Fluctuation theorem in a quantum-dot Aharonov-Bohm interferometer

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In the present study, we investigate the full counting statistics in a two-terminal Aharonov-Bohm interferometer embedded with an interacting quantum dot. We introduce a nonequilibrium saddle-point solution for a cumulant-generating function, which satisfies the fluctuation theorem and accounts for the interaction in the mean-field level approximation. The approximation properly leads to the following two consequences. (i) The nonlinear conductance can be an uneven function of the magnetic field for a noncentrosymmetric mesoscopic system. (ii) The nonequilibrium current fluctuations couple with the charge fluctuations via the Coulomb interaction. As a result, nontrivial corrections appear in the nonequilibrium current noise. Nonlinear transport coefficients satisfy universal relations imposed by microscopic reversibility, though the scattering matrix itself is not reversible. When the magnetic field is applied, the skewness in equilibrium can be finite owing to the interaction. The equilibrium skewness is an odd function of the magnetic field and is proportional to the asymmetric component of the nonlinear conductance. The universal relations predicted can be confirmed experimentally.

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I. INTRODUCTION

Microscopic reversibility is a key ingredient in deriving the Onsager relations and has played a fundamental role in establishing the linear-response theory.[1] Recently, microscopic reversibility was used to develop a new relationship that would be valid beyond the linear-response regime. This is now known as the fluctuation theorem (FT).[2] The FT relates probabilities between positive and negative entropy productions; provides a precise statement for the second law of thermodynamics; and remarkably, reproduces the linear-response theory, the Kubo formula and the Onsager relations.[2]

In the last few years, the full counting statistics (FCS) has been recognized as a suitable framework for the FT in quantum transport.[3–8] FCS provides comprehensive statistical properties for charge transport far from equilibrium.[9–11] It addresses the probability distribution $P(q)$ of the charge q which is transmitted during time τ, and its cumulant-generating function (CGF)

$$\mathcal{F}(\lambda)=\lim_{\tau\to\infty}\ln \mathcal{Z}(\lambda)/\tau, \quad \mathcal{Z}(\lambda)=\sum_q P(q)e^{iq\lambda}. \tag{1}$$

Here $\mathcal{Z}$ is the characteristic function and λ is called the counting field.[9] Recently, the FT was generalized to the quantum transport regime in the presence of interaction and a magnetic field B.[5] For two-terminal systems, the FT is

$$\mathcal{F}(\lambda;B)=\mathcal{F}(-\lambda+i\mathcal{A};-B), \tag{2}$$

$$P(q;B)=P(-q;-B)e^{q\mathcal{A}}, \tag{3}$$

where $\mathcal{A}$ is the affinity $\mathcal{A}=V/T$, the ratio between voltage V and temperature T $(e=\hbar=k_B=1)$. One important consequence from Eq. (2) is the universal relations among transport coefficients.[5] The transport coefficient L is introduced by expanding the current cumulant with respect to $\mathcal{A}$,

$$\langle\langle I^n\rangle\rangle=\left.\frac{\partial^n\mathcal{F}(\lambda;B)}{\partial(i\lambda)^n}\right|_{\lambda=0}=\sum_{m=0}^{\infty}L_m^n(B)\frac{\mathcal{A}^m}{m!}, \tag{4}$$

where $I=q/\tau$. The FT [Eq. (2)] leads to the Kubo formula $L_1^1=L_0^2/2$ and the Onsager relation $L_{1,-}^1=0$, where $L_{m,\pm}^n=L_m^n(B)\pm L_m^n(-B)$ is the symmetrized/antisymmetrized transport coefficient. Furthermore, nontrivial relations among higher-order coefficients are obtained[5]

$$L_{2,-}^1=\frac{1}{3}L_{1,-}^2=\frac{1}{6}L_{0,-}^3, \quad L_{2,+}^1=L_{1,+}^2, \quad L_{0,+}^3=0. \tag{5}$$

This is significant in that the skewness $L_{0,-}^3$ can be finite even in equilibrium and proportional to the asymmetric component of nonlinear conductance $L_{2,-}^1$ as well as the linear response of noise $L_{1,-}^2$. Recently the validity of Eq. (2) was confirmed in a different approach.[12]

The FT [Eq. (2)] has some counterintuitive aspects in a mean-field picture.[7] When an interacting mesoscopic conductor possesses no mirror symmetry, the nonequilibrium charge accumulation inside the conductor is not symmetric in the magnetic field.[13] Then, the potential landscape generated by the nonequilibrium charge accumulation is not symmetric either. This implies that the S matrix is not reversible with respect to a magnetic field $S_{\rm LR}(B)\neq S_{\rm RL}(-B)$, which generates the magnetic-field asymmetric component of the nonlinear conductance (electrical magnetochiral effect[14]) and "violates" the Onsager relation.[7,13,15,16] In fact, in Ref. 7, it was shown that the noninteracting theory of FCS modified by the self-consistent Hartree potential cannot reproduce the FT [Eq. (2)] for the Mach-Zehnder interferometer realized in chiral edge states of the integer quantum-Hall effect. We consider that a more systematic mean-field technique accounting for relevant current fluctuations is necessary to reproduce desired symmetry of the FT. To this end, we consider a quantum dot (QD) embedded in a two-terminal Aharonov-Bohm

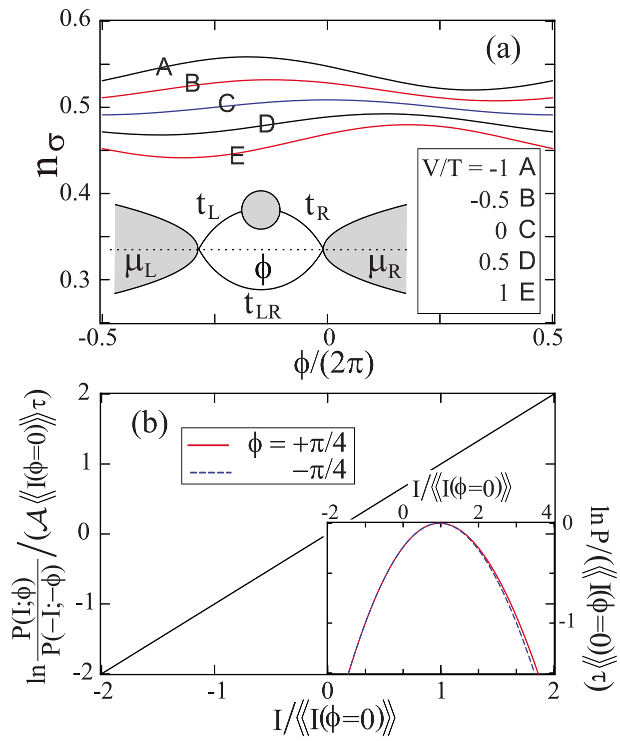

FIG. 1. (Color online) (a) AB phase dependent nonequilibrium charge accumulation. (inset) Quantum-dot AB interferometer. Mirror symmetry along the horizontal axis (dotted line) is absent. (b) Demonstration of the fluctuation theorem [Eq. (3)]. The inset shows probability distributions for positive and negative magnetic fields. Parameters: $\Gamma_L/\Gamma=0.25$, $\Gamma_R/\Gamma=0.75$, $t_{ref}=0.25$, $\epsilon_D=0$, and $U=T=V=\Gamma$.

