Counting statistics in interacting nano-scale conductors

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Abstract
Counting statistics investigates the probability $P(n,t)$ that a number $n$ of electrons traverse a nano-scale conductor during a time span $t$. It is equivalent to consider the zero frequency charge or current correlators, the so-called moments and cumulants, in principle up to infinite order. In this thesis we investigate several aspects of electronic correlations due to interactions. First we investigate the influence of interactions on the counting statistics, considering a generic two-terminal conductor. We show that if the factorial cumulants oscillate as functions of any system parameter or time, then the electrons must be interacting. This statement may be verified in Coulomb blockaded quantum dots, where it is possible to monitor the traversal of electrons in real-time. Moreover, we use a Markovian master equation to describe the first experiment on counting statistics of Andreev events, where two electrons tunnel across a tunnel barrier between a superconducting lead and a normal metallic island. The statistics are strongly super-Poissonian, reflecting that Andreev events occur in avalanches of different sizes. [...]
Counting statistics in interacting nano-scale conductors

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par

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N.B.- La thèse doit porter la déclaration précédente et remplir les conditions énumérées dans les "Informations relatives aux thèses de doctorat à l'Université de Genève".
Abstract

Electronic transport in nano-scale conductors is a very active field of research. When the size of the conductors gets so small and the temperature gets so low that the wave nature of the electrons becomes important, then their physical description must be quantum mechanical. This means that electronic transport in nano-scale conductors is intrinsically probabilistic, as opposed to deterministic, and predictions can only be made on the level of probabilities or for average quantities, as for example the mean current, the noise, but also higher order correlators. What is more, such small conductors may have a very large resistance, on the order of or larger than the inverse conductance quantum $1/(e^2/h) \simeq 26k\Omega$, where $e$ is the elementary charge and $h$ is Planck’s constant. Therefore, the measured currents are typically small and the current noise may take comparable magnitude, resulting in relatively small signal to noise ratios. Large resistances are for example obtained by tunnel barriers formed by a thin insulating layer. In quantum mechanics there is a small but finite probability, decreasing exponentially with the barrier thickness, for electrons to traverse the barrier. This effect is called quantum tunneling. Combining large resistances in series with a tiny capacitance, single-electron charging effects occur, a phenomenon known as Coulomb blockade. The small capacitance reflects the strong Coulomb interactions among electrons on the conductor. In this quantum world where single-electron effects are important, the effect of electrons “seeing” each other, or in other words, correlations among electrons, becomes important. This is in contrast to the macroscopic world, where nearly always quantum correlations are averaged out. Correlations are introduced on the one hand by the Pauli exclusion principle, which states that no two electrons may occupy the same quantum state. This is the case even if we neglect any interactions. On the other hand, any type of effective electron-electron interaction introduces correlations.

Counting statistics investigates the probability $P(n, t)$ that a number $n$ of electrons traverse a nano-scale conductor during a time span $t$. It is equivalent to consider the zero frequency charge or current correlators, the so-called moments
and cumulants, in principle up to infinite order.

In this thesis we investigate several aspects of electronic correlations due to interactions. First we investigate the influence of interactions on the counting statistics, considering a generic two-terminal conductor. We show that if the factorial cumulants oscillate as functions of any system parameter or time, then the electrons must be interacting. This statement may be verified in Coulomb blockaded quantum dots, where it is possible to monitor the traversal of electrons in real-time.

Moreover, we use a Markovian master equation to describe the first experiment on counting statistics of Andreev events, where two electrons tunnel across a tunnel barrier between a superconducting lead and a normal metallic island. The statistics are strongly super-Poissonian, reflecting that Andreev events occur in avalanches of different sizes.

