Counting statistics of cotunneling electrons

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We describe a method for calculating the counting statistics of electronic transport through nanoscale devices with both sequential and cotunneling contributions. The method is based upon a perturbative expansion of the von Neumann equation in Liouvillian space, with current cumulants calculated from the resulting non-Markovian master equation without further approximation. As application, we consider transport through a single quantum dot and discuss the effects of cotunneling on noise and skewness, as well as the properties of various approximation schemes.

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Cotunneling, the transfer of electrons via intermediate "virtual" states, can be an important mechanism in the transport of electrons through quantum dots (QDs).[1,2] In the Coulomb-blockade (CB) regime, sequential tunneling processes are exponentially suppressed and, since it only suffers an algebraic suppression, cotunneling becomes the dominant current-carrying mechanism. Experimental interest in cotunneling has remained high from the earliest experiments on metallic grains[3] and large quantum dots,[4] through to more modern experiments on few-electron single[5–7] and double[8,9] quantum dots.

From a theoretical perspective, cotunneling refers to processes fourth order in the coupling between the system and the leads, an order which also includes, e.g., the pair tunneling of electrons.[10] Such processes can be taken into account in a number of different ways, see for example Refs. 2 and 11–17. Most relevant here is the real-time diagrammatic approach[14–17] in which higher-order tunneling processes are incorporated into a master equation in a systematic fashion. This theory has been extensively developed and successfully applied to numerous transport problems: not just single QDs but also double dots,[18] quantum dot spin valves,[19] carbon nanotubes,[20] and QD interferometers.[21] While such higher-order calculations have typically been restricted to the stationary current, and more recently, the shot noise,[16,17] much interest presently surrounds the full counting statistics (FCS) of the current, i.e., in current correlations beyond the second-order shot noise.[22–24] The last few years has seen the advent of experiments capable of detecting the passage of single electrons through QD systems[25–29] and the experimental determination of FCS. Recently, measurements of the 15th cumulant were reported for a single quantum dot.[29]

In this paper we bring together several strands in the literature to investigate the influence of cotunneling on FCS. We derive a fourth-order master equation for the reduced density matrix of an arbitrary mesoscopic system using the Liouvillian-space perturbation theory of Refs. 30 and 31. We then extend this formalism by showing how counting fields may be added to the reservoir correlation functions. With these in place, the FCS can be calculated and, given that the master equation derived in this approach is non-Markovian, we employ the formalism of Flindt *et al.*[32] to obtain expressions for the current cumulants.

We use this formalism to investigate the transport through a single QD. We study first the single resonant level (SRL) model. Exact solutions exist for this model and this allows an evaluation of various approximation schemes. We then study the effects of interaction on transport through the QD by considering an Anderson model. Of the higher-order cumulants, we focus on the skewness as the first correlator beyond the shot noise. We compare, both on a formal and a numerical level, with the work of Braggio *et al.*,[33] and with the shot noise results of Thielmann *et al.*[17]

This paper is structured as follows: in Sec. I we introduce a general transport model and in Sec. II, Liouville-Laplace space. These sections also serve to fix the notation for Sec. III in which we review the derivation of a quantum master equation with sequential and cotunneling kernels in the Liouvillian perturbation approach as presented in Refs. 30 and 31. Further technical details to be found in Appendix A. In Sec. IV, we show how counting fields can be incorporated in the Liouvillian perturbation theory and discuss how current cumulants can be calculated from the resulting counting-field-dependent Liouvillian. Section V sees application to our model QD systems and is followed by conclusions.

I. TRANSPORT MODEL

We begin by specifying the general transport setup under consideration here. The total Hamiltonian $H=H_{\rm res}+H_{\rm S}+V$ is composed of reservoir, system, and interaction parts. We write the system part in its diagonal basis $H_{\rm S}=\Sigma_a E_a|a\rangle\langle a|$, where $|a\rangle$ is a many-body system state of N_a electrons. We consider a set of reservoirs labeled with an index α that includes spin and any other relevant quantum numbers. We assume noninteracting reservoirs with Hamiltonian

$$H_{\rm res}=\sum_{k,\alpha}(\omega_{k\alpha}+\mu_\alpha)a^\dagger_{k\alpha}a_{k\alpha}, \tag{1}$$

where $\omega_{k\alpha}$ is the energy of the kth mode in lead α, $a_{k\alpha}$ is a lead annihilation operator, and we have included the chemical potential of lead α, μ_α, at this point for convenience.

To ease book keeping, we introduce a compact single index "1" to denote the triple of indices (ξ_1,k_1,α_1).[31] The first index $\xi_1=\pm$ indicates whether a reservoir operator is a creation or annihilation operator

$$a_1 = a_{\xi_1 k_1 \alpha_1} = \begin{cases} a^\dagger_{k_1\alpha_1}, & \xi_1 = + \\ a_{k_1\alpha_1}, & \xi_1 = - \end{cases} . \tag{2}$$

Leaving sums implicit, the reservoir Hamiltonian reads

$$H_{\rm res} = (\omega_{k\alpha} + \mu_\alpha) a_{+k\alpha} a_{-k\alpha} = (\omega_1 + \mu_1) a_1 a_{\bar{1}} \delta_{\xi_1,+}, \tag{3}$$

where $\bar{1}$ denotes $(-\xi_1,\ k_1,\ \alpha_1)$. In equilibrium, the reservoir electrons are distributed according to the Fermi function

$$f(\omega) = \frac{1}{e^{\omega/k_B T} + 1}, \tag{4}$$

which, since we include the chemical potential in Eq. (1) and assume a uniform temperature, is the same for all reservoirs.

Single-electron tunneling between system and reservoirs is described by the Hamiltonian

$$V = \sum_{k\alpha m} t_{k\alpha m} a^\dagger_{k\alpha} d_m + t^*_{k\alpha m} d^\dagger_m a_{k\alpha}, \tag{5}$$

where d_m is the annihilation operator for single-particle level m in the system and $t_{k\alpha m}$ is a tunneling amplitude. We write this interaction as

$$V = \xi_1 t_{1m} a_1 j_{\xi_1 m}, \tag{6}$$

with coefficients $t_{+k\alpha m} = t_{k\alpha m}$ and $t_{-k\alpha m} = t^*_{k\alpha m}$, and system operators in the many-body system basis

$$j_{+m} = \sum_{aa'} \langle a|d_m|a'\rangle \delta(N_a - N_{a'} + 1) |a\rangle\langle a'|,$$

$$j_{-m} = \sum_{aa'} \langle a|d^\dagger_m|a'\rangle \delta(N_a - N_{a'} - 1) |a\rangle\langle a'|. \tag{7}$$

We have made explicit here the change in system charge induced by the operator. Although these operators only depend on ξ_1, we label them with the full 1 index for convenience: $j_{1m} = j_{\xi_1 m}$.

At time $t=0$ we posit a separable total density matrix

$$\rho(t=0) = \rho_{\rm S}(0) \rho^{\rm eq}_{\rm res}, \tag{8}$$

with the system in arbitrary state $\rho_{\rm S}(t_0)$ and reservoirs in thermal equilibrium.

II. LIOUVILLE-LAPLACE SPACE

We now construct the elements required to perform our perturbation calculation in Liouville-Laplace space. In this section and the next, we follow Refs. 30, 31, and 34 to which the reader is referred for further details. The total density matrix evolves according to the von Neumann equation

$$\dot{\rho}(t) = -i[H, \rho(t)] = \mathcal{L}\rho(t), \tag{9}$$

which defines the Liouvillian superoperator $\mathcal{L} = -i[H, \bullet]$. This Liouvillian consists of three parts,

$$\mathcal{L} = \mathcal{L}_{\rm res} + \mathcal{L}_{\rm S} + \mathcal{L}_V \tag{10}$$

with $\mathcal{L}_{\rm res} = -i[H_{\rm res}, \bullet]$, $\mathcal{L}_{\rm S} = -i[H_{\rm S}, \bullet]$, and $\mathcal{L}_V = -i[V, \bullet]$. We write the interaction Liouvillian as

$$\mathcal{L}_V = -i\xi_1 t_{1m} \sum_p A^p_1 J^p_{1m}, \tag{11}$$

where $p = \pm$ is a Keldysh index corresponding to the two parts of the commutator. Superoperators A and J are defined through their actions on arbitrary operator O: for the reservoir, we have

$$A^p_1 O = \begin{cases} a_1 O, & p = + \\ O a_1, & p = - \end{cases} , \tag{12}$$

and analogously for the system

$$J^p_{1m} O = \begin{cases} j_{1m} O, & p = + \\ O j_{1m}, & p = - \end{cases} . \tag{13}$$

