Fluctuation Relations For Adiabatic Quantum Pumping

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We derive an extended fluctuation relation for an open quantum system coupled with two reservoirs under adiabatic one-cycle modulation. We confirm that the geometrical phase caused by the Berry-Sinitsyn-Nemenman curvature in the parameter space generates non-Gaussian fluctuations. This non-Gaussianity is enhanced for the instantaneous fluctuation relation when the bias between the two reservoirs disappears.

I. INTRODUCTION

Adiabatic pumping is a process where an average current is generated even in the absence of an average bias under slow and periodic modulation of multiple parameters of the system. The theory of adiabatic pumping was first proposed by Thouless in isolated quantum systems [11,2]. He showed that charges can be transported by applying a time-periodic potential to one-dimensional isolated quantum systems under a periodic boundary condition. He also clarified that the charge transport in this system is essentially induced by a Berry-phase-like quantity in the parameter space [2,4] before Berry proposed the Berry phase [3]. This phenomenon has been observed experimentally in various processes such as charge transport [5–12] and spin pumping [13].

Later, Brower extended the Thouless pumping to open quantum systems [14]. It was then recognized that the essence of Thouless pumping or geometrical pumping can be described by a classical master equation in which the Berry-Sinitsyn-Nemenman (BSN) phase is the generator of the pumping current [15,16]. There are various papers on geometrical pumping processes in terms of scattering theory [14,17–23], classical master equations [15,16,24,31] and quantum master equations [32,37]. Non-adiabatic pumping processes have also been studied because the pumping current becomes zero in the adiabatic (i.e., zero frequency) limit [38,39].

We discovered the existence of path-dependent excess entropy production induced by the BSN curvature [40–42]. The existence of the path-dependent entropy in systems under cyclic modulation implies that the direct extension of equilibrium thermodynamics to nonequilibrium processes is not possible, at least for such systems. Similarly, the geometrical phase effect plays an important role even in heat engines [43]. Thus, understanding geometrical pumping is important for both applied and fundamental physics.

In nonequilibrium processes, in addition to the expectation values of physical quantities such as electric and heat currents, their fluctuations are also important. Historically, the relationship between fluctuations and responses from base states has been extensively studied, resulting in the Green-Kubo formula for the linear response, the fluctuation-dissipation relation (FDR), Onsager’s reciprocity relation etc. [44]. Furthermore, in the 1990s, the fluctuation theorem (FT) [45–53] was discovered as a relation which holds even in far-from-equilibrium situations. The FT expresses the relative probabilities of typical and rare events such as positive and negative entropy production.

Let us consider an open system in contact with multiple external reservoirs, which is in a nonequilibrium steady state. In this situation, the probability distribution $P_r(J)$ of the current $J$ in time interval $\tau$ satisfies the steady fluctuation theorem [53,56]

$$\lim_{\tau \to \infty} \frac{1}{\tau} \ln \frac{P_r(J)}{P_r(-J)} = A^{st} \tau,$$  \hspace{1cm} (1)

where $A^{st}$ is a steady affinity. For example, when we consider a heat flow, the affinity is given by $A^{st} = \beta_R - \beta_L$, where $\beta_L$ and $\beta_R$ are the inverse temperatures of the left and the right reservoirs, respectively. Moreover, the FT recovers the Green-Kubo formula, the FDR, Onsager’s reciprocity relation and the other nonlinear relations [54]. The FT in Eq. (1) is a direct consequence of Gaussian fluctuations, since the Gaussian form $P_r(J) \sim \exp \left[ -\frac{1}{2A^{st}} (J - \langle J \rangle)^2 \right]$ with the average current $\langle J \rangle$ satisfies Eq. (1).

Therefore, systems by non-Gaussian noises do not satisfy the conventional fluctuation theorem [57,58]. Similary, Ren et. al. indicated that the fluctuation theorem is violated in adiabatic pumping because of the existence of the geometrical phase [59]. Watanabe and Hayakawa [60] analyzed the spin-boson model and verified the violation of the FT in the pumped system. Nevertheless, they could not get a concise form of an extended fluctuation theorem for geometric pumping.
processes. We also need to know relations among the cumulants of the current. If we can construct the extended fluctuation relation between $P_{\tau}(J)$ and $P_{\tau}(-J)$, we can derive the explicit expressions for all cumulants.

In this paper, we derive two types of fluctuation relations for adiabatic pumping processes by using the generalized quantum master equation with the aid of fluctuation theorems (FCTs) [51]. By using these expressions, we also derive nonequilibrium relations corresponding to the FDR and other key results. We have confirmed that the geometrical phase generates non-Gaussian fluctuations [57, 58], and thus, systems under cyclic modulation do not satisfy the fluctuation theorem.

The organization of this paper is as follows. In Sec. II, we introduce the adiabatic approximation and show the general form of the cumulants for the pumping current. In Sec. III, we explain the method used in this paper, the FCS enables us to obtain the probability distribution of the dimensionless current from the system to a reservoir during the time interval $\tau$. When we perform a projective measurement of a dimensionless observable $Q$ (e.g., the dimensionless Hamiltonian $H_R$, the number operator $N_R$, etc.) in the right reservoir at time zero and time $\tau$, the corresponding outcomes are denoted by $Q_0$ and $Q_\tau$, respectively. We assume $[Q, H_R] := QH_R - H_R Q = 0$. The generating function $Z_\tau(\chi)$ for the probability distribution function $P_\tau(q)$ of the transfer $q := Q_\tau - Q_0$ from the target system to the right reservoir in the time interval $\tau$ is given by

$$Z_\tau(\chi) = \int_{-\infty}^{\infty} dq P_\tau(q) e^{i\chi q},$$

where $\chi$ is called the counting field. We introduce the counting field only between the system and the right reservoir as shown in Fig. 1. Note that Eq. (2) can be rewritten as $Z_\tau(\chi) = \text{Tr}[\rho_{\text{tot}}(\tau, \chi)]$ (its derivation is given in Appendix A), where $\rho_{\text{tot}}(t, \chi)$ at time $t$ is the generalized density matrix for the total system defined as $\rho_{\text{tot}}(t, \chi) := U(t, \chi) \rho_{\text{tot}}(0) U^\dagger(t, -\chi)$, $U(t, \chi) := e^{i\chi Q/\hbar}U(t)e^{-i\chi Q/\hbar}$ with $U(t) := \exp[-i \int_0^t dt' \hat{H}_{\text{tot}}(\alpha(t))]$. $\exp_\tau$ stands for the time-ordered exponential form. Furthermore, we introduce the reduced generalized density matrix for the target system $\rho(t, \chi) := \text{Tr}_R[\rho_{\text{tot}}(t, \chi)]$, where $\text{Tr}_R$ is the trace of the reservoir $\nu$. By using $\rho(t, \chi)$, we can calculate the generating function as $Z_\tau(\chi) = \ln \text{Tr}_R[\rho(\tau, \chi)]$, where $\text{Tr}_R$ is the trace in the target system. $\rho(t, \chi)$ satisfies the generalized quantum master equation (GQME) [51]

