

# Inelastic interaction corrections and universal relations for full counting statistics in a quantum contact

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We analyze in detail the interaction correction to full counting statistics (FCS) of electron transfer in a quantum contact originating from the electromagnetic environment surrounding the contact. The correction can be presented as a sum of two terms, corresponding to elastic-inelastic electron transfer. Here we primarily focus on the inelastic correction. For our analysis, it is important to understand more general—universal—relations imposed on FCS only by quantum mechanics and statistics with no regard for a concrete realization of a contact. So we derive and analyze these relations. We reveal that for FCS the universal relations can be presented in a form of detailed balance. We also present several useful formulas for the cumulants. To facilitate the experimental observation of the effect, we evaluate cumulants of FCS at finite voltage and temperature. Several analytical results obtained are supplemented by numerical calculations for the first three cumulants at various transmission eigenvalues.

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## I. INTRODUCTION

Within the past years technological advancements have enabled the fabrication of sufficiently small (nanometer) solid state structures where electrons traverse the system as coherent quantum waves. The electron transport in such a quantum contact can be described with the scattering approach pioneered by Landauer and Büttiker.<sup>1–3</sup> Within this approach, the contact is completely characterized by the set of transmission eigenvalues  $\{T_n\}$ ,  $0 < T_n < 1$ , regardless of its concrete structure. The (differential) conductance is given by the Landauer-Büttiker formula  $G = G_Q \sum_n T_n$ ,  $G_Q = 2e^2/h$ . The same transmission eigenvalues determine superconducting and noise properties of the structure.<sup>4,5</sup> Break junction experiments<sup>6</sup> provide excellent examples of tuning and experimental characterization of  $T_n$ 's in concrete quantum contacts. In the scattering approach, the electron-electron interaction *inside* the contact is commonly disregarded. There is a good reason for that, eventually the same as for electrons in bulk metallic solids. Close to the Fermi energy, the only effect of interaction is to make a quasiparticle from an electron. These quasiparticles do not interact. This means that any contact at sufficiently low energies can be described within a noninteracting scattering approach.

This however presumes an ideal voltage bias of the contact: the electrons are injected and absorbed by reservoirs kept at a certain voltage. This assumption is too ideal: in reality, the contact is embedded in a macroscopic electric circuit, and this *electromagnetic environment* produces voltage fluctuations on the contact. The electrons traversing the contact can emit/absorb energy to/from the environment and interact by means of exchange of photons that are present in the environment. The interaction due to the environment can not be disregarded at low energies and therefore becomes the most important interaction at low temperature and voltage. The environment is completely characterized by a frequency dependent impedance  $Z_\omega$ , in series with the quantum contact.

The influence of the environment on electron transport has been studied in detail for tunnel junctions where all  $T_n$

$\ll 1$ .<sup>7</sup> The tunnel rates in the presence of an environment can be evaluated for arbitrary impedance. For sufficiently large environmental impedances  $ZG_Q/2 = z \gg 1$  the interaction effects are large and the tunnel rates are strongly suppressed below a certain energy. This is termed Coulomb blockade of tunneling in a single junction.<sup>8</sup> The opposite case of small impedance  $z \ll 1$  is more realistic. In this case, the environment provides an interaction correction  $\approx z$  to the rates. This correction can be experimentally identified from its specific voltage and temperature dependence: it is seen as so-called zero-bias anomaly.<sup>9,10</sup>

At arbitrary transmissions, the influence of the environment is more complicated and one cannot evaluate it for an arbitrary impedance (a progress in this direction has been reported in Ref. 11). Still one can investigate the interaction correction  $\approx z$  to the contact conductance, both theoretically and experimentally. It has been demonstrated<sup>12,13</sup> that this correction is related to the second moment of current fluctuations: noise. The correction is proportional to shot noise  $\sim T_n(1-T_n)$ , and disappears at perfect transmissions  $T_n = 1$ . This prediction has been experimentally confirmed.<sup>14</sup>

The environment influences not only the average current, but the whole statistics of electron transfers in the contact, the full counting statistics (FCS). The theory of FCS for a quantum contact within the scattering formalism has been developed in Ref. 15. Later, it has been successfully applied to a variety of systems. The FCS is the statistics of current measurements over a given time interval  $\tau$ ,  $\tau$  being much bigger than the typical time between electron transfers. It gives the probability  $P_\tau(N)$  for  $N$  electrons to be transferred during this time interval. It is convenient to work with the generating function defined as

$$F(V, \chi) = \sum_N P_\tau(N) e^{i\chi N}. \quad (1)$$

The parameter  $\chi$  is frequently called counting field since if one implements the Keldysh formalism for FCS,<sup>16</sup>  $\chi$  enters

the formulation as a field conjugated to electric current. Derivatives of  $\ln F(\chi)$  with respect to  $\chi$  give an infinite set of cumulants of charge transferred, where the first two are related to average current and current noise.

The environment influences the FCS in two distinct ways. A classical effect is governed by the impedance at low frequencies  $\hbar\omega \ll \max(eV, k_B T)$  and scales like  $ZG$ ,  $G$  being the total conductance of the contact. For the two first cumulants—average current and noise—this effect is nothing but voltage division between the contact and the external impedance. The effect is not that trivial for higher cumulants,<sup>17</sup> and the pioneering measurement of the third cumulant<sup>18</sup> was shown to be affected by this environmental effect. However, this effect can be made arbitrarily small by a proper design of the low-frequency impedance to assure  $ZG \ll 1$ .

A more interesting effect comes from the impedance at frequencies  $\hbar\omega \gtrsim \max(eV, k_B T)$  and scales as  $ZG_0$ . This is the interaction correction discussed which eventually leads to Coulomb blockade effects at higher impedance. It has been shown in Ref. 19 that the correction can be separated into inelastic and elastic parts; the latter presents a renormalization of elastic scattering properties of the contact by interaction. It is feasible to observe the interaction correction to FCS in experiments, for instance, with well-characterizable break junctions. Such experiments would be certainly possible for the first three cumulants, and the developments in the field<sup>20–23</sup> suggest that the higher cumulants can be accessed with proper measurement techniques as well. Apparently, the correction will include both the elastic and inelastic parts. To provide theoretical support for these experiments is the main motivation and goal of the present work, which concentrates on the inelastic part of the interaction correction.

In Sec. II we derive the interaction correction to the FCS in first order perturbation in  $z$  starting from the general form of the system-environment Keldysh action. The result allows for the identification of an elastic and inelastic contribution. In Sec. III we explain how one can reinterpret this result in terms of correlations of elementary events of charge transfer. We present concrete analytical results in the next section, particularly for vanishing temperature.

