Symmetry in full counting statistics, fluctuation theorem, and relations among nonlinear transport coefficients in the presence of a magnetic field

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We study the full counting statistics of electron transport through multiterminal interacting quantum dots under a finite magnetic field. Microscopic reversibility leads to a symmetry of the cumulant generating function, which generalizes the fluctuation theorem in the context of the quantum transport. Using the symmetry, we derive the Onsager-Casimir relations in the linear transport regime and universal relations among nonlinear transport coefficients. One of the measurable relations is that the nonlinear conductance, the second-order coefficient with respect to the bias voltage, is connected to the third current cumulant in equilibrium, which can be a finite and uneven function of the magnetic field for two-terminal noncentrosymmetric system.

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I. INTRODUCTION

Full counting statistics (FCS) has become an active topic in the mesoscopic physics.[1–13] FCS addresses the probability distributions of charges transmitted during a measurement time τ. It can characterize the statistical properties of quantum transport in the far-from-equilibrium regime. Since the seminal paper by Levitov and Lesovik,[1(a)] many theories have clarified various aspects of the distributions.[1–8] Recently, experiments have been conducted to measure third current cumulants[11] and the distributions.[12,13] However, FCS has never been applied to study the general aspects of nonequilibrium thermodynamic structures in coherent electron transport. In this paper, we discuss these general aspects by studying symmetries in FCS, which is valid beyond a linear-response regime.

Our argument is based on the microscopic reversibility and is related to the steady-state *fluctuation theorem* (FT) in nonequilibrium statistical mechanics.[14–17] FT is an important theory that holds even in the far-from-equilibrium regime. It is written as

$$\lim_{\tau\to\infty}\frac{1}{\tau}\ln\left[\frac{P(\Delta S)}{P(-\Delta S)}\right]=I_E, \tag{1}$$

where $P(\Delta S)$ is the probability of entropy ΔS produced during time τ, and I_E is the entropy per unit time, $I_E=\Delta S/\tau$. This expression quantifies the probability of negative entropy, which can be finite for a short interval of time in small systems, as demonstrated in colloidal particle experiments.[15] Remarkably, FT can reproduce the Onsager relation and the Kubo formula[16,18] and predicts properties in the far-from-equilibrium regime.

Recently FT was studied with regard to classical mesoscopic electron transport, i.e., the sequential tunneling regime in quantum dots (QDs), with Markovian approximations.[18,19] These works highlighted the relation between FT and FCS. In this paper, we study the general relation between FT and FCS with respect to *coherent electron transports* in generic situations, i.e., transport through a multiterminal interacting quantum dots under a finite magnetic field. In multiterminal electron transport, thermodynamic forces are bias voltages between reservoirs. We consider a cumulant generating function to measure accumulated charge inside each reservoir and derive its different symmetry under finite magnetic fields.[20] Then we demonstrate that it extends the FT to a finite magnetic field in quantum regime.

Important results inferred from the symmetry are relations among nonlinear transport coefficients. This is a main focus of this paper. As a first step, we will show the symmetry of cumulant generating function (CGF) reproduces the linear-response results, such as the Onsager-Casimir relation.[21,22] This is a demonstration of derivation of the Onsager-Casimir relation from the FCS. Furthermore, the symmetry yields other relations among nonlinear transport coefficients beyond the Onsager-Casimir relation. In mesoscopic experiments, large bias voltages can easily induce nonlinear transport, and average current does not satisfy the Onsager-Casimir relation. However, the additional relations among transport coefficients tell us that beyond the Onsager relation, universal relations exist in the nonlinear transport regime. One of interesting results is that the nonlinear conductance, the second-order coefficient with respect to the bias voltage, is connected to the third current cumulant in equilibrium, which can be a finite and uneven function of the magnetic field. Those should be measurable in the present-day experiments.

This paper is organized as follows: In Sec. II, the model we consider is introduced and in Sec. III, the symmetry of the cumulant generating function is explained, whose proof is given in Appendix A. In Sec. IV, the symmetry is employed to obtain a general relation between transport coefficients, and we demonstrate the relation using a three-terminal Aharonov-Bohm interferometer. In Sec. V, the relation between the symmetry and fluctuation theorem is discussed. Finally we summarize the discussions in Sec. VI.