$$\partial_t \rho(t, \chi) = K(\alpha(t), \chi) \rho(t, \chi),$$

where $K(\alpha(t), \chi)$ is the Liouvillian with the counting field $\chi$. The Liouvillian depends on the approximations used. The explicit form of $K(\alpha(t), \chi)$ is given in Appendix B. In this paper, we use the Lindblad form [66], i.e., the Born-Markov approximation with the rotating wave approximation (RWA). Within the Lindblad form, the time evolution of the diagonal and off-diagonal elements of $\rho(t, \chi)$ is decoupled. In particular, the diagonal components satisfy the generalized classical master equation (GCME)

$$\partial_t [\rho(t, \chi)] = K(\alpha(t), \chi) [\rho(t, \chi)],$$

where $[\rho(t, \chi)] := (\rho_0(t, \chi), \ldots, \rho_n(t, \chi))^T$ with $\rho_k(t, \chi) := \langle k|\rho(t, \chi)|k \rangle$ for $1 \leq k \leq n$ and $|k \rangle$ is the energy eigenstate of $H_S$. Elements of the transition matrix $K(\alpha(t), \chi)$ are given in Eq. [B11]. It should be noted that $K(\alpha(t), \chi)$ in the GQME in Eq. (3) is

II. GENERAL FRAMEWORK

![Figure 1. A schematic of the total system which consists of the target system and the left and right reservoirs.](image)

In this paper, we consider a total system in which the target system $S$ interacts with two reservoirs $L$ and $R$ (Fig. 1). The Hamiltonian of the total system contains the system Hamiltonians $H_S$, the reservoir Hamiltonians $H_\nu$ ($\nu = L, R$) and the interaction Hamiltonians $H_{SL}$. The total system is also characterized by a set of control parameters $\alpha = \{\alpha_S, \alpha_L, \alpha_R\}_{\nu = L, R}$. Here, $\alpha_S$ and $\alpha_S$ appear in $H_S$ and $H_{SL}$, respectively, while $\alpha_L$ appears through a parameter in the $\nu$-th reservoir. We assume periodicity of the parameters: $\alpha(t) = \alpha(t + \tau)$, where $\tau$ is the period of the modulation.
not always in the Lindblad form if the transport is coupled with resonances [31]. In this case, we cannot reduce the GQME to the GCME as in Eq. (1). If the system Hamiltonian does not satisfy the commutation relation [33] (e.g. the target system consists of multiple quantum dots that interact each other [39]), we cannot use the Lindblad form [36] and should use the Born-Markov form [31], in which the diagonal and off-diagonal parts are not independent of each other.

Now, let us introduce the angular frequency and the phase of parameter modulation $\Omega := 2 \pi/\tau$ and $\theta := \Omega(t - t_0)$, respectively, where $t_0$ is a time after which the effect of the initial conditions has become negligible and $\rho(t, \chi)$ becomes periodic. Then, we rewrite Eq. (1) as

$$d\theta |\rho(\theta, \chi)\rangle = e^{-i\hat{K}(\alpha(\theta), \chi)} |\rho(\theta, \chi)\rangle, \quad (5)$$

where $\epsilon := \Omega/\Gamma$, $\hat{K}(\alpha(t), \chi) := \Gamma^{-1} K(\alpha(t), \chi)$, and $\Gamma$ characterizes the strength of the interaction between the system and the two reservoirs. Therefore, we can replace $Z_r(\chi)$ by $Z_0(\chi)$. Because we are interested in adiabatic modulation, the parameter $\epsilon$ is assumed to satisfy $\epsilon \ll 1$.

### III. ADIABATIC PUMPING

In this section, we briefly review the method to obtain the cumulant-generating function for the current [59, 60, 62]. First, we adopt the adiabatic approximation

$$|\rho(\theta, \chi)\rangle \simeq \exp \left\{ \frac{1}{\epsilon} \int_0^\theta d\theta' [\lambda(\theta', \chi) - \epsilon \nu(\theta', \chi)] \right\} \times |r(\theta, \chi)\rangle |r(0, \chi)\rangle^{-1}, \quad (6)$$

where $\lambda(\theta, \chi)$ is the eigenvalue of $\hat{K}(\alpha(\theta), \chi)$, which reduces to zero in the limit $\chi \to 0$. Here we define $\nu(\theta, \chi) := \langle \langle \theta, \chi) | \partial_\theta |r(\theta, \chi)\rangle \rangle$, where $\langle \langle \theta, \chi) \rangle$ and $|r(\theta, \chi)\rangle$ are the left and right eigenvectors corresponding to $\lambda(\theta, \chi)$, respectively.

The cumulant generating function for the current $J := \Omega q/(2\pi \Gamma) = \epsilon q/2\pi$ is given by

$$G_c(\chi) = \frac{\epsilon}{2\pi} \ln Z_r(\chi) = \frac{\epsilon}{2\pi} \ln \langle 1 | \rho(2\pi, \chi) \rangle. \quad (7)$$

By using the adiabatic solution (7), we obtain

$$G_c(\chi) = \Lambda(\chi) - \epsilon V(\chi), \quad (8)$$

where

$$\Lambda(\chi) := \frac{1}{2\pi} \int_0^{2\pi} d\theta \lambda(\theta, \chi) \quad (9)$$

is the dynamical part and

$$V(\chi) := \frac{1}{2\pi} \int_0^{2\pi} d\theta \nu(\theta, \chi) = \frac{1}{2\pi} \int_S d\alpha_m d\alpha_n F(\alpha, \chi), \quad (10)$$

is the geometrical part. Here we define $F_{\alpha n}(\alpha, \chi) := \partial_{\alpha m} \langle \langle t(\alpha, \chi) | \phi(\alpha, \chi) \rangle \rangle$ and $S$ is the open surface enclosed by the contour $C$ of parameter control (see Fig. 2). The derivation of Eqs. (9) is given in Appendix C.

The n-th cumulant for the current $J$ can be expressed as

$$\langle J^n \rangle_c = \frac{\partial^n}{\partial (i\chi)^n} G_c(\chi) \bigg|_{\chi = 0}. \quad (11)$$

![Figure 2](image-url) - A schematic of a contour $C$ and the surface enclosed by $C$ in the parameter space spanned by $(\alpha_m, \alpha_n)$.

### IV. FLUCTUATION RELATIONS

This section is the main part of this paper. In this section, we present the general expressions for two types of fluctuation relations for adiabatic pumping processes. In subsection IV.A we discuss the cyclic fluctuation relation and in subsection IV.B we give the general expression for the instantaneous fluctuation relation.

