Frequency-dependent counting statistics in interacting nanoscale conductors

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We present a formalism to calculate finite-frequency current correlations in interacting nanoscopic conductors. We work within the n-resolved density matrix approach and obtain a multi-time cumulant generating function that provides the fluctuation statistics solely from the spectral decomposition of the Liouvillian. We apply the method to the frequency-dependent third cumulant of the current through a single resonant level and through a double quantum dot. Our results, which show that deviations from Poissonian behaviour strongly depend on frequency, demonstrate the importance of finite-frequency higher-order cumulants in fully characterizing transport.

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Following the considerable success of shot-noise in the understanding of transport through mesoscopic systems [1], attention is now turning towards the higher-order statistics of electron current. The so-called Full Counting Statistics (FCS) of electron transport yields all moments (or cumulants) of the probability distribution \( P(n, t) \) of the number of transferred electrons during time \( t \). Despite their difficulty, measurements of the third moment of frequency-dependent current correlators of arbitrary order in the context of the DM approach, — a Quantum Optics technique [14] — a Quantum Optics technique [14], — a Quantum Optics technique [14], promise to open new horizons on the experimental side.

The theory of FCS is now well established in the zero-frequency limit [2, 3, 4]. However, this is by no means the full picture, since the higher-order current correlators at finite frequencies contain much more information than their zero-frequency counterparts. Already at second order (shot-noise), one can extract valuable information about transport time scales and correlations. When the conductor has various intrinsic time scales like, for example, the charge relaxation time and the dwelling time of a chaotic cavity [10], one needs to go beyond second-order in order to fully characterize electronic transport. Apart from this example, and some other notable exceptions [11, 12, 13], the behaviour of finite-frequency correlators beyond shot-noise is still largely unexplored.

In this Rapid Communication, we develop a theory of frequency-dependent current correlators of arbitrary order in the context of the n-resolved density matrix (DM) approach, — a Quantum Optics technique [14] — a Quantum Optics technique [14] — a Quantum Optics technique [14] — a Quantum Optics technique [14] that has recently found application in mesoscopic transport [15]. Within this approach, the DM of the system, \( \rho(t) \), is unravelled into components \( \rho^{(n)}(t) \) in which \( n = n(t) = 0, 1, \ldots \) electrons have been transferred to the collector. Considering a generic mesoscopic system with Hamiltonian \( \mathcal{H} = \mathcal{H}_S + \mathcal{H}_L + \mathcal{H}_T \), where \( \mathcal{H}_S \) and \( \mathcal{H}_L \) refer to the system and leads respectively, and provided that the Born-Markov approximation with respect to the tunnelling term \( \mathcal{H}_T \) is fulfilled, the time-evolution of this n-resolved DM can be written quite generally as

\[
\dot{\rho}^{(n)}(t) = \mathcal{L}_0 \rho^{(n)}(t) + \mathcal{L}_I \rho^{(n-1)}(t),
\]

where the vector \( \rho^{(n)}(t) \) contains the nonzero elements of the DM, written in a suitable many-body basis. The Liouvillian \( \mathcal{L}_0 \) describes the ‘continuous’ evolution of the system, whereas \( \mathcal{L}_I \) describes the quantum jumps of the transfer of an electron to the collector. We make the infinite bias voltage approximation such that the transfer is unidirectional. By construction, this method is very powerful for studying interacting mesoscopic systems that are weakly coupled to the reservoirs, such as coupled quantum dots (QDs) in the Coulomb Blockade (CB) regime [3, 12, 16] or Cooper-pair boxes [17]. Within this framework, our theory of frequency-dependent FCS is of complete generality and therefore of wide applicability.