While investigating the interaction correction in the limit of small voltages, we have found sets of simple relations for cumulants. Further analysis has shown that these relations are not specific for the setup considered and hold for any conductor regardless of its properties and presence or absence of interaction. In fact, these universal relations provide the generalization of the fluctuation-dissipation theorem<sup>24</sup> for FCS. We discuss these relations at length in Sec. V. The derivation is provided in the Appendix.

In the last section we present our numerical results for arbitrary temperature and voltage. We have studied the experimentally interesting case of an RC environment and plot the correction to the first three cumulants for conductors of different transmissions and a diffusive conductor versus  $eV/k_B T$  as obtained from a numerical evaluation of the integrals. In all cases we observe a crossover at  $eV \sim k_B T$  that is related to a transition from thermal to shot noise behavior.

## II. ACTION

The system we consider consists of a quantum conductor which can be described by the set of its transmission probabilities,  $T_n$ , and a frequency dependent environmental impedance,  $z_\omega$  in series. The voltage drop over the whole conductor-environment system is fixed. However, the voltage in the node between the contact and the impedance can fluctuate. For instance, an electron transferred will momentarily charge the node creating a voltage pulse  $\propto z$  that persists for some time and may influence further electron transfer. Thus there will be a fluctuating voltage in the node depending both on the probabilistic nature of the electron transfer in the quantum conductor as well as on the impedance of the environment. We study the corrections to electron transport due to these fluctuations.

It is convenient to work with the phase rather than voltage, which is defined as  $\phi = \int dt eV(t)/\hbar$ . Since we study a quantum mechanical system, we have to describe quantum fluctuations of this quantity. This is most conveniently presented in the language of Keldysh action that expresses physical quantities in the form of a path integral over the fluctuating phase on the Keldysh contour. Since the Keldysh contour consists of two parts, the integration proceeds over two sets of variables  $\phi^\pm(t)$  corresponding to these parts.

The Keldysh action approach to mesoscopic quantum circuits has been pioneered in Ref. 25 and has been extended to cover FCS and arbitrary quantum contacts in Refs. 17 and 26. As usual in the theory of FCS, it is the generating function  $F(\chi)$  which is presented as a path integral over the fluctuating phase. The action in the path integral is a sum of the actions describing the constituent parts of the circuit: the conductor action,  $S_c$ , and the environment action,  $S_{\text{env}}$ ,

$$F(\chi) = \int d\phi^+ d\phi^- \exp\{-iS[\phi^+, \phi^-]\},$$

$$S[\phi^+, \phi^-] = S_c[\phi^+, \phi^-] + S_{\text{env}}[\Phi + \chi/2 - \phi^+, \Phi - \chi/2 - \phi^-]. \quad (2)$$

We use superscripts  $\pm$  to denote the phases at different parts of the contour and use traditional notations  $\varphi(\chi)$  for their half-sum (difference). Current, noise, and higher moments of FCS follow as  $\partial \ln F / \partial \chi|_{\chi=0}$ ,  $\partial^2 \ln F / \partial \chi^2|_{\chi=0}, \dots$ . For a linear environment the action  $S_{\text{env}}$  is a bilinear function of the phases depending on the impedance and temperature  $T$  only,

$$S_{\text{env}} = \frac{1}{2\pi} \int_0^\tau dt \int_0^\tau dt' \left[ (\phi^+(t), \phi^-(t)) A(t-t') \begin{pmatrix} \phi^+(t') \\ \phi^-(t') \end{pmatrix} \right], \quad (3)$$

where the coefficient matrix,  $A$ , depends solely on frequency and temperature:

$$A(\omega) = \begin{pmatrix} -i\omega[z_\omega^{-1} + 2N_\omega \text{Re } z_\omega^{-1}] & 2i\omega N_\omega \text{Re } z_\omega^{-1} \\ -[A^{+-}(-\omega)]^* & -[A^{++}(-\omega)]^* \end{pmatrix}. \quad (4)$$

$N_\omega = \{\exp[\hbar\omega/k_B T] - 1\}^{-1}$  is the Bose-Einstein distribution function.

The most concise way to write the conductor action is in terms of Keldysh-Green functions  $\hat{L}$ ,  $\hat{R}$  of the left-right reservoir:<sup>16</sup>

$$S_c = \frac{i}{2} \sum_n \text{Tr} \ln \left[ 1 + \frac{T_n}{4} (\{\hat{L}, \hat{R}\} - 2) \right]. \quad (5)$$

This assumes that the energy dependence of  $T_n$  can be disregarded on the energy scale of interest. The trace is over Keldysh space and energy or frequencies. In Eq. (5) the fluctuating phase in the node enters in the form of a gauge transform in one of the reservoirs as  $\hat{L} = e^{i\phi} G^{\text{res}} e^{-i\phi}$ ,  $\hat{R} = G^{\text{res}}$ , where  $\phi = \varphi + \chi/2\tau_z$ . The equilibrium Keldysh-Green function depends on time difference (or energy) only and reads in a usual way

$$G^{\text{res}} = \begin{pmatrix} 1 - 2f(\varepsilon) & 2f(\varepsilon) \\ 2(1 - f(\varepsilon)) & 2f(\varepsilon) - 1 \end{pmatrix}, \quad (6)$$

with  $f$  being the Fermi distribution function of the corresponding lead. Equations (2)–(6) define our model and all we have to calculate is the cumulant generating function  $F$  for a given environment  $z_\omega$  and conductor  $T_n$ . However, in the general case this is a formidable task. A natural way to proceed is to assume  $z \ll 1$  and thus to treat the effect of the environment as a perturbation. In zeroth order (no environment), the phases are obviously related to the applied voltage and do not fluctuate:  $\varphi = eVt$ .

Putting this in Eq. (5) and taking the trace gives the well known expression for the generating function of a mesoscopic conductor in terms of its transmission eigenvalues<sup>15</sup>

$$\ln F^{(0)}(\chi) \equiv \frac{\tau}{\hbar} S^{(0)} = \frac{\tau}{\hbar} \sum_n \int \frac{d\varepsilon}{2\pi} \ln \{ 1 + T_n (e^{i\chi} - 1) f_l(1 - f_r) + (e^{-i\chi} - 1) f_r(1 - f_l) \}. \quad (7)$$

Here and in the following indices “ $l, r$ ” refer to the left or right lead. We assign the voltage to the left lead, so that  $f_l(\varepsilon + V) = f_r(\varepsilon) = f(\varepsilon)$ .