By organizing the elements of density matrices into vectors, superoperators such as the Liouvillian take the form of matrices. This is a particularly convenient representation for the system Liouvillian. We write a general system density matrix, $\rho_{\rm S} = \Sigma_{a_1 a_2} \rho_{a_1 a_2} |a_1\rangle\langle a_2|$, as the vector $|\rho_{\rm S}\rangle\rangle = \Sigma_a \rho_a |\phi_a\rangle\rangle$, where the single index a corresponds to the double (a_1, a_2), such that the "ket" $|\phi_a\rangle\rangle$ corresponds to $|a_1\rangle\langle a_2|$. The action of the free system Liouvillian $\mathcal{L}_{\rm S}$ on vector $|\phi_a\rangle\rangle$ is

$$\mathcal{L}_{\rm S} |\phi_a\rangle\rangle \equiv -i[H_{\rm S}, |a_1\rangle\langle a_2|] = -i\Delta_a |\phi_a\rangle\rangle, \tag{14}$$

where $\Delta_a \equiv E_{a_1} - E_{a_2}$ defines the Bohr frequencies. The vectors $|\phi_a\rangle\rangle$ are therefore the right eigenvectors of Liouvillian $\mathcal{L}_{\rm S}$. The left eigenvectors, $\langle\langle\phi_a|$, fulfill

$$\langle\langle\phi_a| \mathcal{L}_{\rm S} = -i\Delta_a \langle\langle\phi_a| \tag{15}$$

and together with the right eigenvectors form a biorthonormal set: $\langle\langle\phi_a|\phi_{a'}\rangle\rangle = \delta_{a,a'}$. We have the closure relation in Liouvillian space[35]

$$\mathbb{1} = \sum_a |\phi_a\rangle\rangle\langle\langle\phi_a| . \tag{16}$$

In general, it is important to make the distinction between left and right eigenvectors because an arbitrary superoperator, in particular, the effective system Liouvillian, will not be Hermitian, and the left and right eigenvectors are therefore not adjoint.

III. EFFECTIVE LIOUVILLIAN

With the definition of the Laplace transform

$$\rho(z) \equiv \int_0^\infty dt e^{-zt} \rho(t), \tag{17}$$

Eq. (9) yields the solution

$$\rho(z) = \frac{1}{z - \mathcal{L}} \rho(0). \tag{18}$$

Tracing Eq. (18) over reservoir degrees of freedom results in an expression for the reduced density matrix of the system that we write

$$\rho_S(z) = \frac{1}{z - \mathcal{W}(z)}\rho_S(0), \tag{19}$$

where $\mathcal{W}(z)$ is the non-Markovian effective dot Liouvillian. This we write as

$$\mathcal{W}(z) = \mathcal{L}_S + \Sigma(z), \tag{20}$$

with $\mathcal{L}_S$ describing the free evolution of the system and $\Sigma(z)$ the self-energy or "memory kernel" arising from coupling with the leads.

In the perturbative approach pursued here, the memory kernel is calculated as the series $\Sigma(z)=\Sigma_n\Sigma^{(n)}(z)$, where n corresponds to the number of interaction Liouvillians $\mathcal{L}_V$ incorporated in that term. Tunneling is governed by the rates

$$\Gamma^{m_2 m_1}_{\xi_1\alpha_1}(\omega) \equiv 2\pi\sum_{k_1} t^{-}_{1m_2}t_{1m_1}\delta(\omega - \omega_{k_1\alpha_1}), \tag{21}$$

the diagonal elements of which are the familiar Fermi golden rule rates

$$\Gamma^{m_1 m_1}_{\xi_1\alpha_1}(\omega) \equiv 2\pi\sum_{k_1} |t_{1m_1}|^2\delta(\omega - \omega_{k_1\alpha_1}). \tag{22}$$

In these terms, the expansion of $\Sigma(z)$ is seen as an expansion in the rates Γ, such that $\Sigma^{(n)}(z)$ is order $\Gamma^{n/2}$. In the current work, we expand up to fourth order in the coupling Hamiltonian (second order in Γ), such that

$$\Sigma(z) \approx \Sigma^{(2)}(z) + \Sigma^{(4)}(z). \tag{23}$$

The first term describes sequential tunneling and the second cotunneling.

Details of the calculation of the memory kernel terms are given in Appendix A. Assuming a constant tunneling density of states $\Gamma(\omega)=\Gamma$, the sequential term reads

$$\Sigma^{(2)}(z) = J^{p_2}_{2m_2}\frac{-p_2}{z - i\xi_2(\omega_2+\mu_2) - \mathcal{L}_S}J^{p_1}_{1m_1}t_{2m_2}t_{1m_1}\gamma^{p_2 p_1}_{21}, \tag{24}$$

where $\gamma^{p_2p_1}_{21}=\langle A^{p_2}_2 A^{p_1}_1\rangle_{\text{eq}}$ is an equilibrium reservoir correlation function which evaluates as

$$\gamma^{p_2 p_1}_{21} = \delta_{21}\bar{p}_1 f(-\xi_1 p_1\omega_1). \tag{25}$$

As described in Appendix A, switching to an integral representation for the lead states and regularizing by setting $z=0^+-i\epsilon$, we obtain

$$\Sigma^{(2)}(z) = -p_1p_2 J^{p_2}_{1m_2}|\phi_a\rangle\rangle\langle\langle\phi_a| J^{p_1}_{1m_1} \times\Gamma^{m_2m_1}_{\xi_1\alpha_1} I^{(2)}_{p_1}(\Delta_a + \xi_1\mu_{\alpha_1} - \epsilon),$$

where the sequential integral evaluates as

$$I^{(2)}_p(\lambda) = \frac{1}{2}f(p\lambda) + \frac{ip}{2\pi}\phi(\lambda)$$

with

$$\phi(\lambda) = \frac{1}{2}\{g(\lambda) + g(-\lambda)\} - \ln\frac{X_C}{2\pi k_B T},$$

$$g(\lambda) = \Psi\left(\frac{1}{2} + \frac{\lambda}{2\pi i k_B T}\right) \tag{26}$$

with Ψ, the digamma function and X_C a high-energy cutoff [see Eq. (A18)].

The cotunneling term has two contributions: "direct" and "exchange," such that $\Sigma^{(4)}(z)=\Sigma^{(4D)}(z)+\Sigma^{(4X)}(z)$. The direct part is given by

$$\Sigma^{(4D)}(z) = p_4p_1 J^{p_4}_{\bar{1}m_4}|\phi_a\rangle\rangle\langle\langle\phi_a| J^{p_3}_{\bar{2}m_3}|\phi_{a'}\rangle\rangle \times\langle\langle\phi_{a'}| J^{p_2}_{2m_2}|\phi_{a''}\rangle\rangle\langle\langle\phi_{a''}| J^{p_1}_{1m_1} \times\Gamma^{m_4m_1}_{\xi_1\alpha_1}\Gamma^{m_3m_2}_{\xi_2\alpha_2} I^D_{p_1p_2}(\lambda_1^{a''},\lambda_2^{a'},\lambda_3^{a}) \tag{27}$$

and the exchange by

$$\Sigma^{(4X)}(z) = -p_4p_1 J^{p_4}_{\bar{2}m_4}|\phi_a\rangle\rangle\langle\langle\phi_a| J^{p_3}_{\bar{1}m_3}|\phi_{a'}\rangle\rangle \times\langle\langle\phi_{a'}| J^{p_2}_{2m_2}|\phi_{a''}\rangle\rangle\langle\langle\phi_{a''}| J^{p_1}_{1m_1} \times\Gamma^{m_3m_1}_{\xi_1\alpha_1}\Gamma^{m_4m_2}_{\xi_2\alpha_2} I^X_{p_1p_2}(\lambda_1^{a''},\lambda_2^{a'},\lambda_3^{a}). \tag{28}$$

The fourth-order integrals I^D and I^X and their functional dependencies are discussed in Appendix A.

IV. COUNTING STATISTICS

The density matrix of Eq. (19) is the Laplace transform of the solution to the non-Markovian master equation[36]

$$\dot{\rho}_S(t) = \int_0^t dt'\mathcal{W}(t-t')\rho(t'), \tag{29}$$

from which we wish to calculate the FCS. The most convenient way to do this is through the introduction of so-called counting fields χ_α, which are associated with processes that transfer electrons to and from leads.[24] In simple situations (e.g., infinite bias limit,[37] phenomenological models[38]), it is relatively straightforward to add counting fields to the relevant master equation. The various derivatives of the resulting "χ-resolved" Liouvillian $\mathcal{W}(\chi,z)$ can then be used to calculate the (zero-frequency) current cumulants as we discuss below.