Finally, we consider finite frequency current noise and show that the noise spectra are in general asymmetric in the applied bias voltage. Using a higher order fluctuation relation, which is an extension of the fluctuation dissipation relation to the non-equilibrium transport regime, we show that this asymmetry is due to a broken electron-hole symmetry, resulting in a finite rectification. We point out that this can occur either due to an asymmetrically applied bias, but more importantly, due to interactions and an inherent chirality of the conductor.
**Résumé**

Le transport électronique dans des conducteurs à l’échelle nanométrique est un domaine de recherche très actuel. Lorsque les conducteurs ont une taille si petite et la température devient si basse que la nature ondulatoire des électrons se révèle, alors leur description physique nécessite d’invoquer la mécanique quantique. Ceci implique que le transport électronique dans des conducteurs à l’échelle nanométrique est intrinsèquement probabiliste, et donc par essence non-déterministe. Des prédictions peuvent uniquement être faites au niveau des probabilités ou pour des quantités moyennées, comme par exemple le courant moyen, le bruit, mais aussi des corrélateurs d’ordre plus élevés. De plus, des conducteurs aussi minuscules peuvent avoir des résistances enormous, de l’ordre de ou plus grandes que l’inverse du quantum de conductance, $1/(e^2/h) \approx 26k\Omega$, où $e$ est la charge élémentaire et $h$ est la constante de Planck. De ce fait, les courants mesurés sont typiquement petits et le bruit peut avoir une amplitude comparable, ce qui engendre des rapports signal sur bruit relativement petits. Une jonction tunnel, formée d’une mince couche isolante entre deux conducteurs, donne par exemple lieu à une grande résistance. En mécanique quantique la probabilité que les électrons traversent la barrière de potentiel formée par la couche isolante est non nulle et décroît exponentiellement avec l’épaisseur de la barrière. Ceci s’appelle l’effet tunnel. En combinant de grandes résistances avec une minuscule capacité, un seul électron peut bloquer le passage d’autres électrons : un phénomène appelé blocage de Coulomb. La minuscule capacité représente la répulsion coulombienne importante entre électrons dans le conducteur. Dans ce monde quantique, les corrélations entre électrons (c’est-à-dire le fait que les électrons “se voient”) jouent un rôle essentiel. Ceci contraste avec le monde macroscopique, dans lequel les corrélations quantiques sont presque toujours moyennées à zéro et donc disparaissent. D’un côté, des corrélations sont introduites par le principe d’exclusion de Pauli exigeant qu’un état électronique puisse accommoder un seul électron ; ceci même si toute interaction est négligée. De l’autre, les interactions entre électrons introduisent aussi des corrélations.
**Counting statistics**, c’est-à-dire la statistique de comptage, analyse la probabilité $P(n, t)$ qu’un nombre $n$ d’électrons traversent un conducteur nanométrique pendant un temps $t$. Ceci est équivalent à considérer les corrélateurs de charge ou de courant à fréquence nulle, c’est-à-dire les moments et cumulants, en principe jusqu’à des ordres infinis.

Dans cette thèse nous étudions plusieurs aspects de corrélations électroniques engendrées par des interactions. Dans un premier temps nous étudions l’influence des interactions sur la statistique de comptage, en considérant un conducteur général connecté à deux électrodes. Nous montrons que si les *cumulants factoriels* oscillent en fonction de n’importe quel paramètre du système ou du temps, alors il doit y avoir des interactions entre les électrons dans le conducteur. Cette assertion pourrait être vérifiée dans des points quantiques sous blocage de Coulomb, où il est possible d’observer le passage d’électrons en temps réel.

Ensuite nous utilisons une équation maîtresse Markovienne pour décrire la première expérience de statistique de comptage d’événements Andreev, où *deux* électrons traversent une jonction tunnel entre une électrode supraconductrice et un îlot de métal normal. La variance de la distribution est fortement supra-poissonienne, ce qui signale que les événements Andreev surviennent en *avalanches* de différentes tailles.