#### A. Cyclic fluctuation relation

Because of Eqs. (2) and (8) the probability distribution function $P_{\epsilon}(J)$ of the current $J$ under one-cycle modulation with the parameter $\epsilon$ is given by

$$P_{\epsilon}(J) = \frac{1}{\epsilon} \int_{-\infty}^{\infty} d\chi e^{-\frac{\epsilon}{2\pi} |\chi J - G_c(\chi)|} = \frac{1}{\epsilon} \int_{-\infty}^{\infty} d\chi e^{-\frac{\epsilon}{2\pi} |\chi J - \Lambda(\chi)| - V(\chi)}. \quad (12)$$

When we consider an adiabatic pumping process ($\epsilon \ll 1$), the contribution of $V(\chi)$ is small. By using the saddle point approximation, $P_{\epsilon}(J)$ can be evaluated as

$$P_{\epsilon}(J) \simeq \frac{1}{\sqrt{\epsilon \Lambda(\chi_c)(\epsilon J)^2}} e^{-\frac{\epsilon}{2\pi} |\langle J \rangle + \epsilon V(\chi_c(J))|}, \quad (13)$$
where we have introduced the large deviation function (LDF)

\[ I(J) := i\chi_c(J)J - \Lambda(\chi_c(J)), \]

where \( \chi_c(J) \) is the saddle point which satisfies \( \partial_\chi \Lambda(\chi) \bigg|_{\chi=\chi_c(J)} = J \) and \( \Lambda^2(\chi_c(J)) := \partial^2_\chi \Lambda(\chi) \bigg|_{\chi=\chi_c(J)} \). It is expected that \( I(J) \) satisfies the symmetry relation

\[ I(J) - I(-J) = -\mathcal{A}J \tag{15} \]

where \( \mathcal{A} \) is the dynamical affinity, which is determined by quantities of the left and the right reservoir. In fact, it was confirmed numerically that \( \mathcal{A} \) is given as

\[ \mathcal{A} = \ln \frac{\int_{0}^{2\pi} d\theta n^+_R(\theta)(1 \pm n^+_F(\theta))}{\int_{0}^{2\pi} d\theta n^+_F(\theta)(1 \pm n^+_R(\theta))} \tag{16} \]

in a system of fermions [62] or bosons (see Appendix F). Here \( n^+_F(\theta) \) is the Bose (+) and Fermi (−) distribution of the \( x \)-th reservoir, respectively. By using Eqs. (13) and (15), we obtain the fluctuation relation

\[ \epsilon \frac{\pi}{2\pi} \ln \frac{P_c(J)}{P_c(-J)} = AJ - \epsilon [V(\chi_c(J)) - V(\chi_c(-J))] - \frac{\epsilon}{4\pi} \ln \frac{\Lambda^2(\chi_c(J))}{\Lambda^2(\chi_c(-J))}, \tag{17} \]

This is one of our main results.

The second term on the right hand side of Eq. (17) stands for the geometrical phase contribution which is much smaller than the first term. When \( V(\chi_c(J)) = 0 \), Eq. (17) reduces to the steady fluctuation theorem in driven systems [62]. Thus, Eq. (17) can be regarded as an extension of the fluctuation theorem for the adiabatic pumping process. If the trajectory \( C \) of the parameter modulation is symmetric with respect to the parameters \( \alpha_n \) and \( \alpha_{n'} \), the average bias is zero, i.e. \( \mathcal{A} = 0 \) and \( V(\chi - \chi(\lambda)) = -V(\chi(\lambda)), \Lambda(\chi) = \Lambda(\lambda) \). In this case, Eq. (17) is reduced to

\[ \ln \frac{P_c(J)}{P_c(-J)} = -4\pi V(\chi_c(J)), \tag{18} \]

which can be expressed only by the geometrical phase. As will be shown in Sec. V, Eq. (18) contains contributions nonlinear in \( J \).

### B. Instantaneous fluctuation relation

In this subsection, let us consider the instantaneous fluctuation relation of our system. If the master equation (5) does not contain any singularities, the cumulant generating function can be written as

\[ G_c(\chi) = \frac{1}{2\pi} \int_{0}^{2\pi} d\theta g(\theta, \chi) = \lim_{N \to \infty} \frac{1}{N} \sum_{n=1}^{N} g_n(\chi), \tag{19} \]

where \( g(\theta, \chi) := \lambda(\theta, \chi) - \epsilon_v(\theta, \chi) \) is the instantaneous cumulant-generating function. Here we discretize \( \theta \) in the interval \([0, 2\pi]\) as in the last expression of Eq. (19), where we have introduced \( \theta_n := n \Delta \theta, \Delta \theta := 2\pi/N, \)

\[ g_n(\chi) := g(\theta_n, \chi), \lambda_n(\chi) := \lambda(\theta_n, \chi), v_n(\chi) := v(\theta_n, \chi). \]

From Eq. (19), the distribution of the current during one cycle can be decomposed into

\[ P_c(J) = \lim_{N \to \infty} N \int_{-\infty}^{\infty} \prod_{n=1}^{N} dJ_n \delta \left( J = \frac{1}{N} \sum_{n=1}^{N} J_n \right) \prod_{n=1}^{N} p_n(J_n), \tag{20} \]

where

\[ p_n(J_n) := \int_{-\infty}^{\infty} d\chi e^{-\frac{\pi}{\epsilon N} [(\chi_J - \chi_n(\chi))]}. \tag{21} \]

is the instantaneous distribution at of the current \( J_n \) at \( \theta = \theta_n \). (The derivation of Eqs. (20) and (21) is explained in Appendix F.) Here we assume \( (\epsilon N)^{-1} \gg 1 \). This means that \( (\epsilon N)^{-1} \) is enough large to relax the system to the instantaneous steady state.

By an argument parallel to that used in the previous subsection, the instantaneous distribution for the current \( J_n \) is given as

\[ p_n(J_n) \sim \frac{1}{(\epsilon N)^{\lambda_n(2)}(\chi_n(J_n))} e^{-\frac{\pi}{\epsilon N} (I_n(J_n) + v_n(\chi_n(J_n)))}, \tag{22} \]

where \( \lambda_n(2) := \partial^2_\chi \lambda_n(\chi) \bigg|_{\chi=\chi_n(J_n)} \) and we have introduced the instantaneous LDF

\[ I_n(J_n) := i\chi_n(J_n)J_n - \lambda_n(\chi_n(J_n)), \tag{23} \]

where \( \chi_n(J_n) \) satisfies \( \partial_\chi \lambda_n(\chi) \bigg|_{\chi=\chi_n(J_n)} = J_n \). Because the instantaneous eigenvalue \( \lambda_n(\chi) \) satisfies the Levitov-Lesovik-Gallavotti-Cohen (LLGC) symmetry

\[ \lambda_n(\chi) = \lambda_n(-\chi + i\mathcal{A}_n), \]

\( I_n(J_n) \) satisfies the symmetry relation

\[ I_n(J_n) - I_n(-J_n) = -\mathcal{A}_n J_n, \tag{24} \]

where \( \mathcal{A}_n \) is the instantaneous affinity, which is given by; for example, \( \mathcal{A}_n = \beta_R(\theta_n) - \beta_L(\theta_n) \) when we control the inverse temperatures \( \beta_L \) and \( \beta_R \) of the left and the right reservoirs.

