},
\]

(3.27)

where \( c \) is an unimportant constant.

Substituting \( \hat{\rho}_{ss,r} = \hat{\rho}_{gc}(\beta, \beta \mu) \) and Eq. (3.27) into the vector potential \( -\text{Tr}_S[\hat{\rho}_{0,r}^+(\partial \hat{\rho}_{ss,r} / \partial \alpha)] \), we obtain

\[
-\text{Tr}_S[\hat{\rho}_{0,r}^+(\alpha) \partial \hat{\rho}_{ss,r}(\alpha) / \partial \alpha] = -\text{Tr}_S[\ln \hat{\rho}_{gc}(\beta, \beta \mu) \partial \hat{\rho}_{gc}(\beta, \beta \mu) / \partial \alpha],
\]

(3.28)

where we used \( \text{Tr}_S[\partial \hat{\rho}_{ss,r} / \partial \alpha] = 0 \). The RHS of Eq. (3.28) is the derivative of the von Neumann entropy, \( S_{VN}(\hat{\rho}_{gc}) := -\text{Tr}_S[\hat{\rho}_{gc} \ln \hat{\rho}_{gc}] \), in terms of the control parameters.

Finally, we note that the the grand-canonical state is the steady solution of QMME in the equilibrium setup also without the RWA: \( \hat{\rho}_{ss} = \hat{\rho}_{gc}(\beta, \beta \mu) \). Combining this fact with Eq. (3.24), we obtain the Clausius equality in the equilibrium setup (even without the RWA). That is, for the quasistatic operations in the equilibrium case, the change of the von Neumann entropy between the initial and final states is equal to the excess entropy production, which equals the total entropy production because the house-keeping part vanishes in the equilibrium states.

### 3.3 Extended Clausius Equality in Weakly Nonequilibrium Regime

We now show that Eq. (3.3) reduces to the extended Clausius equality \cite{16,17,25} in the weakly nonequilibrium setup. In later part of this subsection, we show the following three equations.

\[
-\text{Tr}_S[\hat{\rho}_{0,r}^+(\alpha) \partial \hat{\rho}_{ss,r}(\alpha) / \partial \alpha] = -\text{Tr}_S[\ln (\hat{\rho}_{ss,r}(\alpha) \hat{\theta}^{-1}) \partial \hat{\rho}_{ss,r}(\alpha) / \partial \alpha] + O(\epsilon^2),
\]

(3.29)

\[
\partial \alpha S_{sym}(\hat{\rho}_{ss,r}(\alpha)) = -\text{Tr}_S[\ln (\hat{\rho}_{ss,r}(\alpha) \hat{\theta}^{-1}) \partial \hat{\rho}_{ss,r}(\alpha) / \partial \alpha] + O(\epsilon^2),
\]

(3.30)

\[
S_{sym}(\hat{\rho}_{ss}(\alpha)) = S_{sym}(\hat{\rho}_{ss,r}(\alpha)) + O(\epsilon^2),
\]

(3.31)

\(^6\) As is mentioned in Eq. (2.15), \( \partial \lambda_0^x / \partial (i \chi)|_{x=0} = J_\theta \) is the average entropy flow from the reservoirs into the system. This becomes zero in the equilibrium state because of the second law of thermodynamics.
where $\epsilon$ is a measure of “degree of nonequilibrium” [see the below of Eq. (3.39)], and
\[
S_{\text{sym}}(\hat{\rho}) := -\frac{1}{2} \text{Tr}_S \left[ \hat{\rho} (\ln \hat{\rho} + \ln \hat{\rho}^{-1}) \right] \tag{3.32}
\]
is the symmetrized von Neumann entropy, which is first defined in Ref. [25].

Equations (3.29) and (3.30) lead to
\[
-\text{Tr}_S \left[ \hat{\rho}_{0,r}^{(\alpha)} \frac{\partial \hat{\rho}_{ss,r}^{(\alpha)}(\alpha)}{\partial \alpha} \right] = \frac{\partial}{\partial \alpha} S_{\text{sym}}(\hat{\rho}_{ss,r}(\alpha)) + O(\epsilon^2). \tag{3.33}
\]
Combining this equation with Eqs. (3.4) and (3.31), we have
\[
-\text{Tr}_S \left[ \hat{\rho}_{0,r}^{(\alpha)} \frac{\partial \hat{\rho}_{ss}^{(\alpha)}(\alpha)}{\partial \alpha} \right] = \frac{\partial}{\partial \alpha} S_{\text{sym}}(\hat{\rho}_{ss}(\alpha)) + O(\epsilon^2) + O(\nu^2). \tag{3.34}
\]
Equation (3.34) with the geometrical formula (3.3) implies that the extended Clausius equality holds in the weakly nonequilibrium regime (even without the RWA):
\[
\langle \sigma \rangle^{\text{ex}} = S_{\text{sym}}(\hat{\rho}_{ss}(\alpha^r)) - S_{\text{sym}}(\hat{\rho}_{ss}(\alpha^i)) + O(\epsilon^2 \delta) + O(\nu^2 \delta), \tag{3.35}
\]
where $\delta := \max_{\alpha \in C} |\alpha - \alpha^1|/\bar{\alpha}$, with $\bar{\alpha}$ being a typical values of the control parameters. In particular, if the initial state is an equilibrium state and the reservoir parameter change is included in the external operation, we have $\delta = O(\epsilon)$. Therefore the extended Clausius equality is valid up to $O(\epsilon^2)$ in this case.

Before going to the derivations, we make a remark that the symmetrized von Neumann entropy $S_{\text{sym}}(\hat{\rho}_{ss})$ of the steady state is equal to the von Neumann entropy $S_{\text{VN}}(\hat{\rho}_{ss}) = -\text{Tr}_S [\hat{\rho}_{ss} \ln \hat{\rho}_{ss}]$ if the spectrum of $\hat{H}_S$ is non-degenerate. We can show this as follows. We first note that if $\hat{\rho}$ is time-reversal invariant, $\hat{\rho} \hat{\rho}^{-1} = \hat{\rho}$, then $S_{\text{sym}}(\hat{\rho}) = S_{\text{VN}}(\hat{\rho})$ holds. If $\hat{H}_S$ is non-degenerate, its eigenstates satisfy $\hat{\rho} |E_\nu\rangle = c_\nu |E_\nu\rangle$, where $c_\nu$ is a real constant with $c_\nu^2 = 1$, and the steady state $\hat{\rho}_{ss,r}$ in the RWA is diagonal in the energy eigenstate basis. Therefore, $\hat{\rho}_{ss,r} \hat{\rho}_{ss,r}^{-1} = \hat{\rho}_{ss,r}$ holds. This leads to $S_{\text{sym}}(\hat{\rho}_{ss,r}) = S_{\text{VN}}(\hat{\rho}_{ss,r})$. Furthermore, combining this result with Eqs. (3.31) and (3.71), which will be shown later, we have $S_{\text{sym}}(\hat{\rho}_{ss}) = S_{\text{VN}}(\hat{\rho}_{ss}) + O(\nu^2)$.