Enfin, nous considérons le bruit électronique à fréquence finie et nous montrons qu’en général les spectres de bruit sont asymétriques en fonction de la tension aux bornes. En utilisant une relation de fluctuation de second ordre, qui fait partie de la généralisation du théorème fluctuation-dissipation en régime hors-équilibre, nous montrons que cette asymétrie résulte d’une brisure de symétrie électron-trou, ce qui entraîne une rectification finie. Nous mettons en lumière qu’une rectification finie peut se produire, soit en appliquant une tension asymétriquement, soit, de manière plus importante, par des interactions et une chiralité intrinsèque du conducteur.
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When one is well-balanced and happy, it is just a pleasure to work! On the other hand, when one is sad, exhausted and frightened, work can become a pain. I do not deny that there have been times of trouble during my PhD. I would like to thank my best friends for their love and kindness and for their support, without which I would not stand where I am now. I send my warmest thanks to Roland and Béatrice Marthaler, Laurent Marthaler, Camille and Pierre-Olivier Dion-Labrie, the Gerber family, Carole Durussel, Tatjana Hählen and Christian Tarnutzer, Hayley Finley and Emeric Bron, Dankrad Feist, Thibaud Hotzelier, Philipp Dahinden, Marlene Glauser, Rachel Lahyani, Nicole Lironi, Maher Younan and his family, Christian Kauth, Sebastian Gfeller, Vreni Stauffer, Anne Ramoni, Lucie Christinger and Jacqueline Charvin, and the Brauer family.

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Dania Kambly, Trubschachen, February 2014.
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List of publications

The present thesis consists of an introduction to and an overview of the following publications:


Curriculum Vitae

2003  **Matura**, Gymnasium Burgdorf, Burgdorf, Switzerland.

2004 – 2007 **Bachelor in Physics (BSc)**, École Polytechnique Fédérale de Lausanne (EPFL), Lausanne, Switzerland.

2006 – 2007 **Exchange year** in Physics, Carnegie Mellon University (CMU), Pittsburgh, PA, USA.

2007 **Internship** at ETHZ Zürich with Prof. M. Troyer and at PSI Villigen with Prof. J. Mesot.


2009 **Internship** at MNT–LANS, heig–vd, Yverdon-les-Bains, Switzerland, with Prof. S. Schintke.

2009 – 2014 **PhD** in Theoretical Mesoscopic Physics with Prof. Markus Büttiker, Dr. Christian Flindt and Prof. Eugene V. Sukhorukov at Université de Genève, Geneva, Switzerland.

Schools and conferences attended during my PhD:

- **★** The 5th Windsor Summer School on Quantum Phenomena in Low-Dimensional Materials and Nanostructures, Windsor, UK (poster and two-minute talk).

- **★** DPG Spring Meeting, TU Dresden, Germany, 13th – 18th March 2011 (talk).

- **★** SPG Annual Meeting, EPFL Lausanne, Switzerland, 17th June 2011 (talk).

- **★** NCCR/QSIT Junior–Meeting, Passugg, Switzerland, 21st – 24th June 2011 (talk).

- **★** NCCR meeting on Quantum Systems and Technology, Monte Verità, Ascona, Switzerland, 17th – 22nd June 2012 (poster).

- **★** International Conference on Nanoscience + Technology (ICN+T), Paris, France, 23rd – 27th July 2012 (talk).
Chapter 1

Introduction

1.1 Electron transport in mesoscopic conductors: 
a brief introduction

Nowadays electronic transport in small semiconductor and metallic devices of sizes
down to the nanometer scale, printed circuits or optoelectronic devices are of
tremendous daily importance. On the other hand, research pushes the frontiers
of the feasible more and more forward. The field of Mesoscopic Physics comprises
the basic research on electronic transport properties of mesoscopic devices [1, 2].
The word mesoscopic means that we consider conductors of sizes that are between
the microscopic, i.e. the atomic or molecular level, and the macroscopic world. We
consider true devices, however they are so small that their behavior is quantum
mechanical, that is, the wave nature of the electron plays an important role. This
typically also requires very low temperatures. In terms of physical length scales
this means

\[ l_e \ll L \ll l_\phi \leq l_{in}, \]

where \( l_e \) is the elastic scattering length, which is the mean distance an electron
travels before undergoing an elastic collision. Moreover, \( L \) is the size of the conduc-
tor, which in the case of mesoscopics is much smaller than the dephasing length
\( l_\phi \), the length scale over which an electron keeps its coherence. More accurately, \( l_\phi \)
is the mean length after which the phase of the electronic wave function suffers a
change of \( 2\pi \). It means that an electron can interfere with itself (!) within distances
smaller than \( l_\phi \), for example if there are two possible paths in an interferometer. Fi-
nally, \( l_{in} \) is the inelastic scattering length, the mean length after which an electron

\footnote{In the experiment of Ch. 3 for example the bath temperature was 50mK.}
loses its excess energy and equilibrates. Typical sizes $L$ of mesoscopic conductors range from several µm down to the nanometer scale. As opposed to (1.1), for a large, macroscopic conductor we would have $l_e \ll l \phi \leq l_{in} \ll L$.