The first order correction is proportional to the fluctuations of  $\phi$ , which are small  $\propto z$ . Expanding the logarithm in Eq. (5) to second order in  $\phi$  gives a second order contribution

$$-i\delta S_c^{(2)} = \frac{T_n}{8} \text{Tr} \{ DA^{(2)} \} - \frac{1}{4} \left( \frac{T_n}{4} \right)^2 \text{Tr} \{ DA^{(1)} DA^{(1)} \} \quad (8)$$

$$= \frac{T_n}{8} D \text{Tr} \{ A^{(2)} \} - \frac{1}{4} \left( \frac{T_n}{4} \right)^2 DD^+ \text{Tr} \{ A^{(1)} A^{(1)} \}, \quad (9)$$

where the following relations hold under the trace:

$$A^{(1)} A^{(1)} = 4\phi^2 - 4\phi L^+ \phi L - 2\phi L^+ R^+ \phi L R - 2\phi R^+ L^+ \phi R L + 4\phi L^+ R^+ L^+ \phi R, \quad (10)$$

$$A^{(2)} = \phi L^+ \phi R + \phi R^+ \phi L - \phi^2 (R L + L R), \quad (11)$$

and

$$D^{-1} = 1 + T_n [(e^{i\chi} - 1) f_l(1 - f_r) + (e^{-i\chi} - 1) f_r(1 - f_l)]. \quad (12)$$

All quantities with superscript “+” are taken at energy  $\varepsilon + \omega$  and integration over energy and frequency is implied. For convenience we omitted the explicit dependence on  $\varepsilon, \omega$ . It is easily found from the definition of the trace. The first term in Eq. (9) for instance would read  $T_n/8 \text{Tr} \int d\varepsilon d\omega D_\varepsilon \phi_{-\omega} L_{\varepsilon+\omega} \phi_\omega R_\varepsilon$ , where the trace is understood over Keldysh indices.

These terms are quadratic in phase, and by virtue of the path integral in Eq. (2) are to be replaced with their averages given by the environmental action. These averages read

$$\left\langle \left\langle \begin{pmatrix} |\phi|^2 & \varphi \chi^* \\ \varphi^* \chi & |\chi|^2 \end{pmatrix} \right\rangle \right\rangle \rightarrow \begin{pmatrix} (2N_\omega + 1) \frac{\text{Re } z_\omega}{\omega} & \frac{z_\omega}{\omega} \\ -\frac{z_\omega^*}{\omega} & 0 \end{pmatrix}. \quad (13)$$

After some ordering of terms, the first order correction to the cumulant generating function can be presented as

$$[\ln F(\chi)]^{(1)} = \frac{\tau}{\hbar} \int_0^\infty d\omega \frac{\text{Re } z_\omega}{\omega} [(2N_\omega + 1) S_{\text{el}}^{(1)}(\chi) + N_\omega S_{\text{in}}^{(1)}(\omega, \chi) + (N_\omega + 1) S_{\text{in}}^{(1)}(-\omega, \chi)]. \quad (14)$$

We note that there are three different terms which can be identified as being due to elastic electron transfer, and inelastic transfer with either absorption (positive  $\omega$ ) or emission (negative  $\omega$ ) of energy  $\hbar\omega$  from/to the environment. Explicitly, in terms of filling factors these terms read

$$S_{\text{el}} = -2 \sum_n T_n (1 - T_n) \frac{\partial S^{(0)}}{\partial T_n}, \quad (15)$$

$$S_{\text{in}} = \sum_n \int \frac{d\varepsilon}{2\pi} \{ DD^+ [T_n (f_l - f_l^+) + 2T_n (e^{i\chi} - 1) f_l(1 - f_r^+) + 4T_n^2 (\cos \chi - 1) f_l(1 - f_l^+) (f_r^+ - f_r)] + (1 - D)(1 - T_n - D^+) \} + \left\{ \begin{matrix} l \leftrightarrow r \\ \chi \leftrightarrow -\chi \end{matrix} \right\}. \quad (16)$$

Since the expression is symmetric with respect to exchange  $l \leftrightarrow r$  of the leads and simultaneous change of the sign of the counting field, the cumulants are either even or odd functions of the voltage applied. Following Ref. 19 we present the elastic part of the correction as a change  $\propto z$  of transmission eigenvalues. The analysis of expression (14) shows that the inelastic part is contributed by frequencies of the order of voltage and/or temperature, since the integrand falls off as  $\exp(-\hbar\omega/k_B T)$  at  $\hbar\omega \gg eV, k_B T$  due to restrictions imposed by energy conservation. Elastic processes do not have this restriction and  $S_{\text{el}}$  contributes to the integral at all frequencies.

### III. INTERPRETATION

The advantage of the FCS approach to quantum transport is that in many cases the FCS expression can be reinterpreted

in terms of elementary events, thus providing some insight into the relevant transport properties. A well-known example of such interpretation is provided by the noninteracting Levitov formula.<sup>15</sup> At vanishing temperature, it allows us to present the statistics as a superposition of  $\tau eV/\hbar$  elementary “games” in each transport channel  $n$ , each “game” resulting in either transmission (with chance  $T_n$ ) or reflection (with chance  $1-T_n$ ) of an electron coming to the contact. A recent example of such a re-interpretation concerns spin statistics.<sup>27</sup> In this section, we show how to interpret the inelastic interaction correction given by Eq. (16). As we will see, the interpretation is probably too complicated to be constructive. Still it gives some insight to the form of the expression, in particular, the presence of denominators  $D, D^+$ .

To give an interpretation of the form of  $S_{\text{in}}$  let us go back to relation (7) that holds for non-interacting electrons. In the limit of large measurement time  $\tau$ ,  $\hbar/\tau \ll \max(k_B T, eV)$ , we can discretize the integration over energies. The general structure of the generating function<sup>28</sup> is then

$$F^{(0)}(\chi) = \prod_E \lambda(E), \quad (17)$$

with  $\lambda(E) = D^{-1}$ . The product is taken over discretized energies as well as over transport channels. The right hand side of equation (17) is a product, so each term in this product can be regarded as an independent process (“game”). The concrete form of  $\lambda(E)$  suggests that three distinct outcomes of each process are possible: (i) electrons transmitted from the left reservoir to the right with probability  $P_+ = T_n f_l(1-f_r)$ , (ii) transmission from right to left with  $P_- = T_n f_r(1-f_l)$ , (iii) no transmission ( $P_0 = 1 - P_+ - P_-$ ). Indeed, the generating function for each process is then  $\lambda = \sum_{\alpha} P_{\alpha} X_{\alpha}$  with  $X_{\alpha} = e^{i\alpha\chi}$ . Electrons at different energy (and/or channel) are uncorrelated since the complete generating function factorizes in terms of  $\lambda(E)$ .

