Theoretical studies on quantum pump and excess entropy production: Quantum master equation approach

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Abstract

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by Satoshi Nakajima

In this thesis, we considered quantum systems coupled to several baths. We supposed that the system state is governed by the quantum master equation (QME). We investigated the quantum pump and the excess entropy production. When the set of control parameters $\alpha = \{\alpha^n\}_n$ is modulated between times $t = 0$ and $t = \tau$, the average change of a *time-independent* observable O of the baths is given by

$$\langle \Delta o \rangle = \int_0^\tau dt\ i_O^{\mathrm{ss}}(\alpha_t) + \int_C d\alpha^n\ A_n^O(\alpha) + \langle \Delta o \rangle^{(\mathrm{na})}.$$

Here, the summation symbol for n is omitted, α_t is α at time t, C is the trajectory in the control parameter space, $i_O^{\mathrm{ss}}(\alpha_t)$ is the instantaneous steady current of O and $A_n^O(\alpha)$ is called the Berry-Sinitsyn-Nemenman (BSN) vector. $\langle \Delta o \rangle^{(\mathrm{na})}$ is a non-adiabatic term and order of ω/Γ where ω is the modulation frequency of the control parameters and Γ is the coupling strength between the system and the baths. If ω/Γ is sufficiently small, this pumping is called the quantum adiabatic pump. Similarly, the average entropy production σ under quasistatic ($\omega/\Gamma \to 0$) modulation is given by

$$\sigma = \int_0^\tau dt\ j_\sigma(\alpha_t) + \int_C d\alpha^n\ A_n^\sigma(\alpha).$$

Here, $j_\sigma(\alpha_t)$ is the instantaneous steady entropy production rate and $A_n^\sigma(\alpha)$ is called the BSN vector for entropy production. The second term of the right hand side (RHS) of the above equation is called the excess entropy production, σ_{ex}.

First, we investigated the quantum pump using the full counting statistics with quantum master equation (FCS-QME) approach. We studied the non-adiabatic effect and the showed that the general solution of the QME $\rho(t)$ is decomposed as $\rho(t) = \rho_0(\alpha_t) + \sum_{n=1}^\infty \rho^{(n)}(t) + \sum_{n=0}^\infty \tilde{\rho}^{(n)}(t)$. Here, $\rho_0(\alpha_t)$ is the instantaneous steady state of the QME, $\rho^{(n)}(t)$ and $\tilde{\rho}^{(n)}(t)$ are calculable and order $(\omega/\Gamma)^n$. $\tilde{\rho}^{(n)}(t)$ exponentially damps (like $e^{-\Gamma t}$) as a function of time. We showed that the generalized mater equation (GME) approach provides $\boldsymbol{p}(t) = \boldsymbol{p}_{(\mathrm{ss})}(t) + \delta\boldsymbol{p}(t)$ in the Born approximation. Here, $\boldsymbol{p}$ corresponds to the set of the diagonal components of ρ in the matrix representation by the energy eigenstates, $\boldsymbol{p}_{(\mathrm{ss})}(t)$ corresponds to $\rho_0(\alpha_t) + \sum_{n=1}^\infty \rho^{(n)}(t)$ and the term $\delta\boldsymbol{p}(t)$ originates from non-Markovian effects. We showed that the FCS-QME method provides $(n+1)$-th order pump current from $\rho^{(n)}(t)$. We showed that the quantum pump dose not occur in all orders of the pumping frequency when the system control parameters and the thermodynamic parameters (the temperatures and the chemical potentials of the baths) are fixed under the zero-bias condition.

Next, we studied the quantum adiabatic pump of the quantum dot (QD) system weakly coupled to two leads (L and R) using the FCS-QME. We confirmed the consistency between the FCS-QME approach and the GME approach for a QD of one quantum level with finite Coulomb interaction. We showed that the pumped charge and spin coming from the instantaneous steady current are not negligible when the thermodynamic parameters are not fixed to zero bias. To observe the spin effects, we considered collinear magnetic fields, which affect the spins through the Zeeman effect, with different amplitudes applying to the QDs (B_S) and the leads (B_L and B_R). We focused on the dynamic parameters (B_S, $B_{L/R}$ and the coupling strength between QDs and leads, $\Delta_{L/R}$) as control parameters. In one level QD with the Coulomb interaction U, we studied (B_L, B_S) pump and (Δ_L, B_S) pump for the non-interacting limit ($U = 0$) and the strong interaction limit ($U = \infty$) at zero-bias. The difference depending on U appeared through $n_U(sB_S)$ which is the average number of the electrons with spin s in the QD. For (B_L, B_S) pump, the energy dependences

of the line-width functions are essential. Moreover, we studied the (Δ_L, B_S) pump for finite U at zero-bias. The effect of U appeared through $n_U(sB_S)$. When half-filling condition satisfies, the charge pump does not occur.

We studied quantum diabatic pump for spinless one level QD coupled to two leads. We calculated $\{\rho^{(n)}(t)\}_{n=1}^5$, $\{\tilde{\rho}^{(n)}(t)\}_{n=1}^5$ and particle current up to 6th order and pumped particle numbers.

In the latter part of the thesis, we investigated the excess entropy production. In weakly nonequilibrium regime, we analyzed the BSN vector for the entropy production and showed

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

Here, Tr_S denotes the trace of the system, and ε is a measure of degree of nonequilibrium. $\rho_0^{(-1)}(\alpha)$ is the instantaneous steady state obtained from the QME with reversing the sign of the Lamb shift term. In general, the potential $\mathcal{S}(\alpha)$ such that $A_n^\sigma(\alpha) = \frac{\partial \mathcal{S}(\alpha)}{\partial \alpha^n} + \mathcal{O}(\varepsilon^2)$ dose not exist. This is the most important result of this thesis. The origins of the non-existence of the potential $\mathcal{S}(\alpha)$ are a quantum effect (the Lamb shift term) and the breaking of the time-reversal symmetry. The non-existence of the potential means that the excess entropy essentially depends on the path of the modulation. In contrast, if the system Hamiltonian is non-degenerate or the Lamb shift term is negligible, we obtain $\sigma_{\mathrm{ex}} = S_{\mathrm{vN}}(\rho_0(\alpha_\tau)) - S_{\mathrm{vN}}(\rho_0(\alpha_0)) + \mathcal{O}(\varepsilon^2\delta)$. Here, $S_{\mathrm{vN}}(\rho) = -\mathrm{Tr}_S[\rho \ln \rho]$ is the von Neumann entropy, and δ describes the amplitude of the change of the control parameters. For systems with time-reversal symmetry, there exists a potential $\mathcal{S}(\alpha)$, which is the symmetrized von Neumann entropy. Additionally, we pointed out that the expression of the entropy production obtained in the classical Markov jump process is different from our result and showed that these are approximately equivalent only in the weakly nonequilibrium regime.

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Chapter 1

Introduction

1.1 Background

The properties of the isolated static quantum system in the equilibrium state have been studied deeply. The studies of more general systems are important, however, uncompleted and are actively being studied. This thesis focus on the following three points of view. The first is (1) time-dependence. In the isolated quantum system with time-dependent parameters, the Berry phase [27] is important. The second is (2) open quantum system. The quantum dot (QD) system coupled to several leads is an instance of the open quantum system. A theoretical method to study the open quantum system is the quantum master equation (QME). The third is the (3) nonequilibrium steady state (NESS). The entropy production under operations between NESSs of the classical system is being studied actively.

In particular, in this thesis, we study the quantum pump and the excess entropy production. In a mesoscopic system, even at zero bias, a charge or spin current is induced by a modulation of the control parameters [39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49]. This phenomenon, called the quantum pump, is theoretically interesting because its origins are quantum effects and nonequilibrium effects. The entropy production under operations between NESSs is composed of the time integral of the instantaneous steady entropy production rate and the excess entropy production. The excess entropy production is intensively being studied as a generalization of the entropy concept.

Recently, Ref.[19] had been applied the Berry-Sinitsyn-Nemenman (BSN) phase to the excess entropy production in the classical system. The BSN phase is the "Berry phase" of the modified master equation including the counting fields which is a tool of the full counting statistics (FCS). For quantum system described by the QME, Ref.[23] had applied the BSN phase using the FCS-QME [26] to study the quantum adiabatic pump. The FCS-QME had also been applied the excess entropy production in the quantum system [20]. However, we point out that this study has serious flaws [98].

1.2 Full counting statistics

In this section, we consider two terminals system. In a mesoscopic system, we can see quantum properties through the conducting property. By recent development of experimental techniques, the transfered charge Q within a time interval τ and the variance $\langle (Q - \langle Q \rangle)^2 \rangle$ and higher cumulants can be measured ($\langle \cdots \rangle$ is the statistical average). The notion of obtaining all cumulants is called the full counting statistics

(FCS)[26, 74, 75, 76]. The n-th order cumulant $\langle Q^n \rangle_c$ is defined by

$$\langle Q^n \rangle_c \stackrel{\text{def}}{=} \frac{\partial^n S_\tau(\chi)}{\partial (i\chi)^n}\Big|_{\chi=0}, \tag{1.1}$$

where

$$S_\tau(\chi) = \ln \int dQ\, P_\tau(Q) e^{iQ\chi}, \tag{1.2}$$

is the cumulant generating function of Q. $P_\tau(Q)$ is the probability distribution of Q. χ is called the counting field. The cumulants up to fourth order are given by

$$\begin{aligned}
\langle Q \rangle_c &= \langle Q \rangle, \\
\langle Q^2 \rangle_c &= \langle Q^2 \rangle - \langle Q \rangle^2, \\
\langle Q^3 \rangle_c &= \langle Q^3 \rangle - 3\langle Q^2 \rangle \langle Q \rangle + 2\langle Q \rangle^3 \\
&= \langle (Q - \langle Q \rangle)^3 \rangle, \\
\langle Q^4 \rangle_c &= \langle Q^4 \rangle - 4\langle Q^3 \rangle \langle Q \rangle - 3\langle Q^2 \rangle^2 + 12\langle Q^2 \rangle \langle Q \rangle^2 - 6\langle Q \rangle^4 \\
&= \langle (Q - \langle Q \rangle)^4 \rangle - 3\langle (Q - \langle Q \rangle)^2 \rangle^2.
\end{aligned}$$

The third and fourth cumulants describe the skewness and sharpness, respectively.

The noise $\langle Q^2 \rangle_c$ is composed of the thermal noise (the Johnson-Nyquis noise) and the shot noise. The shot noise appears when $|eV| > k_{\mathrm{B}}T$ where V is the voltage and T is the temperature. The shot noise S relates with the current $I = \frac{\langle Q \rangle}{\tau}$ as

$$S = 2eFI, \tag{1.3}$$

where F is the Fano factor. For classical shot noise (Poisson noise), $F = 1$ holds. Then, effective charge e^* is defined by

$$S = 2e^* I. \tag{1.4}$$

$e^* = e/3$ had been observed for the fractional quantum Hall state $\nu = 1/3$ [88, 89].

The FCS [26, 74, 75, 76] is the method to calculate the generating function. From the FCS of entropy production, the fluctuation theorem [90, 91, 92] is derived [26, 76]. The fluctuation theorem leads to

$$S^{(0)} = 2k_B T G^{(1)}, \tag{1.5}$$

$$S^{(1)} = k_B T G^{(2)}. \tag{1.6}$$

Here, the noise S and the current I are expanded as

$$S = S^{(0)} + S^{(1)}V + S^{(2)}V^2 + \cdots, \tag{1.7}$$

$$I = G^{(1)}V + G^{(2)}V^2 + \cdots. \tag{1.8}$$

(1.5) is the Johnson-Nyquist relation, which can be derived from the linear response theory. (1.6) is a relation of the non-linear response. This relation had been tested by experiments [93, 94].

1.3 Quantum adiabatic pump

In a mesoscopic system, even at zero bias, a charge or spin current is induced by a slow modulation of control parameters [39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49]. This phenomenon, called the quantum adiabatic pump, is theoretically interesting because its origins are quantum effects and nonequilibrium effects. The quantum adiabatic pump is also expected to be applied to the single electron transfer devices and the current standard[50, 51].

1.3.1 Closed system

For a closed quantum system, the Berry phase [27, 95] appear when the parameter of the Hamiltonian is changed adiabatically (slowly). The quantization of the quantum Hall coefficient is proposed by Thouless *et al.* [39] in 1982. In 1983, for the system which only the x-direction is periodic, Thouless showed [39] that the transfered charge by the cyclic adiabatic modulation of the potential is quantized.

1.3.2 Brouwer formula

The adiabatically pumped quantity is described by a geometric expression in the control parameter space, although the pumped quantity coming from second or higher order of the pumping frequency is not geometric. In noninteracting systems, the quantum adiabatic pump had extensively been studied by the Brouwer formula [31, 52, 53, 54, 55, 56, 57, 58, 59], which describes the pumped charge by the scattering matrix. The Brouwer formula is discovered in 1998 by Brouwer [31].

When n-th control parameter α^n is changed to $\alpha^n + \delta\alpha^n$, the change of the average number of the electrons of the bath b is given by $\mathcal{E}_n^{N_b}(\alpha)\delta\alpha^n$. $\mathcal{E}_n^{N_b}(\alpha)$ is called emissivity. For absolute zero temperature,

$$\mathcal{E}_n^{N_b}(\alpha) = \frac{1}{2\pi}\sum_B\sum_{A\in b}\mathrm{Im}\Big[\frac{\partial S_{AB}(\alpha)}{\partial\alpha^n}S^*_{AB}(\alpha)\Big], \tag{1.9}$$

holds [96]. Here, A and B are labels of electron in the baths and $S(\alpha)$ is the scattering matrix. By slow modulation of the control parameters between time $t = 0$ and $t = \tau$, the change of the average number of the electrons of the bath $\langle\Delta N_b\rangle$ is given by

$$\langle\Delta N_b\rangle = \int_0^\tau dt\,\frac{d\alpha_t^n}{dt}\mathcal{E}_n^{N_b}(\alpha_t) = \int_C d\alpha^n\,\mathcal{E}_n^{N_b}(\alpha). \tag{1.10}$$

The summation symbol $\sum_n$ is omitted. C is the trajectory in the control parameters. In particular, for cyclic modulation $\alpha_0 = \alpha_\tau$, using the Stokes theorem,

$$\langle\Delta N_b\rangle = \int_S d\alpha^m\wedge d\alpha^n\,\frac{1}{2}F^{N_b}_{mn}(\alpha), \tag{1.11}$$

holds. S is the surface enclosed by C. $F^{N_b}_{mn}$ is given by

$$\begin{aligned} F^{N_b}_{mn}(\alpha) &\stackrel{\text{def}}{=} \frac{\partial\mathcal{E}_n^{N_b}(\alpha)}{\partial\alpha^m} - \frac{\partial\mathcal{E}_m^{N_b}(\alpha)}{\partial\alpha^n} \\ &= \frac{1}{\pi}\sum_B\sum_{A\in b}\mathrm{Im}\Big[\frac{\partial S_{AB}(\alpha)}{\partial\alpha^n}\frac{\partial S^*_{AB}(\alpha)}{\partial\alpha^m}\Big]. \end{aligned} \tag{1.12}$$

If the electrons of the bath b are labeled by b and energy ε and the scattering is elastic

$$S_{b\varepsilon,b'\varepsilon'}(\alpha) \;=\; S_{b,b'}(\varepsilon,\alpha)\delta_{\varepsilon,\varepsilon'}, \tag{1.13}$$

$F^{N_b}_{mn}(\alpha)$ at zero-bias is given by

$$F^{N_b}_{mn}(\alpha) \;=\; \frac{1}{\pi}\sum_{b'}\mathrm{Im}\Big[\frac{\partial S_{b,b'}(\mu,\alpha)}{\partial\alpha^n}\frac{\partial S^*_{b,b'}(\mu,\alpha)}{\partial\alpha^m}\Big]. \tag{1.14}$$

Here, μ is the chemical potential of the baths.

On the other hand, it is difficult to calculate the scattering matrix in the interacting systems. In the interacting system, the Brouwer formula had only been applied in mean field treatments [60, 61] or in the Toulouse limit [62].

1.3.3 Recent studies of the quantum pump

Recently, the quantum pump in interacting systems have been actively researched. There are three theoretical approaches. The first is the Green's function approach [32, 63, 64, 99, 100]. The second is the generalized master equation (GME) [65, 66, 33, 67, 68, 69, 70, 71] approach which uses the GME that is equivalent[72, 73] to the quantum master equation (QME) derived using the Nakajima-Zwanzig projection operator technique [28]. Particularly, Ref.[69] derived a geometric expression similar to the Brouwer formula and the Berry-Sinitsyn-Nemenman (BSN) vector explained later. The third is the full counting statistics[26, 74, 75] (FCS) with quantum master equation (FCS-QME, which is also called the generalized quantum master equation[26]) approach proposed in Ref.[23].

The adiabatic modulation of the control parameters induces a Berry-phase-like[27] quantity called the BSN phase in the FCS-QME with the Markov approximation. Sinitsyn and Nemenman[22] studied the adiabatically pumped charge using the FCS and had shown that it is characterized by the BSN vector, which results from the BSN phase. The BSN vector was applied to the spin boson system [77]. The FCS-QME approach can treat the Coulomb interaction, which can not be treated in the Brouwer formula. The derived formula of the BSN vector depends on the approximations used for the QME. The Born-Markov approximation with or without the rotating wave approximation [28](RWA) is frequently used. The QME in the Born-Markov approximation without RWA sometimes violates the non-negativity of the system reduced density operator [78]. The QME of the RWA or the coarse-graining approximation[29, 30](CGA) is the Lindblad type which guarantees the non-negativity [28].

Some recent papers [68, 69, 23] showed that the Coulomb interaction induces the quantum pump. In Refs.[68, 69], it was shown that in a one level interacting quantum dot (QD) weakly coupled to two leads, the pumped charge (also spin in Ref.[69]) induced by an adiabatic modulation of the energy level of the QD and the bias between the two leads vanishes in the noninteracting limit. In particular, Yuge *et al.*[23] studied the pumped charge coming from the BSN curvatures by adiabatic modulation of the thermodynamic parameters (the chemical potentials and the temperatures) in spinless QDs weakly coupled to two spinless leads and showed that the BSN curvatures are zero in noninteracting QDs although they are nonzero for finite interaction.

1.4 Thermodynamic entropy

We review the thermodynamic entropy based on Ref.[97].

1.4.1 Principles of thermodynamics

A macro system A is generally imposed internal constraints which describe the characters of the internal structures. For instance, the subsystem of A is enclosed by the wall which does not transmit heat. A can be decomposed to the simple systems $\{A_i\}_i$. The simple system is the macro system which has not internal constraints and of which spatial non-uniformity in the equilibrium state due to the external fields is negligible. The equilibrium state is the state which all macro variables of the system do not change (as functions of time). As a principle, for arbitrary macro system A, it is requested that if A is isolated (static external fields can exist) and is left sufficiently long time, A becomes the equilibrium state. As principles, the followings are requested: (i) If A is in the equilibrium state, the entropy S exist uniquely.
(ii) The entropy S_i of A_i is a function of the internal energy U_i of A_i and the set of additive variables of A_i, $\{X_i^\alpha\}_{\alpha=1}^{m_i}$: $S_i = S_i(U_i, X_i^1, \cdots, X_i^{m_i})$. $U_i, X_i^1, \cdots, X_i^{m_i}$ are called the natural variables.
(iii) $S_i(U_i, X_i^1, \cdots, X_i^{m_i})$ is continuously differentiable for the natural variables. In particular, $k_{\mathrm{B}}\beta_i \stackrel{\mathrm{def}}{=} \partial S_i/\partial U_i$ is positive and its lower limit is 0 and the upper limit does not exist. Here, k_{B} is the Boltzmann constant and β_i is the inverse temperature of A_i.
(iv) A is in the equilibrium state if and only if all A_i are in the equilibrium states and $\hat{S} \stackrel{\mathrm{def}}{=} \sum_i S_i(U_i, X_i^1, \cdots, X_i^{m_i})$ is maximized. The entropy S of A is given by $S = \max_{\{U_i, X_i^1, \cdots, X_i^{m_i}\}} \hat{S}$ where max is the maximization under the permitted area. The values of the natural variables which provide the S are those in the equilibrium state.

1.4.2 Heat and entropy

The work W is the transfered energy described by the macro variables. In general, W is the summation of the mechanical work W^M and the work due to particle transfer W^C and etc. The heat Q is defined by $\mathcal{U} - W$ where $\mathcal{U}$ is the total transfered energy from the external system. Because $\mathcal{U}$ is the change of the internal energy ΔU, $Q = \Delta U - W$ holds.

The process in which a system B can be regarded as always be in the equilibrium state is called the quasistatic process for B.

From the principles of § 1.4.1, the following theorem is derived. We consider a process that a general system A receives the heat from external systems B_1, B_2, $\cdots, B_M$ exchanging mechanical work with the external systems C_1, C_2, $\cdots, C_N$. When A contacts with B_i, A does not contact with $\{B_b\}_{b\neq i}$. The set $\{(b,k) \in \{1,2,\cdots,M\}\times \{1,2,\cdots,N\} | B_b = C_k\}$ may not be an empty set. We suppose that this process is quasistatic process for $\{B_b\}_{b=1}^M$. Then, the change of the entropy ΔS of A satisfies

$$\Delta S \leq \sum_b \int_{i^{(b)}}^{f^{(b)}} k_{\mathrm{B}}\beta_b d'Q. \tag{1.15}$$

Here, β_b is the inverse temperature of B_b, and $i^{(b)}(f^{(b)})$ denotes the initial (final) state contacting B_b. In particular, the equality holds if the following conditions satisfy: (i)

This process is also quasistatic process for A. (ii) While A contacts to B_b, the inverse temperature of A equals to β_b.

In the following of this subsection, we consider a simple system A. We denote the natural variables of the entropy S of A by U and $\{X^\alpha\}_{\alpha=1}^m$. From the principles of § 1.4.1, $S(U, \{X^\alpha\}_{\alpha=1}^m)$ is convex upward for each natural variable. The equation $S = S(U, \{X^\alpha\}_{\alpha=1}^m)$ can solve for U uniquely: $U = U(S, \{X^\alpha\}_{\alpha=1}^m)$. We introduce $T \stackrel{\text{def}}{=} \partial U/\partial S$ and $P_\alpha \stackrel{\text{def}}{=} \partial U/\partial X^\alpha$. T is the temperature of A and $T = 1/(k_{\mathrm{B}}\beta)$ holds with $k_{\mathrm{B}}\beta = \partial S/\partial U$. If X^α is the total particle number N, $\mu \stackrel{\text{def}}{=} \partial U/\partial N$ is the chemical potential.

We denote the work by changing of X^α by W^α. For a quasistatic process for A, the work is defined by

$$d'W^\alpha \stackrel{\text{def}}{=} P_\alpha dX^\alpha \text{ (quasistatic process)}. \tag{1.16}$$

Using $dU = TdS + \sum_\alpha P_\alpha dX^\alpha$ and the definition of the heat,

$$dS = \beta d'Q \text{ (quasistatic process)}, \tag{1.17}$$

hold. This is called the Clausius equality. For the general system (which is not simple system), the Clausius equality holds if the temperature is uniform in the system. In particular, if $\sum_{\alpha=1}^M P_\alpha dX^\alpha = \mu dN$ holds, $d'Q = dU - \mu dN$ and

$$dS = \beta(dU - \mu dN) \text{ (quasistatic process)}, \tag{1.18}$$

hold. Here and in the following this thesis, we set $k_{\mathrm{B}} = 1$. In general process, it is difficult to define the heat. For a quasistatic process for B, Q_B can be defined as explained above. In (1.15), $d'Q$ is defined by $-d'Q_b$ where $d'Q_b$ is the heat to B_b.

In the equilibrium classical (quantum) system, the entropy is given by the Shannon entropy of the probability distribution (von Neumann entropy of the density matrix) of states.

1.5 Nonequilibrium steady state

Let us consider a system A coupled to the baths $\{B_b\}_{b=1}^M$ ($M > 1$). We suppose that $\{B_b\}_{b\in\mathcal{C}}$ are the canonical baths and $\{B_b\}_{b\in\mathcal{G}}$ are the grand canonical baths. We denote the inverse temperature of B_b by β_b and the chemical potential of B_b ($b \in \mathcal{G}$) by μ_b. If all β_b and μ_b are the same ($\beta_b = \beta$ for all b and $\mu_b = \mu$ for all $b \in \mathcal{G}$), the total system is referred as zero-bias or equilibrium. For the nonequilibrium total system fixing (control) parameters, if A is left sufficiently long time and becomes a steady state, this state of A or the total system is called the nonequilibrium steady state (NESS). For quantum system described by the QME, the NESS exists uniquely.

As the instance, we consider spinless one level QD coupled to several leads. $|0\rangle$ ($|1\rangle$) denotes the state that the QD is empty (occupied). The diagonal components $p_n(t) = \langle n|\rho(t)|n\rangle$ ($n = 0, 1$) of the system state ρ are governed by the master equation:

$$\frac{d}{dt}\begin{pmatrix} p_0(t) \\ p_1(t) \end{pmatrix} = K \begin{pmatrix} p_0(t) \\ p_1(t) \end{pmatrix}. \tag{1.19}$$

The Liouvillian is given by

$$K = \sum_b \Gamma_b \begin{pmatrix} -f_b & 1-f_b \\ f_b & -(1-f_b) \end{pmatrix}. \tag{1.20}$$

Here, Γ_b is the line-width function of the lead b, $f_b = [e^{\beta_b(\varepsilon-\mu_b)}+1]^{-1}$ is the Fermi distribution function, ε is the energy level of the QD. In this section the parameters are fixed. The solution of the master equation is

$$\begin{pmatrix} p_0(t) \\ p_1(t) \end{pmatrix} = \begin{pmatrix} 1-F \\ F \end{pmatrix} + e^{-\Gamma t} \begin{pmatrix} -p_1(0)+F \\ p_1(0)-F \end{pmatrix}, \tag{1.21}$$

where $\Gamma = \sum_b \Gamma_b$ and

$$F = \sum_b f_b \frac{\Gamma_b}{\Gamma}. \tag{1.22}$$

The first term of the RHS of (1.21) is the NESS.

1.6 Excess entropy

The investigation of thermodynamic structures of NESSs has been a topic of active research in nonequilibrium statistical mechanics [1, 2, 3, 4, 5, 6, 7, 8, 9]. For instance, the extension of the relations in equilibrium thermodynamics, such as the Clausius equality, to NESSs has been one of the central subjects. Recently there has been a progress in the extension of the Clausius equality to NESSs [10, 11, 12] (see also Refs.[13, 14, 15, 16, 17, 18]). In these studies, the excess heat $Q_{b,\mathrm{ex}}$ (of the bath b) [2], which describes an additional heat induced by a transition between NESSs with time-dependent external control parameters, has been introduced instead of the total heat Q_b. The excess heat $Q_{b,\mathrm{ex}}$ is defined by subtracting from Q_b the time integral of the instantaneous steady heat current from the bath b. In the weakly nonequilibrium regime, it is proposed that there exists a scalar potential $\mathcal{S}$ in the control parameter space which approximately satisfies the extended Clausius equality

$$\sum_b \beta_b Q_{b,\mathrm{ex}} \approx \Delta\mathcal{S}. \tag{1.23}$$

Here, β_b is the inverse temperature of the bath b, $\Delta\mathcal{S} = \mathcal{S}(\alpha_{t_f}) - \mathcal{S}(\alpha_{t_i})$, α_t is the value of the set of the control parameters at time t, and t_i and t_f are initial and final times of the operation. In classical systems, $\mathcal{S}$ is the symmetrized Shannon entropy [11]. In quantum systems with the time-reversal symmetry, $\mathcal{S}$ is the symmetrized von Neumann entropy [12]. In general, the left hand side (LHS) of (1.23) is replaced by the excess entropy $\sigma_{\mathrm{ex}} \stackrel{\mathrm{def}}{=} \sigma - \int_{t_i}^{t_f} dt\, J^{\sigma}_{\mathrm{ss}}(\alpha_t)$ where σ is the average entropy production and $J^{\sigma}_{\mathrm{ss}}(\alpha_t)$ is the instantaneous steady entropy production rate [19, 20, 21]. In the quasistatic operation, the excess entropy is given by

$$\sigma_{\mathrm{ex}} = \Delta\mathcal{S} + \mathcal{O}(\varepsilon^2\delta), \tag{1.24}$$

where ε is a measure of degree of nonequilibrium and δ describes the amplitude of the change of the control parameters. Sagawa and Hayakawa [19] studied the full counting statistics (FCS) of the entropy production for classical systems described

by the Markov jump process and showed that the excess entropy is characterized by the Berry-Sinitsyn-Nemenman (BSN) phase [22].

The method of Ref.[19] was generalized to quantum systems and applied to studies of the quantum pump [23, 24, 25]. We explain the studies of the quantum pump. At $t = 0$ and $t = \tau$, we perform projection measurements of a *time-independent* observable O of the baths and obtain the outcomes $o(0)$ and $o(\tau)$. The generating function of $\Delta o = o(\tau) - o(0)$ is $Z_\tau(\chi) = \int d\Delta o\ P_\tau(\Delta o) e^{i\chi\Delta o}$ where $P_\tau(\Delta o)$ is the probability density distribution of Δo and χ is called the counting field. To calculate the generating function, the method using the quantum master equation (QME) with the counting field (FCS-QME) [26] had been proposed. The solution of the FCS-QME $\rho^\chi(t)$ provides the generating function as $Z_\tau(\chi) = \mathrm{Tr}_S[\rho^\chi(\tau)]$. Tr_S denotes the trace of the system. The Berry phase [27] of the FCS-QME is the BSN phase. The average of the difference of the outcomes is given by $\langle \Delta o \rangle = \int_0^\tau dt\ i^O(t)$ where $i^O(t)$ is the current of an operator O. If the state of the system at $t = 0$ is the instantaneous steady state and the modulation of the control parameters is slow, the relation

$$\langle \Delta o \rangle = \int_0^\tau dt\ i_{\mathrm{ss}}^O(\alpha_t) + \int_C d\alpha^n\ A_n^O(\alpha), \tag{1.25}$$

holds. Here, the summation symbol for n is omitted. $i_{\mathrm{ss}}^O(\alpha_t)$ is the instantaneous steady current of O and $A_n^O(\alpha)$ is the BSN vector derived from the BSN phase. α^n is n-th component of the control parameters, and C is the trajectory from α_0 to α_τ. The derived formula of the BSN vector depends on the approximations used for the QME.

Because of (1.18), the entropy production rate of the bath b is $\dot\sigma_b(t) = \beta_b(t)[i^{H_b}(t) - \mu_b(t) i^{N_b}(t)]$ where μ_b is the chemical potential of the bath b, and $i^{H_b}(t)$ and $i^{N_b}(t)$ are energy and particle currents from the system to the bath b, respectively. H_b and N_b are the Hamiltonian and the total particle number of the bath b, respectively. Then, it is natural to identify $\dot\sigma(t) \stackrel{\mathrm{def}}{=} -\sum_b \dot\sigma_b(t) = \sum_b \beta_b(t)[-i^{H_b}(t) - \mu_b(t)\{-i^{N_b}(t)\}]$ with the average entropy production rate of the system. $\sigma \stackrel{\mathrm{def}}{=} \int_0^\tau dt\ \dot\sigma(t)$ is the average entropy production. Because of (1.25), $\sigma = \int_0^\tau dt\ J_{\mathrm{ss}}^\sigma(\alpha_t) + \int_C d\alpha^n\ A_n^\sigma(\alpha)$ holds with $J_{\mathrm{ss}}^\sigma(\alpha) \stackrel{\mathrm{def}}{=} \sum_b \beta_b[-i_{\mathrm{ss}}^{H_b}(\alpha) - \mu_b\{-i_{\mathrm{ss}}^{N_b}(\alpha)\}]$ and

$$A_n^\sigma(\alpha) \stackrel{\mathrm{def}}{=} \sum_b \beta_b[-A_n^{H_b}(\alpha) - \mu_b\{-A_n^{N_b}(\alpha)\}]. \tag{1.26}$$

Here, $i_{\mathrm{ss}}^{H_b}(\alpha)$ and $i_{\mathrm{ss}}^{N_b}(\alpha)$ are the instantaneous steady currents of the energy and particle from the system to the bath b. $A_n^{H_b}(\alpha)$ and $A_n^{N_b}(\alpha)$ are the BSN vectors of H_b and N_b. The excess entropy production is given by

$$\sigma_{\mathrm{ex}} = \int_C d\alpha^n\ A_n^\sigma(\alpha). \tag{1.27}$$

Yuge *et al.* [20] applied the FCS-QME approach to the excess entropy production of the quantum system. They identified $\sigma' \stackrel{\mathrm{def}}{=} \langle a(\tau) - a(0) \rangle$ with the average entropy production. Here, $a(0)$ and $a(\tau)$ are the outcomes of $A(t) = -\sum_b \beta_b(t)[H_b - \mu_b(t) N_b]$ at $t = 0$ and $t = \tau$. However, σ' is not the average entropy production σ. $\sigma' \approx$

$\mathrm{Tr}_{\mathrm{tot}}[A(\tau)\rho_{\mathrm{tot}}(\tau)] - \mathrm{Tr}_{\mathrm{tot}}[A(0)\rho_{\mathrm{tot}}(0)]$ can be rewritten as

$$\begin{aligned}\sigma' \approx & -\int_0^\tau dt \sum_b \Big[\frac{d\beta_b(t)}{dt}\langle H_b\rangle_t - \frac{d[\beta_b(t)\mu_b(t)]}{dt}\langle N_b\rangle_t\Big] \\ & + \int_0^\tau dt \sum_b \Big[\beta_b(t)\{-\frac{d}{dt}\langle H_b\rangle_t\} - \beta_b(t)\mu_b(t)\{-\frac{d}{dt}\langle N_b\rangle_t\}\Big]. \end{aligned} \tag{1.28}$$

Here, $\langle\bullet\rangle_t \stackrel{\mathrm{def}}{=} \mathrm{Tr}_{\mathrm{tot}}[\bullet\rho_{\mathrm{tot}}(t)]$, $\rho_{\mathrm{tot}}(t)$ is the total system state and $\mathrm{Tr}_{\mathrm{tot}}$ denotes the trace of the total system. The integrand of the second term of the RHS of (1.28) roughly equals to $\dot{\sigma}$ [36]. However, the physical meaning of the first term is not clear. Then, because of the presence of the first term, $\sigma' \neq \sigma$ is concluded. Moreover, they improperly used the FCS-QME applicable only for *time-independent* observable to calculate σ' although $A(t)$ is time-dependent. These two issues are the problems of Ref.[20].

1.7 Aim of this thesis

There are several theoretical approaches to analyze the quantum pump. However, the relations among these are not clear. Then, the first aim of this thesis is to clarify these relations (in particular, the relation between the FCS-QME approach and the GME approach). Moreover, in the previous works, the charge pump had been studied mainly. However, for applications to the spintronics and quantum information processing, the spin degree of freedom is important. Then, we consider the spin degree of freedom and study the spin pump.

Recently, the excess entropy of the classical system is established. However, one for the quantum system is not sufficient as we explained in § 1.6. The second aim of this thesis is to develop the excess entropy of the quantum system. Moreover, we compare between our results and previous results of both classical and quantum systems.

1.8 Outline of the thesis

The outline of the thesis is as follows. First, we review the FCS and the FCS-QME (Chap.2). In § 2.1, we derive the modified von Neumann equation including the counting fields. In § 2.2, we derive and the FCS-QME with the CGA. In § 2.3, we explain the RWA. In § 2.4, we derive the detailed balance condition.

Next, we move to the original results (§ 3.2,§ 3.3 and before (3.21) are review parts). Chap.3 and Chap.4 are based on Ref.[25]. Chap.6, Chap.7 and Chap.8 are based on Ref.[98]. We apply the FCS-QME to the quantum pump (Chap.3). In § 3.1, we derive the expression for current without any approximation and introduce the BSN vector. The BSN vector is also derived from the BSN phase (§ 3.2). In § 3.3, we introduce the BSN curvature used to cyclic adiabatic pump. In § 3.4, we expand the general solution of the QME $\rho(t)$ by the modulation frequency ω as

$$\rho(t) = \rho_0(\alpha_t) + \sum_{n=1}^{\infty}\rho^{(n)}(t) + \sum_{n=0}^{\infty}\tilde{\rho}^{(n)}(t). \tag{1.29}$$

Here, $\rho_0(\alpha_t)$ is the instantaneous steady state of the QME, $\rho^{(n)}(t)$ and $\tilde{\rho}^{(n)}(t)$ are calculable and order $(\omega/\Gamma)^n$. Γ is the coupling strength between the system and the

baths. $\tilde{\rho}^{(n)}(t)$ exponentially damps as a function of time. In the expansion (1.29), a pseudo-inverse of the Liouvillian is used. In § 3.5, we proof the expansion (1.29) is independent of the choice of the pseudo-inverse. In § 3.6, we show that the GME provides an expansion corresponding to $\rho_0(\alpha_t) + \sum_{n=1}^{\infty} \rho^{(n)}(t)$.

In Chap.4, we apply the FCS-QME with the RWA to the quantum adiabatic pump of the quantum dots (QDs) coupled to two leads (L and R). In § 4.1, we explain the model. We show that the pumped charge and spin coming from the instantaneous steady current are not negligible when the thermodynamic parameters are not fixed to zero bias in noninteracting QDs (§ 4.2.2) and an interacting QD (§ 4.3.2). To observe the spin effects, we consider collinear magnetic fields, which relate to spins through the Zeeman effect, with different amplitudes applying to the QDs (B_S) and the leads (B_L and B_R). We focus on the dynamic parameters (B_S, $B_{L/R}$ and the coupling strength between QDs and leads, $\Delta_{L/R}$) as control parameters. In one level QD with the Coulomb interaction U, we analytically calculate the BSN curvatures of spin and charge of (B_L, B_S) pump and (Δ_L, B_S) pump for the noninteracting limit ($U = 0$, § 4.2.3) and the strong interaction limit ($U = \infty$, § 4.3.3) at zero-bias. Moreover, we study the (Δ_L, B_S) pump for finite U at zero-bias (§ 4.3.5).

We study the quantum diabatic pump for spinless one level QD coupled to two leads (Chap.5). We calculate $\{\rho^{(n)}(t)\}_{n=1}^{5}$, $\{\tilde{\rho}^{(n)}(t)\}_{n=1}^{5}$ and particle current up to 6th order and pumped particle numbers.

Next, we introduce the generalized QME (Chap.6) used to analyze the BSN vector of the entropy production. In Chap.7 and Chap.8, we focus on the RWA. In § 7.1, the BSN vector A_n^σ in the equilibrium is discussed. In § 7.2, one of the main result of this thesis

$$A_n^\sigma(\alpha) = -\mathrm{Tr}_S\big[\ln \rho_0^{(-1)}(\alpha) \frac{\partial \rho_0(\alpha)}{\partial \alpha^n}\big] + \mathcal{O}(\varepsilon^2), \tag{1.30}$$

is derived without any assumption on the time-reversal symmetry [98]. $\rho_0^{(-1)}(\alpha)$ is the instantaneous steady state obtained from the QME with reversing the sign of the Lamb shift term. In § 7.3, we consider the time-reversal operation. We show that if the time-reversal symmetry is broken and the system Hamiltonian is degenerated, $\mathcal{S}(\alpha)$ such that $A_n^\sigma(\alpha) = \partial \mathcal{S}(\alpha)/\partial \alpha^n + \mathcal{O}(\varepsilon^2)$ dose not exist. This is the most important result of this thesis. Next we mention the results in the Born-Markov approximation (§ 7.4). In Chap.8, we compare preceding study on of the entropy production in the classical Markov jump process [21, 37] with ours.

At last (Chap.9), we summarize this thesis. In Appendix A, the Liouvillian for the Born-Markov approximation is discussed. In Appendix B, the Liouville space[80, 26] and the matrix representation of the Liouvillian are explained. In Appendix C, we derive (3.23). In Appendix D, we discuss the validity of the adiabatic expansion in Chap.3. In Appendix E, we discuss the derivation of (3.52). In the Appendix F, we discuss the solutions of the GME expanded by the modulation frequency and the coupling strength between the system and the baths. In the Appendix G, we calculate the energy current operator. In the Appendix H, we derive the formula of the derivative of the von Neumann entropy. In the Appendix I, we proof (7.66). In the Appendix J, we explain the definition of entropy production of the Markov jump process and a result of Ref.[21].

Chapter 2

Full counting statistics and quantum master equation

2.1 Full counting statistics

We consider the system S coupled with the bath system B:

$$H_{\rm tot}(t) \;=\; H_S(t) + H_B(t) + H_{\rm int}(t). \tag{2.1}$$

The bath system may contain several baths. The simultaneous eigenstate of a set of the bath's observables $\{O_\mu\}$ is given by

$$O_\mu|\{o_\nu\}, r\rangle \;=\; o_\mu|\{o_\nu\}, r\rangle, \tag{2.2}$$

$$\langle\{o_\nu\}, r|\{o'_\nu\}, s\rangle \;=\; \delta_{r,s}\delta_{\{o_\nu\},\{o'_\nu\}}. \tag{2.3}$$

Here, r and s denote the label of degeneracy, and $\delta_{\{o_\nu\},\{o'_\nu\}} = \prod_{\nu=1}^{n}\delta_{o_\nu,o'_\nu}$ is the kronecker delta. The projection operator to $\{o_\mu\}$ is given by

$$P_{\{o_\mu\}} \;=\; \sum_r |\{o_\mu\}, r\rangle\langle\{o_\mu\}, r|. \tag{2.4}$$

This has the following properties :

$$P_{\{o_\nu\}}P_{\{o'_\nu\}} \;=\; \delta_{\{o_\nu\},\{o'_\nu\}}P_{\{o_\nu\}}, \tag{2.5}$$

$$\sum_{\{o_\nu\}} P_{\{o_\nu\}} \;=\; 1. \tag{2.6}$$

The total system state $\rho_{\rm tot}(t)$ is governed by the von Neumann equation:

$$\frac{d}{dt}\rho_{\rm tot}(t) \;=\; -i[H_{\rm tot}(t), \rho_{\rm tot}(t)]. \tag{2.7}$$

In this thesis, we set $\hbar = 1$. The formal solution is given by

$$\rho_{\rm tot}(t) \;=\; V(t)\rho_{\rm tot}(0)V^\dagger(t), \tag{2.8}$$

where $V(t)$ is the solution of

$$\frac{d}{dt}V(t) \;=\; -iH_{\rm tot}(t)V(t), \tag{2.9}$$

with $V(0) = 1$. At $t = 0$, we perform projection measurements of $\{O_\mu\}$. The probability getting $\{o_\mu^{(0)}\}$ is given by

$$P[\{o_\mu^{(0)}\}] = \mathrm{Tr}_{\mathrm{tot}}[P_{\{o_\mu^{(0)}\}}\rho_{\mathrm{tot}}(0)P_{\{o_\mu^{(0)}\}}]. \tag{2.10}$$

$\mathrm{Tr}_{\mathrm{tot}}$ denotes the trace over the total system. The state just after measuring $\{o_\mu^{(0)}\}$ is

$$\rho_{\mathrm{tot}}^{\{o_\mu^{(0)}\}}(0) = \frac{P_{\{o_\mu^{(0)}\}}\rho_{\mathrm{tot}}(0)P_{\{o_\mu^{(0)}\}}}{P[\{o_\mu^{(0)}\}]}. \tag{2.11}$$

After the time evolution by (2.7), the state at time t is

$$\begin{aligned}\rho_{\mathrm{tot}}^{\{o_\mu^{(0)}\}}(t) &= V(t)\rho_{\mathrm{tot}}^{\{o_\mu^{(0)}\}}(0)V^\dagger(t) \\ &= \frac{V(t)P_{\{o_\mu^{(0)}\}}\rho_{\mathrm{tot}}(0)P_{\{o_\mu^{(0)}\}}V^\dagger(t)}{P[\{o_\mu^{(0)}\}]}.\end{aligned} \tag{2.12}$$

Under this condition, we perform projection measurements of $\{O_\mu\}$ at $t = \tau$. The probability getting $\{o_\mu^{(\tau)}\}$ is given by

$$\begin{aligned}P[\{o_\mu^{(\tau)}\}|\{o_\mu^{(0)}\}] &= \mathrm{Tr}_{\mathrm{tot}}[P_{\{o_\mu^{(\tau)}\}}\rho_{\mathrm{tot}}^{\{o_\mu^{(0)}\}}(\tau)P_{\{o_\mu^{(\tau)}\}}] \\ &= \frac{1}{P[\{o_\mu^{(0)}\}]}\mathrm{Tr}_{\mathrm{tot}}[P_{\{o_\mu^{(\tau)}\}}V(\tau)P_{\{o_\mu^{(0)}\}}\rho_{\mathrm{tot}}(0)P_{\{o_\mu^{(0)}\}}V^\dagger(\tau)P_{\{o_\mu^{(\tau)}\}}].\end{aligned} \tag{2.13}$$

The probability getting $\{o_\mu^{(0)}\}$ at $t = 0$ and $\{o_\mu^{(\tau)}\}$ at $t = \tau$ is

$$\begin{aligned}P[\{o_\mu^{(\tau)}\}, \{o_\mu^{(0)}\}] &= P[\{o_\mu^{(0)}\}] \cdot P[\{o_\mu^{(\tau)}\}|\{o_\mu^{(0)}\}] \\ &= \mathrm{Tr}_{\mathrm{tot}}[P_{\{o_\mu^{(\tau)}\}}V(\tau)P_{\{o_\mu^{(0)}\}}\rho_{\mathrm{tot}}(0)P_{\{o_\mu^{(0)}\}}V^\dagger(\tau)P_{\{o_\mu^{(\tau)}\}}].\end{aligned} \tag{2.14}$$

The probability density distribution of $\{o_\mu^{(\tau)} - o_\mu^{(0)}\}$ is given by

$$P_\tau[\{\Delta o_\mu\}] \stackrel{\mathrm{def}}{=} \mathrm{Prob.}[\{o_\mu^{(\tau)} - o_\mu^{(0)} = \Delta o_\mu\}] \tag{2.15}$$

$$= \sum_{\{o_\mu^{(0)}\},\{o_\mu^{(\tau)}\}} P[\{o_\mu^{(\tau)}\}, \{o_\mu^{(0)}\}] \prod_\mu \delta(o_\mu^{(\tau)} - o_\mu^{(0)} - \Delta o_\mu). \tag{2.16}$$

The generating function is defined by

$$Z_\tau(\chi) \stackrel{\mathrm{def}}{=} \int (\prod_{\nu=1}^n d\Delta o_\nu)\, P_\tau(\{\Delta o_\mu\}) e^{i\sum_{\mu=1}^n \chi_\mu \Delta o_\mu}. \tag{2.17}$$

Here, χ_μ is a real number called the counting field for O_μ. χ denotes the set of the counting fields. The cumulant generating function is defined by

$$S_\tau(\chi) \stackrel{\mathrm{def}}{=} \ln Z_\tau(\chi). \tag{2.18}$$

The n-th order cumulant $\langle \Delta o_{\mu_1}\Delta o_{\mu_2}\dots\Delta o_{\mu_n}\rangle_c$ is given by

$$\langle \Delta o_{\mu_1}\Delta o_{\mu_2}\dots\Delta o_{\mu_n}\rangle_c = \frac{\partial^n S_\tau(\chi)}{\partial(i\chi_{\mu_1})(i\chi_{\mu_2})\cdots(i\chi_{\mu_n})}\Big|_{\chi=0}. \tag{2.19}$$

In particular,

$$\langle \Delta o_\nu \rangle_c = \frac{\partial S_\tau(\chi)}{\partial (i\chi_\nu)}\Big|_{\chi=0} = \langle \Delta o_\nu \rangle, \tag{2.20}$$

is the average of Δo_ν.