From Eqs. (22) and (24), we obtain the instantaneous fluctuation relation

\[ \lim_{\epsilon N \to 0} \frac{1}{2\pi} \ln \frac{p_n(J_n)}{p_n(-J_n)} \sim \mathcal{A}_n J_n - \epsilon [v_n(\chi_n(J_n)) - v_n(\chi_n(-J_n))] \]

\[ - \frac{\epsilon N}{4\pi} \ln \frac{\lambda_n(2)(\chi_n(J_n))}{\lambda_n(2)(\chi_n(-J_n))}. \tag{25} \]
The second term on the right hand side of Eq. (25) expresses the geometrical phase effect at $\theta = \theta_n$, which is much smaller than the first term. If $A_n = 0$, the first and third terms vanish then the geometrical contribution becomes dominant. As will be shown in Sec. V the geometric contribution of Eq. (25) is a nonlinear function of $J_n$.

V. APPLICATION TO THE SPIN-BOSON SYSTEM

The results presented in the previous section can be used for an arbitrary adiabatic pumping process if the process can be described by the master equation Eq. (3) or Eq. (4). To know the explicit contribution of the geometric phase in the extended fluctuation relations such as Eqs. (17), (18) and (25), we need to know the details of the eigenstates and the eigenvalues of the operator $\hat{K}(\alpha(\theta), \chi)$. Here, we apply the general results of Sec. IV to the spin-boson model [63].

A. The spin-boson model

In the spin-boson model, the total Hamiltonian is given by $H_{\text{tot}}(\alpha) = H_S(\alpha_S) + \sum_{\nu=L,R} (H_\nu + H_{\text{SB}_\nu}(\alpha_{S\nu}))$. Each term is given by

$$H_S(\alpha_S) = \frac{\hbar \omega_0}{2} \sigma_z;$$

$$H_\nu = \sum_k \hbar \omega_{k,\nu} b_{k,\nu}^\dagger b_{k,\nu},$$

$$H_{\text{SB}_\nu}(\alpha_{S\nu}) = \hbar \sigma_z \otimes \sum k g_{k,\nu} (b_{k,\nu}^\dagger + b_{k,\nu}),$$

where $\hbar \omega_0$ is the energy gap between the two levels in the target system. $\sigma_z = |1\rangle \langle 1| - |0\rangle \langle 0|$ and $\sigma_x = |1\rangle \langle 0| + |0\rangle \langle 1|$ are Pauli operators, where $|1\rangle$ is the up state and $|0\rangle$ is the down state. $\omega_{k,\nu}$ is the angular frequency at wave number $k$ for the $\nu$-th reservoir and $b_{k,\nu}^\dagger (b_{k,\nu})$ is the boson annihilation (creation) operator for the $\nu$-th reservoir, respectively. Here $g_{k,\nu}$ is the coupling constant, which is related to the spectral density function $D_\nu(\omega) := 2\pi \sum_k \varrho_{k,\nu}^2 \delta(\omega - \omega_{k,\nu})$. For later analysis, we use the line-width $\Gamma_\nu = D_\nu(\omega)$ which is independent of $\omega$. We also introduce the dimensionless line-width $\gamma_\nu := \Gamma_\nu / \Gamma$, where $\Gamma = (\Gamma_L + \Gamma_R)/2$.

We assume that the bosonic reservoirs are always at equilibrium. Thus, the density matrix of the $\nu$-th reservoir is expressed as $\rho_{\nu}^0(\beta_\nu) = e^{-\beta_\nu H_\nu} / Z_{\nu}$ at the inverse temperature $\beta_\nu$, where $Z_\nu = Tr_\nu[e^{-\beta_\nu H_\nu}]$. Now, we can write the control parameters $\alpha = \{\alpha_S = \omega_0, \alpha_{S\nu} = \gamma_\nu, \alpha_\nu = \beta_\nu\}_{\nu=L,R}$ explicitly. In addition, we consider the observable $Q$ is given as $Q = (\hbar \omega_0)^{-1} H_R$. The reduced dynamics of the target system can be described by a Lindblad-type GQME, so the diagonal part of $\rho(\theta, \chi)$ is described by the GCME (5). The details of the GCME (5) are given in Appendix G.

B. Cyclic fluctuation relation for the spin-boson model

Let us calculate the right hand side of Eq. (17) for two types of modulations. In the first case, we control the temperature of the left and right reservoirs as

$$\hat{T}_L(\theta) := (\hbar \omega_0 \beta_L(\theta))^{-1} = \hat{T}_0 + \hat{T}_A \cos(\theta + \pi/4),$$

$$\hat{T}_R(\theta) := (\hbar \omega_0 \beta_R(\theta))^{-1} = \hat{T}_0 + \hat{T}_A \sin(\theta + \pi/4).$$

where $\hat{T}_0$ and $\hat{T}_A$ are the center and the amplitude of the dimensionless temperatures $T_L$ and $T_R$, respectively. For simplicity, we assume $\Gamma_L = \Gamma_R = \Gamma$. In the second case, we control the dimensionless line-width between the target system and the left reservoir and the energy level in the target system such that

$$\gamma_L(\theta) = \gamma_R + \gamma_A \cos(\theta),$$

$$\omega_0(\theta) := \beta \omega_0(\theta) = \omega_c + \omega_A \sin(\theta).$$

where $\gamma_A$ is the amplitude of the dimensionless line-width $\gamma_\nu$. $\omega_c$ and $\omega_A$ are the center and the amplitude of the dimensionless energy gap between two levels in the target system [2]. For simplicity, we assume $\beta_L = \beta_R = \beta$. The corresponding BSN curvatures $F_{mn}(\alpha)$ in Eq. (D2) are plotted in Fig. 4. In both cases, because the affinity satisfies $A = 0$, the geometrical phase effect $V(\chi_c(J))$ plays an important role as in Eq. (18).

1 Of course, the continuous control of the temperatures is not easy but possible as follows. (i) For example, an effective temperature is continuously changed in Ref. [63]. (ii) Since we consider an adiabatic process, it is possible to replace a reservoir by another reservoir having a different temperature and wait for the system to relax to a steady state. If we can repeat this process, we can change the temperatures at the reservoirs as in Eqs. (23) and (30).

2 It is easy to control $\gamma_L(\theta)$ and $\omega_0(\theta)$ in experiments [9][10].
By expanding the right hand side of Eq. (18) with respect to $J$, we obtain

$$\ln \frac{P_2(J)}{P_2(-J)} \simeq A_C[J + B_C J^3 + O(J^5)], \quad (33)$$

where $A_C := -4\pi \partial J V(\chi_C(J))|_{J=0}$ and $B_C := -2\pi \partial^2 J V(\chi_C(J))|_{J=0}/3A_C$ which depend on the contour $C$ of the parameter control. Now $C$ is determined by the set of parameters $\alpha = (\bar{T}_0, \bar{T}_A)$ or $(\omega_C, \omega_A, \gamma_R, \gamma_A)$. With aid of a numerical calculation for both cases, we obtain Figs. 5 and 6 which show that $B_C$ is not negligibly small. This implies the geometrical current or the BSN curvature generates non-Gaussian fluctuations. Our result is consistent with the previous results of Ref. [60].