(AB) interferometer [inset in Fig. 1(a)].[17] We introduce a nonequilibrium saddle-point solution of CGF, which realizes the FT [Eq. (2)] and the lack of reversibility in the S matrix simultaneously. It is achieved by introducing the "counting field for the dot charge" in addition to the dot potential, which are functions of λ. The solution accounts for nonequilibrium charge accumulation and current fluctuations in the Hartree-level approximation. We will also check Eq. (5) explicitly and demonstrate that the equilibrium skewness under the magnetic field is a consequence of the Coulomb interaction.

The outline of the paper is as follows. In Sec. II we introduce the microscopic Hamiltonian and the CGF in the real-time path-integral representation. With the help of this representation, a systematic saddle-point approximation is developed far from equilibrium. In Sec. III, we discuss that our saddle-point solution, which is consistent with the Hartree approximation, properly describes the magnetic-field induced asymmetry in the nonequilibrium charge accumulation and the nonlinear conductance. In Sec. IV, we address the nonequilibrium current noise. It behaves qualitatively in different way than that for noninteracting systems because the current fluctuations couple with the density fluctuations by the Coulomb interaction. This result is consistent with the Hartree approximation *for current noise* with careful treatment of the conservation law.[18] The noninteracting FCS theory with the self-consistent dot potential would fail to account for this coupling between the current fluctuations and the charge fluctuations. Then in Sec. V, we will derive general expressions for the third-order nonlinear transport coefficients [Eqs. (53)–(55)] and demonstrate that they satisfy the extension of the Onsager relations [Eq. (5)] proposed in Ref. 5. Section VI summarizes our results.

II. CUMULANT-GENERATING FUNCTION

A. Quantum-dot Aharonov-Bohm interferometer

The system consists of left (L) and right (R) leads, two arms, and a QD. Electrons can travel through the QD and the lower reference arm [inset in Fig. 1(a)]. The total Hamiltonian is

$$H = \sum_{r=\mathrm{L,R}} H_r + H_D + H_T + H_{\mathrm{ref}}, \tag{6}$$

where the on-site Coulomb interaction U in the QD is accounted for by

$$H_D = \sum_{\sigma=\uparrow,\downarrow} \epsilon_D d_\sigma^\dagger d_\sigma + U d_\uparrow^\dagger d_\uparrow d_\downarrow^\dagger d_\downarrow . \tag{7}$$

The operator d_σ annihilates an electron with spin σ. The leads are modeled by

$$H_r = \sum_{k\sigma} \varepsilon_{rk\sigma} a_{rk\sigma}^\dagger a_{rk\sigma}, \tag{8}$$

where $a_{rk\sigma}$ annihilates electrons in the lead r with spin σ and wave vector k. The tunneling and the reference arm are described as

$$H_T = \sum_{rk\sigma} t_r d_\sigma^\dagger a_{rk\sigma} + \mathrm{H.c.}, \tag{9}$$

$$H_{\mathrm{ref}} = \sum_{kk'\sigma} t_{\mathrm{LR}} e^{i\phi} a_{\mathrm{R}k\sigma}^\dagger a_{\mathrm{L}k'\sigma} + \mathrm{H.c.} \tag{10}$$

The magnetic field B pierces through the ring and the electrons acquire the AB phase ϕ, which satisfies $\phi(B) = -\phi(-B)$. The initial density matrices at both leads are assumed to have an equilibrium distribution with the left and right chemical potentials μ_L and μ_R. For simplicity, we will consider the symmetric case $\mu_L = -\mu_R = V/2$.

B. Nonequilibrium saddle-point approximation

We confine ourselves to high temperature and treat the interaction in a mean-field level approximation.[19] In order to find a proper saddle-point solution out of equilibrium, we employ the real-time path-integral approach.[11,20,21] The characteristic function, which is the partition function in the Keldysh formalism, reads

$$\mathcal{Z}(\lambda) = \int \mathcal{D}[a_{rk\sigma}^*, d_\sigma^*, a_{rk\sigma}, d_\sigma] \exp\left[i \int_C dt \mathcal{L}(t) \right], \tag{11}$$

where C is the closed time path. The Lagrangian is given by

$$\mathcal{L} = \sum_{rk\sigma} a^*_{rk\sigma}(i\partial_t - \varepsilon_{rk\sigma})a_{rk\sigma} + \sum_{\sigma} d^*_{\sigma}(i\partial_t - \epsilon_{\rm D})d_{\sigma} - \sum_{rk\sigma}(t_r e^{i\varphi_r} d^*_{\sigma} a_{rk\sigma} + \text{c.c.}) - U d^*_{\uparrow} d_{\uparrow} d^*_{\downarrow} d_{\downarrow} - \sum_{kk'\sigma}(t_{\rm LR} e^{-i\varphi_{\rm R} + i\varphi_{\rm L} + i\phi} a^*_{{\rm R}k\sigma} a_{{\rm L}k\sigma} + \text{c.c.}), \quad (12)$$

where the phase on the upper/lower branch of the closed time path $\varphi_{r\pm}$ is related to the counting field as $\varphi_{r\pm} = \pm\lambda_r/2$. We introduce the auxiliary field, the dot potential v_σ, via the Stratonovich-Hubbard transformation: $U d^*_\uparrow d_\uparrow d^*_\downarrow d_\downarrow \to \Sigma_\sigma v_\sigma d^*_\sigma d_\sigma - v_\uparrow v_\downarrow / U$,[22]

$$\mathcal{Z}(\lambda) = \int \mathcal{D}[v_\sigma]\mathcal{Z}_0(\lambda, v_\sigma)\exp\left(\frac{i}{U}\int_C dt v_\uparrow(t) v_\downarrow(t)\right), \quad (13)$$

where $\mathcal{Z}_0(\lambda, v_\sigma)$ is the Keldysh partition function for the noninteracting case $U=0$ with a shift in the QD level $\epsilon_{\rm D} \to \epsilon_{\rm D} + v_\sigma(t)$ for spin σ. Although we limit ourselves to the time-independent stationary solution in the nonmagnetic phase,[19] we allow different dot potentials for upper and lower branches of C,[23]

$$v_{\sigma\pm}(t) = v_\pm = v_c \pm i v_q/2. \quad (14)$$

The classical component v_c is the dot potential generated by accumulation of charges with opposite spin. The quantum component v_q plays the role of the counting field for charge in QD.[21,24] Then the total CGF reads

$$\mathcal{F}(\lambda) = \mathcal{F}_0(\lambda) - 2 v_c v_q / U, \quad (15)$$

where $\mathcal{F}_0$ is a bare part related to $\mathcal{Z}_0$. After a number of calculations, the bare part is represented by the S matrix (Appendix A)

$$\mathcal{F}_0(\lambda) = \frac{1}{\pi}\int d\omega \, \text{Tr} \ln[\tilde{1} - \tilde{f}\tilde{K}(\lambda)], \quad (16)$$