Substituting (2.16) to (2.17), we obtain

$$Z_\tau(\chi) = \sum_{\{o_\mu^{(0)}\},\{o_\mu^{(\tau)}\}} P[\{o_\mu^{(\tau)}\},\{o_\mu^{(0)}\}] e^{i\sum_\mu \chi_\mu [o_\mu^{(\tau)} - o_\mu^{(0)}]}. \tag{2.21}$$

Substituting (2.14) to the above equation, we obtain

$$\begin{aligned} &Z_\tau(\chi) \\ &= \sum_{\{o_\mu^{(0)}\},\{o_\mu^{(\tau)}\}} \mathrm{Tr}_{\mathrm{tot}}[P_{\{o_\mu^{(\tau)}\}} V(\tau) P_{\{o_\mu^{(0)}\}} \rho_{\mathrm{tot}}(0) P_{\{o_\mu^{(0)}\}} V^\dagger(\tau) P_{\{o_\mu^{(\tau)}\}}] e^{i\sum_\mu \chi_\mu [o_\mu^{(\tau)} - o_\mu^{(0)}]}. \end{aligned} \tag{2.22}$$

Now, we introduce

$$\bar{\rho}_{\mathrm{tot}}(0) \stackrel{\mathrm{def}}{=} \sum_{\{o_\mu^{(0)}\}} P_{\{o_\mu^{(0)}\}} \rho_{\mathrm{tot}}(0) P_{\{o_\mu^{(0)}\}}. \tag{2.23}$$

Properties

$$e^{-i\chi_\nu O_\nu/2} P_{\{o_\mu^{(0)}\}} = e^{-i\chi_\nu o_\nu^{(0)}/2} P_{\{o_\mu^{(0)}\}},\ P_{\{o_\mu^{(0)}\}} e^{-i\chi_\nu O_\nu/2} = e^{-i\chi_\nu o_\nu^{(0)}/2} P_{\{o_\mu^{(0)}\}}, \tag{2.24}$$

lead

$$e^{-i\sum_\mu \chi_\mu O_\mu/2} \bar{\rho}_{\mathrm{tot}}(0) e^{-i\sum_\mu \chi_\mu O_\mu/2} = \sum_{\{o_\mu^{(0)}\}} e^{-i\chi_\mu o_\mu^{(0)}} P_{\{o_\mu^{(0)}\}} \rho_{\mathrm{tot}}(0) P_{\{o_\mu^{(0)}\}}. \tag{2.25}$$

Then, (2.22) becomes

$$\begin{aligned} &Z_\tau(\chi) \\ &= \sum_{\{o_\mu^{(\tau)}\}} \mathrm{Tr}_{\mathrm{tot}}[P_{\{o_\mu^{(\tau)}\}} V(\tau) e^{-i\sum_\mu \chi_\mu O_\mu/2} \bar{\rho}_{\mathrm{tot}}(0) e^{-i\sum_\mu \chi_\mu O_\mu/2} V^\dagger(\tau) P_{\{o_\mu^{(\tau)}\}}] e^{i\sum_\mu \chi_\mu o_\mu^{(\tau)}}. \end{aligned} \tag{2.26}$$

Moreover, from

$$P_{\{o_\mu^{(\tau)}\}} e^{i\chi_\nu O_\nu/2} = e^{-i\chi_\nu o_\nu^{(\tau)}/2} P_{\{o_\mu^{(\tau)}\}},\ e^{i\chi_\nu O_\nu/2} P_{\{o_\mu^{(\tau)}\}} = e^{i\chi_\nu o_\nu^{(\tau)}/2} P_{\{o_\mu^{(\tau)}\}}, \tag{2.27}$$

we obtain

$$
\begin{aligned}
Z_\tau(\chi) &= \sum_{\{o_\mu^{(\tau)}\}} \mathrm{Tr}_{\mathrm{tot}}[P_{\{o_\mu^{(\tau)}\}} e^{i\chi_\mu O_\mu/2} V(\tau) e^{-i\chi_\mu O_\mu/2} \bar{\rho}_{\mathrm{tot}}(0) e^{-i\chi_\mu O_\mu/2} V^\dagger(\tau) e^{i\chi_\mu O_\mu/2} P_{\{o_\mu^{(\tau)}\}}] \\
&= \sum_{\{o_\mu^{(\tau)}\}} \mathrm{Tr}_{\mathrm{tot}}[P_{\{o_\mu^{(\tau)}\}} P_{\{o_\mu^{(\tau)}\}} e^{i\chi_\mu O_\mu/2} V(\tau) e^{-i\chi_\mu O_\mu/2} \bar{\rho}_{\mathrm{tot}}(0) e^{-i\chi_\mu O_\mu/2} V^\dagger(\tau) e^{i\chi_\mu O_\mu/2}] \\
&= \mathrm{Tr}_{\mathrm{tot}}[\sum_{\{o_\mu^{(\tau)}\}} P_{\{o_\mu^{(\tau)}\}} e^{i\chi_\mu O_\mu/2} V(\tau) e^{-i\chi_\mu O_\mu/2} \bar{\rho}_{\mathrm{tot}}(0) e^{-i\chi_\mu O_\mu/2} V^\dagger(\tau) e^{i\chi_\mu O_\mu/2}] \\
&= \mathrm{Tr}_{\mathrm{tot}}[e^{i\chi_\mu O_\mu/2} V(\tau) e^{-i\chi_\mu O_\mu/2} \bar{\rho}_{\mathrm{tot}}(0) e^{-i\chi_\mu O_\mu/2} V^\dagger(\tau) e^{i\chi_\mu O_\mu/2}] \\
&= \mathrm{Tr}_{\mathrm{tot}}[V_\chi(\tau) \bar{\rho}_{\mathrm{tot}}(0) V_{-\chi}^\dagger(\tau)] \\
&= \mathrm{Tr}_{\mathrm{tot}}[\rho_{\mathrm{tot}}^\chi(\tau)].
\end{aligned} \tag{2.28}
$$

Here, we used (2.5) and (2.6). Here and in the following of this section, $\chi_\mu O_\mu \equiv \sum_\mu \chi_\mu O_\mu$. $V_\chi(t)$ and $\rho_{\mathrm{tot}}^\chi(t)$ are defined by

$$
V_\chi(t) \stackrel{\mathrm{def}}{=} e^{i\chi_\mu O_\mu/2} V(t) e^{-i\chi_\mu O_\mu/2}, \tag{2.29}
$$

$$
\rho_{\mathrm{tot}}^\chi(t) \stackrel{\mathrm{def}}{=} V_\chi(t) \bar{\rho}_{\mathrm{tot}}(0) V_{-\chi}^\dagger(t). \tag{2.30}
$$

$V_\chi(0)$ and $\rho_{\mathrm{tot}}^\chi(0)$ are given by

$$
\begin{aligned}
V_\chi(0) &= 1, \\
\rho_{\mathrm{tot}}^\chi(0) &= \bar{\rho}_{\mathrm{tot}}(0) = \sum_{\{o_\mu^{(0)}\}} P_{\{o_\mu^{(0)}\}} \rho(0) P_{\{o_\mu^{(0)}\}}.
\end{aligned} \tag{2.31}
$$

$V_\chi(t)$ is governed by

$$
\begin{aligned}
\frac{d}{dt} V_\chi(t) &= e^{i\chi_\mu O_\mu/2} \big[\frac{d}{dt} V(t)\big] e^{-i\chi_\mu O_\mu/2} \\
&= e^{i\chi_\mu O_\mu/2} \big[-iH_{\mathrm{tot}}(t) V(t)\big] e^{-i\chi_\mu O_\mu/2} \\
&= -ie^{i\chi_\mu O_\mu/2} H_{\mathrm{tot}}(t) e^{-i\chi_\mu O_\mu/2} e^{i\chi_\mu O_\mu/2} V(t) e^{-i\chi_\mu O_\mu/2} \\
&= -iH_{\mathrm{tot},\chi}(t) V_\chi(t),
\end{aligned} \tag{2.32}
$$

with

$$
H_{\mathrm{tot},\chi}(t) \stackrel{\mathrm{def}}{=} e^{i\chi_\mu O_\mu/2} H_{\mathrm{tot}}(t) e^{-i\chi_\mu O_\mu/2}. \tag{2.33}
$$

$H_{\mathrm{tot},\chi}(t)$ is a Hermitian operator :

$$
H_{\mathrm{tot},\chi}^\dagger(t) = H_{\mathrm{tot},\chi}(t). \tag{2.34}
$$

From the Hermitian conjugate of (2.32), we obtain

$$
\frac{d}{dt} V_{-\chi}^\dagger(t) = iV_{-\chi}^\dagger(t) H_{\mathrm{tot},-\chi}(t). \tag{2.35}
$$

From (2.30), (2.32) and the above equation, $\rho^{\chi}_{\rm tot}(t)$ is governed by

$$\begin{aligned}\frac{d}{dt}\rho^{\chi}_{\rm tot}(t) &= \frac{d}{dt}[V_{\chi}(t)\bar{\rho}_{\rm tot}(0)V^{\dagger}_{-\chi}(t)]\\ &= -iH_{{\rm tot},\chi}(t)V_{\chi}(t)\bar{\rho}_{\rm tot}(0)V^{\dagger}_{-\chi}(t) + iV_{\chi}(t)\bar{\rho}_{\rm tot}(0)V^{\dagger}_{-\chi}(t)H_{{\rm tot},-\chi}(t)\\ &= -i[H_{{\rm tot},\chi}(t)\rho^{\chi}_{\rm tot}(t) - \rho^{\chi}_{\rm tot}(t)H_{{\rm tot},-\chi}(t)]. \end{aligned} \tag{2.36}$$

2.2 Quantum master equation with counting fields

2.2.1 Derivation of FCS-QME

We consider system S weakly coupled to several baths. The total Hamiltonian is given by

$$H_{\rm tot}(\alpha'(t)) = H_S(\alpha_S(t)) + \sum_b [H_b(\alpha'_b(t)) + H_{Sb}(\alpha_{Sb}(t))]. \tag{2.37}$$

$H_S(\alpha_S)$ is the system Hamiltonian and α_S denotes a set of control parameters of the system. $H_b(\alpha'_b)$ is the Hamiltonian of the bath b and α'_b is a set of control parameters. $H_{Sb}(\alpha_{Sb})$ is the coupling Hamiltonian between S and the bath b, and α_{Sb} is a set of control parameters. We suppose that the states of the baths for $b = 1, 2, \cdots, n_{\rm C}$ are the canonical distributions and these for $b = n_{\rm C}+1, \cdots, n_{\rm C}+n_{\rm GC}$ are the grand canonical distributions. We denote $\{1, \cdots, n_{\rm C}\}$ and $\{n_{\rm C}+1, \cdots, n_{\rm C}+n_{\rm GC}\}$ by $\boldsymbol{\mathcal{C}}$ and $\boldsymbol{\mathcal{G}}$. We denote the inverse temperature of the bath b by β_b and the chemical potential of the bath $b \in \boldsymbol{\mathcal{G}}$ by μ_b. α''_b denotes β_b for $b \in \boldsymbol{\mathcal{C}}$ and the set of β_b and $\beta_b\mu_b$ for $b \in \boldsymbol{\mathcal{G}}$. We symbolize the set of all control parameters (α_S, $\{\alpha_{Sb}\}_b$, $\{\alpha'_b\}_b$, $\{\alpha''_b\}_b$) by α, (α_S, $\{\alpha_{Sb}\}_b$, $\{\alpha'_b\}_b$) by α', $\{\alpha''_b\}_b$ by α'', (α'_b, α''_b) by α_b, and $\{\alpha_b\}_b$ by α_B. While α' are dynamical parameters, α'' are thermodynamical parameters. We denote the set of all the linear operators of S by B.

The modified von Neumann equation (2.36) [26] is

$$\frac{d}{dt}\rho^{\chi}_{\rm tot}(t) = -i[H_{\rm tot}(t), \rho^{\chi}_{\rm tot}(t)]_{\chi}. \tag{2.38}$$

Here, $[A, B]_{\chi} \stackrel{\rm def}{=} A_{\chi}B - BA_{-\chi}$ and $A_{\chi} \stackrel{\rm def}{=} e^{i\sum_{\mu}\chi_{O_\mu}O_\mu/2}Ae^{-i\sum_{\mu}\chi_{O_\mu}O_\mu/2}$. χ_{O_μ} is χ_μ of § 2.1. We suppose

$$\rho_{\rm tot}(0) = \rho(0) \otimes \rho_B(\alpha_B(0)), \tag{2.39}$$

where $\rho_B(\alpha_B(0)) \stackrel{\rm def}{=} \bigotimes_b \rho_b(\alpha_b(0))$ and $\rho_b(\alpha_b(0)) \stackrel{\rm def}{=} e^{-\beta_b(0)H_b(\alpha'_b(0))}/Z_b(\alpha_b(0))$ with $Z_b(\alpha_b) \stackrel{\rm def}{=} {\rm Tr}_b[e^{-\beta_b H_b(\alpha'_b)}]$ for $b \in \boldsymbol{\mathcal{C}}$ and $\rho_b(\alpha_b(0)) \stackrel{\rm def}{=} e^{-\beta_b(0)[H_b(\alpha'_b(0))-\mu_b(0)N_b]}/\Xi_b(\alpha_b(0))$ with $\Xi_b(\alpha_b) \stackrel{\rm def}{=} {\rm Tr}_b[e^{-\beta_b[H_b(\alpha'_b)-\mu_b N_b]}]$ for $b \in \boldsymbol{\mathcal{G}}$. ${\rm Tr}_b$ denotes the trace of the bath b and N_b ($b \in \boldsymbol{\mathcal{G}}$) is the total number operator of the bath b. Then,

$$\rho^{\chi}_{\rm tot}(0) = \rho(0) \otimes \sum_{\{o_\nu\}} P_{\{o_\nu\}}\rho_B(\alpha_B(0))P_{\{o_\nu\}}, \tag{2.40}$$

obeys. We suppose $[H_b, N_b] = 0$. We suppose that O_μ commute with H_b and N_b:

$$[O_\mu, H_b] = 0,\ [O_\mu, N_b] = 0. \tag{2.41}$$

Then, $P_{\{o_\nu\}}$ commutes with $\rho_B(\alpha_B(0))$ and

$$\rho^\chi_{\rm tot}(0) = \rho(0) \otimes \rho_B(\alpha_B(0)), \tag{2.42}$$

holds because (2.5) and (2.6).

We defined

$$\rho^\chi(t) \stackrel{\rm def}{=} {\rm Tr}_B[\rho^\chi_{\rm tot}(t)], \tag{2.43}$$

which provides the generating function

$$Z_\tau(\chi) = {\rm Tr}_S[\rho^\chi(t=\tau)]. \tag{2.44}$$

${\rm Tr}_B$ denotes the trace over all baths' degrees of freedom. We assume $\rho_{\rm tot}(t) \approx \rho(t) \otimes \rho_B(\alpha_B(t))$ $(0 < t \leq \tau)$, where

$$\rho_B(\alpha_B(t)) \stackrel{\rm def}{=} \bigotimes_b \rho_b(\alpha_b(t)), \tag{2.45}$$

$$\rho_b(\alpha_b(t)) \stackrel{\rm def}{=} \begin{cases} e^{-\beta_b(t)H_b(\alpha'_b(t))}/Z_b(\alpha_b(t)) & b \in \boldsymbol{\mathcal{C}} \\ e^{-\beta_b(t)[H_b(\alpha'_b(t))-\mu_b(t)N_b]}/\Xi_b(\alpha_b(t)) & b \in \boldsymbol{\mathcal{G}} \end{cases}. \tag{2.46}$$

and

$$\rho(t) \stackrel{\rm def}{=} {\rm Tr}_B[\rho_{\rm tot}(t)]. \tag{2.47}$$

First, we introduce the CGA. An operator in the interaction picture corresponding to $A(t)$ is defined by

$$A^I(t) = U_0^\dagger(t)A(t)U_0(t), \tag{2.48}$$

with

$$\frac{dU_0(t)}{dt} = -i[H_S(\alpha_S(t)) + \sum_b H_b(\alpha'_b(t))]U_0(t), \tag{2.49}$$

and $U_0(0) = 1$. The system reduced density operator in the interaction picture is given by

$$\rho^{I,\chi}(t) = {\rm Tr}_B[\rho^{I,\chi}_{\rm tot}(t)], \tag{2.50}$$

where

$$\rho^{I,\chi}_{\rm tot}(t) = U_0^\dagger(t)\rho^\chi_{\rm tot}(t)U_0(t). \tag{2.51}$$

$\rho^{I,\chi}_{\rm tot}(t)$ is governed by

$$\frac{d\rho^{I,\chi}_{\rm tot}(t)}{dt} = -i[H^I_{\rm int}(t), \rho^{I,\chi}_{\rm tot}(t)]_\chi, \tag{2.52}$$

with

$$H_{\rm int} \stackrel{\rm def}{=} \sum_b H_{Sb}. \tag{2.53}$$

Up to the second order perturbation in $H_{\rm int}$, we obtain

$$\begin{aligned}\rho^{I,\chi}(t+\tau_{\rm CG}) &= \rho^{I,\chi}(t) \\ &\quad - \int_t^{t+\tau_{\rm CG}} du \int_t^u ds\, {\rm Tr}_B\{[H^I_{\rm int}(u), [H^I_{\rm int}(s), \rho^{I,\chi}(t)\rho_B(\alpha_B(t))]_\chi]_\chi\} \\ &\equiv \rho^{I,\chi}(t) + \tau_{\rm CG}\hat{L}^\chi_{\tau_{\rm CG}}(t)\rho^{I,\chi}(t), \end{aligned} \tag{2.54}$$

using the large-reservoir approximation

$$\rho^{I,\chi}_{\rm tot}(t) \approx \rho^{I,\chi}(t) \otimes \rho_B(\alpha_B(t)), \tag{2.55}$$

and supposing

$${\rm Tr}_B[H^I_{\rm int}(u)\rho_B(\alpha_B(t))] = 0. \tag{2.56}$$

The arbitrary parameter $\tau_{\rm CG}$ (> 0) is called the coarse-graining time. The CGA [29, 30] is defined by

$$\frac{d}{dt}\rho^{I,\chi}(t) = \hat{L}^\chi_{\tau_{\rm CG}}(t)\rho^{I,\chi}(t). \tag{2.57}$$

In the Schrödinger picture, (2.57) is described as

$$\frac{d\rho^\chi(t)}{dt} = -i[H_S(\alpha_S(t)), \rho^\chi(t)] + \sum_b \mathcal{L}^\chi_{b,\tau_{\rm CG}}(\alpha_t)\rho^\chi(t). \tag{2.58}$$

At $\chi = 0$, this is the Lindblad type. If $\tau_{\rm CG} \ll \tau$, the super-operator $\mathcal{L}^\chi_{b,\tau_{\rm CG}}$ is described as a function of the set of control parameters at time t. $\alpha_t = \alpha(t)$ is the value of α at time t. In this thesis, we suppose

$$\tau_{\rm CG} \ll \tau. \tag{2.59}$$

Moreover, $\tau_{\rm CG}$ should be much shorter than the relaxation time of the system, τ_S:

$$\tau_{\rm CG} \ll \tau_S. \tag{2.60}$$

For the adiabatic modulation, $\tau_S \ll \tau$ should hold, then $\tau_{\rm CG} \ll \tau_S \ll \tau$ holds.

In general, the FCS-QME is given by

$$\frac{d\rho^\chi(t)}{dt} = -i[H_S(\alpha_S(t)), \rho^\chi(t)] + \sum_b \mathcal{L}^\chi_b(t)\rho^\chi(t), \tag{2.61}$$

with the initial condition

$$\rho^\chi(0) = \rho(0). \tag{2.62}$$

$\mathcal{L}^\chi_b(t)$ describes the coupling effects between S and the bath b and depends on used approximations. In this thesis, we suppose

$$\mathcal{L}^\chi_b(t) = \mathcal{L}^\chi_b(\alpha_t). \tag{2.63}$$

The Born-Markov approximation without or within the RWA and the CGA satisfy this equation. Then, the FCS-QME is given by

$$\frac{d\rho^{\chi}(t)}{dt} = \hat{K}^{\chi}(\alpha_t)\rho^{\chi}(t). \tag{2.64}$$

Here,

$$\hat{K}^{\chi}(\alpha)\bullet = -i[H_S(\alpha_S), \bullet] + \sum_b \mathcal{L}_b^{\chi}(\alpha)\bullet, \tag{2.65}$$

is the Liouvillian. Here and in the following, $\bullet$ denotes an arbitrary liner operator of the system.

The Born-Markov approximation is given by

$$\frac{d\rho^{I,\chi}(t)}{dt} = -\int_0^{\infty} ds\ \mathrm{Tr}_B\Big\{[H_{\mathrm{int}}^I(t), [H_{\mathrm{int}}^I(t-s), \rho^{I,\chi}(t)\rho_B(\alpha_B(t))]_{\chi}]_{\chi}\Big\}. \tag{2.66}$$

2.2.2 Coarse-graining approximation

In general, the interaction Hamiltonian is given by

$$H_{Sb}(\alpha_{Sb}) = \sum_{\mu} s_{b\mu}R_{b,\mu}(\alpha_{Sb}) = \sum_{\mu} R_{b,\mu}^{\dagger}(\alpha_{Sb})s_{b\mu}^{\dagger}. \tag{2.67}$$

Here, $s_{b\mu}$ is an operator of the system and $R_{b,\mu}(\alpha_{Sb})$ is an operator of the bath b. We suppose

$$\mathrm{Tr}_b[\rho_b(\alpha_b(t))R_{b,\mu}(\alpha_{Sb}(s))] = 0, \tag{2.68}$$

corresponding to (2.56). Then,

$$\begin{aligned}
&\mathrm{Tr}_B\Big\{[H_{\mathrm{int}}^I(u), [H_{\mathrm{int}}^I(s), \rho^{I,\chi}(t)\rho_B(\alpha_B(t))]_{\chi}]_{\chi}\Big\} \\
&= \sum_b \sum_{\mu,\nu} \Big(s_{b\nu}^{I\dagger}(u)s_{b\mu}^I(s)\rho^{I,\chi}(t)\mathrm{Tr}_b[R_{b,\nu,\chi}^{I\dagger}(u)R_{b,\mu,\chi}^I(s)\rho_b(\alpha_b(t))] \\
&\quad -s_{b\mu}^I(s)\rho^{I,\chi}(t)s_{b\nu}^{I\dagger}(u)\mathrm{Tr}_b[R_{b,\mu,\chi}^I(s)\rho_b(\alpha_b(t))R_{b,\nu,-\chi}^{I\dagger}(u)] \\
&\quad -s_{b\nu}^I(u)\rho^{I,\chi}(t)s_{b\mu}^{I\dagger}(s)\mathrm{Tr}_b[R_{b,\nu,\chi}^I(u)\rho_b(\alpha_b(t))R_{b,\mu,-\chi}^{I\dagger}(s)] \\
&\quad +\rho^{I,\chi}(t)s_{b\mu}^{I\dagger}(s)s_{b\nu}^I(u)\mathrm{Tr}_b[\rho_b(\alpha_b(t))R_{b,\mu,-\chi}^{I\dagger}(s)R_{b,\nu,-\chi}^I(u)]\Big),
\end{aligned} \tag{2.69}$$

holds. In the calculation of $\mathrm{Tr}_b[R_{b,\nu,\chi}^{I\dagger}(u)R_{b,\mu,\chi}^I(s)\rho_b(\alpha_b(t))]$, the values of the control parameters can be approximated by α_t. Then, we obtain

$$\mathrm{Tr}_b[R_{b,\nu,\chi}^{I\dagger}(u)R_{b,\mu,\chi}^I(s)\rho_b] \approx \mathrm{Tr}_b[\rho_b R_{b,\nu}^{\dagger}(u-s)R_{b,\mu}] \equiv C_{b,\nu\mu}(u-s), \tag{2.70}$$

$$\mathrm{Tr}_b[R_{b,\mu,\chi}^I(s)\rho_b R_{b,\nu,-\chi}^{I\dagger}(u)] \approx \mathrm{Tr}_b[\rho_b R_{b,\nu,-2\chi}^{\dagger}(u-s)R_{b,\mu}] \equiv C_{b,\nu\mu}^{\chi}(u-s), \tag{2.71}$$

$$\mathrm{Tr}_b[R_{b,\nu,\chi}^I(u)\rho_b R_{b,\mu,-\chi}^{I\dagger}(s)] \approx \mathrm{Tr}_b[\rho_b R_{b,\mu,-2\chi}^{\dagger}(s-u)R_{b,\nu}] = C_{b,\mu\nu}^{\chi}(s-u), \tag{2.72}$$

$$\mathrm{Tr}_b[\rho_b R_{b,\mu,-\chi}^{I\dagger}(s)R_{b,\nu,-\chi}^I(u)] \approx \mathrm{Tr}_b[\rho_b R_{b,\mu}^{\dagger}(s-u)R_{b,\nu}] = C_{b,\mu\nu}(s-u), \tag{2.73}$$

with

$$R_{b,\nu}^{\dagger}(v) = e^{iH_b(\alpha_b(t))v}R_{b,\nu}^{\dagger}(\alpha_{Sb}(t))e^{-iH_b(\alpha_b(t))v}. \tag{2.74}$$

Here, $\rho_b = \rho_b(\alpha_b(t))$ and $R_{b,\mu} = R_{b,\mu}(\alpha_b(t))$. Then, (2.69) becomes

$$\begin{aligned}
&\mathrm{Tr}_B\Big\{[H^I_{\mathrm{int}}(u), [H^I_{\mathrm{int}}(s), \rho^{I,\chi}(t)\rho_B(\alpha_B(t))]_\chi]_\chi\Big\} \\
&= \sum_b \sum_{\mu,\nu} \Big(s^{I\dagger}_{b\nu}(u) s^I_{b\mu}(s) \rho^{I,\chi}(t) C_{b,\nu\mu}(u-s) - s^I_{b\mu}(s)\rho^{I,\chi}(t) s^{I\dagger}_{b\nu}(u) C^\chi_{b,\nu\mu}(u-s) \\
&\quad - s^I_{b\nu}(u)\rho^{I,\chi}(t) s^{I\dagger}_{b\mu}(s) C^\chi_{b,\mu\nu}(s-u) + \rho^{I,\chi}(t) s^{I\dagger}_{b\mu}(s) s^I_{b\nu}(u) C_{b,\mu\nu}(s-u)\Big), \qquad (2.75)
\end{aligned}$$

and

$$\begin{aligned}
&\mathcal{L}^\chi_{b,\tau_{\mathrm{CG}}}(\alpha_t)\bullet \\
&= -\frac{1}{\tau_{\mathrm{CG}}}\int_t^{t+\tau_{\mathrm{CG}}} du \int_t^u ds \sum_{\mu,\nu} \Big(s^{I\dagger}_{b\nu}(u,t) s^I_{b\mu}(s,t) \bullet C_{b,\nu\mu}(u-s) \\
&\quad - s^I_{b\mu}(s,t) \bullet s^{I\dagger}_{b\nu}(u,t) C^\chi_{b,\nu\mu}(u-s) \\
&\quad - s^I_{b\nu}(u,t) \bullet s^{I\dagger}_{b\mu}(s,t) C^\chi_{b,\mu\nu}(s-u) + \bullet s^{I\dagger}_{b\mu}(s,t) s^I_{b\nu}(u,t) C_{b,\mu\nu}(s-u)\Big), \qquad (2.76)
\end{aligned}$$

holds. Here,

$$s^I_{b\mu}(s,t) = U_S(t) U_S^\dagger(s) s_{b\mu} U_S(s) U_S^\dagger(t). \qquad (2.77)$$

and $U_S(t)$ is the solution of $\frac{dU_S(t)}{dt} = -iH_S(\alpha_S(t))U_S(t)$ for $U_S(0) = 1$. In the calculation of $s^I_{b\mu}(s,t)$, the values of the control parameters can be approximated by α_t. Then, we obtain

$$s^I_{b\mu}(s,t) = \sum_\omega e^{-i\omega(s-t)} s_{b\mu}(\omega), \qquad (2.78)$$

$$s^{I\dagger}_{b\nu}(u,t) = \sum_\omega e^{i\omega(u-t)} [s_{b\nu}(\omega)]^\dagger. \qquad (2.79)$$

Here, the eigenoperator $s_{b\mu}(\omega)$ is defined by

$$s_{b\mu}(\omega) = \sum_{n,m,r,s} \delta_{\omega_{mn},\omega} |E_n, r\rangle\langle E_n, r| s_{b\mu} |E_m, s\rangle\langle E_m, s|, \qquad (2.80)$$

with $\omega_{mn} = E_m - E_n$ and

$$H_S|E_n, r\rangle = E_n|E_n, r\rangle. \qquad (2.81)$$

r denotes the label of the degeneracy. ω is one of the elements of $\{\omega_{mn} |\ \langle E_n, r|s_{b\mu}|E_m, s\rangle \neq 0\ ^\exists\mu\}$. $s_{b\mu}(\omega)$ and ω depend on α_S. The eigenoperators satisfy

$$\sum_\omega s_{b\mu}(\omega) = s_{b\mu}, \qquad (2.82)$$

and

$$[H_S, s_{b\mu}(\omega)] = -\omega s_{b\mu}(\omega). \qquad (2.83)$$

Then, we obtain

$$\begin{aligned}\mathcal{L}^{\chi}_{b,\tau_{\rm CG}}(\alpha)\bullet \;=\; -\frac{1}{\tau_{\rm CG}}\int_t^{t+\tau_{\rm CG}} du \int_t^{t+\tau_{\rm CG}} ds \sum_{\mu,\nu}\sum_{\omega,\omega'}\theta(u-s)\\ \times\Big(\Big\{[s_{b\nu}(\omega')]^\dagger s_{b\mu}(\omega)\bullet C_{b,\nu\mu}(u-s)\\ -s_{b\mu}(\omega)\bullet[s_{b\nu}(\omega')]^\dagger C^{\chi}_{b,\nu\mu}(u-s)\Big\}e^{-i\omega(s-t)}e^{i\omega'(u-t)}\\ +\Big\{-s_{b\mu}(\omega)\bullet[s_{b\nu}(\omega')]^\dagger C^{\chi}_{b,\nu\mu}(s-u)\\ +\bullet[s_{b\nu}(\omega')]^\dagger s_{b\mu}(\omega)C_{b,\nu\mu}(s-u)\Big\}e^{i\omega'(s-t)}e^{-i\omega(u-t)}\Big). \qquad (2.84)\end{aligned}$$

In last two terms, we swapped μ and ν. $\theta(u-s)$ is the step function.

Now, we introduce

$$\Phi^{\chi}_{b,\nu\mu}(\Omega) \stackrel{\rm def}{=} \int_{-\infty}^{\infty} du\, C^{\chi}_{b,\nu\mu}(u)e^{i\Omega u}. \qquad (2.85)$$

Then,

$$\begin{aligned}\int_{-\infty}^{\infty} du\, C^{\chi}_{b,\nu\mu}(u)\theta(u)e^{i\omega u} &= \frac{1}{2\pi}\int_0^{\infty} du \int_{-\infty}^{\infty} d\Omega\, \Phi^{\chi}_{b,\nu\mu}(\Omega)e^{-i\Omega u}e^{i\omega u}\\ &= \frac{1}{2\pi}\int_{-\infty}^{\infty} d\Omega\left[\pi\delta(\Omega-\omega)-i\frac{\rm P}{\Omega-\omega}\right]\Phi^{\chi}_{b,\nu\mu}(\Omega)\\ &= \frac{1}{2}\Phi^{\chi}_{b,\nu\mu}(\omega)-\frac{i}{2}\Psi^{\chi}_{b,\nu\mu}(\omega)=\Phi^{(+)\chi}_{b,\nu\mu}(\omega), \qquad (2.86)\end{aligned}$$

holds. Here, P denotes the Cauchy principal value and

$$\Psi^{\chi}_{b,\nu\mu}(\omega) \stackrel{\rm def}{=} \frac{\rm P}{\pi}\int_{-\infty}^{\infty} d\Omega\, \frac{\Phi^{\chi}_{b,\nu\mu}(\Omega)}{\Omega-\omega}, \qquad (2.87)$$

$$\Phi^{(\pm)\chi}_{b,\nu\mu}(\Omega) \stackrel{\rm def}{=} \frac{1}{2}\Phi^{\chi}_{b,\nu\mu}(\omega)\mp\frac{i}{2}\Psi^{\chi}_{b,\nu\mu}(\omega). \qquad (2.88)$$

(2.86) leads

$$C^{\chi}_{b,\nu\mu}(u-s)\theta(u-s) \;=\; \int_{-\infty}^{\infty} d\Omega\, \frac{\Phi^{(+)\chi}_{b,\nu\mu}(\Omega)}{2\pi}e^{-i\Omega(u-s)}. \qquad (2.89)$$

Similarly,

$$C^{\chi}_{b,\nu\mu}(s-u)\theta(u-s) \;=\; \int_{-\infty}^{\infty} d\Omega\, \frac{\Phi^{(-)\chi}_{b,\nu\mu}(\Omega)}{2\pi}e^{i\Omega(u-s)}, \qquad (2.90)$$

holds. Then, we obtain

$$\begin{aligned}\mathcal{L}^{\chi}_{b,\tau_{\rm CG}}(\alpha)\bullet \;=\; & -\frac{1}{\tau_{\rm CG}}\int_t^{t+\tau_{\rm CG}} du \int_t^{t+\tau_{\rm CG}} ds \int_{-\infty}^{\infty}\frac{d\Omega}{2\pi}\sum_{\mu,\nu}\sum_{\omega,\omega'} \\ & \times\Big(\Big\{[s_{b\nu}(\omega')]^{\dagger}s_{b\mu}(\omega)\bullet\Phi^{(+)}_{b,\nu\mu}(\Omega) \\ & -s_{b\mu}(\omega)\bullet[s_{b\nu}(\omega')]^{\dagger}\Phi^{(+)\chi}_{b,\nu\mu}(\Omega)\Big\}e^{-i\Omega(u-s)}e^{-i\omega(s-t)}e^{i\omega'(u-t)} \\ & +\Big\{-s_{b\mu}(\omega)\bullet[s_{b\nu}(\omega')]^{\dagger}\Phi^{(-)\chi}_{b,\nu\mu}(\Omega) \\ & +\bullet[s_{b\nu}(\omega')]^{\dagger}s_{b\mu}(\omega)\Phi^{(-)}_{b,\nu\mu}(\Omega)\Big\}e^{i\Omega(u-s)}e^{i\omega'(s-t)}e^{-i\omega(u-t)}\Big), \end{aligned} \tag{2.91}$$

with $\Phi^{(\pm)}_{b,\nu\mu}=\Phi^{(\pm)\chi}_{b,\nu\mu}\big|_{\chi=0}$. The integrals for u and s are performed as

$$\int_t^{t+\tau_{\rm CG}} du\; e^{-i\Omega u}e^{i\omega'(u-t)} \;=\; \tau_{\rm CG}e^{-i\Omega t-i[\Omega-\omega']\tau_{\rm CG}/2}{\rm sinc}([\Omega-\omega']\tau_{\rm CG}/2), \tag{2.92}$$

$$\int_t^{t+\tau_{\rm CG}} ds\; e^{i\Omega s}e^{-i\omega(s-t)} \;=\; \tau_{\rm CG}e^{i\Omega t+i[\Omega-\omega]\tau_{\rm CG}/2}{\rm sinc}([\Omega-\omega]\tau_{\rm CG}/2), \tag{2.93}$$

then

$$\begin{aligned}\mathcal{L}^{\chi}_{b,\tau_{\rm CG}}(\alpha)\bullet \;=\; & -\sum_{\mu,\nu}\sum_{\omega,\omega'}\frac{e^{-i(\omega-\omega')/\tau_{\rm CG}}}{2\pi}\int_{-\infty}^{\infty}d\Omega\;\Big([s_{b\nu}(\omega')]^{\dagger}s_{b\mu}(\omega)\bullet\Phi^{(+)}_{b,\nu\mu}(\Omega) \\ & -s_{b\mu}(\omega)\bullet[s_{b\nu}(\omega')]^{\dagger}\Phi^{(+)\chi}_{b,\nu\mu}(\Omega) \\ & -s_{b\mu}(\omega)\bullet[s_{b\nu}(\omega')]^{\dagger}\Phi^{(-)\chi}_{b,\nu\mu}(\Omega)+\bullet[s_{b\nu}(\omega')]^{\dagger}s_{b\mu}(\omega)\Phi^{(-)}_{b,\nu\mu}(\Omega)\Big) \\ & \times\tau_{\rm CG}{\rm sinc}\frac{[\Omega-\omega']\tau_{\rm CG}}{2}{\rm sinc}\frac{[\Omega-\omega]\tau_{\rm CG}}{2}, \end{aligned} \tag{2.94}$$

holds. Here, ${\rm sinc}(x)=\sin x/x$. The above equation can be rewritten as

$$\mathcal{L}^{\chi}_{b,\tau_{\rm CG}}(\alpha)\bullet \;=\; -i[h_{b,\tau_{\rm CG}}(\alpha),\bullet]+\Pi^{\chi}_{b,\tau_{\rm CG}}(\alpha)\bullet, \tag{2.95}$$

$$\begin{aligned}\Pi^{\chi}_{b,\tau_{\rm CG}}(\alpha)\bullet \;=\; & \sum_{\omega,\omega'}\sum_{\mu,\nu}\Big[\Phi^{\chi}_{b,\mu\nu}(\tau_{\rm CG},\omega,\omega')s_{b\nu}(\omega')\bullet[s_{b\mu}(\omega)]^{\dagger} \\ & -\frac{1}{2}\Phi_{b,\mu\nu}(\tau_{\rm CG},\omega,\omega')\bullet[s_{b\mu}(\omega)]^{\dagger}s_{b\nu}(\omega') \\ & -\frac{1}{2}\Phi_{b,\mu\nu}(\tau_{\rm CG},\omega,\omega')[s_{b\mu}(\omega)]^{\dagger}s_{b\nu}(\omega')\bullet\Big], \end{aligned} \tag{2.96}$$

with

$$h_{b,\tau_{\rm CG}}(\alpha) \;=\; -\frac{1}{2}\sum_{\omega,\omega'}\sum_{\mu,\nu}\Psi_{b,\mu\nu}(\tau_{\rm CG},\omega,\omega')[s_{b\mu}(\omega)]^{\dagger}s_{b\nu}(\omega'). \tag{2.97}$$

Here,

$$\begin{aligned} & X^{\chi}(\tau_{\rm CG},\omega,\omega') \\ & \;=\; \frac{e^{i(\omega-\omega')\tau_{\rm CG}/2}}{2\pi}\int_{-\infty}^{\infty}d\Omega\;X^{\chi}(\Omega)\tau_{\rm CG}{\rm sinc}\Big(\frac{\tau_{\rm CG}(\Omega-\omega)}{2}\Big){\rm sinc}\Big(\frac{\tau_{\rm CG}(\Omega-\omega')}{2}\Big), \end{aligned} \tag{2.98}$$

with $X = \Phi_{b,\mu\nu}, \Psi_{b,\mu\nu}$. $\Pi_{b,\tau_{\rm CG}} = \Pi^{\chi}_{b,\tau_{\rm CG}}\big|_{\chi=0}$ is the Lindblad type. By the way, from

$$[C_{b,\mu\nu}(t)]^* = C_{b,\nu\mu}(-t), \tag{2.99}$$

relations

$$[\Phi_{b,\mu\nu}(\Omega)]^* = \Phi_{b,\nu\mu}(\Omega), \tag{2.100}$$

and $[\Psi_{\mu\nu}(\Omega)]^* = \Psi_{\nu\mu}(\Omega)$ hold. Then,

$$[\Phi_{b,\mu\nu}(\tau_{\rm CG},\omega,\omega')]^* = \Phi_{b,\nu\mu}(\tau_{\rm CG},\omega',\omega), \tag{2.101}$$

holds.

For super-operator $\mathcal{J}$, $\mathcal{J}^\dagger$ is defined by

$$\mathrm{Tr}_S(Y^\dagger \mathcal{J} X) = \mathrm{Tr}_S([\mathcal{J}^\dagger Y]^\dagger X), \tag{2.102}$$

where $X, Y \in \mathrm{B}$. If $\mathcal{J}\bullet = \sum_a A_a \bullet B_a$ holds,

$$\mathcal{J}^\dagger \bullet = \sum_a A_a^\dagger \bullet B_a^\dagger, \tag{2.103}$$

is obtained. Here, $A_a, B_a \in \mathrm{B}$. (2.101) leads

$$\begin{aligned}\Pi^\dagger_{b,\tau_{\rm CG}}(\alpha)\bullet = \sum_{\omega,\omega'}\sum_{\mu,\nu}\Big[&\Phi_{b,\mu\nu}(\tau_{\rm CG},\omega,\omega')[s_{b\mu}(\omega)]^\dagger \bullet s_{b\nu}(\omega') \\ &-\frac{1}{2}\Phi_{b,\mu\nu}(\tau_{\rm CG},\omega,\omega')\bullet[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega') \\ &-\frac{1}{2}\Phi_{b,\mu\nu}(\tau_{\rm CG},\omega,\omega')[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega')\bullet\Big].\end{aligned} \tag{2.104}$$

This leads

$$\Pi^\dagger_{b,\tau_{\rm CG}}(\alpha)1 = 0, \tag{2.105}$$

which means the conservation of the probability.