We are thus led to the central question of the formal part of this work: how can one add counting fields to the complex kernels arising from the Liouvillian perturbation theory? This is clearly not trivial since, at nth order each term in the Liouvillian can transfer a total of up to n electrons to and from the various reservoirs. Furthermore, there is also the issue of how the counting fields should enter elements of the Liouvillian that concern changes in internal coherences of the system. Within the real-time diagrammatic approach, this question is answered by introducing counting fields into the tunnel Hamiltonian of Eq. (5), which then accrue different signs depending on the Keldysh branch on which they act.[33]

We assert here that the correct assignment of counting fields can also be obtained within the Liouvillian perturbation scheme by replacing each bath contraction $\gamma^{p_2p_1}_{21}$ in the memory kernel $\Sigma(z)$ by the counting-field-dependent analog $\gamma^{p_2p_1}_{21}(\chi)$, which we define as

$$\gamma_{21}^{p_2p_1}(\chi) = \gamma_{21}^{p_2p_1} \exp\left[is_{\alpha_1}\xi_1\left(\frac{p_1 - p_2}{2}\right)\chi_{\alpha_1}\right] \tag{30}$$

with $s_\alpha = \pm 1$ a factor given by the sign convention for current flow in lead α. To see that $\gamma_{21}^{p_2p_1}(\chi)$ adds counting fields at the correct points, let us consider an example with $-\xi_2 = \xi_1 = +$. Then, for $-p_2 = p_1 = +$, the contraction $\gamma_{21}^{p_2p_1}$ is proportional to trace of $a_{k\alpha}^\dagger \rho a_{k\alpha}$, a state with one more electron in lead α than ρ itself. This process should therefore be associated with counting-field factor, $e^{is_\alpha\chi_\alpha}$, which is as obtained from Eq. (30). On the other hand, for $p_2 = p_1 = +$, the contraction is proportional to the trace of $a_{k\alpha}^\dagger a_{k\alpha}\rho$, a state with the same number of electrons as ρ. This term should therefore obtain no counting field, and this is what is returned by Eq. (30). In this way, and through comparison with previous calculations,[17,33] it can be seen that Eq. (30) assigns the counting fields correctly. The addition of counting fields through Eq. (30) is simpler than determining the corresponding counting-field-dependent real-time diagrams and can easily be extended to higher orders. In the following we will only count electrons in a single lead, for which we choose $s_\alpha = 1$.

Once in possession of the χ-resolved Liouvillian, the cumulant generating function (CGF) $\mathcal{F}(\chi) = -tz_0(\chi)$ is obtained from the solution $z_0(\chi)$ of the equation

$$z_0 - \lambda_0(\chi; z_0) = 0, \tag{31}$$

where $\lambda_0(\chi; z_0)$ is the eigenvalue of $\mathcal{W}(\chi; z)$ that develops adiabatically from zero as χ is increased from zero.[39] In the Markovian case, λ_0 is independent of z and the CGF is simply $\mathcal{F}(\chi) = -t\lambda_0(\chi)$. The non-Markovian case is less straightforward, however. We follow here the approach of Ref. 32, which uses Eq. (31) to derive expressions for the cumulants themselves, bypassing an explicit evaluation of the CGF itself. This approach can deliver the cumulants up to, in principle, arbitrary order (>20 in Ref. 40) and is applicable to systems of large size, unlike methods that explicitly require the eigenvalue $\lambda_0(\chi, z)$.

We first define

$$\mathcal{J}(\chi, \epsilon) = \mathcal{W}(\chi, z = 0^+ - i\epsilon) - \mathcal{W}(\chi = 0, z = 0^+), \tag{32}$$

along with the derivatives

$$\mathcal{J}' = \partial_\chi \mathcal{J}|_{\chi,\epsilon\to 0}, \quad \dot{\mathcal{J}} = \partial_\epsilon \mathcal{J}|_{\chi,\epsilon\to 0}, \tag{33}$$

and analogously for higher orders. We define the left and right null vectors of $\mathcal{W}(0, 0^+)$ via

$$\mathcal{W}(0,0^+)\ |\psi_0\rangle\rangle = \langle\langle\psi_0|\ \mathcal{W}(0,0^+) = 0, \tag{34}$$

which we assume to be unique. The vector $|\psi_0\rangle\rangle$ corresponds to the stationary density matrix of the system, and multiplication with $\langle\langle\psi_0|$, corresponds to taking the trace over system states.[39] We define the stationary state "expectation value" $\langle\langle\bullet\rangle\rangle = \langle\langle\psi_0|\bullet|\psi_0\rangle\rangle$ and the projectors $\mathcal{P} = |\psi_0\rangle\rangle\langle\langle\psi_0|$ and $\mathcal{Q} = \mathbb{1} - \mathcal{P}$. Finally, we require the pseudoinverse

$$\mathcal{R}(\epsilon) = \mathcal{Q}\frac{1}{i\epsilon + \mathcal{W}(0,0^+ - i\epsilon)}\mathcal{Q}. \tag{35}$$

From Refs. 32 and 41, the first three current cumulants are

$$\langle I\rangle = \langle I\rangle_c = \langle I\rangle^m \tag{36}$$

$$S = \langle I\rangle^2{}_c = S^m + 2\langle I\rangle\ \langle\langle \dot{\mathcal{J}}' - \mathcal{J}'\mathcal{R}\dot{\mathcal{J}}\rangle\rangle, \tag{37}$$

$$\begin{aligned} S^{(3)} = \langle I\rangle^3{}_c = S^{(3m)} &- \frac{3S}{2\langle I\rangle}(S^m - S) \\ &- 3i\langle I\rangle\langle\langle \ddot{\mathcal{J}}' - 2\dot{\mathcal{J}}'\mathcal{R}\mathcal{J}' - \mathcal{J}'\mathcal{R}\ddot{\mathcal{J}}\rangle\rangle \\ &+ 6i\langle I\rangle\langle\langle \mathcal{J}'\mathcal{R}(\mathcal{R}\dot{\mathcal{J}}\mathcal{P}\mathcal{J}' + \dot{\mathcal{J}}')\rangle\rangle \\ &- 6i\langle I\rangle\langle\langle \mathcal{J}'\mathcal{R}(\mathcal{J}'\mathcal{R}\dot{\mathcal{J}} + \dot{\mathcal{J}}\mathcal{R}\mathcal{J}')\rangle\rangle \\ &+ 3i\langle I\rangle^2\langle\langle \ddot{\mathcal{J}}' + 2\mathcal{J}'\mathcal{R}\dot{\mathcal{J}}\mathcal{R}\dot{\mathcal{J}}\rangle\rangle \\ &- 3i\langle I\rangle^2\langle\langle \mathcal{J}'\mathcal{R}\ddot{\mathcal{J}} + 2\dot{\mathcal{J}}'\mathcal{R}\dot{\mathcal{J}}\rangle\rangle, \end{aligned} \tag{38}$$

where

$$i\langle I\rangle^m = \langle\langle \mathcal{J}'\rangle\rangle \tag{39}$$

$$i^2 S^m = \langle\langle \mathcal{J}'' - 2\mathcal{J}'\mathcal{R}\mathcal{J}'\rangle\rangle \tag{40}$$

$$\begin{aligned} i^3 S^{(3m)} = &\langle\langle \mathcal{J}''' - 3\mathcal{J}''\mathcal{R}\mathcal{J}' - 3\mathcal{J}'\mathcal{R}\mathcal{J}''\rangle\rangle \\ &- 6\langle\langle \mathcal{J}'\mathcal{R}(\mathcal{R}\mathcal{J}'\mathcal{P} - \mathcal{J}'\mathcal{R})\mathcal{J}'\rangle\rangle, \end{aligned} \tag{41}$$

are the cumulants in the Markov approximation. In these expressions, it is understood that the pseudoinverse is evaluated at $\epsilon = 0$. Although it is only practicable to explicitly write down the cumulants up to third order, the high-order cumulants can be obtained recursively.[32]

Reference 33 took a different approach to calculating the cumulants. There it was taken that $\mathcal{W}$ is known to a given order in some small parameter and the CGF then calculated to the same order. For problems such as considered here, this means that the CGF, and hence all the cumulants, are calculated rigorously up to order Γ^2. This is to be contrasted with the above cumulants which, if expanded, have contributions at all orders in Γ since the pseudoinverse Eq. (35) has terms of order Γ (and Γ^2) in the denominator. While the method of Ref. 33 may naively seem more consistent, there are several good reasons why the approach described here might be preferable. First, from Ref. 42 we know that in the infinite bias limit, the effective Liouvillian is given exactly by $\mathcal{L}_S$ plus the rate part of $\Sigma^{(2)}(\epsilon = 0)$. In order to recover the FCS correctly in this limit then, no further approximations should be made when calculating the cumulants. A good example is the shot noise of double quantum dot, for which Eq. (40) reproduces exactly the infinite bias results of,[43] whereas the approach of Ref. 33 would only provide an approximation. It should be noted that this difference is only evident for models with internal quantum degrees of freedom. Second, Refs. 30, 44, and 45 make the point that, in certain circumstances, by treating incoming and outgoing processes unequally, a strict order-by-order approach can lead to unphysical results

for the current. Finally, from a many-body perspective, it seems more natural that once one has summed an infinite class of diagrams to obtain the propagator Ω to leave it in this linked form in subsequent expressions. In the next section, we shall compare the results of these two methods for the single QD models.