$$\tilde{K}(\lambda) = \tilde{1} - e^{i\tilde{\lambda}} \mathcal{S}^\dagger(v_-) e^{-i\tilde{\lambda}} \mathcal{S}(v_+), \quad (17)$$

$$\mathcal{S}(v) = \begin{bmatrix} S_{\rm LL}(v) & S_{\rm LR}(v) \\ S_{\rm RL}(v) & S_{\rm RR}(v) \end{bmatrix}, \quad (18)$$

where $\tilde{1}$ is a unit matrix and $\tilde{\lambda} = \text{diag}(\lambda, 0)$ with $\lambda = \lambda_{\rm L} - \lambda_{\rm R}$. Here the CGF depends only on the difference between two counting fields because of the charge conservation.[5] $\tilde{f} = \text{diag}(f_{\rm L}, f_{\rm R})$ consists of the Fermi-distribution function

$$f_r(\omega) = \frac{1}{\exp[(\omega - \mu_r)/T] + 1}. \quad (19)$$

When the potential v is independent of the magnetic field, the S matrix is reversible

$$S_{rr'}(v; B) = S_{r'r}(v; -B). \quad (20)$$

Explicit forms are given as

$$S_{rr}(v) = 1 - \frac{i\Gamma_r + t_{\rm ref}\sqrt{\Gamma_{\rm L}\Gamma_{\rm R}}\cos\phi - t_{\rm ref}^2 \epsilon(v)/2}{\Delta(v)}, \quad (21)$$

$$S_{\rm RL}(v) = [e^{i\phi} t_{\rm ref}\epsilon(v) - \sqrt{\Gamma_{\rm L}\Gamma_{\rm R}}]/\Delta(v), \quad (22)$$

$$\Delta(v) = \frac{t_{\rm ref}\sqrt{\Gamma_{\rm L}\Gamma_{\rm R}}\cos\phi}{2} - \left(1 + \frac{t_{\rm ref}^2}{4}\right)\epsilon(v) + i\frac{\Gamma}{2}, \quad (23)$$

where $\epsilon(v) = \epsilon_{\rm D} + v - \omega$. The tunnel coupling $\Gamma = \Gamma_{\rm L} + \Gamma_{\rm R}$ is written with the density of states of the lead as $\Gamma_r = 2\pi t_r^2 \rho_r$. Hopping through the reference arm is characterized by $t_{\rm ref} = 2\pi t_{\rm LR}\sqrt{\rho_{\rm L}\rho_{\rm R}}$. It appears that the CGF [Eqs. (16)–(18)] is that for the joint probability distribution of current and charge.[21,24]

However, the "charge counting field" v_q is a function of λ and now is determined by coupled saddle-point equations

$$v_c = \frac{U}{2}\frac{\partial \mathcal{F}_0}{\partial v_q}, \quad v_q = \frac{U}{2}\frac{\partial \mathcal{F}_0}{\partial v_c}. \quad (24)$$

As we will discuss in the Sec. III, if the counting field is zero $\lambda=0$, the quantum component is zero $v_q=0$ since the upper and lower branches of the closed time path are symmetric. Then the first equation is reduced to the ordinary Hartree equation. However, for finite λ, the upper branch and the lower branch are no longer symmetric. Then the quantum component v_q, the charge counting field, is expected to be finite. Then we have to determine v_q self-consistently as well.

III. MAGNETIC-FIELD-INDUCED ASYMMETRY IN NONLINEAR TRANSPORT

In this section, we will demonstrate that the saddle-point solution [Eq. (24)] captures magnetic-field asymmetry in the nonlinear transport regime.[7,15] For $\lambda=0$, Eq. (24) possesses a trivial solution: $v_q=0$ and $v_c=v^*$ determined by the nonequilibrium Hartree equation,

$$v^* = \frac{U}{2\pi}\frac{\partial}{\partial \epsilon_{\rm D}}\int d\omega \, \text{Im Tr} \, \tilde{f} \ln \mathcal{S}(v^*) = \frac{U}{2\pi}\int d\omega A_\sigma(\omega), \quad (25)$$

$$A_\sigma = \sum_r (\Gamma_r + t_{\rm ref}^2 \Gamma_{\bar{r}}/4)[f_r(\omega) - 1/2]/|\Delta(v^*)|^2 + t_{\rm ref}\sqrt{\Gamma_{\rm L}\Gamma_{\rm R}}\sin\phi[f_{\rm L}(\omega) - f_{\rm R}(\omega)]/|\Delta(v^*)|^2, \quad (26)$$

where $\bar{r} = {\rm L/R}$ for $r = {\rm R/L}$. Figure 1(a) shows the AB flux (the magnetic field) dependence of charge accumulation inside the QD, $n_\sigma = v^*/U + 1/2$. In equilibrium $V=0$, n_σ is an even function of the magnetic field. For $V \neq 0$, because of the second line of Eq. (26), which is related to the lack of mirror symmetry, the charge accumulation becomes an uneven function of the AB flux $n_\sigma(\phi) \neq n_\sigma(-\phi)$.

The average of the charge current is obtained by differentiating the CGF in terms of λ.

$$\frac{d\mathcal{F}}{d(i\lambda)} = \frac{\partial \mathcal{F}_0}{\partial(i\lambda)} + \sum_{\alpha=c,q}\left(\frac{\partial \mathcal{F}_0}{\partial v_\alpha}\frac{dv_\alpha}{d(i\lambda)} - 2\frac{v_{\bar{\alpha}}}{U}\frac{dv_\alpha}{d(i\lambda)}\right) = \frac{\partial \mathcal{F}_0}{\partial(i\lambda)}, \tag{27}$$

where $\bar{\alpha}=c/q$ for $\alpha=q/c$. All contributions except $\mathcal{F}_0$ cancel because of the conditions [Eq. (24)]. Then, the Landauer formula with the transmission probability $\mathcal{T}=|S_{\rm LR}(v^*)|^2$ is obtained,

$$\langle\langle I\rangle\rangle = \left.\frac{d\mathcal{F}_0}{d(i\lambda)}\right|_{\lambda=0} = \frac{1}{\pi}\int d\omega \mathcal{T}(\omega)[f_{\rm L}(\omega)-f_{\rm R}(\omega)], \tag{28}$$

$$\mathcal{T}(\omega) = \frac{\Gamma_{\rm L}\Gamma_{\rm R} + t_{\rm ref}^2\epsilon(v^*)^2 - 2t_{\rm ref}\epsilon(v^*)\sqrt{\Gamma_{\rm L}\Gamma_{\rm R}}\cos\phi}{|\Delta(v^*)|^2}. \tag{29}$$

The transmission probability [Eq. (29)] fully accounts for the phase coherence. Because an electron can travel around the ring many times, it picks up AB phase many times.[25] Then the denominator of the transmission probability [Eq. (23)] also depends on the AB phase, which causes higher harmonics of the AB oscillations. In the absence of interaction $U=0$, and thus $v^*=0$, the transmission probability is symmetric in the magnetic field.[17] For finite U and V, because the nonequilibrium charge accumulation is not symmetric in the magnetic field $v^*(B)\neq v^*(-B)$, the reversibility of S matrix breaks down,

$$S_{\rm LR}[v^*(B);B] \neq S_{\rm RL}[v^*(-B);-B]. \tag{30}$$

It leads to the magnetic-field asymmetry in the nonlinear conductance.