In this n-resolved picture, electrons are transferred to the leads via quantum jumps and there exists no quantum coherence between states in the system and those in the leads. Thus, although the system itself may be quantum, the measured current may be considered a classical stochastic variable and therefore amenable to classical counting [18]. This observation allows us to derive various generalizations of classical stochastic results, and obtain a multi-time cumulant generating function in terms of local propagators. We illustrate our method by calculating the frequency-resolved third cumulant (skewness) for two paradigms of CB mesoscopic transport: the single resonant level (SRL) and the double quantum dot (DQD).

Equation (1) can be solved by Fourier transformation. Defining \( \rho(\chi, t) = \sum_n \rho^{(n)}(t) e^{i n \chi} \), we obtain \( \rho(\chi, t) = M(\chi) \rho(\chi, t) \), with \( M(\chi) = \mathcal{L}_0 + e^{i \chi} \mathcal{L}_I \). Let \( N_v \) be the dimension of \( M(\chi) \), and \( \lambda_i(\chi) \); \( i = 1, \ldots, N_v \) its eigenvalues. In the \( \chi \to 0 \) limit, one of these eigenvalues, \( \lambda_1(\chi) \) say, tends to zero and the corresponding eigenvector gives the stationary DM for the system. This single eigenvalue is sufficient to determine the zero-frequency FCS [1]. In contrast, here we need all \( N_v \) eigenvalues. Using
the spectral decomposition, \( M(\chi) = V(\chi) \Lambda(\chi) V^{-1}(\chi) \), with \( \Lambda(\chi) \) the diagonal matrix of eigenvalues and \( V(\chi) \) the corresponding matrix of eigenvectors, the DM of the system at an arbitrary time \( t \) is given by

\[
\rho(\chi, t) = \Omega(\chi, t) - \delta_{\chi t} \rho(t_0),
\]

where \( \Omega(\chi; t) \equiv e^{M(\chi)t} = V(\chi)e^{A(\chi)t}V^{-1}(\chi) \) is the propagator in \( \chi \)-space, and \( \rho(\chi, t_0) \) is the (normalized) state of the system at \( t_0 \), at which time no electrons have passed so that \( \rho^{(n)}(t_0) = \delta_{\chi t} \rho(t_0) \) and thus \( \rho(\chi, t_0) \equiv \rho(t_0) \). The propagator in \( n \)-space, \( G(n, t) \equiv \int \frac{d^2\pi}{(2\pi)^{1/2}} e^{-im\chi} \Omega(\chi, t) \), such that \( \rho^{(n)}(t) = G(n, t - t_0) \rho(t_0) \), for \( t > t_0 \), fulfills the property: \( G(n - n', t - t_0) \equiv \sum_{i} G(n - n', t - t')G(n' - n, t' - t_0) \), for \( t > t' > t_0 \), \( (n' \equiv n(t') , n_0 \equiv n(t_0)) \). This is an operator version of the Chapman-Kolmogorov equation [19].

The joint probability of obtaining \( n_1 \) electrons after \( t_1 \) and \( n_2 \) electrons after \( t_2 \), namely \( P^{\geq}(n_1, t_1; n_2, t_2) \) (the superscript ‘\( \geq \)’ implies \( t_2 > t_1 \)), can be written in terms of these propagators by evolving the local probabilities \( P(n, t) = \text{Tr}\{\rho^{(n)}(t)\} \) [20], and taking into account \( P(n_2, t_2) = \sum_{n_1} \rho^{\geq}(n_1, t_1; n_2, t_2) \), such that

\[
P^{\geq}(n_1, t_1; n_2, t_2) = \text{Tr}\{G(n_2 - n_1, t_2 - t_1) \\
\times \text{Tr}\{\rho^{(n_1)}(t_1)\} \text{Tr}\{\rho^{(n_2)}(n_1, t_1)\} \}.
\]

The total joint probability reads \( P(n_1, t_1; n_2, t_2) = \mathcal{T}P^{\geq}(n_1, t_1; n_2, t_2) = P^{\geq}(n_1, t_1; n_2, t_2) \theta(t_2 - t_1) + P^{\leq}(n_1, t_1; n_2, t_2) \theta(t_1 - t_2) \) where \( \mathcal{T} \) is the time-ordering operator, \( \theta(t) \) the unit step function defined as \( \theta(t) = 1 \) for \( t \geq 0 \), zero otherwise, and where \( P^{\leq}(n_1, t_1; n_2, t_2) \) is the joint probability with \( t_2 < t_1 \). It should be noted that, in contrast to the local probability \( P(n, t) \), the joint probability \( P(n_1, t_1; n_2, t_2) \) contains information about the correlations at different times.