Before doing so, we note that comparing the foregoing expressions for the current and the shot noise with those of, e.g., Ref. 17, we can identify the derivatives of $\mathcal{J}$ with respect to χ and z with the various "current superoperator blocks" of the real-time diagrammatic approach. For example, differentiating $\mathcal{J}$ once with respect to $i\chi$ and setting $\chi \to 0$ yields a superoperator similar to $\mathcal{W}$ but with an additional forefactor $\xi_1(p_1-p_2)/2$. Under the summation, the p_2 contribution cancels, leaving a forefactor $\xi_1 p_1/2$. This forefactor is the same as arises in replacing a single tunnel vertex with a current vertex in the self-energy, which is the recipe for obtaining the current superoperator block used to calculate the stationary current in the diagrammatic approach. Using a very different approach, we thus reproduce the real-time current and shot-noise expressions. The advantage of the present method is that it is now easy to obtain to the higher cumulants, whereas with the diagrammatic approach, this requires some effort.

V. TRANSPORT THROUGH A SINGLE QUANTUM DOT

We model the transport through a single Zeeman-split level with the Anderson Hamiltonian[46]

$$H = \sum_{\sigma} \epsilon_{\sigma} d_{\sigma}^{\dagger} d_{\sigma} + U n_{\uparrow} n_{\downarrow} + H_{\mathrm{res}} + V, \tag{42}$$

where ϵ_{σ} is the energy of a spin-σ electron in the dot and U is the interaction energy. The reservoir and interaction Hamiltonians are as Eqs. (1) and (5), with index α including both lead ($=L,R$) and spin index. In the limit of large level splitting we can address one and only one Zeeman level. We then recover the SRL model, the transport properties of which can be obtained exactly from scattering theory.[47,48] The SRL thus provides a useful benchmark against which to compare approximate methods.

We will discuss results obtained in several different approximate schemes. We denote as "O(4)" the results obtained by calculating the cumulants as outlined in the previous section with the fourth-order effective Liouvillian containing both sequential and cotunneling terms. The second-order "O(2)" solution is obtained in the same way but with sequential terms only. We also consider a scheme in which we expand the cumulants and truncate at fourth order. In this way we recover the FCS results of Braggio *et al.*[33] and the shot-noise results of Thielmann *et al.*[17] This approach we label as "O(4) trunc." Finally, we compare with results in the Markovian approximation, which we label with "Mark."

We calculated results with and without the imaginary parts of the fourth-order self-energy. For the models studied here (no internal coherences), these terms only contribute to the non-Markovian terms and it was found that their influence on the results was negligible in all instances studied here. In the results presented below, these parts of the self-energy have been neglected as this considerably reduces the computational effort.

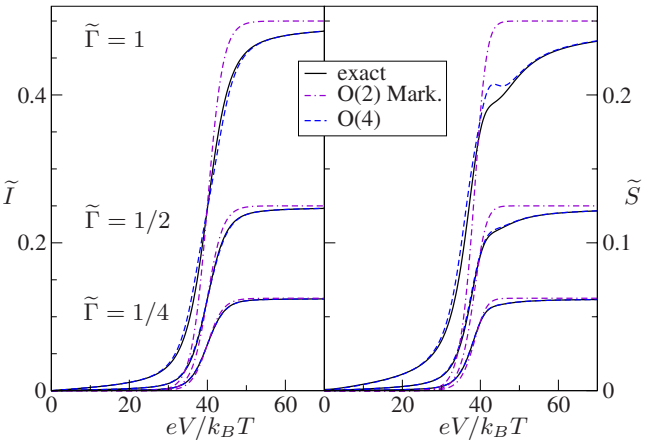

FIG. 1. (Color online) Stationary current $\widetilde{I}=\langle I\rangle/k_BT$ (left) and zero-frequency shot noise $\widetilde{S}=S/k_BT$ (right) as a function of applied bias eV for the single resonant level model with level located at $\varepsilon = 20k_BT$, chemical potentials $\mu_L=-\mu_R=eV/2$, and bandwidth $D = 10^3 k_BT$. Results are shown for three different couplings: $\widetilde{\Gamma} = \Gamma_L/k_BT=\Gamma_R/k_BT=\frac{1}{4},\frac{1}{2},1$ and three calculational schemes: exact, full fourth-order [O(4)], and second-order Markovian [O(2) Mark.] Whereas the O(2) Markovian results show obvious deviations from the exact results for these couplings, the O(4) solution gives good agreement except around the top of the shot-noise step for $\widetilde{\Gamma}=1$.

A. Single resonant level

The calculation of the first three cumulants in the scattering approach is discussed in Appendix B. In the infinite bias limit, we have[49]

$$\langle I\rangle = \frac{\Gamma_L\Gamma_R}{\Gamma}, \quad S = \langle I\rangle \frac{\Gamma_L^2+\Gamma_R^2}{\Gamma^2},$$

$$S^{(3)} = \frac{\langle I\rangle}{\Gamma^4}(\Gamma_L^4 - 2\Gamma_L^3\Gamma_R + 6\Gamma_L^2\Gamma_R^2 - 2\Gamma_L\Gamma_R^3 + \Gamma_R^4),$$

or, for symmetric rates ($\Gamma_L=\Gamma_R$), $\langle I\rangle=\Gamma_L/2$, $S=\Gamma_L/4$, and $S^{(3)}=\Gamma_L/8$.

Figure 1 shows the current and shot noise as a function of applied bias for different values of the coupling $\widetilde{\Gamma}=\Gamma_L/k_BT = \Gamma_R/k_BT$. A step occurs at a bias of $eV \approx 2\varepsilon$ ($=40k_BT$ here) as the level enters the transport window. For $\widetilde{\Gamma} \lesssim \frac{1}{4}$ agreement between the our O(4) fourth-order calculation and the exact result is excellent across the whole bias range. For greater couplings, $\widetilde{\Gamma} \gtrsim 1/2$, deviation from the exact solution is seen in the shot noise around the top of the step which signals the start of the break down of our approach. The difference between the second-order Markovian and the exact solution is stark. In the CB regime ($eV \lesssim 30$ here) the sequential current is almost totally suppressed but the cotunneling current is still considerable. The O(2) Mark. solution also shows significant error in the high-bias ($eV \gtrsim 2\varepsilon$) regime, which arises largely from the Markovian approximation.

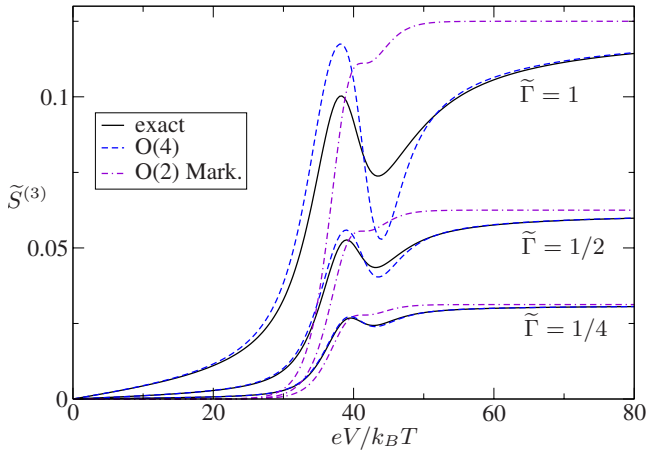

FIG. 2. (Color online) Zero-frequency skewness $\widetilde{S}^{(3)}=S^{(3)}/k_BT$ of SRL with the same parameters as in Fig. 1. For a given coupling, agreement with the exact solution is worse than for shot noise, but still good at low couplings (e.g., $\widetilde{\Gamma}=1/4$).