How can this picture change if one introduces electron-electron interaction via the environment? It is clear that interaction will bring about all kinds of correlations between electrons at different energies and the simple picture presented above is not true anymore. The major change is, that the factorization in uncorrelated elementary events does not hold. Presumably, the generating function of an elementary event  $\Lambda$  will depend on many different energies. The relation (16) suggests that in lowest order in  $z$  it depends on two energies only,  $\Lambda(E, E')$ , where  $|E - E'| = \hbar\omega$  is the energy of an absorbed or emitted photon. With this accuracy, the cumulant generating function can be expressed as a product over pairs of energies

$$F^{(0)}(\chi) + F^{(1)}(\chi) = \prod_{E, E'} \Lambda(E, E'). \quad (18)$$

Without interactions,

$$\Lambda(E, E') = \begin{cases} \lambda(E) & \text{if } E = E', \\ 1 & \text{if } E \neq E' \end{cases} \quad (19)$$

so that electrons with different energy are uncorrelated. If  $\delta\Lambda(E, E')$  is the interaction correction to  $\Lambda$ , the change of the cumulant generating function reads

$$[\ln F(\chi)]^{(1)} = \sum_{E, E'} \frac{\delta\Lambda(E, E')}{\lambda(E)\lambda(E')}. \quad (20)$$

This explains already the presence of denominators in (16). In addition, we conclude from (16) that contains a single sum over transport channels, that the elementary events do not involve electrons in different channels, even though they involve electrons at different energies. This is probably valid only for the first order correction.

To proceed, let us note that the correction  $\delta\Lambda(E, E')$  consists of terms to be divided into three classes. First, there will be terms presenting *new events*,  $\delta\Lambda_{\text{new}}(E, E')$  not taking place for noninteracting electrons. An example is an electron transfer from the left to the right with photon emission. As we can assert from (16) it comes with probability  $\propto T_n f_l(E) \times [1 - f_r(E')](1 + N_{E-E'})$ . Another example is a two-particle process consisting of elastic electron transfer at energy  $E$  accompanied by inelastic transfer, its probability being proportional to  $T_n^2 f_l(E) f_r(E) [1 - f_l(E')][1 - f_r(E)](1 + N_{E-E'})$ . Secondly, since the probabilities of elementary *old events* are modified by interaction, there will be terms depending on a single energy only, those can be seen as the modification of  $\lambda(E)$ ,  $\lambda(E) \rightarrow \lambda(E) + \delta\Lambda_{\text{old}}(E, E)$ . They are incorporated into the elastic part of the correction. Finally, the environment will introduce *correlations* among pairs of old events, represented by  $\delta\Lambda_{\text{corr}}(E, E')$ . For instance, the correlation between left-right transfer at energy  $E$  and right-left transfer at energy  $E'$  will come with a factor  $T_n^2 f_l(E) [1 - f_r(E)] f_r(E') [1 - f_l(E')]$ .

These three contributions simply add up in the correction to the generating function,

$$[\ln F(\chi)]^{(1)} = \sum_E \frac{\delta\Lambda_{\text{old}}(E, E)}{\lambda(E)} + 2 \sum_{E > E'} \frac{\delta\Lambda_{\text{new}}(E, E') + \delta\Lambda_{\text{corr}}(E, E')}{\lambda(E)\lambda(E')}. \quad (21)$$

One recognizes this structure in the relations for  $S_{\text{el}}$  (first term) and for  $S_{\text{in}}$  (second term). In principle, in this way one can recover the correction to the generating function of an elementary event  $\delta\Lambda$  and find the (corrections to) probabilities of all possible outcomes, new ones as well as old ones. This gives a reinterpretation of the correction: any term of Eq. (16) is assigned to a term of one of the three classes discussed.

However, the procedure is cumbersome and hardly practical because of the large number of possible processes and outcomes. For a two-electron process, each incoming electron can be in one of four possible states (coming from the left or the right at  $E$  or  $E'$ ), the same for outgoing electrons. This gives in total  $2^8$  terms: it looks like a somewhat lengthy interpretation of a relatively compact Eq. (16). This prevented us from accomplishing this program explicitly. We are satisfied with the fact that the combinations of electron and photon filling factors make sense for the terms contrib-

uting to  $\delta\Lambda_{\text{new,corr}}$ . The picture and the interpretation are expected to become even more involved for the corrections of higher orders in  $z$ .

#### IV. ANALYTICAL RESULTS

At vanishing temperature we can easily perform the integration over  $\varepsilon$  in Eq. (14). The full correction to FCS then reads

$$[\ln F(\chi)]^{(1)} = \frac{\tau}{\hbar} eV S_{\text{el}} \int_{eV/\hbar}^{\infty} d\omega \frac{\text{Re } z_{\omega}}{\omega},$$

$$S_{\text{el}} = -2 \sum_n T_n (1 - T_n) \frac{e^{i\chi} - 1}{1 + T_n (e^{i\chi} - 1)}. \quad (22)$$

It has a simple and interesting structure revealing the relationship between the elastic and inelastic part of the correction. If we took the elastic part only, by virtue of (7) we would obtain a similar expression. The difference is that the integration over  $\omega$  would start at zero. Therefore, the inelastic part of the correction precisely cancels the modification of the elastic transmission for photon energies in the interval  $0 < \hbar\omega < eV$ . It is indeed expected from general reasoning<sup>19</sup> that the low-energy divergences present in the elastic correction are cut off at energies  $\simeq eV$ . Our somewhat unexpected result is that at vanishing temperatures this cut-off is sharp and clear. In agreement with expectations, the inelastic  $S_{\text{in}}$  only contributes at frequencies  $0 < \hbar\omega < eV$ , reflecting the fact that the only energy source for inelastic processes is given by the voltage.

The correction to the  $m$ th cumulant  $S^{(m)}$  is given by derivatives of the above relation, we find:

$$\delta S^{(m)} = -2 \frac{\tau}{\hbar} eV \sum_n T_n (1 - T_n)$$

$$\times \left. \frac{d^m}{d(i\chi)^m} \frac{e^{i\chi} - 1}{1 + T_n (e^{i\chi} - 1)} \right|_{\chi=0} \int_{eV/\hbar}^{\infty} d\omega \frac{\text{Re } z_{\omega}}{\omega}. \quad (23)$$

Importantly, the correction is proportional to the  $(m+1)$ th cumulant for the non-interacting case,  $\delta S^{(m)} \propto S^{(m+1)}$ . This generalizes the results<sup>12,13</sup> for the average current. The environment enters the corrections as an integral over the impedance and affects every cumulant in the same way.