Result [21] may be alternatively derived using the Bayes formula for the conditional density operator [21]:

\[
P^{\geq}(n_1, t_1; n_2, t_2) = P(n_1, t_1)P^{\geq}(n_2, t_2|n_1, t_1) \\
\quad = \text{Tr}\{\rho^{(n_1)}(t_1)\} \text{Tr}\{\rho^{(n_2)}(n_1, t_1)\} \\
\quad = \text{Tr}\{\rho^{(n_1)}(t_1)\} \text{Tr}\{\rho^{(n_2)}(n_1, t_1)\} \}.
\]

The normalization in the denominator accounts for the collapse \( n = n_1 \) at \( t = t_1 \) using Von Neumann’s projection postulate [21]. Equation [21] is recovered when \( \rho^{(n)}(t_1) \) is written as a time evolution from \( t_0 \).

The two-time cumulant generating function (CGF) associated with these joint probabilities is

\[
e^{-\mathcal{F}(x_1, x_2; t_1, t_2)} = \sum_{n_1, n_2} P(n_1, t_1; n_2, t_2)e^{in_1x_1 + in_2x_2},
\]

which, using Eq. [22], and \( e^{-\mathcal{F}} = e^{-\mathcal{F}^{\geq}} = \mathcal{T}e^{-\mathcal{F}} \), gives

\[
e^{-\mathcal{F}(x_2; x_1; t_2; t_1)} = \mathcal{T} \text{Tr}\{\Omega(x_2, t_2 - t_1) \\
\times \Omega(x_1, t_2 - t_1 - t_0) \rho(t_0)\}.
\]

The above procedure can be easily generalized to obtain the N-time CGF, which reads:

\[
e^{-\mathcal{F}(x; t)} = \mathcal{T} \text{Tr}\{\prod_{k=1}^{N} \Omega(\sigma_k; \tau_{N-k}) \rho(t_0)\},
\]

where \( \sigma_k \equiv \sum_{i=N-k+1}^{N} \chi_i \), \( x \equiv (\chi_1, \ldots, \chi_N) \), \( t \equiv (t_1, \ldots, t_N) \) and \( \tau_k \equiv t_{k+1} - t_k \) [22]. The multi-time CGF in Eq. [23] contains a product of local-time propagators, and expresses the Markovian character of the problem. It allows one to obtain all the frequency-dependent cumulants from the spectral decomposition of \( M(\chi) \). The N-time current-cumulant \( (\epsilon = 1) \) is calculated using:

\[
S^{(N)}(t_1, \ldots, t_N) \equiv \langle \delta I(t_1) \cdots \delta I(t_N) \rangle = \\
\partial_{\tau_1} \cdots \partial_{\tau_N} \langle \langle n(t_1) \cdots n(t_N) \rangle \rangle = \\
= -(i)^{N-1} \partial_{\tau_1} \cdots \partial_{\tau_N} \partial_{\delta_n} \mathcal{F}(x; t) |_{x=0}.
\]

The Fourier transform of \( S^{(N)} \) with respect to the time intervals \( \tau_k \), gives the Nth-order correlation functions as functions of \( N - 1 \) frequencies. In particular, the frequency-dependent skewness is a function of two frequencies which, as a consequence of time-symmetrization and the Markovian approximation, has the symmetries \( S^{(3)}(\omega, \omega') = S^{(3)}(\omega', \omega) = S^{(3)}(\omega, \omega - \omega') = S^{(3)}(\omega' - \omega, \omega') \), and is therefore real. The Nth-order Fano-factor is defined as \( F^{(N)} \equiv S^{(N)} / \langle I \rangle \).