When the wave nature of carriers is important, the physical description of the transport is quantum mechanical, that is, inherently statistical and not deterministic. What is more, transport is a non-equilibrium phenomenon. To simplify matters, one can consider a constant bias voltage $V$, such that the system may reach a steady state. Moreover, one can consider linear response, that is, say, the current $I$ resulting from the applied bias $V$ to linear order in $V$. On the other hand, for time dependent driving a periodic bias voltage can be applied and the analyzed quantities can be averaged over a large number of periods.

Semiconductor or metallic devices can be designed such that the electrons are constrained to move in less than 3D. In a two-dimensional electron gas (2DEG) electrons move in a plane. The energy for motion perpendicular to the plane is quantized, and typically only the ground state is occupied, whereas the motion in the plane is hardly constrained. 2DEGs can be realized for example at the interface joining two semiconductors with different band gaps and Fermi energies, as for example in AlGaAs/GaAs structures. Another example are Si MOSFETs, where a large positive voltage on a metallic gate on top of a p-doped Si layer covered with SiOx induces a 2DEG under the gate. Requirement in both cases is that a narrow minimum is formed at the interface in the band bending (or band edge) diagram. Another example of two-dimensional electron motion are graphene sheets, where the free electrons are massless Dirac fermions.

When a perpendicular magnetic field is applied to a 2DEG, the allowed electron energies become discrete. They are called Landau levels. At the edges of the 2DEG they are bent upwards, which can be understood from the fact that the electrons are confined to the sample. Consequentially, edge states form along the edges of the 2DEG at the places where the Fermi energy crosses the Landau levels. Electrons travel ballistically and without backscattering along the edge for values of the magnetic field such that the Fermi energy lies between two Landau levels, whereas the bulk becomes insulating. When the magnetic field takes values such that the Fermi energy lies within a (broadened) Landau level, the longitudinal

\[ 2 \text{If equilibration is achieved via the exchange of energy with surrounding electrons, then } l_{in} \text{ is the mean length after which an electron loses an energy of } k_B T \text{ with } k_B \text{ the Boltzmann constant and } T \text{ the electron temperature.} \]

\[ 3 \text{The Fermi energy is the energy of the topmost occupied state at zero temperature. At low temperatures transport typically takes place at energies close to the Fermi energy.} \]
resistance rises, a phenomenon known as Schubnikov-de Haas oscillations. The resistance transverse to the current flow exhibits plateaus, with the transverse conductance taking the quantized values $G_H = G_Q \nu$, where $G_Q = e^2/h$ is the conductance quantum, with $e$ the elementary charge and $h$ the Planck constant, and where $\nu$ is the number of filled edge channels, the so-called filling factor. This quantization of the transverse conductance at low temperatures and high magnetic fields is called the quantum Hall effect (QHE). The electron motion along an edge state is free and one-dimensional.

The shape of a 2DEG can be engineered by adding gate electrodes. With this and in many cases using the quantum Hall effect, a great variety of systems have been designed and studied. The most prominent example is the quantum point contact (QPC). Two negatively charged gate electrodes (split-gate) on top of the heterostructure deplete the 2DEG underneath and create a narrow constriction with a size on the order of the Fermi wavelength. This constriction is an electron wave guide, quantizing the transverse motion. Transverse conduction channels arise resulting in the quantization of conductance, which is described by the Landauer formula with unit transmission for each of the $N$ channels, that is, $G = 2G_Q N$. The factor of two arises because of the spin degeneracy. As the width of the QPC is made larger, it accommodates more transverse channels. This yields a step-like behavior of the conductance as a function of gate voltage, with steps of height $2G_Q$. When a large perpendicular magnetic field is applied to the QPC only a few ($\nu$) edge states traverse the constriction, yielding a conductance of $G = G_Q \nu$ since the spin degeneracy is lifted in the presence of a magnetic field.