2.2.3 Concrete model

In this subsection, we consider $b = n_{\rm C}+1, \cdots, n_{\rm C}+n_{\rm GC}$. Now we suppose

$$H_{Sb}(\alpha_{Sb}) = \sum_\alpha a^\dagger_\alpha B_{b\alpha} + \text{h.c.},\ B_{b\alpha} = \sum_{k,\sigma} V_{bk\sigma,\alpha}(\alpha_{Sb})c_{bk\sigma}\ (b \in \boldsymbol{\mathcal{G}}), \tag{2.106}$$

where a_α and $c_{bk\sigma}$ are single-particle annihilation operators of the system and of the bath b. Using

$$\mathrm{Tr}_b[\rho_b B^I_{b\alpha}(t')B^I_{b\beta}(t'')] = 0 = \mathrm{Tr}_b[\rho_b B^{I\dagger}_{b\alpha}(t')B^{I\dagger}_{b\beta}(t'')], \tag{2.107}$$

we obtain

$$\begin{aligned}
\mathcal{L}^{\chi}_{b,\tau_{\rm CG}}(\alpha)\bullet &= -i[h_{b,\tau_{\rm CG}}(\alpha),\bullet]+\Pi^{\chi}_{b,\tau_{\rm CG}}(\alpha)\bullet,\\
\Pi^{\chi}_{b,\tau_{\rm CG}}(\alpha)\bullet &= \sum_{\omega,\omega'}\sum_{\alpha,\beta}\Big[\Phi^{-,\chi}_{b,\alpha\beta}(\tau_{\rm CG},\omega,\omega')a_\beta(\omega')\bullet[a_\alpha(\omega)]^\dagger\\
&\quad -\frac{1}{2}\Phi^{-}_{b,\alpha\beta}(\tau_{\rm CG},\omega,\omega')\bullet[a_\alpha(\omega)]^\dagger a_\beta(\omega')\\
&\quad -\frac{1}{2}\Phi^{-}_{b,\alpha\beta}(\tau_{\rm CG},\omega,\omega')[a_\alpha(\omega)]^\dagger a_\beta(\omega')\bullet\\
&\quad +\Phi^{+,\chi}_{b,\alpha\beta}(\tau_{\rm CG},\omega,\omega')[a_\beta(\omega')]^\dagger\bullet a_\alpha(\omega)\\
&\quad -\frac{1}{2}\Phi^{+}_{b,\alpha\beta}(\tau_{\rm CG},\omega,\omega')\bullet a_\alpha(\omega)[a_\beta(\omega')]^\dagger\\
&\quad -\frac{1}{2}\Phi^{+}_{b,\alpha\beta}(\tau_{\rm CG},\omega,\omega')a_\alpha(\omega)[a_\beta(\omega')]^\dagger\bullet\Big],
\end{aligned}\tag{2.108}$$

and

$$\begin{aligned}
h_{b,\tau_{\rm CG}}(\alpha) &= \sum_{\omega,\omega'}\sum_{\alpha,\beta}\Big[-\frac{1}{2}\Psi^{-}_{b,\alpha\beta}(\tau_{\rm CG},\omega,\omega')[a_\alpha(\omega)]^\dagger a_\beta(\omega')\\
&\quad +\frac{1}{2}\Psi^{+}_{b,\alpha\beta}(\tau_{\rm CG},\omega,\omega')a_\alpha(\omega)[a_\beta(\omega')]^\dagger\Big].
\end{aligned}\tag{2.109}$$

The eigenoperators $a_\alpha(\omega)$ are given by

$$a_\alpha(\omega) = \sum_{n,m,r,s}\delta_{\omega_{mn},\omega}|E_n,r\rangle\langle E_n,r|a_\alpha|E_m,s\rangle\langle E_m,s|.\tag{2.110}$$

ω is one of the elements of $\{\omega_{mn}|\ \langle E_n,r|a_\alpha|E_m,s\rangle\neq 0\ \ ^{\exists}\alpha\}$. $a_\alpha(\omega)$ satisfy

$$\sum_{\omega}a_\alpha(\omega)=a_\alpha,\tag{2.111}$$

and

$$[H_S,a_\alpha(\omega)]=-\omega a_\alpha(\omega),\ [N_S,a_\alpha(\omega)]=-a_\alpha(\omega).\tag{2.112}$$

N_S is total number operator of the system. Here and in the following, we suppose

$$[N_S,H_S]=0.\tag{2.113}$$

If $n_{\rm GC}=0$, existence of N_S and the above equation are not required. In (2.108) and (2.109),

$$\begin{aligned}
&X^{\pm,\chi}(\tau_{\rm CG},\omega,\omega')\\
&\quad=\frac{e^{\pm i(\omega-\omega')\tau_{\rm CG}/2}}{2\pi}\int_{-\infty}^{\infty}d\Omega\,X^{\pm,\chi}(\Omega)\tau_{\rm CG}{\rm sinc}\Big(\frac{\tau_{\rm CG}(\Omega-\omega)}{2}\Big){\rm sinc}\Big(\frac{\tau_{\rm CG}(\Omega-\omega')}{2}\Big),
\end{aligned}\tag{2.114}$$

and $X^{\pm}(\tau_{\rm CG},\omega,\omega') = X^{\pm,\chi}(\tau_{\rm CG},\omega,\omega')\big|_{\chi=0}$. Here, $X^{\pm,\chi}(\Omega)$ denotes one of $\Phi^{\pm,\chi}_{b,\alpha,\beta}(\Omega)$, $\Psi^{\pm,\chi}_{b,\alpha\beta}(\Omega)$, where

$$\Phi^{-,\chi}_{b,\alpha\beta}(\Omega) \;=\; \int_{-\infty}^{\infty} du\ {\rm Tr}_b[\rho_b B^I_{b\alpha,-2\chi}(u) B^\dagger_{b\beta}] e^{i\Omega u}, \tag{2.115}$$

$$\Phi^{+,\chi}_{b,\alpha\beta}(\Omega) \;=\; \int_{-\infty}^{\infty} du\ {\rm Tr}_b[\rho_b B^{\dagger I}_{b\alpha,-2\chi}(u) B_{b\beta}] e^{-i\Omega u}, \tag{2.116}$$

$$\Psi^{\pm,\chi}_{b,\alpha\beta}(\Omega) \stackrel{\rm def}{=} \frac{\rm P}{\pi}\int_{-\infty}^{\infty} d\Omega'\ \frac{\Phi^{\pm,\chi}_{b,\alpha\beta}(\Omega')}{\Omega'-\Omega}. \tag{2.117}$$

We set $\{O_\mu\} = \{N_b\}_{b\in\boldsymbol{\mathcal{G}}} + \{H_b\}_b$, where

$$N_b \;=\; \sum_{k,\sigma} c^\dagger_{bk\sigma} c_{bk\sigma}. \tag{2.118}$$

Whenever H_b is an element of $\{O_\mu\}$, we suppose α'_b are fixed. We introduce the eigenoperator

$$B_{b\alpha}(\Omega_b) \;=\; \sum_{n,m,r,s} \delta_{\Omega_{b,mn},\Omega_b} |E_{b,n},r\rangle\langle E_{b,n},r|B_{b\alpha}|E_{b,m},s\rangle\langle E_{b,m},s|, \tag{2.119}$$

with $\Omega_{b,mn} = E_{b,m} - E_{b,n}$ and $H_b|E_{b,n},r\rangle = E_{b,n}|E_{b,n},r\rangle$. r denotes the label of the degeneracy. Ω_b is one of the elements of $\{\Omega_{b,mn}|\ \langle E_{b,n},r|B_{b\alpha}|E_{b,m},s\rangle \neq 0\ \ ^\exists\alpha\}$. The relations

$$\sum_{\Omega_b} B_{b\alpha}(\Omega_b) = B_{b\alpha}, \tag{2.120}$$

and

$$[H_b, B_{b\alpha}(\Omega_b)] = -\Omega_b B_{b\alpha}(\Omega_b),\ [N_b, B_{b\alpha}(\Omega_b)] = -B_{b\alpha}(\Omega_b) \tag{2.121}$$

hold. Then, we obtain

$$B^I_{b\alpha,-2\chi}(u) \;=\; \sum_{\Omega_b} B_{b\alpha}(\Omega_b) e^{-i\Omega_b u + i\chi_{H_b}\Omega_b + i\chi_{N_b}}, \tag{2.122}$$

$$B^{\dagger I}_{b\alpha,-2\chi}(u) \;=\; \sum_{\Omega_b} [B_{b\alpha}(\Omega_b)]^\dagger e^{i\Omega_b u - i\chi_{H_b}\Omega_b - i\chi_{N_b}}, \tag{2.123}$$

and

$$\begin{aligned}\Phi^{-,\chi}_{b,\alpha\beta}(\Omega) \;&=\; 2\pi\sum_{\Omega_b}\delta(\Omega-\Omega_b) e^{i\chi_{H_b}\Omega_b + i\chi_{N_b}} {\rm Tr}_b(\rho_b B_{b\alpha}(\Omega_b) B^\dagger_{b\beta}) \\ &=\; e^{i\chi_{H_b}\Omega + i\chi_{N_b}} 2\pi\sum_{\Omega_b}\delta(\Omega-\Omega_b){\rm Tr}_b(\rho_b B_{b\alpha}(\Omega_b)[B_{b\beta}(\Omega_b)]^\dagger),\end{aligned} \tag{2.124}$$

$$\begin{aligned}\Phi^{+,\chi}_{b,\alpha\beta}(\Omega) \;&=\; 2\pi\sum_{\Omega_b}\delta(\Omega-\Omega_b) e^{-i\chi_{H_b}\Omega_b - i\chi_{N_b}} {\rm Tr}_b(\rho_b [B_{b\alpha}(\Omega_b)]^\dagger B_{b\beta}) \\ &=\; e^{-i\chi_{H_b}\Omega - i\chi_{N_b}} 2\pi\sum_{\Omega_b}\delta(\Omega-\Omega_b){\rm Tr}_b(\rho_b[B_{b\alpha}(\Omega_b)]^\dagger B_{b\beta}(\Omega_b)).\end{aligned} \tag{2.125}$$

Here, we used (2.120) and $\mathrm{Tr}_b(\rho_b B_{b\alpha}(\Omega_b)[B_{b\beta}(\Omega'_b)]^\dagger) = 0$ and $\mathrm{Tr}_b(\rho_b[B_{b\alpha}(\Omega_b)]^\dagger B_{b\beta}(\Omega'_b)) = 0$ for $\Omega_b \neq \Omega'_b$. Then, we obtain

$$\Phi^{\pm,\chi}_{b,\alpha\beta}(\Omega) = e^{\mp(i\chi_{H_b}\Omega + i\chi_{N_b})}\Phi^{\pm}_{b,\alpha\beta}(\Omega), \tag{2.126}$$

with $\Phi^{\pm}_{b,\alpha\beta}(\Omega) = \Phi^{\pm,\chi}_{b,\alpha\beta}(\Omega)\big|_{\chi=0}$ and

$$\Psi^{-}_{b,\alpha\beta}(\Omega) = 2\sum_{\Omega_b} \mathrm{P}\frac{1}{\Omega_b - \Omega}\mathrm{Tr}_b(\rho_b B_{b\alpha}(\Omega_b)[B_{b\beta}(\Omega_b)]^\dagger), \tag{2.127}$$

$$\Psi^{+}_{b,\alpha\beta}(\Omega) = 2\sum_{\Omega_b} \mathrm{P}\frac{1}{\Omega_b - \Omega}\mathrm{Tr}_b(\rho_b [B_{b\alpha}(\Omega_b)]^\dagger B_{b\beta}(\Omega_b)). \tag{2.128}$$

$\Phi^{\pm}_{b,\alpha\beta}(\Omega)$ satisfy

$$[\Phi^{\pm}_{b,\alpha\beta}(\Omega)]^* = \Phi^{\pm}_{b,\beta\alpha}(\Omega), \tag{2.129}$$

$$\Phi^{+}_{b,\alpha\beta}(\Omega) = e^{-\beta_b(\Omega-\mu_b)}\Phi^{-}_{b,\beta\alpha}(\Omega). \tag{2.130}$$

The latter is the Kubo-Martin-Schwinger (KMS) condition. (2.130) is derived from $\rho_b B_{b\alpha}(\Omega_b) = e^{\beta_b(\Omega_b-\mu)}B_{b\alpha}(\Omega_b)\rho_b$ (derived from (2.121)) and (2.124) and (2.125).

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

$$H_b(\alpha'_b) = \sum_{k,\sigma} \varepsilon_{bk\sigma}(\alpha'_b) c^\dagger_{bk\sigma} c_{bk\sigma}, \tag{2.131}$$

and $\{O_\mu\} = \{N_{b\sigma}\}_{b\sigma}$ with

$$N_{b\sigma} = \sum_k c^\dagger_{bk\sigma} c_{bk\sigma}. \tag{2.132}$$

In this case, α'_b can depend on time and

$$\Phi^{-,\chi}_{b,\alpha\beta}(\Omega) = 2\pi\sum_{k,\sigma} V_{bk\sigma,\alpha}V^*_{bk\sigma,\beta}F^-_b(\varepsilon_{bk\sigma})e^{i\chi_{b\sigma}}\delta(\varepsilon_{bk\sigma}-\Omega), \tag{2.133}$$

$$\Phi^{+,\chi}_{b,\alpha\beta}(\Omega) = 2\pi\sum_{k,\sigma} V^*_{bk\sigma,\alpha}V_{bk\sigma,\beta}F^+_b(\varepsilon_{bk\sigma})e^{-i\chi_{b\sigma}}\delta(\varepsilon_{bk\sigma}-\Omega), \tag{2.134}$$

$$\Psi^{-,\chi}_{b,\alpha\beta}(\Omega) = 2\sum_{k,\sigma} V_{bk\sigma,\alpha}V^*_{bk\sigma,\beta}F^-_b(\varepsilon_{bk\sigma})e^{i\chi_{b\sigma}}\mathrm{P}\frac{1}{\varepsilon_{bk\sigma}-\Omega}, \tag{2.135}$$

$$\Psi^{+,\chi}_{b,\alpha\beta}(\Omega) = 2\sum_{k,\sigma} V^*_{bk\sigma,\alpha}V_{bk\sigma,\beta}F^+_b(\varepsilon_{bk\sigma})e^{-i\chi_{b\sigma}}\mathrm{P}\frac{1}{\varepsilon_{bk\sigma}-\Omega}, \tag{2.136}$$

hold. $\chi_{b\sigma}$ denotes the counting fields for $N_{b\sigma}$. If the baths are fermions, $F^+_b(\varepsilon) = f_b(\varepsilon) \overset{\mathrm{def}}{=} [\exp(\beta_b(\varepsilon-\mu_b))+1]^{-1}$ and $F^-_b(\varepsilon) = 1 - f_b(\varepsilon)$. If the baths are bosons, $F^+_b(\varepsilon) = n_b(\varepsilon) \overset{\mathrm{def}}{=} [\exp(\beta_b(\varepsilon-\mu_b))-1]^{-1}$ and $F^-_b(\varepsilon) = 1 + n_b(\varepsilon)$.

(2.106) can be generalized as

$$H_{Sb}(\alpha_{Sb}) = \sum_{n,\xi} s^\dagger_{(n)\xi} B_{b,(n)\xi} + \mathrm{h.c.}\ (b \in \boldsymbol{\mathcal{G}}), \tag{2.137}$$

with

$$[s_{(n)\xi}(\omega), N_S] = -ns_{(n)\xi}(\omega),\ [B_{b,(n)\xi}(\Omega_b), N_b] = -nB_{b,(n)\xi}(\Omega_b). \tag{2.138}$$

Here, $n = 1, 2, \cdots$, and $s_{(n)\xi}(\omega)$ and $B_{b,(n)\xi}(\Omega_b)$ are the eigenoperators.

2.3 Rotating wave approximation

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

$$\Phi^\chi_{b,\mu\nu}(\tau_{\rm CG}, \omega, \omega') \approx \Phi^\chi_{b,\mu\nu}(\omega)\delta_{\omega,\omega'},\ \Psi^\chi_{b,\mu\nu}(\tau_{\rm CG}, \omega, \omega') \approx \Psi^\chi_{b,\mu\nu}(\omega)\delta_{\omega,\omega'}, \tag{2.139}$$

hold because of the fact that

$$\lim_{\tau_{\rm CG}\to\infty} \tau_{\rm CG}{\rm sinc}\frac{\tau_{\rm CG}(\Omega - \omega)}{2}{\rm sinc}\frac{\tau_{\rm CG}(\Omega - \omega')}{2} = 2\pi\delta_{\omega,\omega'}\delta(\Omega - \omega). \tag{2.140}$$

If H_S is time independent, this RWA is equivalent to usual RWA. $\mathcal{L}^\chi_b(\alpha)$ is given by

$$\mathcal{L}^\chi_b(\alpha)\bullet = \Pi^\chi_b(\alpha) \bullet -i[h_b(\alpha), \bullet], \tag{2.141}$$

where $h_b(\alpha)$ is a Hermitian operator describing the Lamb shift. $H_{\rm L}(\alpha) \stackrel{\rm def}{=} \sum_b h_b(\alpha)$ is called the Lamb shift Hamiltonian. $\Pi^\chi_b(\alpha)$ and $h_b(\alpha)$ are given by

$$\begin{aligned}\Pi^\chi_b(\alpha)\bullet &= \sum_\omega\sum_{\mu,\nu}\Big[\Phi^\chi_{b,\mu\nu}(\omega)s_{b\nu}(\omega) \bullet [s_{b\mu}(\omega)]^\dagger \\ &\quad -\frac{1}{2}\Phi_{b,\mu\nu}(\omega) \bullet [s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega) - \frac{1}{2}\Phi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega) \bullet \Big], \qquad (2.142)\\ h_b(\alpha) &= -\frac{1}{2}\sum_\omega\sum_{\mu,\nu}\Psi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega). \qquad (2.143)\end{aligned}$$

Because of (2.83), $h_b(\alpha)$ commutes with $H_S(\alpha_S)$:

$$[h_b(\alpha), H_S(\alpha_S)] = 0. \tag{2.144}$$

We introduce projection super-operators $\mathcal{P}(\alpha_S)$ and $\mathcal{Q}(\alpha_S)$ by

$$\mathcal{P}(\alpha_S)|E_n, r\rangle\langle E_m, s| = \delta_{E_n,E_m}|E_n, r\rangle\langle E_m, s|, \tag{2.145}$$

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

$$\mathcal{Q}\hat{K}^\chi\mathcal{P} = 0 = \mathcal{P}\hat{K}^\chi\mathcal{Q}, \tag{2.146}$$

hold. This implies that the right eigenvalue equations (3.3) are decomposed into two closed systems of equations for $\mathcal{P}\rho^\chi_n$ and for $\mathcal{Q}\rho^\chi_n$. Thus, ρ^χ_n is an element of ${\rm B_P}$ or ${\rm B_Q}$. In particular, $\rho^\chi_0 \in {\rm B_P}$. Then, the matrix representation of $\rho_0(\alpha)$ by $|E_n, r\rangle$ is block diagonalized. This implies

$$[H_S(\alpha_S), \rho_0(\alpha)] = 0. \tag{2.147}$$

For (2.106), $\Pi_b^\chi(\alpha)$ in (2.141) is given by

$$\Pi_b^\chi(\alpha)\bullet = \sum_\omega \sum_{\alpha,\beta} \Big[\Phi_{b,\alpha\beta}^{-,\chi}(\omega) a_\beta(\omega) \bullet [a_\alpha(\omega)]^\dagger - \frac{1}{2}\Phi_{b,\alpha\beta}^{-}(\omega) \bullet [a_\alpha(\omega)]^\dagger a_\beta(\omega)$$
$$-\frac{1}{2}\Phi_{b,\alpha\beta}^{-}(\omega)[a_\alpha(\omega)]^\dagger a_\beta(\omega) \bullet + \Phi_{b,\alpha\beta}^{+,\chi}(\omega)[a_\beta(\omega)]^\dagger \bullet a_\alpha(\omega)$$
$$-\frac{1}{2}\Phi_{b,\alpha\beta}^{+}(\omega) \bullet a_\alpha(\omega)[a_\beta(\omega)]^\dagger - \frac{1}{2}\Phi_{b,\alpha\beta}^{+}(\omega) a_\alpha(\omega)[a_\beta(\omega)]^\dagger \bullet \Big]. \quad (2.148)$$

The Lamb shift is given by

$$h_b(\alpha) = \sum_\omega \sum_{\alpha,\beta} \Big(-\frac{1}{2}\Psi_{b,\alpha\beta}^{-}(\omega)[a_\alpha(\omega)]^\dagger a_\beta(\omega) + \frac{1}{2}\Psi_{b,\alpha\beta}^{+}(\omega) a_\alpha(\omega)[a_\beta(\omega)]^\dagger \Big) \quad (2.149)$$

The second equation of (2.112) leads

$$[h_b(\alpha), N_S] = 0. \quad (2.150)$$

2.4 Detailed balance condition

In this section, we consider the RWA. If we suppose (2.106),

$$\Pi_b(\bullet e^{-\beta_b(H_S-\mu_b N_S)}) = (\Pi_b^\dagger \bullet) e^{-\beta_b(H_S-\mu_b N_S)} \ (b \in \boldsymbol{\mathcal{G}}), \quad (2.151)$$

holds using (2.130). This is the detailed balance condition. If we suppose (2.137), the above relation also holds. From $\mathcal{L}_b^\dagger 1 = \Pi_b^\dagger 1 = 0$ (see (2.105)) and (2.151) for $\bullet = 1$ lead

$$\Pi_b e^{-\beta_b(H_S-\mu_b N_S)} = \mathcal{L}_b e^{-\beta_b(H_S-\mu_b N_S)} = 0, \quad (2.152)$$

using (2.144) and (2.150). If the bath b is fermion, (2.106) or (2.137) are general.

In the following of this section, we consider canonical baths ($b \in \boldsymbol{\mathcal{C}}$). (2.143) leads

$$\Pi_b^\dagger(\alpha)\bullet = \sum_\omega \sum_{\mu,\nu} \Big[\Phi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^\dagger \bullet s_{b\nu}(\omega)$$
$$-\frac{1}{2}\Phi_{b,\mu\nu}(\omega) \bullet [s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega) - \frac{1}{2}\Phi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega) \bullet \Big]. \quad (2.153)$$

Then, we obtain

$$(\Pi_b^\dagger(\alpha)\bullet) e^{-\beta_b H_S} = \sum_\omega \sum_{\mu,\nu} \Big[\Phi_{b,\mu\nu}(\omega) e^{-\beta_b \omega}[s_{b\mu}(\omega)]^\dagger \bullet e^{-\beta_b H_S} s_{b\nu}(\omega)$$
$$-\frac{1}{2}\Phi_{b,\mu\nu}(\omega) \bullet e^{-\beta_b H_S}[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega)$$
$$-\frac{1}{2}\Phi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega) \bullet e^{-\beta_b H_S} \Big], \quad (2.154)$$

using (2.83). Then,

$$\begin{aligned}
&\Pi_b(\bullet e^{-\beta_b H_S}) - (\Pi_b^\dagger \bullet) e^{-\beta_b H_S} \\
&= \sum_\omega \sum_{\mu,\nu} \Big[\Phi_{b,\mu\nu}(\omega) s_{b\nu}(\omega) \bullet e^{-\beta_b H_S} [s_{b\mu}(\omega)]^\dagger \\
&\quad - \Phi_{b,\mu\nu}(\omega) e^{-\beta_b \omega} [s_{b\mu}(\omega)]^\dagger \bullet e^{-\beta_b H_S} s_{b\nu}(\omega) \Big] \\
&= \sum_\omega \sum_{\mu,\nu} \Big[\phi_{b,\mu\nu}(\omega) [s_{b\nu}(\omega)]^\dagger \bullet e^{-\beta_b H_S} s_{b\mu}(\omega) \\
&\quad - \Phi_{b,\mu\nu}(\omega) e^{-\beta_b \omega} [s_{b\mu}(\omega)]^\dagger \bullet e^{-\beta_b H_S} s_{b\nu}(\omega) \Big],
\end{aligned} \tag{2.155}$$

holds. Here, we used

$$\sum_\omega \sum_{\mu,\nu} \Phi_{b,\mu\nu}(\omega) s_{b\nu}(\omega) \bullet [s_{b\mu}(\omega)]^\dagger = \sum_\omega \sum_{\mu,\nu} \phi_{b,\mu\nu}(\omega) [s_{b\nu}(\omega)]^\dagger \bullet s_{b\mu}(\omega), \tag{2.156}$$

with

$$\phi_{b,\mu\nu}(\omega) = \int_{-\infty}^{\infty} du\, D_{b,\mu\nu}(u) e^{-i\omega u}, \tag{2.157}$$

$$D_{b,\mu\nu}(u) = \mathrm{Tr}_b[\rho_b R_{b,\mu}^I(u) R_{b,\nu}^\dagger],\ \rho_b = e^{-\beta_b H_b}/\mathrm{Tr}_b(e^{-\beta_b H_b}). \tag{2.158}$$

Using

$$\begin{aligned}
\mathrm{Tr}_b[\rho_b R_\mu^I(u) R_\nu^\dagger] &= \mathrm{Tr}_b[R_{b,\mu}^I(u + i\beta_b) \rho_b R_{b,\nu}^\dagger] \\
&= \mathrm{Tr}_b[\rho_b R_{b,\nu}^{I\dagger}(-u - i\beta_b) R_{b,\mu}] = C_{b,\nu\mu}(-u - i\beta_b),
\end{aligned} \tag{2.159}$$

$\phi_{b,\mu\nu}(\omega)$ is given by

$$\begin{aligned}
\phi_{b,\mu\nu}(\omega) &= \int_{-\infty}^{\infty} du\, C_{b,\nu\mu}(-u - i\beta_b) e^{-i\omega u} \\
&= \int_{-\infty}^{\infty} du \int_{-\infty}^{\infty} d\Omega\, \Phi_{b,\nu\mu}(\Omega) e^{i\Omega u - \beta_b \Omega} e^{-i\omega u} \\
&= \Phi_{b,\nu\mu}(\omega) e^{-\beta_b \omega}.
\end{aligned} \tag{2.160}$$

Substituting this into (2.155), we obtain

$$\Pi_b(\bullet e^{-\beta_b H_S}) = (\Pi_b^\dagger \bullet) e^{-\beta_b H_S}\ (b \in \boldsymbol{\mathcal{C}}). \tag{2.161}$$

Substituting $\bullet = 1$ to this equation, we get

$$\Pi_b e^{-\beta_b H_S} = 0. \tag{2.162}$$

If $n_{\mathrm{GC}} > 0$, we suppose

$$[s_{b\mu}(\omega), N_S] = 0\ (b \in \boldsymbol{\mathcal{C}}). \tag{2.163}$$

Then,

$$\Pi_b(\bullet e^{-\beta_b (H_S - \mu_b' N_S)}) = (\Pi_b^\dagger \bullet) e^{-\beta_b (H_S - \mu_b' N_S)}\ (b \in \boldsymbol{\mathcal{C}}), \tag{2.164}$$

and

$$\Pi_b e^{-\beta_b(H_S-\mu'_b N_S)} = \mathcal{L}_b e^{-\beta_b(H_S-\mu'_b N_S)} = 0, \tag{2.165}$$

hold. (2.164) is the detailed balance condition. Here, μ'_b is an arbitrary real number, and we used

$$[h_b(\alpha), N_S] = 0, \tag{2.166}$$

derived from (2.163). (2.163) and (2.104) lead

$$\Pi_b^\dagger N_S = 0 \ (b \in \boldsymbol{\mathcal{C}}). \tag{2.167}$$

Chapter 3

FCS-QME and quantum pump

3.1 Currents

Generally, $\mathcal{L}_b^\chi(\alpha)$ has the form:

$$\mathcal{L}_b^\chi(\alpha)\bullet = \sum_a c_{ba}^\chi(\alpha) A_a \bullet B_a, \tag{3.1}$$

where A_a and B_a belong to B and depend on α_S, and $c_{ba}^\chi(\alpha)$ is a complex number which depends on α_S, α_{Sb} and α_b. If and only if $A_a, B_a \neq 1$, $c_{ba}^\chi(\alpha)$ depends on χ. In this chapter, we assume only Markov property (i.e., $\hat{K}^\chi$ just depends on α_t). At $\chi = 0$, the FCS-QME becomes the quantum master equation (QME)

$$\frac{d\rho(t)}{dt} = \hat{K}(\alpha_t)\rho(t). \tag{3.2}$$

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

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

$$\hat{K}^\chi(\alpha)|\rho_n^\chi(\alpha)\rangle\rangle = \lambda_n^\chi(\alpha)|\rho_n^\chi(\alpha)\rangle\rangle, \tag{3.3}$$

$$\langle\langle l_n^\chi(\alpha)|\hat{K}^\chi(\alpha) = \lambda_n^\chi(\alpha)\langle\langle l_n^\chi(\alpha)|. \tag{3.4}$$

In the Liouville space, $A \in$ B is described by $|A\rangle\rangle$. The inner produce is defined by $\langle\langle A|B\rangle\rangle = \mathrm{Tr}_S(A^\dagger B)$ ($A, B \in$ B). In particular, $\langle\langle 1|A\rangle\rangle = \mathrm{Tr}_S A$ holds. A superoperator which operates to a liner operator of the system becomes an operator of the Liouville space. The left eigenvectors $l_n^\chi(\alpha)$ and the right eigenvectors $\rho_m^\chi(\alpha)$ satisfy

$$\langle\langle l_n^\chi(\alpha)|\rho_m^\chi(\alpha)\rangle\rangle = \delta_{nm}. \tag{3.5}$$

The mode which has the eigenvalue with the maximum real part is assigned by the label $n = 0$. Because the conservation of the probability $\frac{d}{dt}\langle\langle 1|\rho(t)\rangle\rangle = \langle\langle 1|\hat{K}(\alpha_t)|\rho(t)\rangle\rangle = 0$ leads

$$\langle\langle 1|\hat{K}(\alpha) = 0, \tag{3.6}$$

in the limit $\chi \to 0$, $\lambda_0^\chi(\alpha)$ becomes 0 and $\langle\langle l_0^\chi(\alpha)|$ becomes $\langle\langle 1|$ (i.e., $l_0(\alpha)$ is identity operator). In addition, $|\rho_0(\alpha)\rangle\rangle$ determined by

$$\hat{K}(\alpha)|\rho_0(\alpha)\rangle\rangle = 0, \tag{3.7}$$

represents the *instantaneous steady state*.

The formal solution of the FCS-QME (2.64) is

$$|\rho^{\chi}(t)\rangle\rangle = \mathrm{T}\exp\Big[\int_0^t ds\ \hat{K}^{\chi}(\alpha_s)\Big]|\rho(0)\rangle\rangle, \tag{3.8}$$

where T denotes the time-ordering operation. Using this, we obtain the averages

$$\begin{aligned}\langle \Delta o_\mu\rangle_t &= \frac{\partial}{\partial(i\chi_{O_\mu})}\langle\langle 1|\rho^{\chi}(t)\rangle\rangle\Big|_{\chi=0} \\ &= \int_0^t du\ \langle\langle 1|\hat{K}^{O_\mu}(\alpha_u)|\rho(u)\rangle\rangle \equiv \int_0^t du\ i_{O_\mu}(u),\end{aligned} \tag{3.9}$$

where $X^{O_\mu}(\alpha) \stackrel{\text{def}}{=} \frac{\partial X^{\chi}(\alpha)}{\partial(i\chi_{O_\mu})}\big|_{\chi=0}$ when X is an (super)operator or c-number. Here, we used $\langle\langle 1|\hat{K}(\alpha) = 0$. Moreover, using $\langle\langle l_0(\alpha)| = \langle\langle 1|$, $\lambda_0(\alpha) = 0$ and (3.4), we obtain

$$\langle\langle 1|\hat{K}^{O_\mu}(\alpha) = \lambda_0^{O_\mu}(\alpha)\langle\langle 1| - \langle\langle l_0^{O_\mu}(\alpha)|\hat{K}(\alpha). \tag{3.10}$$

Here, $\langle\langle l_0^{O_\mu}(\alpha)|$ is defined by $\frac{\partial\langle\langle l_0^{\chi}(\alpha)|}{\partial(i\chi_{O_\mu})}\big|_{\chi=0}$, then

$$l_0^{O_\mu} = -\frac{\partial l_0^{\chi}(\alpha)}{\partial(i\chi_{O_\mu})}\Big|_{\chi=0}, \tag{3.11}$$

holds. The current $i_{O_\mu}(t)$ is given by [24]

$$\begin{aligned} i_{O_\mu}(t) &= \langle\langle 1|\hat{K}^{O_\mu}(\alpha_t)|\rho(t)\rangle\rangle \\ &= \lambda_0^{O_\mu}(\alpha_t) - \langle\langle l_0^{O_\mu}(\alpha_t)|\hat{K}(\alpha_t)|\rho(t)\rangle\rangle \\ &= \lambda_0^{O_\mu}(\alpha_t) - \langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\rho(t)\rangle\rangle. \end{aligned} \tag{3.12}$$

The current can also be written as

$$i_{O_\mu}(t) = \langle\langle 1|W^{O_\mu}(\alpha_t)|\rho(t)\rangle\rangle, \tag{3.13}$$

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

$$\langle\langle 1|W^{O_\mu}(\alpha) = \langle\langle 1|\hat{K}^{O_\mu}(\alpha), \tag{3.14}$$

i.e., $\mathrm{Tr}_S[W^{O_\mu}(\alpha)\bullet] = \mathrm{Tr}_S[\hat{K}^{O_\mu}(\alpha)\bullet]$ for any $\bullet \in \mathrm{B}$. Therefore, using (3.1), the current operator is given by

$$W^{O_\mu}(\alpha) = \sum_{b,a} c_{ba}^{O_\mu}(\alpha)B_a A_a. \tag{3.15}$$

Using (3.10), the *instantaneous steady current* is given by

$$\langle\langle 1|W^{O_\mu}(\alpha)|\rho_0(\alpha)\rangle\rangle = \lambda_0^{O_\mu}(\alpha) \equiv i_{O_\mu}^{\mathrm{ss}}(\alpha). \tag{3.16}$$

In the following, we suppose $\rho(0) = \rho_0(\alpha_0)$. In this case, as we will show, $\rho(t) = \rho_0(\alpha_t) + \mathcal{O}(\omega/\Gamma)$ holds where $\omega = 2\pi/\tau$ and

$$\Gamma = \min_{n\neq 0,\alpha\in C}\{-\mathrm{Re}[\lambda_n(\alpha)]\}. \tag{3.17}$$

In $\omega \ll \Gamma$ limit, we obtain

$$i_{O_\mu}(t) = i^{\rm ss}_{O_\mu}(\alpha_t) - \langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle + \mathcal{O}(\frac{\omega^2}{\Gamma}), \tag{3.18}$$

which leads to

$$\langle \Delta o_\mu \rangle_\tau = \int_0^\tau dt\ i^{\rm ss}_{O_\mu}(\alpha_t) + \int_C d\alpha^n\ A_n^{O_\mu}(\alpha) + \mathcal{O}(\frac{\omega}{\Gamma}). \tag{3.19}$$

Here, α^n is the n-th component of the control parameters, C is the trajectory from α_0 to α_τ,

$$A_n^{O_\mu}(\alpha) \stackrel{\rm def}{=} -\langle\langle l_0^{O_\mu}(\alpha)|\frac{\partial}{\partial \alpha^n}|\rho_0(\alpha)\rangle\rangle, \tag{3.20}$$

is the BSN vector, and the summation symbol $\sum_n$ is omitted. As we will show, the BSN vector is also given by [25]

$$A_n^{O_\mu}(\alpha) = \langle\langle 1|W^{O_\mu}(\alpha)\mathcal{R}(\alpha)\frac{\partial}{\partial \alpha^n}|\rho_0(\alpha)\rangle\rangle, \tag{3.21}$$

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

$$\mathcal{R}(\alpha)\hat{K}(\alpha) = 1 - |\rho_0(\alpha)\rangle\rangle\langle\langle 1|. \tag{3.22}$$

In the research of adiabatic pumping, the expression of (3.19) is essential. In Refs.[23, 24, 25], (3.19) with (3.20) was used to study the quantum pump. On the other hand, in Ref.[34], (3.19) was derived using the generalized master equation [33] and without using the FCS. In Ref.[34], $A_n^{O_\mu}(\alpha)$ was described by the quantity corresponding to the current operator and the pseudo-inverse of the Liouvillian, as shown in (3.21). In this chapter, we show the equivalence between the FCS-QME approach and the generalized master equation approach (with the Born-approximation) for all orders of the pumping frequency [25] (see also Ref.[35]).

3.2 Berry-Sinitsyn-Nemenman phase

The expression of (3.19) was originally derived like the following. The formal solution of the FCS-QME is expanded as

$$|\rho^\chi(t)\rangle\rangle = \sum_n c_n^\chi(t) e^{\int_0^t ds\ \lambda_n^\chi(\alpha_s)}|\rho_n^\chi(\alpha_t)\rangle\rangle. \tag{3.23}$$

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

$$c_0^\chi(\tau) = c_0^\chi(0)\exp\Big[-\int_0^\tau dt\ \langle\langle l_0^\chi(\alpha_t)|\frac{d}{dt}|\rho_0^\chi(\alpha_t)\rangle\rangle\Big], \tag{3.24}$$

using (C.8) and the fact that the second term of RHS of (C.8) for $m = 0$ exponentially damps as a function of time. Here, the argument of the exponential function is called the BSN phase. Substituting this expression and $c_0^\chi(0) = \langle\langle l_0^\chi(\alpha_0)|\rho_0(\alpha_0)\rangle\rangle$ into (3.23), we obtain the expression of $\rho^\chi(\tau)$ which provides (3.19). However, when we

consider only the average of Δo_μ, the BSN phase is not essential. All informations of the counting fields up to the first order are included in W^{O_μ}.

Substituting (3.24) and $c_0^\chi(0) = \langle\langle l_0^\chi(\alpha_0)|\rho(0)\rangle\rangle$ into (3.23), we obtain

$$|\rho^\chi(\tau)\rangle\rangle \approx \langle\langle l_0^\chi(\alpha_0)|\rho(0)\rangle\rangle e^{-\int_0^\tau dt\ \langle\langle l_0^\chi(\alpha_t)|\frac{d}{dt}|\rho_0^\chi(\alpha_t)\rangle\rangle} e^{\int_0^\tau dt\ \lambda_0^\chi(\alpha_t)}|\rho_0^\chi(\alpha_\tau)\rangle\rangle, \quad (3.25)$$

and the cumulant generating function $S_\tau(\chi) = \ln Z_\tau(\chi) = \ln\langle\langle 1|\rho^\chi(\tau)\rangle\rangle$:

$$\begin{aligned} S_\tau(\chi) &= \int_0^\tau dt\ \lambda_0^\chi(\alpha_t) - \int_C d\alpha^n\ \langle\langle l_0^\chi(\alpha)|\frac{\partial|\rho_0^\chi(\alpha)\rangle\rangle}{\partial\alpha^n} \\ &\quad + \ln\langle\langle l_0^\chi(\alpha_0)|\rho(0)\rangle\rangle + \ln\langle\langle 1|\rho_0^\chi(\alpha_\tau)\rangle\rangle. \end{aligned} \quad (3.26)$$

(3.26) is the same with Yuge *et al.*[23] except for that χ denotes a multi-counting field. The averages $\langle\Delta o_\mu\rangle_\tau = \frac{\partial S_\tau(\chi)}{\partial(i\chi_\mu)}\big|_{\chi=0}$ are

$$\langle\Delta o_\mu\rangle_\tau = \int_0^\tau dt\ \lambda_0^{O_\mu}(\alpha_t) + \int_C d\alpha^n\ A_n^{O_\mu}(\alpha) + \langle\langle l_0^{O_\mu}(\alpha_0)|\rho(0)\rangle\rangle + \langle\langle 1|\rho_0^\mu(\alpha_0)\rangle\rangle \quad (3.27)$$

Here, we used $-\int_C d\alpha^n\ \langle\langle l_0(\alpha)|\frac{\partial|\rho_0^\mu(\alpha)\rangle\rangle}{\partial\alpha^n} = -\langle\langle 1|\rho_0^\mu(\alpha_\tau)\rangle\rangle + \langle\langle 1|\rho_0^\mu(\alpha_0)\rangle\rangle$ because $\langle\langle l_0(\alpha)|\frac{\partial|\rho_0^\mu(\alpha)\rangle\rangle}{\partial\alpha^n} = \frac{\partial}{\partial\alpha^n}\langle\langle 1|\rho_0^\mu(\alpha)\rangle\rangle$. The integrand of the first time integral, $\lambda_0^{O_\mu}(\alpha_t)$, are the instantaneous steady currents of O_μ at time t; if the control parameters are fixed to α and the state is $\rho_0(\alpha)$, the current of O_μ is $\lambda_0^{O_\mu}(\alpha)$. The third and fourth terms of the right side of (3.27) cancel if the initial condition is the instantaneous steady state $\rho_0(\alpha_0)$.

3.3 Cyclic pump

For $\alpha_\tau = \alpha_0$, the second term of the right side of (3.27) can be described as a surface integral over the surface S enclosed by C using the Stokes theorem :

$$\langle\Delta o_\mu\rangle_\tau = \langle\Delta o_\mu\rangle_\tau^{\rm ss} + \langle\Delta o_\mu\rangle_S^{\rm Berry}, \quad (3.28)$$

$$\langle\Delta o_\mu\rangle_\tau^{\rm ss} = \int_0^\tau dt\ \lambda_0^{O_\mu}(\alpha_t), \quad (3.29)$$

$$\langle\Delta o_\mu\rangle_S^{\rm Berry} = \int_S d\alpha^m \wedge d\alpha^n\ \frac{1}{2}F_{mn}^{O_\mu}(\alpha). \quad (3.30)$$

Here, $\wedge$ is the wedge product and the summation symbol $\sum_{n,m}$ is omitted. BSN curvature $F_{mn}^{O_\mu}(\alpha)$ is given by

$$F_{mn}^{O_\mu}(\alpha) = \frac{\partial A_n^{O_\mu}(\alpha)}{\partial\alpha^m} - \frac{\partial A_m^{O_\mu}(\alpha)}{\partial\alpha^n}. \quad (3.31)$$

Yuge *et al.*[23] focus on only the second term of (3.28) subtracting the first term, and they did not evaluate $\langle\Delta o_\mu\rangle_\tau^{\rm ss}$. In § 4.2.2, we show that this contribution is usually dominant if the thermodynamic parameters are modulated although the steady currents $\lambda_0^{O_\mu}(\alpha_t)$ are zero if the thermodynamic parameters are fixed to zero bias.

3.4 Expansion by frequency

Applying the pseudo-inverse $\mathcal{R}(\alpha)$ to the QME (3.2), we obtain

$$\Big[1-\mathcal{R}(\alpha_t)\frac{d}{dt}\Big]|\delta\rho(t)\rangle\rangle \;=\; \mathcal{R}(\alpha_t)\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle, \tag{3.32}$$

with $\delta\rho(t) \stackrel{\text{def}}{=} \rho(t)-\rho_0(\alpha_t)$. One of the solution of (3.32) is

$$|\delta\rho_{(\text{ss})}(t)\rangle\rangle \;=\; \sum_{n=1}^{\infty}\Big[\mathcal{R}(\alpha_t)\frac{d}{dt}\Big]^n|\rho_0(\alpha_t)\rangle\rangle \equiv \sum_{n=1}^{\infty}|\rho^{(n)}(t)\rangle\rangle. \tag{3.33}$$

$\langle\langle 1|\rho^{(n)}(t)\rangle\rangle = 0$ holds (we show this at § 3.5). The general solution of (3.32) is

$$|\delta\rho(t)\rangle\rangle \;=\; |\delta\rho_{(\text{ss})}(t)\rangle\rangle + |\tilde{\rho}(t)\rangle\rangle, \tag{3.34}$$

where $\tilde{\rho}(t)$ is the solution of

$$\Big[1-\mathcal{R}(\alpha_t)\frac{d}{dt}\Big]|\tilde{\rho}(t)\rangle\rangle = 0, \tag{3.35}$$

with $\tilde{\rho}(0) = \delta\rho(0)-\delta\rho_{(\text{ss})}(0)$. By the way, applying $\hat{K}(\alpha)$ to (3.22) from the left, we obtain

$$\hat{K}(\alpha)\mathcal{R}(\alpha)\hat{K}(\alpha) = \hat{K}(\alpha). \tag{3.36}$$

This leads

$$\hat{K}(\alpha)\mathcal{R}(\alpha) = 1-|\sigma(\alpha)\rangle\rangle\langle\langle 1|,\;\; \langle\langle 1|\sigma(\alpha)\rangle\rangle = 1. \tag{3.37}$$

Applying $\hat{K}(\alpha)$ to (3.32) from the left and using the above relation and $\langle\langle 1|\tilde{\rho}(t)\rangle\rangle = 0$, we obtain

$$\frac{d}{dt}|\tilde{\rho}(t)\rangle\rangle \;=\; \hat{K}(\alpha)|\tilde{\rho}(t)\rangle\rangle, \tag{3.38}$$

which is the same form with the original QME. The solution is $|\tilde{\rho}(t)\rangle\rangle = \hat{U}(t)|\tilde{\rho}(0)\rangle\rangle$ with

$$\hat{U}(t) \stackrel{\text{def}}{=} \mathrm{T}\exp\Big[\int_0^t ds\;\hat{K}(\alpha_s)\Big]. \tag{3.39}$$

Because $\langle\langle 1|\tilde{\rho}(0)\rangle\rangle = 0$, $|\tilde{\rho}(t)\rangle\rangle$ is described as $|\tilde{\rho}(t)\rangle\rangle = \sum_{n\neq 0} c'_n(t)e^{\int_0^t ds\;\lambda_n(\alpha_s)}|\rho_n(\alpha_s)\rangle\rangle$. This damps exponentially as a function of time. Then, the state reaches to a "steady state"

$$\rho_{(\text{ss})} \stackrel{\text{def}}{=} \rho_0(\alpha_t)+\delta\rho_{(\text{ss})}(t). \tag{3.40}$$

$\tilde{\rho}(0) = \delta\rho(0)-\delta\rho_{(\text{ss})}(0)$ is the difference of the initial state from the "steady state". We introduce

$$|\tilde{\rho}^{(n)}(t)\rangle\rangle \stackrel{\text{def}}{=} -\hat{U}(t)|\rho^{(n)}(0)\rangle\rangle, \tag{3.41}$$

$$|\tilde{\rho}^{(0)}(t)\rangle\rangle \stackrel{\text{def}}{=} \hat{U}(t)|\delta\rho(0)\rangle\rangle. \tag{3.42}$$

The general solution of the QME is given by

$$|\delta\rho(t)\rangle\rangle = \sum_{n=1}^{\infty}\left[|\rho^{(n)}(t)\rangle\rangle + |\tilde{\rho}^{(n)}(t)\rangle\rangle\right] + |\tilde{\rho}^{(0)}(t)\rangle\rangle. \tag{3.43}$$

$\langle\langle 1|\tilde{\rho}^{(n)}(t)\rangle\rangle = -\langle\langle 1|\rho^{(n)}(0)\rangle\rangle = 0$ and $\langle\langle 1|\tilde{\rho}^{(0)}(t)\rangle\rangle = \langle\langle 1|\delta\rho(0)\rangle\rangle = 0$ holds. The current $i_{O_\mu}(t)$ is given by

$$i_{O_\mu}(t) = i^{\rm ss}_{O_\mu}(\alpha_t) + \delta i^{(\rm ss)}_{O_\mu}(t) + \tilde{i}_{O_\mu}(t), \tag{3.44}$$

$$\delta i^{(\rm ss)}_{O_\mu}(t) \stackrel{\rm def}{=} \langle\langle 1|W^{O_\mu}(\alpha_t)|\delta\rho_{(\rm ss)}(t)\rangle\rangle, \tag{3.45}$$

$$\tilde{i}_{O_\mu}(t) \stackrel{\rm def}{=} \langle\langle 1|W^{O_\mu}(\alpha_t)|\tilde{\rho}(t)\rangle\rangle. \tag{3.46}$$

$$\delta i^{(\rm ss)}_{O_\mu}(t) = \sum_{n=1}^{\infty} i^{(n)}_{O_\mu}(t),\ i^{(n)}_{O_\mu}(t) \stackrel{\rm def}{=} \langle\langle 1|W^{O_\mu}(\alpha_t)|\rho^{(n)}(t)\rangle\rangle, \tag{3.47}$$

$$\tilde{i}_{O_\mu}(t) = \sum_{n=0}^{\infty} \tilde{i}^{(n)}_{O_\mu}(t),\ \tilde{i}^{(n)}_{O_\mu}(t) \stackrel{\rm def}{=} \langle\langle 1|W^{O_\mu}(\alpha_t)|\tilde{\rho}^{(n)}(t)\rangle\rangle. \tag{3.48}$$

Let's consider the relation between (3.12) and (3.44). In § 3.2, we used χ-adiabatic approximation (3.25), which becomes $|\rho(t)\rangle\rangle \approx |\rho_0(\alpha_t)\rangle\rangle$ at $\chi = 0$. Substituting it to (3.13), we obtain $i_{O_\mu}(t) \approx i^{\rm ss}_{O_\mu}(t)$. So, we cannot obtain $\delta i^{(\rm ss)}_{O_\mu}(t) + \tilde{i}_{O_\mu}(t)$. However, from the χ_{O_μ} derivative of (3.25), we obtain

$$i_{O_\mu}(t) \approx \lambda_0^{O_\mu}(\alpha_t) - \langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle. \tag{3.49}$$

This is equivalent to (3.27) for $\rho(0) = \rho_0(\alpha_0)$. (3.49) suggests

$$i^{(1)}_{O_\mu}(t) = -\langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle. \tag{3.50}$$

In fact, this is equivalent to $i^{(1)}_{O_\mu}(t) = \langle\langle 1|W^{O_\mu}(\alpha_t)|\rho^{(1)}(t)\rangle\rangle$, namely

$$i^{(1)}_{O_\mu}(t) = \langle\langle 1|W^{O_\mu}(\alpha_t)\mathcal{R}(\alpha_t)\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle, \tag{3.51}$$

because of

$$\langle\langle 1|W^{O_\mu}(\alpha)\mathcal{R}(\alpha) = -\langle\langle l_0^{O_\mu}(\alpha)| + c^{O_\mu}(\alpha)\langle\langle 1|. \tag{3.52}$$

Here, $c^{O_\mu}(\alpha)$ are constants shown in (E.10). We prove (3.52) in Appendix E. (3.52) leads (3.21) and

$$\begin{aligned} i^{(n+1)}_{O_\mu}(t) &= \langle\langle 1|W^{O_\mu}(\alpha_t)\mathcal{R}(\alpha_t)\frac{d}{dt}|\rho^{(n)}(t)\rangle\rangle \\ &= -\langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\rho^{(n)}(t)\rangle\rangle. \end{aligned} \tag{3.53}$$

By the way, (3.12) is

$$i_{O_\mu}(t) = i^{\rm ss}_{O_\mu}(\alpha_t) - \langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\rho(t)\rangle\rangle. \tag{3.54}$$