C. Relations among cumulants

Let us discuss the relations among cumulants. The cyclic fluctuation relation (33) can be rewritten as the integral form

$$\left\langle e^{-A_C(J + B_C J^3)} \right\rangle \simeq 1. \quad (34)$$

We expand $n$-th cumulant with $A_C$ in Eq. (33) as

$$\left\langle J^n \right\rangle = \sum_m L_{nm} A_C^m / m!. \quad (35)$$

From Eqs. (34) and (35), we obtain the violation of the FDR as

$$2L_{11} - L_{20} + 2B_C(L_{31} + 3L_{11}L_{20} - L_{40}) = 0 \quad (36)$$
as the balance of terms of order $O(A_C^3)$. Similarly, we also obtain the violation of the nonlinear relation

$$L_{12} - L_{21} + B_C(L_{32} + 3L_{12}L_{20} + 6L_{11}L_{21} - 2L_{41}) = 0$$

as the balance of terms of order $O(A_C^3)$. Note that we can obtain more relations as the balance of terms of any order $O(A_C^n)$. The derivations of Eqs. (36) and (37) are given in Appendix 1. The violations of the conventional relations in Eqs. (36) and (37) are caused by the non-Gaussianity $B_C$ which originates from the BSN curvature.

D. Instantaneous fluctuation relation

Let us discuss the instantaneous fluctuation relation in this subsection. Here, we control temperatures in the right and the left reservoirs as in Eqs. (29) and (30). At $\theta_n = \pi n$ and $A_n = 0$ the geometrical contribution (the second term on the right hand side of Eq. (25)) is dominant. Its explicit behaviour is plotted as in Fig. 7. This result implies that the fluctuation is highly non-Gaussian, in contrast to the conventional fluctuation theorem.

VI. CONCLUSION

In this paper, we derived the cyclic and the instantaneous fluctuation relations given in Eqs. (17) and (25), respectively, for adiabatic pumping processes. We applied these results to the spin-boson model and clarified the existence of non-Gaussianity as the geometric phase contribution in the fluctuation relations as in Eq. (33) (Fig. 5). We confirmed that the non-Gaussianity $V_3(n)$ in Eq. (33) is not small. From the cyclic fluctuation relation, we obtained the relations among cumulants (36) and (37), which show the violation of the FDR and other conventional relations among cumulants. Our results indicate that the conventional fluctuation theorem should be extended to include non-Gaussian fluctuations if the geometric phase effect exists under cyclic modulation of parameters.

Our future tasks are as follows: (i) Because our analysis is restricted to the adiabatic case, we will have to try to extend our analysis to the non-adiabatic case. If we restrict our interest to a two-level system like the spin-boson system, we can use the analytic solution of the generalized master equation [4]. (ii) We can analyze the entropy production by a parallel method reported in Refs. [10][12], in which the excess entropy production can be expressed by the geometric phase. (iii) Shortcuts to adiabaticity (STA) can be used for the non-adiabatic pumping in which a finite pumping current can be realized under finite speed modulation. Therefore, we expect that the universal work-fluctuation relation discussed by Funo et al. [65] can be generalized to...
include non-Gaussian fluctuations as mentioned in this paper. (iv) We will have to discuss the linear response around a cyclic adiabatic state obtained in this paper by changing the modulation perturbatively. This linear response theory is expected to be different from that obtained from the Green-Kubo formula [60].

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Appendix A: Full counting statistics

We briefly summarize the method of the full counting statistics (FCS) in this appendix. As mentioned in Sec. II, we perform the projective measurement on Hamiltonian $H_R$, the number operator $N_R$, etc.) in the right reservoir at time zero and time $\tau$, the corresponding outcomes are denoted by $Q_0$ and $Q_\tau$, respectively. The joint probability $P(Q_0,Q_\tau)$ to get the outcome $Q_0$ and $Q_\tau$ is given as

$$P(Q_\tau,Q_0) = \text{Tr} \left[ P_{Q,R}(\tau) P^{\text{tot}}(0) P_{Q,R} U^{\dag}(\tau) P_{Q,R} \right], \quad \text{(A1)}$$

where $P_{Q,R}$ is the projection operator corresponding to the outcome $Q_\tau$, $U(\tau) := e^{iH_{R,k}\tau}/\hbar$ and $P^{\text{tot}}(0)$ is the density matrix of the total system at $t = 0$. The probability distribution $P_\tau(q)$ of the transfer $q = Q_\tau - Q_0$ during the time interval $\tau$ is given by

$$P_\tau(q) = \sum_{Q_\tau,Q_0} \delta(q - Q_\tau + Q_0) P(Q_\tau,Q_0). \quad \text{(A2)}$$

Then, using Eq. (A1), the generating function (2) can be rewritten as

$$Z_\tau(\chi) := \int_0^\infty dq P_\tau(q)e^{i\chi q} \quad = \sum_{Q_\tau,Q_0} P(Q_\tau,Q_0)e^{i\chi(Q_\tau-Q_0)} \quad = \text{Tr} \left[ e^{i\chi Q/2} U(\tau)e^{-i\chi Q/2} P^{\text{tot}}(0)e^{-i\chi Q/2} U^{\dag}(\tau) e^{i\chi Q/2} \right]$$

$$= \text{Tr}[\rho_{\text{tot}}(\tau,\chi)], \quad \text{(A3)}$$

where we have introduced $\rho_{\text{tot}}(\tau,\chi) := U(\tau,\chi)\rho^{\text{tot}}(0)U^{\dag}(\tau,-\chi)$ and $U(\tau,\chi) := e^{i\chi Q/2} U(\tau)e^{-i\chi Q/2}$. Note that $\rho_{\text{tot}}(0,\chi) = \rho_{\text{tot}}(0)$.

Appendix B: Details of the Liouvillian in Eqs. (3) and (4)

The Born-Markov form of the Liouvillian in Eq. (3) is given as [51]

$$[H_S, L_k] = -\hbar \omega_k L_k, \quad [H_S, L_k^\dagger] = \hbar \omega_k L_k^\dagger. \quad \text{(B3)}$$

Here $B_{\nu,k}$ is the operator of $\nu$-th reservoir which satisfies

$$[H_S, B_{\nu,k}] = -\hbar \omega_{\nu,k} B_{\nu,k}, \quad [H_S, B_{\nu,k}^\dagger] = \hbar \omega_{\nu,k} B_{\nu,k}^\dagger. \quad \text{(B4)}$$

In addition, $Q$ and $B_R$ satisfy

$$[Q, B_{R,k}] = -q_{R,k}B_{R,k}, \quad [Q, B_{R,k}^\dagger] = q_{R,k}B_{R,k}^\dagger, \quad \text{(B5)}$$

where $\omega_{R,k} = \hbar \omega_{R,k}$ when $Q = H_R$, $q_{R,k} = 1$ when $Q = N_R$, respectively. Furthermore, we assume that $|\omega_k \pm \omega_l|^{-1}$ is smaller than the time scale of the target system (RWA). By using this assumption, the
where we note that \( q_{L,\nu} = 0 \). Here, \( \gamma_{\nu, k \pm}(\alpha(t)) \) is the dissipation coefficient defined as

\[
\gamma_{\nu, k \pm}(\alpha(t)) := \frac{1}{\hbar^2} \left[ \int_{-\infty}^{\infty} ds \, e^{i\omega_k s} \langle B^\dagger_{\nu, k} B_{\nu, k}(s) \rangle \pm \frac{i}{\hbar} \right] \delta(\omega_k - \omega_{\nu, k}) \langle B^\dagger_{\nu, k} B_{\nu, k} \rangle, \tag{B8}
\]