A common and experimentally interesting model for a frequency-dependent impedance is that of an RC environment,  $z_{\omega} = z(1 + i\omega/\omega_c)^{-1}$ . The impedance is cut at  $\omega_c = 1/(RC)$  and approaches a constant value of  $z$  at  $\omega \ll \omega_c$ . The integral governing the correction evaluates to

$$\int_{eV/\hbar}^{\infty} d\omega \frac{\text{Re } z_{\omega}}{\omega} = \ln \sqrt{1 + \frac{\hbar^2 \omega_c^2}{e^2 V^2}} \approx \ln \frac{\hbar \omega_c}{eV}, \quad \text{if } \hbar \omega_c \gg eV. \quad (24)$$

That is, it diverges logarithmically at sufficiently low voltages  $eV \ll \hbar \omega_c$ . This is the well known zero bias anomaly. As

has been shown it holds for any cumulant. Different cumulants have the same functional dependence on voltage and can be scaled by the prefactor of Eq. (24), which depends only on the transmission probabilities.

Even if  $z \ll 1$  the correction  $\propto z \ln(\hbar \omega_c / eV)$  can become big at sufficiently small voltages,  $eV/\hbar \omega_c \simeq e^{-1/2z}$ . It has been shown in Ref. 19 that in this case one has to implement the renormalization procedure neglecting the inelastic part. The elastic correction can be consolidated in the energy dependent renormalization of transmission eigenvalues given by

$$\frac{dT_n(E)}{d \log E} = 2z T_n(E) [1 - T_n(E)]. \quad (25)$$

We do not consider this here. Rather, we expect that finite temperature will lead to a rounding off of the singularity at small voltages in the same way as for the current correction.

Equation (14) is too complex at finite temperatures so that it is hard and non-instructive to perform the integration over energies  $\varepsilon$ . However, the correction to any cumulant of finite order derived from the generating function is an integral over a finite polynomial of Fermi functions. This integration can easily be done for arbitrary temperature and voltage. The analytical formulas obtained in this way are too long to be of any use except numerical evaluation. The correction to any cumulant is proportional to  $T_n(1 - T_n)$ , thereby vanishing at perfect and vanishing transmission.

For the correction to the average current we find

$$\delta I = -2e \sum_n T_n (1 - T_n)$$

$$\times \int_0^{\infty} d\omega \frac{\text{Re } z_{\omega}}{\omega} \frac{\omega \sinh \frac{eV}{k_B T} - \frac{eV}{\hbar} \sinh \frac{\hbar \omega}{k_B T}}{\cosh \frac{eV}{k_B T} - \cosh \frac{\hbar \omega}{k_B T}} \quad (26)$$

$$= -e \int_0^{\infty} d\omega \frac{\text{Re } z_{\omega}}{\omega} \langle |\Delta I|_{\omega}^2 \rangle, \quad (27)$$

where  $\langle |\Delta I|_{\omega}^2 \rangle$  is the finite-frequency current correlator without environment.<sup>13,29</sup> The correction to the current is thus related to the noise in the absence of an environment.

The correction to the  $m$ th cumulant is an  $(m+1)$ th order polynomial in  $T_n$ . The term linear in  $T_n$  has the same functional dependence as that in (27). The reason for this is that in the limit of small  $T_n$  the full counting statistics is that of a tunnel junction: electron transfers are rare and consequently independent. We get a superposition of two Poissonian statistics for electrons tunneling from the left to the right and from the right to the left expressed as

$$\ln F(\chi) = \tau [\Gamma_{LR}(V, T)(e^{i\chi} - 1) + \Gamma_{RL}(V, T)(e^{-i\chi} - 1)], \quad (28)$$

$\Gamma_{LR,RL}$  being tunneling rates in these two directions. The interaction in this limit modifies  $\Gamma_{LR,RL}$ , this being the only effect on FCS. We will see below that these two rates are related by the detailed balance condition  $\Gamma_{LR}$

$=\Gamma_{LR} \exp(-eV/k_B T)$ . From this it follows that in the tunneling limit

$$\frac{e}{\tau} S^{(m)} = \begin{cases} I & \text{if } m \text{ odd,} \\ I \coth\left(\frac{eV}{2k_B T}\right) & \text{if } m \text{ even,} \end{cases} \quad (29)$$

for any interactions.

Analytical work in the limit of small voltages  $eV \ll k_B T$  gave us some relations between the cumulants. However, we have recognized that these relations are not specific for the interaction correction but are of general nature. That is why we discuss them in the next section.

### V. UNIVERSAL RELATIONS FOR CUMULANTS

The detailed investigation of the interaction corrections to FCS is hardly effective without appreciation of universal relations for FCS cumulants that hold with no regard for interaction and/or concrete structure of the conductor. This is why in the course of this research we had to understand the general constraints imposed on the FCS by laws of quantum mechanics and thermodynamics.

We show in the Appendix that this universal relation can be most generally expressed in the following form:

$$F(V, \chi) = F(V, -\chi + ieV/k_B T). \quad (30)$$

A didactic representation of this relation can be obtained by recalling the definition of  $F$  as generating function of the probability distribution of a certain number  $N$  of particle transfers,

$$F(V, \chi) = \sum_N P_N e^{i\chi N}. \quad (31)$$

Applying (30), we observe that the probabilities of opposite number of particles transferred are related by

$$P_N(V) = P_{-N}(V) e^{eVN/k_B T}. \quad (32)$$

This relation is well known for independent tunnelling events (see, e.g., Refs. 7 and 8) and is referred to as *detailed balance* condition. We thus demonstrate that the detailed balance holds for any  $N$  irrespective of possible interactions and correlations in and beyond the conductor.

Whatever didactic, the detailed balance condition is not easy to apply to cumulants. We do this with the universal relation (30) that obviously holds for  $\ln F$  as well. A series of relations for cumulants is obtained by taking derivatives of this relation with respect to voltage at  $V=0$ . First of all, we just go to the equilibrium limit  $V \rightarrow 0$  to obtain

$$\ln F(\chi) = \ln F(-\chi), \quad (33)$$

all even cumulants thus vanish at equilibrium. This relation and *all subsequent relations* till the end of this Section are valid only in the limit  $V \rightarrow 0$ .

Taking the first derivative with respect to voltage, we arrive at

$$\frac{\partial}{\partial eV} [\ln F(\chi) - \ln F(-\chi)] = -\frac{i}{k_B T} \frac{\partial \ln F(\chi)}{\partial \chi}. \quad (34)$$

If we expand this in  $\chi$ , we obtain a series of equations that relate even cumulants with voltage derivatives of odd cumulants

$$S^{(2n+2)} = \frac{2k_B T}{e} \frac{\partial S^{(2n+1)}}{\partial V}. \quad (35)$$

The first equation in this series is nothing but Johnson's noise formula,

$$e^2 S^{(2)} = 2k_B T \frac{\partial I}{\partial V}, \quad (36)$$

that relates zero-voltage conductance and equilibrium current noise.