Substituting

$$\rho(t) = \rho_0(\alpha_t) + \sum_{n=1} \rho^{(n)}(t) + \sum_{n=0} \tilde{\rho}^{(n)}(t) = \rho_{(\mathrm{ss})}(t) + \sum_{n=0} \tilde{\rho}^{(n)}(t), \tag{3.55}$$

to the RHS of (3.54), ρ_0 provides $i^{(1)}_{O_\mu}$ and $\rho^{(n)}$ provides $i^{(n+1)}_{O_\mu}$. $\rho_{(\mathrm{ss})}$ provides $\delta i^{(\mathrm{ss})}_{O_\mu}$. $\tilde{\rho}^{(n)}$ provides $\tilde{i}^{(n)}_{O_\mu}$:

$$\begin{aligned}
&-\langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\tilde{\rho}^{(n)}(t)\rangle\rangle \\
&= -\langle\langle l_0^{O_\mu}(\alpha_t)|\hat{K}(\alpha_t)|\tilde{\rho}^{(n)}(t)\rangle\rangle \\
&= \langle\langle 1|W^{O_\mu}(\alpha_t)\mathcal{R}(\alpha_t)\hat{K}(\alpha_t)|\tilde{\rho}^{(n)}(t)\rangle\rangle - c^{O_\mu}(\alpha_t)\langle\langle 1|\hat{K}(\alpha_t)|\tilde{\rho}^{(n)}(t)\rangle\rangle \\
&= \langle\langle 1|W^{O_\mu}(\alpha_t)(1-|\rho_0(\alpha_t)\rangle\rangle\langle\langle 1|)|\tilde{\rho}^{(n)}(t)\rangle\rangle \\
&= \tilde{i}^{(n)}_{O_\mu}.
\end{aligned} \tag{3.56}$$

The third and fourth terms of (3.27), $\langle \Delta o_\mu\rangle^{3+4}_\tau = \langle\langle l_0^{O_\mu}(\alpha_0)|\rho(0)\rangle\rangle + \langle\langle 1|\rho_0^{O_\mu}(\alpha_0)\rangle\rangle$, result from this relaxation. The contribution of $\langle \Delta o_\mu\rangle_\tau$ from $\delta\rho(0)$ is

$$\begin{aligned}
\langle \Delta o_\mu\rangle^{\mathrm{ini}}_\tau &\overset{\mathrm{def}}{=} -\int_0^\tau dt\, \langle\langle l_0^{O_\mu}(\alpha_t)|\frac{d}{dt}|\tilde{\rho}^{(0)}(t)\rangle\rangle \\
&= \langle\langle l_0^{O_\mu}(\alpha_0)|\tilde{\rho}^{(0)}(0)\rangle\rangle - \langle\langle l_0^{O_\mu}(\alpha_\tau)|\tilde{\rho}^{(0)}(\tau)\rangle\rangle \\
&\quad + \int_0^\tau dt\, \frac{d\langle\langle l_0^{O_\mu}(\alpha_t)|}{dt}|\tilde{\rho}^{(0)}(t)\rangle\rangle.
\end{aligned} \tag{3.57}$$

The first term of the right side of (3.57) is $\langle \Delta o_\mu\rangle^{3+4}_\tau$. Because we can obtain $\langle\langle l_0^{O_\mu}(\alpha)|\rho_0(\alpha)\rangle\rangle + \langle\langle 1|\rho_0^{O_\mu}(\alpha)\rangle\rangle = 0$ from the normalization $\langle\langle l_0^{\chi}(\alpha)|\rho_0^{\chi}(\alpha)\rangle\rangle = 1$, $\langle \Delta o_\mu\rangle^{3+4}_\tau$ is given by

$$\langle \Delta o_\mu\rangle^{3+4}_\tau = \langle\langle l_0^{O_\mu}(\alpha_0)\big[|\rho(0)\rangle\rangle - |\rho_0(\alpha_0)\rangle\rangle\big] = \langle\langle l_0^{O_\mu}(\alpha_0)|\tilde{\rho}^{(0)}(0)\rangle\rangle. \tag{3.58}$$

The second term of the right side of (3.57) is exponentially small since $\tilde{\rho}^{(0)}(\tau) \sim e^{-\Gamma\tau}$. The order of the third term is $\mathcal{O}(\frac{\omega}{\Gamma})$ with $\omega = 2\pi/\tau$ because $\frac{d\langle\langle l_0^{O_\mu}(\alpha_t)|}{dt} = \mathcal{O}(\omega)$ and the integral range is restricted up to $1/\Gamma$ since $\tilde{\rho}^{(0)}(t) \sim e^{-\Gamma t}$. Hence

$$\langle \Delta o_\mu\rangle^{\mathrm{ini}}_\tau = \langle \Delta o_\mu\rangle^{3+4}_\tau + \mathcal{O}(\frac{\omega}{\Gamma}). \tag{3.59}$$

Since $\frac{d}{dt}\alpha_t = \mathcal{O}(\omega)$ and $\mathcal{R}(\alpha_t) = \mathcal{O}(\frac{1}{\Gamma})$,

$$\rho^{(n)}(t) = \mathcal{O}(\frac{\omega}{\Gamma})^n. \tag{3.60}$$

In Appendix D, we discuss the reasonable range of n of $\rho^{(n)}(t)$ and show that with the larger non-adiabaticity ($\frac{\omega}{\Gamma}$), the reasonable range becomes wider. We have

$$\tilde{\rho}^{(n)} = \mathcal{O}(\frac{\omega^n}{\Gamma^n}e^{-\Gamma t}),\ \tilde{\rho}^{(0)} = \mathcal{O}(e^{-\Gamma t}). \tag{3.61}$$

The above equations and $W^{O_\mu} = \mathcal{O}(\Gamma)$ lead

$$i^{(n)}_{O_\mu} = \mathcal{O}(\frac{\omega^n}{\Gamma^{n-1}}),\ \tilde{i}^{(n)}_{O_\mu} = \mathcal{O}(\frac{\omega^n}{\Gamma^{n-1}}e^{-\Gamma t}),\ \tilde{i}^{(0)}_{O_\mu} = \mathcal{O}(\Gamma e^{-\Gamma t}). \tag{3.62}$$

This leads

$$\langle \Delta o_\mu \rangle_\tau^{(n)} \stackrel{\text{def}}{=} \int_0^\tau dt\, i_{O_\mu}^{(n)}(t) = \mathcal{O}(\frac{\omega^{n-1}}{\Gamma^{n-1}}), \tag{3.63}$$

$$\widetilde{\langle \Delta o_\mu \rangle}_\tau^{(n)} \stackrel{\text{def}}{=} \int_0^\tau dt\, \tilde{i}_{O_\mu}^{(n)}(t) = \mathcal{O}(\frac{\omega^{n}}{\Gamma^{n}}), \tag{3.64}$$

$$\langle \Delta o_\mu \rangle_\tau^{\text{ini}} = \int_0^\tau dt\, \tilde{i}_{O_\mu}^{(0)}(t) = \mathcal{O}(1). \tag{3.65}$$

In particular, the contribution from the BSN vector is

$$\langle \Delta o_\mu \rangle_\tau^{\text{BSN}} \stackrel{\text{def}}{=} \langle \Delta o_\mu \rangle_\tau^{(1)} = -\int_C d\alpha^n\, \langle\langle l_0^{O_\mu}(\alpha)| \frac{\partial |\rho_0(\alpha)\rangle\rangle}{\partial \alpha^n} = \mathcal{O}(1). \tag{3.66}$$

Moreover, although the BSN phase is derived under the χ-adiabatic condition which makes (3.24) and $c_n^\chi(\tau)e^{\Lambda_n^\chi(\tau)} \approx 0$ $(n \neq 0)$ appropriate, its origin is probably a non-adiabatic effect that comes from $\frac{\omega}{\Gamma}$, because (3.50) shows that the BSN phase has the information of the non-adiabatic part of the QME $(\delta\rho(t) = \rho(t) - \rho_0(\alpha_t))$.

For the RWA, at equilibrium (zero-bias) case, $\rho_0(\alpha)$ is the grand canonical distribution

$$\rho_{\text{gc}}(\alpha_S; \beta, \beta\mu) \stackrel{\text{def}}{=} \frac{e^{-\beta(H_S(\alpha_S)-\mu N_S)}}{\Xi(\alpha_S; \beta, \beta\mu)}. \tag{3.67}$$

Cf.(A.11). Here, $\Xi(\alpha_S; \beta, \beta\mu) \stackrel{\text{def}}{=} \text{Tr}_S[e^{-\beta(H_S(\alpha_S)-\mu N_S)}]$, β is the inverse temperature of all baths and μ is the chemical potential for $b \in \boldsymbol{\mathcal{G}}$. (3.67) is derived from (2.152) and (2.165). At zero-bias, for pumping by only α' (α'' are fixed), (3.20), (3.33) and (3.41) lead that the pumping dose not occur in all orders of ω when α_S are fixed.

3.5 Arbitrariness of pseudo-inverse

General solution of $\mathcal{R}\hat{K}(\alpha_t) = 1 - |\rho_0(\alpha_t)\rangle\rangle\langle\langle 1|$ is given by

$$\mathcal{R}(t) = |\rho_i(t)\rangle\rangle\langle\langle 1| + \mathcal{R}_0(\alpha_t), \tag{3.68}$$

where $\mathcal{R}_0(\alpha)$ is one of the solution of

$$\mathcal{R}_0(\alpha)\hat{K}(\alpha) = 1 - |\rho_0(\alpha)\rangle\rangle\langle\langle 1|. \tag{3.69}$$

$\rho_i(t)$ can depend on the initial values of the QME. In the following of this section, we show that $|\rho^{(n)}(t)\rangle\rangle = \left[\mathcal{R}(t)\frac{d}{dt}\right]^n |\rho_0(\alpha_t)\rangle\rangle$ is independent of $\rho_i(t)$. Then, $\rho^{(n)}$ and $\tilde{\rho}^{(n)}$ are independent of the choice of the pseudo-inverse.

$\rho^{(1)}(t)$ is given by

$$\begin{aligned} |\rho^{(1)}(t)\rangle\rangle &= |\rho_i(t)\rangle\rangle\langle\langle 1|\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle + \mathcal{R}_0(\alpha_t)\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle \\ &= \mathcal{R}_0(\alpha_t)\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle. \end{aligned} \tag{3.70}$$

Then, $\rho^{(1)}(t)$ is independent of the choice of the pseudo-inverse. Next, $\rho^{(2)}(t)$ is given by

$$|\rho^{(2)}(t)\rangle\rangle = |\rho_i(t)\rangle\rangle \frac{d}{dt}\left[\langle\langle 1|\mathcal{R}_0(\alpha_t)\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle\right] + \mathcal{R}_0(\alpha_t)\frac{d}{dt}\left[\mathcal{R}_0(\alpha_t)\frac{d}{dt}|\rho_0(\alpha_t)\rangle\rangle\right] \tag{3.71}$$

By the way, applying $\langle\langle 1|$ to (3.69), we obtain

$$\langle\langle 1|\mathcal{R}_0(\alpha)\hat{K}(\alpha) = \langle\langle 1| - \langle\langle 1|\rho_0(\alpha)\rangle\rangle\langle\langle 1| = 0. \tag{3.72}$$

This leads

$$\langle\langle 1|\mathcal{R}_0(\alpha) = \mathcal{C}(\alpha)\langle\langle 1|. \tag{3.73}$$

Then, $|\rho^{(2)}(t)\rangle\rangle$ and $|\rho^{(n)}(t)\rangle\rangle$ do not depend on the choice of the pseudo-inverse. In fact,

$$|\rho^{(n+1)}(t)\rangle\rangle = |\rho_i(t)\rangle\rangle \frac{d}{dt}\langle\langle 1|\rho^{(n)}(t)\rangle\rangle + \mathcal{R}_0(\alpha_t)\frac{d}{dt}|\rho^{(n)}(t)\rangle\rangle, \tag{3.74}$$

leads

$$\langle\langle 1|\rho^{(n+1)}(t)\rangle\rangle = \langle\langle 1|\rho_i(t)\rangle\rangle \frac{d}{dt}\langle\langle 1|\rho^{(n)}(t)\rangle\rangle + \mathcal{C}(\alpha_t)\frac{d}{dt}\langle\langle 1|\rho^{(n)}(t)\rangle\rangle. \tag{3.75}$$

Then, $\langle\langle 1|\rho^{(n)}(t)\rangle\rangle = 0$ leads $\langle\langle 1|\rho^{(n+1)}(t)\rangle\rangle = 0$. Because of this and $\langle\langle 1|\rho^{(1)}(t)\rangle\rangle = 0$ derived from (3.70) and (3.73), we obtain

$$\langle\langle 1|\rho^{(n)}(t)\rangle\rangle = 0 \quad (n = 1, 2, 3, \cdots). \tag{3.76}$$

This and (3.74) lead

$$|\rho^{(n+1)}(t)\rangle\rangle = \mathcal{R}_0(\alpha_t)\frac{d}{dt}|\rho^{(n)}(t)\rangle\rangle. \tag{3.77}$$

3.6 Generalized mater equation approach

It is important to recognize the relations between the FCS-QME approach and the GME approach [66, 33, 67, 68, 69, 70, 71]. In the GME approach, $p_i(t) = \langle i|\rho(t)|i\rangle$ are governed by the generalized master equation (GME)

$$\frac{d}{dt}p_i(t) = \sum_j \int_{-\infty}^{t} dt'\, W_{ij}(t,t')p_j(t'), \tag{3.78}$$

where $|i\rangle$ are the energy eigenstates of the system Hamiltonian. The kernel $W_{ij}(t,t')$ can include the higher order contribution of the tunneling interaction between baths and the system. In the GME, $p_j(t')$ is given by $p_j(t) + \sum_{k=1}^{\infty}\frac{(t'-t)^k}{k!}\frac{d^k p_j(t)}{dt^k}$ [66, 33]. Moreover, $W_{ij}(t,t')$ and $p_j(t)$ are expanded as $W_{ij}(t,t') = \sum_{n=0}^{\infty}\sum_{m=1}^{\infty} W_{ij(m)}^{(n)}(t;t-t')$ and $p_j(t) = \sum_{n=0}^{\infty}\sum_{m=-n}^{\infty} p_{j(m)}^{(n)}(t)$, where $W_{ij(m)}^{(n)}(t;t-t')$ and $p_{j(m)}^{(n)}(t)$ are of the order of $\omega^n\Gamma^m$. In particular, $W_{ij(m)}^{(0)}(t;t-t') = W_{ij(m)}^{(0)}(\alpha_t;t-t')$ is the kernel where the control parameters are fixed to α_t. Up to the second order of the tunneling interaction (in the following we consider this level of approximation), we obtain (see

Appendix F) [33, 70]

$$0 = \sum_j K_{ij}^{(0)}(\alpha_t) p_j^{(0)}(\alpha_t), \tag{3.79}$$

$$\frac{dp_{i(-n)}^{(n)}(t)}{dt} = \sum_j K_{ij}^{(0)}(\alpha_t) p_{j(-n-1)}^{(n+1)}(t), \tag{3.80}$$

for $n = 0, 1, \cdots$, with

$$K_{ij}^{(0)}(\alpha_t) = \int_{-\infty}^{t} dt' \, W_{ij(1)}^{(0)}(\alpha_t, t - t'), \tag{3.81}$$

which is the instantaneous Liouvillian corresponding to our $\hat{K}(\alpha_t)$. (3.79) is just the definition of the instantaneous steady state $p_j^{(0)}(\alpha_t) \equiv p_{j(0)}^{(0)}(t)$, which satisfies $\sum_i p_i^{(0)}(\alpha_t) = 1$. Additionally, $p_{i(m)}^{(n)}(t)$ for $n \geq 1$ satisfies $\sum_i p_{i(m)}^{(n)}(t) = 0$. The conservation of the probability leads to $\sum_i K_{ij}^{(0)}(\alpha_t) = 0$, which corresponds to our $\langle\langle 1|\hat{K}(\alpha_t) = 0$. The charge or spin current $i_{O_\mu}(t)$ is given by [69, 70]

$$i_{O_\mu}(t) = \sum_{i,j} w_{ij}^{O_\mu}(\alpha_t) p_j(t), \tag{3.82}$$

corresponding to our (3.13). $w_{ij}^{O_\mu}(\alpha_t)$ is the instantaneous current matrix of O_μ in the present approximation, which corresponds to our $W^{O_\mu}(\alpha_t)$ and is linear in Γ (see (8.15)). $i_{O_\mu}(t)$ can be rewritten as

$$i_{O_\mu}(t) = \sum_j w_j^{O_\mu}(\alpha_t) p_j(t), \; w_j^{O_\mu}(\alpha) \stackrel{\text{def}}{=} \sum_i w_{ij}^{O_\mu}(\alpha). \tag{3.83}$$

$w_j^{O_\mu}(\alpha)$ corresponds to $\langle j|W^{O_\mu}(\alpha_t)|j\rangle$. Substituting $p_j(t) \approx \sum_{n=0}^{\infty} p_{j(-n)}^{(n)}(t)$ into (3.82), we obtain

$$i_{O_\mu}(t) = \sum_{n=0}^{\infty} i_{O_\mu}^{(n)}(t), \; i_{O_\mu}^{(n)}(t) \stackrel{\text{def}}{=} \sum_{i,j} w_{ij}^{O_\mu}(\alpha_t) p_{j(-n)}^{(n)}(t). \tag{3.84}$$

(3.80) for $n = 0$ leads to [69]

$$p_{j(-1)}^{(1)}(t) = \sum_i R_{ji}(\alpha_t) \frac{dp_i^{(0)}(\alpha_t)}{dt}. \tag{3.85}$$

Here, $R_{ji}(\alpha_t)$ is the pseudo-inverse of $K_{ij}^{(0)}(\alpha_t)$ corresponding to our $\mathcal{R}(\alpha_t)$ and it is given by [69]

$$R_{ji}(\alpha_t) = (\tilde{K}^{-1})_{ji}, \; \tilde{K}_{ji} = K_{ji}^{(0)} - K_{jj}^{(0)}. \tag{3.86}$$

Substituting (3.85) into (3.84), we obtain [69]

$$i^{(1)}_{O_\mu}(t) = \sum_i \varphi_i^{O_\mu}(\alpha_t)\frac{dp_i^{(0)}(\alpha_t)}{dt}, \tag{3.87}$$

$$\varphi_i^{O_\mu}(\alpha_t) = \sum_{k,j} w_{kj}^{O_\mu}(\alpha_t)R_{ji}(\alpha_t) = \sum_j w_j^{O_\mu}(\alpha_t)R_{ji}(\alpha_t). \tag{3.88}$$

A similar method has been used in Ref.[66]. $\varphi_i^{O_\mu}(\alpha_t)$ and (3.87) respectively correspond to our $\langle\langle 1|W^{O_\mu}(\alpha)\mathcal{R}(\alpha)$ and (3.51). Moreover, (3.80) for arbitrary n leads to

$$p^{(n+1)}_{j(-n-1)}(t) = \sum_i R_{ji}(\alpha_t)\frac{dp^{(n)}_{i(-n)}(t)}{dt}, \tag{3.89}$$

which corresponds to our (3.33). Because of these relations, the GME approach is equivalent to the FCS-QME approach in the calculation up to the second order of the tunneling interaction. Additionally, we discuss corrections due to the non-adiabatic effect of the FCS-QME in Appendix D. The first equation of (D.7) is consistent with $p^{(1)}_{j(0)}(t) = \mathcal{O}(\omega\tau_B)$ derived in Appendix F. Here, τ_B is the relaxation time of the baths.

In this chapter, we proved the equivalence between (3.12) and (3.44) using a key relation (3.52) and showed the origin of the BSN phase is a non-adiabatic effect, and connected the FCS-QME approach and the GME approach [69]. These are among the most important results of the first half of this thesis.

Chapter 4

Quantum adiabatic pump

4.1 Model

In this chapter, we consider quantum dots (QDs) (denoted by a symbol S) weakly coupled to several leads. The total Hamiltonian is $H_{\rm tot}(\alpha'(t)) = H_S(\alpha_S(t)) + \sum_b [H_b(\alpha'_b(t)) + H_{Sb}(\alpha_{Sb}(t))]$. Here, $H_S(\alpha_S(t))$ is the system (QDs) Hamiltonian, $H_b(\alpha'_b(t))$ is the Hamiltonian of the lead b, and $H_{Sb}(\alpha_{Sb}(t))$ is the tunneling interaction Hamiltonian between S and the lead b. To observe the spin effects, we suppose that the leads and the system are applied to collinear magnetic fields with different amplitudes, which relate to spins through the Zeeman effect. The leads are noninteracting:

$$H_b(\alpha'_b(t)) = \sum_{k,\sigma} (\varepsilon_{bk} + \sigma g_b B_b(t)) c^\dagger_{bk\sigma} c_{bk\sigma}. \tag{4.1}$$

Here, $\sigma = \uparrow, \downarrow = \pm 1$ is spin label,

$$g_b = \frac{1}{2} \mu_{\rm B} g^*_b, \tag{4.2}$$

where g^*_b is the g-factor of the lead b, $\mu_{\rm B}$ is the Bohr magneton and $B_b(t)$ is the strength of the magnetic field of the lead b. $c^\dagger_{bk\sigma}(c_{bk\sigma})$ is the creation (annihilation) operator of an electron with spin σ and momentum k in the lead b. The system Hamiltonian is

$$H_S(\alpha_S(t)) = \sum_{n,m,s,s'} \varepsilon_{ns,ms'}(B_S(t)) a^\dagger_{ns} a_{ms'} + H_{\rm Coulomb}, \tag{4.3}$$

where $a^\dagger_{ns}$ is the creation operator of an electron with orbital n and spin s. $\varepsilon_{ns,ms'}(B_S(t))$ means the energy of the electron for $n = m, s = s'$ and the tunneling amplitude between orbitals for $(n, s) \neq (m, s')$ which depends on the magnetic field of the system. $H_{\rm Coulomb}$ denotes Coulomb interaction. The tunneling interaction Hamiltonian is

$$H_{Sb}(\alpha_{Sb}(t)) = \sum_{k,\sigma,n,s} \sqrt{\Delta_b(t)} v_{bk\sigma,ns} a^\dagger_{ns} c_{bk\sigma} + \text{h.c.}, \tag{4.4}$$

where $\Delta_b(t)$ is a dimensionless parameter, and $v_{bk\sigma,ns}$ is the tunneling amplitude.

We assume B_S, $\{B_b\}_b$ and $\{\Delta_b\}_b$ are control parameters (denoted $\alpha' = (B_S, \{B_b\}_b, \{\Delta_b\}_b)$ and are called the dynamic parameters). The thermodynamic parameters (the chemical potentials and inverse temperatures of the leads, $\{\mu_b\}_b$ and $\{\beta_b\}_b$) are also considered as control parameters in § 4.2.2 and § 4.3.2. We denote $\alpha'' = \{\beta_b, \mu_b\}_b$ and $\alpha = \alpha' + \alpha''$. Yuge *et al.*[23] chose the set of control parameters as only α''. However we are interested in α' for the reason explained in § 4.2.2.

We choose the measured observables $\{O_\mu\} = \{N_{b\sigma}\}_{b,\sigma=\uparrow,\downarrow}$ with $N_{b\sigma} = \sum_k c^\dagger_{bk\sigma} c_{bk\sigma}$. The pumped charge (spin) of the lead b is given by $\langle \Delta N_{b\uparrow}\rangle \pm \langle \Delta N_{b\downarrow}\rangle$. $\langle \Delta N_{b\sigma}\rangle$ are calculated by (3.28). In fact, what we call the pumped charge, $\langle \Delta N_{b\uparrow}\rangle + \langle \Delta N_{b\downarrow}\rangle$, is the pumped electron number (actual pumped charge is given by $-e[\langle \Delta N_{b\uparrow}\rangle + \langle \Delta N_{b\downarrow}\rangle]$, where e (> 0) is the elementary charge).

In § 4.2.3 and § 4.3 we consider a one level system

$$H_S(\alpha_S(t)) = \sum_{s=\uparrow,\downarrow} \omega_s(B_S(t)) a^\dagger_s a_s + U a^\dagger_\uparrow a_\uparrow a^\dagger_\downarrow a_\downarrow, \tag{4.5}$$

as a special model of (4.3). Here, $s = \uparrow, \downarrow = \pm 1$,

$$\omega_s(B_S) = \omega_0 + s g_S B_S, \tag{4.6}$$

with ω_0 the electron energy at $B_S = 0$, and

$$g_S = \frac{1}{2}\mu_{\mathrm{B}} g^*_S, \tag{4.7}$$

where g^*_S is the g factor of the QD.

In the following of this chapter, we apply the FCS-QME with RWA.

4.2 Non-interacting system

In this section, we consider a noninteracting system ($H_{\mathrm{Coulomb}} = 0$). The system Hamiltonian (4.3) can be diagonalized

$$H_S = \sum_{i=1}^{2N} \tilde{\omega}_i b^\dagger_i b_i, \tag{4.8}$$

by a unitary transform $a_{ns} = \sum_{i=1}^{2N} U_{ns,i} b_i$. The tunneling interaction Hamiltonian (4.4) is

$$H_{Sb} = \sum_{k,\sigma,i} W_{bk\sigma,i} b^\dagger_i c_{bk\sigma} + \text{h.c.}, \tag{4.9}$$

with

$$W_{bk\sigma,i} = \sum_{n,s} \sqrt{\Delta_b} v_{bk\sigma,ns} U^*_{ns,i}. \tag{4.10}$$

In § 4.2.1, the Liouvillian and its instantaneous steady state are explained. In § 4.2.2, we consider the contribution of (3.29) and show that this cannot be neglected in general if the chemical potentials and the temperatures are not fixed. In § 4.2.3, we calculate the BSN curvatures for two combinations of modulated control parameters (B_L, B_S) and (Δ_L, B_S).

4.2.1 Liouvillian

The Liouvillian in the RWA is given by

$$\hat{K}^{\chi}(\alpha) = \sum_{i=1}^{2N} \hat{K}_i^{\chi}(\alpha), \tag{4.11}$$

$$\hat{K}_i^{\chi}(\alpha)\bullet = -i[\tilde{\omega}_i b_i^\dagger b_i, \bullet] + \hat{\Pi}_i^{\chi}(\alpha)\bullet - i[H_{\mathrm{L},i}, \bullet], \tag{4.12}$$

if $\{\tilde{\omega}_i\}$ are not degenerated. Here, super-operator $\hat{\Pi}_i^{\chi}(\alpha)$ operates to an arbitrary operator $\bullet$ as

$$\begin{aligned}\hat{\Pi}_i^{\chi}(\alpha)\bullet = \Big\{&\Phi_{b,i}^{+,\chi} b_i^\dagger \bullet b_i - \frac{1}{2}\Phi_i^+ \bullet b_i b_i^\dagger - \frac{1}{2}\Phi_i^+ b_i b_i^\dagger \bullet \\ &+\Phi_{b,i}^{-,\chi} b_i \bullet b_i^\dagger - \frac{1}{2}\Phi_i^- \bullet b_i^\dagger b_i - \frac{1}{2}\Phi_i^- b_i^\dagger b_i \bullet \Big\},\end{aligned} \tag{4.13}$$

with

$$\Phi_i^{\pm,\chi} = 2\pi \sum_{b,k,\sigma} |W_{bk\sigma,i}|^2 f_b^{\pm}(\tilde{\omega}_i) e^{\mp i\chi_{b\sigma}} \delta(\varepsilon_{bk} + \sigma g_b B_b - \tilde{\omega}_i). \tag{4.14}$$

Here, $f_b^+(\omega) = [e^{\beta_b(\omega-\mu_b)}+1]^{-1}$ is the Fermi distribution function, $f_b^-(\omega) = 1 - f_b^+(\omega)$, $\chi_{b\sigma}$ is the counting field for $N_{b\sigma}$. The Lamb shift Hamiltonian is given by

$$H_{\mathrm{L},i} = \Omega_i(\alpha') b_i^\dagger b_i, \tag{4.15}$$

with

$$\Omega_i(\alpha') = -\frac{1}{2}\Big(\Psi_i^- + \Psi_i^+\Big), \tag{4.16}$$

$$\Psi_i^{\pm} = 2\sum_{b,k,\sigma} |W_{bk\sigma,i}|^2 f_b^{\pm}(\tilde{\omega}_i) \mathrm{P}\frac{1}{\varepsilon_{bk} + \sigma g_b B_b - \tilde{\omega}_i}. \tag{4.17}$$

Here, P denotes the Cauchy principal value. $\Phi_i^{\pm,\chi}$ satisfies

$$\Phi_i^{\pm} = \Phi_i^{\pm,\chi}\big|_{\chi=0} = \sum_{b,\sigma} \Phi_{b\sigma,i}^{\pm}, \tag{4.18}$$

and

$$\frac{\Phi_i^{\pm,\chi}}{\partial(i\chi_{b\sigma})}\Big|_{\chi=0} = \mp\Phi_{b\sigma,i}^{\pm}, \tag{4.19}$$

with

$$\Phi_{b\sigma,i}^{\pm} = 2\pi \sum_{k} |W_{bk\sigma,i}|^2 f_b^{\pm}(\tilde{\omega}_i) \delta(\varepsilon_{bk} + \sigma g_b B_b - \tilde{\omega}_i). \tag{4.20}$$

We set

$$\Gamma_i = \sum_{b,\sigma} \Gamma_{b\sigma,i} = \sum_{b} \Gamma_{b,i}, \tag{4.21}$$

with

$$\Gamma_{b\sigma,i} = 2\pi \sum_k |W_{bk\sigma,i}|^2 \delta(\varepsilon_{bk} + \sigma g_b B_b - \tilde{\omega}_i). \tag{4.22}$$

Then,

$$\Gamma_i = \Phi_i^+ + \Phi_i^-, \tag{4.23}$$

and

$$\Phi_{b\sigma,i}^{\pm} = \Gamma_{b\sigma,i} f_b^{\pm}(\tilde{\omega}_i), \tag{4.24}$$

hold. The matrix representation of $\hat{K}_i^{\chi}(\alpha)$ (see Appendix B) by the number states of $b_i^\dagger b_i$ ($|0\rangle_i$ and $|1\rangle_i$) is a 4×4 matrix which is block diagonalized to $\{|0\rangle_{ii}\langle 0|, |1\rangle_{ii}\langle 1|\}$ space and $\{|0\rangle_{ii}\langle 1|, |1\rangle_{ii}\langle 0|\}$ space. The $\{|0\rangle_{ii}\langle 0|, |1\rangle_{ii}\langle 1|\}$ part is given by

$$K_i^{\chi}(\alpha) = \begin{pmatrix} -\Phi_i^+ & \Phi_i^{-,\chi} \\ \Phi_i^{+,\chi} & -\Phi_i^- \end{pmatrix} \begin{matrix} |00\rangle\rangle_i \\ |11\rangle\rangle_i \end{matrix}. \tag{4.25}$$

$\{|0\rangle_{ii}\langle 1|, |1\rangle_{ii}\langle 0|\}$ part does not relate to the instantaneous steady state of $\hat{K}_i^{\chi}(\alpha)$. The eigenvalue of the instantaneous steady state of $\hat{K}_i^{\chi}(\alpha)$ is given by

$$\lambda_{0,i}^{\chi}(\alpha) = -\frac{\Phi_i^+(\alpha) + \Phi_i^-(\alpha)}{2} + \sqrt{D_i^{\chi}(\alpha)}, \tag{4.26}$$

with

$$D_i^{\chi}(\alpha) = [\Phi_i^+ + \Phi_i^-]^2/4 - [\Phi_i^+\Phi_i^- - \Phi_i^{-,\chi}\Phi_i^{+,\chi}]. \tag{4.27}$$

The corresponding left and right eigenvectors are $|\rho_{0,i}^{\chi}(\alpha)\rangle\rangle = C_i^{\chi}(\alpha)|00\rangle\rangle_i + E_i^{\chi}(\alpha)|11\rangle\rangle_i$ and $\langle\langle l_{0,i}^{\chi}(\alpha)| = {}_i\langle\langle 00| + v_i^{\chi}(\alpha)_i\langle\langle 11|$ with $C_i^{\chi}(\alpha) = \frac{\Phi_i^{-,\chi}\Phi_i^{+,\chi}}{[\lambda_{0,i}^{\chi}+\Phi_i^+]^2+\Phi_i^{-,\chi}\Phi_i^{+,\chi}}$, $E_i^{\chi}(\alpha) = \frac{\Phi_i^{+,\chi}(\lambda_{0,i}^{\chi}+\Phi_i^+)}{[\lambda_{0,i}^{\chi}+\Phi_i^+]^2+\Phi_i^{-,\chi}\Phi_i^{+,\chi}}$, and

$$v_i^{\chi}(\alpha) = \frac{\Phi_i^+ - \Phi_i^- + 2\sqrt{D_i^{\chi}(\alpha)}}{2\Phi_i^{+,\chi}}. \tag{4.28}$$

At $\chi_{b\sigma} = 0$, $E_i^{\chi}(\alpha)$ becomes

$$E_i(\alpha) = \frac{\Phi_i^+}{\Phi_i^+ + \Phi_i^-} \tag{4.29}$$

and $C_i^{\chi}(\alpha)$ becomes $C_i(\alpha) = 1 - E_i(\alpha)$. We have

$$\lambda_0^{\chi}(\alpha) = \sum_i \lambda_{0,i}^{\chi}(\alpha), \tag{4.30}$$

$$\rho_0(\alpha) = \bigotimes_i \rho_{0,i}(\alpha), \tag{4.31}$$

$$l_0^{\chi}(\alpha) = \bigotimes_i l_{0,i}^{\chi}(\alpha). \tag{4.32}$$

4.2.2 Instantaneous steady currents

The instantaneous steady current is given by $i^{\rm ss}_{b\sigma}(\alpha) = \frac{\partial \lambda^{\chi}_0(\alpha)}{\partial(i\chi_{b\sigma})}\big|_{\chi=0}$. (4.30) leads to

$$i^{\rm ss}_{b\sigma}(\alpha) = \sum_i i^{\rm ss}_{b\sigma,i}(\alpha). \tag{4.33}$$

Here, $i^{\rm ss}_{b\sigma,i}(\alpha) = \frac{\partial \lambda^{\chi}_{0,i}(\alpha)}{\partial(i\chi_{b\sigma})}\big|_{\chi=0}$ are calculated from (4.26) as

$$i^{\rm ss}_{b\sigma,i}(\alpha) = \frac{\Phi^-_{b\sigma,i}\Phi^+_i - \Phi^-_i\Phi^+_{b\sigma,i}}{\Gamma_i}. \tag{4.34}$$

From (4.18), we obtain

$$\sum_{b,\sigma} i^{\rm ss}_{b\sigma,i}(\alpha) = 0. \tag{4.35}$$

From (4.24), we obtain

$$i^{\rm ss}_{b\sigma,i}(\alpha) = \frac{\Gamma_{b\sigma,i}\sum_{b'(\neq b)}\Gamma_{b',i}[f_{b'}(\tilde{\omega}_i) - f_b(\tilde{\omega}_i)]}{\Gamma_i}. \tag{4.36}$$

$i^{\rm ss}_{b\sigma,i}(\alpha)$ vanishes at zero bias ($\beta_b = \beta$, $\mu_b = \mu$). Let us consider the modulation of only the thermodynamic parameters (α'') similar to Refs.[23, 79, 24, 83]. The factor depending on α'' of $i^{\rm ss}_{b\sigma,i}(\alpha_t)$ is $f_{\beta_{b'}(t),\mu_{b'}(t)}(\tilde{\omega}_i) - f_{\beta_b(t),\mu_b(t)}(\tilde{\omega}_i)$ with $f_{\beta,\mu}(\omega) = [e^{\beta(\omega-\mu)}+1]^{-1}$. Hence

$$\langle \Delta N_{b\sigma}\rangle^{\rm ss}_\tau = \sum_i \frac{\Gamma_{b\sigma,i}}{\Gamma_i}\sum_{b'(\neq b)}\Gamma_{b',i}\int_0^\tau dt\,[f_{\beta_{b'}(t),\mu_{b'}(t)}(\tilde{\omega}_i) - f_{\beta_b(t),\mu_b(t)}(\tilde{\omega}_i)], \tag{4.37}$$

is generally nonzero and is much lager than $\langle \Delta N_{b\sigma}\rangle^{\rm Berry}_S$ because the period τ is large for adiabatic pumps. Similarly, we can show that $\langle \Delta N_{b\sigma}\rangle^{\rm ss}_\tau$ is generally nonzero for interacting system (§ 4.3.2). Reference[24] considered special modulations of only thermodynamic parameters which satisfy $\langle \Delta N_{b\sigma}\rangle^{\rm ss}_\tau = 0$. In fact, the instantaneous steady currents are always zero for arbitrary modulations of only the dynamics parameters at zero bias.

The pumped charge and spin due to the instantaneous steady currents (backgrounds) are generally nonzero even if the time averages of the bias are zero. References[68, 69] (two leads case) chose $V = \mu_L - \mu_R$ as one of the modulating parameters and considered a pumping such that $\frac{1}{\tau}\int_0^\tau dt\, V(t) = 0$ and $\langle \Delta N_{b\sigma}\rangle^{\rm ss}_\tau \neq 0$. In such pumping, the (thermal or voltage) bias is effectively nonzero.

Even if the backgrounds do not vanish, one can detect the BSN curvatures by subtracting the backgrounds by using zero-frequency measurements or by lock-in measurements. However, if one wants to apply the adiabatic pump to the current standard[50, 51], the instantaneous steady currents should be zero at all times because the backgrounds are sensitive to the velocity of the modulation of the control parameters and its trajectory. In contrast, the pumped charge and spin due to the BSN curvatures are robust against the modulation of the velocity and the trajectory. Hence, if one wants to directly apply the BSN curvatures to, for instance, the current standard, one should fix the thermodynamic parameters at zero bias.

4.2.3 BSN curvatures

In the following of this subsection, we consider one level system of which the Hamiltonian is (4.5) at $U = 0$. The instantaneous steady state is given by $|\rho_0(\alpha)\rangle\rangle = \otimes_{s=\uparrow,\downarrow}|\rho_{0,s}(\alpha)\rangle\rangle$ because the Liouvillian is described by a summation ($\hat{K}^\chi = \sum_{s=\uparrow,\downarrow}\hat{K}^\chi_s$). Similarly, the corresponding left eigenvalue is given by $\langle\langle l^\chi_0(\alpha)| = \otimes_{s=\uparrow,\downarrow}\langle\langle l^\chi_{0,s}(\alpha)|$. The BSN vectors are given by

$$A^{b\sigma}_n(\alpha) = -\sum_{s=\uparrow,\downarrow} v^{b\sigma}_s(\alpha')\frac{\partial E_s(\alpha)}{\partial\alpha^n}, \tag{4.38}$$

where

$$v^{b\sigma}_s(\alpha') = \frac{\partial v^\chi_s(\alpha)}{\partial(i\chi_{b\sigma})}\Big|_{\chi=0} = \frac{\Gamma_{b\sigma,s}}{\Gamma_s}, \tag{4.39}$$

with

$$\Gamma_{b\sigma,s}(\alpha') = 2\pi\Delta_b\sum_k |v_{bk\sigma,s}|^2\delta(\varepsilon_{bk} + \sigma g_b B_b - \omega_0 - s g_S B_S). \tag{4.40}$$

$v^{b\sigma}_s(\alpha')$ dose not depend on α''. $\sum_{b,\sigma} v^{b\sigma}_s(\alpha') = 1$ leads

$$\sum_{b,\sigma} A^{b\sigma}_n(\alpha) = -\sum_{s=\uparrow,\downarrow}\frac{\partial E_s(\alpha)}{\partial\alpha^n}. \tag{4.41}$$

This equation and (4.35) lead

$$\sum_{b,\sigma}\langle\Delta N_{b\sigma}\rangle = -\sum_{s=\uparrow,\downarrow}[E_s(\alpha_\tau) - E_s(\alpha_0)]. \tag{4.42}$$

The RHS is (-1) times the change of the total electron average number of the QD. The above equation describes the conservation of the total electron number. (4.38) leads to an expression of the BSN curvatures

$$F^{b\sigma}_{mn}(\alpha) = -\sum_{s=\uparrow,\downarrow}\Big[\frac{\partial v^{b\sigma}_s(\alpha')}{\partial\alpha^m}\frac{\partial E_s(\alpha)}{\partial\alpha^n} - (m\leftrightarrow n)\Big]. \tag{4.43}$$

We emphasize that (4.43) is consistent with the results of Refs.[68, 69, 23], which showed that the pumped charge (and also spin in Ref.[69]) vanishes at the noninteracting limit in these settings. The set of control parameters α was α'' (for Ref.[23]) and $\{\omega_0, V = \mu_L - \mu_R\}$ (for Refs.[68, 69]). If α^m or α^n is an element of α'', $F^{b\sigma}_{mn}(\alpha)$ is consistently zero. In Refs.[68, 69], the line-width functions were energy-independent, namely $\Gamma_{b\sigma,s}(\alpha') = \delta_{\sigma,s}\Gamma_b$=constant. Hence $\frac{\partial\Gamma_{b\sigma,s}(\alpha')}{\partial\omega_0} = 0 = \frac{\partial\Gamma_{b\sigma,s}(\alpha')}{\partial V}$ and $F^{b\sigma}_{\omega_0,V}(\alpha) = 0$ hold consistently.

To calculate $F^{b\sigma}_{mn}(\alpha)$, we need to assume the energy dependences of $\Gamma_{b\sigma,s}$. For the simplicity, we assume that

$$\begin{aligned}\Gamma_{b\sigma,s} &= \delta_{\sigma,s}[\Gamma_b + \Gamma'_b\cdot(s g_S B_S - \sigma g_b B_b)]\\ &= \delta_{\sigma,s}\Delta_b[\gamma_b + \gamma'_b\cdot(s g_S B_S - \sigma g_b B_b)],\end{aligned} \tag{4.44}$$

where Γ'_b are energy differential coefficients of the line-width functions at $B_b = B_S = 0$. Namely, we disregard spin flips induced by tunneling between the QD

and the leads. (4.44) is always appropriate when $|\Gamma'_b(g_S B_S - g_b B_b)| \ll \Gamma_b$ is satisfied. Additionally, we fix α'' to zero bias ($\beta_b = \beta$, $\mu_b = \mu$), in which $E_s(\alpha)$ is given by $E_s(\alpha) = f(\omega_0 + s g_S B_S)$ with $f(\omega) = [e^{\beta(\omega-\mu)}+1]^{-1}$. In the following this subsection, we suppose two leads ($b = L, R$) case. $(\alpha^m, \alpha^n) = (B_L, B_S), (\Delta_L, B_S)$ components of the charge and spin BSN curvatures of the lead L are

$$\begin{aligned}
&F^{L\uparrow}_{B_L,B_S} \pm F^{L\downarrow}_{B_L,B_S} \\
&\quad = g_S g_L \Gamma'_L [f'(\omega_0 + g_S B_S) \pm f'(\omega_0 - g_S B_S)] \frac{\Gamma_R}{\Gamma^2_{\rm tot}} \\
&\qquad - g_S g_L \Gamma'_L [f'(\omega_0 + g_S B_S) \mp f'(\omega_0 - g_S B_S)] \\
&\qquad\quad \times \Big(\Gamma'_L (g_S B_S - g_L B_L) \frac{2\Gamma_R}{\Gamma^3_{\rm tot}} + \Gamma'_R (g_S B_S - g_R B_R) \frac{\Gamma_R - \Gamma_L}{\Gamma^3_{\rm tot}} \Big), \qquad (4.45) \\
&F^{L\uparrow}_{\Delta_L,B_S} \pm F^{L\downarrow}_{\Delta_L,B_S} \\
&\quad = -g_S [f'(\omega_0 + g_S B_S) \mp f'(\omega_0 - g_S B_S)] \frac{\gamma_L \gamma_R \Delta_R}{(\gamma_L \Delta_L + \gamma_R \Delta_R)^2} \\
&-g_S [f'(\omega_0 + g_S B_S) \pm f'(\omega_0 - g_S B_S)] \gamma'_L (g_S B_S - g_L B_L) \frac{\gamma_R \Delta_R - \gamma_L \Delta_L}{(\gamma_L \Delta_L + \gamma_R \Delta_R)^2}. \qquad (4.46)
\end{aligned}$$

Here $f'(\omega) = \frac{\partial f(\omega)}{\partial \omega}$ and $\Gamma_{\rm tot} = \Gamma_L + \Gamma_R$. The pumped charge (spin) induced by a slow cycle modulation of (α^n, B_S) ($\alpha^n = B_L, \Delta_L$) are given by

$$\langle \Delta N_{L\uparrow} \rangle \pm \langle \Delta N_{L\downarrow} \rangle = \int_{S^n} d\alpha^n dB_S \, (F^{L\uparrow}_{\alpha^n,B_S} \pm F^{L\downarrow}_{\alpha^n,B_S}), \qquad (4.47)$$

where S^n are areas enclosed by the trajectories of (α^n, B_S). $F^{L\uparrow}_{\alpha^n,B_S} \pm F^{L\downarrow}_{\alpha^n,B_S}$ ($\alpha^n = B_L, \Delta_L$) are invariant under the transformation $\gamma_b \to c\gamma$, $\gamma'_b \to c\gamma'_b$ (for any $c > 0$). Hence relevant quantities are $\gamma'_b/\Gamma_{\rm tot}$. The coupling strength $\Gamma_{\rm tot}$ itself is not important. $F^{L\uparrow}_{B_L,B_S} \pm F^{L\downarrow}_{B_L,B_S}$ are proportional to $g_S g_L$ and $F^{L\uparrow}_{\Delta_L,B_S} \pm F^{L\downarrow}_{\Delta_L,B_S}$ are proportional to g_S. The first terms of the right side of (4.45) and (4.46) are dominant terms. In the limit $\gamma'_L \to 0$, $F^{L\uparrow}_{B_L,B_S} \pm F^{L\downarrow}_{B_L,B_S}$ and the second term of (4.46) vanish; however, the dominant term of (4.46) remains. At $\omega_0 = \mu$, $f'(\omega_0 + g_S B_S) - f'(\omega_0 - g_S B_S)$ vanish. Hence, at $\omega_0 = \mu$, the dominant terms of the spin BSN curvature of (B_L, B_S) pump and the charge BSN of (Δ_L, B_S) pump vanish. The contour plots of these BSN curvatures are shown in Figs. 4.1(a) and 1(b) and Figs. 4.2(a) and 2(b). The details are explained in § 4.3.3.

It is important to remark that $(\alpha^m, \alpha^n) = (B_L, B_R), (\Delta_L, \Delta_R)$ components of the charge and spin BSN curvatures are zero at zero bias because, in (4.43), $E_s(\alpha) = f(\omega_0 + s g_S B_S)$ are independent of $B_{L/R}$ and $\Delta_{L/R}$. As we showed in § 3.4, for general model, the pumping dose not occur for all orders of the pumping frequency when α_S are fixed.