For the Lindblad form, The diagonal elements of \( \rho(t, \chi) \) satisfy the GCME (4). The \((mn)\)-component of the transition matrix \( K(\alpha(t), \chi) \) in Eq. (4) is given as

\[
k_{mn}(t, \chi) := q_{mn}^L(t) + k_{mn}^R(t) e^{-i\chi q_{mn}^R}, \tag{B11}
\]

where

\[
k_{mn}^\nu(t) := \sum_k [k_{mn}^{\nu, k+}(t) + k_{mn}^{\nu, k-}(t)] \quad \tag{B12}
\]

with

\[
k_{mn}^{\nu, k+}(t) := \gamma_{\nu, k -}(\alpha(t)) |n| L^+_{k \nu} |m\rangle^2, \tag{B13}
\]

\[
k_{mn}^{\nu, k-}(t) := \gamma_{\nu, k +}(\alpha(t)) |n| L^-_{k \nu} |m\rangle^2. \tag{B14}
\]

Here \( q_{mn}^R = E_m - E_n \) for \( Q = H_R \), where \( E_l \) is \( l \)-th eigenenergy of \( H_S \), i.e. \( H_S = \sum_l E_l |l\rangle \langle l| \). For \( Q = N_R \), \( q_{mn}^R \) is given by \( q_{mn}^R = m - n \).

**Appendix C: Adiabatic approximation**

Let us assume that the density matrix \( \rho(\theta, \chi) \) is parallel to the right eigenvector \( |r(\theta, \chi)\rangle \) as

\[
|\rho(\theta, \chi)\rangle = C(\theta, \chi)|r(\theta, \chi)\rangle, \tag{C1}
\]

where the function \( C(\theta, \chi) \) will be determined later.

By using the \( \text{GCME}(4) \) and the normalization condition \( \langle l(\theta, \chi)|r(\theta, \chi)\rangle = 1 \), we get

\[
\partial_\theta C(\theta, \chi) = C(\theta, \chi) \left[ e^{-1} \lambda(\theta, \chi) - v(\theta, \chi) \right], \tag{C2}
\]

where \( v(\theta, \chi) = \langle l(\theta, \chi)|\partial_\theta|r(\theta, \chi)\rangle \). Equation (C2) can be solved as

\[
C(\theta, \chi) = C(0, \chi) \exp \left[ \int_0^\theta d\theta' e^{-1} \lambda(\theta', \chi) - v(\theta', \chi) \right]. \tag{C3}
\]

From the \( \text{normalization condition} \) \( \langle 1|\rho(0, \chi)\rangle = 1 \), we get

\[
C(0, \chi) = \langle 1| r(0, \chi) \rangle ^{-1}. \]

Therefore, we obtain Eqs. (6).

**Appendix D: Pumping current**

Although we are not interested in the average current for the adiabatic pumping process, it is useful to write its explicit form for the convenience to compare our results with the results in the literature. The average current can be decomposed to two parts as \( \langle J \rangle = \langle J \rangle^\text{dyn} + \langle J \rangle^\text{geo} \). The first part is the dynamic current expressed as

\[
\langle J \rangle^\text{dyn} = \frac{1}{2\pi} \int_0^{2\pi} d\theta J^\text{st}(\theta), \tag{D1}
\]

where \( J^\text{st}(\theta) := \partial_\lambda \lambda(\theta, \chi) |_{\chi=0} \) is the instantaneous steady current. The second part is the geometrical current expressed as

\[
\langle J \rangle^\text{geo} = -\frac{\epsilon}{2\pi} \int_S d\alpha_m d\alpha_n F_{mn}(\alpha), \tag{D2}
\]

where \( F_{mn}(\alpha) := \partial_\chi \mathcal{F}(\alpha, \chi) |_{\chi=0} \) is the BSN curvature \([15, 16]\) in the parameter space. Note that even if the average bias is zero such that \( \langle J \rangle^\text{dyn} = 0 \), \( \langle J \rangle^\text{geo} \) is generally not zero.

**Appendix E: Derivation of Eq. (13)**

The current distribution is given in Eq. (13). The LDF is given in (14). We expand \( \Lambda(\chi) \) with respect to
\[ i\chi \text{ around } \chi = \chi_c(J) \text{ as} \]
\[
\Lambda(\chi) = \Lambda(\chi_c) + \Lambda^{(1)}(\chi_c)(i\chi - i\chi_c) + \frac{1}{2}\Lambda^{(2)}(\chi_c)(i\chi - i\chi_c)^2 + O((i\chi - i\chi_c)^3), \quad (E1)
\]

where we have introduced \( \Lambda^{(n)}(\chi_c(J)) := \partial^n \Lambda(\chi)|_{\chi=\chi_c(J)} \). Then, we get
\[
i\chi J - \Lambda(\chi) \simeq I(J) + \frac{1}{2}\Lambda^{(2)}(\chi_c(J))(\chi - \chi_c(J))^2. \quad (E2)
\]

Therefore we obtain Eq. (13) as
\[
\begin{align*}
P_c(J) & \simeq e^{-\frac{1}{2}|I(J)|} \times \int_{-\infty}^{\infty} \frac{d\chi}{\epsilon} e^{\frac{\epsilon}{2}\Lambda^{(2)}(\chi_c(J))(\chi - \chi_c(J))^2} e^{-2\pi V(\chi)} \\
& \simeq \frac{1}{\sqrt{\epsilon\Lambda^{(2)}(\chi_c(J))}} e^{-\frac{\epsilon}{2}|I(J)|+V(\chi_c(J))}. \quad (E3)
\end{align*}
\]

**Appendix F: Derivation of Eq. (20)**

In this appendix, we explain the details of the derivation of Eqs. (20) and (22). From the definition of \( P_c(J) \) in Eq. (13) we can write
\[
P_c(J) = \frac{1}{\epsilon} \int_{-\infty}^{\infty} d\chi e^{-\frac{2\pi}{\epsilon}(i\chi J-G(\chi))} \]
\[
= \frac{1}{\epsilon} \int_{-\infty}^{\infty} d\chi \exp \left[ -\frac{2\pi}{\epsilon} \left\{ i\chi J - \frac{1}{2\pi} \int_0^{2\pi} d\theta g(\chi, \theta) \right\} \right] \quad (F1)
\]