The next series is obtained by taking the second derivative with respect to voltage and making use of (34),

$$\frac{\partial^2}{\partial V^2} [\ln F(\chi) - \ln F(-\chi)] = -\frac{ie}{k_B T} \frac{\partial^2}{\partial V \partial \chi} [\ln F(\chi) + \ln F(-\chi)] \quad (37)$$

which is only practical if even(odd) cumulants are *not* even(odd) functions of voltage, that is, in the absence of electron-hole symmetry. Since our model is electron-hole symmetric, this relation is of no immediate relevance. The first relation in the series reads

$$\frac{\partial^2 I}{\partial V^2} = -\frac{e^2}{k_B T} \frac{\partial S^{(2)}}{\partial V}. \quad (38)$$

It relates dc current induced by low-frequency a.c. voltage (rectification effect) to low-frequency current noise proportional to dc voltage applied. This relation was discussed in detail in Ref. 30 in the context of the photovoltaic effect.

Taking the third derivative with respect to voltage and making use of (38) we obtain another series:

$$\begin{aligned} \frac{\partial^3}{\partial V^3} [\ln F(\chi) - \ln F(-\chi)] &= \frac{ie^3}{(k_B T)^3} \frac{\partial^3 \ln F(\chi)}{\partial \chi^3} \\ &\quad - \frac{3ie}{k_B T} \frac{\partial^3}{\partial V^2 \partial \chi} [\ln F(\chi) \\ &\quad + \ln F(-\chi)]. \end{aligned} \quad (39)$$

The first relation in the series can be rewritten as

$$2e^2 \frac{\partial^2 S^{(2)}}{\partial V^2} = \frac{1}{3} \left( 2k_B T \frac{\partial^3 I}{\partial V^3} - \frac{e^4}{(k_B T)^2} S^{(4)} \right). \quad (40)$$

The left-hand side gives the change of the current noise induced by low-frequency ac voltage at nonmatching frequency. This response coefficient, and its importance, has been recently discussed in Ref. 31, where it has been termed "noise thermal impedance." The authors have conjectured the relation of this coefficient to the fourth cumulant.

It is easy to see that Eq. (30) holds for the elastic part of the FCS even before integration over energies in (7), since for any energy

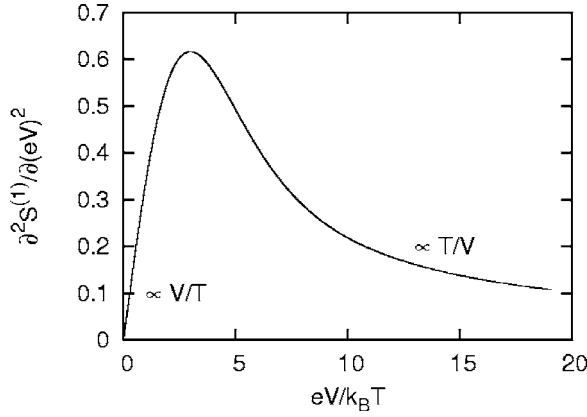


FIG. 1. Second derivative with respect to voltage of the correction to the average current,  $\partial^2 S^{(1)}/\partial(eV)^2 \equiv \hbar k_B T / [2e^2 R G_Q \sum_n T_n (1 - T_n)] \partial^2 I / \partial(eV)^2$  vs  $eV/k_B T$ .

$$\frac{f_l(1-f_r)}{f_r(1-f_l)} = e^{eV/k_B T}. \quad (41)$$

The corresponding proof for the inelastic contribution can be done only after integration over energies, is cumbersome, and has provided a good check for the validity of expression (16).

## VI. NUMERICAL RESULTS

We restrict the numerical analysis to the RC-environment model discussed above. There are three energy scales in the system: voltage, temperature, and the cutoff frequency of the environment,  $\omega_c$ . It is especially interesting to study the case  $\hbar\omega_c \gg eV, k_B T$ . In this case it is expected that all cumulants show the logarithmic divergence at small voltages/temperature, which is well known for the conductance:  $\delta G \sim G_Q \ln[\hbar\omega_c / \max(eV, k_B T)]$ . This motivated us to the following choice of the presentation of the results: we plot the second derivative of the correction to the first three cumulants with respect to voltage versus  $eV/k_B T$  (Figs. 1–3). It is the second derivative that approaches a limit independent of  $\omega_c$  upon increasing the ratios  $\hbar\omega_c/eV, \hbar\omega_c/k_B T$ . To illustrate the dependence on transmission and to assess differences be-

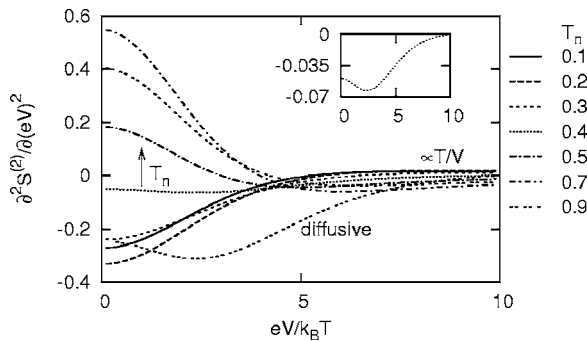


FIG. 2. Second derivative with respect to voltage of the correction to the noise,  $\partial^2 S^{(2)}/\partial(eV)^2 \equiv \hbar k_B T / [2e^2 R G_Q] \partial^2 I^{(2)}/\partial(eV)^2$  vs  $eV/k_B T$ . The general tendency as a function of transmission  $T_n$  is indicated by an arrow. The inset shows a zoom for  $T_n=0.4$ .

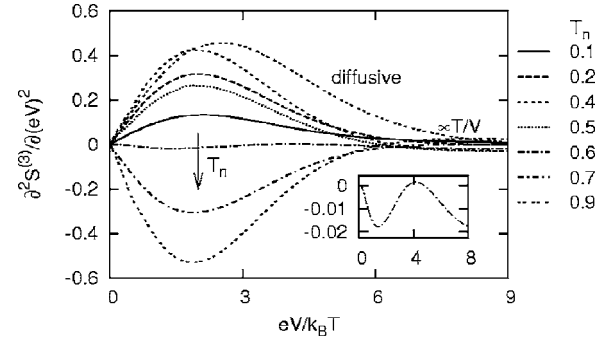


FIG. 3. Second derivative with respect to voltage of the correction to the third cumulant,  $\partial^2 S^{(3)}/\partial(eV)^2 \equiv \hbar k_B T / [2e^3 R G_Q] \partial^2 I^{(3)}/\partial(eV)^2$  vs  $eV/k_B T$ . The general tendency as a function of transmission  $T_n$  is indicated by an arrow. The inset shows a zoom for  $T_n=0.6$ .

tween specific conductors (ballistic, tunnel,...), we plot the results for a single-channel conductor with transmission values ranging from 0.1 to 0.9. Another interesting reference system is a diffusive conductor. The results for a diffusive conductor can be obtained by averaging over transmission eigenvalues with the distribution function  $\rho(T_n) = (T_n \sqrt{1-T_n})^{-1}$ .<sup>4,5</sup> It should be noted that this procedure assumes that the dwell time in the conductor  $\tau_{\text{dwell}}$  is sufficiently short<sup>32</sup> such that  $\hbar/\tau_{\text{dwell}} \approx E_{\text{th}} \approx D/L^2 \gg eV, k_B T$ . Under these conditions the energy dependence of  $T_n$  can be disregarded and the averaging is possible. Since all cumulants are polynomials in  $T_n$  this is equivalent to replacing  $T_n^m \rightarrow \sqrt{\pi} \Gamma(m) / \Gamma(m+1/2)$ .