Another prominent example encountered later in this thesis is the quantum dot (QD). In a QD electronic motion is constrained in all three directions of space, so that the electrons are confined and their energy takes on discrete values. There are numerous ways to fabricate quantum dots with different potential applications. When electrodes are added on top of a heterostructure with a 2DEG at the interface, and a negative voltage is applied to deplete the 2DEG underneath, a potential minimum can be created which forms the QD. Quantum dots formed in this way can be designed, and connected to other structures as well as to electronic leads. If the dot is connected directly to electronic leads, we say it is open. However, if it is weakly coupled via tunnel barriers with resistances greater than $\sim 1/G_Q$.

---

4. See e.g. in 3.
5. i.e. the inverse of the resistance
6. with the mass corresponding to the effective mass.
7. The Fermi wavelength, $\lambda_F$, is 40nm in GaAs(100) and 112-135nm in Si(100) 3.
the dot can be operated in the Coulomb blockade regime \cite{14}. When an electron is
to be added to the dot, it will interact with other charges on the dot via Coulomb
interaction. Typically, the dot capacitance $C$ is very small, so that the charging
energy to be paid to add one more electron to the dot, $\Delta E = e^2/C$, is much larger
than the applied bias and the temperature. Thus, one extra electron on the dot
will block the tunneling.

The Coulomb blockade is also the working principle of the single electron tran-
sistor (SET) \cite{17}. Typically, a small metallic island of small capacitance $C$ is
connected via tunnel barriers of resistance $R_t$ to the source and drain leads. A gate
voltage $V_g$ polarizes the island such that the energetically most favorable island
charge is the integer closest to the continuous gate charge $N_g = C_g V_g/e$, with $C_g$
the gate capacitance. The energy to promote the island to the $N^{th}$ charge state
is given by Eq. (3.1), where the charging energy $E_C = e^2/2C$ is the energy to be
paid to add or take out one electron, given that the island is in its ground state.
The difference to the QD systems described above is that the density of states on
the metallic island is continuous. The condition that a SET may operate, i.e. that
single-electron charging effects are observed, are the same as we mentioned for the
QD, namely \cite{14}

\begin{equation}
R_t \gg \frac{\hbar}{e^2} = \frac{1}{G_Q},
\end{equation}

\begin{equation}
\frac{e^2}{2C} \gg k_B T,
\end{equation}

where $k_B$ is the Boltzmann constant and $T$ the temperature. An electron micro-
graph of a SET connected to superconducting leads is shown at the top of Fig. 3.1.
It represents the charge detector, since due to the capacitive coupling its current $I_d$
is very sensitive to the charge state of the adjacent large metallic island.

Electrons in normal metallic leads connecting the different mesoscopic conduc-
tors may in most cases be modeled as noninteracting electrons moving in 1D.
Moreover, the leads are treated as semi-infinite since they lead into large electron
reservoirs. The reservoirs are typically considered to be at equilibrium. Because of
the small width of the leads, the lateral confinement leads to the quantization of the
transverse part of the wave function, resulting in transverse conduction channels.
The longitudinal momentum $k$ of electrons at the Fermi edge is given by

\begin{equation}
E_F = E_n + \frac{\hbar^2 k^2}{2m},
\end{equation}
where $E_F$ is the Fermi energy, $m$ is the effective electron mass and $E_n$ is the transverse energy of the $n^{th}$ conduction channel. For definiteness, the transverse spectrum $E_n$ can be computed using infinite hard-walls [18] or a parabolic potential [19] for the wire.

Considering the mesoscopic conductor itself and not its connectors to the macroscopic electronics, there are for example quantum wires or carbon nanotubes, which are effectively 1D.

Let us consider a mesoscopic conductor connected to several electronic leads. A generic two-lead setup is sketched in Fig. 1.1. If the electrons in the whole setup, that is, not only in the leads but also in the conductor, can be treated as noninteracting, then the Landauer-Büttiker scattering matrix formalism may be applied [20, 21, 22, 23, 24]. In this formalism the only source of correlations are the fermionic statistics, or in other words, the Pauli principle, which forbids that two fermions occupy the same state. The scattering matrix is specific for a given conductor and links the complete set of outgoing single-particle scattering states to the complete set of ingoing single-particle scattering states [23, 24]. The Landauer-Büttiker formula [25, 26], Eq. (1.5), then expresses the mean current in lead $\alpha$. This is done in terms of the reflection probability $R_{\alpha\alpha}$ in lead $\alpha$ and the total transmission probabilities $T_{\alpha\beta}$ of electrons from lead $\beta$ into lead $\alpha$, quantities which are readily obtained from the scattering matrix [22, 23].