where \( g(\theta, \chi) := \lambda(\theta, \chi) - \epsilon c(\theta, \chi) \). As mentioned in Sec. IVB, we discretize variables in the interval \([0, 2\pi]\) into \( N \) pieces. Then, we rewrite Eq. (F1) as
\[
P_c(J) = \frac{1}{\epsilon} \lim_{N \rightarrow \infty} \int_{-\infty}^{\infty} d\chi \exp \left[ -\frac{2\pi}{\epsilon} \left\{ i\chi J - \frac{1}{N} \sum_{n=1}^{N} g_n(\chi) \right\} \right] \]
\[
= \frac{1}{\epsilon} \lim_{N \rightarrow \infty} \int_{-\infty}^{\infty} d\chi e^{-\frac{2\pi}{\epsilon} i\chi J} \prod_{n=1}^{N} e^{\frac{2\pi}{\epsilon g_n(\chi)}}, \quad (F2)
\]

where we have used \( g_n(\chi) = \int_{-\infty}^{\infty} d\chi_n \delta(\chi - \chi_n) g_n(\chi_n) \). By using \( \delta(x) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dk \, e^{ikx} \), Eq. (F2) can be rewritten further as
\[
P_c(J) = \frac{1}{\epsilon} \lim_{N \rightarrow \infty} \int_{-\infty}^{\infty} d\chi e^{-\frac{2\pi}{\epsilon} i\chi J} \prod_{n=1}^{N} \int_{-\infty}^{\infty} d\chi_n \int_{-\infty}^{\infty} dJ_n e^{\frac{2\pi}{\epsilon} (i\chi_n J_n - g_n(\chi_n))} \\
= \frac{1}{\epsilon} \lim_{N \rightarrow \infty} \prod_{n=1}^{N} \int_{-\infty}^{\infty} dJ_n \int_{-\infty}^{\infty} d\chi_n \int_{-\infty}^{\infty} d\chi_n \prod_{n=1}^{N} e^{\frac{2\pi}{\epsilon} (i\chi_n J_n - g_n(\chi_n))} \\
= \frac{1}{\epsilon} \lim_{N \rightarrow \infty} \prod_{n=1}^{N} \int_{-\infty}^{\infty} dJ_n \int_{-\infty}^{\infty} d\chi_n \prod_{n=1}^{N} e^{\frac{2\pi}{\epsilon} \left[i\chi_n J_n - g_n(\chi_n)\right]} \\
= \lim_{N \rightarrow \infty} \prod_{n=1}^{N} \int_{-\infty}^{\infty} dJ_n \prod_{n=1}^{N} e^{\frac{2\pi}{\epsilon} \left[i\chi_n J_n - g_n(\chi_n)\right]} \prod_{n=1}^{N} p_n(J_n). \quad (F3)
\]

Here we obtain the explicit expression of \( p_n(J_n) \) as Eq. (22).

**Appendix G: Detail of the spin-boson model**

The diagonal element of \( \rho(\theta, \chi) \) is given as \( \rho(\theta, \chi) = (\langle 0|\rho(\theta, \chi)|0 \rangle, \langle 1|\rho(\theta, \chi)|1 \rangle)^T \). Each element of the transition matrix \( \hat{K}(\alpha(\theta), \chi) \) in the spin-boson model is
The BSN curvature of the second case in Sec.V is given as
\[ \lambda(\theta, \chi) := \Gamma(\theta) \frac{1}{\Gamma(\theta)} \frac{\lambda(\theta, \chi) + k_{10}(\theta, \chi)}{10}, \]
with
\[ n_{\nu}(\theta) := (e^{\beta_{\nu}(\theta)\omega_{0}(\theta)} - 1)^{-1}, \]
\[ \gamma_{\nu}(\theta) := \frac{\Gamma_{\nu}(\theta)}{\Gamma}, \]
\[ \Gamma := \frac{1}{2\pi} \int_{0}^{2\pi} d\beta \frac{\Gamma_{L}(\theta) + \Gamma_{R}(\theta)}{2}. \]

The eigenvalue \( \lambda(\theta, \chi) \) of \( K(\alpha(\theta), \chi) \) and corresponding eigenvectors \( \langle l(\theta, \chi) \rangle, \langle r(\theta, \chi) \rangle \) are given as
\[ \lambda(\theta, \chi) = -\frac{k_{10}(\theta) + k_{10}(\theta)}{2} + \sqrt{\left( \frac{k_{10}(\theta) - k_{10}(\theta)}{2} \right)^{2} + k_{10}(\theta, \chi)k_{10}(\theta, \chi)}, \]
\[ \langle l(\theta, \chi) \rangle = \frac{1}{c(\theta, \chi)} \left( \frac{\lambda(\theta, \chi) + k_{10}(\theta, \chi)}{10} \right), \]
\[ \langle r(\theta, \chi) \rangle = \frac{1}{c(\theta, \chi)} \left( \frac{\lambda(\theta, \chi) + k_{10}(\theta, \chi)}{10} \right), \]
\[ c(\theta, \chi) := 1 + \frac{(\lambda(\theta, \chi) + k_{10}(\theta, \chi))^{2}}{k_{10}(\theta, \chi)k_{10}(\theta, \chi)}. \]

The BSN curvature \( F_{mn}(\alpha) \) in Eq. (12) of the first case in Sec.V is given as
\[ F(\hat{T}_{L}, \hat{T}_{R}) = \frac{n_{L}^{2}n_{R}^{2}}{8\hat{T}_{L}^{2}\hat{T}_{R}^{2}(1 + n_{L} + n_{R})^{3}}. \]

Similarly, the BSN curvature of the second case in Sec.V is given as
\[ F(\gamma_{L}, \hat{\omega}_{0}) = \frac{(\gamma_{L} + 1 - \hat{\omega}_{0})n_{L}(1 + n_{L})}{(\gamma_{L} + 1)(1 + 2n_{L})}. \]

Appendix H: Numerical check of Eq. (15) for the spin-boson model

We derive the LLGC symmetry for \( \lambda(\theta, \chi) \) in the spin-boson model. The eigenvalue \( \lambda(\theta, \chi) \) depends on \( \chi \) through \( k_{01}(\theta, \chi)k_{10}(\theta, \chi) \). For any \( \chi \), we obtain
\[ 0 = k_{01}(\theta, \chi)k_{10}(\theta, \chi) - k_{10}(\theta, -\chi + i\mathcal{A}(\theta))k_{10}(\theta, -\chi + i\mathcal{A}(\theta)) = (k_{10}(\theta)k_{01}(\theta) - k_{10}(\theta)k_{10}(\theta)e^{i\mathcal{A}(\theta)}) \times (e^{i\chi} + e^{-i\chi}e^{-i\mathcal{A}(\theta)}), \]
where
\[ \mathcal{A}(\theta) = \ln \frac{k_{01}(\theta)k_{10}(\theta)}{k_{10}(\theta)k_{01}(\theta)} = \ln \frac{n_{L}(1 + n_{L})}{n_{L}(1 + n_{R})}. \]
This reduces to \( \mathcal{A}(\theta) = \hbar\omega_{0}(\beta_{R}(\theta) - \beta_{L}(\theta)) \). Therefore we obtain the LLGC symmetry \( \lambda(\theta, \chi) = \lambda(\theta, -\chi + i\mathcal{A}(\theta)) \). This leads the symmetry relation \( \Lambda(\chi) \) for the instantaneous LDF \( I_{n}(\chi) \).