Figure 2 plots the skewness which, in contrast to the shot noise, show a pronounced undulation at onset. The O(2) Markovian solution provides only the coarsest description of this behavior, whereas it is reproduced by the O(4) solution. For $\widetilde{\Gamma}\lesssim\frac{1}{4}$, the quantitative agreement with the exact result is good. Nevertheless, for a given coupling, the error is larger for skewness than for the noise. Figures 3 and 4 compare the O(4) and O(4) trunc results at $\widetilde{\Gamma}=1/2$. We choose this value

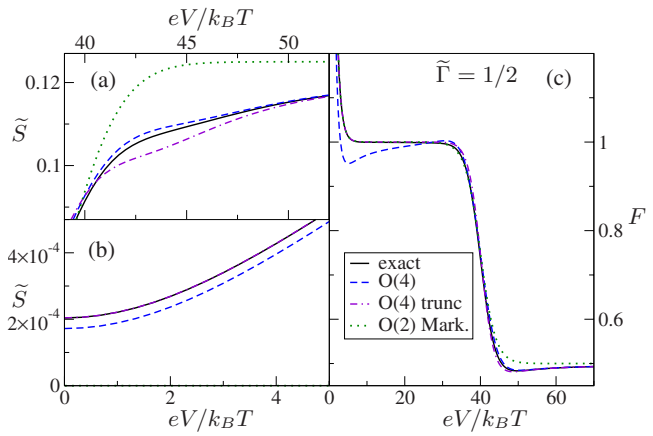

FIG. 3. (Color online) Zero-frequency shot noise (left panels) and Fano factor $F=S/\langle I\rangle$ (right panel) of SRL as a function of applied bias eV. The tunnel rate is fixed at $\widetilde{\Gamma}=1/2$ here but otherwise the parameters are as Fig. 1. In addition to the approximation schemes discussed in Fig. 1, results are also shown here from a rigorous expansion of the cumulants to fourth order [O(4) trunc]. Around the current step [panel (a)], the full O(4) solution describes the behavior better than O(4) trunc. However, in the low bias, Coulomb-blockade regime [panel (b)], it is the O(4) trunc solution that matches the exact solution better. Panel (c) illustrates the dangers of considering the Fano factor alone: although O(4) trunc reproduces the exact Fano factor extremely well in the Coulomb-blockade regime, so does the O(2) Markovian solution, which we know gives the current and shot noise individually extremely poorly.

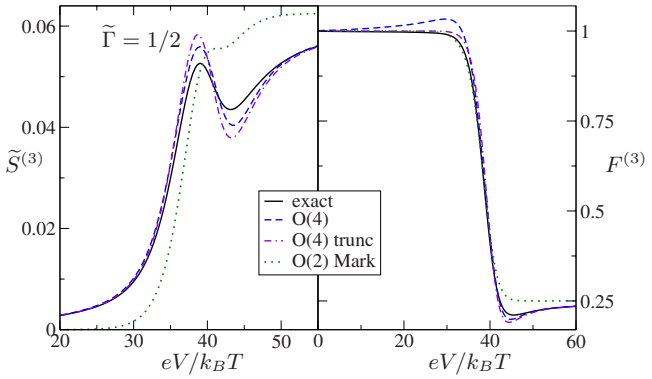

FIG. 4. (Color online) Zero-frequency skewness (left) and associated Fano factor $F^{(3)}=S^{(3)}/\langle I\rangle$ (right) of the SRL model as a function of applied bias eV. Same parameters and labels as Fig. 3. Once again, the O(4) solution performs better in the onset region.

to highlight the differences between the two solutions which, for smaller couplings, are negligible.

Near the top of the step [Figs. 3(a) and 4(a)], the O(4) and O(4) trunc solutions are noticeably different, with our O(4) results significantly closer to the exact result. Deep in the CB regime [Fig. 3(b)], the two approximate solutions differ once more, but this time the O(4) trunc solution is more accurate. The difference is small (a difference in $\widetilde{S}$ of 2×10^{-5} near zero bias); it plays, however, a disproportionate role in determining the Fano factors in the CB regime as Figs. 3(c) and 4(b) show. From the exact solution, we know that noise Fano factor diverges at low bias (fluctuation dissipation theorem), whereas the skewness Fano factor $F^{(n)}\equiv S^{(n)}/\langle I\rangle$ tends to unity. In this regime, both Fano factors are reproduced better by the O(4) trunc solution than by solution O(4).

Figure 5 show the nth-order Fano factors up to and including $n=7$. Below resonance (e.g., $eV/k_BT=30$ in Fig. 5), the O(4) solution predicts superPoissonian cumulants, with magnitude increasing with increasing order n. This is clearly incorrect behavior as the SRL is a noninteracting model for which we expect sub-Poissonian statistics. In this regime the O(4) trunc solution remains sub-Poissonian and thus provides a better description. At or above resonance, both solutions predict sub-Poissonian behavior, with differences appearing between the two solutions which increase with the order of the cumulant. As the 15th cumulant has recently been measured,[29] it is, in principle, possible to test the differences in the predictions of these calculational schemes.

B. Anderson model

The current, shot noise, and Fano factor of the Anderson model with cotunneling were investigated in Ref. 17, and, as Fig. 6 shows, the present calculation broadly reproduces these results. The situation in which the lower dot level lies below the transport window is of particular interest. As observed in Ref. 17, increasing the applied bias results in a large peak in the Fano factor around the point where the upper dot level enters the transport window. The peak exists in the sequential tunnel limit, but its height, width, and location are markedly altered by inelastic cotunneling processes.

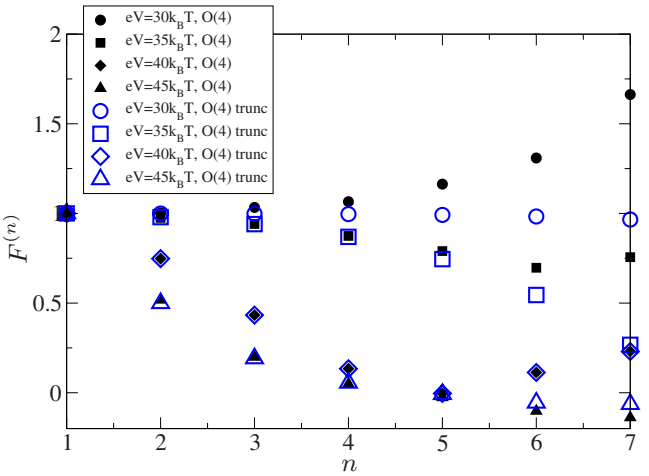

FIG. 5. (Color online) The nth-order Fano factors, $F^{(n)} \equiv S^{(n)}/\langle I \rangle$, up to order $n=7$ for the single resonant level model with parameters as in Fig. 1 and with $\tilde{\Gamma}=1/4$. At low bias, the O(4) solution predicts spurious superPoissonian cumulants, whereas the O(4) trunc cumulants are sub-Poissonian. Around and above resonance, both sets of results are sub-Poissonian with differences between the two results becoming more significant with increasing order. Note that the O(4) and O(4) trunc results are virtually coincident for $eV/k_BT=40$.

No peak occurs in the shot noise itself; only in the Fano factor is this feature visible.

Figure 7 shows our results for the skewness in this situation. It is immediately clear that the skewness Fano factor also shows a peak and that this is even more pronounced than that of the noise. Furthermore, the skewness itself exhibits a sharp peak, as inset Fig. 7(b) shows. As with the shot noise, the presence of cotunneling significantly reduces the height and overall area of the peak in the skewness Fano factor. This superPoissonian behavior indicates a significantly bunched electron flow, which can nicely be explained with the dynamical channel blockade model of Refs. 50 and 51, in which a single level (here, the lower) is but weakly coupled to the collector. In the simple sequential picture of Ref. 51, the shot noise and skewness Fano factors are predicted to be $F_2=(1+p)/(1-p)$ and $F_3=(1+4p+p^2)/(1-p)^2$, where $1/p$ is parameter corresponding to the number of ways in which the dot can be filled. With $p=1/3$ (corresponding to three ways of filling the dot: from the left and right into the lower level, and from the left only into the upper level), we obtain $F_2=2$ and $F_3=11/2$, which are almost exactly the values obtained by our sequential O(2) results at the tops of the peaks. Cotunneling reduces the heights of peaks, and good agreement with the dynamical channel blockade model can be obtained with the choice $p=0.272$.