At  $eV/k_B T \gg 1$  the corrections are dominated by  $S_{e1}$  and the emission term  $S_{\text{in}}^{(1)}(-\omega, \chi)$ , since the environment does not provide any energy at  $T \rightarrow 0$ . The functions plotted  $\propto z/V$  for all cumulants, and are given by Eq. (23). As discussed in the paragraph below that equation, this leads to a suppression of the conductance (and any other cumulant) at small voltages which is termed zero-bias anomaly.

Let us now discuss differences between cumulants, starting with the current (Fig. 1). An apparent feature of the current correction is that the corrections to different conductors can be scaled to one curve by the common prefactor  $\sum_n T_n (1 - T_n)$  [Eq. (26)]. The corresponding curve for a diffusive conductor can be obtained, following from averaging over transmissions, by multiplication with 2/3 (and removing the dependence on  $T_n$  in the normalization). This feature is unique for the correction to the current and independent of the choice of a specific environment. Higher order cumulants have a more involved dependence on transmission eigenvalues and environment.

In the opposite limit of large temperatures,  $eV \ll k_B T$ , the environment provides the energy for inelastic electron transfer. Consequently the absorption term,  $S_{\text{in}}^{(1)}(\omega, \chi)$  becomes more important in Eq. (14). It is expected that in the same way as for the zero-bias anomaly the correction to the conductance is logarithmically diverging with temperature. Due to the choice of presentation, this term is not visible in Fig. 1. What is shown in this plot and the following is the lowest order term in the expansion in powers of  $eV/k_B T$ . For the

current (and any odd cumulant) this is a linear term, following from the symmetry with respect to inversion of voltage as explained below Eq. (16). In summary, the correction to the current shows a crossover from a temperature to voltage dominated regime at  $eV \sim k_B T$ .

In Fig. 2 we plotted the second derivative with respect to voltage of the correction to the noise for several single channel conductors and a diffusive conductor. At small voltages, all curves start with zero slope since noise is an even function of voltage. Interestingly with increasing voltage they all cross the x-axis at  $eV \sim k_B T$ , before approaching zero. As expected, changes occur on the scale of temperature. The limit of vanishing temperature,  $eV/k_B T \gg 1$ , namely the proportionality to  $z/V$ , can be discussed along the same lines as for the current. However there are several striking differences that were absent in the correction of the current.

Unlike for the current, curves for different transmission can not be reconciled by scaling, rather we observe a strong dependence on the value of the transmission. This dependence is nonmonotonous. However at small voltages, conductors with  $T_n \lesssim 1$  have positive correction while those with  $T_n \gg 1$  have a negative. This sign change could have been conjectured since it is well known that corrections to the  $m$ th cumulant are related to the unperturbed  $(m+1)$ th cumulant. Hence, the correction to noise should be related to the third cumulant, whose dependence on the transmission eigenvalues  $[\propto T_n(1-T_n)(1-2T_n)]$  changes sign at intermediate transmissions.

This behavior can be interpreted by looking at the extreme cases of  $T_n \ll 1$  and  $T_n \approx 1$ . It is plausible that either conductor in the absence of an environment produces little shot noise since in the first case the current is “most of the time” zero with only rare transfers of charges. In the second limit, electrons are transferred with probability close to one and are only occasionally reflected.

The same conductors embedded in an environment however will feel a suppression of current due to fluctuations of the voltage in the node as discussed in Sec. II. For the noise of the tunnel conductor that means that the rare electron transfers being the source of noise, will be suppressed, furthermore leading to a reduction of noise (negative correction). If  $T_n \lesssim 1$ , the suppressed conductance means that the quasi-constant flow of electrons will be interrupted more often, resulting in an enhancement of noise (positive correction). Consequently there will be a crossover at intermediate values of the transmission, which can be seen in Fig. 1.

Comparing the shape of the curves we observe that the maximum (absolute) value lies for most conductors at  $V=0$ . Remarkable exceptions are the diffusive conductor and such with transmission close to the crossover ( $T_n=0.4$ , inset Fig. 2). We might argue that the diffusive conductor inherits this feature from intermediate transmissions, or rather that it is an “interference” effect determined by the coefficients of lowest and highest power in transmission ( $T_n, T_n^3$ ) in the expression for the correction to the noise.

The corresponding plot of the correction to the third cumulant, which reflects the asymmetry of electron transfer, is presented in Fig. 3. It shares features of both current and noise correction. Due to the different symmetry with respect to voltage inversion, the corrections to the third cumulant

start at zero with linear slope. Again there is a crossover at  $eV \sim k_B T$  and a decay with  $z/V$  at large voltages reflecting the zero bias anomaly. The dependence on  $T_n$  is nontrivial, which is not surprising since the expression contains four terms of different power in  $T_n$ , each of which can have a distinct dependence on  $eV/k_B T$ . However, ballistic or tunnel conductors at small voltages separate in the same way (albeit with opposite sign) as for the correction to noise to the lower or upper part of the plot. Comparing curves of small  $T_n$  with those for the current, we recover the tunneling limit discussed in Sec. IV. Interestingly the correction for intermediate transmission appears to be more feature-rich than for the limiting cases. This is clearly an indication that terms of different power in  $T_n$  contribute with equal weight to the interaction correction while the correction for a tunnel contact is dominated by just one term.

As the main result of our numerical analysis we note, that the corrections to cumulants strongly depend on the transmission of the contact. They can have either sign and a distinct dependence on the ratio of voltage and temperature. Both of these facilitate the experimental detection of environmental effects on transport properties. The plots in this section were obtained for an RC environment. In principle one could obtain results for any given  $z_\omega$ . At least qualitatively we expect the corrections due to a (physical) environment to be similar to those presented.