Though the S matrix is not reversible, the CGF [Eq. (16)] with Eqs. (24), fulfills the FT [Eq. (2)]. The bare CGF [Eq. (16)] is for the joint probability distribution of current and charge dwelling inside the QD. It is a function of the two counting fields, λ and v_q, and the QD potential v_c. We observe that it possesses the symmetry

$$\mathcal{F}_0(\lambda,v_q,v_c;B) = \mathcal{F}_0(-\lambda+i\mathcal{A},v_q,v_c;-B). \tag{31}$$

The equation means that under the magnetic-field reversal, the counting field for current changes as $\lambda\to-\lambda+i\mathcal{A}$ while that for charge remains unchanged $v_q\to v_q$. The difference appears because, under the time-reversal operation, the current operator changes sign while the charge operator does not. After solving the self-consistent Eq. (24), both v_c and v_q get the dependence of the magnetic field B and the counting field for current λ. However they still satisfy conditions $v_c(\lambda;B)=v_c(-\lambda+i\mathcal{A};-B)$ and $v_q(\lambda;B)=v_q(-\lambda+i\mathcal{A};-B)$. The conditions ensure that our saddle-point approximation preserves the FT [Eq. (2)]. Equation (31) is another key symmetry. We will utilize it to derive general relations among current- and density-correlation functions in order to obtain simple expressions for the third-order nonlinear transport coefficients [Eqs. (53)–(55)] in Sec. V.

Figure 1(b) demonstrates the FT [Eq. (3)], though probability distributions for positive and negative magnetic fields are different [inset of Fig. 1(b)]. Then we conclude that the magnetic-field asymmetry does not necessarily contradict the FT. If we substitute $v_q=0$ and $v_c=v^*$ in Eqs. (16)–(18), our CGF may be compatible with that in Ref. 7 at the formal level. However, this approximation does not necessary preserve the FT [Eq. (2)].

IV. NONEQUILIBRIUM NOISE

In the presence of the Coulomb interaction, the nonequilibrium noise becomes qualitatively different from that for noninteracting systems since the current fluctuations and the density fluctuations couple by the Coulomb interaction. It was demonstrated before that the Hartree-level approximation is able to capture this physics once the conservation law is properly accounted for.[18] In this section, we derive the current noise out of equilibrium and check that our saddle-point approximation gives a consistent result with the previous theory.[18] We point out that for this it is crucial to determine self-consistently v_q as well as v_c.

Let us consider the derivative of Eq. (24) with respect to the counting field

$$\frac{dv_{\bar{\alpha}}}{d(i\lambda)} = \frac{U}{2}\frac{\partial^2\mathcal{F}_0}{\partial v_\alpha\,\partial(i\lambda)} + \frac{U}{2}\sum_{\alpha'=c,q}\frac{\partial^2\mathcal{F}_0}{\partial v_\alpha\,\partial v_{\alpha'}}\frac{dv_{\alpha'}}{d(i\lambda)}. \tag{32}$$

We introduce a symmetric matrix $U_{\alpha\beta}=U_{\beta\alpha}$ satisfying the following relation,

$$\sum_{\gamma=c,q}\left(\frac{1-\delta_{\alpha\gamma}}{U} - \frac{1}{2}\frac{\partial^2\mathcal{F}_0}{\partial v_\alpha\,\partial v_\gamma}\right)U_{\gamma\beta} = \delta_{\alpha\beta}, \tag{33}$$

where $\delta_{\alpha\beta}$ is the Kronecker's delta. Then we solve Eq. (32) as

$$\frac{dv_{\bar{\alpha}}}{d(i\lambda)} = \sum_{\alpha'}\frac{U_{\alpha\alpha'}}{2}\frac{\partial^2\mathcal{F}_0}{\partial v_{\alpha'}\,\partial(i\lambda)}. \tag{34}$$

For $\lambda=0$, which implies that $v_q=0$ and $v_c=v^*$, the four components are $U_{cc}|_{\lambda=0}=U_{\rm eff}^2 S_{NN}$, $U_{cq}|_{\lambda=0}=U_{cq}|_{\lambda=0}=U_{\rm eff}$, and $U_{qq}|_{\lambda=0}=0$. Here the Coulomb interaction is screened,

$$U_{\rm eff} = U/(1-U\chi_{NN}). \tag{35}$$

The bare density-density response function χ_{NN} and the density-density correlation function (charge noise) S_{NN} are given by

$$\chi_{NN} = \left.\frac{1}{2}\frac{\partial^2\mathcal{F}_0}{\partial v_c\,\partial v_q}\right|_{\lambda=0} = \frac{\partial n_\sigma}{\partial\epsilon_{\rm D}} \tag{36}$$

$$S_{NN} = \left.\frac{1}{2}\frac{\partial^2\mathcal{F}_0}{\partial v_q^2}\right|_{\lambda=0} \tag{37}$$

$$= \frac{1}{2\pi}\int d\omega\left[\frac{(1+t_{\rm ref}^2/4)^2\Gamma^2}{4|\Delta(\epsilon^*)|^4} - A_\sigma(\omega)^2\right]. \tag{38}$$

In order to obtain Eq. (38), one need to be careful about the analytic properties [one can simply add $(\partial^2\mathcal{F}_0/\partial v_c^2)/8|_{\lambda=0}$, which is zero from the normalization condition or the causality[26]].

We calculate the current noise by performing the second derivative of the CGF in terms of the counting field λ. With the help of Eq. (34), the derivative of Eq. (27) reads

$$\frac{d^2\mathcal{F}}{d(i\lambda)^2} = \frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)^2} + \sum_\alpha \frac{dv_\alpha}{d(i\lambda)}\frac{\partial^2\mathcal{F}_0}{\partial v_\alpha \partial(i\lambda)} \tag{39}$$

$$= \frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)^2} + \sum_{\alpha,\alpha'} \frac{\partial^2\mathcal{F}_0}{\partial v_\alpha \partial(i\lambda)}\frac{U_{\alpha\alpha'}}{2}\frac{\partial^2\mathcal{F}_0}{\partial v_{\alpha'} \partial(i\lambda)}. \tag{40}$$