**Derivation of Eq. (3.29)**

In the weakly nonequilibrium regime, we can use a perturbative analysis with respect to the thermodynamic forces $\{\epsilon_1,b := \beta_{b} - \bar{\beta}\}_b$ and $\{\epsilon_2,b := \beta_{b}\mu_{b} - \bar{\beta}\bar{\mu}\}_b$, where $\bar{\beta}$ and $\bar{\beta}\bar{\mu}$ are reference values of the forces; we expand $\hat{K}_{\alpha}^{x}, \hat{\rho}_{0,r}^{(\alpha)}$ and $\hat{\rho}_{ss,r}$ as
\[
\hat{K}_{\alpha}^{x}(\alpha) = \hat{K}_{\alpha}^{x} + \sum_{b} \left( \epsilon_1,b \hat{K}_{1,b}^{x} + \epsilon_2,b \hat{K}_{2,b}^{x} \right) + O(\epsilon^2), \tag{3.36}
\]
\[
\hat{\rho}_{ss,r}(\alpha) = \hat{\rho}_{ss,r} + \sum_{b} \left( \epsilon_1,b \hat{\rho}_{1,b} + \epsilon_2,b \hat{\rho}_{2,b} \right) + O(\epsilon^2), \tag{3.37}
\]
\[
\hat{\rho}_{0,r}^{(\alpha)}(\alpha) = \hat{\rho}_{0,r}^{(\alpha)} + \sum_{b} \left( \epsilon_1,b \hat{k}_{1,b} + \epsilon_2,b \hat{k}_{2,b} \right) + O(\epsilon^2), \tag{3.38}
\]
where $\hat{K}_{\alpha}^{x} := \hat{K}_{\alpha}^{x}(\alpha_S,\alpha_B)$, $\hat{\rho}_{ss,r} := \hat{\rho}_{ss,r}(\alpha_S,\alpha_B)$, $\hat{\rho}_{0,r}^{(\alpha)} := \hat{\rho}_{0,r}^{(\alpha)}(\alpha_S,\alpha_B)$, and
\[
\frac{\partial \hat{K}_{\alpha}^{x}(\alpha)}{\partial \alpha_{1,b}} \bigg|_{\alpha_B = \bar{\alpha}_B} = \frac{\partial \hat{K}_{\alpha}^{x}(\alpha)}{\partial \alpha_{1,b}} \bigg|_{\alpha_B = \bar{\alpha}_B}, \tag{3.39}
\]
with \( i = 1, 2 \), \( \alpha_{1,b} = \beta_b \), and \( \alpha_{2,b} = \beta_b \mu_b \). Here, \( \overline{\alpha}_B = \{ \overline{\beta}, \overline{\mu} \}_b \) is the set of the reservoir parameters all of which are set to the reference values, \( \epsilon = \max_b \epsilon_b \), and \( \epsilon_b = \max \{ \epsilon_{1,b}/\overline{\beta}, \epsilon_{2,b}/\overline{\mu} \} \). In the following we derive the relation between \( \hat{\rho}_{ss,i} \) and \( \hat{\ell}_{0,i} \) from the equations which determine \( \overline{\rho}_{ss,i}, \hat{\rho}_{i,b}, \overline{\ell}_{0,i}, \) and \( \hat{k}_{i,b} \).

We first investigate \( \overline{\rho}_{ss,i} \) and \( \hat{\rho}_{i,b} \). Substituting Eq. (3.36) with \( \chi = 0 \) and Eq. (3.37) into the right eigenvalue equation \( \mathcal{K}_r(\alpha)\hat{\rho}_{ss,i}(\alpha) = 0 \), we obtain

\[
\mathcal{K}_r\overline{\rho}_{ss,i} = 0 \tag{3.40}
\]

in \( O(\epsilon^0) \), and

\[
\mathcal{K}_r\hat{\rho}_{i,b} + \overline{\ell}_{0,i} \mathcal{K}_r\overline{\rho}_{ss,i} = 0 \tag{3.41}
\]

in \( O(\epsilon_i\epsilon_b) \) with \( i = 1, 2 \). Because Eq. (3.40) is identical to the steady state equation for the equilibrium case, we have

\[
\overline{\rho}_{ss,i} = \hat{\rho}_{gc}(\overline{\beta}, \overline{\mu}). \tag{3.42}
\]

Using this result, we rewrite the second term on the LHS of Eq. (3.41) as

\[
\overline{\ell}_{0,i} \mathcal{K}_r\overline{\rho}_{ss,i} = v\mathcal{L}_{b,r} \left( \frac{\partial(\beta_b \hat{H}_S - \beta_b \mu_b \hat{N}_S)}{\partial \alpha_{i,b}} \bigg|_{\alpha_{i,b} = \overline{\alpha}_i} \hat{\rho}_{gc}(\overline{\beta}, \overline{\mu}) \right), \tag{3.43}
\]

where \( \mathcal{L}_{b,r} := \mathcal{L}_{b,r}(\alpha_S, \overline{\alpha}_B) \). We can derive this equation from the \( \alpha_{i,b} \)-derivative of the eigenvalue equation \( \mathcal{L}_{b,r} \hat{\rho}_{gc}(\beta_b, \beta_b \mu_b) = 0 \). Therefore we can rewrite Eq. (3.41) as

\[
\mathcal{K}_r\hat{\rho}_{i,b} + v\mathcal{L}_{b,r} \left( \hat{H}_S \hat{\rho}_{gc}(\overline{\beta}, \overline{\mu}) \right) = 0, \tag{3.44}
\]

\[
\mathcal{K}_r\hat{\rho}_{2,b} - v\mathcal{L}_{b,r} \left( \hat{N}_S \hat{\rho}_{gc}(\overline{\beta}, \overline{\mu}) \right) = 0. \tag{3.45}
\]

To proceed further, we note that the following relation holds for any \( \hat{Y} \in \mathbf{B} \):

\[
\mathcal{L}_{b,r}(\hat{Y} \hat{\rho}_{gc}(\overline{\beta}, \overline{\mu})) = (\mathcal{L}_{b,r}^\dagger \hat{Y}) \hat{\rho}_{gc}(\overline{\beta}, \overline{\mu}), \tag{3.46}
\]

where \( \hat{\rho}_{gc}^{-1}(\overline{\beta}, \overline{\mu}) := Z_{gc}(\overline{\beta}, \overline{\mu})e^{-\hat{H}_S - \beta_b \mu_b \hat{N}_S} \). Equation (3.46) is the detailed balance condition for the QMME [28]. We can derive this equation with the help of the KMS condition (2.32). By using Eq. (3.46), we rewrite Eqs. (3.44) and (3.45) as

\[
\mathcal{K}_r\hat{\rho}_{i,b} + \hat{\rho} \left( \mathcal{L}_{b,r}^\dagger \hat{H}_S \right) \hat{\rho}_{gc}(\overline{\beta}, \overline{\mu}) = 0, \tag{3.47}
\]

\[
\mathcal{K}_r\hat{\rho}_{2,b} - \hat{\rho} \left( \mathcal{L}_{b,r}^\dagger \hat{N}_S \right) \hat{\rho}_{gc}(\overline{\beta}, \overline{\mu}) = 0. \tag{3.48}
\]