## VII. CONCLUSION

We have studied the interaction correction to full counting statistics of electron transport in a quantum contact. It was shown that the interaction can be modeled by an environmental impedance  $Z_\omega$  in series with the contact. In Sec. II we presented a formulation of the problem in terms of a non-equilibrium Keldysh action. Assuming  $ZG_Q \ll 1$  we proceeded perturbatively and calculated the correction to the cumulant generating function, that is, to any cumulant [Eq. (14)]. This correction splits into three parts corresponding to elastic electron transfer and inelastic transitions with absorption/emission of energy from the environment.

We looked in detail at the structure of the interaction correction and found a re-interpretation in terms of elementary events. This provided a deeper insight into the physics involved and presented a basic check for the obtained expression. Since the full expression, Eq. (14) is a complicated function of temperature and voltage that is not easily understood, we looked at certain limiting cases. In the limit of vanishing temperature we found a particularly simple expression for the correction to any cumulant, Eq. (23). For the opposite limit of vanishing voltage, we realized that any expression between cumulants is due to a universal relation of detailed balance for the generating function that holds irrespective of the concrete structure of the quantum contact and possible interactions, Eq. (30). In order to bridge those limits and to enable the experimental observation of environmental effects on electron transport in a quantum contact, we calculated numerically the correction due to an environment to the first three cumulants for arbitrary voltage, temperature, and different transmission eigenvalues. We have shown that the



corrections show an interesting crossover behavior from voltage to thermal noise at  $eV \simeq k_B T$  as well as a specific non-trivial dependence on transmission eigenvalues. The presented analytical and numerical results facilitate the measurement of the interaction correction.

### ACKNOWLEDGMENTS

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### APPENDIX A: DERIVATION OF THE UNIVERSAL RELATION FOR FCS

In this appendix, we will present the derivation of the universal relation (30) that is valid for FCS of any conductor regardless of its concrete realization. A well-known example of a relation of this type is provided by the fluctuation-dissipation theorem that relates the linear response and Gaussian fluctuations.<sup>24</sup> The same approach can be extended to provide similar relations for nonlinear frequency-dependent responses and non-Gaussian fluctuations<sup>33-35</sup> using Hermiticity, time reversibility, and KMS<sup>36,37</sup> relations. These results cannot be immediately used for our purpose since they are formulated in terms of relations between multipoint Keldysh Green functions rather than in terms of generating functions.

We apply this approach to the most general generating functional of current fluctuations where both voltage applied  $V$  and counting field  $\chi$  depend on time. The derivatives of the functional with respect to  $\chi(t)$  give averages of current operators. The Hamiltonian in the presence of the voltage source can be written as

$$\hat{H}(t) = H_0 - \hbar \Phi(t) \hat{I} / e, \quad \dot{\Phi}(t) = eV(t) / \hbar, \quad (\text{A1})$$

$\hat{I}$  being the operator of full current in the conductor.

We make use of the interaction picture introducing  $\hat{I}(t) = e^{i\hat{H}_0 t / \hbar} \hat{I} e^{-i\hat{H}_0 t / \hbar}$ . The generating function reads<sup>16,17,26</sup>

$$F(\{\phi^+(t)\}, \{\phi^-(t)\}) = \langle \hat{U}^\dagger(\{\phi^-(t)\}) \hat{U}(\{\phi^+(t)\}) \rangle, \quad (\text{A2})$$

$$\hat{U}(\{\phi^+(t)\}) = \tilde{T} \exp\left(i \int dt \phi^+(t) \hat{I}(t) / e\right), \quad (\text{A3})$$

$$\hat{U}^\dagger(\{\phi^-(t)\}) = \tilde{T} \exp\left(-i \int dt \phi^-(t) \hat{I}(t) / e\right), \quad (\text{A4})$$

$$\langle \dots \rangle = \text{Tr}(\dots \hat{\rho}); \quad \hat{\rho} = e^{-\hat{H} / k_B T} / \text{Tr}(e^{-\hat{H} / k_B T}), \quad (\text{A5})$$

where  $\tilde{T}(\tilde{T})$  stands for (anti)time-ordering of the operators and  $\phi^\pm(t) = \varphi(t) \pm \chi(t) / 2$ . This expression is formally equivalent

to the generating functional for multi-point Keldysh Green functions used in Refs. 34 and 35. The only difference is that the Green functions generated are those of current operators.

We shall assume time-reversibility of the Hamiltonian. Since in this case

$$\hat{H}^T = \hat{H}; \quad \hat{I}^T = -\hat{I}; \quad \hat{I}^T(t) = -\hat{I}(-t) \quad (\text{A6})$$

we observe the following transposing rule for  $S$ -operators:

$$(\hat{U}(\{\phi(t)\}))^T = \hat{U}(\{-\phi(-t)\}). \quad (\text{A7})$$

Transposing operators in the average (A2), we obtain

$$F(\{\phi^+(t)\}, \{\phi^-(t)\}) = \langle \hat{U}(\{-\phi^+(-t)\}) \hat{U}^\dagger(\{-\phi^-(-t)\}) \rangle.$$

In comparison with (A2),  $U, U^\dagger$  are interchanged. We want them back to their original positions. The way to do this is to make use of KMS relations: For any operator  $\hat{A}$

$$\hat{\rho} \hat{A}(t) = \hat{A}(t + i\hbar / k_B T) \hat{\rho}. \quad (\text{A8})$$

We do this commutation with all operators comprising  $U^\dagger$  to obtain

$$\hat{\rho} \hat{U}(\{-\phi^+(-t)\}) = \tilde{T} \exp\left(-i \int dt \phi^+(-t) \hat{I}(t + i\hbar / k_B T) / e\right) \hat{\rho}. \quad (\text{A9})$$

We shift now the time argument of  $\phi^+$  by  $i\hbar / k_B T$  to obtain

$$\begin{aligned} & \tilde{T} \exp\left(-i \int dt \phi^+(-t) \hat{I}(t + i\hbar / k_B T) / e\right) \\ &= \hat{U}(\{-\phi^+(-t + i\hbar / k_B T)\}). \end{aligned}$$

This step looks rather heuristic. Since nothing is assumed concerning the analytical properties of  $\phi$  as a function of complex time, the complex shift may be ambiguous. However we note that we are mainly interested in  $\phi^\pm(t)$  that change at time scales much bigger than  $\hbar / k_B T$ : for those, we expect no ambiguity.

Finally, we cycle operators under the sign of trace to obtain

$$F(\{\phi^+(t)\}, \{\phi^-(t)\}) = F(\{-\phi^+(-t + i\hbar / k_B T)\}, \{-\phi^-(-t)\}). \quad (\text{A10})$$

For quasistationary  $V, \chi$  we substitute  $\phi^\pm = eVt / \hbar \pm \chi / 2$  and neglect the dependence on time-independent phase to arrive at

$$F(V, \chi) = F(V, -\chi + ieV / k_B T) \quad (\text{A11})$$

which is the universal relation to prove.

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