Then, by fixing $\lambda=0$, we obtain the full form of the nonequilibrium current noise as follows:

$$\langle\langle I^2\rangle\rangle = 2(S_{II} + 2S_{IN}U_{\text{eff}}\chi_{IN} + \chi_{IN}^2 S_{NN}U_{\text{eff}}^2), \tag{41}$$

where the bare current-density response and the current-density correlation functions are

$$\chi_{IN} = \frac{1}{2}\frac{\partial^2\mathcal{F}_0}{\partial v_c \partial(i\lambda)}\bigg|_{\lambda=0} = \frac{1}{2}\frac{\partial\langle\langle I\rangle\rangle}{\partial\epsilon_{\text{D}}}, \tag{42}$$

$$S_{IN} = \frac{1}{2}\frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)\,\partial v_q}\bigg|_{\lambda=0}$$
$$= \frac{1}{2\pi}\int d\omega\{t_{\text{ref}}\sqrt{\Gamma_{\text{L}}\Gamma_{\text{R}}}\sin\phi[f_{\text{L}}(\omega) + f_{\text{R}}(\omega) - 2f_{\text{R}}(\omega)$$
$$\times f_{\text{L}}(\omega)]/|\Delta(v^*)|^2 - A_\sigma(\omega)\mathcal{T}(\omega)[f_{\text{L}}(\omega) - f_{\text{R}}(\omega)]\}. \tag{43}$$

The current noise [Eq. (41)] is not just the bare current-current correlation. Namely, it is the quantum-noise formula for noninteracting systems[27] modified with the self-consistent potential v^*,

$$S_{II} = \frac{1}{2}\frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)^2}\bigg|_{\lambda=0}$$
$$= \frac{1}{2\pi}\int d\omega\mathcal{T}(\omega)[f_{\text{L}}(\omega) + f_{\text{R}}(\omega) - 2f_{\text{L}}(\omega)f_{\text{R}}(\omega)]$$
$$- \mathcal{T}(\omega)^2[f_{\text{L}}(\omega) - f_{\text{R}}(\omega)]^2. \tag{44}$$

The second and third terms of Eq. (41) are the result of the Coulomb interaction out of equilibrium [similar corrections also appear in the dynamical conductance of a mesoscopic capacitor[28]]. In equilibrium they vanish since the average current is zero $\langle\langle I\rangle\rangle=0$ and consequently $\chi_{IN}=0$. In the absence of the reference arm $t_{\text{ref}}=0$, Eq. (41) reproduces the theory of the noise for the nonequilibrium Anderson model in the Hartree-level approximation [Eqs. (68) and (89) of Ref. 18]. This theory[18] is based on the diagrammatic language. Diagrammatically, the second and third terms of Eq. (41) correspond to vertex corrections. If we only substitute $v_q=0$ and $v_c=v^*$ in Eqs. (16)–(18), we fail to account for these terms. In order to be consistent with this theory,[18] the quantum component v_q is crucial.

V. NONLINEAR TRANSPORT COEFFICIENTS

Now we come to the relations among the third-order transport coefficients [Eq. (5)]. First, we can check that the bare parts vanish for our case,

$$\partial_{i\lambda}^{3-n}\partial_{\mathcal{A}}^n\mathcal{F}_0(0,B)|_{\mathcal{A}=0} = 0 \tag{45}$$

($n=0,1,2,3$). Then, the skewness following the derivative of Eq. (39) in terms of λ reads (Appendix B)

$$L_0^3 = 6U_{\text{eff}}^{\text{eq.}}S_{IN}^{\text{eq.}}\chi_{II,N}^{\text{eq.}}, \quad \chi_{II,N} = \frac{\partial S_{II}}{\partial\epsilon_{\text{D}}}, \tag{46}$$

where $\chi_{II,N}$ is the linear response of the noise. The superscript eq. specifies that $\mathcal{A}$ is fixed to 0. Equation (46) reveals that the equilibrium skewness is caused by the Coulomb interaction. The other transport coefficients are calculated in the same manner.

$$L_1^2 = 2U_{\text{eff}}^{\text{eq.}}\chi_{NI}^{\text{eq.}}\chi_{II,N}^{\text{eq.}} + 4U_{\text{eff}}^{\text{eq.}}S_{IN}^{\text{eq.}}\chi_{I,IN}^{\text{eq.}}, \tag{47}$$

$$L_2^1 = 4U_{\text{eff}}^{\text{eq.}}\chi_{NI}^{\text{eq.}}\chi_{I,IN}^{\text{eq.}}, \tag{48}$$

where

$$\chi_{NI} = \frac{\partial n_\sigma}{\partial\mathcal{A}}, \quad \chi_{I,IN} = \frac{\partial\chi_{IN}}{\partial\mathcal{A}}. \tag{49}$$

Figure 2(a) shows the AB flux dependence of third-order nonlinear transport coefficients. It appears that the coefficients behave independently. However, as shown in panel (b), the extension of the Onsager relations [Eq. (5)] is satisfied perfectly.

Further the universal relations [Eq. (5)] within the saddle-point approximation can be proven with the help of the symmetry [Eq. (31)]. In parallel to the discussions for the nonlinear transport coefficients, the symmetry [Eq. (31)] provides general relations among the current- and density-correlation functions for noninteracting case (Appendix C),

$$S_{IN+} = 0, \quad S_{IN-} = 2\chi_{NI-}, \tag{50}$$

$$\chi_{I,IN-} = 0, \quad \chi_{II,N+} = 2\chi_{I,IN+}, \tag{51}$$

where the correlation functions with subscripts $\pm$ denotes the even/odd components in the magnetic field, such as

$$S_{IN\pm} = \frac{S_{IN}(B) \pm S_{IN}(-B)}{2}. \tag{52}$$

Equations (50) and (51) simplify the nonlinear transport coefficients [Eqs. (46)–(48)] as

$$L_{2-}^1 = \frac{L_{1-}^2}{3} = \frac{L_{0-}^3}{6} = 4U_{\text{eff}}^{\text{eq.}}\chi_{NI-}^{\text{eq.}}\chi_{I,IN+}^{\text{eq.}}, \tag{53}$$

$$L_{1+}^2 = L_{2+}^1 = 4U_{\text{eff}}^{\text{eq.}}\chi_{NI+}^{\text{eq.}}\chi_{I,IN+}^{\text{eq.}}, \tag{54}$$

$$L_{0+}^3 = 0. \tag{55}$$

Equations (53)–(55) precisely demonstrate the universal relations among nonlinear the transport coefficients [Eq. (5)].

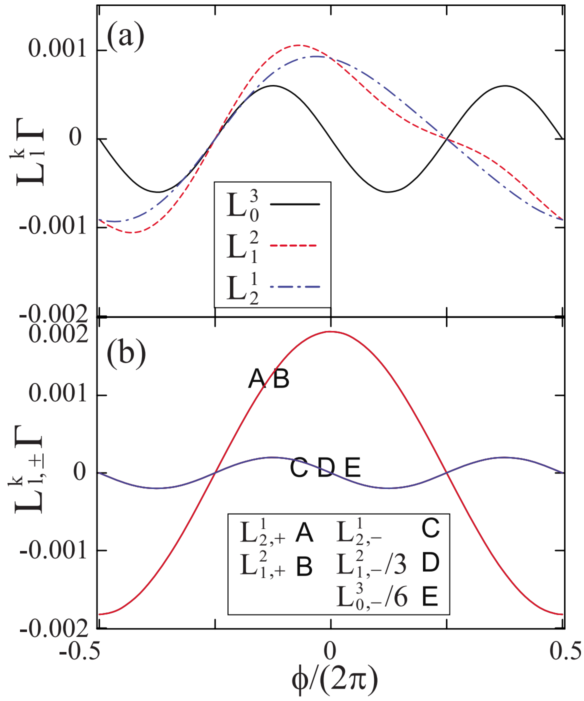

FIG. 2. (Color online) (a) Aharonov-Bohm flux dependent third-order nonlinear transport coefficients and (b) the extension of the Onsager relations. The parameters are the same as those in Fig. 1.