$$\langle I_{\alpha} \rangle = \frac{e}{\hbar} \int dE \left[ (M_{\alpha} - R_{\alpha\alpha}) f_{\alpha} - \sum_{\beta} T_{\alpha\beta} f_{\beta} \right]$$

$$\simeq \frac{e}{\hbar} \int dE \left( -\frac{\partial f}{\partial E} \right) \left[ (M_{\alpha} - R_{\alpha\alpha}) \mu_{\alpha} - \sum_{\beta} T_{\alpha\beta} \mu_{\beta} \right]. \quad (1.5)$$

---

$^8$Current conservation requires that the scattering matrix be unitary and if the problem is time-reversal symmetric the scattering matrix must also be symmetric [24].
In Eq. (1.5) above, \(e\) is the elementary charge, \(h\) is Planck’s constant and \(M_\alpha\) is the number of transverse channels in lead \(\alpha\). What is more, \(f_\alpha\) is the Fermi function in reservoir \(\alpha\), evaluated at \(\mu_\alpha\), whereas \(f\) is the Fermi function at the equilibrium chemical potential \(\mu\). The second line in (1.5) expresses the current to linear order in the applied bias \(eV_\alpha\).

The scattering matrix formalism allows us to analyze and describe not only the transport, but also the fluctuations and higher order zero-frequency current correlators, namely the full counting statistics (FCS).

When interactions between electron quasiparticles become important, they can be treated on the mean-field level within scattering theory, or dephasing can be taken into account by adding voltage probes. However, for stronger interactions we need to use other formalisms. One can use circuit theory using Green’s functions. Another example are Markovian master equations that we will be using in Chs. 2 and 3 to describe transport through Coulomb blockaded conductors. The advantage of master equations is that the interactions are treated non-perturbatively. The price to pay is that we typically do perturbation theory in the tunneling Hamiltonian that links the conductor to the leads. The relevant tunneling rates are determined using the Fermi golden rule, which corresponds to the first order in the tunneling Hamiltonian.

Interactions introduce correlations among electrons traversing a mesoscopic conductor. Since charge and current correlators can be measured, it is natural to ask in what way they may be used to characterize a mesoscopic conductor and its inherent physical processes. The most commonly experimentally accessible are the mean current and the noise, corresponding to the first and second order correlators.

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9 The chemical potential in lead \(\alpha\) reads \(\mu_\alpha = \mu + eV_\alpha\) and due to charge conservation, i.e. due to the unitarity of the scattering matrix, we have \(R_{\alpha\alpha} + \sum_\beta T_{\alpha\beta} = M_\alpha\).

10 The finite frequency current correlators in a noninteracting theory do not capture current conservation. In fact, potential fluctuations in the reservoirs, that is, electron-hole excitations of different frequencies, generate momentary charge pileup in the mesoscopic conductor. The finite frequency noise captures this effect, whereas the zero frequency (dc) limit describes fluctuations averaged over long times. The accumulated charges on the conductor create a local screening potential redistributing charges in the capacitively coupled close neighborhood, generating displacement currents. This response can clearly only be described by a formalism that includes Coulomb interactions. In order to express current conservation, also these latter currents must be taken into account. In scattering theory there is a way to introduce Coulomb interactions self-consistently order by order so as to restore current conservation. Typically, the density of states on the conductor is calculated in the noninteracting framework and then capacitive coupling to a nearby gate is considered.
In this thesis . . .