II. MODEL HAMILTONIAN

We consider mesoscopic QDs connected to m electron reservoirs. The total Hamiltonian consists of the Hamiltonians of the reservoirs, H_r $(r=1,\ldots,m)$; the QDs, H_d; the Coulomb interaction inside the QDs, $H_{\rm int}$; and the tunneling H_T:

$$H=\sum_{r=1}^{m} H_r + H_d + H_{\rm int} + H_T, \quad (2)$$

$$H_d=\sum_{ij\sigma} t_{ij} d_{i\sigma}^{\dagger} d_{j\sigma}, \quad (3)$$

$$H_{\rm int}=\frac{1}{2}\sum_{ij\sigma\sigma'} U_{i\sigma j\sigma'} d_{i\sigma}^{\dagger} d_{j\sigma'}^{\dagger} d_{j\sigma'} d_{i\sigma}, \quad (4)$$

where $d_{i\sigma}$ annihilates an electron with spin σ at site i. The Hamiltonian for the reservoir r is

$$H_r=\sum_{k\sigma} \varepsilon_{rk} a_{rk\sigma}^{\dagger} a_{rk\sigma}, \quad (5)$$

where $a_{rk\sigma}$ annihilates an electron with spin σ and the wave vector k. The tunneling Hamiltonian between the reservoirs and the QDs is described as

$$H_T=\sum_{rki\sigma} t_{rki} d_{i\sigma}^{\dagger} a_{rk\sigma} + \text{H.c.} \quad (6)$$

When the magnetic field B is applied, the hopping and tunneling matrix elements acquire the phases as $t_{ij}=|t_{ij}|\exp(i\phi_{ij})$ and $t_{rki}=|t_{rki}|\exp(i\phi_{ri})$. The phases are odd functions of the magnetic field: $\phi(-B)=-\phi(B)$.

The initial density matrix at $t=-\tau/2$ is assumed to be of decoupled form, where each reservoir and QDs are in equilibrium at a uniform temperature β^{-1} and the respective chemical potential:

$$\rho_0=\prod_s \frac{\exp[-\beta(H_s-\mu_s N_s)]}{\text{Tr}\{\exp[-\beta(H_s-\mu_s N_s)]\}}, \quad (7)$$

where s is the index of the reservoir $r=1,\ldots,m$ and the QDs d, i.e., $s=1,\ldots,m,d$. Here, μ_s is the chemical potential at the part s, and N_s is the number operator: $N_r=\Sigma_{k\sigma} a_{rk\sigma}^{\dagger} a_{rk\sigma}$, and $N_d=\Sigma_{i\sigma} d_{i\sigma}^{\dagger} d_{i\sigma}$. The charge current operators between reservoir r and QDs are expressed as

$$I_r=-\sum_{ik\sigma} i t_{rki} d_{i\sigma}^{\dagger} a_{rk\sigma} + \text{H.c.} \quad (8)$$

III. CUMULANT GENERATING FUNCTION AND ITS SYMMETRY

We introduce the generating function for the transmitted charge during time τ,

$$q_r=\int_{-\tau/2}^{\tau/2} dt I_r(t).$$

Here we derive the characteristic function with different interpretation from Levitov and Lesovik,[1(a)] basically following the idea in Ref. 23. First, we observe the reservoir r to measure charge. Suppose the observed charge for the part r is $Q_{r,i}$. The wave function collapses into an eigenfunction which is a product eigenstate of each part of the number operator. Suppose the wave function to be φ_i. After a free time evolution during τ, we again observe the reservoirs to measure the charge and suppose those to be $Q_{r,j}$ and the wave function to be φ_j. From these two measurements, we find the amount of changes in the charge in each part as $Q_{r,i}-Q_{r,j}$. Thus, the conditional probability $P_{i\to j}(\{Q_r\})$ of finding the amount of change to be Q_r in this one step is written as

$$P_{i\to j}(\{Q_r\})=\prod_{r=1}^{m} \delta[Q_r-(Q_{r,i}-Q_{r,j})]|\langle\varphi_j|e^{-iH\tau}|\varphi_i\rangle|.$$

By iterating these steps for the same initial density matrix ρ_0, we can obtain the probability of change in the charge in the form

$$P(\{Q_r\})=\sum_{i,j} P_{i\to j}(\{Q_r\})\langle\varphi_i|\rho_0|\varphi_i\rangle = \prod_{r=1}^{m}\frac{1}{2\pi}\int d\chi_r \mathcal{Z}(\{\chi\};B)\exp\Big(-i\sum_{r=1}^{m}\chi_r Q_r\Big), \quad (9)$$

where $\mathcal{Z}(\{\chi\};B)$ is the characteristic function given by

$$\mathcal{Z}(\{\chi_r\};B)=\langle V^{\dagger}e^{iH\tau}V^2 e^{-iH\tau}V^{\dagger}\rangle, \quad (10)$$

$$V=\prod_{r=1}^{m}\exp[-i\chi_r N_r/2], \quad (11)$$

where the operator V contains the counting fields for charge χ_r. The symbol $\langle\cdots\rangle$ denotes the average over the initial state ρ_0. This characteristic function satisfies the normalization condition $\mathcal{Z}(\{0\};B)=1$. Equation (10) can be rewritten in the familiar form, where the counting fields play roles of fictitious gauge fields,[1,2,9] since $V^{\dagger}H_T V$ transforms the tunneling matrix element as $t_{rki}\to t_{rki}\exp(-i\chi_r/2)$. Hence, Eq. (10) is the compact expression of the original characteristic function of Levitov and Lesovik.[1(a)]