Here we observe a finite skewness [panel (a)], which may be counterintuitive since in equilibrium, we expect that all odd cumulants vanish, which means $P(I)=P(-I)$. However, in the presence of the magnetic field it is not necessarily the case. It is because in order to keep physical processes invariant under the time reversal, one must reverse the magnetic field $B\rightarrow -B$. Therefore, we may prove an equality not $P(I;B)=P(-I;B)$ but Eq. (3), $P(I;B)=P(-I;-B)$. Then one would conclude that the odd cumulants are zero only for $B=0$. Indeed the result properly describes this fact, namely, the skewness in equilibrium is odd in magnetic field and zero for $\phi=0$ [panel (a)].

We note that our results can be obtained using the Hartree approximation based on the nonequilibrium self-consistent Φ-derivable approximation.[29,30] In this scheme, the Keldysh generating function consists of an infinite number of closed diagrams each of which satisfies the symmetry [Eq. (2)], as shown in Ref. 5.

VI. SUMMARY

We studied the full counting statistics of a quantum-dot Aharonov-Bohm interferometer and have developed a nonequilibrium Hartree approximation, which satisfies the fluctuation theorem and describes the magnetic-field asymmetry in the nonlinear transport. We have also shown that the equilibrium skewness as well as the asymmetric component of the nonlinear conductance are results of the Coulomb interaction. They satisfy the extension of the Onsager relations [Eq. (5)],[5] which may be measured by the currently available experiments.[13] In the present paper, though we discussed the quantum-dot Aharonov-Bohm interferometer, the mean-field approximation developed in the present paper can be applicable for general systems. However, they are limited to the high-temperature regime, where electron-electron correlations play minor role. For low temperatures the Kondo correlations grow and the Hartree approximation fails.[25] Also for Mach-Zehnder interferometers, the importance of the electron correlations beyond the Hartree approximation is addressed recently.[31] It would be interesting to investigate the fluctuation theorem for the strongly correlated systems in the nonequilibrium steady state as a future problem.

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APPENDIX A: *S*-MATRIX REPRESENTATION

Usually the CGF is expressed either by the Keldysh-Green function (GF) or the *S*-matrix. To our knowledge, the direct transformation between two representations is not straightforward. In this appendix, we derive the *S*-matrix representation form the GF representation. In the limit of long measurement time, after tracing out fermions, we obtain the following form, which is written with matrices of Keldysh GFs for the dot $\mathbf{g}_D$ and the reservoirs $\mathbf{g}_{\text{res}}$ as

$$\mathcal{F}_0=\frac{1}{2\pi}\sum_\sigma\int d\omega\,\ln\det\begin{pmatrix}\mathbf{g}_D^{-1} & -\mathbf{t}^\dagger\\ -\mathbf{t} & \mathbf{g}_{\text{res}}^{-1}-\mathbf{t}_{\text{ref}}\end{pmatrix},\tag{A1}$$

where submatrices are defined in the Keldysh space as

$$\mathbf{t}=\begin{pmatrix}\tilde{t} & \tilde{0}\\ \tilde{0} & -\tilde{t}\end{pmatrix},\quad \mathbf{t}_{\text{ref}}=\begin{pmatrix}\tilde{t}_{\text{ref}} & \tilde{0}\\ \tilde{0} & -\tilde{t}_{\text{ref}}\end{pmatrix}.\tag{A2}$$

The submatrices denoted with tilde are

$$\tilde{t}=\begin{pmatrix}t_{\text{L}}\\ t_{\text{R}}\end{pmatrix},\quad \tilde{t}_{\text{ref}}=\begin{pmatrix}0 & t_{\text{LR}}e^{-i\phi}\\ t_{\text{LR}}e^{i\phi} & 0\end{pmatrix},\tag{A3}$$

and $\tilde{0}$ is a 2×2 zero matrix. For simplicity, let us assume that initially the QD is empty $\mu_D\rightarrow\infty$ (It is a trivial assumption since in the limit of long measurement time, the CGF does not depend on the initial QD state[5]). The matrix of the QD Keldysh GF reads

$$\mathbf{g}_D(\omega)^{-1}=\begin{pmatrix}g_{D+}(\omega+i0)^{-1} & 0\\ 2i0 & -g_{D-}(\omega-i0)^{-1}\end{pmatrix},\tag{A4}$$

where $g_{D\pm}(\omega)^{-1}=\omega-\epsilon_{\text{D}}-v_\pm$. In the following, we safely neglect positive infinitesimal since the reservoir GF dominates the analytic property.[5] The matrix of reservoir Keldysh GF is given as

$$\mathbf{g}_{\mathrm{res}}(\omega) = 2i\pi\tilde{\rho} \otimes \boldsymbol{\tau}^0 \begin{bmatrix} \tilde{f}(\omega) - \tilde{1}/2 & \tilde{f}(\omega)e^{i\tilde{\lambda}} \\ (\tilde{f}(\omega) - \tilde{1})e^{-i\tilde{\lambda}} & \tilde{f}(\omega) - \tilde{1}/2 \end{bmatrix}, \tag{A5}$$

where $\tilde{\rho}=\mathrm{diag}(\rho_L,\rho_R)$, $\tilde{\lambda}=\mathrm{diag}(\lambda_L,\lambda_R)$ and $\tilde{f}(\omega)=\mathrm{diag}[f_L(\omega),f_R(\omega)]$. The matrix $\boldsymbol{\tau}^0$ is an unit matrix in the Keldysh space. The form given in Eq. (A1) is not complete since it does not satisfy the normalization condition $\mathcal{F}_0(0)=0$. Therefore, we have to subtract a constant. We choose such a constant as Eq. (A1) with empty reservoirs $\mu_r \to -\infty$,

$$\frac{1}{2\pi}\sum_\sigma \int d\omega \ln \det \begin{pmatrix} \mathbf{g}_D^{-1} & -\mathbf{t}^\dagger \\ -\mathbf{t} & \mathbf{g}_{\mathrm{res}}^{e-1} - \mathbf{t}_{\mathrm{ref}} \end{pmatrix}, \tag{A6}$$

where $\mathbf{g}^e_{\mathrm{res}}$ is the reserver GF without electrons, namely, $\mathbf{g}_{\mathrm{res}}$ replaced $\tilde{f}$ with the zero matrix $\tilde{0}$. Though the matrix Eq. (A6) is a function of the counting fields, after performing the determinant, the counting fields disappears. Then by subtracting this constant from Eq. (A1), we obtain a complete form of the CGF as

$$\mathcal{F}_0 = \frac{1}{2\pi}\sum_\sigma \int d\omega \ln \det \mathbf{X}(\omega), \tag{A7}$$

$$\mathbf{X}(\omega) = [1 - \mathbf{g}_{\mathrm{res}}(\omega)\mathbf{V}(\omega)][1 - \mathbf{g}^e_{\mathrm{res}}\mathbf{V}(\omega)]^{-1}, \tag{A8}$$

$$\mathbf{V}(\omega) = \mathbf{t}_{\mathrm{ref}} + \mathbf{t}\mathbf{g}_D(\omega)\mathbf{t}^\dagger. \tag{A9}$$