. . . we propose to go beyond the characterization of mesoscopic conductors by their mean current and noise. We consider the zero-frequency charge or current correlators, i.e. the moments and cumulants, in principle up to infinite orders. This gives credit to the quantum mechanical nature of transport in mesoscopic structures: as indicated in Fig. 1.1 electrons are not transferred deterministically, however there is a probability \( P(n, t) \) that \( n \) electrons traverse the conductor during the time \( t \). This probability distribution \( P(n, t) \) contains the full counting statistics, as we shall develop further in Sec. 1.2.2. The present chapter introduces the basic concepts and the results needed to elaborate the later chapters. In Ch. 2 we show that signatures of interactions may be seen in the factorial cumulants. The result applies to any generic two-terminal conductor, as sketched in Fig. 1.1. Several examples of Coulomb blockaded QDs are considered. Ch. 3 is a collaboration with an experimental group, where we have measured and calculated the full counting statistics of Andreev events. In this process, electrons tunnel in pairs through a tunnel barrier between superconducting leads and a normal metallic island. To our knowledge, this is the first experiment where the counting statistics of Andreev events have been measured, and we obtain an excellent fit to our model employing the Markovian master equation formalism. In Ch. 4 we exemplify that the noise spectra are in general asymmetric in the applied bias voltage. For this purpose we analyze a toy model of a quantum wire.

1.2 Counting statistics

1.2.1 Analyzing the noise

It is not the mean current alone that characterizes a mesoscopic conductor. Even though in many cases the measured signal corresponds to a current or voltage and the noise needs to be reduced to a minimum, the noise itself contains valuable additional information about the conductor and the nature of the charge transport. This has been pointed out in the seminal paper “The noise is the signal” by R. Landauer [39]. The fermionic nature of the electrons, that is, the Pauli principle, typically reduces the noise. Qualitatively, in a train of electrons the different wave packets tend to be arranged with minimal overlap, which regularizes the train and thus reduces the noise [22, 40, 41, 42, 43]. This effect cannot be seen in the mean current. Also, for example Coulomb interactions affect the noise
What is more, current-current (cross) correlators can be used to probe the quantum nature of electrons in electronic interferometers. In such interferometers the mean current typically plays no direct role. Using quantum Hall edge states as the analogue of optical fiber and QPCs as beam splitters, one can perform the electronic analogue of quantum optics. For example the Hanbury-Brown-Twiss \cite{44, 45, 46, 47} or Hong-Ou-Mandel type geometries \cite{48, 49, 50}, as well as the Mach-Zehnder interferometer \cite{51, 52, 53, 54} have been investigated.

There are two distinct contributions to the noise (see e.g. \cite{40}). First, the occupation numbers of the reservoir states fluctuate due to temperature, which generates fluctuations in the transport called thermal noise. As $T \to 0$ the thermal noise vanishes. Second, the discrete nature of the charge carriers produces what Schottky \cite{55} called shot noise and what is also referred to as partition noise. Partition noise occurs because at a potential barrier quantum particles have a certain probability to tunnel or else to be reflected, however the particle does this as a discrete entity. Partition noise is present even at zero temperature.

Schottky studied current fluctuations in vacuum tubes and found that for small frequencies the noise is proportional to the mean current \cite{55}, that is,

$$S = 2e \langle I \rangle . \quad (1.6)$$

Eq. (1.6) corresponds to the shot noise produced by uncorrelated events where the time intervals $\Delta t$ between detection times of electrons is distributed according to a Poissonian as $p(\Delta t) = (1/\tau) e^{-\Delta t/\tau}$, with $\tau = \langle \Delta t \rangle$ \cite{40}. In eq. (1.6) current and noise are equivalent. However, as soon as subsequent events are correlated through Fermi statistics or interactions among electrons this is no longer the case. A prominent example is noninteracting electrons at zero temperature. The shot noise is determined by the transmission eigenvalues $\{T_n\}$ of the conductor \cite{56, 22, 57, 40} and the noise power spectral density reads

$$S = 2 e^2 |eV| \sum_n T_n (1 - T_n). \quad (1.7)$$

Recalling that to first order in bias voltage $V$ the mean current reads $\langle I \rangle = \frac{e^2}{h} V \sum_n T_n$ and $0 \leq T_n \leq 1$, we see that the shot noise (1.7) is reduced with respect to the classical Poissonian value (1.6). This is due to the Fermi statistics inducing correlations among the electrons \cite{56, 22}.