We consider the general properties of the CGF. The CGF is defined at a stationary state as

$$\mathcal{F}(\{\chi_r\};B)=\lim_{\tau\to\infty}\frac{1}{\tau}\ln \mathcal{Z}(\{\chi_r\};B). \quad (12)$$

The CGF generates cumulants by the derivatives with respect to the counting fields. The average current between the terminal and the QDs and the second order of cumulant of the currents are respectively computed as

$$\langle\langle I_r\rangle\rangle=\frac{\partial\mathcal{F}(\{0\};B)}{\partial i\chi_r}=\lim_{\tau\to\infty}\frac{1}{\tau}\langle q_r\rangle,$$

$$\langle\langle I_r I_{r'}\rangle\rangle=\frac{\partial^2\mathcal{F}(\{0\};B)}{\partial i\chi_r\,\partial i\chi_{r'}}=\lim_{\tau\to\infty}\frac{1}{2\tau}(\langle\{q_r,q_{r'}\}\rangle-2\langle q_r\rangle\langle q_{r'}\rangle).$$

The main interests on the CGF in this paper are its symmetry and experimentally measurable properties inferred from it. As shown in Appendix A, the CGF has the following symmetry:

$$\mathcal{F}(\{\boldsymbol{\chi}_r\};B) = \mathcal{F}(\{-\boldsymbol{\chi}_r + i\mathcal{A}_r\};-B), \tag{13}$$

where $\boldsymbol{\chi}_r$ is the difference between the counting fields of the reservoirs r and m, and $\mathcal{A}_r$ is the affinity (thermodynamic force), which are written as

$$\boldsymbol{\chi}_r = \chi_r - \chi_m, \tag{14}$$

$$\mathcal{A}_r = \beta(\mu_r - \mu_m). \tag{15}$$

We can easily check that this symmetry is satisfied in the CGF by Levitov and Lesovik.[1(a)] Relation (13) provides quite much information on cumulant measurements in realistic experiments. Especially it gives additional general relations between transport coefficients. Below we derive some of the information from symmetry (13), and discuss the experimental setup used to measure the results.

IV. GENERAL RELATION AMONG TRANSPORT COEFFICIENTS

A. Two-terminal case

We consider the two-terminal case ($m=2$) as the simplest case. The symmetry is written in the form

$$\mathcal{F}(\chi;B) = \mathcal{F}(-\chi + i\mathcal{A};-B), \tag{16}$$

where, for simplicity, we used the notations $\chi = \boldsymbol{\chi}_1 = \chi_1 - \chi_2$ and $\mathcal{A} = \mathcal{A}_1 = \beta(\mu_1 - \mu_2)$. Symmetry (16) predicts general relations among nonlinear transport coefficients. Let us fix the chemical potential of the second terminal as $\mu_2 = \mu$ and measure the charge current. Then we compute the kth current cumulant,

$$\langle\langle I^k \rangle\rangle = \frac{\partial^k \mathcal{F}(0;B)}{\partial(i\chi)^k}. \tag{17}$$

In general, the arbitrary order of cumulant is a function of bias voltage $V = \mu_1 - \mu_2$. Therefore the cumulant $\langle\langle I_1^k \rangle\rangle$ can be expanded as

$$\langle\langle I^k \rangle\rangle = L_0^k(B) + L_1^k(B)\beta V + \frac{1}{2!}L_2^k(B)(\beta V)^2 + \cdots$$
$$= \sum_{\ell=0}^{\infty} \frac{1}{\ell!} L_\ell^k(B)\mathcal{A}^\ell. \tag{18}$$

The variable L_ℓ^k is the nonlinear transport coefficient we consider. For instance, $L_1^1(B)$ is the linear conductance multiplied by β^{-1}, and $L_0^2(B)$ is the second order of cumulant in the equilibrium state. Definition (18) is a natural extension of the linear conductance. We symmetrize the transport coefficients and the CGF as

$$L_{\ell,\pm}^k = L_\ell^k(B) \pm L_\ell^k(-B), \tag{19}$$

$$\mathcal{F}_\pm(\chi) = \mathcal{F}(\chi;B) \pm \mathcal{F}(\chi;-B). \tag{20}$$