Multiplying \( \hat{\rho}_{gc}^{-1}(\overline{\beta}, \overline{\mu}) \) from the right and taking the time reversal, we obtain

\[
\hat{\theta} \left( \mathcal{K}_r\hat{\rho}_{i,b} \right) \hat{\theta}^{-1} \hat{\rho}_{gc}^{-1}(\overline{\beta}, \overline{\mu}) + \hat{v} \mathcal{L}_{b,r}^\dagger \hat{H}_S = 0, \tag{3.49}
\]

\[
\hat{\theta} \left( \mathcal{K}_r\hat{\rho}_{2,b} \right) \hat{\theta}^{-1} \hat{\rho}_{gc}^{-1}(\overline{\beta}, \overline{\mu}) - \hat{v} \mathcal{L}_{b,r}^\dagger \hat{N}_S = 0, \tag{3.50}
\]

where we used \( \mathcal{L}_{b,r}^\dagger = \mathcal{L}_{b,r}^\dagger, \hat{H}_S = \hat{H}_S, \) and \( \hat{N}_S = \hat{N}_S \). Furthermore, we can rewrite the first terms on the LHS as

\[
\hat{\theta} \left( \mathcal{K}_r\hat{\rho}_{i,b} \right) \hat{\theta}^{-1} \hat{\rho}_{gc}^{-1}(\overline{\beta}, \overline{\mu}) = \mathcal{K}_r^\dagger \left[ \hat{\theta} \hat{\rho}_{i,b} \hat{\theta}^{-1} \hat{\rho}_{gc}^{-1}(\overline{\beta}, \overline{\mu}) \right]. \tag{3.51}
\]
We can derive this equation from Eq. \(3.46\). Therefore, we can rewrite Eqs. \(3.49\) and \(3.50\) as

\[
\begin{align*}
\mathcal{K}_r^\dagger \left( \partial \hat{\rho}_{1,b} \hat{\theta}^{-1} \hat{\rho}_{gc}^{-1} (\beta, \beta \mu) \right) + v \mathcal{L}_{b,1}^\dagger \hat{N}_S &= 0, \quad (3.52) \\
\mathcal{K}_r^\dagger \left( \partial \hat{\rho}_{2,b} \hat{\theta}^{-1} \hat{\rho}_{gc}^{-1} (\beta, \beta \mu) \right) - v \mathcal{L}_{b,1}^\dagger \hat{N}_S &= 0. \quad (3.53)
\end{align*}
\]

We next investigate \( \ell_{0,r}^0 \) and \( \hat{k}_{i,b} \). We differentiate the left eigenvalue equation of \( \mathcal{K}_r^\dagger \) with respect to \( i \chi \) and set \( \chi = 0 \):

\[
\mathcal{K}_r^\dagger \left( \alpha \right) \hat{1} + \mathcal{K}_r^\dagger \left( \alpha \right) \hat{k}_{i,r} \left( \alpha \right) = - J_\sigma \left( \alpha \right) \hat{1},
\]

where we used \( \lambda_0^\ast \mid_{\chi=0} = 0, \partial \lambda_0^\ast / \partial (i \chi) \mid_{\chi=0} = - J_\sigma \), and \( \hat{\kappa}_0^\ast = \hat{1} \). In the NESSs close to equilibrium, we can write the average entropy flow \( J_\sigma \) in a quadratic form: \( J_\sigma = \sum_{i,j=1,2} \sum_{b,b'} L_{i,b,j,b'} \epsilon_{i,b} \epsilon_{j,b'} \) with a phenomenological coefficient \( L_{i,b,j,b'} \). This implies that the RHS of Eq. \(3.54\) is \( O(\epsilon^2) \). By substituting Eqs. \(3.39\) and \(3.38\) into Eq. \(3.54\), we obtain

\[
\mathcal{K}_r^\dagger \hat{1} + \mathcal{K}_r^\dagger \ell_{0,r}^0 = 0
\]

in \( O(\epsilon^0) \), and

\[
\partial_{i,b} \mathcal{K}_r^\dagger \hat{1} + \mathcal{K}_r^\dagger \ell_{i,b} + \partial_{b} \mathcal{K}_r^\dagger \ell_{0,r}^0 = 0
\]

in \( O(\epsilon, b) \) with \( i = 1, 2 \). Here, \( \mathcal{K}_r^\dagger := \mathcal{K}_r^\dagger \left( \alpha_S, \alpha_B \right) \), and \( \partial_{i,b} \mathcal{K}_r^\dagger := \left( \partial \mathcal{K}_r^\dagger / \partial \alpha_{i,b} \right) \mid_{\alpha_S = \alpha_B} \). Equation \(3.55\) is identical to Eq. \(3.23\), so that

\[
\ell_{0,r}^0 = - \beta \hat{H}_S + \beta \mu \hat{N}_S + c \hat{1}.
\]

We rewrite the first term on the LHS of Eq. \(3.56\) as

\[
\begin{align*}
\partial_{i,b} \mathcal{K}_r^\dagger \hat{1} &= v \frac{\partial (\mathcal{L}_{b,1}^\dagger)}{\partial \alpha_{i,b}} \mid_{\alpha_S = \alpha_B} \\
&= v \frac{\partial [\mathcal{L}_{b,1}^\dagger (\beta \hat{H}_S - \beta \mu \hat{N}_S)]}{\partial \alpha_{i,b}} \mid_{\alpha_S = \alpha_B} \\
&= v \partial_{b,b} \mathcal{K}_r^\dagger (\beta \hat{H}_S - \beta \mu \hat{N}_S) + v \mathcal{L}_{b,1}^\dagger \frac{\partial (\beta \hat{H}_S - \beta \mu \hat{N}_S)}{\partial \alpha_{i,b}} \mid_{\alpha_S = \alpha_B},
\end{align*}
\]

where \( \partial_{i,b} \mathcal{L}_{b,1}^\dagger := \left( \partial \mathcal{L}_{b,1}^\dagger / \partial \alpha_{i,b} \right) \mid_{\alpha_S = \alpha_B} \). Here, we used Eq. \(3.25\) in the second line. By using Eq. \(3.57\), we also rewrite the third term on the LHS of Eq. \(3.56\) as

\[
\begin{align*}
\partial_{i,b} \mathcal{K}_r^\dagger \left( - \beta \hat{H}_S + \beta \mu \hat{N}_S + c \hat{1} \right) &= - v \partial_{i,b} \mathcal{L}_{b,1}^\dagger \left( \beta \hat{H}_S - \beta \mu \hat{N}_S \right),
\end{align*}
\]

where we used the fact that \( \partial_{i,b} \mathcal{K}_r^\dagger \hat{1} = 0 \) holds (we can derive this by differentiating the left eigenvalue equation \( \mathcal{K}_r^\dagger \hat{1} = 0 \) with respect to \( \alpha_{i,b} \)). Equation \(3.59\) cancels out the first term of Eq. \(3.58\). Thus Eq. \(3.56\) yields