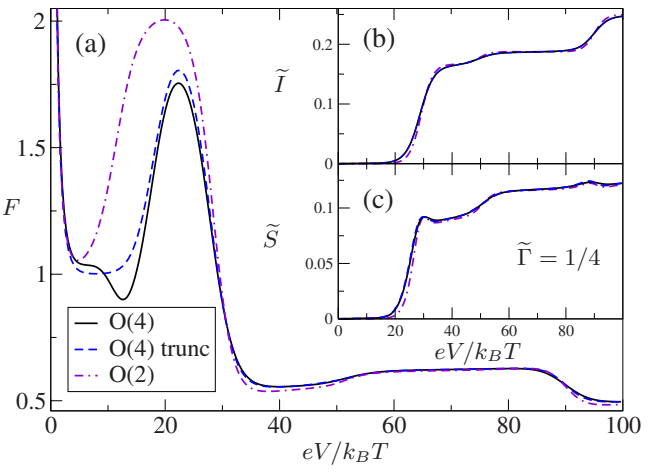

FIG. 6. (Color online) (b) Current, (c) noise, and (a) Fano factor for the Anderson model as a function of applied bias eV. Parameters were chosen as in Ref. 17: $\Gamma_L=\Gamma_R=\frac{1}{4}k_BT$, $\mu_L=-\mu_R=\frac{1}{2}eV$, $\epsilon_\uparrow=-15k_BT$, $\epsilon_\downarrow=5k_BT$, $U=40k_BT$, and bandwidth $D=10^3k_BT$. A prominent peak is observed in the Fano factor around a bias such that the transport window includes the upper level while the lower level is still included. Cotunneling reduces the size of the peak.

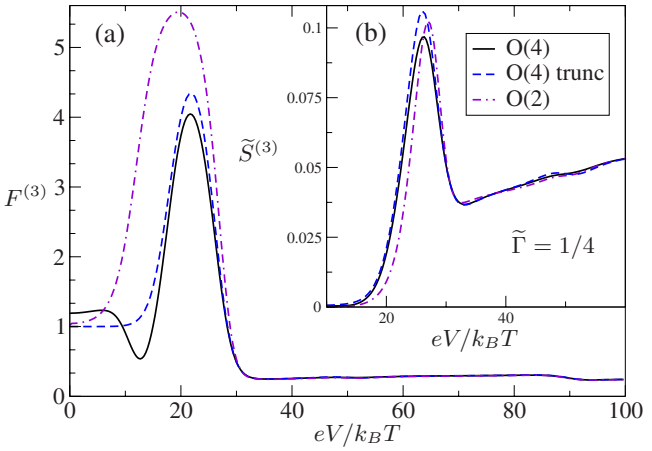

FIG. 7. (Color online) (b) Skewness and (a) skewness Fano factor $F^{(3)}$ for the Anderson model with parameters and labeling as Fig. 6. Not only the skewness Fano factor but the skewness itself show a large peak as the top level enters the transport window.

Both noise and skewness figures show results for O(4) and O(4) trunc solutions. At high bias, these solutions agree closely with one another and both predict the same position and widths of the Fano factor peaks. However, the heights of the peaks are given differently in the two approaches with the O(4) trunc peaks being somewhat higher. Differences between the two solutions are most pronounced at low bias, when the Coulomb blockade is in effect. For example, the O(4) solution predicts a small sub-Poissonan dip before the superPoissonian peak, which is absent in the O(4) trunc results. From our studies of the SRL model, we expect the O(4) trunc prediction to be more accurate in this regime, and this is borne out by the fact that the skewness Fano factor in the O(4) calculation does not tend to unity with decreasing bias. Conversely, based on the SRL results, we expect the height of the Fano factor peaks to be better described by the O(4) solution, i.e., the lower of the two values.

VI. CONCLUSIONS

We have described a method for calculating the counting statistics of an arbitrary mesoscopic system taking into account both sequential tunneling and cotunneling of electrons.

This method is based upon a perturbative expansion of the von Neumann equation in Liouville-Laplace space. The key element that we add to this formalism here is a prescription for how counting fields can be included in the reservoir contractions, such that we obtain a χ-dependent kernel from which all the current cumulants may be calculated. We believe that this Liouvillian approach is particularly convenient and could, in principle, be extended to higher orders or other computational schemes.

Once in possession of a χ-dependent kernel, current cumulants can be obtained, and we have discussed here two different approaches: the pseudoinverse method advocated here after the kernels have been derived in which no further approximations are made and the approach of Ref. 33 in which cumulants are calculated to order Γ^2. From our study of simple quantum-dot models we can draw some tentative conclusions about the relative merits of these two approaches. For very small Γ or at high bias, the results are very similar. For moderate Γ, the full pseudoinverse method gives slightly better results around resonance, whereas, below resonance, with the system in the CB regime, the truncated scheme provides a more reliable description—and, in particular, in the calculation of the Fano factors. It should be borne in mind that, in obtaining the Fano factors in this Coulomb-blockade regime, one is dividing one very small quantity by another and therefore even small absolute errors can effect Fano factors quite dramatically. By truncating rigorously at second order, cancellation between numerator and denominator is far better than in the O(4) calculation, yielding better Fano factors. As a warning, however, not to take the finer details of the Fano factor too seriously, we observe that the second-order Markovian solution in the CB regime reproduces the exact Fano factors better than any of the fourth-order results. This is deceptive since the individual current and shot noise obtained with this method are vastly different from the exact results.

As an example of the application of this formalism, we have studied the shot noise, and, in particular, the skewness of the current through an interacting single QD. We predict a sharp peak in the skewness itself around the inelastic cotunneling conditions and have discussed an interpretation in terms of dynamical channel blockade.

Future work includes the study of transport models with internal quantum degrees of freedom, such as the double quantum dot. It is anticipated that for such models, the advantages of the nontruncated FCS calculation will be more evident.

ACKNOWLEDGMENTS

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APPENDIX A: DERIVATION OF EFFECTIVE SYSTEM LIOUVILLIAN

Following Refs. 30, 31, and 34 we start by defining the system operators

$$g_{k\alpha} = \sum_m t_{k\alpha m} j_m, \tag{A1}$$

such that the interaction Hamiltonian of Eq. (5) can be written as $V=\xi_1 a_1 g_1$. Correspondingly, in Liouville space we have

$$\mathcal{L}_V = -i\xi_1 \sum_p p\sigma^p A_1^p G_1^p \tag{A2}$$

with A as before, and G defined via

$$G_1^p O = \sigma^p \begin{cases} g_1 O, & p=+ \\ -O g_1, & p=- \end{cases}. \tag{A3}$$

The object σ^p is a dot-space superoperator with matrix elements[31]

$$(\sigma^p)_{ss',\bar{s}\bar{s}'} = \delta_{s\bar{s}}\delta_{s'\bar{s}'} \begin{cases} 1, & N_s - N_{s'} = \text{even} \\ p, & N_s - N_{s'} = \text{odd} \end{cases}, \tag{A4}$$

where, N_s is the number of electrons in state s. Note that $G_1^p = p\sigma^p t_{1m} J_{1m}^p$.

The reduced density matrix of the dot is given by tracing out the electron reservoirs

$$\rho_S(z) = \mathrm{Tr}_R\{\rho(z)\} = \mathrm{Tr}_R\left\{\frac{1}{z-\mathcal{L}}\rho(0)\right\}. \tag{A5}$$

This we expand in powers of $\mathcal{L}_V$ to obtain

$$\rho_S(z) = \mathrm{Tr}_R\{[\Omega_0(z) + \Omega_0(z)\mathcal{L}_V\Omega_0(z) + \cdots]\rho(0)\} \tag{A6}$$

with free propagator $\Omega_0(z)=[z-\mathcal{L}_{\mathrm{res}}-\mathcal{L}_S]^{-1}$. With substitution of Eq. (A2), a typical term of the expansion Eq. (A6) reads

$$(-i)^n \left(\prod_{l=1}^{n} \xi_l p_l\right) \mathrm{Tr}_R\{\Omega_0(z)\sigma^{p_n} A_n^{p_n} G_n^{p_n} \ldots \sigma^{p_1} A_1^{p_1} G_1^{p_1} \Omega_0(z)\rho(0)\}. \tag{A7}$$