Symmetry (16) leads to the following symmetry for the symmetrized CGF:

$$\mathcal{F}_\pm(\chi) = \pm \mathcal{F}_\pm(-\chi + i\mathcal{A}). \tag{21}$$

The symmetrized coefficient $L_{\ell,\pm}^k$ can be computed using $\mathcal{F}_\pm(\chi)$ as $L_{\ell,\pm}^k = \partial^{k+\ell}\mathcal{F}_\pm(0)/\partial(i\chi)^k \partial\mathcal{A}^\ell$. Note that the CGF is a function of $\mathcal{A}$ as well as χ. By taking derivatives with respect to the affinity and counting field for both sides of the equality of $\mathcal{F}_\pm$, we immediately obtain the general relations among the transport coefficients as

$$L_{\ell,\pm}^k = \pm \sum_{n=0}^{\ell} \binom{\ell}{n} (-1)^{n+k} L_{\ell-n,\pm}^{k+n}, \tag{22}$$

Note the trivial relation

$$L_{\ell,\pm}^0 = 0,$$

due to the normalization condition. Then, this simplifies relation (22). We consider equations satisfying $\mathcal{N} = k + \ell$ in Eq. (22). Equations for $\mathcal{N}=2$ yield linear-response results such as Onsager-Casimir relations and Kubo formula,

$$L_{1,-}^1 = 0, \tag{23}$$

$$L_1^1(B) = \frac{1}{2}L_0^2(B). \tag{24}$$

Relations among higher-order transport coefficients can be obtained in the same manner. We list some of the relations for $\mathcal{N}=3$:

$$L_{2,+}^1 = L_{1,+}^2, \quad L_{0,+}^3 = 0, \tag{25}$$

$$L_{2,-}^1 = \frac{1}{3}L_{1,-}^2 = \frac{1}{6}L_{0,-}^3. \tag{26}$$

In mesoscopic experiments, large bias voltages can easily produce finite higher-order coefficients, and the Onsager-Casimir relations can be violated.[24–26] Equations (25) and (26) demonstrate that beyond the Onsager relation, universal relations exist in the nonlinear transport regime.

Equations (25) are the symmetric parts of the relations and measurable even in the absence of the magnetic field. The first equation in Eq. (25) is the easiest relation to measure. In inhomogeneous systems, those coefficients should be finite. Equation (26) is one of the remarkable results in the presence of finite magnetic field. In noninteracting cases, arbitrary cumulants are symmetric in magnetic field, i.e., $\mathcal{F}_-(\chi)=0$.[24,25,27] However, in noncentrosymmetric systems, the coefficient $L_{2,-}^1$ can be generally finite as shown in the experiment.[28] In this case, Eq. (26) shows that $L_{0,-}^3$ can be finite. This implies the third order of cumulant can be finite and asymmetric in the magnetic field even in the equilibrium state. We expect that this nontrivial relation can be observed in the setup of the experiments.[26]

B. Multiterminal case

We discuss the multiterminal case. We consider the situation that only the chemical potentials μ_1 and μ_2 are varied, while other terminals have a definite chemical potential μ. The charge currents are measured at terminals 1 and 2. Then we consider the cumulant of I_1 and I_2:

$$\langle\langle I_1^{k_1} I_2^{k_2}\rangle\rangle = \frac{\partial^{k_1+k_2}\mathcal{F}(\{0\};B)}{\partial(i\chi_1)^{k_1}\,\partial(i\chi_2)^{k_2}}. \tag{27}$$

The nonlinear transport coefficient is defined as in the two-terminal case,

$$L_{\ell_1\ell_2}^{k_1k_2}(B) = \left.\frac{\partial^{\ell_1+\ell_2}\langle\langle I_1^{k_1} I_2^{k_2}\rangle\rangle}{\partial\mathcal{A}_1^{\ell_1}\,\partial\mathcal{A}_2^{\ell_2}}\right|_{\mathcal{A}_1=\mathcal{A}_2=0}. \tag{28}$$

The affinities $\mathcal{A}_j(j=1,2)$ are written as $\mathcal{A}_j=\beta(\mu_j-\mu)$ with the fixed chemical potential μ for terminals 3–m. We symmetrize the coefficients and the CGF as

$$L_{\ell_1\ell_2,\pm}^{k_1k_2} = L_{\ell_1\ell_2}^{k_1k_2}(B) \pm L_{\ell_1\ell_2}^{k_1k_2}(-B). \tag{29}$$