\[
\begin{align*}
\mathcal{K}_r^\dagger \ell_{1,b} + v \mathcal{L}_{b,1}^\dagger \hat{H}_S &= 0, \quad (3.60) \\
\mathcal{K}_r^\dagger \ell_{2,b} - v \mathcal{L}_{b,1}^\dagger \hat{N}_S &= 0. \quad (3.61)
\end{align*}
\]
Then, combining Eq. (3.52) with Eq. (3.60) and Eq. (3.53) with Eq. (3.61), we obtain
\[
\mathcal{K}_i^\dagger \left( k_{i,b} - \hat{\theta} \hat{\rho}_{i,b} \hat{\theta}^{-1} \hat{\rho}^{-1}_{\text{gc}} (\bar{\beta}, \bar{\beta} \mu) \right) = 0,
\]
for \( i = 1, 2 \). This results in
\[
k_{i,b} = \hat{\theta} \hat{\rho}_{i,b} \hat{\theta}^{-1} \hat{\rho}^{-1}_{\text{gc}} (\bar{\beta}, \bar{\beta} \mu) + c_{i,b} \hat{1},
\]
where \( c_{i,b} \) is a constant. Combining the results of Eqs. (3.37), (3.38), (3.42), (3.57), and (3.63), we obtain
\[
\hat{\theta}_0 (\alpha) = \ln \hat{\rho}_{\text{gc}} (\bar{\beta}, \bar{\beta} \mu) + \sum_b (\epsilon_{1,b} \hat{\rho}_{1,b} \hat{\theta}^{-1} + \epsilon_{2,b} \hat{\rho}_{2,b} \hat{\theta}^{-1}) \hat{\rho}^{-1}_{\text{gc}} (\bar{\beta}, \bar{\beta} \mu) + c \hat{1} + O(\epsilon^2)
\]
\[
= \ln \hat{\theta} \hat{\rho}_{\text{ss},r} (\alpha) \hat{\theta}^{-1} + c \hat{1} + O(\epsilon^2),
\]
where \( c \) is a constant. Finally, substituting this result into the vector potential, we obtain Eq. (3.29):
\[
-\text{Tr}_S \left[ \hat{\theta}_0^\dagger (\alpha) \frac{\partial \hat{\rho}_{\text{ss},r} (\alpha)}{\partial \alpha} \right] = -\text{Tr}_S \left[ \left( \ln \hat{\theta} \hat{\rho}_{\text{ss},r} (\alpha) \hat{\theta}^{-1} \right) \frac{\partial \hat{\rho}_{\text{ss},r} (\alpha)}{\partial \alpha} \right] + O(\epsilon^2),
\]
where we used \( \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r}}{\partial \alpha} \right] = 0 \).

**Derivation of Eq. (3.30)**

The difference between the RHS and LHS of Eq. (3.30) is written as
\[
- \frac{\partial}{\partial \alpha} S_{\text{sym}} (\hat{\rho}_{\text{ss},r} (\alpha)) - \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r} (\alpha)}{\partial \alpha} \ln \hat{\rho}_{\text{ss},r} (\alpha) \right]
\]
\[
= \frac{1}{2} \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r}}{\partial \alpha} (\ln \hat{\rho}_{\text{ss},r} - \ln \hat{\rho}_{\text{ss},r}) \right] + \frac{1}{2} \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r}}{\partial \alpha} \right] + \frac{1}{2} \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r}}{\partial \alpha} \hat{\rho}_{\text{ss},r} \hat{\rho}_{\text{ss},r} \right]. \tag{3.65}
\]
The second term on the RHS vanishes because of the normalization condition. From Eq. (3.37) and the time-reversal invariance of \( \hat{\rho}_{\text{ss},r} = \hat{\rho}_{\text{gc}} (\bar{\beta}, \bar{\beta} \mu) \), we can show \( \hat{\rho}_{\text{ss},r} - \hat{\rho}_{\text{ss},r} = \epsilon \hat{\psi} + O(\epsilon^2) \). Therefore, we have
\[
\ln \hat{\rho}_{\text{ss},r} - \ln \hat{\rho}_{\text{ss},r} = \ln (\hat{\rho}_{\text{ss},r} + \epsilon \hat{\psi}) - \ln \hat{\rho}_{\text{ss},r} + O(\epsilon^2)
\]
\[
= \epsilon \hat{\rho}_{\text{ss},r} \hat{\psi} + O(\epsilon^2). \tag{3.66}
\]
Using this result, we can evaluate the first term on the RHS of (3.65) as
\[
\frac{1}{2} \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r}}{\partial \alpha} (\ln \hat{\rho}_{\text{ss},r} - \ln \hat{\rho}_{\text{ss},r}) \right] = \frac{\epsilon}{2} \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r} \hat{\psi} \hat{\rho}_{\text{ss},r} \hat{\psi} \hat{\rho}_{\text{ss},r}^{-1}}{\partial \alpha} \right] + O(\epsilon^2)
\]
\[
= \epsilon \text{Tr}_S \left[ \frac{\partial (\hat{\theta} \hat{\rho}_{\text{ss},r} \hat{\theta}^{-1}) \hat{\theta} \hat{\psi} \hat{\theta}^{-1} \hat{\rho}_{\text{ss},r} \hat{\psi} \hat{\rho}_{\text{ss},r}^{-1}}{\partial \alpha} \right] + O(\epsilon^2)
\]
\[
= - \frac{\epsilon}{2} \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r} \hat{\psi} \hat{\rho}_{\text{ss},r}^{-1}}{\partial \alpha} \right] + O(\epsilon^2), \tag{3.67}
\]
where we used that \( \text{Tr}_S \hat{Y} = \text{Tr}_S \hat{\theta} \hat{Y} \hat{\theta}^{-1} \) if \( \text{Tr}_S \hat{Y} \) is real in the second line, and \( \hat{\theta} \hat{\psi} \hat{\theta}^{-1} = \hat{\psi} = -\hat{\psi} \) in the third line. We can evaluate the third term on the RHS of (3.65) as
\[
\frac{1}{2} \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r} \hat{\psi} \hat{\rho}_{\text{ss},r}^{-1}}{\partial \alpha} \hat{\psi} \hat{\rho}_{\text{ss},r} \hat{\psi} \hat{\rho}_{\text{ss},r}^{-1} \right] = \frac{1}{2} \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r}}{\partial \alpha} (\hat{\psi} \hat{\rho}_{\text{ss},r} + \epsilon \hat{\psi}) \hat{\rho}_{\text{ss},r} \hat{\psi} \hat{\rho}_{\text{ss},r}^{-1} \right] + O(\epsilon^2)
\]
\[
= \frac{\epsilon}{2} \text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r} \hat{\psi} \hat{\rho}_{\text{ss},r}^{-1}}{\partial \alpha} \right] + O(\epsilon^2), \tag{3.68}
\]
where in the second line we used \( \text{Tr}_S [\frac{\partial \hat{\rho}_{\text{ss},r}}{\partial \alpha}] = 0 \) and \( \epsilon \hat{\rho}_{\text{ss},r}^{-1} = \epsilon \hat{\rho}_{\text{ss},r}^{-1} + O(\epsilon^2) \).