4.3 Interacting system

In this section, we study the interacting system (4.5). First, we explain the Liouvillian for $0 \le U \le \infty$ (§ 4.3.1). Next, the instantaneous steady charge and spin currents are calculated at $U = \infty$ (§ 4.3.2). In § 4.3.3, we confirm the consistency between our results and Ref.[69] for $0 \le U \le \infty$. The BSN curvatures corresponding to (4.45) and (4.46) are calculated at $U = \infty$ and differences of the results between $U = 0$

and $U = \infty$ are discussed (§ 4.3.3). Finally, in § 4.3.5, we study the pumping for $0 \leq U \leq \infty$ in the wide-band limit (i.e., (4.44) with $\Gamma'_b = 0$).

4.3.1 Liouvillian

We explain the Liouvillian for $k_{\rm B}T > \Gamma$, in which the Born-Markov approximation is appropriate. The matrix representation of the Liouvillian of the RWA by the number states $\{|n_\uparrow n_\downarrow\rangle\}$ ($n_s = 0, 1$ are the numbers of an electron with spin $s = \uparrow, \downarrow$) is a 16×16 matrix which is block diagonalized to the "diagonal" space (spanned by $\{|n_\uparrow n_\downarrow\rangle\langle n_\uparrow n_\downarrow|\}_{n_\uparrow, n_\downarrow = 0,1}$) and the "off-diagonal" space (spanned by $\{|n_\uparrow n_\downarrow\rangle\langle m_\uparrow m_\downarrow|\}_{(n_\uparrow, n_\downarrow) \neq (m_\uparrow, m_\downarrow)}$). The "diagonal" block is given by

$$K^\chi(\alpha) = \begin{pmatrix} -[\Phi^+_\uparrow + \Phi^+_\downarrow] & \Phi^{-,\chi}_\uparrow & \Phi^{-,\chi}_\downarrow & 0 \\ \Phi^{+,\chi}_\uparrow & -[\Phi^-_\uparrow + \phi^+_\downarrow] & 0 & \phi^{-,\chi}_\downarrow \\ \Phi^{+,\chi}_\downarrow & 0 & -[\Phi^-_\downarrow + \phi^+_\uparrow] & \phi^{-,\chi}_\uparrow \\ 0 & \phi^{+,\chi}_\downarrow & \phi^{+,\chi}_\uparrow & -[\phi^-_\uparrow + \phi^-_\downarrow] \end{pmatrix} \begin{matrix} |0000\rangle\rangle \\ |1010\rangle\rangle \\ |0101\rangle\rangle \\ |1111\rangle\rangle \end{matrix} \tag{4.48}$$

with

$$\begin{aligned} \phi^{\pm,\chi}_s &= 2\pi \sum_b \Delta_b \sum_{k,\sigma} |v_{bk\sigma,s}|^2 f^\pm_b(\omega_0 + s g_S B_S + U) \\ &\quad \times e^{\mp i\chi_{b\sigma}} \delta(\varepsilon_{bk} + \sigma g_b B_b - \omega_0 - s g_S B_S - U), \end{aligned} \tag{4.49}$$

and $\Phi^{\pm,\chi}_s = \phi^{\pm,\chi}_s|_{U=0}$. $\phi^{\pm,\chi}_s$ satisfies

$$\phi^\pm_s = \phi^{\pm,\chi}_s\big|_{\chi=0} = \sum_{b,\sigma} \phi^\pm_{b\sigma,s}, \tag{4.50}$$

and

$$\frac{\phi^{\pm,\chi}_s}{\partial(i\chi_{b\sigma})}\Big|_{\chi=0} = \mp\phi^\pm_{b\sigma,s}, \tag{4.51}$$

with

$$\begin{aligned} \phi^\pm_{b\sigma,s} &= 2\pi\Delta_b \sum_k |v_{bk\sigma,s}|^2 f^\pm_b(\omega_0 + s g_S B_S + U) \\ &\quad \times e^{\mp i\chi_{b\sigma}} \delta(\varepsilon_{bk} + \sigma g_b B_b - \omega_0 - s g_S B_S - U). \end{aligned} \tag{4.52}$$

The off-diagonal block is a (12×12)-diagonal matrix, which dose not relate to the instantaneous steady state. At $U = 0$, $K^\chi(\alpha)$ becomes $K^\chi_\uparrow(\alpha) \otimes 1_\downarrow + 1_\uparrow \otimes K^\chi_\downarrow(\alpha)$, where $K^\chi_s(\alpha)(s = \uparrow, \downarrow)$ are given by (4.25) and 1_s are identity matrices. In the opposite limit $U \to \infty$, $K^\chi(\alpha)$ reduces to

$$K^{\chi(\infty)}(\alpha) = \begin{pmatrix} -[\Phi^+_\uparrow + \Phi^+_\downarrow] & \Phi^{-,\chi}_\uparrow & \Phi^{-,\chi}_\downarrow \\ \Phi^{+,\chi}_\uparrow & -\Phi^-_\uparrow & 0 \\ \Phi^{+,\chi}_\downarrow & 0 & -\Phi^-_\downarrow \end{pmatrix} \begin{matrix} |0000\rangle\rangle \\ |1010\rangle\rangle, \\ |0101\rangle\rangle \end{matrix} \tag{4.53}$$

because the density of states of the leads vanish at high energy ($\phi^\pm_s \to 0$).

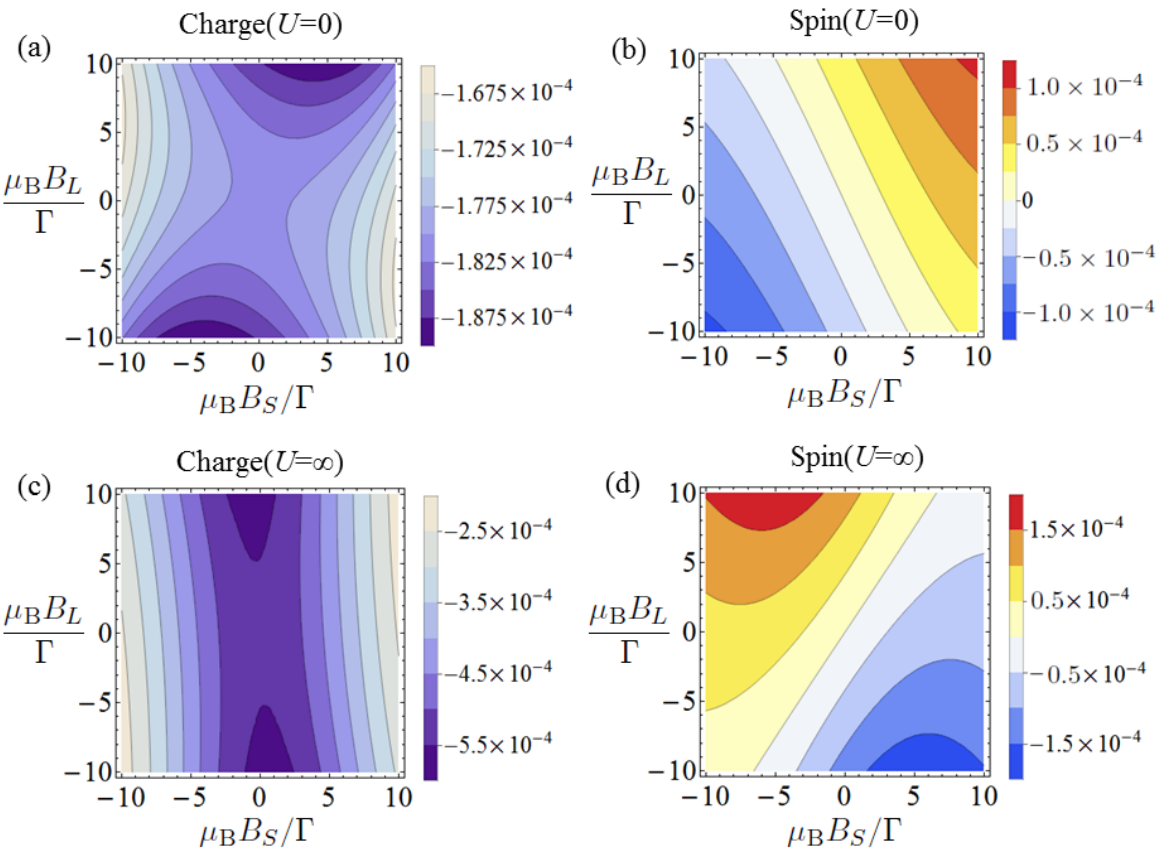

FIGURE 4.1: (a) BSN curvature of charge of (B_L, B_S) pump, $[F^{L\uparrow}_{B_L,B_S} + F^{L\downarrow}_{B_L,B_S}]/\left(\frac{\mu_B}{\Gamma}\right)^2$ at $U = 0$, (b) the BSN curvature of spin, $[F^{L\uparrow}_{B_L,B_S} - F^{L\downarrow}_{B_L,B_S}]/\left(\frac{\mu_B}{\Gamma}\right)^2$ at $U = 0$, (c) $[F^{L\uparrow}_{B_L,B_S} + F^{L\downarrow}_{B_L,B_S}]/\left(\frac{\mu_B}{\Gamma}\right)^2$ at $U = \infty$, and (d) $[F^{L\uparrow}_{B_L,B_S} - F^{L\downarrow}_{B_L,B_S}]/\left(\frac{\mu_B}{\Gamma}\right)^2$ at $U = \infty$. The values of the parameters used for these plots are $\Gamma_L = \Gamma_R = \Gamma$, $\Gamma'_L = \Gamma'_R = 0.1$, $\beta = 0.5/\Gamma$, $\omega_0 = \mu - 3\Gamma$, and $B_R = 0$, and all g factors (g^*_L, g^*_R, g^*_S) are -0.44 (bulk GaAs).

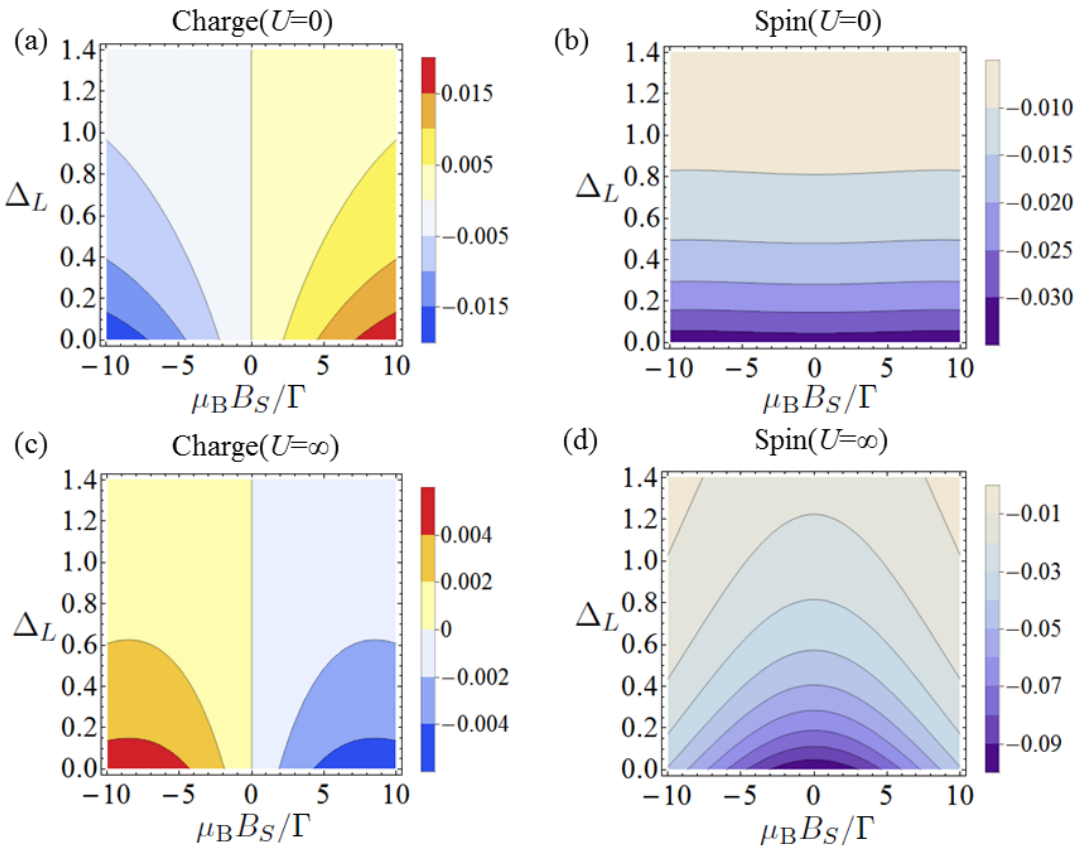

FIGURE 4.2: (a) BSN curvature of charge of (Δ_L, B_S) pump, $[F^{L\uparrow}_{\Delta_L,B_S} + F^{L\downarrow}_{\Delta_L,B_S}]/\frac{\mu_B}{\Gamma}$ at $U = 0$, (b) the BSN curvature of spin, $[F^{L\uparrow}_{\Delta_L,B_S} - F^{L\downarrow}_{\Delta_L,B_S}]/\frac{\mu_B}{\Gamma}$ at $U = 0$, (c) $[F^{L\uparrow}_{\Delta_L,B_S} + F^{L\downarrow}_{\Delta_L,B_S}]/\frac{\mu_B}{\Gamma}$ at $U = \infty$, and (d) $[F^{L\uparrow}_{\Delta_L,B_S} - F^{L\downarrow}_{\Delta_L,B_S}]/\frac{\mu_B}{\Gamma}$ at $U = \infty$. The values of the parameters used for these plots are $\gamma_L = \Gamma_R = \Gamma$, $\gamma'_L = \Gamma'_R = 0.1$, and $B_L = 0$ and other conditions are the same as Fig. 4.1.

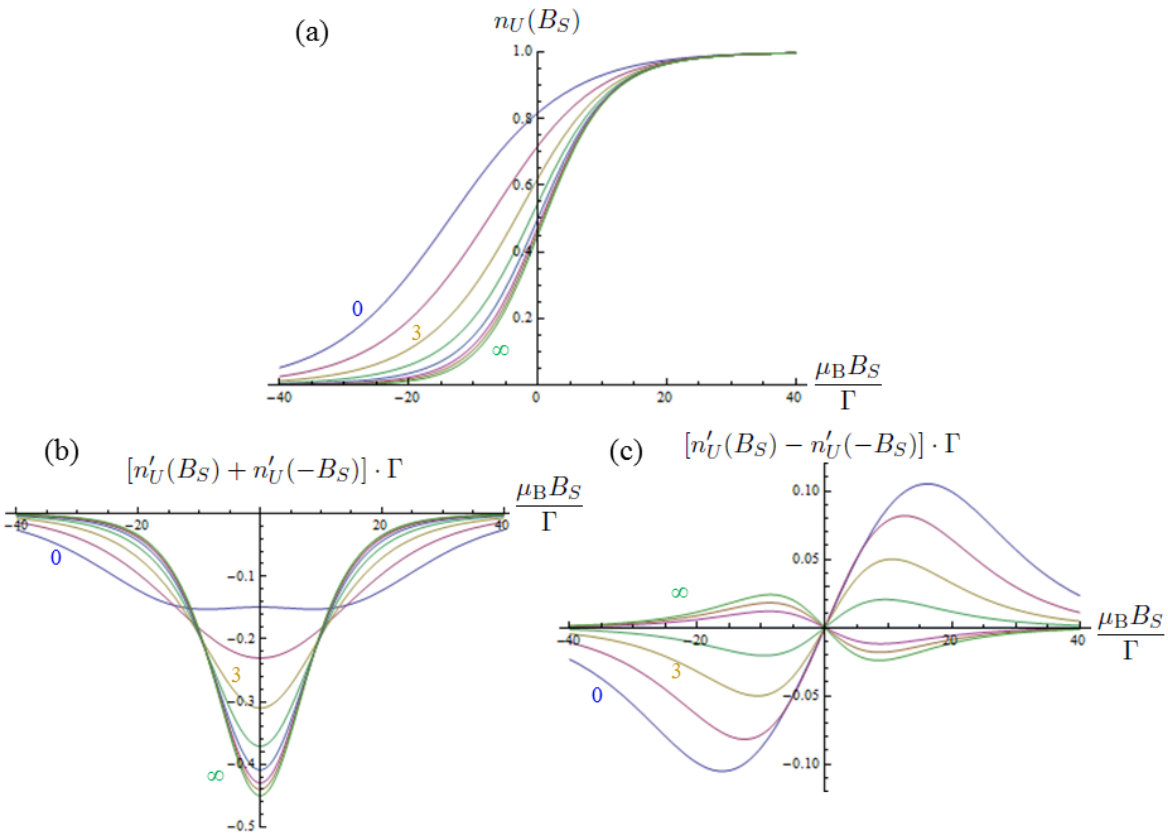

FIGURE 4.3: (a)$n_S(B)$, (b)$[n'_S(B) + n'_S(-B)] \cdot \Gamma$, (c)$[n'_S(B) - n'_S(-B)] \cdot \Gamma$ for U/Γ =0, 1.5, 3, 4,5, 6, 7.5, 9, ∞. Here, $n'_U(\pm B_S) = \frac{1}{g_S}\frac{\partial n_U(B)}{\partial B}|_{B=\pm B_S}$ and $n_U(B_S)$ is defined by (4.92). The conditions are the same as Fig. 4.1.

4.3.2 Instantaneous steady currents for $U \to \infty$

In this subsection, we set $U = \infty$. The characteristic polynomial of $K^{\chi(\infty)}$ is denoted as

$$C_3(\chi, \lambda) = \det(K^{\chi(\infty)} - \lambda) = \sum_{n=0}^{2} C_n(\chi)\lambda^n - \lambda^3. \tag{4.54}$$

Because of $C_0(0) = 0$, $\lambda = 0$ is one of the solutions at $\chi = 0$. Now we set $\chi_{b\sigma}$ as infinitesimal and other counting fields are zero. Then, the eigenvalue corresponding to the instantaneous steady state is given by $\lambda = \lambda_0^\chi = i\chi_{b\sigma} \cdot i_{b\sigma}^{\rm ss} + \mathcal{O}(\chi_{b\sigma}^2)$. It leads to $0 = C_3(\chi, \lambda_0^\chi) = C_1(0) i\chi_{b\sigma} i_{b\sigma}^{\rm ss} + i\chi_{b\sigma} C_0^{b\sigma}$ with $C_0^{b\sigma} = \frac{\partial C_0(\chi)}{\partial(i\chi_{b\sigma})}\big|_{\chi=0}$, and we obtain

$$i_{b\sigma}^{\rm ss} = -\frac{C_0^{b\sigma}}{C_1(0)}, \tag{4.55}$$

with $C_1(0) = -[\Phi_\uparrow^+\Phi_\downarrow^- + \Phi_\uparrow^-\Phi_\downarrow^+ + \Phi_\uparrow^-\Phi_\downarrow^-]$. From $C_0(\chi) = -[\Phi_\uparrow^+ + \Phi_\downarrow^+]\Phi_\uparrow^-\Phi_\downarrow^- + \Phi_\downarrow^{-,\chi}\Phi_\uparrow^-\Phi_\downarrow^{+,\chi} + \Phi_\downarrow^-\Phi_\uparrow^{-,\chi}\Phi_\uparrow^{+,\chi}$, we have

$$i_{b\sigma}^{\rm ss} = \frac{\Phi_\uparrow^-(\Phi_{b\sigma,\downarrow}^-\Phi_\downarrow^+ - \Phi_\downarrow^-\Phi_{b\sigma,\downarrow}^+) + \Phi_\downarrow^-(\Phi_{b\sigma,\uparrow}^-\Phi_\uparrow^+ - \Phi_\uparrow^-\Phi_{b\sigma,\uparrow}^+)}{\Phi_\uparrow^+\Phi_\downarrow^- + \Phi_\uparrow^-\Phi_\downarrow^+ + \Phi_\uparrow^-\Phi_\downarrow^-}. \tag{4.56}$$

The total instantaneous steady current vanishes:

$$\sum_{b,\sigma} i_{b\sigma}^{\rm ss} = 0. \tag{4.57}$$

$i_{b\sigma}^{\rm ss}$ can be rewritten as

$$i_{b\sigma}^{\rm ss} = \frac{\sum_{s=\uparrow,\downarrow}\Phi_{-s}^-\Gamma_{b\sigma,s}\sum_{b'(\neq b)}\Gamma_{b',s}[f_{b'}(\omega_s) - f_b(\omega_s)]}{\Phi_\uparrow^+\Phi_\downarrow^- + \Phi_\uparrow^-\Phi_\downarrow^+ + \Phi_\uparrow^-\Phi_\downarrow^-}. \tag{4.58}$$

Here, Φ_{-s}^- ($s = \uparrow, \downarrow$) describes $\Phi_\downarrow^-$ for $s = \uparrow$ and $\Phi_\downarrow^-$ for $s = \downarrow$. At zero bias, the instantaneous steady currents vanish. Similar to § 4.2.2, $\langle \Delta N_{b\sigma}\rangle_\tau^{\rm ss}$ are generally nonzero when α'' is not fixed at zero bias.

4.3.3 BSN curvatures for $U \to \infty$

The instantaneous steady state $\rho_0(\alpha)$ and corresponding left eigenvector $l_0^\chi(\alpha)$ are written as

$$\rho_0 = \rho_0|00\rangle\langle 00| + \rho_\uparrow|10\rangle\langle 10| + \rho_\downarrow|01\rangle\langle 01| + \rho_2|11\rangle\langle 11|, \tag{4.59}$$

and

$$l_0^\chi = |00\rangle\langle 00| + l_\uparrow^\chi|10\rangle\langle 10| + l_\downarrow^\chi|01\rangle\langle 01| + l_2^\chi|11\rangle\langle 11|. \tag{4.60}$$

The BSN vectors are given by

$$A_n^{b\sigma}(\alpha) = -\sum_{c=\uparrow,\downarrow,2} l_c^{b\sigma}(\alpha)\frac{\partial\rho_c(\alpha)}{\partial\alpha^n}, \tag{4.61}$$

where $l_c^{b\sigma}(\alpha) = \frac{\partial[l_c^{\chi}(\alpha)]^*}{\partial(i\chi_{b\sigma})}\big|_{\chi=0}$. It leads to the BSN curvatures

$$F_{mn}^{b\sigma}(\alpha) = -\sum_{c=\uparrow,\downarrow,2} \frac{\partial l_c^{b\sigma}(\alpha)}{\partial\alpha^m}\frac{\partial\rho_c(\alpha)}{\partial\alpha^n} - (m \leftrightarrow n). \tag{4.62}$$

We confirmed the consistency between our results and Ref.[69], which studied the similar system for $0 \leq U \leq \infty$ using the wide-band limit. As we explained in Chap.3, $\varphi_\kappa^{O_\mu}(\alpha)$ of (3.88) corresponds to $-\langle\langle l_0^{O_\mu}(\alpha)|$, namely $-l_c^{b\sigma}(\alpha)$. In the condition of the wide-band limit, we calculated $l_c^{b\sigma}(\alpha)$ $(c =\uparrow,\downarrow,2)$ for $0 \leq U \leq \infty$ and confirmed numerically the correspondence between $\varphi_c^{O_\mu}(\alpha)$ $(c =\uparrow,\downarrow,2)$ and $-[l_c^{b\uparrow}(\alpha) \pm l_c^{b\downarrow}(\alpha)]$ for the charge and spin pump.

Particularly, in the limit $U \to \infty$, ρ_2 vanishes and $F_{mn}^{b\sigma}(\alpha)$ reduces to

$$F_{mn}^{b\sigma(\infty)}(\alpha) = -\sum_{s=\uparrow,\downarrow} \frac{\partial l_s^{b\sigma(\infty)}(\alpha)}{\partial\alpha^m}\frac{\partial\rho_s^{(\infty)}(\alpha)}{\partial\alpha^n} - (m \leftrightarrow n), \tag{4.63}$$

where $\rho_s^{(\infty)}(\alpha)$ and $l_s^{b\sigma(\infty)}(\alpha)$ are the limits $U \to \infty$ of $\rho_s(\alpha)$ and $l_s^{b\sigma}(\alpha)$, respectively. From (4.53) we obtain

$$\rho_s^{(\infty)}(\alpha) = \frac{\Phi_s^+\Phi_{-s}^-}{\Phi_\uparrow^-\Phi_\downarrow^- + \Phi_\uparrow^-\Phi_\downarrow^+ + \Phi_\uparrow^+\Phi_\downarrow^-}, \tag{4.64}$$

$$\{l_s^{(\infty)}(\alpha)\}^* = \frac{\Phi_s^{-,\chi}}{\Phi_s^- + \lambda_0^\chi}, \tag{4.65}$$

and

$$l_s^{b\sigma(\infty)}(\alpha) = \frac{\Phi_{b\sigma,s}^- - i_{b\sigma}^{ss}(\alpha)}{\Phi_s^-}. \tag{4.66}$$

(4.57) leads $\sum_{b,\sigma} l_s^{b\sigma(\infty)}(\alpha) = 1$. Then, we obtain

$$\sum_{b,\sigma} A_n^{b\sigma(\infty)}(\alpha) = -\sum_{c=\uparrow,\downarrow,2} \frac{\partial\rho_c^{(\infty)}(\alpha)}{\partial\alpha^n}. \tag{4.67}$$

This equation and (4.57) lead

$$\sum_{b,\sigma}\langle\Delta N_{b\sigma}\rangle = -\sum_{s=\uparrow,\downarrow}[\rho_c^{(\infty)}(\alpha_\tau) - \rho_c^{(\infty)}(\alpha_0)]. \tag{4.68}$$

The RHS is (-1) times the change of the total electron average number of the QD. The above equation describes the conservation of the total electron number. In the following of this subsection, we fix α'' to zero bias ($\beta_b = \beta$, $\mu_b = \mu$) and suppose (4.44). Then, $l_s^{b\sigma(\infty)}(\alpha)$ equals $v_s^{b\sigma}(\alpha')$ given by (4.39) and $\rho_s^{(\infty)}(\alpha)$ are given by

$$\rho(sB_S) = \frac{e^{-\beta(\omega_s-\mu)}}{1 + e^{-\beta(\omega_\downarrow-\mu)} + e^{-\beta(\omega_\uparrow-\mu)}}. \tag{4.69}$$

We emphasize that $F_{mn}^{b\sigma(\infty)}(\alpha)$ can be obtained by just a replacement,

$$E_s(\alpha) = f(\omega_s) \to \rho(sB_S), \tag{4.70}$$

in (4.43). In the following this subsection, we suppose two leads ($b = L, R$) case. The charge and spin BSN curvatures of $(B_L, B_S), (\Delta_L, B_S)$ pump are given by a replacement $f'(\omega_0 \pm g_S B_S) \to \rho'(\pm B_S)$ in (4.45) and (4.46), where $\rho'(B_S) \stackrel{\text{def}}{=} \frac{1}{g_S}\frac{\partial \rho(B_S)}{\partial B_S}$. Similar to $U = 0$, the charge and spin BSN curvatures of $(B_L, B_R), (\Delta_L, \Delta_R)$ pump are zero.

In Figs. 4.1(a)-4.1(d), we plot the BSN curvatures of (B_L, B_S) pump normalized by $(\mu_{\rm B}/\Gamma)^2$, where $\Gamma = \Gamma_L = \Gamma_R$ and $\mu_{\rm B} = 57.88\ \mu$eV/T is the Bohr magneton. For $U = 0$, the charge and spin BSN curvatures are shown in Fig. 4.1(a) and Fig. 4.1(b), and for $U = \infty$ these are shown in Figs. 4.1(c) and 4.1(d). The horizontal and vertical axes of these plots are the strength of the magnetic fields B_S and B_L normalized by $\Gamma/\mu_{\rm B}$. The values of the parameters used for these plots are $\Gamma_L = \Gamma_R = \Gamma$, $\Gamma'_L = \Gamma'_R = 0.1$, $\beta = 0.5/\Gamma$, $\omega_0 = \mu - 3\Gamma$, $B_R = 0$, and $g^*_L = g^*_R = g^*_S = -0.44$ (bulk GaAs). The BSN curvatures of (Δ_L, B_S) pump normalized by $\mu_{\rm B}/\Gamma$ are shown similarly in Figs. 4.2(a)-4.2(d). In all plots, $\gamma_L = \Gamma_R = \Gamma$, $\gamma'_L = \Gamma'_R = 0.1$, $B_L = 0$, and other conditions are the same as in Fig. 4.1. In Figs. 4.1 and 4.2, the maximum values of $|\Gamma'_b(g_S B_S - g_b B_b)|/\Gamma_b$ are 0.44 and 0.22 (<1), respectively. The pumped charges and spins are given by (4.47).

Figure 4.3(a) shows the instantaneous average numbers of the up spin electron of the QD, $n_U(B_S)$ defined by (4.92) at U/Γ =0, 1.5, 3, 4.5, 6, 7.5, 9, ∞ for $\beta = 0.5/\Gamma$, $\omega_0 = \mu - 3\Gamma$, and $g_S = -0.44 \times \mu_{\rm B}/2$. In particular, $n_0 = f(\omega_0 + g_S B_S)$ and $n_\infty(B_S) = \rho(B_S)$ hold. Because two electrons cannot occupy a QD at $U = \infty$, the magnetic field dependence of $\rho(B_S)$ is more sensitive than $f(\omega_0 + g_S B_S)$. Figures 4.3(b) and 4.3(c) show $n'_U(B_S) \pm n'_U(-B_S)$ normalized by $1/\Gamma$, where $n'_U(\pm B_S) = \frac{1}{g_S}\frac{\partial n_U(B)}{\partial B}|_{B=\pm B_S}$.

In Figs. 4.2(a) and 4.2(c), the charge BSN curvatures of (Δ_L, B_S) pump vanish at $B_S = 0$. This is because the first term of (4.46) vanishes since $n'(B_S) - n'(-B_S) = 0$ (n denotes n_0 or n_∞) for $B_S = 0$ and the second term vanishes since $g_S B_S - g_b B_b = 0$ for $B_S = 0 = B_L$. Similarly, in Figs. 4.1(b) and 4.1(d), the spin BSN curvatures of (B_L, B_S) pump vanish at $B_S = 0 = B_L$. The zero lines in these plots relate to the cancellation between the first and second terms of (4.45). Figures. 4.1(a), 4.1(c) and Figs. 4.1(b), 4.1(d) are respectively symmetric and antisymmetric under the transformation $(B_S, B_L) \to (-B_S, -B_L)$. Similarly, Figs. 4.2(b), 4.2(d) and Figs. 4.2(a), 4.2(c) are respectively symmetric and antisymmetric under the transformation $B_S \to -B_S$. We emphasize that pure charge and pure spin pumps are respectively realized for (B_L, B_S) pump and (Δ_L, B_S) pump such that the areas S^n in (4.47) are symmetric under the above transformations. An instance of symmetric area of (B_L, B_S) pump is a disk of which the center is $B_S = 0 = B_L$.

In $\omega_0 > \mu$ region, the larger $\omega_0 - \mu$, the less difference between $U = 0$ and $U = \infty$ becomes. The Coulomb interaction prevents two electrons from occupying the QD. This effect is conspicuous in the $\omega_0 < \mu$ region, although it is not important in the $\omega_0 > \mu$ region.

As shown in Figs. 4.1(a), 4.1(c) and Figs. 4.2(b), 4.2(d), the B_S dependence of the charge BSN curvature of (B_L, B_S) pump and the spin BSN curvature of (Δ_L, B_S) pump at $U = 0$ are more gentle than those at $U = \infty$. It results from the behavior of $n'(B_S) + n'(-B_S)$ as shown in Fig. 4.3(b).

As shown in Figs. 4.1(b), 4.1(d) and in Figs. 4.2(a), 4.2(c), the B_S dependence of the spin BSN curvature of (B_L, B_S) pump and the charge BSN curvature of (Δ_L, B_S) pump are opposite. This is because the leading term (in weak magnetic field region) of these are proportional to $n'(B_S) - n'(-B_S)$ and its B_S dependence is opposite in $U = 0$ and $U = \infty$ for $\omega_0 - \mu < 0$ as indicated in Fig. 4.3(b). This inversion is realized for only $\omega_0 - \mu < 0$ region. At $\omega_0 = \mu$, $f'(\omega_0 + g_S B_S) - f'(\omega_0 - g_S B_S)$ vanish. In

$\omega_0 > \mu$ region, the signs of $f'(\omega_0 + g_S B_S) - f'(\omega_0 - g_S B_S)$ and $\rho'(B_S) - \rho'(-B_S)$ are the same.

In Figs. 4.1 and 4.2, absolute values of the normalized BSN curvatures are smaller than unity. However, we can improve this problem by tuning g factors. The first and second terms of the right side of (4.45) are the second and third order in the g factors, and the first and second terms of the right side of (4.46) are the first and second order in the g factors. If all g factors change to -20 (for example for the materials like InAs, InSb), the first, second, and third order terms become about 45, 2 000, and 90 000 times. In fact, for these values of g factors, the assumption (4.44) is not appropriate for magnetic fields that are not small; we need concrete energy dependence of the line-width functions.

4.3.4 Instantaneous steady currents

The characteristic polynomial of (4.48) is

$$\det(K^\chi - \lambda) = \sum_{n=0}^{3} c_n(\chi)\lambda^n + \lambda^4. \tag{4.71}$$

Similar to (4.3.2), we obtain

$$i_{b\sigma}^{\rm ss} = -\frac{c_0^{b\sigma}}{c_1(0)}, \tag{4.72}$$

with $c_0^{b\sigma} = \frac{\partial c_0(\chi)}{\partial(i\chi_{b\sigma})}\big|_{\chi=0}$. $c_0(\chi)$ is given by

$$\begin{aligned} c_0(\chi) \;=\; & K_{00}K_{\uparrow\uparrow}K_{\downarrow\downarrow}K_{22} - K_{00}K_{\downarrow\downarrow}\phi_\downarrow^{-,\chi}\phi_\downarrow^{+,\chi} - K_{00}K_{\uparrow\uparrow}\phi_\uparrow^{-,\chi}\phi_\uparrow^{+,\chi} \\ & -K_{\downarrow\downarrow}K_{22}\Phi_\uparrow^{-,\chi}\Phi_\uparrow^{+,\chi} - K_{22}K_{\uparrow\uparrow}\Phi_\downarrow^{-,\chi}\Phi_\downarrow^{+,\chi} \\ & -\Phi_\uparrow^{-,\chi}\Phi_\downarrow^{+,\chi}\phi_\downarrow^{-,\chi}\phi_\uparrow^{+,\chi} + \Phi_\uparrow^{-,\chi}\Phi_\uparrow^{+,\chi}\phi_\uparrow^{-,\chi}\phi_\uparrow^{+,\chi} \\ & +\Phi_\downarrow^{-,\chi}\Phi_\downarrow^{+,\chi}\phi_\downarrow^{-,\chi}\phi_\downarrow^{+,\chi} - \Phi_\downarrow^{-,\chi}\Phi_\uparrow^{+,\chi}\phi_\uparrow^{-,\chi}\phi_\downarrow^{+,\chi}. \end{aligned} \tag{4.73}$$

Then, we obtain

$$\begin{aligned} c_0^{b\sigma} \;=\; & -K_{00}K_{\downarrow\downarrow}[\phi_{b\sigma,\downarrow}^-\phi_\downarrow^+ - \phi_\downarrow^-\phi_{b\sigma,\downarrow}^+] - K_{00}K_{\uparrow\uparrow}[\phi_{b\sigma,\uparrow}^-\phi_\uparrow^+ - \phi_\uparrow^-\phi_{b\sigma,\uparrow}^+] \\ & -K_{\downarrow\downarrow}K_{22}[\Phi_{b\sigma,\uparrow}^-\Phi_\uparrow^+ - \Phi_\uparrow^-\Phi_{b\sigma,\uparrow}^+] - K_{22}K_{\uparrow\uparrow}[\Phi_{b\sigma,\downarrow}^-\Phi_\downarrow^+ - \Phi_\downarrow^-\Phi_{b\sigma,\downarrow}^+] \\ & -[\Phi_{b\sigma,\uparrow}^-\Phi_\downarrow^+ - \Phi_\uparrow^-\Phi_{b\sigma,\downarrow}^+]\phi_\downarrow^-\phi_\uparrow^+ - \Phi_\uparrow^-\Phi_\downarrow^+[\phi_{b\sigma,\downarrow}^-\phi_\uparrow^+ - \phi_\downarrow^-\phi_{b\sigma,\uparrow}^+] \\ & +[\Phi_{b\sigma,\uparrow}^-\Phi_\uparrow^+ - \Phi_\uparrow^-\Phi_{b\sigma,\uparrow}^+]\phi_\uparrow^-\phi_\uparrow^+ + \Phi_\uparrow^-\Phi_\uparrow^+[\phi_{b\sigma,\uparrow}^-\phi_\uparrow^+ - \phi_\uparrow^-\phi_{b\sigma,\uparrow}^+] \\ & +[\Phi_{b\sigma,\downarrow}^-\Phi_\downarrow^+ - \Phi_\downarrow^-\Phi_{b\sigma,\downarrow}^+]\phi_\downarrow^-\phi_\downarrow^+ + \Phi_\downarrow^-\Phi_\downarrow^+[\phi_{b\sigma,\downarrow}^-\phi_\downarrow^- - \phi_\downarrow^+\phi_{b\sigma,\downarrow}^+] \\ & -[\Phi_{b\sigma,\downarrow}^-\Phi_\uparrow^+ - \Phi_\downarrow^-\Phi_{b\sigma,\uparrow}^+]\phi_\uparrow^-\phi_\downarrow^+ - \Phi_\downarrow^-\Phi_\uparrow^+[\phi_{b\sigma,\uparrow}^-\phi_\downarrow^+ - \phi_\uparrow^-\phi_{b\sigma,\downarrow}^+]. \end{aligned} \tag{4.74}$$

Here,

$$\phi_{b\sigma,s}^\pm = 2\pi\Delta_b \sum_k |v_{bk\sigma,s}|^2 f_b^\pm(\omega_s + U)\delta(\varepsilon_{bk} + \sigma g_b B_b - \omega_s - U), \tag{4.75}$$

and $\Phi^{\pm}_{b\sigma,s} = \phi^{\pm}_{b\sigma,s}\big|_{U=0}$. $c_1(0)$ is given by

$$\begin{aligned} c_1(0) \;=\; & -K_{00}K_{\uparrow\uparrow}K_{\downarrow\downarrow} - K_{00}K_{\uparrow\uparrow}K_{22} - K_{00}K_{\downarrow\downarrow}K_{22} - K_{\uparrow\uparrow}K_{\downarrow\downarrow}K_{22} \\ & +K_{\downarrow\downarrow}(\phi^-_\downarrow\phi^+_\downarrow + \Phi^-_\uparrow\Phi^+_\uparrow) + K_{00}(\phi^-_\downarrow\phi^+_\downarrow + \phi^-_\uparrow\phi^+_\uparrow) \\ & +K_{\uparrow\uparrow}(\phi^-_\uparrow\phi^+_\uparrow + \Phi^-_\downarrow\Phi^+_\downarrow) + K_{22}(\Phi^-_\uparrow\Phi^+_\uparrow + \Phi^-_\downarrow\Phi^+_\downarrow). \end{aligned} \tag{4.76}$$

$\sum_{b,\sigma} c_0^{b\sigma} = 0$ leads $\sum_{b,\sigma} i^{\rm ss}_{b\sigma} = 0$. At zero-bias, $i^{\rm ss}_{b\sigma}$ vanishes.

4.3.5 BSN curvatures

$(l^\chi_c)^*$ are given by

$$(l^\chi_\uparrow)^* \;=\; \frac{[-K_{\downarrow\downarrow}+\lambda^\chi_0][-K_{00}+\lambda^\chi_0]\phi^{+,\chi}_\downarrow + \phi^{+,\chi}_\uparrow\Phi^{-,\chi}_\uparrow\Phi^{+,\chi}_\downarrow - \phi^{+,\chi}_\downarrow\Phi^{-,\chi}_\downarrow\Phi^{+,\chi}_\downarrow}{[-K_{\downarrow\downarrow}+\lambda^\chi_0]\Phi^{+,\chi}_\uparrow\phi^{+,\chi}_\downarrow + [-K_{\uparrow\uparrow}+\lambda^\chi_0]\Phi^{+,\chi}_\downarrow\phi^{+,\chi}_\uparrow}, \tag{4.77}$$

$$(l^\chi_\downarrow)^* \;=\; \frac{[-K_{\uparrow\uparrow}+\lambda^\chi_0][-K_{00}+\lambda^\chi_0]\phi^{+,\chi}_\uparrow + \phi^{+,\chi}_\downarrow\Phi^{-,\chi}_\downarrow\Phi^{+,\chi}_\uparrow - \phi^{+,\chi}_\uparrow\Phi^{-,\chi}_\uparrow\Phi^{+,\chi}_\uparrow}{[-K_{\uparrow\uparrow}+\lambda^\chi_0]\Phi^{+,\chi}_\downarrow\phi^{+,\chi}_\uparrow + [-K_{\downarrow\downarrow}+\lambda^\chi_0]\Phi^{+,\chi}_\uparrow\phi^{+,\chi}_\downarrow}, \tag{4.78}$$

$$(l^\chi_2)^* \;=\; \frac{-\Phi^{-,\chi}_\uparrow + [-K_{\uparrow\uparrow}+\lambda^\chi_0](l^\chi_\uparrow)^*}{\phi^{+,\chi}_\downarrow}. \tag{4.79}$$

Similarly, we obtain $\rho_c = \rho_0 r_c$ ($c = \uparrow, \downarrow, 2$) with

$$r_\uparrow \;=\; \frac{K_{\downarrow\downarrow}K_{00}\phi^-_\downarrow + \phi^-_\uparrow\Phi^+_\uparrow\Phi^-_\downarrow - \phi^-_\downarrow\Phi^+_\downarrow\Phi^-_\downarrow}{-K_{\downarrow\downarrow}\Phi^-_\uparrow\phi^-_\downarrow - K_{\uparrow\uparrow}\Phi^-_\downarrow\phi^-_\uparrow}, \tag{4.80}$$

$$r_\downarrow \;=\; \frac{K_{\uparrow\uparrow}K_{00}\phi^-_\uparrow + \phi^-_\downarrow\Phi^+_\downarrow\Phi^-_\uparrow - \phi^-_\uparrow\Phi^+_\uparrow\Phi^-_\uparrow}{-K_{\uparrow\uparrow}\Phi^-_\downarrow\phi^-_\uparrow - K_{\downarrow\downarrow}\Phi^-_\uparrow\phi^-_\downarrow}, \tag{4.81}$$

$$r_2 \;=\; \frac{-\Phi^+_\uparrow - K_{\uparrow\uparrow}r_\uparrow}{\phi^-_\downarrow}, \tag{4.82}$$

$$\rho_0 \;=\; \frac{1}{1+\sum_{c=\uparrow,\downarrow,2} r_c}. \tag{4.83}$$

In the following of this subsection, we suppose zero-bias. Then, ρ_0 and ρ_c become

$$\rho_0 \;=\; \frac{1}{\Xi},\; \rho_s = \frac{e^{-\beta(\omega_s-\mu)}}{\Xi},\; \rho_2 = \frac{e^{-\beta(\omega_\uparrow+\omega_\downarrow+U-2\mu)}}{\Xi}. \tag{4.84}$$

Here,

$$\Xi \;=\; 1 + e^{-\beta(\omega_\uparrow-\mu)} + e^{-\beta(\omega_\downarrow-\mu)} + e^{-\beta(\omega_\uparrow+\omega_\downarrow+U-2\mu)}. \tag{4.85}$$

In the following of this subsection, we suppose that the line-width functions do not depend on the energy. Then, we obtain

$$l^{b\sigma}_s \;=\; \frac{\Gamma_{b\sigma,s}}{\Gamma} = v^{b\sigma}_s, \tag{4.86}$$

$$l^{b\sigma}_2 \;=\; \frac{\Gamma_{b\sigma,\uparrow}+\Gamma_{b\sigma,\downarrow}}{\Gamma} = l^{b\sigma}_\uparrow + l^{b\sigma}_\downarrow. \tag{4.87}$$

Substituting these two equations to (4.62), we obtain

$$F^{b\sigma}_{mn}(\alpha) = -\sum_{s=\uparrow,\downarrow}\Big(\frac{\partial}{\partial\alpha^m}\frac{\Gamma_{b\sigma,s}}{\Gamma}\Big)\frac{\partial n_s(\alpha)}{\partial\alpha^n} - (m \leftrightarrow n). \tag{4.88}$$

Here,

$$n_s = \rho_s + \rho_2 = \frac{e^{-\beta(\omega_s-\mu)} + e^{-\beta(2\omega_0+U-2\mu)}}{\Xi}, \tag{4.89}$$

is the average number of the electrons in the QD with spin s. Because the line-width functions are energy-independent, the BSN curvatures of (B_b, B_S)-pump vanish. In the following this subsection, we suppose two leads ($b = L, R$) case. If we suppose

$$\Gamma_{b\sigma,s}(\omega) = \delta_{\sigma,s}\Gamma_b = \delta_{\sigma,s}\Delta_b\gamma_b, \tag{4.90}$$

the BSN curvatures of (Δ_L, B_S)-pump are given by

$$F^{L\uparrow}_{\Delta_L,B_S} \pm F^{L\downarrow}_{\Delta_L,B_S} = -g_S[n'_U(B_S) \mp n'_U(-B_S)]\frac{\gamma_L\gamma_R\Delta_R}{(\gamma_L\Delta_L+\gamma_R\Delta_R)^2}, \tag{4.91}$$

where

$$n_U(B_S) \stackrel{\text{def}}{=} n_\uparrow = \frac{e^{-\beta(\omega_\uparrow-\mu)} + e^{-\beta(2\omega_0+U-2\mu)}}{1+e^{-\beta(\omega_\uparrow-\mu)}+e^{-\beta(\omega_\downarrow-\mu)}+e^{-\beta(2\omega_0+U-2\mu)}}, \tag{4.92}$$

$$n'_U(B_S) \stackrel{\text{def}}{=} \frac{1}{g_S}\frac{\partial n_U(B)}{\partial B}\Big|_{B=\pm B_S}. \tag{4.93}$$

Because $n_0 = f(\omega_0 + g_S B_S)$ and $n_\infty(B_S) = \rho(B_S)$, (4.91) confirms with the results of § 4.2.3 and § 4.3.3. $n'_U(sB_S)$ and $n'_U(B_S) \mp n'_U(-B_S)$ are given by

$$\begin{aligned} &n'_U(sB_S) \\ &= -\beta\frac{e^{-\beta(\omega_0-\mu)}[e^{-s\beta g_S B_S} + e^{s\beta g_S B_S}e^{-\beta[2(\omega_0-\mu)+U]} + 2e^{-\beta(\omega_0-\mu)}]}{\{1+e^{-\beta(\omega_0-\mu)}[e^{\beta g_S B_S}+e^{-\beta g_S B_S}]+e^{-\beta[2(\omega_0-\mu)+U]}\}^2}, \end{aligned} \tag{4.94}$$

$$\begin{aligned} &n'_U(B_S) - n'_U(-B_S) \\ &= \beta[1-e^{-\beta[2(\omega_0-\mu)+U]}]\frac{e^{-\beta(\omega_0-\mu)}(e^{\beta g_S B_S} - e^{-\beta g_S B_S})}{\{1+e^{-\beta(\omega_0-\mu)}[e^{\beta g_S B_S}+e^{-\beta g_S B_S}]+e^{-\beta[2(\omega_0-\mu)+U]}\}^2}, \end{aligned} \tag{4.95}$$

$$\begin{aligned} &n'_U(B_S) + n'_U(-B_S) \\ &= -\beta\frac{e^{-\beta(\omega_0-\mu)}[1+e^{-\beta[2(\omega_0-\mu)+U]}](e^{\beta g_S B_S}+e^{-\beta g_S B_S}) + 4e^{-2\beta(\omega_0-\mu)}}{\{1+e^{-\beta(\omega_0-\mu)}[e^{\beta g_S B_S}+e^{-\beta g_S B_S}]+e^{-\beta[2(\omega_0-\mu)+U]}\}^2}. \end{aligned} \tag{4.96}$$

In particular, at

$$\omega_0 - \mu = -\frac{U}{2}, \tag{4.97}$$

$n'_U(B_S) - n'_U(-B_S) = 0$ and $F^{L\uparrow}_{\Delta_L,B_S} + F^{L\downarrow}_{\Delta_L,B_S} = 0$ hold. (4.97) is called the half-filling condition. Under this condition, pure spin pump is realized. $n'_U(B_S) - n'_U(-B_S)$ is proportional to $F_U \stackrel{\text{def}}{=} 1 - e^{-\beta[2(\omega_0-\mu)+U]}$. This factor becomes $F_0 = 1 - e^{-\beta[2(\omega_0-\mu)]}$ at $U = 0$ and $F_\infty = 1$ at $U \to \infty$. If $\omega_0 - \mu < 0$, $F_0 < 0$ holds and $n'_U(B_S) - n'_U(-B_S)$ is negative for $0 \le U < -2(\omega_0 - \mu)$ and 0 for $U = -2(\omega_0 - \mu)$ and positive for

$U > -2(\omega_0 - \mu)$. If $\omega_0 - \mu > 0$, $n'_U(B_S) - n'_U(-B_S)$ is always positive.