The completely same argument as in the two-terminal case leads to the general relations among the transport coefficients,

$$L_{\ell_1\ell_2,\pm}^{k_1k_2} = \pm\sum_{n_1=0}^{\ell_1}\sum_{n_2=0}^{\ell_2}\binom{\ell_1}{n_1}\binom{\ell_2}{n_2}(-1)^n L_{\ell_1-n_1\ell_2-n_2,\pm}^{k_1+n_1k_2+n_2}, \tag{30}$$

where $n=n_1+n_2+k_1+k_2$. With the equalities $L_{\ell_1\ell_2,\pm}^{0\,0}=0$ relation (30) can be simplified further. We consider equations satisfying $\mathcal{N}=k_1+k_2+\ell_1+\ell_2$ in Eq. (30). Equations for $\mathcal{N}=2$ yield Onsager-Casimir relations,[22]

$$L_{01}^{10}(B) = L_{10}^{01}(-B), \tag{31}$$

and so on. The Kubo formula $L_{01,+}^{01}=L_{00,+}^{02}/2$, etc., are also obtained. Some of the relations among higher orders for $\mathcal{N}=3$ are listed as

$$L_{20,+}^{10} = L_{10,+}^{20}, \tag{32}$$

$$L_{01,+}^{11} = L_{02,+}^{10} = 2L_{11,+}^{01} - L_{10,+}^{02}, \tag{33}$$

$$L_{01,-}^{20} = L_{20,-}^{01} + 2L_{11,-}^{10}, \tag{34}$$

$$L_{20,-}^{10} = L_{10,-}^{20}/3 = L_{00,-}^{30}/6, \quad L_{00,+}^{30} = 0. \tag{35}$$

C. Example: Three-terminal Aharonov-Bohm interferometer

The simplest setup for demonstrating the relations would be a three-terminal Aharonov-Bohm (AB) ring with the threefold symmetry. For simplicity, we consider the ring consisting of three noninteracting QDs, each of which connects to a reservoir (inset of Fig. 1). The Hamiltonian of the ring is given as

$$H_d = \sum_\sigma\sum_{i=1}^{3} \varepsilon d_{i\sigma}^\dagger d_{i\sigma} - te^{i\phi/3}d_{i+1\sigma}^\dagger d_{i\sigma} + \text{H.c.}, \tag{36}$$

where $d_{4\sigma}=d_{1\sigma}$. The explicit form of the cumulant generating function is calculated using Eq. (B2) in Appendix B, where tunnel coupling (B4) is given as $\Gamma_{rij}=\Gamma\delta_{ij}\delta_{ri}$. The explicit form of CGF is written as

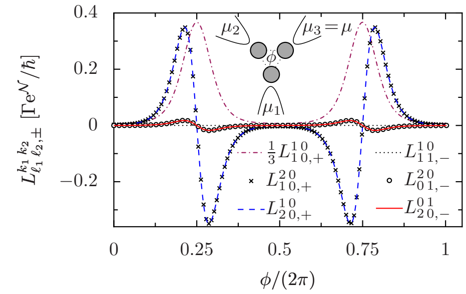

FIG. 1. (Color online) The flux dependence of nonlinear transport coefficients (the unit is $t/\hbar$). Parameters: $\varepsilon=\mu$, $t/\Gamma=10$, and $t\beta=10$. Equations (32) and (34) are satisfied. $L_{11,-}^{10}$ is always zero.

$$\mathcal{F}_0 = \frac{1}{\pi}\int_{-\infty}^{\infty} d\omega\,\ln\Bigg\{1+\sum_{j,k=1}^{3} f_j^+(\omega)f_k^-(\omega)[e^{i(\chi_j-\chi_k)}-1] \times\Bigg\{\mathcal{T}_{\text{even}}(\omega) - \sum_{\ell=1}^{3}\mathcal{T}_{\text{odd}}(\omega)\varepsilon_{jk\ell}[1-2f_\ell^+(\omega)]\Bigg\}\Bigg\}, \tag{37}$$

where $\epsilon_{jk\ell}$ is the totally antisymmetric tensor and $f_r^\pm(\omega)$ is the Fermi/hole distribution,

$$f_r^\pm(\omega) = 1/\{\exp[\pm\beta_r(\omega-\mu_r)]+1\}.$$

The transmission probabilities, which are even and odd functions of the AB phase ϕ, are written as

$$\mathcal{T}_{\text{even}} = \bar{t}^2(1/4+\bar{t}^2+z^2-2\bar{t}z\cos\phi)/\Delta, \tag{38}$$

$$\mathcal{T}_{\text{odd}} = \bar{t}^3\sin\phi/\Delta, \tag{39}$$

where $z=(\omega-\varepsilon)/\Gamma$, $\bar{t}=t/\Gamma$, and

$$\Delta = (z^2+1/4)^3 + \bar{t}^2(3/8-6z^4) + \bar{t}^3 z[(4z^4-3)-12\bar{t}^2]\cos\phi + 9\bar{t}^4(z^2+1/4) + 4\bar{t}^6\cos^2\phi.$$

Now, we can check that the CGF depends on the counting-field difference in Eq. (37), and symmetry (13) is satisfied. Figure 1 shows the linear conductance $L_{10,+}^{10}$ as well as the nonlinear coefficients. Though the overall structures generally depend on temperature regions, relations (32) and (34) hold.