Substituting these results into the first and third terms on the RHS of (3.65), we obtain
\[
\frac{\partial}{\partial \alpha} S_{\text{sym}} (\hat{\rho}_{\text{ss},r} (\alpha)) = -\text{Tr}_S \left[ \frac{\partial \hat{\rho}_{\text{ss},r} (\alpha)}{\partial \alpha} \ln \hat{\rho}_{\text{ss},r} (\alpha) \right] + O(\epsilon^2). \tag{3.69}
\]
Derivation of Eq. (3.31)

We note that the symmetrized von Neumann entropy is written as

\[ S_{\text{sym}}(\hat{\rho}) = \frac{1}{2} \left\{ S_{\nu}^{\text{vN}}(\hat{\rho}) + \hat{S}(\hat{\rho}) \right\}, \quad (3.70) \]

where \( \hat{S}(\hat{\rho}) := -\text{Tr}_{S} \left[ \hat{\rho} \ln \hat{\rho} \right] \). In the following, we derive

\[ S_{\nu}^{\text{vN}}(\hat{\rho}_{ss}) = S_{\nu}^{\text{vN}}(\hat{\rho}_{ss,r}) + O(v^2), \quad (3.71) \]

\[ \hat{S}(\hat{\rho}_{ss}) = \hat{S}(\hat{\rho}_{ss,r}) + O(v^2). \quad (3.72) \]

These lead to Eq. (3.31).

First, we consider \( S_{\nu}^{\text{vN}} \). We represent \( \hat{\rho}_{ss,r} \) by a matrix in the basis of the eigenstates of \( \hat{H}_S \). Then, because \( \hat{\rho}_{ss,r} \) is in \( P \), \( \hat{\rho}_{ss,r} \) is represented by a block diagonal matrix. Each block is in the degenerate subspace that has a single energy eigenvalue; i.e., the non-vanishing matrix elements are \( \{ \langle E_{\nu, n}|\hat{\rho}_{ss,r}|E_{\nu, n'} \rangle \}_\nu \). Therefore, if we take the appropriate linear combination \( |E_{\nu, l} \rangle \) of \( \{|E_{\nu, n} \rangle \}_n \), which is also an energy eigenstate, in each degenerate subspace, we can diagonalize \( \hat{\rho}_{ss,r} \). Using this diagonalizing basis, we have

\[ S_{\nu}^{\text{vN}}(\hat{\rho}_{ss,r}) = -\sum_{\nu, l} [\hat{\rho}_{ss,r}]_{\nu, l} \ln[\hat{\rho}_{ss,r}]_{\nu, l}, \quad (3.73) \]

where \( [\hat{\rho}_{ss,r}]_{\nu, l} := \langle E_{\nu, l} |\hat{\rho}_{ss,r}|E_{\nu, l} \rangle \). We assume that there is no degeneracy in the eigenvalues of \( \hat{\rho}_{ss,r} \); i.e., \( [\hat{\rho}_{ss,r}]_{\nu, l} \neq [\hat{\rho}_{ss,r}]_{\nu', l'} \) if \( \nu \neq \nu' \) or \( l \neq l' \).

As we showed in Sec 3.1, we can write the steady state without the RWA as

\[ \hat{\rho}_{ss} = \hat{\rho}_{ss,r} + v\hat{\eta} + O(v^2). \quad (3.74) \]

We evaluate the eigenvalue \( e_{\nu, l} \) of \( \hat{\rho}_{ss} \) by regarding \( \hat{\rho}_{ss,r} \) as the unperturbed part and \( v\hat{\eta} \) as the perturbation:

\[ e_{\nu, l} = [\hat{\rho}_{ss,r}]_{\nu, l,v, l'} + v\Delta_{\nu, l} + O(v^2). \quad (3.75) \]

Because we assume the non-degeneracy in the eigenvalues of \( \hat{\rho}_{ss,r} \), we can use the perturbation theory for the non-degenerate case to obtain

\[ \Delta_{\nu, l} = \langle E_{\nu, l} |\hat{\eta}|E_{\nu, l} \rangle. \quad (3.76) \]

Because \( \hat{\eta} \in Q \), as we showed in the previous subsection, \( \Delta_{\nu, l} \) vanishes. Therefore we obtain

\[ S_{\nu}^{\text{vN}}(\hat{\rho}_{ss}) = -\sum_{\nu, l} e_{\nu, l} \ln e_{\nu, l} \]

\[ = -\sum_{\nu, l} [\hat{\rho}_{ss,r}]_{\nu, l} \ln[\hat{\rho}_{ss,r}]_{\nu, l} + O(v^2). \quad (3.77) \]

Comparing this with Eq. (3.73), we derive Eq. (3.71).

Next, we consider \( \hat{S} \). We note that \( \hat{\rho}_{ss,r} \) is the steady solution of \( \hat{K}_r \); i.e., \( \hat{K}_r\hat{\rho}_{ss,r} = 0 \). Similarly to the case of \( \hat{\rho}_{ss,r} \) we can show \( \hat{\rho}_{ss,r} \in P \). Therefore by the same argument as the above, we can diagonalize \( \hat{\rho}_{ss,r} \) by taking the appropriate energy eigenstates \( |E_{\nu, l} \rangle \). We note that \( \{|E_{\nu, l} \rangle \}_l \) is different from \( \{|E_{\nu, l} \rangle \}_l \) in the above, and that \( \hat{\rho}_{ss,r} \) is not diagonalized in the basis of \( \{|E_{\nu, l} \rangle \}_l \). Using this basis, we have

\[ \hat{S}(\hat{\rho}_{ss,r}) = -\sum_{\nu, l} [\hat{\rho}_{ss,r}]_{\nu, l} \ln[\hat{\rho}_{ss,r}]_{\nu, l}, \quad (3.78) \]
where \( [\tilde{\rho}_{ss,i},_{\nu}l] = \langle E_{\nu}, l | \tilde{\rho}_{ss,i} | E_{\nu}, l \rangle \) and \( [\tilde{\rho}_{ss,i},_{\nu}l] = \langle E_{\nu}, l | \tilde{\rho}_{ss,i} | E_{\nu}, l \rangle \).

The time reversal \( \tilde{\rho}_{ss} \) of the steady state \( \hat{\rho}_{ss} \) without the RWA is the steady solution of \( \tilde{K} \). Taking the time reversal of Eq. (3.74), we have
\[
\tilde{\rho}_{ss} = \tilde{\rho}_{ss} + v\bar{\eta} + O(v^2).
\]
(3.79)

By an argument similar to that in the previous subsection, we can show \( \bar{\eta} \in \mathbb{Q} \). Therefore, as in the above, we can evaluate the eigenvalue \( \tilde{e}_{\nu,l} \) of \( \tilde{\rho}_{ss} \) as
\[
\tilde{e}_{\nu,l} = [\tilde{\rho}_{ss,r},_{\nu}l] + O(v^2).
\]
(3.80)

Therefore we obtain
\[
\tilde{S}(\tilde{\rho}_{ss}) = -\sum_{\nu,l} [\tilde{\rho}_{ss,l},_{\nu}l] \ln \tilde{e}_{\nu,l}
\]
\[
= -\sum_{\nu,l} [\tilde{\rho}_{ss,l},_{\nu}l] \ln [\tilde{\rho}_{ss,l},_{\nu}l] + O(v^2).
\]
(3.81)

Comparing this with Eq. (3.78), we derive Eq. (3.72).

4 Example: Spinless Electron System in Quantum Dots

In this section, we investigate the excess entropy production for quasistatic operations in a simple electron model to demonstrate the general results in the previous section. We consider a spinless electron system in \( N \) quantum dots connected to \( N_B \) electron reservoirs. An example of the system is illustrated in Fig. 1.