We focus on a cyclic pump of an area $\Delta_L^- \leq \Delta \leq \Delta_L^+$, $B_S^- \leq B_S \leq B_S^+$. The pumped charge and spin are given by

$$\begin{aligned}
&\langle \Delta N_{L\uparrow} \rangle \pm \langle \Delta N_{L\downarrow} \rangle \\
&= \int_{\Delta_L^-}^{\Delta_L^+} d\Delta_L \int_{B_S^-}^{B_S^+} dB_S \, (F^{L\uparrow}_{\Delta_L, B_S} \pm F^{L\downarrow}_{\Delta_L, B_S}) \\
&= -\int_{\Delta_L^-}^{\Delta_L^+} d\Delta_L \, \frac{\gamma_L \gamma_R \Delta_R}{(\gamma_L \Delta_L + \gamma_R \Delta_R)^2} \int_{B_S^-}^{B_S^+} dB_S \, g_S [n'_U(B_S) \mp n'_U(-B_S)] \\
&= -\gamma_R \Delta_R \Big[\frac{1}{\gamma_L \Delta_L^- + \gamma_R \Delta_R} - \frac{1}{\gamma_L \Delta_L^+ + \gamma_R \Delta_R} \Big] \\
&\quad \times [n_U(B_S^+) - n_U(B_S^-) \pm \{ n_U(-B_S^+) - n_U(-B_S^-) \}].
\end{aligned} \tag{4.98}$$

In particular, if $\gamma_L \Delta_L^- \ll \gamma_R \Delta_R$, $\gamma_L \Delta_L^+ \gg \gamma_R \Delta_R$,

$$\langle \Delta N_{L\uparrow} \rangle \pm \langle \Delta N_{L\downarrow} \rangle = -[n_U(B_S^+) - n_U(B_S^-) \mp \{ n_U(-B_S^-) - n_U(-B_S^+) \}], \tag{4.99}$$

holds. For instance, if the g-factor of the system is negative and $B_S^\pm = \pm\infty$,

$$\langle \Delta N_{L\uparrow} \rangle + \langle \Delta N_{L\downarrow} \rangle = 0, \tag{4.100}$$

$$\langle \Delta N_{L\uparrow} \rangle - \langle \Delta N_{L\downarrow} \rangle = -2, \tag{4.101}$$

hold.

Chapter 5

Quantum diabatic pump

5.1 Spinless one level quantum dot

In this section, we consider spinless one level QD coupled to two leads ($b = L, R$). $|0\rangle$ ($|1\rangle$) denotes the state that the QD is empty (occupied). The diagonal components $p_n = \langle n|\rho|n\rangle$ ($n = 0, 1$) of the system state ρ are governed by the master equation:

$$\frac{d}{dt}\begin{pmatrix} p_0(t) \\ p_1(t) \end{pmatrix} = K(\alpha_t)\begin{pmatrix} p_0(t) \\ p_1(t) \end{pmatrix}. \tag{5.1}$$

The Liouvillian is given by

$$K = \sum_b \Gamma_b \begin{pmatrix} -f_b & 1-f_b \\ f_b & -(1-f_b) \end{pmatrix}. \tag{5.2}$$

Here, Γ_b is the line-width function of the lead b, $f_b = [e^{\beta_b(\varepsilon-\mu_b)}+1]^{-1}$ is the Fermi distribution function, β_b and μ_b are inverse temperature and chemical potential of the lead b, ε is the energy level of the QD. The right eigenvectors of the Liouvillian are the instantaneous steady state

$$p^{\rm ss}(\alpha) = \begin{pmatrix} p_0^{\rm ss}(\alpha) \\ p_1^{\rm ss}(\alpha) \end{pmatrix} = \begin{pmatrix} 1-F(\alpha) \\ F(\alpha) \end{pmatrix}, \tag{5.3}$$

and

$$\begin{pmatrix} -1 \\ 1 \end{pmatrix}, \tag{5.4}$$

with the eigenvalue $(-\Gamma)$. Here,

$$F(\alpha) \stackrel{\rm def}{=} \frac{\sum_b \Gamma_b f_b}{\Gamma},\ \Gamma \stackrel{\rm def}{=} \sum_b \Gamma_b. \tag{5.5}$$

As a specialty of this model, (5.4) is time-independent. We introduce $p_n^{(m)} = \langle n|\rho^{(m)}|n\rangle$ ($m = 1, 2, \cdots$) and $\tilde{p}_n^{(m)} = \langle n|\tilde{\rho}^{(m)}|n\rangle$ ($m = 0, 1, \cdots$). $\tilde{p}_n^{(0)}$ are given by

$$\begin{pmatrix} \tilde{p}_0^{(0)}(\alpha_t) \\ \tilde{p}_1^{(1)}(\alpha_t) \end{pmatrix} = e^{-\int_0^t ds\ \Gamma(s)}[p(0) - p^{\rm ss}(\alpha_0)] = e^{-\int_0^t ds\ \Gamma(s)}\begin{pmatrix} -p_1(0) + F(\alpha_0) \\ p_1(0) - F(\alpha_0) \end{pmatrix}. \tag{5.6}$$

We suppose $p(0) = {}^t(p_0(0), p_1(0)) = p^{\rm ss}(\alpha_0)$. Then, $\tilde{p}_n^{(0)} = 0$ holds. We choose the $\tilde{K}^{-1}(\alpha)$ of (3.86) as the pseudo-inverse of $K(\alpha)$. We have

$$\tilde{K}^{-1} = \frac{1}{\Gamma}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \tag{5.7}$$

From $\rho^{(n)}(t) = [\mathcal{R}(\alpha_t)\frac{d}{dt}]^n \rho_0(\alpha_t)$, we obtain $p^{(n)} = {}^t(p_0^{(n)}, p_1^{(n)})$ as

$$\begin{aligned} p^{(1)}(\alpha_t) &= \tilde{K}^{-1}(\alpha_t)\frac{d}{dt}p^{\rm ss}(\alpha_t) \\ &= \frac{1}{\Gamma(t)}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}\frac{d}{dt}\begin{pmatrix} 1-F(\alpha_t) \\ F(\alpha_t) \end{pmatrix} \\ &= -\frac{1}{\Gamma(t)}\frac{d}{dt}\begin{pmatrix} -F(\alpha_t) \\ F(\alpha_t) \end{pmatrix}, \end{aligned} \tag{5.8}$$

and

$$\begin{aligned} p^{(n+1)}(t) &= \tilde{K}^{-1}(\alpha_t)\frac{d}{dt}p^{(n)}(t) \\ &= -\frac{1}{\Gamma(t)}\frac{d}{dt}\begin{pmatrix} -p_1^{(n)}(t) \\ p_1^{(n)}(t) \end{pmatrix}. \end{aligned} \tag{5.9}$$

$\tilde{p}^{(n)} = {}^t(-\tilde{p}_1^{(n)}, \tilde{p}_1^{(n)})$ $(n = 1, 2, \cdots)$ is given by

$$\tilde{p}_1^{(n)}(t) = -e^{-\int_0^t ds\, \Gamma(s)} p_1^{(n)}(0). \tag{5.10}$$

For by only modulating Γ_b at zero-bias, the pump dose not occur for all orders ($p^{(n)}(t) = 0$) because $p^{\rm ss}(\alpha)$ dose not change.

We consider the particle current to the lead b. From discussion of § 4.2.3, we obtain

$$\langle\langle l_0^{N_b}| = (0, \frac{\Gamma_b}{\Gamma}). \tag{5.11}$$

Then, we get

$$i_{N_b}^{(1)} = -\langle\langle l_0^b|\frac{d}{dt}p^{\rm ss}(\alpha_t) = -\frac{\Gamma_b(t)}{\Gamma(t)}\frac{d}{dt}F(t), \tag{5.12}$$

and

$$i_{N_b}^{(n+1)} = -\frac{\Gamma_b(t)}{\Gamma(t)}\frac{d}{dt}p_1^{(n)}(t). \tag{5.13}$$

$\tilde{i}^{(n)}_{N_b}$ is given by

$$
\begin{aligned}
\tilde{i}^{(n)}_{N_b} &= -\frac{\Gamma_b(t)}{\Gamma(t)}\frac{d}{dt}\tilde{p}^{(n)}_1(t) \\
&= -\frac{\Gamma_b(t)}{\Gamma(t)}[K(\alpha_t)\tilde{p}^{(n)}(t)]_1 \\
&= -\frac{\Gamma_b(t)}{\Gamma(t)}\Big[\Gamma(t)\begin{pmatrix} -F(\alpha_t) & 1-F(\alpha_t) \\ F(\alpha_t) & -1+F(\alpha_t)\end{pmatrix}\begin{pmatrix} -\tilde{p}^{(n)}_1(t) \\ \tilde{p}^{(n)}_1(t)\end{pmatrix}\Big]_1 \\
&= -\frac{\Gamma_b(t)}{\Gamma(t)}\Big[-\Gamma(t)\begin{pmatrix} -\tilde{p}^{(n)}_1(t) \\ \tilde{p}^{(n)}_1(t)\end{pmatrix}\Big]_1 \\
&= \Gamma_b(t)\tilde{p}^{(n)}_1(\alpha_t) \\
&= -\Gamma_b(t)e^{-\int_0^t ds\,\Gamma(s)}p^{(n)}_1(0).
\end{aligned}
\tag{5.14}
$$

5.2 Numerical calculation

We set the time-dependence of the control parameters as

$$\Gamma(t) = \Gamma_L(t) + \Gamma_R,\ \Gamma_L(t) = \gamma[1 + g\sin\omega(t+\delta))],\ \Gamma_R = \gamma, \tag{5.15}$$

$$f_L(t) = f_R(t) = f(t) = \frac{1}{e^{\beta(\varepsilon(t)-\mu)}+1},\ \varepsilon(t) - \mu = \varepsilon_0 \sin\omega t. \tag{5.16}$$

For the numerical calculation, we set

$$g = 0.5,\ \omega = 0.3\gamma,\ \beta\varepsilon_0 = 1,\ \delta = 0, \frac{\pi}{2}. \tag{5.17}$$

Γ of (3.17) is given by $\gamma(2-g) = 1.5\gamma$. Then,

$$\frac{\omega}{\Gamma} = \frac{0.3}{1.5} = 0.2, \tag{5.18}$$

holds.

For $\delta = \pi/2$, the pumped particle numbers of the first one cyclic are given by

$$
\begin{aligned}
\langle \Delta N_L \rangle &= 7.69464 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{\mathrm{BSN}} + \sum_{n=1}^{5} [\widetilde{\langle \Delta N_L \rangle}^{(n)} + \langle \Delta N_L \rangle^{(n+1)}] &= 7.69583 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{\mathrm{BSN}} &= 9.71762 \times 10^{-2}, \\
\widetilde{\langle \Delta N_L \rangle}^{(1)} &= -1.79649 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{(2)} &= 0, \\
\widetilde{\langle \Delta N_L \rangle}^{(2)} &= 0, \\
\langle \Delta N_L \rangle^{(3)} &= -0.270724 \times 10^{-2}, \\
\widetilde{\langle \Delta N_L \rangle}^{(3)} &= 0.0336304 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{(4)} &= 0, \\
\widetilde{\langle \Delta N_L \rangle}^{(4)} &= 0, \\
\langle \Delta N_L \rangle^{(5)} &= 0.0133644 \times 10^{-2}, \\
\widetilde{\langle \Delta N_L \rangle}^{(5)} &= -0.00156459 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{(6)} &= 0.
\end{aligned}
\tag{5.19}
$$

Figure 5.1(b) shows that $p_1(t)$ and $f(t)$, Fig.5.1(a) shows that $\delta p_1(t) \stackrel{\mathrm{def}}{=} p_1(t) - f(t)$, $p_1^{(1)}(t)$ and $p_1^{(1)}(t) + p_1^{(2)}(t)$, and Fig.5.1(c) shows that $\delta p_1 - p_1^{(1)} - p_1^{(2)}$ and $p_1^{(3)}(t)$.

For $\delta = 0$, the pumped particle numbers of the first one cyclic are given by

$$
\begin{aligned}
\langle \Delta N_L \rangle &= -0.466997 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{\mathrm{BSN}} + \sum_{n=1}^{5} [\widetilde{\langle \Delta N_L \rangle}^{(n)} + \langle \Delta N_L \rangle^{(n+1)}] &= -0.464558 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{\mathrm{BSN}} &= 0, \\
\widetilde{\langle \Delta N_L \rangle}^{(1)} &= -1.9376 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{(2)} &= 1.52006 \times 10^{-2}, \\
\widetilde{\langle \Delta N_L \rangle}^{(2)} &= -0.0726599 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{(3)} &= 0, \\
\widetilde{\langle \Delta N_L \rangle}^{(3)} &= 0.0572197 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{(4)} &= -0.0462914 \times 10^{-2}, \\
\widetilde{\langle \Delta N_L \rangle}^{(4)} &= 0.0148158 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{(5)} &= 0, \\
\widetilde{\langle \Delta N_L \rangle}^{(5)} &= -0.00221088 \times 10^{-2}, \\
\langle \Delta N_L \rangle^{(6)} &= 0.00210926 \times 10^{-2}.
\end{aligned}
\tag{5.20}
$$

Figure 5.2(b) shows that $p_1(t)$ and $f(t)$, Fig.5.2(a) shows that $\delta p_1(t) = p_1(t) - f(t)$, $p_1^{(1)}(t)$ and $p_1^{(1)}(t) + p_1^{(2)}(t)$, and Fig.5.2(c) shows that $\delta p_1 - p_1^{(1)} - p_1^{(2)}$ and $p_1^{(3)}(t)$.

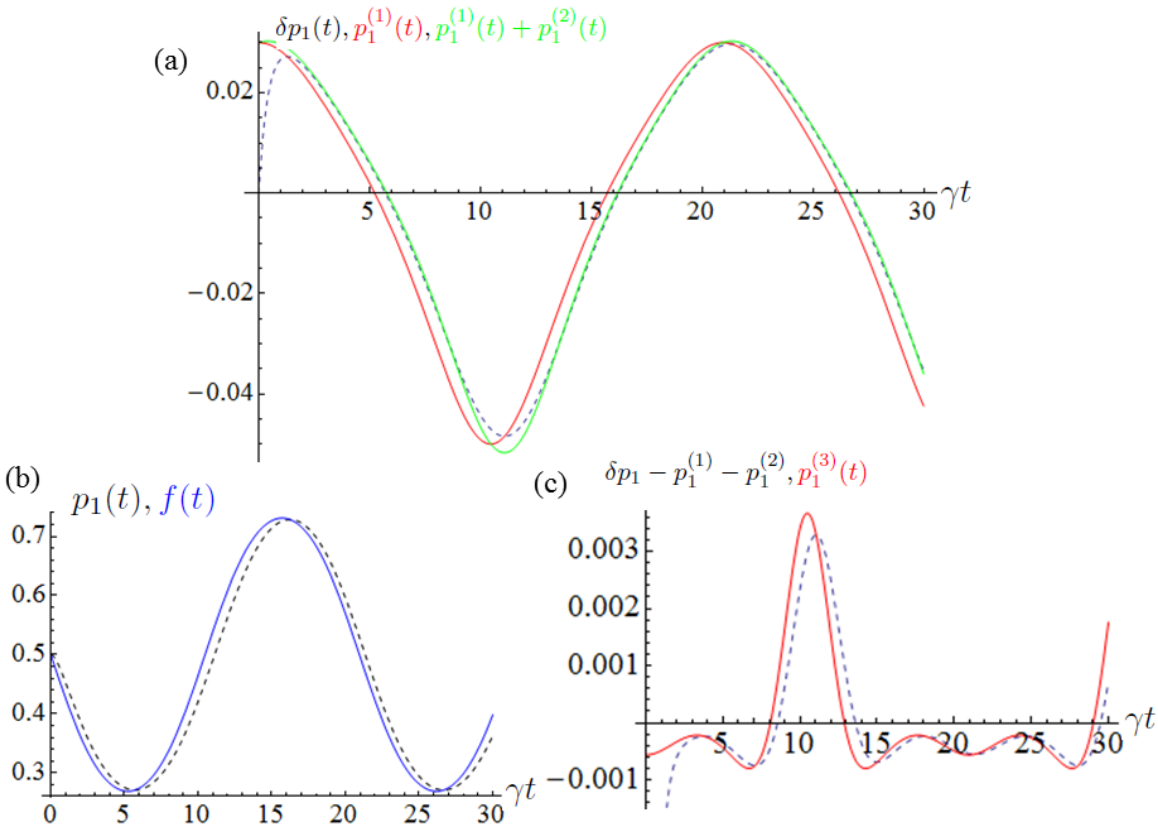

FIGURE 5.1: (a)$\delta p_1(t) \stackrel{\text{def}}{=} p_1(t) - f(t)$(dashed line), $p_1^{(1)}(t)$(red line) and $p_1^{(1)}(t) + p_1^{(2)}(t)$, (b)$p_1(t)$(dashed line) and $f(t)$, (c)$\delta p_1 - p_1^{(1)} - p_1^{(2)}$(dashed line) and $p_1^{(3)}(t)$ for $\delta = \pi/2$.

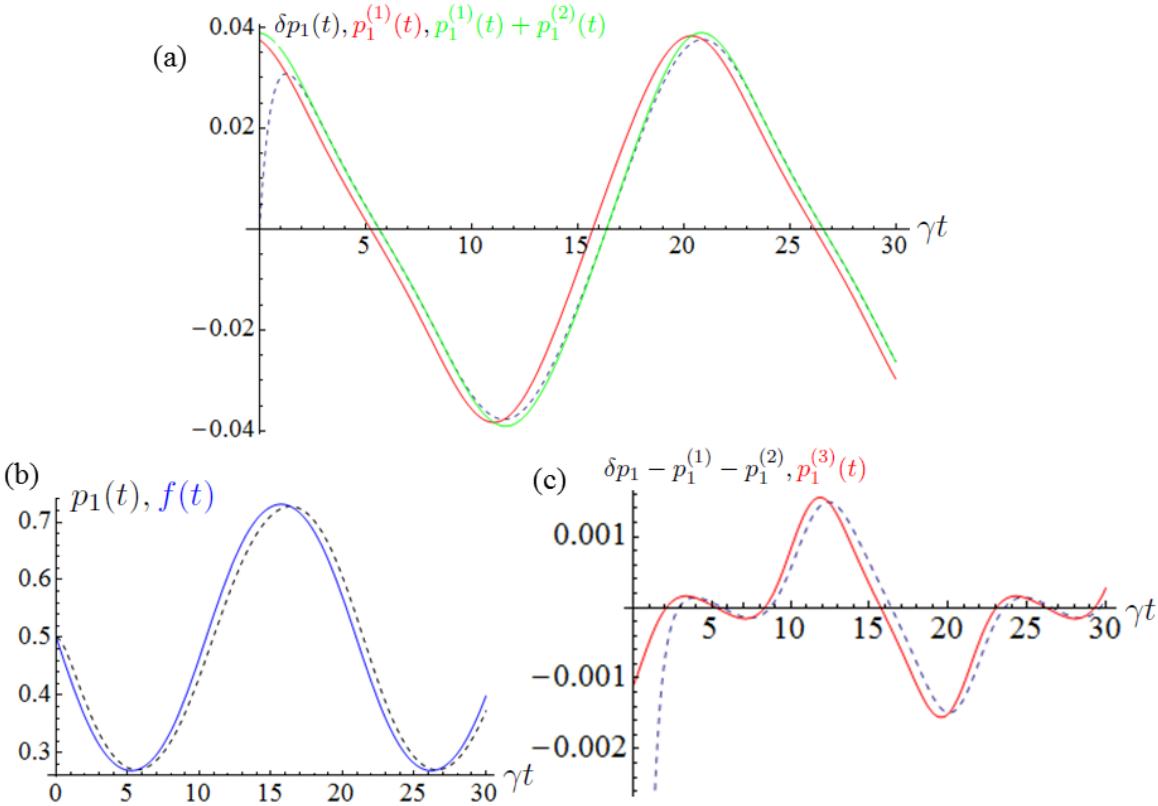

FIGURE 5.2: (a)$\delta p_1(t) = p_1(t) - f(t)$(dashed line), $p_1^{(1)}(t)$(red line) and $p_1^{(1)}(t) + p_1^{(2)}(t)$, (b)$p_1(t)$(dashed line) and $f(t)$, (c)$\delta p_1 - p_1^{(1)} - p_1^{(2)}$(dashed line) and $p_1^{(3)}(t)$ for $\delta = 0$.

Chapter 6

Generalized quantum master equation for entropy production

6.1 Definition of entropy production

In this chapter and Chap.7 and Chap.8, we suppose that $\{H_b\}_b$ are time-independent.

It is natural to identify the average entropy production rate with

$$\dot{\sigma}(t) \stackrel{\text{def}}{=} \sum_{b\in\mathcal{C}} \beta_b(t)[-i_{H_b}(t)] + \sum_{b\in\mathcal{G}} \beta_b(t)[-i_{H_b}(t) - \mu_b(t)\{-i_{N_b}(t)\}]. \tag{6.1}$$

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

$$W^\sigma(\alpha) \stackrel{\text{def}}{=} \sum_{b\in\mathcal{C}} \beta_b[-W^{H_b}(\alpha)] + \sum_{b\in\mathcal{G}} \beta_b[-W^{H_b}(\alpha) - \mu_b\{-W^{N_b}(\alpha)\}]. \tag{6.2}$$

The average entropy production is given by

$$\begin{aligned} \sigma &\stackrel{\text{def}}{=} \int_0^\tau dt\, \dot{\sigma}(t) \\ &= \int_0^\tau dt\, J_\sigma^{\text{ss}}(\alpha_t) + \int_C d\alpha^n\, A_n^\sigma(\alpha) + \mathcal{O}(\frac{\omega}{\Gamma}), \end{aligned} \tag{6.3}$$

where

$$J_\sigma^{\text{ss}}(\alpha) \stackrel{\text{def}}{=} \sum_{b\in\mathcal{C}} \beta_b[-i_{H_b}^{\text{ss}}(\alpha)] + \sum_{b\in\mathcal{G}} \beta_b[-i_{H_b}^{\text{ss}}(\alpha) - \mu_b\{-i_{N_b}^{\text{ss}}(\alpha)\}], \tag{6.4}$$

and

$$A_n^\sigma(\alpha) \stackrel{\text{def}}{=} \sum_{b\in\mathcal{C}} \beta_b[-A_n^{H_b}(\alpha)] + \sum_{b\in\mathcal{G}} \beta_b[-A_n^{H_b}(\alpha) - \mu_b\{-A_n^{N_b}(\alpha)\}]. \tag{6.5}$$

Here, we used (3.18) for $\{O_\mu\} = \{H_b\}_b + \{N_b\}_{b\in\mathcal{G}}$. The excess entropy production is defined by

$$\sigma_{\text{ex}} \stackrel{\text{def}}{=} \sigma - \int_0^\tau dt\, J_\sigma^{\text{ss}}(\alpha_t) = \int_C d\alpha^n\, A_n^\sigma(\alpha) + \mathcal{O}(\frac{\omega}{\Gamma}). \tag{6.6}$$

While we can calculate the average of the entropy production, our formalism is not compatible to discuss the higher moments of the entropy production. Although (3.19) is the average of the difference between outcomes at $t = \tau$ and $t = 0$ of O_μ, σ is not that of some bath's operator if α'' are modulated.

6.2 Introduction of generalized QME

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

$$\frac{d}{dt}\rho^\lambda(t) = \mathcal{K}^\lambda(\alpha_t)\rho^\lambda(t), \tag{6.7}$$

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

$$\mathcal{K}^\lambda(\alpha)\bullet = -i[H_S(\alpha_S), \bullet] + \sum_b \mathcal{L}_b^\lambda(\alpha)\bullet, \tag{6.8}$$

with

$$\mathcal{L}_b^\lambda(\alpha)\bullet = \sum_a c_{ba}^\lambda(\alpha) A_a \bullet B_a, \tag{6.9}$$

and

$$c_{ba}^\lambda(\alpha)\big|_{\lambda=0} = c_{ba}. \tag{6.10}$$

While $c_{ba}^\chi(\alpha)$ of (3.1) depend on χ if and only if $A_a, B_a \neq 1$, $c_{ba}^\lambda(\alpha)$ can depend on λ for all a. We suppose that the solution of (6.7) satisfies

$$\mathrm{Tr}_S[\rho'(\tau)] = \sigma, \tag{6.11}$$

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

$$\langle\langle 1|\mathcal{K}'(\alpha) = \langle\langle 1|W^\sigma(\alpha). \tag{6.12}$$

Let's consider

$$\langle\langle l_0^\lambda(\alpha)|\mathcal{K}^\lambda(\alpha) = \lambda_0^\lambda(\alpha)\langle\langle l_0^\lambda(\alpha)|, \tag{6.13}$$

corresponding to (3.4) for $n = 0$. Similar to (3.16) and (3.20),

$$\lambda_0'(\alpha) \;=\; \langle\langle 1|W^\sigma(\alpha)|\rho_0(\alpha)\rangle\rangle = J_\sigma^{\mathrm{ss}}(\alpha), \tag{6.14}$$

and

$$A_n^\sigma(\alpha) \;=\; -\langle\langle l_0'(\alpha)|\frac{\partial}{\partial\alpha^n}|\rho_0(\alpha)\rangle\rangle = \langle\langle 1|W^\sigma(\alpha)\mathcal{R}(\alpha)\frac{\partial}{\partial\alpha^n}|\rho_0(\alpha)\rangle\rangle, \tag{6.15}$$

hold. Although $\lambda_0^\lambda(\alpha)$ and $l_0^\lambda(\alpha)$ depend on the choice of $\mathcal{K}^\lambda(\alpha)$, $\lambda_0'(\alpha)$ and $A_n^\sigma(\alpha)$ do not depend, as can be seen in the RHS of the (6.14) and (6.15). The LHS of (6.12) is given by

$$\langle\langle 1|\mathcal{K}'(\alpha) = \langle\langle 1|\sum_{b,a} c_{ba}'(\alpha) B_a A_a. \tag{6.16}$$

Using this and (3.15), (6.12) becomes

$$\sum_{b,a} c_{ba}'(\alpha) B_a A_a = \sum_a [-\sum_b \beta_b c_{ba}^{H_b}(\alpha) + \sum_{b\in\mathcal{G}} \beta_b \mu_b c_{ba}^{N_b}(\alpha)] B_a A_a. \tag{6.17}$$

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

"Higher moments" $\frac{\partial^n}{\partial(i\lambda)^n}\mathrm{Tr}_S[\rho^\lambda(\tau)]\big|_{\lambda=0}$ $(n = 2, 3, \cdots)$ depend on the choice of $\mathcal{K}^\lambda(\alpha)$ and seems have no physical meening. In contrast, the higher moments of the entropy production could be considered for the classical Markov jump process. In Appendix J, we review the entropy production of the Markov jump process [21, 37], and in Chap.8, we compare that and (6.3).

6.3 Current operators

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

$$w^{X_b}(\alpha) \stackrel{\text{def}}{=} -[\mathcal{L}_b^\dagger(\alpha)X_S]^\dagger = -\mathcal{L}_b^\dagger(\alpha)X_S \ (X = N, H). \tag{6.18}$$

For a super-operator $\mathcal{J}$, $\mathcal{J}^\dagger$ is defined by $\langle\langle \mathcal{J}^\dagger X|Y\rangle\rangle = \langle\langle X|\mathcal{J}Y\rangle\rangle$ $(X, Y \in \mathrm{B})$.

$$\mathcal{L}_b^\dagger(\alpha)\bullet = \sum_a c_{ba}^*(\alpha)A_a^\dagger \bullet B_a^\dagger, \tag{6.19}$$

holds. $w^{X_b}(\alpha)$ is a Hermitian operator and is given by

$$w^{X_b}(\alpha) = -\sum_a c_{ba}(\alpha)B_aX_SA_a \ (X = N, H). \tag{6.20}$$

In general, for the RWA,

$$w^{H_b} = W^{H_b}(\alpha) = \sum_\omega\sum_{\mu,\nu}\omega\Phi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega), \tag{6.21}$$

holds (Appendix G). For the Born-Markov approximation and the CGA, $w^{H_b}(\alpha) \neq W^{H_b}(\alpha)$. From (2.163), (2.166) and (2.167),

$$w^{N_b}(\alpha) = 0 \ (b \in \boldsymbol{\mathcal{C}}), \tag{6.22}$$

holds for the RWA, the Born-Markov approximation, and the CGA. In the following, we set

$$W^{N_b}(\alpha) \stackrel{\text{def}}{=} w^{N_b}(\alpha) = 0 \ (b \in \boldsymbol{\mathcal{C}}), \tag{6.23}$$

and

$$i_{N_b}^{\text{ss}}(\alpha) \stackrel{\text{def}}{=} \mathrm{Tr}_S[w^{N_b}(\alpha)\rho_0(\alpha)] = 0 \ (b \in \boldsymbol{\mathcal{C}}). \tag{6.24}$$

Here, we suppose (2.106) for $b \in \boldsymbol{\mathcal{G}}$. The generalization to (2.137) case is straightforward. For $\{O_\mu\} = \{N_b\}_{b\in\boldsymbol{\mathcal{G}}} + \{H_b\}_b$, (2.126) holds in (2.148). For the Born-Markov approximation and the CGA, $w^{N_b}(\alpha) = W^{N_b}$, however, $w^{H_b}(\alpha) \neq W^{H_b}(\alpha)$. For the

RWA,

$$
\begin{aligned}
w^{N_b}(\alpha) &= W^{N_b}(\alpha) \\
&= \sum_{\omega} \sum_{\alpha,\beta} \left\{ \Phi^{-}_{b,\alpha\beta}(\omega)[a_\alpha(\omega)]^\dagger a_\beta(\omega) - \Phi^{+}_{b,\alpha\beta}(\omega) a_\alpha(\omega)[a_\beta(\omega)]^\dagger \right\} \ (b \in \boldsymbol{\mathcal{G}}), \qquad (6.25) \\
w^{H_b}(\alpha) &= W^{H_b}(\alpha) \\
&= \sum_{\omega} \sum_{\alpha,\beta} \left\{ \omega\Phi^{-}_{b,\alpha\beta}(\omega)[a_\alpha(\omega)]^\dagger a_\beta(\omega) - \omega\Phi^{+}_{b,\alpha\beta}(\omega) a_\alpha(\omega)[a_\beta(\omega)]^\dagger \right\} \ (b \in \boldsymbol{\mathcal{G}}), \qquad (6.26)
\end{aligned}
$$

hold. Therefore, (6.2) and (6.18) imply that $W^\sigma(\alpha)$ is given by

$$
W^\sigma(\alpha) = \sum_b \mathcal{L}^\dagger_b(\alpha)(\beta_b H_S - \beta_b \nu_b N_S) = \sum_b \Pi^\dagger_b(\alpha)(\beta_b H_S - \beta_b \nu_b N_S). \qquad (6.27)
$$

Here,

$$
\nu_b \stackrel{\text{def}}{=} \begin{cases} \mu'_b & b \in \boldsymbol{\mathcal{C}} \\ \mu_b & b \in \boldsymbol{\mathcal{G}} \end{cases} . \qquad (6.28)
$$

μ'_b is an arbitrary real number.

Chapter 7

Geometrical expression of excess entropy production

In this chapter and Chap.8, we focus on the RWA. We use

$$W^{H_b} = w^{H_b} \ (b \in \boldsymbol{\mathcal{C}}), \tag{7.1}$$
$$W^{H_b} = w^{H_b}, \ W^{N_b} = w^{N_b} \ (b \in \boldsymbol{\mathcal{G}}), \tag{7.2}$$

and

$$\Pi_b(\bullet e^{-\beta_b(H_S-\nu_b N_S)}) = (\Pi_b^\dagger \bullet) e^{-\beta_b(H_S-\nu_b N_S)}, \tag{7.3}$$

and

$$[h_b(\alpha), N_S] = 0. \tag{7.4}$$

If we suppose (2.163) for $b \in \boldsymbol{\mathcal{C}}$ and (2.106) or (2.137) for $b \in \boldsymbol{\mathcal{G}}$, these relations hold. If $n_{\mathrm{GC}} = 0$, existence of N_S, (2.163), (7.2) and (7.4) are not required and the system S does not have to be described by the annihilation and creation operators (S can be spin chain or few level system, etc.). Using $\mathcal{L}_b^\dagger 1 = \Pi_b^\dagger 1 = 0$ (see (2.105)) for (7.3) with $\bullet = 1$, we obtain

$$\Pi_b e^{-\beta_b(H_S-\nu_b N_S)} = \mathcal{L}_b e^{-\beta_b(H_S-\nu_b N_S)} = 0. \tag{7.5}$$

Here, we used (2.144) and (7.4).

7.1 Equilibrium state

In this section, we consider equilibrium state $\beta_b = \beta$ (for all b) and $\mu_b = \mu$ ($b \in \boldsymbol{\mathcal{G}}$), and α denotes the set of $(\alpha_S, \{\alpha_{Sb}\}_b, \beta, \beta\mu)$. We show that $A_n^\sigma(\alpha)$ is a total derivative of the von Neumann entropy of the instantaneous steady state. Differentiating (6.13) by λ, we obtain

$$\langle\langle l_0'(\alpha)|\hat{K}(\alpha) + \langle\langle 1|\mathcal{K}'(\alpha) = \lambda_0'(\alpha)\langle\langle 1|. \tag{7.6}$$

In the RHS, $\lambda_0'(\alpha) = J_\sigma^{\mathrm{ss}}(\alpha) = 0$ holds. The second term of the LHS is $\langle\langle 1|W^\sigma(\alpha)$. (6.27) leads

$$W^\sigma(\alpha) = \beta \sum_b \mathcal{L}_b^\dagger(\alpha)[H_S - \mu N_S] = \beta \hat{K}^\dagger(\alpha)[H_S - \mu N_S], \tag{7.7}$$

i.e.,

$$\langle\langle\beta[H_S-\mu N_S]|\hat{K}(\alpha)=\langle\langle 1|W^{\sigma}(\alpha). \tag{7.8}$$

Then, (7.6) leads

$$\left[\langle\langle l_0'(\alpha)|+\langle\langle\beta[H_S-\mu N_S]|\right]\hat{K}(\alpha)=0. \tag{7.9}$$

This implies

$$\langle\langle l_0'(\alpha)|=-\langle\langle\beta[H_S-\mu N_S]|+c(\alpha)\langle\langle 1|, \tag{7.10}$$

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

$$\rho_0(\alpha)=\rho_{\rm gc}(\alpha_S;\beta,\beta\mu)\stackrel{\rm def}{=}\frac{e^{-\beta(H_S(\alpha_S)-\mu N_S)}}{\Xi(\alpha_S;\beta,\beta\mu)}, \tag{7.11}$$

with $\Xi(\alpha_S;\beta,\beta\mu)\stackrel{\rm def}{=}{\rm Tr}_S[e^{-\beta(H_S(\alpha_S)-\mu N_S)}]$. This is derived from (7.5) (Cf.(A.11)). Then,

$$\{l_0'(\alpha)\}^{\dagger}=\ln\rho_{\rm gc}(\alpha_S;\beta,\beta\mu)+c'(\alpha)1,\ c'(\alpha)=c(\alpha)+\ln\Xi(\alpha_S;\beta,\beta\mu), \tag{7.12}$$

holds. Substituting this equation into (6.15), we obtain

$$A_n^{\sigma}(\alpha)=\frac{\partial}{\partial\alpha^n}S_{\rm vN}(\rho_{\rm gc}(\alpha_S;\beta,\beta\mu)), \tag{7.13}$$

using (H.1).

7.2 Weakly nonequilibrium regime

We introduce

$$\varepsilon_{1,b}\stackrel{\rm def}{=}\beta_b-\overline{\beta},\ \varepsilon_{2,b}\stackrel{\rm def}{=}\begin{cases}0 & b\in\boldsymbol{\mathcal{C}}\\ \beta_b\mu_b-\overline{\beta\mu} & b\in\boldsymbol{\mathcal{G}}\end{cases},\ \varepsilon\stackrel{\rm def}{=}\max_b\left\{\frac{|\varepsilon_{1,b}|}{\overline{\beta}},\frac{|\varepsilon_{2,b}|}{|\overline{\beta\mu}|}\right\}, \tag{7.14}$$

where $\overline{\beta}$ and $\overline{\beta\mu}$ are the reference values, which satisfy

$$\min_b\beta_b\le\overline{\beta}\le\max_b\beta_b, \tag{7.15}$$

$$\min_{b\in\boldsymbol{\mathcal{G}}}\beta_b\mu_b\le\overline{\beta\mu}\le\max_{b\in\boldsymbol{\mathcal{G}}}\beta_b\mu_b. \tag{7.16}$$

ε is a measure of degree of nonequilibrium. We consider $\varepsilon\ll 1$ regime. Now, we introduce

$$\hat{K}_{\kappa}(\alpha)\bullet\stackrel{\rm def}{=}-i[H_S(\alpha_S)+\kappa H_{\rm L}(\alpha),\bullet]+\sum_b\Pi_b(\alpha)\bullet, \tag{7.17}$$

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

$$\hat{K}_{\kappa}(\alpha)\rho_0^{(\kappa)}(\alpha)=0. \tag{7.18}$$

Here, κ is a real parameter satisfying $-1 \leq \kappa \leq 1$. $\langle\langle 1|\hat{K}_\kappa(\alpha) = 0$ holds. In the following, we show

$$A_n^\sigma(\alpha) = -\mathrm{Tr}_S\Big[\ln \rho_0^{(-1)}(\alpha)\frac{\partial \rho_0(\alpha)}{\partial \alpha^n}\Big] + \mathcal{O}(\varepsilon^2). \tag{7.19}$$

We use the following notations:

$$\alpha_{1,b} \stackrel{\mathrm{def}}{=} \beta_b,\ \alpha_{2,b} \stackrel{\mathrm{def}}{=} \beta_b\nu_b,\ \overline{X} \stackrel{\mathrm{def}}{=} X\big|_{\alpha_{i,b}=\overline{\alpha_i}}. \tag{7.20}$$

Here, $\overline{\alpha_1} = \overline{\beta}$ and $\overline{\alpha_2} = \overline{\beta\mu}$.

We expand $\rho_0^{(\kappa)}$ and l_0' as

$$\rho_0^{(\kappa)}(\alpha) = \overline{\rho_0^{(\kappa)}} + \sum_b(\varepsilon_{1,b}\rho_{1,b}^{(\kappa)} + \varepsilon_{2,b}\rho_{2,b}^{(\kappa)}) + \mathcal{O}(\varepsilon^2), \tag{7.21}$$

$$l_0'(\alpha) = \overline{l_0'(\alpha)} + \sum_b(\varepsilon_{1,b}k_{1,b} + \varepsilon_{2,b}k_{2,b}) + \mathcal{O}(\varepsilon^2), \tag{7.22}$$

with

$$\overline{\rho_0^{(\kappa)}} = \rho_{\mathrm{gc}},\ \overline{l_0'(\alpha)} = -\overline{\beta}H_S + \overline{\beta\mu}N_S + \overline{c}^*1 = \ln\rho_{\mathrm{gc}} + \overline{c'}^*1. \tag{7.23}$$

Here, $\rho_{\mathrm{gc}} \stackrel{\mathrm{def}}{=} \rho_{\mathrm{gc}}(\alpha_S;\overline{\beta},\overline{\beta\mu})$, $\overline{c}$ and $\overline{c'}$ are the same with $c(\alpha)$ and $c'(\alpha)$ in § 7.1.