V. QUANTUM FLUCTUATION THEOREM UNDER MAGNETIC FIELDS

Equation (16) can be regarded as an extension of the FT to a finite magnetic field in the quantum regime. For simplicity, we consider the two-terminal case. Following the standard definition, the entropy is defined as the Joule heating

$$\Delta S = \mathcal{A}q_1. \tag{40}$$

Introducing the counting field $\xi=\mathcal{A}^{-1}\chi$, the probability distribution of the entropy is computed as

$$P(\Delta S;B) = \frac{1}{2\pi}\int d\xi e^{-i\Delta S\xi}\mathcal{Z}(\mathcal{A}\xi;B). \tag{41}$$

The saddle-point analysis with symmetry (16), as in Ref. 9, yields the relation

$$\lim_{\tau\to\infty}\frac{1}{\tau}\ln\left[\frac{P(\Delta S;B)}{P(-\Delta S;-B)}\right] = I_E. \tag{42}$$

In the case of $B=0$, this is the usual expression of the FT. This formula generalizes FT in the quantum regime under a finite magnetic field. At a uniform temperature, the entropy production is proportional to the charge current. In this case, Eq. (42) quantifies the probability of backflow of charge currents. In the recent experiment[13] at a finite temperature, the finite probability of backflow was measured. It is expected that the probabilities of positive current and negative current should satisfy Eq. (42).

VI. SUMMARY

The symmetry in the FCS was derived from the microscopic reversibility. This leads to universal relations among nonlinear transport coefficients. We demonstrated them in the three-terminal AB interferometer. In the noncentrosymmetric system, the third cumulant can be finite even in equilibrium and is an odd function of the AB phase. We expect that this nontrivial relation should be measured in future experiments.

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APPENDIX A: DERIVATION OF SYMMETRY (13)

We derive symmetry (13) using a wider class of symmetry. This wider class of symmetry is given in Eq. (A9). Equation (A9) is the symmetry of the cumulant generating function for measuring not only transmitted charge but also heat. Let s be the index of reservoir r and QDs d, i.e., $s=1,\ldots,m,d$. We consider the case that each part s has different temperature β_s^{-1}. The density matrix at the initial time $t=-\tau/2$ is assumed to be

$$\rho_0 = \prod_s \frac{\exp[-\beta_s(H_s-\mu_s N_s)]}{\mathrm{Tr}\{\exp[-\beta_s(H_s-\mu_s N_s)]\}}. \tag{A1}$$

Let us consider the characteristic function which generates accumulated charge and heat at each part as

$$\mathcal{Z}(\{\chi_{\alpha s}\};B) = \langle W^\dagger e^{iH\tau}W^2 e^{-iH\tau}W^\dagger\rangle, \tag{A2}$$

$$W = \prod_s \exp[-i(\chi_{hs}H_s + \chi_{cs}N_s)/2], \tag{A3}$$

where χ_{cs} and χ_{hs} are the counting fields for measuring charge and heat currents at the part s, respectively. Note some differences between this characteristic function and Eq. (10). One difference is that this includes the counting field for heat, and the other difference is that this has counting fields of QDs, χ_{cd} and χ_{hd}. In the absence of the counting fields of QDs ($\chi_{cd}=\chi_{hd}=0$), we can derive Eq. (A2) in the same kind of protocol as in the Sec. III. Counting fields of QDs in this form are introduced only for convenience in calculations. As clarified later, counting fields of QDs disappear in the CGF, which is defined as

$$\mathcal{F}(\{\chi_{\alpha s}\};B) = \lim_{\tau\to\infty}\frac{1}{\tau}\ln\,\mathcal{Z}(\{\chi_{\alpha s}\};B). \tag{A4}$$

Here, $\alpha=c$ or h. Important physics in deriving symmetry (A9) is the microscopic reversibility. Let Θ be the time-reversal operator,[29] which satisfies

$$\Theta i\Theta^{-1} = -i, \tag{A5}$$

$$\langle n|\mathcal{O}|n'\rangle = \langle\tilde{n}'|\Theta\mathcal{O}^\dagger\Theta^{-1}|\tilde{n}\rangle, \tag{A6}$$

where $\mathcal{O}$ is an arbitrary operator and $|\tilde{n}\rangle=\Theta|n\rangle$. Using the operator Θ, we obtain the relation

$$\mathrm{Tr}[W^\dagger e^{iH\tau}W^2e^{-iH\tau}W^\dagger\rho_0] = \sum_n\langle n|W^\dagger e^{iH\tau}W^2e^{-iH\tau}W^\dagger\rho_0|n\rangle = \sum_n\langle\tilde{n}|\rho_0W^\dagger e^{-iH\tau}W^2e^{iH\tau}W^\dagger|\tilde{n}\rangle|_{B\to-B} = \mathrm{Tr}[\rho_0W^\dagger e^{-iH\tau}W^2e^{iH\tau}W^\dagger]|_{B\to-B}. \tag{A7}$$