We assume that each dot has a single level. The Hamiltonian of the total system is given in the form of Eq. (2.2), where
\[
\hat{H}_S = \sum_{i=1}^{N} \varepsilon_i \hat{d}_i^\dagger \hat{d}_i + \sum_{(ii')} t_{ii'} (\hat{d}_i^\dagger \hat{d}_{i'} + \text{h.c.}) + U \sum_{(ii')} \hat{d}_i^\dagger \hat{d}_i \hat{d}_{i'}^\dagger \hat{d}_{i'},
\]
(4.1)
\[
\hat{H}_b = \sum_k \hbar \Omega_{bk} \hat{c}_{bk}^\dagger \hat{c}_{bk},
\]
(4.2)
\[
\hat{H}_{Sb} = \sum_k \sum_{i=1}^{N} \xi_{ibk} (\hat{d}_i^\dagger \hat{c}_{bk} + \text{h.c.}).
\]
(4.3)
Here, \( \varepsilon_i \) is the energy level of the \( i \)th dot, \( t_{i'i'} \) is the transition probability amplitude between the \( i \)th and \( i' \)th dots, \( U \) is the interdot potential energy, \( \hbar \Omega_{bk} \) is the energy of the \( k \)th mode in the \( b \)th reservoir, and \( \xi_{i'bk} \) is the transition probability amplitude between the \( i' \)th dot and the \( k \)th mode in the \( b \)th reservoir.

In the second and third terms in the RHS of Eq. (4.1), the sum is taken over the neighboring dots. The creation \( \hat{b}_k \) and annihilation \( \hat{a}_k \) operators of an electron in the \( i \)th dot (\( k \)th mode in the \( b \)th reservoir) satisfies the canonical anti-commutation relations: \( \{ \hat{b}_k, \hat{b}_{k'}^\dagger \} = \delta_{kk'}, \{ \hat{b}_k, \hat{d}_{k'}^\dagger \} = \{ \hat{d}_k, \hat{d}_{k'}^\dagger \} = 0, \{ \hat{b}_k, \hat{d}_{k'} \} = 0, \{ \hat{d}_k, \hat{d}_{k'} \} = 0 \). We assume that the \( b \)th reservoir is in the equilibrium state with inverse temperature \( \beta_b \) and chemical potential \( \mu_b \) (\( b = 1, 2, \ldots, N_B \)). Note that the control parameters are \( \{ \varepsilon_i \}, \{ t_{i'i'} \}, U \) (system parameters) and \( \{ \beta_b, \mu_b \} \) (reservoir parameters) in this model.

4.1 RWA analysis of noninteracting system \( (U = 0) \)

We first analyze the noninteracting case \( (U = 0) \) using the RWA. In this case, by using a linear transformation of the operators \( \hat{d}_i^\dagger, \hat{d}_i, \)

\[
\hat{d}_i^\dagger = \sum_{j=1}^N W_{ij}^* \hat{a}_j^\dagger, \\
\hat{d}_i = \sum_{j=1}^N W_{ij} \hat{a}_j,
\]

we can diagonalize the system Hamiltonian \( \hat{H}_S \):

\[
\hat{H}_S = \sum_{j=1}^N \hbar \omega_j \hat{a}_j^\dagger \hat{a}_j,
\]

where \( \hbar \omega_j \) is the \( j \)th mode energy of the noninteracting system. We can also write the eigenstate \( |E_i\rangle \) of \( \hat{H}_S \) as \( |E_i\rangle = \bigotimes_j |\nu_j\rangle \). Here, \( |\nu_j\rangle \) is either of the empty state \( |0_j\rangle \) or singly-occupied state \( |1_j\rangle \) in the \( j \)th mode Hilbert space \( (\hat{a}_j |0_j\rangle = 0 \text{ and } |1_j\rangle = \hat{a}_j^\dagger |0_j\rangle). \)

By the above transformation, we can rewrite the system-reservoir coupling Hamiltonian as

\[
\hat{H}_{SB} = \sum_{k,j=1}^N (\zeta_{jbk} \hat{a}_j^\dagger \hat{c}_{bk} + \text{h.c.}),
\]

where \( \zeta_{jbk} = \sum_i W_{ij}^* \xi_{ik} \). Note that \( \zeta_{jbk} \) depends on the control parameters although \( \xi_{ik} \) is not included in the control parameters, because \( W_{ij} \) depends on \( \{ \varepsilon_i \} \) and \( \{ t_{i'i'} \} \).

Now we take the correspondence between the present model and the generic model in Sec. 2. Equation (4.7) is in the form of Eq. (4.11), where \( \hat{X}_{h,l} \rightarrow \hat{a}_j \) and \( \hat{B}_{h,l} \rightarrow \sum_k \zeta_{jbk} \hat{c}_{bk} \). The spectral functions of the reservoir given in Eqs. (4.29) and (4.30) read

\[
\Phi_{b,j'j}^+(\omega) = 2\pi \sum_k |\zeta_{jbk}|^2 \delta(\Omega_{bk} - \omega) f^{b,j'}_k(\omega) \delta_{jj'},
\]

\[
\Phi_{b,j'j}^-(\omega) = 2\pi \sum_k |\zeta_{jbk}|^2 \delta(\Omega_{bk} - \omega) f^{b,j'}_k(\omega) \delta_{jj'},
\]

where \( f^{b,j'}_k(\omega) = 1/(1 + e^{\beta_b(\omega - \mu_b)}) \) is the Fermi distribution function and \( f^{b}_{k}(\omega) = 1 - f_{k}^{b,\dagger}(\omega) \). Note that \( \Phi_{b,j'j}^{\pm}(\omega) \) depends on the system parameters as well as the reservoir parameters since \( \zeta_{jbk} \) depends on \( \{ \varepsilon_i \} \) and \( \{ t_{i'i'} \} \).
As is mentioned in the previous section, it is sufficient to investigate $\mathcal{PK}_r\mathcal{P}$ to calculate the excess entropy production $\langle \sigma \rangle^{\alpha}$ for quasistatic operations. Furthermore, because $\hat{H}_S$ is non-degenerate in the present model, each eigenspace is spanned only by a single energy eigenstate $|E_{\nu}\rangle$. Using the above-mentioned facts, we obtain the matrix representation of the GQMME generator for the noninteracting model within the RWA in the following form:

$$\begin{align*}
\langle E_{\nu}|(\mathcal{K}_r^\lambda|E_{\nu}\rangle|E_{\nu}\rangle\rangle = \sum_{j=1}^{N} I_2 \otimes \cdots \otimes I_2 \otimes [\mathcal{K}_j^\lambda] \otimes I_2 \otimes \cdots \otimes I_2,
\end{align*}$$

(4.10)

where $I_2$ is the $2 \times 2$ identity matrix, and

$$[\mathcal{K}_j^\lambda] = -\frac{u^2}{\hbar^2} \left( -\sum_b \Phi_{b,jj}^+(\omega_j) e^{i\chi_b(\hbar\omega_j - \mu_b)} - \sum_b \Phi_{b,jj}^-(\omega_j) e^{-i\chi_b(\hbar\omega_j - \mu_b)} \right).$$

(4.11)