In the case where the jump matrix \( \mathcal{L}_J \) contains a single element, \( (\mathcal{L}_J)_{ij} = \Gamma \delta_{ij} \delta_{\epsilon j} \), which is the case for a wide class of models including our two examples below, all the correlation functions can be expressed solely in terms of the eigenvalues \( \lambda_k \) of \( \langle I \rangle \), and the \( N_\epsilon \) coefficients \( c_k \equiv \langle V_\epsilon^{-1} \mathcal{L}_J V_\epsilon \rangle_{kk} = \Gamma_R V_{\beta k} V_{\epsilon k}^{-1} \). The second-order Fano factor then has the simple, general form

\[
F^{(2)}(\omega) = 1 - 2 \sum_{k=2}^{N_\epsilon} \frac{c_k \lambda_k}{\omega^2 + \lambda_k^2},
\]

which has also been derived in other ways [23]. The skewness has the form \( F^{(3)}(\omega, \omega') = -2 + \sum_{i=1}^{N_\epsilon} F^{(2)}(\nu_i) + F^{(3)}(\omega, \omega') \), with \( \nu_1 = \omega, \nu_2 = \omega' \), and \( \nu_3 = \omega - \omega' \). \( F^{(3)}(\omega, \omega') \) is an irreducible contribution, the form of which is too cumbersome to be given here. The high-frequency limit of the skewness is \( F^{(3)}(\omega, \infty) = F^{(2)}(\omega) \).

As a first example we consider a SRL, described by \( \mathcal{P} = (\rho_{00}, \rho_{11})^{T} \) and \( M(\chi) = \begin{pmatrix} -\Gamma_L & e^{i\chi} \Gamma_R \\ \Gamma_L & -\Gamma_R \end{pmatrix} \), in the basis of ‘empty’ and ‘populated’ states, \( \{0\}, \{1\} \). Employing Eq. [25], we obtain the known results for the current and noise, and arrive at our result for the skewness:

\[
F^{(3)}(\omega, \omega') = 1 - 2 \Gamma_L \Gamma_R \Gamma_L \Gamma_R \prod_{i=1}^{3} (\Gamma^2 + v_i^2),
\]

with \( \gamma_1 = \Gamma_L^2 + \Gamma_R^2, \gamma_2 = 3 \Gamma^2, \) and \( \Gamma = \Gamma_L + \Gamma_R \). The
minimum at a finite frequency $\omega$ with respect to the Poissonian value of unity and to the noise is suppressed throughout frequency space both with respect to the many body ‘empty’ state $|0\rangle$, the Hamiltonian reads $\mathcal{H}_S = \epsilon (|L\rangle\langle L| - |R\rangle\langle R|) + T_c (|L\rangle\langle R| + |R\rangle\langle L|)$, with detuning $\epsilon$ and coupling strength $T_c$. The two levels $|L\rangle$, $|R\rangle$, are coupled to their respective leads with rates $\Gamma_L$ and $\Gamma_R$. The DM vector is now $\rho = (\rho_{00}, \rho_{LL}, \rho_{RR}, \text{Re}(\rho_{LR}), \text{Im}(\rho_{LR}))^T$, and the Liouvillian in this basis reads:

$$M(\chi) = \begin{pmatrix}
-\Gamma_L & 0 & e^{i\chi} & 0 & 0 \\
\Gamma_L & 0 & 0 & 0 & -2T_c \\
0 & 0 & -\Gamma_R & 0 & 2T_c \\
0 & 0 & 0 & -\frac{1}{2} & 2\epsilon \\
T_c & -T_c & -2\epsilon & -\frac{1}{2} \Gamma_R & 0
\end{pmatrix}.$$  