Using the definition of W, this is simplified as

$$\mathcal{Z}(\{\chi_{\alpha s}\};B) = \mathcal{Z}(\{-\chi_{\alpha s}+iA_{\alpha s}\};-B), \tag{A8}$$

where $A_{cs}=\beta_s\mu_s$ and $A_{hs}=-\beta_s$. This is the key equality to obtain the symmetry in the CGF. Relation (A8) is obtained using the special form of QDs' counting field and initial condition. However, the detailed form of the initial condition of QDs does not affect steady-state properties. As clarified later, counting fields of QDs disappear in the CGF also.

At a stationary state, no extra charge and energy are accumulated inside the QDs. Because both the energy and number of charge inside QDs have upper bounds, any orders of fluctuations cannot increase linearly in time. This implies that the CGF cannot generate any finite values by taking derivatives with respect to $\chi_{\alpha d}$. This means that the CGF does not include the counting fields of QDs. Furthermore, from the gauge invariance and translational invariance in time, the CGF should depend only on differences between counting fields of reservoirs. This counting-field dependence is proven in Appendix B.

Now, suppose the following two ingredients: (1) relation (A8) holds and (2) the CGF does not include counting fields of QDs and depends on the difference between counting fields of reservoirs, that is, the CGF can be regarded as the function $\boldsymbol{\chi}_{\alpha r}=\chi_{\alpha r}-\chi_{\alpha m}$. The second ingredient means that the function Z is expanded using the CGF which is a function of $\boldsymbol{\chi}_{\alpha r}$ as

$$\mathcal{Z}(\{\chi_{\alpha s}\};B)=e^{\tau\mathcal{F}(\{\boldsymbol{\chi}_{\alpha r}\};B)}+\text{lower terms of }\tau.$$

Note that the first ingredient holds for arbitrary τ. Taking account of these properties, we can immediately obtain the symmetry

$$F(\{\boldsymbol{\chi}_{\alpha r}\};B)=F(\{-\boldsymbol{\chi}_{\alpha r}+i\mathcal{A}_{\alpha r}\};-B), \tag{A9}$$

where $\mathcal{A}_{\alpha r}$ is the affinity (thermodynamics force) $\mathcal{A}_{\alpha r}=A_{\alpha r}-A_{\alpha m}$. At a uniform temperature without counting fields for heat, it leads to symmetry (13).

APPENDIX B: COUNTING-FIELD DEPENDENCE OF THE CGF

We present the proof that the CGF depends only on $\boldsymbol{\chi}_{\alpha r}$. We use the Schwinger-Keldysh approach.[5,9,30] The whole expression of the CGF consists of the noninteracting part $\mathcal{F}_0$ for $H_{\rm int}=0$ and the interacting part $\mathcal{F}_{\rm int}$:

$$\mathcal{F}=\mathcal{F}_0+\mathcal{F}_{\rm int}. \tag{B1}$$

We follow the standard procedure with perturbation series for $H_{\rm int}$. First we consider F_0 obtained as

$$\mathcal{F}_0(\{\chi_{\alpha s}\};B)=-\frac{1}{2\pi}\int_{-\infty}^{\infty}d\omega\,\ln\det\hat{g}(\omega)+\text{const}. \tag{B2}$$

The inverse of the matrix Green's function $\hat{g}(\omega)$ is given in the Keldysh space as

$$[\hat{g}^{-1}]_{i\sigma,j\sigma'}=\omega\delta_{ij}\hat{\tau}_3-t_{ij}\hat{\tau}_3-\sum_r\hat{\tau}_3\hat{\Sigma}_{rij}(\omega)\hat{\tau}_3-\hat{\eta}_d \tag{B3}$$

for $\sigma=\sigma'$ and 0 otherwise. The operator $\hat{\tau}_3$ is the Pauli matrix $\hat{\tau}_3=\text{diag}(1,-1)$. In the wide-band limit

$$\Gamma_{rij}=2\pi\sum_k t_{rki}t^*_{rkj}\delta(\omega-\varepsilon_{rk}), \tag{B4}$$

the self-energy matrices are written as

$$\hat{\Sigma}_{rij}(\omega)=\begin{pmatrix}\Sigma^{++}_{rij}(\omega) & \Sigma^{+-}_{rij}(\omega)\\ \Sigma^{-+}_{rij}(\omega) & \Sigma^{--}_{rij}(\omega)\end{pmatrix}, \tag{B5}$$

$$\Sigma^{\pm\pm}_{rij}(\omega)=-i\Gamma_{rij}[1/2-f^+_r(\omega)], \tag{B6}$$

$$\Sigma^{\pm\mp}_{rij}(\omega)=\pm i\Gamma_{rij}f^{\pm}_r(\omega)\exp\{\pm i(\chi_{hr}\omega+\chi_{cr})\}. \tag{B7}$$