Evaluating the action of the σ^p superoperators we obtain a factor $\Pi_l^{\mathrm{odd}} p_l$ and, as the G operators also contain σ, they evaluate at different positions in the chain as

$$G_l^{p_l} O = (p_l)^{l+1} \begin{cases} g_l O, & p_l = + \\ O g_l, & p_l = - \end{cases}, \tag{A8}$$

or, in other words, $G_l^{p_l} \to (p_l)^{l+1} t_{lm_l} J_{lm_l}^{p_l}$. Our typical term then looks like

$$(-i)^n \left(\prod_{l}^{\mathrm{all}} \xi_l p_l\right)\left(\prod_{l'}^{\mathrm{odd}} p_{l'}\right) \times \mathrm{Tr}_R\{\Omega_0(z) A_n^{p_n} G_n^{p_n} \ldots A_1^{p_1} G_1^{p_1} \Omega_0(z)\rho(0)\}, \tag{A9}$$

and the next task is to separate dot and reservoir degrees of freedom. For this we can use the dot-reservoir superoperator commutation relation $A_1^p G_{1'}^{p'} = -pp' G_{1'}^{p'} A_1^p$. We will also need the following relations: $\mathrm{Tr}_R\, \mathcal{L}_{\mathrm{res}}=0$, $\mathcal{L}_{\mathrm{res}}\rho_{\mathrm{res}}^{\mathrm{eq}}=0$, and $A_1^p \mathcal{L}_{\mathrm{res}} = (\mathcal{L}_{\mathrm{res}} - x_1)A_1^p$ with

$$x_1 = -i\xi_1(\omega_1 + \mu_{\alpha_1}). \tag{A10}$$

The commutation of the dot operators through the free propagators therefore changes the argument of the propagator

$$A_1^p \Omega_0(z) = \Omega_0(z + x_1) A_1^p. \tag{A11}$$

Bringing all the A operators to the right of the G operators generates a factor which exactly cancels with the first product in Eq. (A9). Our term becomes

$$(-i)^n \left(\prod_{l'}^{\text{odd}} p_{l'} \right) \mathrm{Tr}_R \{ \Omega_S(z) G_n^{p_n} \Omega_S(z_{n-1}) G_{n-1}^{p_{n-1}} \ldots \times G_2^{p_2} \Omega_S(z_1) G_1^{p_1} \Omega_S(z) A_n^{p_n} \ldots A_1^{p_1} \rho(0) \} \tag{A12}$$

with free dot propagator

$$\Omega_S(z) = \frac{1}{z - \mathcal{L}_S}, \tag{A13}$$

and $z_m = z + \Sigma_{l=m+1}^n x_l$; $1 \le m \le n-1$.

The reservoir expectation values, $\mathrm{Tr}_{\text{res}}\{A_n^{p_n} \ldots A_1^{p_1} \rho_{\text{res}}^{\text{eq}}\} = \langle A_n^{p_n} \ldots A_1^{p_1} \rangle_{\text{eq}}$, can be evaluated with the rules of Wick's theorem in Liouville space, which read:[30] (1) decompose $\langle \ldots \rangle_{\text{res}}$ into pair contractions, (2) add minus sign for each interchange of A, (3) omit factor $(\Pi_{l'}^{\text{odd}} p_{l'})$ arising from σ superoperators, and (4) each pair contraction then contributes a factor

$$\langle A_2^{p_2} A_1^{p_1} \rangle = \gamma_{21}^{p_2 p_1} = \delta_{2\bar{1}} p_1 f(-\xi_1 p_1 \omega_1). \tag{A14}$$

With these rules, our typical term becomes

$$(-i)^n \Omega_S(z_n) G_n^{p_n} \Omega_S(z_{n-1}) G_{n-1}^{p_{n-1}} \ldots G_2^{p_2} \Omega_S(z_1) G_1^{p_1} \Omega_S(z) \rho_S(t_0) \times \left(\sum_{\text{decomps}} (-1)^{N_P} \prod \gamma_{ij} \right), \tag{A15}$$

where the last factor indicates a sum over all pair decompositions with the relevant Wick sign $(-1)^{N_P}$.

Comparison of this expression with Eqs. (19) and (20) allows us to identify the self-energy as

$$\Sigma(z) = \sum_n^{\text{even}} (-i)^n \left(\sum_{\text{irred.}} (-1)^{N_P} \prod \gamma_{ij} \right) \times G_n^{p_n} \Omega_S(z_{n-1}) G_{n-1}^{p_{n-1}} \ldots G_2^{p_2} \Omega_S(z_1) G_1^{p_1},$$

where the sum is over *irreducible* contractions only.

1. Second order

At second order, there is only one contraction, and we have

$$\Sigma^{(2)}(z) = G_2^{p_2} \frac{-1}{z + x_2 - \mathcal{L}_S} G_1^{p_1} \gamma_{21}^{p_2 p_1} = G_{\bar{1}}^{p_2} \frac{-p_1 f(-\xi_1 p_1 \omega_1)}{z + i\xi_1(\omega_1 + \mu_{\alpha_1}) - \mathcal{L}_S} G_1^{p_1} = -G_{\bar{1}}^{p_2} |\phi_a\rangle\rangle\langle\langle\phi_a| G_1^{p_1} \frac{p_1 f(-\xi_1 p_1 \omega_1)}{z + i\xi_1(\omega_1 + \mu_{\alpha_1}) + i\Delta_a},$$

where a summation over a has been left implicit. Re-expressing the G superoperators in terms of J superoperators and tunnel amplitudes, we have

$$\Sigma^{(2)}(z) = -p_1 p_2 J_{\bar{1}m_2}^{p_2} |\phi_a\rangle\rangle\langle\langle\phi_a| J_{1m_1}^{p_1} \times t_{\bar{1}m_2} t_{1m_1} \frac{f(-\xi_1 p_1 \omega_1)}{z + i\xi_1(\omega_1 + \mu_{\alpha_1}) + i\Delta_a}. \tag{A16}$$

With rates defined as in Eq. (21) in the constant tunneling density of states approximation, Eq. (A16) becomes

$$\Sigma^{(2)}(z) = -p_1 p_2 J_{\bar{1}m_2}^{p_2} |\phi_a\rangle\rangle\langle\langle\phi_a| J_{1m_1}^{p_1} \Gamma_{\xi_1\alpha_1}^{m_2 m_1} I_{p_1}^{(2)}(z; \Delta_a, \xi_1, \mu_1), \tag{A17}$$

where we have converted the sum over lead degrees of freedom into the integral

$$I_{p_1}^{(2)} = \frac{1}{2\pi} \int d\omega_1 \frac{f(-\xi_1 p_1 \omega_1)}{z + i\xi_1(\omega_1 + \mu_{\alpha_1}) + i\Delta_a}.$$

To evaluate this integral, we regularize it by setting $z \to 0^+ - i\epsilon$ with ϵ wholly real. The self-energy then becomes

$$\Sigma^{(2)}(z = 0^+ - i\epsilon) = -p_1 p_2 J_{\bar{1}m_2}^{p_2} |\phi_a\rangle\rangle\langle\langle\phi_a| J_{1m_1}^{p_1} \times \Gamma_{\xi_1\alpha_1}^{m_2 m_1} I_{p_1}^{(2)}(\Delta_a + \xi_1 \mu_1 - \epsilon)$$

with

$$I_p^{(2)}(\lambda) = \frac{i}{2\pi} \int d\omega_1 \mathcal{D}(\omega - \lambda) \frac{f(\omega_1)}{i0^+ + p\omega_1 - \lambda}.$$

In this integral we have introduced the Lorentzian cutoff function

$$\mathcal{D}(\omega) = \frac{X_C^2}{\omega^2 + X_C^2} \tag{A18}$$

with cutoff energy X_C assumed much larger than all other energy scales in the problem. This cutoff insures that individual integrals remain finite. Contour integration yields

$$I_p^{(2)}(\lambda) = \frac{1}{2} f(p\lambda) + \frac{ip}{2\pi} \phi(\lambda)$$

with the function

$$\phi(\lambda) = \frac{1}{2}[g(\lambda) + g(-\lambda) - g(\lambda + iX_C) - g(-\lambda + iX_C)],$$

in terms of the digamma function of Eq. (26). In the limit of large X_C, we obtain the form given in Eq. (26), correct to order $1/X_C$.

2. Fourth order

At fourth order, there are two linked contractions, (41)(32) and (42)(31), which we label "*D*" for direct and "*X*" for exchange. The direct contribution reads

$$\Sigma^{(4D)}(z) = G_{\bar{1}}^{p_4}\Omega_S(z_3)G_{\bar{2}}^{p_3}\Omega_S(z_2)G_2^{p_2}\Omega_S(z_1)G_1^{p_1}\gamma_{41}^{p_4p_1}\gamma_{32}^{p_3p_2}. \tag{A19}$$