Comparison of the quantum-mechanical level-splitting $\Delta \equiv 2\sqrt{T_c^2 + \epsilon^2}$ with the incoherent rates $\Gamma_{L,R}$ divides the dynamical behaviour of the system into two regimes. For $\Delta \ll \Gamma_{L,R}$, all eigenvalues of $M(0)$ are real, and correspondingly, the noise and skewness are slowly-varying functions of their frequency arguments. In this regime, dephasing induced by the leads suppresses interdot coherence, and the transport is largely incoherent. In the opposite regime, $\Delta \gg \Gamma_{L,R}$, two of the eigenvalues form a complex pair, $\lambda_L \approx \pm i\Delta - \Gamma_R/2 + O(\Gamma/\Delta)^2$, which signals the persistence of coherent oscillations in the dots. The finite-frequency correlators then show resonant features at $\Delta$, since these eigenvalues enter into the denominators, as in Eq. (7), giving rise to poles such as $\omega = \Delta - i\Gamma_R/2$. The structure of the skewness is similar to the SRL for weak coupling ($\Delta \ll \Gamma_{L,R}$), but much richer in the strong coupling regime ($\Delta \gg \Gamma_{L,R}$) (Fig. 2). Now the skewness exhibits a series of rapid increases. From the origin outwards in the $\omega$-$\omega'$ plane, we observe first a minimum at finite frequency and then inflexion points at $\omega \sim \Gamma_R$, $|\omega'| = \Delta$, $|\omega''| = \Delta$ and $|\omega - \omega'| = \Delta$. Fig. 2 shows sections in the $\omega$-$\omega'$ plane, and the resonant behaviour, in the form of Fano shapes, is most pronounced in $F(\omega, -\omega)$. Starting from high frequencies, the onset of antibunching occurs at $\omega = \Delta$. At higher frequencies, the system has no information about correlations and is Poissonian. The overall behaviour is seen in Fig. 2(c) where we plot $F(\omega, -\omega)$ as a function of $T_c$ and $\omega$. The line $\omega = \Delta$ delimits two regions: at high frequencies the skewness is Poissonian. At resonance, and after a small super-Poissonian region at $\omega \gtrsim \Delta$, the system becomes sub-Poissonian (and even negative, for certain internal couplings). In the limit $\omega \to 0$, our results qualitatively agree with those of Ref. [27] for a noninteracting DQD: as a function of $T_c$, the skewness presents two minima and a maximum (where the noise is minimum, not shown). In our case, however, the maximum occurs around $\Delta = \Gamma_R/2$ — half that of the noninteracting case. Finally, we plot $dF(\omega, -\omega)/d\omega$ as a function of both $\epsilon$ and $\omega$ (Fig. 2(b)) where the resonances at $\omega = \Delta$, $\omega = \Delta/2$ and $\omega \sim \Gamma_R$ are clearly resolved. In contrast, the deriv-
the skewness shows rapid increases along the lines $\omega = \Delta$, $\Delta = 0$. The skewness is strongly suppressed for small $\omega$, always sub-Poissonian. The skewness is strongly suppressed for $\omega > \Delta$ and $\omega = \Delta/2$. For $\omega > \Delta$ the system is Poissonian (slightly super-Poissonian for $\omega \gtrsim \Delta$), while for $\omega < \Delta$ the transport is always sub-Poissonian. The skewness is strongly suppressed at low frequencies. (d) The derivative $dF^{(3)}(\omega, -\omega)/d\omega$ as a function of frequency and detuning $\epsilon$ for $T_c = 3\Gamma_L = 3\Gamma_R$. Resonances occur at $\omega = \Delta$, $\Delta/2$ and $\sim \Gamma_R$.

Deviation from Poissonian behaviour of higher-order cumulants are frequency-dependent, such that a comprehensive analysis in the frequency domain is needed in order to fully characterize correlations and statistics in electronic transport.

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