The term $\hat{\eta}_d$ has an infinitesimal contribution dependent on the counting fields and the initial state of the QDs. This is crucial from the causality[30] but negligible compared with the self-energy terms from reservoirs. This is plausible because the steady state should not depend on the initial state of the QDs. Let us consider the rotation operator in the Keldysh space,

$$\hat{R}=\exp[i(\chi_{hm}\omega+\chi_{cm})\hat{\tau}_3/2], \tag{B8}$$

and apply it to Green's function

$$\hat{R}^\dagger\hat{g}_{i\sigma j\sigma'}(\omega)\hat{R}. \tag{B9}$$

As easily checked, this does not change Eq. (B2). This only replaces $\chi_{\alpha r}$ with $\boldsymbol{\chi}_{\alpha r}$ in Green's function. Hence, we can regard the CGF as a function of $\boldsymbol{\chi}_{\alpha r}$.

We next consider the interaction part $F_{\rm int}$ based on the linked cluster expansions. It is formally written as[31]

$$\mathcal{F}_{\rm int}=\lim_{\tau\to\infty}\ln[e^{iS^+_{\rm int}-iS^-_{\rm int}}e^{iS_J}|_{J_{i\sigma}=J^*_{i\sigma}=0}]/\tau,$$

$$S_J=-\sum_{ij\sigma}\int_{-\tau/2}^{\tau/2}dtdt'\hat{J}_{i\sigma}(t)^\dagger\hat{\tau}_3\hat{g}_{i\sigma j\sigma}(t,t')\hat{\tau}_3\hat{J}_{j\sigma}(t'),$$

$$S^{\pm}_{\rm int}=-\int_{-\tau/2}^{\tau/2}dtH_{\rm int}\left[-i\frac{d}{dJ^*_{i\sigma\pm}(t)},i\frac{d}{dJ_{i\sigma\pm}(t)}\right], \tag{B10}$$

where the Grassmann source field in the Keldysh space $\hat{J}_{i\sigma}={}^t(J_{i\sigma+},J_{i\sigma-})$ is used. The function $H_{\rm int}[\cdots]$ is obtained by substituting the derivatives of the Grassmann numbers for the fermion operators in the Hamiltonian $H_{\rm int}$. Let us consider the Fourier transformation

$$J_{i\sigma s}(t)=\tau^{-1}\sum_\omega J_{i\sigma s}(\omega)e^{-i\omega t},$$

$$J^*_{i\sigma s}(t)=\tau^{-1}\sum_\omega J^*_{i\sigma s}(\omega)e^{i\omega t},$$

where $s=+$ or $-$. Then S_J and $S^s_{\rm int}$ are written as

$$S_J=-\frac{1}{\tau}\sum_{i,j,\sigma}\sum_\omega\hat{J}_{i\sigma}(\omega)^\dagger\hat{\tau}_3\hat{g}_{i\sigma j\sigma}(\omega)\hat{\tau}_3\hat{J}_{j\sigma}(\omega), \tag{B11}$$

$$S^s_{\rm int}=-\frac{1}{\tau^3}\sum_{\substack{\omega_1,\omega_2\\ \omega_3,\omega_4}}\sum_{ij\sigma\sigma'}\frac{U_{ij}}{2}\frac{d}{dJ^*_{i\sigma s}(\omega_1)}\frac{d}{dJ^*_{j\sigma' s}(\omega_2)}\times\frac{d}{dJ_{j\sigma' s}(\omega_3)}\frac{d}{dJ_{i\sigma s}(\omega_4)}\delta_{\omega_1+\omega_2,\omega_3+\omega_4}. \tag{B12}$$

For this representation, we consider the canonical transformation which does not change the value of Eq. (B10),

$$\widetilde{J}_{i\sigma s}(\omega) = J_{i\sigma s}(\omega)\exp\{-is(\chi_{hm}\omega + \chi_{cm})/2\}, \quad \text{(B13)}$$

$$\widetilde{J}^{*}_{i\sigma s}(\omega) = J^{*}_{i\sigma s}(\omega)\exp\{is(\chi_{hm}\omega + \chi_{cm})/2\}. \quad \text{(B14)}$$

This is equivalent to transformation (B9). Thus, $\mathcal{F}_{\rm int}$ can be regarded as a function of $\boldsymbol{\chi}_{\alpha r}$.

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