One can check that Eq. (A7) satisfies the normalization condition. The matrix appears in the integrand of Eq. (A7) is transformed in the following way,

$$\mathbf{X} = \left(\mathbf{M}^{-1} - 2i\pi\tilde{\rho}\tilde{f} \otimes \boldsymbol{\tau}^0 \begin{bmatrix} \tilde{1} & e^{i\tilde{\lambda}} \\ e^{-i\tilde{\lambda}} & \tilde{1} \end{bmatrix} \right)\mathbf{M}, \tag{A10}$$

$$\mathbf{M} = \begin{bmatrix} \tilde{T}_+^{-1} & \tilde{0} \\ 2\pi i\tilde{\rho}e^{-i\tilde{\lambda}} & -\tilde{T}_-^{\dagger -1} \end{bmatrix}^{-1}. \tag{A11}$$

The T matrix,

$$\tilde{T}^{-1}_\pm(\omega) = [\tilde{t}_{\mathrm{ref}} + \tilde{t}g_{D\pm}(\omega)\tilde{t}^\dagger]^{-1} + i\pi\tilde{\rho}, \tag{A12}$$

appears in the diagonal components. Since the determinant of $\mathbf{M}$ does not depend on the counting field, $\det \mathbf{M}=\det(\tilde{T}_+)\det(-\tilde{T}_-)$, the determinant of Eq. (A10) can be simplified

$$\det \mathbf{X} = \det \begin{bmatrix} \tilde{1} + (\tilde{S}_+ - \tilde{1})\tilde{f} & (\tilde{S}_+ - \tilde{1})\tilde{f}e^{i\tilde{\lambda}} \\ (\tilde{1} - \tilde{S}_-^\dagger)(\tilde{1} - \tilde{f})e^{-i\tilde{\lambda}} & \tilde{1} + (\tilde{S}_-^\dagger - \tilde{1})\tilde{f} \end{bmatrix}, \tag{A13}$$

where submatrices are expressed with the S matrix,

$$\tilde{S}_\pm(\omega) = \tilde{1} - 2\pi i\tilde{\rho}^{1/2}\tilde{T}_\pm(\omega)\tilde{\rho}^{1/2}. \tag{A14}$$

Then by utilizing the property of the determinant

$$\det \begin{bmatrix} \tilde{v} & \tilde{w} \\ \tilde{x} & \tilde{y} \end{bmatrix} = \det(\tilde{y}\tilde{w}^{-1}\tilde{v}\tilde{w} - \tilde{x}\tilde{w}), \tag{A15}$$

Equation (A13) can be calculated as

$$\det \mathbf{X} = \det[\tilde{1} + \tilde{f}(e^{i\tilde{\lambda}}\tilde{S}_-^\dagger e^{-i\tilde{\lambda}}\tilde{S}_+ - \tilde{1})]. \tag{A16}$$

Further after the gauge transformation, we can replace $\tilde{\lambda}=\mathrm{diag}(\lambda_L,\lambda_R)$ with $\tilde{\lambda}=\mathrm{diag}(\lambda_L-\lambda_R,0)$. The CGF only depends on the difference between the two counting fields because of the charge conservation.[5] Then by combining Eqs. (A7) and (A16), we obtain Eqs. (16)–(18) [we used the notation $\mathcal{S}(v_\pm)$ for the S matrix $\tilde{S}_\pm$ in Eqs. (16)–(18)]. Though here we considered the QD AB interferometer, generalizations to multiterminal QDs (Ref. 5) are straightforward.

APPENDIX B: THIRD-ORDER NONLINEAR TRANSPORT COEFFICIENTS

In this appendix we derive the third-order transport coefficients. Let us consider the skewness. The derivative of Eq. (39) in terms of the counting field reads

$$\frac{d^3\mathcal{F}}{d(i\lambda)^3} = \frac{d^3\mathcal{F}_0}{d(i\lambda)^3} + \sum_{\alpha=c,q}\left(2\frac{dv_\alpha}{d(i\lambda)}\frac{\partial^3\mathcal{F}_0}{\partial(i\lambda)^2\,\partial v_\alpha} + \frac{\partial\mathcal{F}_0}{\partial(i\lambda)\,\partial v_\alpha} \times \frac{d^2v_\alpha}{d(i\lambda)^2} + \sum_{\beta=c,q}\frac{dv_\beta}{d(i\lambda)}\frac{dv_\alpha}{d(i\lambda)}\frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)\,\partial v_\beta\,\partial v_\alpha}\right). \tag{B1}$$

The second line of this equation contains an unknown term, namely, the second derivative of the auxiliary field in terms of the counting field $d^2v_\alpha/d(i\lambda)^2$. A self-consistent equation for this term is derived from the derivative of Eq. (32) in terms of the counting field. The solution is expressed with $U_{\alpha\beta}$ introduced in Eq. (33),

$$\frac{d^2v_\alpha}{d(i\lambda)^2} = \sum_{\beta=c,q}\left\{\frac{\partial^3\mathcal{F}_0}{\partial(i\lambda)^2\,\partial v_\beta} + \sum_{\gamma=c,q}\left[2\frac{dv_\gamma}{d(i\lambda)}\frac{\partial^2\mathcal{F}_0}{\partial v_\gamma\,\partial v_\beta} + \sum_{\delta=c,q}\frac{dv_\delta}{d(i\lambda)}\frac{dv_\gamma}{d(i\lambda)}\frac{\partial^3\mathcal{F}_0}{\partial v_\delta\,\partial v_\gamma\,\partial v_\beta}\right]\right\}\frac{U_{\beta\alpha}}{2}. \tag{B2}$$

By substituting Eq. (B2) into Eq. (B1) and by using Eq. (34), we obtain the following form containing terms $dv_\alpha/d(i\lambda)$, $[dv_\alpha/d(i\lambda)]^2$, and $[dv_\alpha/d(i\lambda)]^3$ as

$$\frac{d^3\mathcal{F}}{d(i\lambda)^3} = \frac{d^3\mathcal{F}_0}{d(i\lambda)^3} + \sum_\alpha 3\frac{dv_\alpha}{d(i\lambda)}\frac{\partial^3\mathcal{F}_0}{\partial(i\lambda)^2\,\partial v_\alpha} + \sum_{\alpha,\beta} 3\frac{dv_\alpha}{d(i\lambda)}\frac{dv_\beta}{d(i\lambda)}\frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)\,\partial v_\alpha\,\partial v_\beta} + \sum_{\alpha,\beta,\gamma}\frac{dv_\alpha}{d(i\lambda)}\frac{dv_\beta}{d(i\lambda)}\frac{dv_\gamma}{d(i\lambda)}\frac{\partial^3\mathcal{F}_0}{\partial v_\alpha\,\partial v_\beta\,\partial v_\gamma}. \tag{B3}$$

In the diagrammatic language,[18] the first derivative $dv_\alpha/d(i\lambda)$ corresponds to a vertex correction. The coefficients 3 in the first and second lines are numbers of equivalent diagrams with one vertex correction and two vertex corrections, respectively.