First, we investigate $k_{i,b}$ in (7.22). (7.6) can be rewritten as

$$\hat{K}^\dagger(\alpha)l_0'(\alpha) + [\mathcal{K}'(\alpha)]^\dagger 1 = J_\sigma^{\mathrm{ss}}(\alpha). \tag{7.24}$$

Here,

$$J_\sigma^{\mathrm{ss}}(\alpha) = \mathcal{O}(\varepsilon^2), \tag{7.25}$$

holds because $i_{H_b}^{\mathrm{ss}}(\alpha), i_{N_b}^{\mathrm{ss}}(\alpha) = \mathcal{O}(\varepsilon)$ and

$$J_\sigma^{\mathrm{ss}}(\alpha) = \sum_b(-i_{H_b}^{\mathrm{ss}}(\alpha)\varepsilon_{1,b} + i_{N_b}^{\mathrm{ss}}(\alpha)\varepsilon_{2,b}), \tag{7.26}$$

since

$$\sum_b i_{X_b}^{\mathrm{ss}}(\alpha) = -\mathrm{Tr}_S[X_S\sum_b\mathcal{L}_b(\alpha)\rho_0(\alpha)] = 0 \quad (X = N, H). \tag{7.27}$$

Then we obtain

$$\overline{\partial_{i,b}\mathcal{K}'}^\dagger 1 + \overline{K}^\dagger k_{i,b} + \overline{\partial_{i,b}\mathcal{L}_b}^\dagger\overline{l_0'} = 0, \tag{7.28}$$

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

$$\begin{aligned}\overline{\partial_{i,b}\mathcal{K}'}^\dagger 1 &= \frac{\partial[\mathcal{K}']^\dagger 1}{\partial\alpha_{i,b}}\Big|_{\alpha_{i,b}=\overline{\alpha_i}} \\ &= \frac{\partial\mathcal{L}_b^\dagger[\alpha_{1,b}H_S - \alpha_{2,b}N_S]}{\partial\alpha_{i,b}}\Big|_{\alpha_{i,b}=\overline{\alpha_i}} \\ &= \overline{\partial_{i,b}\mathcal{L}_b}^\dagger[\overline{\beta}H_S - \overline{\beta\mu}N_S] + \overline{\Pi_b}^\dagger\frac{\partial[\alpha_{1,b}H_S - \alpha_{2,b}N_S]}{\partial\alpha_{i,b}}.\end{aligned} \tag{7.29}$$

The third term of the LHS becomes

$$\begin{aligned}\overline{\partial_{i,b}\mathcal{L}_b}^\dagger \overline{l'_0} &= \overline{\partial_{i,b}\mathcal{L}_b}^\dagger(-\overline{\beta}H_S + \overline{\beta\mu}N_S + c1) \\ &= -\overline{\partial_{i,b}\mathcal{L}_b}^\dagger(\overline{\beta}H_S - \overline{\beta\mu}N_S). \end{aligned} \tag{7.30}$$

Here, we used $\overline{\partial_{i,b}\mathcal{L}_b}^\dagger 1 = 0$ derived from $\hat{K}^\dagger 1 = 0$. Then, (7.28) becomes

$$\overline{K}^\dagger k_{1,b} + \overline{\Pi_b}^\dagger H_S = 0, \tag{7.31}$$

$$\overline{K}^\dagger k_{2,b} - \overline{\Pi_b}^\dagger N_S = 0. \tag{7.32}$$

Next, we show the relation between $k_{i,b}$ and $\rho_{i,b}^{(-1)}$. (7.18) leads

$$\overline{K_\kappa}\rho_{i,b}^{(\kappa)} + \overline{\partial_{i,b}\mathcal{L}_b}\rho_{\mathrm{gc}} = 0, \tag{7.33}$$

in $\mathcal{O}(\varepsilon_{i,b})$. Here, $\overline{K_\kappa} \stackrel{\mathrm{def}}{=} \overline{\hat{K}_\kappa}$. By the way, (7.5) is

$$\mathcal{L}_b\rho_{\mathrm{gc}}(\alpha_S;\beta_b,\beta_b\nu_b) = 0. \tag{7.34}$$

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

$$\overline{\partial_{i,b}\mathcal{L}_b}\rho_{\mathrm{gc}} = -\overline{\mathcal{L}_b}\frac{\overline{\rho_{\mathrm{gc}}(\alpha_S;\beta_b,\beta_b\nu_b)}}{\partial\alpha_{i,b}} = \overline{\mathcal{L}_b}\frac{\partial[\alpha_{1,b}H_S - \alpha_{2,b}N_S]}{\partial\alpha_{i,b}}\rho_{\mathrm{gc}}(\alpha_S;\overline{\beta},\overline{\beta\mu}). \tag{7.35}$$

Substituting these equations into (7.33), we obtain

$$\overline{K}_\kappa\rho_{1,b}^{(\kappa)} + \overline{\Pi_b}(H_S\rho_{\mathrm{gc}}) = 0, \tag{7.36}$$

$$\overline{K}_\kappa\rho_{2,b}^{(\kappa)} - \overline{\Pi_b}(N_S\rho_{\mathrm{gc}}) = 0. \tag{7.37}$$

Now, we use (7.3), namely,

$$\overline{\Pi_b}(\bullet\rho_{\mathrm{gc}}) = (\overline{\Pi_b}^\dagger\bullet)\rho_{\mathrm{gc}}. \tag{7.38}$$

Using this relation, we rewire (7.36) and (7.37) as

$$\overline{K}_\kappa\rho_{1,b}^{(\kappa)} + (\overline{\Pi_b}^\dagger H_S)\rho_{\mathrm{gc}} = 0, \tag{7.39}$$

$$\overline{K}_\kappa\rho_{2,b}^{(\kappa)} - (\overline{\Pi_b}^\dagger N_S)\rho_{\mathrm{gc}} = 0. \tag{7.40}$$

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

$$(\overline{K}_\kappa\rho_{1,b}^{(\kappa)})\rho_{\mathrm{gc}}^{-1} + \overline{\Pi_b}^\dagger H_S = 0, \tag{7.41}$$

$$(\overline{K}_\kappa\rho_{2,b}^{(\kappa)})\rho_{\mathrm{gc}}^{-1} - \overline{\Pi_b}^\dagger N_S = 0. \tag{7.42}$$

(7.38) can be rewritten as

$$(\overline{\Pi_b}Y)\rho_{\mathrm{gc}}^{-1} = \overline{\Pi_b}^\dagger(Y\rho_{\mathrm{gc}}^{-1}), \tag{7.43}$$

for any $Y = \bullet\rho_{\mathrm{gc}} \in \mathrm{B}$ by multiplying ρ_{gc}^{-1} from the right. (7.43) leads

$$(\overline{\Pi}\rho_{i,b}^{(\kappa)})\rho_{\mathrm{gc}}^{-1} = \overline{\Pi}^\dagger(\rho_{i,b}^{(\kappa)}\rho_{\mathrm{gc}}^{-1}), \tag{7.44}$$

where $\overline{\Pi} \stackrel{\text{def}}{=} \sum_b \overline{\Pi_b}$. By the way, $[H_S(\alpha_S), \rho_0^{(\kappa)}(\alpha)] = 0$ holds similarly to (2.147). Differentiating this equation by $\alpha_{i,b}$, we obtain

$$[H_S(\alpha_S), \rho_{i,b}^{(\kappa)}] = 0. \tag{7.45}$$

This relation leads

$$(\overline{H_\kappa^\times} \rho_{i,b}^{(\kappa)}) \rho_{\rm gc}^{-1} = \overline{H_\kappa^\times} (\rho_{i,b}^{(\kappa)} \rho_{\rm gc}^{-1}) = \overline{H_{-\kappa}^\times}^\dagger (\rho_{i,b}^{(\kappa)} \rho_{\rm gc}^{-1}), \tag{7.46}$$

where $H_\kappa^\times \bullet \stackrel{\text{def}}{=} -i[H_S(\alpha_S) + \kappa H_{\rm L}(\alpha), \bullet]$. We used $(H_\kappa^\times)^\dagger = -H_\kappa^\times$. In the first equality, we used that $\rho_{\rm gc}$ commutes with H_S and $H_{\rm L}$. (7.44) and (7.46) lead

$$(\overline{K}_\kappa \rho_{i,b}^{(\kappa)}) \rho_{\rm gc}^{-1} = \overline{K}_{-\kappa}^\dagger (\rho_{i,b}^{(\kappa)} \rho_{\rm gc}^{-1}). \tag{7.47}$$

Substituting this into (7.41) and (7.42), we obtain

$$\overline{K}_{-\kappa}^\dagger (\rho_{1,b}^{(\kappa)} \rho_{\rm gc}^{-1}) + \overline{\Pi_b}^\dagger H_S = 0, \tag{7.48}$$

$$\overline{K}_{-\kappa}^\dagger (\rho_{2,b}^{(\kappa)} \rho_{\rm gc}^{-1}) - \overline{\Pi_b}^\dagger N_S = 0. \tag{7.49}$$

Subtracting (7.48) ((7.49)) for $\kappa = -1$ from (7.31) ((7.32)), we obtain

$$\overline{K}^\dagger (k_{i,b} - \rho_{i,b}^{(-1)} \rho_{\rm gc}^{-1}) = 0. \tag{7.50}$$

This means

$$k_{i,b} = \rho_{i,b}^{(-1)} \rho_{\rm gc}^{-1} + \overline{c_{i,b}} 1, \tag{7.51}$$

where $\overline{c_{i,b}}$ is unknown complex number. Using this relation, (7.22) becomes

$$\begin{aligned} l_0'(\alpha) &= \ln \rho_{\rm gc}(\alpha_S; \overline{\beta}, \overline{\beta\mu}) + C(\alpha) 1 + \sum_b \sum_{i=1}^2 \varepsilon_{i,b} \rho_{i,b}^{(-1)} \rho_{\rm gc}^{-1} + \mathcal{O}(\varepsilon^2) \\ &= \ln \rho_0^{(-1)}(\alpha) + C(\alpha) 1 + \mathcal{O}(\varepsilon^2). \end{aligned} \tag{7.52}$$

Substituting this equation into (6.15), we obtain (7.19). Here, $C(\alpha) \stackrel{\text{def}}{=} \overline{c'}^* + \sum_{b,i} \overline{c_{i,b}} \varepsilon_{i,b}$. We supposed $[\rho_{\rm gc}, \rho_{i,b}^{(-1)}] = 0$, which leads $\ln \rho_0^{(-1)}(\alpha) = \ln \rho_{\rm gc} + \sum_{i,b} \varepsilon_{i,b} \rho_{i,b}^{(-1)} \rho_{\rm gc}^{-1} + \mathcal{O}(\varepsilon^2)$. This supposition is satisfied if $[N_S, \rho_0^{(-1)}(\alpha)] = \mathcal{O}(\varepsilon^2)$ (which leads $[N_S, \rho_{i,b}^{(-1)}] = 0$) or $\overline{\beta\mu} = 0$ holds. If H_S is non-degenerate, $[N_S, \rho_0^{(-1)}(\alpha)] = 0$ holds, then $[N_S, \rho_{i,b}^{(-1)}] = 0$, $[\rho_{\rm gc}, \rho_{i,b}^{(-1)}] = 0$ and (7.52) hold. If $n_{\rm GC} = 0$, $\rho_{\rm gc}$ is replaced by the canonical distribution and (7.52) holds without any assumption.

If

$$[H_{\rm L}(\alpha), \rho_0^{(\kappa)}(\alpha)] = 0, \tag{7.53}$$

holds, $\rho_0^{(\kappa)}(\alpha)$ is independent of κ ($\rho_0^{(\kappa)}(\alpha) = \rho_0(\alpha)$), then (7.19) becomes

$$A_n^\sigma(\alpha) = \frac{\partial}{\partial \alpha^n} S_{\rm vN}(\rho_0(\alpha)) + \mathcal{O}(\varepsilon^2), \tag{7.54}$$

using (H.1). (7.53) holds if H_S is non-degenerate. (7.54) can be shown from $[\overline{H_{\rm L}}, \rho_{i,b}^{(1)}] = 0$, which is weaker assumption than (7.53) and is derived from (7.53) for $\kappa = 1$. If we

neglect the Lamb shift Hamiltonian, namely we consider the QME for $\hat{K}_0(\alpha)$, (7.54) holds (with a replacement $\rho_0 \to \rho_0^{(0)}$). From (7.54), we obtain

$$\sigma_{\rm ex} = S_{\rm vN}(\rho_0(\alpha_\tau)) - S_{\rm vN}(\rho_0(\alpha_0)) + \mathcal{O}(\varepsilon^2\delta), \tag{7.55}$$

with $\delta = \max_{n,\alpha\in C} \frac{|\alpha^n - \alpha_0^n|}{|\bar{\alpha}^n|}$. $\bar{\alpha}^n$ is typical value of the n-th control parameter.

Yuge *et al.* [20] considered the outputs of $A(t) = -\sum_b \beta_b(t)[H_b - \mu_b(t)N_b]$ (for $n_{\rm C} = 0$) at $t = 0$ and $t = \tau$ as $a(0)$ and $a(\tau)$, and errorneously identified $a(\tau) - a(0)$ with the entropy production. To analyze $\sigma' \stackrel{\rm def}{=} \langle a(\tau) - a(0)\rangle$, improperly, they took $\chi_{H_b} \to -\beta_b\lambda$ and $\chi_{N_b} \to \beta_b\mu_b\lambda$ limit of the FCS-QME (2.64) only valid for time independent observables. The obtained Liouvillian (of which the Lamb shift Hamiltonian is neglected) incidentally satisfy (6.12). Using that Liouvillian, for the time-reversal symmetric system, Yuge *et al.* studied the relation between $A_n^\sigma(\alpha)$ and the symmetrized von Neumann entropy. In contrast, up to here, we do not suppose the time-reversal symmetry. In § 7.3, we consider the time-reversal operations and show that the potential $\mathcal{S}(\alpha)$ such that $A_n^\sigma(\alpha) = \partial\mathcal{S}/\partial\alpha^n + \mathcal{O}(\varepsilon^2)$ dose not exist if the time-reversal symmetry is broken.

7.3 Time-reversal operations

We define the time-reversal operation. We denote the time-reversal operator of the system by θ. We also define

$$\tilde{Y} \stackrel{\rm def}{=} \theta Y \theta^{-1}, \tag{7.56}$$

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

$$\tilde{\mathcal{J}}\tilde{Y} \stackrel{\rm def}{=} \theta(\mathcal{J}Y)\theta^{-1}, \tag{7.57}$$

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

$$i[\tilde{H}_{\rm L}(\alpha), \tilde{\rho}_0(\alpha)] + \sum_b \tilde{\Pi}_b(\alpha)\tilde{\rho}_0(\alpha) = 0, \tag{7.58}$$

using (2.147). If

$$\tilde{H}_{\rm L}(\alpha) = H_{\rm L}(\alpha), \;\; \tilde{\Pi}_b(\alpha) = \Pi_b(\alpha), \tag{7.59}$$

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

$$\tilde{\rho}_0(\alpha) = \rho_0^{(-1)}(\alpha), \tag{7.60}$$

holds. If the total Hamiltonian is time-reversal invariant, (7.59) holds [38]. If (7.59) holds and we neglect the Lamb shift Hamiltonian, the instantaneous steady state is time-reversal invariant: $\tilde{\rho}_0^{(0)} = \rho_0^{(0)}$.

As we will show, for time-reversal symmetric system,

$$\frac{\partial}{\partial\alpha^n} S_{\rm sym}(\rho_0(\alpha)) = -{\rm Tr}_S\Big[\ln\tilde{\rho}_0(\alpha)\frac{\partial\rho_0(\alpha)}{\partial\alpha^n}\Big] + \mathcal{O}(\varepsilon^2), \tag{7.61}$$

holds. Here,

$$S_{\rm sym}(\rho) \stackrel{\rm def}{=} -{\rm Tr}_S\big[\rho\frac{1}{2}(\ln\rho+\ln\tilde{\rho})\big], \tag{7.62}$$

is the symmetrized von Neumann entropy. Combining (7.19) with (7.60), we obtain

$$A_n^\sigma(\alpha) = \frac{\partial}{\partial\alpha^n}S_{\rm sym}(\rho_0(\alpha)) + \mathcal{O}(\varepsilon^2), \tag{7.63}$$

then, the equation (7.55) with $S_{\rm vN} \to S_{\rm sym}$ holds. As analogy, we consider

$$S'(\alpha) \stackrel{\rm def}{=} -{\rm Tr}_S\big[\rho_0(\alpha)\frac{1}{2}(\ln\rho_0(\alpha)+\ln\rho_0^{(-1)}(\alpha))\big], \tag{7.64}$$

for generally non-time-reversal symmetric system. The difference between $\partial S'(\alpha)/\partial\alpha^n$ and the first term of the RHS of (7.19) is

$$\begin{aligned}&\frac{\partial S'(\alpha)}{\partial\alpha^n} - \Big(-{\rm Tr}_S\Big[\ln\rho_0^{(-1)}(\alpha)\frac{\partial\rho_0(\alpha)}{\partial\alpha^n}\Big]\Big)\\ &= -\frac{1}{2}{\rm Tr}_S\big[\frac{\partial\rho_0}{\partial\alpha^n}(\ln\rho_0-\ln\rho_0^{(-1)})\big] - \frac{1}{2}{\rm Tr}_S\big[\rho_0\frac{\partial}{\partial\alpha^n}\ln\rho_0^{(-1)}\big].\end{aligned} \tag{7.65}$$

To calculate the RHS of this equation, we use formulas

$$\begin{aligned}\ln(A+\delta B) &= \ln A + \int_0^\infty ds\,\Big(\delta\frac{1}{A+s}B\frac{1}{A+s}\\ &\qquad -\delta^2\frac{1}{A+s}B\frac{1}{A+s}B\frac{1}{A+s}+\mathcal{O}(\delta^3)\Big),\end{aligned} \tag{7.66}$$

$$\frac{\partial}{\partial\alpha^n}\ln A(\alpha) = \int_0^\infty ds\,\frac{1}{A(\alpha)+s}\frac{\partial A(\alpha)}{\partial\alpha^n}\frac{1}{A(\alpha)+s}, \tag{7.67}$$

where $A, B, A(\alpha) \in$ B and δ is small real number. We proof (7.66) in Appendix I. (7.67) is derived from (7.66). $\rho_0 - \rho_0^{(-1)} = \varepsilon\psi + \mathcal{O}(\varepsilon^2)$ holds because $\overline{\rho_0^{(\kappa)}} = \rho_{\rm gc}(\alpha_S;\overline{\beta},\overline{\beta\mu})$. Then, the first term of the RHS of (7.65) is given by

$$\begin{aligned}&-\frac{1}{2}{\rm Tr}_S\big[\frac{\partial\rho_0}{\partial\alpha^n}(\ln\rho_0-\ln\rho_0^{(-1)})\big]\\ &= -\frac{\varepsilon}{2}\int_0^\infty ds\,{\rm Tr}_S\Big[\frac{\partial\rho_0}{\partial\alpha^n}\frac{1}{\rho_0^{(-1)}+s}\psi\frac{1}{\rho_0^{(-1)}+s}\Big]+\mathcal{O}(\varepsilon^2).\end{aligned} \tag{7.68}$$

The second term of the RHS of (7.65) is given by

$$\begin{aligned}&-\frac{1}{2}{\rm Tr}_S\big[\rho_0\frac{\partial}{\partial\alpha^n}\ln\rho_0^{(-1)}\big]\\ &= -\frac{1}{2}\int_0^\infty ds\,{\rm Tr}_S\Big[\frac{\partial\rho_0^{(-1)}}{\partial\alpha^n}\frac{1}{\rho_0^{(-1)}+s}(\rho_0^{(-1)}+\varepsilon\psi)\frac{1}{\rho_0^{(-1)}+s}\Big]+\mathcal{O}(\varepsilon^2)\\ &= -\frac{1}{2}{\rm Tr}_S\big[\frac{\partial\rho_0^{(-1)}}{\partial\alpha^n}\big] - \frac{\varepsilon}{2}\int_0^\infty ds\,{\rm Tr}_S\Big[\frac{\partial\rho_0^{(-1)}}{\partial\alpha^n}\frac{1}{\rho_0^{(-1)}+s}\psi\frac{1}{\rho_0^{(-1)}+s}\Big]+\mathcal{O}(\varepsilon^2)\\ &= -\frac{\varepsilon}{2}\int_0^\infty ds\,{\rm Tr}_S\Big[\frac{\partial\rho_0^{(-1)}}{\partial\alpha^n}\frac{1}{\rho_0+s}\psi\frac{1}{\rho_0+s}\Big]+\mathcal{O}(\varepsilon^2)\\ &= -\frac{\varepsilon}{2}\int_0^\infty ds\,{\rm Tr}_S\Big[\frac{\partial(\theta\rho_0^{(-1)}\theta^{-1})}{\partial\alpha^n}\frac{1}{\tilde{\rho}_0+s}\tilde{\psi}\frac{1}{\tilde{\rho}_0+s}\Big]+\mathcal{O}(\varepsilon^2).\end{aligned} \tag{7.69}$$

Here, we used $\varepsilon(\rho_0^{(-1)}+s)^{-1}=\varepsilon(\rho_0+s)^{-1}+\mathcal{O}(\varepsilon^2)$ and $\mathrm{Tr}_S\bullet=\mathrm{Tr}_S\tilde{\bullet}$ if $\mathrm{Tr}_S\bullet$ is real. In general, the RHS of (7.65) is not $\mathcal{O}(\varepsilon^2)$. However, if $\tilde{\rho}_0=\rho_0^{(-1)}$ holds, the RHS of (7.65) becomes $\mathcal{O}(\varepsilon^2)$ since $\tilde{\psi}=-\psi$, then (7.61) holds. In the proof of (7.61), Yuge *et al.* [20] used incorrect equations $\frac{\partial}{\partial\alpha^n}\ln\tilde{\rho}_0=\tilde{\rho}_0^{-1}\frac{\partial\tilde{\rho}_0}{\partial\alpha^n}$ and $\ln\rho_0-\ln\tilde{\rho}_0=\varepsilon\psi\tilde{\rho}_0^{-1}+\mathcal{O}(\varepsilon^2)$.

We introduce the BSN curvature

$$F^{\sigma}_{mn}(\alpha)=\frac{\partial A^{\sigma}_n}{\partial\alpha^m}-\frac{\partial A^{\sigma}_m}{\partial\alpha^n}. \tag{7.70}$$

$F^{\sigma}_{mn}(\alpha)=\mathcal{O}(\varepsilon^2)$ and the existence of $\mathcal{S}(\alpha)$ such that $A^{\sigma}_n(\alpha)=\partial\mathcal{S}(\alpha)/\partial\alpha^n+\mathcal{O}(\varepsilon^2)$ are equivalent. If $F^{\sigma}_{mn}(\alpha)=\mathcal{O}(\varepsilon)$ holds, $\mathcal{S}(\alpha)$ does not exist. $F^{\sigma}_{mn}(\alpha)$ is given by

$$F^{\sigma}_{mn}(\alpha)\;=\;f_{mn}(\alpha)-f_{nm}(\alpha)+\mathcal{O}(\varepsilon^2), \tag{7.71}$$

where

$$f_{mn}(\alpha)\overset{\text{def}}{=}-\mathrm{Tr}_S\Big(\frac{\partial\ln\rho_0^{(-1)}}{\partial\alpha^m}\frac{\partial\rho_0}{\partial\alpha^n}\Big). \tag{7.72}$$

$f_{mn}(\alpha)$ is given by

$$\begin{aligned} f_{mn}(\alpha)\;&=\;-\int_0^\infty ds\;\mathrm{Tr}_S\Big(\frac{1}{\rho_0^{(-1)}+s}\frac{\partial\rho_0^{(-1)}}{\partial\alpha^m}\frac{1}{\rho_0^{(-1)}+s}\frac{\partial\rho_0}{\partial\alpha^n}\Big)\\ &=\;\int_0^\infty ds\;[\mathcal{F}^{(0)}_{mn}(s)+\mathcal{F}^{(1)}_{mn}(s)+\mathcal{O}(\varepsilon^2)], \end{aligned} \tag{7.73}$$

with

$$\mathcal{F}^{(0)}_{mn}(s)=-\mathrm{Tr}_S\Big(\sigma_s\frac{\partial\rho_{\mathrm{gc}}}{\partial\alpha^m}\sigma_s\frac{\partial\rho_{\mathrm{gc}}}{\partial\alpha^n}\Big), \tag{7.74}$$

and

$$\sigma_s\overset{\text{def}}{=}\frac{1}{\rho_{\mathrm{gc}}+s}. \tag{7.75}$$

$\mathcal{F}^{(1)}_{mn}(s)$ is given by

$$\mathcal{F}^{(1)}_{mn}(s)\;=\;\mathcal{F}^{(1,0)}_{mn}(s)+\mathcal{F}^{(1,1)}_{mn}(s), \tag{7.76}$$

$$\mathcal{F}^{(1,0)}_{mn}(s)\;=\;\mathrm{Tr}_S\Big(\sigma_s\eta^{(-1)}\sigma_s\frac{\partial\rho_{\mathrm{gc}}}{\partial\alpha^m}\sigma_s\frac{\partial\rho_{\mathrm{gc}}}{\partial\alpha^n}+\sigma_s\frac{\partial\rho_{\mathrm{gc}}}{\partial\alpha^m}\sigma_s\eta^{(-1)}\sigma_s\frac{\partial\rho_{\mathrm{gc}}}{\partial\alpha^n}\Big), \tag{7.77}$$

$$\mathcal{F}^{(1,1)}_{mn}(s)\;=\;\mathrm{Tr}_S\Big(-\sigma_s\frac{\partial\eta^{(-1)}}{\partial\alpha^m}\sigma_s\frac{\partial\rho_{\mathrm{gc}}}{\partial\alpha^n}-\sigma_s\frac{\partial\rho_{\mathrm{gc}}}{\partial\alpha^m}\sigma_s\frac{\partial\eta^{(1)}}{\partial\alpha^n}\Big), \tag{7.78}$$

where

$$\eta^{(\kappa)}(\alpha)\overset{\text{def}}{=}\sum_b\sum_{i=1,2}\varepsilon_{i,b}\rho^{(\kappa)}_{i,b}. \tag{7.79}$$

$\rho_0 = \rho_{\rm gc} + \eta^{(1)} + \mathcal{O}(\varepsilon^2)$ and $\rho_0^{(-1)} = \rho_{\rm gc} + \eta^{(-1)} + \mathcal{O}(\varepsilon^2)$ hold. Because of $\mathcal{F}_{mn}^{(0)}(s) - \mathcal{F}_{nm}^{(0)}(s) = 0$ and $\mathcal{F}_{mn}^{(1,0)}(s) - \mathcal{F}_{nm}^{(1,0)}(s) = 0$, we obtain

$$\begin{aligned} F_{mn}^{\sigma}(\alpha) &= \int_0^\infty ds\, [\mathcal{F}_{mn}^{(1,1)}(s) - \mathcal{F}_{nm}^{(1,1)}(s)] + \mathcal{O}(\varepsilon^2) \\ &= \zeta_{mn} - \zeta_{nm} + \mathcal{O}(\varepsilon^2), \end{aligned} \tag{7.80}$$

with

$$\begin{aligned} \zeta_{mn} &= \int_0^\infty ds\, \mathrm{Tr}_S\Big(\sigma_s \frac{\partial[\eta^{(1)} - \eta^{(-1)}]}{\partial\alpha^m} \sigma_s \frac{\partial \rho_{\rm gc}}{\partial\alpha^n}\Big) \\ &= \mathrm{Tr}_S\Big(\frac{\partial[\eta^{(1)} - \eta^{(-1)}]}{\partial\alpha^m} \frac{\partial \ln \rho_{\rm gc}}{\partial\alpha^n}\Big) \\ &= -\mathrm{Tr}_S\Big(\frac{\partial[\eta^{(1)} - \eta^{(-1)}]}{\partial\alpha^m} \frac{\partial[\overline{\beta} H_S(\alpha_S) - \overline{\beta\mu} N_S]}{\partial\alpha^n}\Big). \end{aligned} \tag{7.81}$$

Here, we used $\mathrm{Tr}_S\Big(\frac{\partial[\eta^{(1)} - \eta^{(-1)}]}{\partial\alpha^m} \frac{\partial \ln \Xi}{\partial\alpha^n}\Big) = 0$ because $\mathrm{Tr}_S \eta^{(1)} = 0 = \mathrm{Tr}_S \eta^{(-1)}$. $\overline{\beta}$ and $\overline{\beta\mu}$ are functions of α''. Using $\mathrm{Tr}_S \bullet = \mathrm{Tr}_S \tilde{\bullet}$ if $\mathrm{Tr}_S \bullet$ is real, we obtain

$$\zeta_{mn} = -\mathrm{Tr}_S\Big(\frac{\partial[\theta\eta^{(1)}\theta^{-1} - \theta\eta^{(-1)}\theta^{-1}]}{\partial\alpha^m} \frac{\partial[\overline{\beta} \tilde{H}_S(\alpha_S) - \overline{\beta\mu} \tilde{N}_S]}{\partial\alpha^n}\Big). \tag{7.82}$$

For time-reversal symmetric system, $\tilde{H}_S = H_S$, $\tilde{N}_S = N_S$ and $\theta\eta^{(1)}\theta^{-1} = \eta^{(-1)}$ hold. Then, the above equation becomes $\zeta_{mn} = -\zeta_{mn}$, namely, $\zeta_{mn} = 0$ and $F_{mn}^{\sigma}(\alpha) = \mathcal{O}(\varepsilon^2)$ hold. However, if the time-reversal symmetry is broken, $\theta\eta^{(1)}\theta^{-1} \neq \eta^{(-1)}$ holds in general. Then, $\zeta_{mn} \neq -\zeta_{mn}$ namely $\zeta_{mn} \neq 0$ hold. ζ_{mn} is not symmetric for m and n. Then, if the time-reversal symmetry is broken and H_S is degenerated, $\mathcal{S}(\alpha)$ dose not exist in general. This is the most important result of this thesis.

7.4 Born-Markov approximation

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

$$A_n^{\sigma,\mathrm{BM}}(\alpha) = A_n^{\sigma}(\alpha) + \mathcal{O}(v^2), \tag{7.83}$$

$$S_{\mathrm{vN}}(\rho_0^{\mathrm{BM}}(\alpha)) = S_{\mathrm{vN}}(\rho_0(\alpha)) + \mathcal{O}(v^2), \tag{7.84}$$

$$S_{\mathrm{sym}}(\rho_0^{\mathrm{BM}}(\alpha)) = S_{\mathrm{sym}}(\rho_0(\alpha)) + \mathcal{O}(v^2), \tag{7.85}$$

hold [20]. Here, $v = u^2$ and $u(\ll 1)$ describes the order of H_{Sb}. Then, if (7.54) holds, we obtain

$$A_n^{\sigma,\mathrm{BM}}(\alpha) = \frac{\partial}{\partial\alpha^n} S_{\mathrm{vN}}(\rho_0^{\mathrm{BM}}(\alpha)) + \mathcal{O}(\varepsilon^2) + \mathcal{O}(v^2). \tag{7.86}$$

For time-reversal symmetric system,

$$A_n^{\sigma,\mathrm{BM}}(\alpha) = \frac{\partial}{\partial\alpha^n} S_{\mathrm{sym}}(\rho_0^{\mathrm{BM}}(\alpha)) + \mathcal{O}(\varepsilon^2) + \mathcal{O}(v^2), \tag{7.87}$$

holds.

Chapter 8

Comparison of two definitions of entropy production

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

$$\frac{dp_n(t)}{dt} = \sum_{m=1}^{\mathcal{N}} K_{nm}(\alpha_t) p_m(t). \tag{8.1}$$

The Liouvillian is given by

$$K_{nm}(\alpha) = \sum_b K_{nm}^{(b)}(\alpha), \tag{8.2}$$

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

$$\sigma^{\mathrm{C}} = \int_0^\tau dt \sum_{n,m} \sigma_{nm}^{\mathrm{C}}(\alpha_t) p_m(t), \tag{8.3}$$

where

$$\sigma_{nm}^{\mathrm{C}}(\alpha) = -K_{nm}(\alpha) \ln \frac{K_{nm}(\alpha)}{K_{mn}(\alpha)}. \tag{8.4}$$

We denote the solution of the QME with RWA by $\rho(t)$. We suppose $p_n(t) \stackrel{\mathrm{def}}{=} \langle n|\rho(t)|n\rangle$ is governed by (8.1) with

$$K_{nm}^{(b)}(\alpha) = (\Pi_b(\alpha))_{nn,mm}. \tag{8.5}$$

Here, $|n\rangle$ is the energy eigenstate of $H_S(\alpha_S)$,

$$(\Pi_b(\alpha)\bullet)_{nm} = \sum_{k,l} (\Pi_b(\alpha))_{nm,kl} (\bullet)_{kl}, \ (\bullet)_{kl} \stackrel{\mathrm{def}}{=} \langle k| \bullet |n\rangle. \tag{8.6}$$

This supposition implies (7.53). A sufficient condition by which $p_n(t)$ obeys (8.1) is below: (1) $H_S(\alpha_S)$ is non-degenerate and (2) $\{\alpha^n \in \alpha_S | \frac{\partial}{\partial \alpha^n}|n\rangle \neq 0\}$ are fixed. The eigenenergy can depend on $\{\alpha^n \in \alpha_S | \frac{\partial}{\partial \alpha^n}|n\rangle = 0\}$. We show that our average

entropy production (6.3) is given by a similar expression of (8.3):

$$\sigma = \int_0^\tau dt \sum_{n,m} \sigma_{nm}(\alpha_t) p_m(t). \tag{8.7}$$

Here,

$$\sigma_{nm}(\alpha) \stackrel{\text{def}}{=} \sum_b K_{nm}^{(b)}(\alpha)\theta_{nm}^{(b)}(\alpha) = -\sum_b K_{nm}^{(b)}(\alpha) \ln \frac{K_{nm}^{(b)}(\alpha)}{K_{mn}^{(b)}(\alpha)}, \tag{8.8}$$

with

$$\theta_{nm}^{(b)}(\alpha) \stackrel{\text{def}}{=} \begin{cases} -\ln \frac{K_{nm}^{(b)}(\alpha)}{K_{mn}^{(b)}(\alpha)} & K_{nm}^{(b)}(\alpha) \neq 0 \\ 0 & K_{nm}^{(b)}(\alpha) = 0 \end{cases}. \tag{8.9}$$

Because of (6.18), (6.25) and (6.26), the particle and energy currents are given by $i_{X_b} = \mathrm{Tr}_S[W^{X_b}\rho(t)]$ with $W^{X_b} = -(\Pi_b^\dagger X_S)^\dagger$ $(X = H, N)$. (6.20) leads

$$(W^{X_b})_{nm} = -\sum_{k,l} (\Pi_b)_{lk,mn}(X_S)_{kl}. \tag{8.10}$$

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

$$i_{X_b} = \sum_m (W^{X_b})_{mm} p_m(t), \tag{8.11}$$

holds. Substituting $(W^{X_b})_{mm} = -\sum_n K_{nm}^{(b)}(X_S)_{nn}$ into (8.11), we obtain

$$\begin{aligned} i_{X_b} &= -\sum_{n,m} K_{nm}^{(b)}(X_S)_{nn} p_m(t) \\ &= \sum_{n,m} K_{nm}^{(b)}[(X_S)_{mm} - (X_S)_{nn}] p_m(t). \end{aligned} \tag{8.12}$$

This equation leads

$$\dot{\sigma}(t) = -\sum_{n,m}\sum_b K_{nm}^{(b)} \beta_b(t)\{[(H_S)_{mm} - (H_S)_{nn}] - \nu_b(t)[(N_S)_{mm} - (N_S)_{nn}]\} p_m(t). \tag{8.13}$$

Using the local detailed balance condition derived from (7.3)

$$\ln \frac{K_{nm}^{(b)}(\alpha)}{K_{mn}^{(b)}(\alpha)} = \beta_b\{[(H_S)_{mm} - (H_S)_{nn}] - \nu_b[(N_S)_{mm} - (N_S)_{nn}]\}, \tag{8.14}$$

we obtain (8.7). For $b \in \mathcal{C}$, $(N_S)_{mm} - (N_S)_{nn} = 0$ holds for n and m such that $K_{nm}^{(b)}(\alpha) \neq 0$.

(8.12) can be rewritten as

$$i_{X_b} = \sum_{n,m} w_{nm}^{X_b}(\alpha_t) p_m(t), \; w_{nm}^{X_b}(\alpha_t) \stackrel{\text{def}}{=} K_{nm}^{(b)}[(X_S)_{mm} - (X_S)_{nn}]. \tag{8.15}$$

This $w_{nm}^{X_b}(\alpha_t)$ corresponds to $w_{ij}^{O_\mu}(\alpha_t)$ of (3.82).

Now we introduce $\boldsymbol{\mathcal{A}}_{nm} = \{b|K_{nm}^{(b)} \neq 0\}$. From the assumption, $\boldsymbol{\mathcal{A}}_{nm} = \boldsymbol{\mathcal{A}}_{mn}$ holds. If we suppose (2.163) for $b \in \boldsymbol{\mathcal{C}}$ and (2.137) for $b \in \boldsymbol{\mathcal{G}}$, $\boldsymbol{\mathcal{A}}_{nm} = \boldsymbol{\mathcal{C}}$ for $(N_S)_{mm} = (N_S)_{nn}$ and $\boldsymbol{\mathcal{A}}_{nm} = \boldsymbol{\mathcal{G}}$ for $(N_S)_{mm} \neq (N_S)_{nn}$. Then, (8.14) means

$$\ln \frac{K_{nm}^{(b)}(\alpha)}{K_{mn}^{(b)}(\alpha)} = \beta_b[(H_S)_{mm} - (H_S)_{nn}] \ (b \in \boldsymbol{\mathcal{C}}), \tag{8.16}$$

with $(N_S)_{mm} = (N_S)_{nn}$ and

$$\ln \frac{K_{nm}^{(b)}(\alpha)}{K_{mn}^{(b)}(\alpha)} = \beta_b\{[(H_S)_{mm} - (H_S)_{nn}] - \mu_b[(N_S)_{mm} - (N_S)_{nn}]\} \ (b \in \boldsymbol{\mathcal{G}}). \tag{8.17}$$

with $(N_S)_{mm} \neq (N_S)_{nn}$.

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

$$[\mathcal{K}^\lambda(\alpha)]_{nm} \stackrel{\text{def}}{=} \sum_b K_{nm}^{(b)}(\alpha) e^{i\lambda\theta_{nm}^{(b)}(\alpha)}. \tag{8.18}$$

Then, we obtain

$$\frac{\partial}{\partial(i\lambda)}\Big|_{\lambda=0} \sum_{n,m} \Big[\mathrm{T}\exp\Big[\int_0^\tau dt\, \mathcal{K}^\lambda(\alpha_t)\Big]\Big]_{nm} p_m(0) = \int_0^\tau dt \sum_{n,m} \sigma_{nm}(\alpha_t) p_m(t) = \sigma. \tag{8.19}$$

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

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

$$\sigma_{nm}^{\mathrm{C}}(\alpha) = \sigma_{nm}(\alpha) + \mathcal{O}(\varepsilon^2). \tag{8.20}$$

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

$$K_{nm}^{(b)} = \bar{K}_{nm}^{(b)} + \sum_{i=1,2} \varepsilon_{i,b} K_{nm}^{i,b} + \mathcal{O}(\varepsilon^2), \tag{8.21}$$

then we obtain

$$\sigma_{nm}^{\mathrm{C}}(\alpha) = \sigma_{nm}^{\mathrm{C}(0,1)} + \sigma_{nm}^{\mathrm{C}(2)}(\alpha) + \mathcal{O}(\varepsilon^3), \tag{8.22}$$

$$\sigma_{nm}(\alpha) = \sigma_{nm}^{(0,1)} + \sigma_{nm}^{(2)}(\alpha) + \mathcal{O}(\varepsilon^3), \tag{8.23}$$

with

$$\sigma_{nm}^{\mathrm{C}(0,1)} = -\bar{K}_{nm} \ln \frac{\bar{K}_{nm}}{\bar{K}_{mn}} + \sum_{i,b} \varepsilon_{i,b} \Big[K_{nm}^{i,b} \ln \frac{\bar{K}_{nm}}{\bar{K}_{mn}} + K_{nm}^{i,b} - K_{mn}^{i,b} \frac{\bar{K}_{nm}}{\bar{K}_{mn}}\Big], \tag{8.24}$$

$$\sigma_{nm}^{(0,1)} = \sum_{b \in \boldsymbol{\mathcal{A}}_{nm}} \Big(- \bar{K}_{nm}^{(b)} \ln \frac{\bar{K}_{nm}^{(b)}}{\bar{K}_{mn}^{(b)}} + \sum_i \varepsilon_{i,b} \Big[K_{nm}^{i,b} \ln \frac{\bar{K}_{nm}^{(b)}}{\bar{K}_{mn}^{(b)}} + K_{nm}^{i,b} - K_{mn}^{i,b} \frac{\bar{K}_{nm}^{(b)}}{\bar{K}_{mn}^{(b)}}\Big]\Big). \tag{8.25}$$

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

$$\bar{K}_{nm} = \sum_{b \in \boldsymbol{\mathcal{A}}_{nm}} \bar{K}_{nm}^{(b)}, \ \bar{K}_{mn} = \sum_{b \in \boldsymbol{\mathcal{A}}_{nm}} \bar{K}_{mn}^{(b)}. \tag{8.26}$$

(8.14) leads $\bar{K}^{(b)}_{nm}/\bar{K}^{(b)}_{mn}$ is independent of $b \in \boldsymbol{\mathcal{A}}_{nm}$. Then, we obtain

$$\frac{\bar{K}^{(b)}_{nm}}{\bar{K}^{(b)}_{mn}} = \frac{\bar{K}_{nm}}{\bar{K}_{mn}} \ (b \in \boldsymbol{\mathcal{A}}_{nm}). \tag{8.27}$$

The above relation and (8.26) lead

$$\sigma^{\mathrm{C}(0,1)}_{nm} = \sigma^{(0,1)}_{nm}, \tag{8.28}$$

and (8.20). (8.20) leads

$$\sigma^{\mathrm{C}}_{\mathrm{ex}} = \sigma_{\mathrm{ex}} + \mathcal{O}(\varepsilon^2\delta). \tag{8.29}$$

Here, $\sigma^{\mathrm{C}}_{\mathrm{ex}}$ is given by (J.13). Then, (J.12), the result of Ref.[21], coincides with (7.55) when $p_n(t) = \langle n|\rho(t)|n\rangle$ is governed by the master equation (8.1).

Chapter 9

Conclusion

9.1 General conclusion

In this thesis, for open systems described by the quantum master equation (QME), we investigated the quantum pump and the excess entropy production.

First, we investigated quantum pump using the FCS-QME (full counting statistics with quantum master equation) approach. We studied the non-adiabatic effect and the showed that the general solution of the QME $\rho(t)$ is decomposed as $\rho(t) = \rho_0(\alpha_t) + \sum_{n=1}^{\infty} \rho^{(n)}(t) + \sum_{n=0}^{\infty} \tilde{\rho}^{(n)}(t)$ (Chap.3). Here, α_t is the value of the set of the control parameters at time t and $\rho_0(\alpha_t)$ is the instantaneous steady state of the QME, $\rho^{(n)}(t)$ and $\tilde{\rho}^{(n)}(t)$ are calculable and order $(\omega/\Gamma)^n$ where ω is the modulation frequency of the control parameters and Γ is the coupling strength between the system and the baths. $\tilde{\rho}^{(n)}(t)$ exponentially damps (like $e^{-\Gamma t}$) as a function of time. We showed that the generalized mater equation (GME) approach provides $\boldsymbol{p}(t) = \boldsymbol{p}_{(\mathrm{ss})}(t) + \delta\boldsymbol{p}(t)$ in the Born approximation (Appendix § F). Here, $\boldsymbol{p}$ corresponds to the set of the diagonal components of ρ in the matrix representation by the energy eigenstates, $\boldsymbol{p}_{(\mathrm{ss})}(t)$ corresponds to $\rho_0(\alpha_t) + \sum_{n=1}^{\infty} \rho^{(n)}(t)$ and the the term $\delta\boldsymbol{p}(t)$ originates from non-Markovian effects. The FCS-QME picks out one higher order non-adiabatic piece of information from the solution of the QME, namely, if we have $\rho^{(n)}(t)$, the FCS-QME method provides $(n+1)$-th order pump currents. Moreover, we showed that the Berry-Sinitsyn-Nemenman (BSN) phase derived under the "adiabatic" condition which makes the Berry phase like treatment appropriate has the non-adiabatic (first order of ω) information. We showed that the quantum pump dose not occur in all orders of the pumping frequency when the system control parameters and the thermodynamic parameters (the temperatures and the chemical potentials of the baths) are fixed under the zero-bias condition.

Next, we studied the quantum adiabatic pump of the quantum dot (QD) system weakly coupled to two leads (L and R) in § 4.2 and § 4.3 using the FCS-QME with the rotating wave approximation (RWA) defined as the long coarse-graining time limit of the coarse-graining approximation (CGA). We confirmed the consistency between the FCS-QME approach and the GME approach for a QD of one quantum level with finite Coulomb interaction (§ 4.2.3 and § 4.3.3). We showed that the pumped charge and spin coming from the instantaneous steady current are not negligible when the thermodynamic parameters are not fixed to zero bias (§ 4.2.2 and § 4.3.2). To observe the spin effects, we consider collinear magnetic fields, which affect the spins through the Zeeman effect, with different amplitudes applying to the QDs (B_S) and the leads (B_L and B_R). We focused on the dynamic parameters (B_S, $B_{L/R}$ and the coupling strength between QDs and leads, $\Delta_{L/R}$) as control parameters. In one level QD with the Coulomb interaction U, we analytically calculated the BSN curvatures of spin and charge of (B_L, B_S) pump and (Δ_L, B_S) pump for the noninteracting limit ($U = 0$) and the strong interaction limit ($U = \infty$) at zero-bias. The difference depending on

U appeared through $n_U(sB_S)$ which is the average number of the electrons with spin s in the QD. For (B_L, B_S) pump, the energy dependences of the line-width functions are essential. Moreover, we studied the (Δ_L, B_S) pump for finite U at zero-bias (§ 4.3.5). The effect of U appeared through $n_U(sB_S)$. When half-filling condition is satisfied, the charge pump does not occur.

We studied the quantum diabatic pump for spinless one level QD coupled to two leads (Chap.5). We calculated $\{\rho^{(n)}(t)\}_{n=1}^5$, $\{\tilde{\rho}^{(n)}(t)\}_{n=1}^5$ and particle current up to 6th order and pumped particle numbers.

In § 1.6, we newly defined average entropy production rate $\dot{\sigma}(t)$ using the average energy and particle currents, which are calculated by using the FCS-QME. Next, we introduced the generalized QMEs (GQMEs) providing $\dot{\sigma}(t)$ (Chap.6). The GQMEs do not relate the higher moments (thus and the FCS) of the entropy production. We can calculate only the average of the entropy production. In § 7.2, using the GQME, in weakly nonequilibrium regime, we analyzed the BSN vector for the entropy production, $A_n^\sigma(\alpha)$, which provides the excess entropy production $\sigma_{\rm ex}$ under quasistatic operations between nonequilibrium steady states as $\sigma_{\rm ex} = \int_C d\alpha^n\, A_n^\sigma(\alpha)$, and showed $A_n^\sigma(\alpha) = -{\rm Tr}_S\big[\ln \rho_0^{(-1)}(\alpha)\frac{\partial \rho_0(\alpha)}{\partial \alpha^n}\big] + \mathcal{O}(\varepsilon^2)$. Here, α is the set of the control parameters and α^n is n-th component of the control parameters, C is the trajectory in the control parameter space, ${\rm Tr}_S$ denotes the trace of the system, and ε is a measure of degree of nonequilibrium. $\rho_0^{(-1)}(\alpha)$ is the instantaneous steady state obtained from the QME with reversing the sign of the Lamb shift term. In general, the potential $\mathcal{S}(\alpha)$ such that $A_n^\sigma(\alpha) = \frac{\partial \mathcal{S}(\alpha)}{\partial \alpha^n} + \mathcal{O}(\varepsilon^2)$ dose not exist (§ 7.3). This is the most important result of this thesis. The origins of the non-existence of the potential $\mathcal{S}(\alpha)$ are a quantum effect (the Lamb shift term) and the breaking of the time-reversal symmetry. The non-existence of the potential means that the excess entropy essentially depends on the path of the modulation. In this case, it is important to consider the generalization of the entropy concept. In contrast, if the system Hamiltonian is non-degenerate or the Lamb shift term is negligible, we obtain $\sigma_{\rm ex} = S_{\rm vN}(\rho_0(\alpha_{t_f})) - S_{\rm vN}(\rho_0(\alpha_{t_i})) + \mathcal{O}(\varepsilon^2\delta)$. Here, $S_{\rm vN}(\rho) = -{\rm Tr}_S[\rho \ln \rho]$ is the von Neumann entropy, t_i and t_f are the initial and final times of the operation, and δ describes the amplitude of the change of the control parameters. For time-reversal symmetric system, we showed that $\mathcal{S}(\alpha)$ is the symmetrized von Neumann entropy. Additionally, we pointed out that preceding expression of the entropy production in the classical Markov jump process is different from ours and showed that these are approximately equivalent in the weakly nonequilibrium regime. We also checked that the definition of the average entropy production in the classical Markov jump process by Ref.[19] is equivalent to ours.

9.2 Future perspective

$\rho_0^{(-1)}$ and $A_n^\sigma(\alpha)$ should be calculated for concrete model in which the system Hamiltonian is degenerated or/and the time-reversal symmetry is broken. For instance, multi-level QD system applying the magnetic field is a candidate.

If $\mathcal{S}(\alpha)$ does not exist, the path dependence of the excess entropy is essential. The path dependence and the path of which the excess entropy is minimized should be studied.