When we consider the cyclic modulation in the limit \( \epsilon \to 0 \), the rate function \( I(\mathcal{J}) \) can be evaluated only from the dynamical part. We expect that the logarithmic form of Eq. (15) satisfies Eq. (16) if we replace the numerator and the denominator in the right hand side of Eq. (15) in the spin-boson model, we numerically check the validity of Eq. (15). We control the temperature of the left and right reservoirs as
\[ \hat{T}_{L}(\theta) = \hat{T}_{L0} + \hat{T}_{LA}\cos(\theta + \pi/4), \]
\[ \hat{T}_{R}(\theta) = \hat{T}_{R0} + \hat{T}_{RA}\sin(\theta + \pi/4). \]

From the numerical calculation of the left and right hand sides of Eq. (15) in the spin-boson model, we have confirmed the symmetry relation \( \Lambda(\chi) \) numerically (Fig. 8). Note that, from our numerical calculation, \( \Lambda(\chi) = \Lambda(-\chi + i\mathcal{A}) \) seems to hold.

Appendix I: Derivation of Eqs. (36) and (37)

Eq. (34) can be rewritten as
\[ 0 = \sum_{n=1}^{\infty} (-A_{C})^{n}(\mathcal{J} + B_{C}\mathcal{J}^{3})^{n}. \]
Each moment \( \langle J^{n} \rangle \) can be rewritten by cumulants \( \langle J^{n} \rangle_{c} \) as
\[ \langle J \rangle = \langle J \rangle_{c}, \]
\[ \langle J^{2} \rangle = \langle J^{2} \rangle_{c} + \langle J \rangle^{2}, \]
\[ \langle J^{3} \rangle = \langle J^{3} \rangle_{c} + 3\langle J^{2} \rangle_{c}\langle J \rangle_{c} + \langle J \rangle^{3}. \]
By using Eq. (35), we obtain

\[0 = -A_C L_{10} + A_C^2 \left[ -L_{11} + \frac{1}{2} L_{20} - B_C (L_{13} + 3L_{20}L_{11} - L_{40}) \right]
+ A_C^3 \left[ -\frac{L_{12}}{2} + \frac{L_{21}}{2} - B_C \left( \frac{L_{32}}{2} + 3L_{21}L_{11} + \frac{3}{2} L_{20}L_{12} - L_{41} \right) \right] + O(A_C^4).
\]

Then, we obtain Eqs. (36) and (37) as the balance of terms of order \(O(A_C^2)\) and \(O(A_C^3)\), respectively.

![Figure 8. The verification of the symmetry relation of the large deviation function, Eq. (14), in the spin-boson model. The points are the plots of \(I(J) - I(-J)\) at \((\hat{T}_{R0}, \hat{T}_{LA}) = (5, 4)\) and \((\hat{T}_{L0}, \hat{T}_{LA}) = (10, 9), (6, 5), (5, 4)\) (their colors are red, purple, blue, respectively). The solid lines are the plots of \(-AJ\) at \((\hat{T}_{R0}, \hat{T}_{LA}) = (5, 4)\) and \((\hat{T}_{L0}, \hat{T}_{LA}) = (10, 9), (6, 5), (5, 4)\) (their colors are orange, magenta and cyan, respectively).]
[33] T. Brandes and T. Vorrath, Phys. Rev. B 66, 075341 (2002).
[34] E. Cota, R. Aguado, and G. Platero, Phys. Rev. Lett. 94, 107202 (2005).
[35] J. Splettstoesser, M. Governale, J. König, and R. Fazio, Phys. Rev. B 74, 085305 (2006).
[36] F. Reckermann, J. Splettstoesser, and M. R. Wegewijs, Phys. Rev. Lett. 104, 226803 (2010).
[37] T. Yuge, T. Sagawa, A. Sugita, and H. Hayakawa, Phys. Rev. B 86, 235308 (2012).
[38] K. L. Watanabe and H. Hayakawa, Prog. Theor. Exp. Phys. 2014, 113A01 (2014).
[39] S. Nakajima, M. Taguchi, T. Kubo, and Y. Tokura, Phys. Rev. B 92, 195420 (2015).
[40] T. Sagawa and H. Hayakawa, Phys. Rev. E 84, 051110 (2011).
[41] T. Yuge, T. Sagawa, A. Sugiura, and H. Hayakawa, J. Stat. Phys. 153, 412 (2013).
[42] S. Nakajima and Y. Tokura, J. Stat. Phys. 169, 902 (2017).
[43] S. K. Giri and H. P. Goswami, Phys. Rev. E 96, 052129 (2017).
[44] M. Toda, R. Kubo et al., *Statistical Physics II: Nonequilibrium Statistical Mechanics*, (Springer, Berlin, 2013), 2nd ed.
[45] D. J. Evans, E. G. D. Cohen, and G. P. Morriss, Phys. Rev. Lett. 71, 2401 (1993).
[46] L. S. Levitov and G. B. Lesovik, Pis’ma Zh. Eksp. Teor. Fiz. 58, 225 (1993) [JETP Lett. 58, 230 (1993)].
[47] G. Gallavotti and E. G. D. Cohen, Phys. Rev. Lett. 74, 2694 (1995).
[48] J. L. Lebowitz and H. Spohn, J. Stat. Phys. 95, 333 (1999).
[49] C. Jarzynski, Phys. Rev. Lett. 78, 2690 (1997).
[50] G. E. Crooks, J. Stat. Phys. 90, 1481 (1998).
[51] M. Esposito, U. Harbola, and S. Mukamel, Rev. Mod. Phys. 81, 1665 (2009).
[52] M. Esposito and C. Van den Broeck, Phys. Rev. Lett. 104, 090601 (2010).
[53] D. J. Evans and G. Morriss, *Statistical Mechanics of Nonequilibrium Liquids*, 2nd ed. (Cambridge University Press, Cambridge, UK, 2008).
[54] D. Andrieux and P. Gaspard, J. Stat. Mech. (2007) P02006.
[55] K. Saito and A. Dhar, Phys. Rev. Lett. 99, 180601 (2007).
[56] J. D. Noh and J.-M. Park, Phys. Rev. Lett. 108, 240603 (2012).
[57] K. Kanazawa, T. Sagawa, and H. Hayakawa, Phys. Rev. E 87, 052124 (2013).
[58] T. G. Sano and H. Hayakawa, Phys. Rev. E 89, 032104 (2014).
[59] J. Ren, P. Hänggi, and B. Li, Phys. Rev. Lett. 104, 170601 (2010).
[60] K. L. Watanabe and H. Hayakawa, Phys. Rev. E 96, 022118 (2017).
[61] A. A. Svidzinsky, K. E. Dorfman, M. O. Scully, Coherent Opt. Phenom. 1, 7 (2012).
[62] H. P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University Press, 2002).
[63] Y. Izumida, Euro. Phys. Lett. 121, 50004 (2018).
[64] K. Funo, J. N. Zhang, C. Chatou, K. Kim, M. Ueda, and A. del Campo, Phys. Rev. Lett. 118, 100602 (2017).
[65] E. Potaniina, C. Flindt, M. Moskalets, and K. Brandner, arXiv:1906.04297.