In equilibrium $V=0$, the expression can be simplified further. First, we check that the bare contributions vanish Eq. (45). Second, we pay attention to consequences of a trivial fact that the current is zero in equilibrium $\langle I\rangle=0$,

$$\left.\frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)\,\partial v_c}\right|_{\lambda=\mathcal{A}=0}=\left.\frac{\partial^3\mathcal{F}_0}{\partial(i\lambda)\,\partial v_c^2}\right|_{\lambda=\mathcal{A}=0}=0. \tag{B4}$$

Third, we use consequences of the normalization condition or the causality $U_{qq}|_{\lambda=0}=0$ and $(\partial^3\mathcal{F}_0/\partial v_c^3)|_{\lambda=0}=0$. Then first derivatives of classical and quantum components of the auxiliary field read

$$\left.\frac{dv_c}{d(i\lambda)}\right|_{\lambda=\mathcal{A}=0}=\left.\frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)\,\partial v_q}\frac{U_{qc}}{2}\right|_{\lambda=\mathcal{A}=0}, \tag{B5}$$

$$\left.\frac{dv_q}{d(i\lambda)}\right|_{\lambda=\mathcal{A}=0}=0. \tag{B6}$$

From the relations mentioned above, we can observe that only a term in Eq. (B3) explicitly proportional to U_{qc} remains. Then the final form is

$$\left.\frac{d^3\mathcal{F}}{d(i\lambda)^3}\right|_{\lambda=\mathcal{A}=0}=3\left.\frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)\,\partial v_q}\frac{U_{qc}}{2}\frac{\partial^3\mathcal{F}_0}{\partial(i\lambda)^2\,\partial v_c}\right|_{\lambda=\mathcal{A}=0}. \tag{B7}$$

After rewriting it using the current-density correlation [Eq. (43)] and the current-current correlation [Eq. (44)], we obtain the skewness in equilibrium L_0^3, Eq. (46).

The other two third-order nonlinear transport coefficients L_1^2 and L_2^1 can be calculated in a similar manner. We find that, again, only terms explicitly proportional to U_{qc} remain for $\lambda=\mathcal{A}=0$,

$$\left.\frac{d^3\mathcal{F}}{d\mathcal{A}d(i\lambda)^2}\right|_{\lambda=\mathcal{A}=0}=\left(\frac{\partial^2\mathcal{F}_0}{\partial\mathcal{A}\,\partial v_q}\frac{U_{qc}}{2}\frac{\partial^3\mathcal{F}_0}{\partial(i\lambda)^2\,\partial v_c}+2\frac{\partial^2\mathcal{F}_0}{\partial(i\lambda)\,\partial v_q}\frac{U_{qc}}{2}\frac{\partial^3\mathcal{F}_0}{\partial\mathcal{A}\,\partial(i\lambda)\,\partial v_c}\right)\Bigg|_{\lambda=\mathcal{A}=0}, \tag{B8}$$

$$\left.\frac{d^3\mathcal{F}}{d\mathcal{A}^2d(i\lambda)}\right|_{\lambda=\mathcal{A}=0}=2\left.\frac{\partial^2\mathcal{F}_0}{\partial\mathcal{A}\,\partial v_q}\frac{U_{qc}}{2}\frac{\partial^3\mathcal{F}_0}{\partial\mathcal{A}\,\partial(i\lambda)\,\partial v_c}\right|_{\lambda=\mathcal{A}=0}. \tag{B9}$$

After rewriting it, we obtain Eqs. (47) and (48).

APPENDIX C: RELATIONS AMONG BARE CORRELATION FUNCTIONS

In this appendix, we demonstrate that the saddle-point approximation satisfy the universal relations [Eq. (5)]. For this purpose, we utilize the symmetry of the "bare" CGF [Eq. (31)]. From this symmetry, it is possible to derive general relations among current- and density-correlation functions and simplifies Eqs. (46)–(48). First we symmetrize the bare CGF as

$$\mathcal{F}_{0\pm}(\lambda,v_c,v_q;B)=\mathcal{F}_0(\lambda,v_c,v_q;B)\pm\mathcal{F}_0(\lambda,v_c,v_q;-B). \tag{C1}$$

Then the symmetry [Eq. (31)] is written as

$$\mathcal{F}_{0\pm}(\lambda,v_c,v_q;B)=\pm\mathcal{F}_{0\pm}(-\lambda+i\mathcal{A},v_c,v_q;B). \tag{C2}$$

The derivative in terms of $\mathcal{A}$ and v_q for the both side of Eq. (C2) reads

$$\frac{\partial^2}{\partial v_q\,\partial\mathcal{A}}\mathcal{F}_{0\pm}=\pm\frac{\partial}{\partial v_q}\left(\frac{\partial}{\partial\mathcal{A}}-\frac{\partial}{\partial(i\lambda)}\right)\mathcal{F}_{0\pm}. \tag{C3}$$

After fixing $\lambda=0$ and $\mathcal{A}=0$, we obtain Eq. (50),

$$S_{IN+}=0,\quad S_{IN-}=2\chi_{NI-}.$$

In the same way, from the equality

$$\frac{\partial^3\mathcal{F}_{0\pm}}{\partial v_c\,\partial\mathcal{A}\,\partial(i\lambda)}=\pm\frac{\partial}{\partial v_c}\left(\frac{\partial}{\partial\mathcal{A}}-\frac{\partial}{\partial(i\lambda)}\right)\left(-\frac{\partial}{\partial(i\lambda)}\right)\mathcal{F}_{0\pm}, \tag{C4}$$

we derive Eq. (51),

$$\chi_{I,IN-}=0,\quad \chi_{II,N+}=2\chi_{I,IN+}.$$

Equations (50) and (51) simplify the transport coefficients Eqs. (46)–(48) as follows:

$$L_0^3=24U_{\rm eff}^{\rm eq.}\chi_{NI-}^{\rm eq.}\chi_{I,IN+}^{\rm eq.}, \tag{C5}$$

$$L_2^1=4U_{\rm eff}^{\rm eq.}\chi_{NI-}^{\rm eq.}\chi_{I,IN+}^{\rm eq.}+4U_{\rm eff}^{\rm eq.}\chi_{NI+}^{\rm eq.}\chi_{I,IN+}^{\rm eq.}, \tag{C6}$$

$$L_1^2=12U_{\rm eff}^{\rm eq.}\chi_{NI-}^{\rm eq.}\chi_{I,IN+}^{\rm eq.}+2U_{\rm eff}^{\rm eq.}\chi_{NI+}^{\rm eq.}\chi_{I,IN+}^{\rm eq.}. \tag{C7}$$

These equations lead the compact expressions for symmetrized and antisymmetrized third-order transport coefficients, Eqs. (53)–(55). They satisfy the universal relations among nonlinear transport coefficients [Eq. (5)].

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