Appendix A

Born-Markov approximation

We denote $\mathcal{L}_b^\chi$ in the Born-Markov approximation by $\mathcal{L}_{b(\mathrm{BM})}^\chi$. From (2.66) and (2.75), we obtain

$$\begin{aligned}\mathcal{L}_{b(\mathrm{BM})}^\chi \bullet \;=\; -\int_0^\infty ds \sum_{\mu,\nu} \Big(& s_{b\nu}^\dagger s_{b\mu}^I(t-s,t) \bullet C_{b,\nu\mu}(s) - s_{b\mu}^I(t-s,t) \bullet s_{b\nu}^\dagger C_{b,\nu\mu}^\chi(s) \\ & - s_{b\nu} \bullet s_{b\mu}^{I\dagger}(t-s,t) C_{b,\mu\nu}^\chi(-s) + \bullet s_{b\mu}^{I\dagger}(t-s,t) s_{b\nu} C_{b,\mu\nu}(-s)\Big). \end{aligned} \tag{A.1}$$

$C_{b,\mu\nu}(s)$ damps exponentially as $e^{-|s|/\tau_b}$ where τ_b is the relaxation time of the bath b. Then, in the calculations of $s_{b\mu}^I(t-s,t)$ and $s_{b\mu}^{I\dagger}(t-s,t)$, the values of the control parameters can be approximated by $\alpha_S(t)$. Then, we obtain

$$s_{b\mu}^I(t-s,t) = \sum_\omega e^{i\omega s} s_{b\mu}(\omega),\; s_{b\mu}^{I\dagger}(t-s,t) = \sum_\omega e^{-i\omega s} [s_{b\mu}(\omega)]^\dagger, \tag{A.2}$$

and

$$\begin{aligned}\mathcal{L}_{b(\mathrm{BM})}^\chi \bullet \;=\; -\int_0^\infty ds \sum_{\mu,\nu}\sum_\omega \Big(& \Big\{ s_{b\nu}^\dagger s_{b\mu}(\omega) \bullet C_{b,\nu\mu}(s) - s_{b\mu}(\omega) \bullet s_{b\nu}^\dagger C_{b,\nu\mu}^\chi(s) \Big\} e^{i\omega s} \\ & + \Big\{ - s_{b\nu} \bullet [s_{b\mu}(\omega)]^\dagger C_{b,\mu\nu}^\chi(-s) + \bullet [s_{b\mu}(\omega)]^\dagger s_{b\nu} C_{b,\mu\nu}(-s) \Big\} e^{-i\omega s}\Big). \end{aligned} \tag{A.3}$$

Here,

$$\begin{aligned}\int_0^\infty ds\, C_{b,\nu\mu}^\chi(s) e^{i\omega s} \;&=\; \int_0^\infty ds \int_{-\infty}^\infty d\Omega\, \frac{1}{2\pi} \Phi_{b,\nu\mu}^\chi(\Omega) e^{i(\omega-\Omega)s} \\ &=\; \int_{-\infty}^\infty d\Omega\, \frac{1}{2\pi} \Big[\pi\delta(\Omega-\omega) - i\mathrm{P}\frac{1}{\Omega-\omega}\Big] \Phi_{b,\nu\mu}^\chi(\Omega) \\ &=\; \Phi_{b,\nu\mu}^{(+)\chi}(\omega), \end{aligned} \tag{A.4}$$

and

$$\int_0^\infty ds\, C_{b,\mu\nu}^\chi(-s) e^{-i\omega s} \;=\; \Phi_{b,\mu\nu}^{(-)\chi}(\omega), \tag{A.5}$$

hold. Then, we get

$$\begin{aligned}\mathcal{L}_{b(\mathrm{BM})}^\chi \bullet \;=\; -\sum_{\mu,\nu}\sum_\omega \Big(& s_{b\mu}^\dagger s_{b\nu}(\omega) \bullet \Phi_{b,\mu\nu}^{(+)}(\omega) - s_{b\nu}(\omega) \bullet s_{b\mu}^\dagger \Phi_{b,\mu\nu}^{(+)\chi}(\omega) \\ & - s_{b\nu} \bullet [s_{b\mu}(\omega)]^\dagger \Phi_{b,\mu\nu}^{(-)\chi}(\omega) + \bullet [s_{b\mu}(\omega)]^\dagger s_{b\nu} \Phi_{b,\mu\nu}^{(-)}(\omega)\Big) \\ =\; \mathcal{L}_{b(\mathrm{BM})}^{\Phi,\chi} \bullet + & \mathcal{L}_{b(\mathrm{BM})}^{\Phi,\chi} \bullet . \end{aligned} \tag{A.6}$$

Here,

$$\mathcal{L}^{\Phi,\chi}_{b(\mathrm{BM})}\bullet = -\frac{1}{2}\sum_{\mu,\nu}\sum_{\omega}\Big(\Phi_{b,\mu\nu}(\omega)s^{\dagger}_{b\mu}s_{b\nu}(\omega)\bullet - \Phi^{\chi}_{b,\mu\nu}(\omega)s_{b\nu}(\omega)\bullet s^{\dagger}_{b\mu}$$
$$-\Phi^{\chi}_{b,\mu\nu}(\omega)s_{b\nu}\bullet[s_{b\mu}(\omega)]^{\dagger} + \Phi_{b,\mu\nu}(\omega)\bullet[s_{b\mu}(\omega)]^{\dagger}s_{b\nu}\Big), \tag{A.7}$$

$$\mathcal{L}^{\Psi,\chi}_{b(\mathrm{BM})}\bullet = \frac{i}{2}\sum_{\mu,\nu}\sum_{\omega}\frac{1}{2}\Big(\Psi_{b,\mu\nu}(\omega)s^{\dagger}_{b\mu}s_{b\nu}(\omega)\bullet - \Psi^{\chi}_{b,\mu\nu}(\omega)s_{b\nu}(\omega)\bullet s^{\dagger}_{b\mu}$$
$$+\Psi^{\chi}_{b,\mu\nu}(\omega)s_{b\nu}\bullet[s_{b\mu}(\omega)]^{\dagger} - \bullet\Psi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^{\dagger}s_{b\nu}\Big). \tag{A.8}$$

For (2.106), we obtain

$$\mathcal{L}^{\Phi,\chi}_{b(\mathrm{BM})}\bullet = -\frac{1}{2}\sum_{\alpha,\beta}\sum_{\omega}\Big(\Phi^{-}_{b,\alpha\beta}(\omega)a^{\dagger}_{\alpha}a_{\beta}(\omega)\bullet - \Phi^{-,\chi}_{b,\alpha\beta}(\omega)a_{\beta}(\omega)\bullet a^{\dagger}_{\alpha}$$
$$-\Phi^{-,\chi}_{b,\alpha\beta}(\omega)a_{\beta}\bullet[a_{\alpha}(\omega)]^{\dagger} + \Phi^{-}_{b,\alpha\beta}(\omega)\bullet[a_{\alpha}(\omega)]^{\dagger}a_{\beta}$$
$$+\Phi^{+}_{b,\alpha\beta}(\omega)a_{\alpha}[a_{\beta}(\omega)]^{\dagger}\bullet - \Phi^{+,\chi}_{b,\alpha\beta}(\omega)[a_{\beta}(\omega)]^{\dagger}\bullet a_{\alpha}$$
$$-\Phi^{+,\chi}_{b,\alpha\beta}(\omega)a^{\dagger}_{\beta}\bullet a_{\alpha}(\omega) + \Phi^{+}_{b,\alpha\beta}(\omega)\bullet a_{\alpha}(\omega)a^{\dagger}_{\beta}\Big), \tag{A.9}$$

and

$$\mathcal{L}^{\Psi,\chi}_{b(\mathrm{BM})}\bullet = \frac{i}{2}\sum_{\alpha,\beta}\sum_{\omega}\Big(\Psi^{-}_{b,\alpha\beta}(\omega)a^{\dagger}_{\alpha}a_{\beta}(\omega)\bullet - \Psi^{-,\chi}_{b,\alpha\beta}(\omega)a_{\beta}(\omega)\bullet a^{\dagger}_{\alpha}$$
$$+\Psi^{-,\chi}_{b,\alpha\beta}(\omega)a_{\beta}\bullet[a_{\alpha}(\omega)]^{\dagger} - \Psi^{-}_{b,\alpha\beta}(\omega)\bullet[a_{\alpha}(\omega)]^{\dagger}a_{\beta}$$
$$-\Psi^{+}_{b,\alpha\beta}(\omega)a_{\alpha}[a_{\beta}(\omega)]^{\dagger}\bullet + \Psi^{+,\chi}_{b,\alpha\beta}(\omega)[a_{\beta}(\omega)]^{\dagger}\bullet a_{\alpha}$$
$$-\Psi^{+,\chi}_{b,\alpha\beta}(\omega)a^{\dagger}_{\beta}\bullet a_{\alpha}(\omega) + \Psi^{+}_{b,\alpha\beta}(\omega)\bullet a_{\alpha}(\omega)a^{\dagger}_{\beta}\Big). \tag{A.10}$$

By the way,

$$\mathcal{L}^{\Phi}_{b(\mathrm{BM})}e^{-\beta_b(H_S-\mu N_S)} = 0, \tag{A.11}$$

holds. Here, $\mathcal{L}^{\Phi}_{b(\mathrm{BM})} = \mathcal{L}^{\Phi,\chi}_{b(\mathrm{BM})}\big|_{\chi=0}$. Because of (2.130), 1st and 7th terms of (A.9) cancel in the LHS of (A.11). Similarly, 2nd and 8th, 3rd and 5th, 4th and 6th terms of (A.9) cancel. If $\mathcal{L}^{\Psi}_{b(\mathrm{BM})}$ is negligible, ρ_0 becomes (3.67) at zero-bias.

Appendix B

Liouville space

By following correspondence, an arbitrary linear operator (which operates to the Hilbert space) $\bullet = \sum_{n,m}\langle n| \bullet |m\rangle|n\rangle\langle m|$ is mapped to a vector of the Liouville space[26, 80], $|\bullet\rangle\rangle = \sum_{n,m}\langle n| \bullet |m\rangle|nm\rangle\rangle$:

$$|n\rangle\langle m| \longleftrightarrow |nm\rangle\rangle, \tag{B.1}$$

$$\mathrm{Tr}(|m\rangle\langle n|n'\rangle\langle m'|) \longleftrightarrow \langle\langle nm|n'm'\rangle\rangle, \tag{B.2}$$

$$\mathrm{Tr}(A^\dagger B) \longleftrightarrow \langle\langle A|B\rangle\rangle, \tag{B.3}$$

$$\mathrm{Tr}(\bullet) \longleftrightarrow \langle\langle 1|\bullet\rangle\rangle. \tag{B.4}$$

Here, $\{|n\rangle\}$ is an arbitrarily complete orthonormal basis. The inner product of the Liouville space is defined by the Hilbert-Schmidt product [(B.3)]. The Hermitian conjugate of $|\bullet\rangle\rangle$ is defined as $\langle\langle\bullet| = (|\bullet\rangle\rangle)^\dagger = \sum_{n,m}\langle n|\bullet|m\rangle^*\langle\langle nm|$. An arbitrary linear super-operator $\hat{J}$ which operates to any operator $(\bullet)$ is mapped to a corresponding operator of the Liouville space $(\check{J})$ as

$$|\hat{J}\bullet\rangle\rangle = \check{J}|\bullet\rangle\rangle. \tag{B.5}$$

The matrix representation of $\check{J}$ (or $\hat{J}$) is defined by

$$J_{nm,kl} = \langle\langle nm|\check{J}|kl\rangle\rangle. \tag{B.6}$$

In the main text of this thesis, both $\check{J}$ and $\hat{J}$ are denoted by $\hat{J}$.

Generally, the Liouvillian $\hat{K}^\chi$ operates to an operator $\bullet$ as

$$\hat{K}^\chi\bullet = -i[H_S, \bullet] + \hat{\Pi}^\chi\bullet, \tag{B.7}$$

$$\hat{\Pi}^\chi\bullet = \sum_a c_a^\chi A_a \bullet B_a, \tag{B.8}$$

where H_S is the system Hamiltonian, $\hat{\Pi}^\chi$ is the dissipator, A_a, B_a are operators, and $c_a^\chi(\alpha)$ is a complex number. The matrix representation of (B.7) is given by

$$\sum_{k,l} K^\chi_{nm,kl}\bullet_{kl} = \sum_{k,l}\big[-i\{(H_S)_{nk}\delta_{lm} - \delta_{nk}(H_S)_{lm}\}\bullet_{kl} + \{\sum_a c_a^\chi (A_a)_{nk}(B_a)_{lm}\}\bullet_{kl}\big], \tag{B.9}$$

where $\bullet_{kl} = \langle k| \bullet |l\rangle$. Hence the matrix representation of $\hat{K}^{\chi}$ is given by

$$K^{\chi}_{nm,kl} = -iH_{nm,kl} + \Pi^{\chi}_{nm,kl}, \tag{B.10}$$

$$H_{nm,kl} = (H_S)_{nk}\delta_{lm} - \delta_{nk}(H_S)_{lm}, \tag{B.11}$$

$$\Pi^{\chi}_{nm,kl} = \sum_a c^{\chi}_a (A_a)_{nk}(B_a)_{lm}. \tag{B.12}$$

Finally, we consider the current operators defined by (3.14). $\hat{K}^{O_\mu} = \frac{\partial \hat{K}^{\chi}(\alpha)}{\partial(i\chi_{O_\mu})}\Big|_{\chi=0}$ is given by

$$\hat{K}^{O_\mu}\bullet = \sum_a c^{O_\mu}_a A_a \bullet B_a. \tag{B.13}$$

Hence the current operators defined by (3.14) are given by

$$W^{O_\mu} = \sum_a c^{O_\mu}_a B_a A_a. \tag{B.14}$$

Appendix C

The time evolutions of $c_n^\chi(t)$

In this chapter, we derive the time evolution equations of $c_n^\chi(t)$ of (3.23). The LHS of the FCS-QME, $\frac{d}{dt}|\rho^\chi(t)\rangle\rangle = \hat{K}^\chi(\alpha_t)|\rho^\chi(t)\rangle\rangle$, is

$$\begin{aligned}\frac{d}{dt}|\rho^\chi(t)\rangle\rangle &= \sum_n \Big\{ \frac{dc_n^\chi(t)}{dt} e^{\Lambda_n^\chi(t)}|\rho_n^\chi(\alpha_t)\rangle\rangle + c_n^\chi(t)e^{\Lambda_n^\chi(t)}\lambda_n(\alpha_t)|\rho_n^\chi(\alpha_t)\rangle\rangle \\ &\quad + c_n^\chi(t)e^{\Lambda_n^\chi(t)}\frac{d}{dt}|\rho_n^\chi(\alpha_t)\rangle\rangle \Big\}. \end{aligned} \tag{C.1}$$

And the RHS of the FCS-QME is

$$\begin{aligned}\hat{K}^\chi(\alpha_t)|\rho^\chi(t)\rangle\rangle &= \sum_n c_n^\chi(t)e^{\Lambda_n^\chi(t)}\hat{K}^\chi(\alpha_t)|\rho_n^\chi(\alpha_t)\rangle\rangle \\ &= \sum_n c_n^\chi(t)e^{\Lambda_n^\chi(t)}\lambda_n(\alpha_t)|\rho_n^\chi(\alpha_t)\rangle\rangle. \end{aligned} \tag{C.2}$$

Hence we obtain

$$\sum_n \Big\{ \frac{dc_n^\chi(t)}{dt} e^{\Lambda_n^\chi(t)}|\rho_n^\chi(\alpha_t)\rangle\rangle + c_n^\chi(t)e^{\Lambda_n^\chi(t)}\frac{d|\rho_n^\chi(\alpha_t)\rangle\rangle}{dt} \Big\} = 0. \tag{C.3}$$

Applying $\langle\langle l_m^\chi(\alpha_t)|$ to (C.3), and using $\langle\langle l_n^\chi(\alpha)|\rho_m^\chi(\alpha)\rangle\rangle = \delta_{nm}$, we obtain

$$\frac{d}{dt}c_m^\chi(t) = -\sum_n c_n^\chi(t)e^{\Lambda_n^\chi(t)-\Lambda_m^\chi(t)}\langle\langle l_m^\chi(\alpha_t)|\frac{d|\rho_n^\chi(\alpha_t)\rangle\rangle}{dt}. \tag{C.4}$$

By the way, the time derivative of (3.3), $\hat{K}^\chi(\alpha_t)|\rho_n^\chi(\alpha_t)\rangle\rangle = \lambda_n^\chi(\alpha_t)|\rho_n^\chi(\alpha_t)\rangle\rangle$, is

$$\frac{d\hat{K}^\chi(\alpha_t)}{dt}|\rho_n^\chi(\alpha_t)\rangle\rangle + \hat{K}^\chi(\alpha_t)\frac{d|\rho_n^\chi(\alpha_t)\rangle\rangle}{dt} = \frac{d\lambda_n^\chi(\alpha_t)}{dt}|\rho_n^\chi(\alpha_t)\rangle\rangle + \lambda_n^\chi(\alpha_t)\frac{d|\rho_n^\chi(\alpha_t)\rangle\rangle}{dt} \tag{C.5}$$

Applying $\langle\langle l_m^\chi(\alpha_t)|$ to this equation, we obtain

$$\begin{aligned}&\langle\langle l_m^\chi(\alpha_t)|\frac{d\hat{K}^\chi(\alpha_t)}{dt}|\rho_n^\chi(\alpha_t)\rangle\rangle + \lambda_m^\chi(\alpha_t)\langle\langle l_m^\chi(\alpha_t)|\frac{d|\rho_n^\chi(\alpha_t)\rangle\rangle}{dt} \\ &= \frac{d\lambda_n^\chi(\alpha_t)}{dt}\delta_{mn} + \lambda_n^\chi(\alpha_t)\langle\langle l_m^\chi(\alpha_t)|\frac{d|\rho_n^\chi(\alpha_t)\rangle\rangle}{dt}, \end{aligned} \tag{C.6}$$

and it leads to

$$\langle\langle l_m^\chi(\alpha_t)|\frac{d|\rho_n^\chi(\alpha_t)\rangle\rangle}{dt} = -\frac{\langle\langle l_m^\chi(\alpha_t)|\frac{d\hat{K}^\chi(\alpha_t)}{dt}|\rho_n^\chi(\alpha_t)\rangle\rangle}{\lambda_m^\chi(\alpha_t) - \lambda_n^\chi(\alpha_t)}, \tag{C.7}$$

for $\lambda_m^\chi(\alpha_t) \neq \lambda_n^\chi(\alpha_t)$. Substituting this to (C.4), we obtain

$$\begin{aligned}\frac{dc_m^\chi(t)}{dt} &= -\langle\langle l_m^\chi(\alpha_t)|\frac{d}{dt}|\rho_m^\chi(\alpha_t)\rangle\rangle c_m^\chi(t) \\ &\quad + \sum_{n(\neq m)} c_n^\chi(t) e^{\Lambda_n^\chi(t)-\Lambda_m^\chi(t)} \frac{\langle\langle l_m^\chi(\alpha_t)|\frac{d\hat{K}^\chi(\alpha_t)}{dt}|\rho_n^\chi(\alpha_t)\rangle\rangle}{\lambda_m^\chi(\alpha_t)-\lambda_n^\chi(\alpha_t)}. \end{aligned} \tag{C.8}$$

The above equation can also be written as

$$\frac{d\tilde{c}_m^\chi(t)}{dt} = \sum_{n(\neq m)} \tilde{c}_n^\chi(t) e^{\Lambda_n^\chi(t)-\Lambda_m^\chi(t)+\eta_m^\chi(t)-\eta_n^\chi(t)} \frac{\langle\langle l_m^\chi(\alpha_t)|\frac{d\hat{K}^\chi(\alpha_t)}{dt}|\rho_n^\chi(\alpha_t)\rangle\rangle}{\lambda_m^\chi(\alpha_t)-\lambda_n^\chi(\alpha_t)}, \tag{C.9}$$

where $\tilde{c}_m^\chi(t) = c_m^\chi(t) e^{\eta_m^\chi(t)}$ with

$$\begin{aligned}\eta_m^\chi(t) &= \int_0^t ds\ \langle\langle l_m^\chi(\alpha_s)|\frac{d}{ds}|\rho_m^\chi(\alpha_s)\rangle\rangle \\ &= \sum_k \int_C d\alpha^k\ \langle\langle l_m^\chi(\alpha)|\frac{\partial}{\partial\alpha^k}|\rho_m^\chi(\alpha)\rangle\rangle. \end{aligned} \tag{C.10}$$

Here, C is the trajectory from α_0 to α_t, α^k are the k-th component of the control parameters, and $\eta_m^\chi(t) = \mathcal{O}(1)$ since $\langle\langle l_m^\chi(\alpha_t)|\frac{d}{dt}|\rho_m^\chi(\alpha_t)\rangle\rangle = \mathcal{O}(\omega)$ with $\omega = 2\pi/\tau$. In the RHS of (C.9), the dominant term is $n = 0$ if $m \neq 0$ because $\mathrm{Re}\lambda_0^\chi(\alpha) > \mathrm{Re}\lambda_n^\chi(\alpha)$. Using $\frac{d\hat{K}^\chi(\alpha_t)}{dt} = \mathcal{O}(\Gamma\omega)$, $\lambda_n^\chi(\alpha_t) = \mathcal{O}(\Gamma)$, $e^{\eta_n^\chi(t)} = \mathcal{O}(1)$ and $c_0^\chi(t)e^{\Lambda_0^\chi} = \mathcal{O}(1)$, we obtain

$$\frac{d\tilde{c}_m^\chi(t)}{dt} = \mathcal{O}(e^{-\Lambda_m^\chi(t)}\omega), \tag{C.11}$$

and

$$c_m^\chi(t)e^{\Lambda_m^\chi(t)} = \mathcal{O}\big(\omega \int_0^t ds\ e^{\Lambda_m^\chi(t)-\Lambda_m^\chi(s)}\big) = \mathcal{O}\big(\frac{\omega}{\Gamma}\big). \tag{C.12}$$

For $\chi = 0$, (C.12) is also derived from

$$\delta\rho(t) = \rho(t) - \rho_0(\alpha_t) = \sum_{m\neq 0} c_m(t) e^{\Lambda_m(t)} \rho_m(\alpha_t), \tag{C.13}$$

and (3.33) and (3.60).

Appendix D

The validity of the adiabatic expansion

In the derivation of the QME with CGA, when going from (2.57) to (2.58), we used the following type of approximation:

$$\int_t^{t+T} du \int_t^u ds\, G([\alpha]_s^u; s, u; t) \approx \int_t^{t+T} du \int_t^u ds\, G([\alpha_t]; s, u; t). \tag{D.1}$$

Here, $G([\alpha]_s^u, s, u, t) \sim e^{-(u-s)/\tau_B}$ and $[\alpha]_s^u = (\alpha_{t'})_{t'=s}^u$ is the control parameter trajectory and $[\alpha_t]$ is the trajectory which $\alpha_{t'} = \alpha_t$ $(s \leq t' \leq u)$. τ_B is the relaxation time of the baths. Similarly, in the Born-Markov approximation (BM), when going from

$$\frac{d\rho^{I,\chi}(t)}{dt} = -\int_0^t du\, \mathrm{Tr}_B\Big\{[H_{\mathrm{int}}^I(t), [H_{\mathrm{int}}^I(u), \rho^{I,\chi}(t)\rho_B(\alpha_t)]_\chi]_\chi\Big\}, \tag{D.2}$$

to (2.64), we used

$$\int_0^t du\, G([\alpha]_u^t; u, t; t) \approx \int_{-\infty}^t du\, G([\alpha_t]; u, t; t). \tag{D.3}$$

Considering the corrections of the above approximations, the QME is given by

$$\frac{d|\rho(t)\rangle\rangle}{dt} = \mathcal{K}(t)|\rho(t)\rangle\rangle, \tag{D.4}$$

$$\mathcal{K}(t) = \hat{K}(\alpha_t) + \hat{K}^{(1)}(t),\ \hat{K}^{(1)}(t) = \mathcal{O}(\Gamma\omega\tau_X), \tag{D.5}$$

with $\omega = 2\pi/\tau$ and $\tau_X = \tau_{\mathrm{CG}}$ for CGA; $\tau_X = \tau_B$ for BM. $\hat{K}^{(1)}(t)$ corresponds to $\boldsymbol{K}_{[1]}^{(1)}(t)$ of Appendix F. The discussions of § 3.4 are correct after replacing $K(\alpha_t) \to \mathcal{K}(t)$, $\mathcal{R}(\alpha_t) \to \bar{\mathcal{R}}(t)$ and $\rho_0(\alpha_t) \to \bar{\rho}_0(t)$. Here, $\bar{\rho}_0(t)$ and $\bar{\mathcal{R}}(t)$ are defined by $\mathcal{K}(t)|\bar{\rho}_0(t)\rangle\rangle = 0$ and $\bar{\mathcal{R}}(t)\mathcal{K}(t) = 1 - |\bar{\rho}_0(t)\rangle\rangle\langle\langle 1|$, respectively. (3.33) is corrected to

$$|\bar{\delta}\rho_{(\mathrm{ss})}(t)\rangle\rangle = \sum_{n=1}^{\infty}\Big[\bar{\mathcal{R}}(t)\frac{d}{dt}\Big]^n|\bar{\rho}_0(t)\rangle\rangle \equiv \sum_{n=1}^{\infty}|\bar{\rho}^{(n)}(t)\rangle\rangle, \tag{D.6}$$

with $\bar{\delta}\rho(t) \stackrel{\mathrm{def}}{=} \rho(t) - \bar{\rho}_0(t)$. The corrections are given by

$$\bar{\rho}_0 = \rho_0[1 + \mathcal{O}(\omega\tau_X)],\ \bar{\mathcal{R}} = \mathcal{R}[1 + \mathcal{O}(\omega\tau_X)], \tag{D.7}$$

and

$$\bar{\rho}^{(n)}(t) - \rho^{(n)}(t) = \mathcal{O}\Big((\frac{\omega}{\Gamma})^n\omega\tau_X\Big). \tag{D.8}$$

Next, we consider the reasonable range of n of $\rho^{(n)}(t)$. Because $\rho^{(n)}(t) = \mathcal{O}(\frac{\omega}{\Gamma})^n$ and $\bar{\rho}_0(t) - \rho_0(\alpha_t) = \mathcal{O}(\omega\tau_X)$, the reasonable range is $n \leq n_{\max}$, where $n_{\max}$ is determined by

$$(\frac{\omega}{\Gamma})^{n_{\max}+1} < \omega\tau_X \ll (\frac{\omega}{\Gamma})^{n_{\max}}. \tag{D.9}$$

Let us consider that reasonable concrete values of the parameters in the model of § 4.1: $\omega = 10^p$ MHz, $\Gamma = 10$ μeV=0.116 K, $1/\Gamma = 65.8$ ps, $\tau_{\mathrm{CG}} = 1$ ps, and $\tau_B = 0.1$ ps. These values lead to

$$\omega\tau_{\mathrm{CG}} = 10^{-6+p},\ \omega\tau_B = 10^{-7+p},\ \frac{\omega}{\Gamma} = 10^{-4.18+p}, \tag{D.10}$$

and $n_{\max} = [\tilde{n}_{\max}]$ with

$$\tilde{n}_{\max} = \frac{-6+p}{-4.18+p}\ (\mathrm{CGA}),\ \frac{-7+p}{-4.18+p}\ (\mathrm{BM}). \tag{D.11}$$

Here, $[n]$ means the biggest integer below n. At $p = 0$, $\tilde{n}_{\max} = 1.44$ (CGA), 1.67 (BM) and at $p = 3$, $\tilde{n}_{\max} = 2.54$ (CGA), 3.39 (BM). The larger the non-adiabaticity ($\frac{\omega}{\Gamma}$), the larger $n_{\max}$ becomes.

Appendix E

Proof of (3.52)

First, using (3.22) and (3.16), we obtain

$$\langle\langle 1|W^{O_\mu}(\alpha)\mathcal{R}(\alpha)\hat{K}(\alpha) = \langle\langle 1|W^{O_\mu}(\alpha) - \lambda_0^{O_\mu}(\alpha)\langle\langle 1|. \tag{E.1}$$

Next, $\langle\langle l_0(\alpha)| = \langle\langle 1|$, $\lambda_0(\alpha) = 0$, and (3.4) and (3.14) lead to

$$\langle\langle l_0^{O_\mu}(\alpha)|\hat{K}(\alpha) = \lambda_0^{O_\mu}(\alpha)\langle\langle 1| - \langle\langle 1|W^{O_\mu}(\alpha). \tag{E.2}$$

Hence, we obtain

$$\left[\langle\langle 1|W^{O_\mu}(\alpha)\mathcal{R}(\alpha) + \langle\langle l_0^{O_\mu}(\alpha)|\right]\hat{K}(\alpha) = 0, \tag{E.3}$$

and it leads to (3.52). To prove (3.52) only (3.22) is required and $\hat{K}(\alpha)\mathcal{R}(\alpha) = 1 - |\rho_0(\alpha)\rangle\rangle\langle\langle 1|$ is not necessary. Additionally, the pseudo-inverse of the GME approach (3.86) satisfies

$$\sum_j R_{ij}K_{jk}^{(0)} = \delta_{ik} - p_i^{(0)} \neq \sum_j K_{ij}^{(0)}R_{jk}, \tag{E.4}$$

which corresponds to our

$$\mathcal{R}(\alpha)\hat{K}(\alpha) = 1 - |\rho_0(\alpha)\rangle\rangle\langle\langle 1| \neq \hat{K}(\alpha)\mathcal{R}(\alpha). \tag{E.5}$$

(3.52) is shown also as follows. (3.22) and $\langle\langle 1|\hat{K}(\alpha) = 0$ lead to $\hat{K}(\alpha)\mathcal{R}(\alpha)\hat{K}(\alpha) = \hat{K}(\alpha)$, which implies

$$\hat{K}(\alpha)\mathcal{R}(\alpha) \;=\; 1 - |\sigma(\alpha)\rangle\rangle\langle\langle 1|,\;\; \langle\langle 1|\sigma(\alpha)\rangle\rangle = 1. \tag{E.6}$$

Applying $\langle\langle 1|$ to (3.22), we obtain $\langle\langle 1|\mathcal{R}(\alpha)\hat{K}(\alpha) = 0$, which is equivalent to

$$\langle\langle 1|\mathcal{R}(\alpha) \;=\; \mathcal{C}(\alpha)\langle\langle 1|. \tag{E.7}$$

By the way, differentiating (3.4) for $n = 0$ by $i\chi_{O_\mu}$, we obtain

$$\langle\langle l_0^{O_\mu}(\alpha)|\hat{K}(\alpha) + \langle\langle 1|\hat{K}^{O_\mu}(\alpha) \;=\; \langle\langle 1|\lambda_0^{O_\mu}(\alpha). \tag{E.8}$$

Applying $\mathcal{R}(\alpha)$ to this equation and using (E.6) and (E.7), we obtain[19, 84]

$$\langle\langle l_0^{O_\mu}(\alpha)| \;=\; -\langle\langle 1|\hat{K}^{O_\mu}(\alpha)\mathcal{R}(\alpha) + c^{O_\mu}(\alpha)\langle\langle 1|, \tag{E.9}$$

$$c^{O_\mu}(\alpha) \;=\; \mathcal{C}(\alpha)\lambda_0^{O_\mu}(\alpha) + \langle\langle l_0^{O_\mu}(\alpha)|\sigma(\alpha)\rangle\rangle. \tag{E.10}$$

(E.9) becomes (3.52) because of (3.14). Particularly, Yuge[84] used

$$\mathcal{R}(\alpha) = -\lim_{s\to\infty}\int_0^s dt\, e^{\hat{K}(\alpha)t}(1-|\rho_0(\alpha)\rangle\rangle\langle\langle 1|), \tag{E.11}$$

which satisfies (E.6) and (E.7) with $\sigma(\alpha) = \rho_0(\alpha)$, $\mathcal{C}(\alpha) = 0$ and (3.22) (in Ref.[84], $\mathcal{C}(\alpha)$ was incorrectly set to -1).

Appendix F

Generalized mater equation and frequency-expansion

The first half of this chapter is based on [33]. The GME is

$$\frac{d}{dt}\boldsymbol{p}(t) = \int_{-\infty}^{t} dt'\, \boldsymbol{W}(t,t')\boldsymbol{p}(t'), \tag{F.1}$$

where $\boldsymbol{p} = {}^t(p_1, p_2, \cdots, p_n)$. $\boldsymbol{W}(t,t')$ functionally depends on α_t. We expand $\boldsymbol{p}$ and $\boldsymbol{W}$ by the modulation frequency ω of the control parameters:

$$\boldsymbol{p}(t) = \sum_{k=0}^{\infty} \boldsymbol{p}^{(k)}(t),\ \boldsymbol{W}(t,t') = \sum_{k=0}^{\infty} \boldsymbol{W}^{(k)}(t; t-t'). \tag{F.2}$$

$\boldsymbol{p}^{(k)}$ and $\boldsymbol{W}^{(k)}$ are proportional to ω^k. In general, $\boldsymbol{p}(t)$ should contain a term which exponentially damps as $e^{-\Gamma\cdot(t-t_0)}$. Here, Γ is the coupling strength between the system and the baths. However, this method suppose $t_0 \to -\infty$. The RHS of (F.1) becomes

$$\begin{aligned}
\int_{-\infty}^{t} dt'\, \boldsymbol{W}(t,t')\boldsymbol{p}(t') &= \sum_{p,q} \int_{-\infty}^{t} dt'\, \boldsymbol{W}^{(p)}(t; t-t')\boldsymbol{p}^{(q)}(t') \\
&= \sum_{p,q,k} \int_{-\infty}^{t} dt'\, \boldsymbol{W}^{(p)}(t; t-t') \frac{(-1)^k (t-t')^k}{k!} \frac{d^k \boldsymbol{p}^{(q)}(t)}{dt^k} \\
&= \sum_{p,q,k} \frac{1}{k!} \frac{\partial^k}{\partial z^k}\Big|_{z=0} \int_{-\infty}^{t} dt'\, \boldsymbol{W}^{(p)}(t; t-t') e^{-z(t-t')} \frac{d^k \boldsymbol{p}^{(q)}(t)}{dt^k} \\
&\equiv \sum_{p,q,k} \frac{1}{k!} \partial^k \boldsymbol{K}^{(p)}(t) \frac{d^k \boldsymbol{p}^{(q)}(t)}{dt^k}.
\end{aligned} \tag{F.3}$$

Then, (F.1) becomes

$$\frac{d}{dt}\boldsymbol{p}^{(n)}(t) = \sum_{p,q,k}^{(n)} \frac{1}{k!} \partial^k \boldsymbol{K}^{(p)}(t) \frac{d^k \boldsymbol{p}^{(q)}(t)}{dt^k}. \tag{F.4}$$

$\sum_{p,q,k}^{(n)}$ is the summation over terms which have the same order with the LHS. How to count the time derivatives of $\boldsymbol{p}$ in this expansion depends on the considered frequency regime. In this chapter, we consider the regime $\omega \lesssim \Gamma$, for which the system quickly relaxes to an oscillatory steady state with the frequency of α_t; i.e., each time

derivative introduces one power in ω. Then,

$$n+1=p+q+k, \tag{F.5}$$

holds.

Now, we expand $\boldsymbol{p}^{(q)}$ and $\boldsymbol{K}^{(p)}(t)$ by Γ :

$$\boldsymbol{K}^{(p)}(t)=\sum_{j_K=1}^{\infty}\boldsymbol{K}^{(p)}_{[j_K]}(t),\ \boldsymbol{p}^{(k)}(t)=\sum_{j_p^{(k)}=-k}^{\infty}\boldsymbol{p}^{(k)}_{[j_p^{(k)}]}(t). \tag{F.6}$$

$_{[j]}$ indicates terms of order Γ^j. This matching requires the expansion for $\boldsymbol{p}^{(k)}(t)$ to start from Γ^{-k}. The matching condition for Γ is

$$j_p^{(n)}=j_K+j_p^{(q)}. \tag{F.7}$$

In the following, we consider the Born approximation: $\boldsymbol{K}^{(p)}(t)=\boldsymbol{K}^{(p)}_{[1]}(t)$. Then, (F.7) becomes $j_p^{(n)}=1+j_p^{(q)}$. This can be rewritten as

$$n+1=q+j_n-j_q, \tag{F.8}$$

where $j_p^{(n)}=-n+j_n$ and $j_p^{(q)}=-q+j_q$. The above equation and (F.5) lead

$$p+k+j_q=j_n. \tag{F.9}$$

First, we consider $j_n=0$. Then, solution of (F.9) is only $(p,k,j_q)=(0,0,0)$. Then, we obtain

$$\frac{d\boldsymbol{p}^{(k)}_{[-k]}(t)}{dt}=\boldsymbol{K}^{(0)}_{[1]}(\alpha_t)\boldsymbol{p}^{(k+1)}_{[-(k+1)]}(t)\quad(k=0,1,\cdots). \tag{F.10}$$

Where, $\boldsymbol{K}^{(0)}_{[1]}(t)$ is function of only α_t. Because the LHS of (F.1) does not have terms of $\mathcal{O}(\omega^0)$, we get

$$0=\boldsymbol{K}^{(0)}_{[1]}(\alpha_t)\boldsymbol{p}^{(0)}_{[0]}. \tag{F.11}$$

This is the definition of the instantaneous steady state.

Reference[33] considered only the solutions of $j_n=0$. However, the solutions of $j_n>0$ should also be considered. We consider $j_n=1$. Then, the solutions of (F.9) are $(p,k,j_q)=(1,0,0)$, $(0,1,0)$, $(0,0,1)$:

$$\frac{d\boldsymbol{p}^{(k)}_{[-k+1]}(t)}{dt}=\boldsymbol{K}^{(1)}_{[1]}(t)\boldsymbol{p}^{(k)}_{[-k]}(t)+\partial\boldsymbol{K}^{(0)}_{[1]}(\alpha_t)\frac{d\boldsymbol{p}^{(k)}_{[-k]}(t)}{dt}+\boldsymbol{K}^{(0)}_{[1]}(\alpha_t)\boldsymbol{p}^{(k+1)}_{[-k]}(t). \tag{F.12}$$

Here,

$$\boldsymbol{p}^{(k)}_{[-k]}(t)=\mathcal{O}(\frac{\omega^k}{\Gamma^k}),\ \frac{d\boldsymbol{p}^{(k)}_{[-k]}(t)}{dt}=\mathcal{O}(\frac{\omega^{k+1}}{\Gamma^k}), \tag{F.13}$$

and

$$\boldsymbol{K}^{(1)}_{[1]}(t)=\mathcal{O}(\Gamma\omega\tau_B),\ \partial\boldsymbol{K}^{(0)}_{[1]}(\alpha_t)=\mathcal{O}(\Gamma\tau_B), \tag{F.14}$$

hold. τ_B is the relaxation time of the baths. Then, we obtain

$$\boldsymbol{K}^{(1)}_{[1]}(t)\boldsymbol{p}^{(k)}_{[-k]}(t) = \mathcal{O}(\frac{\omega^{k+1}}{\Gamma^{k-1}}\tau_B),\ \partial\boldsymbol{K}^{(0)}_{[1]}(\alpha_t)\frac{d\boldsymbol{p}^{(k)}_{[-k]}(t)}{dt} = \mathcal{O}(\frac{\omega^{k+1}}{\Gamma^{k-1}}\tau_B). \tag{F.15}$$

(F.12) leads

$$\boldsymbol{p}^{(k+1)}_{[-k]}(t) \ = \ \boldsymbol{R}(\alpha_t)\frac{d\boldsymbol{p}^{(k)}_{[-k+1]}(t)}{dt} - \boldsymbol{R}(\alpha_t)\Big[\boldsymbol{K}^{(1)}_{[1]}(t)\boldsymbol{p}^{(k)}_{[-k]}(t) + \partial\boldsymbol{K}^{(0)}_{[1]}(\alpha_t)\frac{d\boldsymbol{p}^{(k)}_{[-k]}(t)}{dt}\Big]. \tag{F.16}$$

Here, $\boldsymbol{R}(\alpha)$ is the pseudo-inverse of $\boldsymbol{K}^{(0)}_{[1]}(\alpha)$:

$$\boldsymbol{R}(\alpha)\boldsymbol{K}^{(0)}_{[1]}(\alpha) \ = \ 1 - \boldsymbol{p}^{(0)}_{[0]}(\alpha)e,\ e = (1, \cdots, 1). \tag{F.17}$$

(F.16) for $k = 0$ leads

$$\boldsymbol{p}^{(1)}_{[0]}(t) \ = \ \mathcal{O}(\omega\tau_B). \tag{F.18}$$

Here, we used $\boldsymbol{p}^{(0)}_{[1]}(t) = 0$. Then, considering $\boldsymbol{p}^{(k)}_{[-k]}$ smaller than $\mathcal{O}(\omega\tau_B)$ is meaningless. This result (F.18) is the same order with that discussed in Appendix D.

Under the Born approximation, the difference between the QME and the GME is

$$\sum_{k=0}^{\infty}\sum_{j_p^{(k)}=-k+1}^{\infty}\boldsymbol{p}^{(k)}_{[j_p^{(k)}]}(t) = \boldsymbol{p}^{(1)}_{[0]}(t) + \cdots. \tag{F.19}$$

The origin of this is the non-Markovian property of the GME.

Appendix G

Energy current operator

Similar to (2.119), we introduce

$$[R^\dagger_{b,\mu}](\Omega_b) = \sum_{n,m,r,s} \delta_{\Omega_{b,mn},\Omega_b} |E_{b,n},r\rangle\langle E_{b,n},r|R^\dagger_{b,\mu}|E_{b,m},s\rangle\langle E_{b,m},s|, \tag{G.1}$$

with $\Omega_{b,mn} = E_{b,m} - E_{b,n}$ and $H_b|E_{b,n},r\rangle = E_{b,n}|E_{b,n},r\rangle$. r denotes the label of the degeneracy. Ω_b is one of the elements of $\{\Omega_{b,mn}|\ \langle E_{b,n},r|R^\dagger_{b\mu}|E_{b,m},s\rangle \neq 0\ \ ^\exists \mu\}$. We set $\{O_\mu\} = \{H_b\}_b$. Then,

$$R^{I\dagger}_{b,\mu,-2\chi}(u) = \sum_{\Omega_b} [R^\dagger_{b,\mu}](\Omega_b) e^{-i\Omega_b u + i\chi_{H_b}\Omega_b}, \tag{G.2}$$

holds. Using this, we obtain

$$\Phi^\chi_{b,\alpha\beta}(\Omega) = 2\pi \sum_{\Omega_b} \delta(\Omega - \Omega_b) e^{i\chi_{H_b}\Omega_b} \mathrm{Tr}_b(\rho_b [R^\dagger_{b,\mu}](\Omega_b) R_{b,\nu}). \tag{G.3}$$

This means

$$\Phi^\chi_{b,\mu\nu}(\Omega) = e^{i\chi_{H_b}\Omega} \Phi_{b,\mu\nu}(\Omega). \tag{G.4}$$

Using this, we obtain

$$W^{H_b}(\alpha) = \sum_\omega \sum_{\mu,\nu} \omega \Phi_{b,\mu\nu}(\omega) [s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega), \tag{G.5}$$

for the RWA.

Using (2.153), we obtain

$$\begin{aligned} w^{H_b}(\alpha) &= -\sum_\omega \sum_{\mu,\nu} \Big[\Phi_{b,\mu\nu}(\omega) [s_{b\mu}(\omega)]^\dagger H_S s_{b\nu}(\omega) \\ &\quad -\frac{1}{2}\Phi_{b,\mu\nu}(\omega) H_S [s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega) - \frac{1}{2}\Phi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega) H_S \Big] \\ &= -\sum_\omega \sum_{\mu,\nu} \Phi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^\dagger [H_S, s_{b\nu}(\omega)] \\ &= \sum_\omega \sum_{\mu,\nu} \omega \Phi_{b,\mu\nu}(\omega)[s_{b\mu}(\omega)]^\dagger s_{b\nu}(\omega). \end{aligned} \tag{G.6}$$

Then,

$$W^{H_b}(\alpha) = w^{H_b}(\alpha), \tag{G.7}$$

holds.

Appendix H

Derivative of the von Neumann entropy

We show that

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

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

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

Using (7.67), the second term of the RHS of the above equation becomes

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

Then, we obtain (H.1).

Appendix I

Proof of (7.66)

For an arbitrary operator X,

$$\frac{1}{1+\delta X} = (1+\delta X)^{-1} = 1 - \delta X + \delta^2 X^2 - \delta^3 X^3 + \cdots , \tag{I.1}$$

holds if the absolute value of a real number δ is sufficiently small. Using this equation,

$$\begin{aligned}\frac{1}{A+\delta B} &= [A(1+\delta A^{-1}B)]^{-1} = (1+\delta A^{-1}B)^{-1}A^{-1} \\ &= \frac{1}{A} - \delta\frac{1}{A}B\frac{1}{A} + \delta^2\frac{1}{A}B\frac{1}{A}B\frac{1}{A} - \delta^3\frac{1}{A}B\frac{1}{A}B\frac{1}{A}B\frac{1}{A} + \cdots ,\end{aligned} \tag{I.2}$$

holds for an arbitrary operator A which has A^{-1}. Here, we used (I.1) for $X = A^{-1}B$. For an arbitrary operator Y which has Y^{-1},

$$\int_0^a ds\, \frac{1}{Y+s} = \ln(Y+a) - \ln Y, \tag{I.3}$$

holds for a real number a. Using this equation for $Y = A$ and $Y = A+\delta B$, we obtain

$$\begin{aligned}\ln(A+\delta B) &= \ln A + \ln(A+\delta B+a) - \ln(A+a) \\ &\quad + \int_0^a ds\, \Big(\frac{1}{A+s} - \frac{1}{A+\delta B+s}\Big).\end{aligned} \tag{I.4}$$

Using this equation and (I.2), we get

$$\begin{aligned}\ln(A+\delta B) &= \ln A + \ln(A+\delta B+a) - \ln(A+a) \\ &\quad + \int_0^a ds\, \Big(\delta\frac{1}{A+s}B\frac{1}{A+s} - \delta^2\frac{1}{A+s}B\frac{1}{A+s}B\frac{1}{A+s} + \cdots\Big).\end{aligned} \tag{I.5}$$

Because the second and third terms of the RHS are

$$\ln(A+B+a) - \ln(A+a) = \ln\Big(1+\frac{A+B}{a}\Big) - \ln\Big(1+\frac{A}{a}\Big) = \mathcal{O}(\frac{1}{a}), \tag{I.6}$$

we obtain

$$\ln(A+\delta B) = \ln A + \int_0^\infty ds\, \Big(\delta\frac{1}{A+s}B\frac{1}{A+s} - \delta^2\frac{1}{A+s}B\frac{1}{A+s}B\frac{1}{A+s} + \cdots\Big), \tag{I.7}$$

for $a \to \infty$. The above equation is (7.66).

We show (I.6). Substituting $A = 1$ to (I.5), we get

$$\begin{aligned}
\ln(1+\delta B) &= \ln(1+\delta B+a) - \ln(1+a) \\
&\quad + \int_0^a ds \left(\delta \frac{1}{(1+s)^2} B - \delta^2 \frac{1}{(1+s)^3} B^2 + \cdots\right) \\
&= \ln(1+\delta B+a) - \ln(1+a) \\
&\quad + \int_0^a ds \left(\delta \frac{1}{(1+s)^2} B - \delta^2 \frac{1}{(1+s)^3} B^2 + \cdots\right) \\
&= \ln\left(1+\frac{\delta B}{a+1}\right) + \sum_{n=1}^{\infty} \frac{(-1)^{n-1}}{n} \delta^n B^n \left(1 - \frac{1}{(1+a)^n}\right). \qquad \text{(I.8)}
\end{aligned}$$

Using this equation for $a \to \infty$, we have

$$\ln(1+\delta B) = \sum_{n=1}^{\infty} \frac{(-1)^{n-1}}{n} \delta^n B^n, \qquad \text{(I.9)}$$

which leads to (I.6).

Appendix J

Definition of entropy production of the Markov jump process

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

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

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

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

The probability to find the system in a state n is $p_n(t)$ and it obeys the master equation (8.1). We suppose the trajectory of the control $\hat{\alpha} = \big(\alpha(t)\big)_{t=0}^{\tau}$ is smooth. Now we introduce

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

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

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

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

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

The integral over all the paths is defined by

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

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

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

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

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

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

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

In particular, the average is given by

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

where

$$\sigma_{nm}^{\rm C}(\alpha) \stackrel{\rm def}{=} K_{nm}(\alpha)\theta_{nm}(\alpha) = -K_{nm}(\alpha)\ln\frac{K_{nm}(\alpha)}{K_{mn}(\alpha)}. \tag{J.11}$$

According to Ref.[21], for a quasistatic operation,

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

holds where

$$\sigma_{\rm ex}^{\rm C} \stackrel{\rm def}{=} \sigma^{\rm C} - \int_0^\tau dt\,\sum_{n,m}\sigma_{nm}^{\rm C}(\alpha_t)p_m^{\rm ss}(\alpha_t), \tag{J.13}$$

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

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