This evaluates as Eq. (27) with the integral

$$I^D_{P_1P_2}(\lambda_1^{a''},\lambda_2^{a'},\lambda_3^a) = \frac{1}{(2\pi)^2}\frac{-i}{\lambda_3^a - \lambda_1^{a''}}\int d\omega_1 \int d\omega_2 \times \frac{f(p_1\omega_1)f(p_2\omega_2)}{(i0^+ + \omega_1 + \omega_2 - \lambda_2^{a'})(i0^+ + \omega_1 - \lambda_3^a)} + (\lambda_1^{a''} \leftrightarrow \lambda_3^a) \tag{A20}$$

with $\lambda_1^{a''} = \xi_1\mu_{\alpha_1} + \Delta_{a''} - \epsilon$, $\lambda_2^{a'} = \xi_1\mu_{\alpha_1} + \xi_2\mu_{\alpha_2} + \Delta_{a'} - \epsilon$, and $\lambda_3^a = \xi_1\mu_{\alpha_1} + \Delta_a - \epsilon$. Note that here the $(\lambda_1^{a''} \leftrightarrow \lambda_3^a)$ symbol includes the preintegral forefactor. Use of partial fraction decomposition allows this integral to be written as

$$I^D_{P_1P_2}(\lambda_1,\lambda_2,\lambda_3) = \frac{-i}{(2\pi)^2}\frac{I^{(4)}_{p_1p_2}(\lambda_2,\lambda_3) - I^{(4)}_{p_1p_2}(\lambda_2,\lambda_1)}{\lambda_3 - \lambda_1} \tag{A21}$$

with

$$I^{(4)}_{p_1p_2}(\lambda_2,\lambda_3) = \int d\omega_1 d\omega_2 \frac{f(p_1\omega_1)f(p_2\omega_2)}{(i0^+ + \omega_1 + \omega_2 - \lambda_2)(i0^+ + \omega_1 - \lambda_3)} = \frac{2\pi}{i}\int d\omega_1 \mathcal{D}(\omega - \lambda_3)\frac{f(p_1\omega_1)}{(i0^+ + \omega_1 - \lambda_3)} I^{(2)}_{p_2}(\lambda_2 - \omega), \tag{A22}$$

where we have explicitly included the cutoff function. Similarly, the exchange term is

$$\Sigma^{(4X)}(z) = -G_{\bar{2}}^{p_4}\Omega_S(z_3)G_{\bar{1}}^{p_3}\Omega_S(z_2)G_2^{p_2}\Omega_S(z_1)G_1^{p_1}\gamma_{42}^{p_4p_2}\gamma_{31}^{p_3p_1}, \tag{A23}$$

which yields Eq. (28) with exchange integral

$$I^X_{p_1p_2}(\lambda_1^{a''},\lambda_2^{a'},\lambda_3^a) = \frac{1}{(2\pi)^2}\frac{-i}{\lambda_2^{a'} - \lambda_3^a - \lambda_1^{a''}}\int d\omega_1 \int d\omega_2 f(p_1\omega_1)f(p_2\omega_2) \times \left(\frac{1}{i0^+ + \omega_1 - \lambda_1^{a''}} + \frac{1}{i0^+ + \omega_2 - \lambda_3^a}\right) \times \left(\frac{1}{i0^+ + \omega_1 + \omega_2 - \lambda_2^{a'}} - \frac{1}{i0^+ + \omega_1 + \omega_2 - \lambda_1^{a''} - \lambda_3^a}\right) \tag{A24}$$

with $\lambda_1^{a''} = \xi_1\mu_{\alpha_1} + \Delta_{a''} - \epsilon$, $\lambda_2^{a'} = \xi_1\mu_{\alpha_1} + \xi_2\mu_{\alpha_2} + \Delta_{a'} - \epsilon$, and $\lambda_3^a = \xi_2\mu_{\alpha_2} + \Delta_a - \epsilon$. This integral can then be written as

$$I^X_{p_1p_2}(\lambda_1,\lambda_2,\lambda_3) = \frac{-i}{(2\pi)^2}\left\{\frac{I^{(4)}_{p_1p_2}(\lambda_2,\lambda_1) - I^{(4)}_{p_1p_2}(\lambda_1+\lambda_3,\lambda_1)}{\lambda_2 - \lambda_3 - \lambda_1} + \frac{I^{(4)}_{p_2p_1}(\lambda_2,\lambda_3) - I^{(4)}_{p_2p_1}(\lambda_1+\lambda_3,\lambda_3)}{\lambda_2 - \lambda_3 - \lambda_1}\right\}. \tag{A25}$$

Both Eqs. (A21) and (A25) are written as differential quotients such that for the cases in which the denominators disappear [e.g., $\lambda_1 = \lambda_3$ in Eq. (A21)] the integrals can be expressed as derivatives.[30]

It then remains to evaluate the integral of Eq. (A22) via contour integration. The imaginary part of this integral was discussed in Ref. 30 and reads

$$\mathrm{Im}[I^{(4)}_{p_1p_2}(\lambda_2,\lambda_3)] = p_1p_2F(\lambda_2,\lambda_3) + p_1\tilde{F}(\lambda_3) \tag{A26}$$

with

$$F(\lambda_2,\lambda_3) = -\pi\left\{-b(\lambda_2)[\phi(-\lambda_3) - \phi(\lambda_2 - \lambda_3)] - \frac{1}{2}\phi(\lambda_3) + \phi(\lambda_2 - \lambda_3)f(\lambda_3)\right\},$$

$$\tilde{F}(\lambda_3) = -\frac{\pi}{2}\phi(\lambda_3), \tag{A27}$$

and $b(x) = (e^{x/T} - 1)^{-1}$ the Bose-Einstein distribution. For the real part, we find

$$\mathrm{Re}[I^{(4)}_{p_1p_2}(\lambda_2,\lambda_3)] = p_1p_2G(\lambda_2,\lambda_3) + p_1\tilde{G}(\lambda_3) + p_1p_2H(\lambda_2,\lambda_3) + \mathrm{const}, \tag{A28}$$

where the constant cancels in both I^D and I^X. In the large X_C limit, we can approximate the constituent functions as

$$G(\lambda_2,\lambda_3) = -\pi^2\left[b(\lambda_2) + \frac{1}{2}\right][f(\lambda_{32}) - f(\lambda_3)] + \frac{1}{2}\left(\phi(\lambda_{32})[g(\lambda_3) + g(-\lambda_3)] - \log\left(\frac{X_C}{2\pi T}\right)\{\phi(\lambda_{32} + iX_C) + \phi(-\lambda_{32} + iX_C)]\}\right), \tag{A29}$$

$$\tilde{G}(\lambda_3) = -\frac{\pi^2}{2}f(\lambda_3), \tag{A30}$$

$$H(\lambda_2,\lambda_3) = \pi\,\mathrm{Im}\sum_{k=\mathrm{odd}}\left[g(-\lambda_2+i\pi k)\left\{\frac{1}{\lambda_{32}-iX_C+i\pi k}+\frac{1}{\lambda_{32}+iX_C+i\pi k}-\frac{2}{\lambda_{32}+i\pi k}\right\}-g(-\lambda_2+iX_C+i\pi k)\left\{\frac{1}{\lambda_{32}+i\pi k}+\frac{1}{\lambda_{32}+2iX_C+i\pi k}-\frac{2}{\lambda_{32}+iX_C+i\pi k}\right\}\right], \tag{A31}$$

with $\lambda_{ij}\equiv\lambda_i-\lambda_j$ and with the sum over all odd integers, $k=1,3,5,\ldots$ For numerical evaluation, this sum can simply be truncated at a sufficiently large odd integer.

APPENDIX B: FIRST THREE CUMULANTS IN SCATTERING APPROACH

In the two-terminal scattering formalism,[47,48] the average current and shot noise at finite temperature and bias are given by the well-known expressions

$$I = \frac{1}{2\pi}\int dE T(E)[f_L(E)-f_R(E)],$$

$$S = \frac{1}{2\pi}\int dE\{T(E)[f_L(1-f_L)+f_R(1-f_R)] + T(E)[1-T(E)](f_L-f_R)^2\},$$

where $T(E)$ is the transmission probability of the device and f_X is the Fermi function of lead X. We have set here $e=\hbar=1$ and define the correlation functions in agreement with those of FCS. The corresponding expression for the skewness at finite temperature and bias is less well known. However, from the results for the symmetrized correlator of Ref. 52 (the appropriate quantity here), we have

$$S^{(3)} = S^{(3)}_{\mathrm{SYM}} = \frac{1}{2\pi}\int dE\{3S_{ioo}(E)-S_{ooo}(E)\} \tag{B1}$$

with

$$S_{ioo} = (1-T)^2 f_L(1-f_L)(1-2f_L) + T(1-T)f_L(1-f_L)(1-2f_R),$$

$$S_{ooo} = (1-T)^3 f_L(1-f_L)(1-2f_L) + T(1-T)^2 a_{LR} + T^2(1-T)a_{RL} + T^3 f_R(1-f_R)(1-2f_R),$$

and

$$a_{XY} = f_X(1-f_X)(1-2f_Y) + f_X(1-f_Y)(1-2f_X) + f_Y(1-f_X)(1-2f_X). \tag{B2}$$

In the infinite bias limit, $f_L=1$ and $f_R=0$, we recover

$$S^{(3)}_{\mathrm{SYM}} = \frac{1}{2\pi}\int dE\{T(E)[1-T(E)][1-2T(E)]\},$$

which is the more familiar expression for skewness in the scattering approach.

From Ref. 48 the transmission probability of the single resonant level at energy ϵ with partial widths Γ_L and Γ_R is

$$T(E) = \frac{\Gamma_L\Gamma_R}{(E-\epsilon)^2+(\Gamma_L+\Gamma_R)^2/4}. \tag{B3}$$

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