Accordingly we can also decompose $\hat{r}_{0,r}^\lambda$ and $\hat{r}_{0,r}^\lambda$ as $\hat{r}_{0,r}^\lambda = \bigotimes_j \hat{r}_{0,j}^\lambda$ and $\hat{r}_{0,r}^\lambda = \bigotimes_j \hat{r}_{0,j}^\lambda$, where $\hat{r}_{0,j}^\lambda$ and $\hat{r}_{0,j}^\lambda$ are respectively the left and right eigenvectors of $\mathcal{K}_j^\lambda$, corresponding to the eigenvalue with the maximum real part. By diagonalizing Eq. (4.11), we obtain

$$\begin{align*}
\begin{pmatrix}
0_j | \hat{r}_{0,j}^\lambda \rangle | 0_j \rangle \\
1_j | \hat{r}_{0,j}^\lambda \rangle | 1_j \rangle 
\end{pmatrix} &= \begin{pmatrix}
w_j^+(\chi) \\
w_j^-(\chi)
\end{pmatrix},
\end{align*}$$

(4.12)

$$\begin{align*}
\begin{pmatrix}
0_j | \hat{r}_{0,j}^\lambda \rangle | 0_j \rangle \\
1_j | \hat{r}_{0,j}^\lambda \rangle | 1_j \rangle 
\end{pmatrix} &= C_j(\chi) \begin{pmatrix}
1 \\
w_j^-(\chi)
\end{pmatrix},
\end{align*}$$

(4.13)

where

$$w_j^\pm(\chi) = \frac{\sum_b \{ \Phi_{b,jj}^+(\omega_j) - \Phi_{b,jj}^-(\omega_j) \}}{2 \sum_b \Phi_{b,jj}^\pm(\omega_j) e^{\pm i\chi_b(\hbar\omega_j - \mu_b)}} + \sqrt{D_j(\chi)},$$

(4.14)

$$D_j(\chi) = \left[ \sum_b \{ \Phi_{b,jj}^+(\omega_j) - \Phi_{b,jj}^-(\omega_j) \} \right]^2 + 4 \sum_{bb'} \Phi_{b,jj}^+(\omega_j) \Phi_{b',jj}^-(\omega_j) e^{i\chi_b(\hbar\omega_j - \mu_b)} e^{i\chi_{b'}(\hbar\omega_j - \mu_{b'})}.$$

(4.15)

From the normalization condition for $\chi = 0$, $\text{Tr}_{S} r_{0,r}^\lambda = 1$, we have $C_j(0) = \sum_b \Phi_{b,jj}^-(\omega_j)/\gamma_j(\omega_j)$, where $\gamma_j(\omega) := \sum_b \gamma_{bj}(\omega)$ and $\gamma_{bj}(\omega) := \Phi_{b,jj}^+(\omega) + \Phi_{b,jj}^-(\omega)$. We thus obtain the vector potential for the excess entropy production in Eq. (3.33) for this noninteracting model:

$$-\text{Tr}_{S} \left[ \hat{r}_{0,r}^\lambda \frac{\partial \hat{\rho}_{se,r}}{\partial \alpha} \right] = \sum_{j=1}^{N} \frac{\partial w_j^+(\chi)}{\partial (i\chi)} \bigg|_{\chi=0} \frac{\partial (C_j(0)w_j^- (0))}{\partial \alpha}$$

$$= \sum_{j=1}^{N} \frac{\sum_b \beta_b(\hbar\omega_j - \mu_b)\gamma_{bj}(\omega_j)}{\gamma_j(\omega_j)} \frac{\partial}{\partial \alpha} \left( \sum_b \Phi_{b,jj}^+(\omega_j) \right).$$

(4.16)

We can show $\gamma_{bj}(\omega) = 2\pi \sum_k |\zeta_{ijk}|^2 \delta(\Omega_{jk} - \omega)$ and $\Phi_{b,jj}^+(\omega) = \gamma_{bj}(\omega)f^+(\omega)$ with the aid of Eqs. (4.8) and (1.19).

As a special case of the external operations, we investigate the quasistatic operation where we modulate only the parameters of a single reservoir, say $\beta_L$ and $\mu_L$. In this case, we can write the vector potential (4.16) as the derivative of a scalar function $F$ of $\beta_L$ and $\mu_L$. That is,

$$-\text{Tr}_{S} \left[ \hat{r}_{0,r}^\lambda \frac{\partial \hat{\rho}_{se,r}}{\partial \alpha_L} \right] = \frac{\partial F(\beta_L, \mu_L)}{\partial \alpha_L},$$

(4.17)
Therefore, for this special case of the quasistatic operation in the noninteracting model, the excess entropy production is written as the difference of the initial and final values of the scalar function $F$. We note that this scalar function $F$ is not equal to the von Neumann entropy $S_{\text{vn}}(\hat{\rho}_{ss,r})$ of the steady state ($S_{\text{vn}}(\hat{\rho}_{ss,r}) = S_{\text{s}}(\hat{\rho}_{ss,r})$ for non-degenerate $\hat{H}$, which is given by

$$S_{\text{vn}}(\hat{\rho}_{ss,r}) = -\sum_{j=1}^{N} \sum_{s=\pm} \frac{b \gamma_{b}\gamma(b)\{f_{L}^{+}(\omega) \ln f_{L}^{+}(\omega) + f_{L}^{-}(\omega) \ln f_{L}^{-}(\omega)\}}{\gamma_{J}(\omega)^{2}}. \quad (4.18)$$

We can derive this from Eq. (4.13) with $\chi = 0$.

4.2 Analysis without RWA

Next, we numerically analyze the model (4.1)-(4.3) without using the RWA. We here restrict our interest to the four-dot system $(i = 1, 2, 3, 4)$ with two reservoirs $(b = L, R)$, as illustrated in Fig. 1. In this case, the coupling constant $\xi_{ibk}$ in Eq. (4.3) is given by $\xi_{ilk} = \xi_{1lk}\delta_{i1}$ and $\xi_{ilk} = \xi_{4lk}\delta_{i4}$. The spectral functions of the reservoirs are defined as $\Gamma_{L}(\omega) = 2\pi \sum_{k} |\xi_{1lk}|^{2}\delta(\Omega_{L} - \omega)$ and $\Gamma_{R}(\omega) = 2\pi \sum_{k} |\xi_{4lk}|^{2}\delta(\Omega_{R} - \omega)$. We also assume the wide band limit and the symmetric coupling, i.e., $\Gamma_{L}(\omega) = \Gamma_{R}(\omega) = \text{const.} =: \Gamma$.

We use the geometrical formula (3.3) to obtain the excess entropy production $\langle \sigma \rangle_{\text{ex}}$ for the quasistatic operations. That is, we numerically solve the eigenvalue problem of the GQMME generator $\mathcal{K}$, calculate the vector potential $-\text{Tr}_{S} \left[ \sigma_{0}^{f} \partial \hat{\rho}_{ss}/\partial \alpha \right]$, and integrate it along the curve of the operation in the parameter space.

Excess Entropy Production for Modulation of Reservoir Parameters of Single Reservoir

We here calculate $\langle \sigma \rangle_{\text{ex}}$ for an operation of the parameters $\beta_{L}$ and $\mu_{L}$ of the reservoir L. We set the initial condition to $\mu_{L}^{0} = \mu_{R}$ and $\beta_{L}^{0} = \beta_{R}^{0} =: \beta$ (equilibrium condition), and the final condition to $\mu_{L}^{f} > \mu_{R}$ and $\beta_{L}^{f} = \beta_{R}$ (nonequilibrium condition). We calculate $\langle \sigma \rangle_{\text{ex}}$ for four paths (denoted by A, B, C, and D) connecting these two conditions in the parameter space which are illustrated in the inset of Fig. 2(a). For the paths B–D, we set middle conditions to $\beta_{L}^{0} > \beta_{R}$. In Fig. 2 we plot the results as a function of the difference between the initial and final values of $\mu_{L}$ (with fixing the value of $\beta_{R}^{0}$). We show the results for noninteracting ($\beta U = 0$) and interacting ($\beta U = 8$) systems in Figs. 2(a) and (b), respectively.

In Fig. 2(a), we observe that the data of $\langle \sigma \rangle_{\text{ex}}$ for all of the paths agree in the whole range of $\mu_{L}^{0} - \mu_{L}^{f}$. Furthermore, the change $\Delta F$ of the scalar function given in Eq. (4.18) (plotted as a solid line), quantitatively agrees with these data. These results indicates that the statement in the RWA analysis on the noninteracting system described around Eqs. (4.17) and (4.18) in the previous subsection is valid even in the non-RWA analysis.

For interacting systems, this statement is not valid. We can clearly see this breakdown in Fig. 2(b), where the results for the different paths show different behaviors in the range of large $\beta(\mu_{L}^{f} - \mu_{L}^{0}) (\geq 4)$. 

\footnote{For $U = 0$, the spectral function $\Gamma_{b}(\omega)$ is related with $\gamma_{b}(\omega)$ in the previous subsection:

\begin{align*}
\gamma_{L}(\omega) &= |W_{ij}|^{2}\Gamma_{L}(\omega), \\
\gamma_{R}(\omega) &= |W_{ij}|^{2}\Gamma_{R}(\omega).
\end{align*}

Therefore, $\gamma_{b}(\omega)$ depends on the system parameters even in the wide band limit, since $W_{ij}$ does.}
In contrast, for small $\beta(\mu^f_L - \mu^i_L)$ ($\ll 1$), the results for all of the paths are nearly equal. Moreover, in this range, these results almost agree with the change of the von Neumann entropies $S_{vN}(\hat{\rho}_{ss,t})$ between the initial and final steady states (calculated within the RWA and plotted as dashed lines) both in Figs. 2(a) and (b). These observations are consistent with the fact that the extended Clausius equality holds in the weakly nonequilibrium regime.

In Fig. 2(b), we also show the results for the interacting system analyzed in the RWA (plotted as solid lines). To obtain these data, we numerically solve the eigenvalue problem of the RWA-GQMME generator $\mathcal{K}^\chi$ instead of the non-RWA-GQMME generator $\mathcal{K}^\chi$. We see that all of the data agree with those without the RWA for the corresponding paths. This result is consistent with the equivalence between the RWA and non-RWA shown in Sec. 3.1.

**Excess Entropy Production for Cycle Process of Dot Energy Levels**

We next investigate the excess entropy production for system parameter operations. In Fig. 3, we plot $\langle \sigma \rangle^{ex}$ for the cycle operations of the dot energy levels under a nonequilibrium condition of $\Delta \mu := \mu_L - \mu_R > 0$ (and the same temperature condition $\beta_L = \beta_R =: \beta$). We modulate the energy levels along the circle given by

$$\varepsilon_1 = \varepsilon_2 = \varepsilon_c + \varepsilon_r \cos \phi,$$

$$\varepsilon_3 = \varepsilon_4 = \varepsilon_c + \varepsilon_r \sin \phi,$$
\[ \beta (\mu_L - \mu_R) < \sigma \]

\[ \beta U = 0 \quad \textcircles{2} \]
\[ \beta U = 8 \quad \text{triangles} \]

Figure 3: (Color online) Excess entropy production in the four-dot system for a quasistatic cycle operation of the dot energy levels under the nonequilibrium condition of \( \mu_L > \mu_R \) with \( \beta_L = \beta_R = \beta \) [see Eqs. (4.20) and (4.21) for the detail of the operation]. The results for a noninteracting \( (\beta U = 0) \) circle and an interacting \( (\beta U = 8) \) triangles systems are plotted against \( \beta (\mu_L - \mu_R) \). The solid lines are the results analyzed within the RWA. The parameters are set to \( \beta \varepsilon_c = 10, \beta \varepsilon_r = 3, \beta t_{i'j} = 4 \) (for all \( i, i' \)), \( \beta \Gamma = 0.01 \), and \( \beta \mu_R = 4 \).

We observe that \( \langle \sigma \rangle_{\text{ex}} \approx 0 \) in the weakly nonequilibrium regime \( (\beta \Delta \mu \lesssim 1) \) for both the noninteracting \( (\beta U = 0) \) and interacting \( (\beta U = 8) \) systems. This indicates that the extended Clausius equality is valid in this regime. In the strongly nonequilibrium regime \( (\beta \Delta \mu \gtrsim 2) \), in contrast, \( \langle \sigma \rangle_{\text{ex}} \) takes nonzero values, which implies the failure of the extension of the Clausius equality with the excess entropy production (even in the noninteracting system).

We also show the results analyzed in the RWA in Fig. 3 (plotted as solid lines). To obtain these results, we use Eq. (4.16) for the noninteracting system, whereas for the interacting system we numerically solve the eigenvalue problem of the RWA-GQMME generator \( K^\chi \) instead of the non-RWA-GQMME generator \( K^\chi \). We see that the data within the RWA agree with those without the RWA in the whole range of \( \Delta \mu \). This result is consistent with the equivalence between the RWA and non-RWA shown in Sec. 3.1.

5 Summary and Discussions

For open quantum systems described by the QMME, we have derived a geometrical expression for the excess entropy production during an arbitrary quasistatic operation that connects two NESSs. In the derivation, we have used the technique of the full counting statistics and the adiabatic approximation. This result implies that the scalar thermodynamic potential for arbitrary NESSs cannot be defined from the excess entropy production for the quasistatic operation, and that the vector potential \( -\text{Tr}_S \left[ \partial_i^j (\partial \rho_{ss}/\partial \alpha) \right] \) plays a crucial role in the steady state thermodynamics (SST).

We have also shown that the result of the excess entropy production within the RWA is equivalent to that without the RWA. This is helpful for the investigation of the SST in the framework of the QMME,
because the RWA makes calculation easier (in particular, if the system Hamiltonian is non-degenerate, the form of the QMME is equivalent to that of the classical Markov jump process).

In the weakly nonequilibrium regime, with the aid of the RWA, we have derived the extended Clausius equality from the geometrical expression. This result extends the validity range of the equality derived in classical systems [16] [17] [24] and quantum heat conducting systems [25] to the systems described by the QMME (including electrical conducting systems).

As an example, we have investigated a spinless electron system in quantum dots. We have found that in the noninteracting systems there exists a scalar potential for the operation on a single reservoir, but that this is not valid in the interacting systems.

There are many issues to be studied in the future. One of the important issues is the way of constructing a thermodynamic potential in the SST. We are considering two directions. One is that we construct the thermodynamics by using the vector potential as the thermodynamic potential. For this purpose, it is important to investigate the thermodynamic structure from the geometrical viewpoint [35]. The other is that we restrict the class of systems of the SST to “macroscopic” systems. By the restriction, it may be possible to construct a scalar potential from the excess entropy production.

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