The current noise as such corresponds to the zero-frequency current-current correlator. Thus, Eqs. (1.6) and (1.7) correlate currents from the same reservoir.
at zero frequency, $\omega = 0$. At finite frequency the noise for a stationary process is defined as \[ S_{\alpha \beta}(\omega) = \frac{1}{2} \int dt \, e^{i\omega t} \langle \Delta \hat{I}_\alpha(t) \Delta \hat{I}_\beta(0) + \Delta \hat{I}_\beta(0) \Delta \hat{I}_\alpha(t) \rangle, \] (1.8)

where $\hat{I}_\alpha$ is the current operator in reservoir $\alpha$ and $\Delta \hat{I}_\alpha = \hat{I}_\alpha - \langle \hat{I}_\alpha \rangle$. The diagonal elements in (1.8) are noise spectra, whereas the off-diagonal elements correspond to current-current cross-correlators between different reservoirs. In this thesis we concentrate on two-terminal conductors with a source (S) and a drain (D) electrode, that is, $\alpha, \beta \in \{S, D\}$.

At zero frequency, $\omega = 0$, the equilibrium noise spectra are related to the conductance matrix $G_{\alpha \beta} = \frac{d}{dV} \langle I_\alpha \rangle \big|_{V_\beta = 0}$ by the first fluctuation relation \[ S_{\alpha \beta}(0) = 4k_B T G_{\alpha \beta}, \] (1.9)

which corresponds to the Nyquist-Johnson noise. Eq. (1.9) shows that from equilibrium noise measurements we get the same information as from conductance measurements. However, in the presence of transport this ceases to be the case and we do get additional information from noise measurements. To this end, for stationary transport one can analyze the noise as an expansion in bias voltage:

\[ S = S^{(0)} + S^{(1)} V + \ldots \] (1.10)

Similarly, the charge current can be expanded in powers of the voltage

\[ I = GV + \frac{1}{2} G^{(2)} V^2 + \ldots \] (1.11)

where $G$ is the linear response conductance and $G^{(2)}$ is the second order conductance (rectification coefficient). The noise expansion coefficient of order $m - 1$ is related to the $m^{th}$ order conductance by a fluctuation relation \[58, 59, 60, 61, 62\]. The second order relation, that we shall be using in Ch. 4, reads

\[ S^{(1)} = 2k_B T \cdot G^{(2)}. \] (1.12)

$S^{(1)} V$ is the near-equilibrium excess noise to linear order in voltage. As the temperature is lowered, the region of validity of this expansion shrinks since at zero-

\[11\]This is one of the possible definitions and the one we shall adopt here. The expression is symmetrized.
temperature and at small voltages we have a shot noise that is linear in voltage with a slope that is singular at $V = 0$, Eq. (1.7).

1.2.2 From current and noise to higher order statistics: Full Counting Statistics

Parts of this section have been taken, in part verbatim, from publication (iii).

In the previous section we have shown that in many cases one can get additional information from noise measurements compared to what can be inferred from the mean current. Extrapolating this idea and following ideas from quantum optics [63, 64] it is the purpose of Full Counting Statistics to characterize a mesoscopic conductor via the full charge or current statistics [31, 40, 65], including an infinite number of moments or cumulants. The idea is that high order statistics provide additional information to what is available from the mean current and the noise.

Electron transport through a mesoscopic conductor is a quantum process and therefore its description is inherently statistical. Transport in a generic two-terminal setup as in Fig. 1.1 is described by a probability distribution $P(n, t)$ that $n$ electrons have traversed the conductor during the measuring time $t$. Since the transport is possibly bi-directional, $n$ can take positive or negative values. For concreteness we choose electron flow from the source to the drain as the positive direction of the current. The probability distribution $P(n, t)$ yields the full counting statistics. An equivalent and very convenient description uses the probability generating function (GF)

$$G(z, t) = \sum_n P(n, t) z^n. \quad (1.13)$$

The normalization condition for the probabilities, $\sum_n P(n, t) = 1$, implies for the GF that $G(z = 1, t) = 1$. The GF has several analogies to the partition function in Statistical Mechanics. In the long-time limit, as $t \to \infty$, the logarithm of the GF becomes linear in time [65, 66] just as the free energy (which is proportional to the logarithm of the partition function) typically becomes linear in the volume of the system in the thermodynamic limit. Moreover, the GF of a transport process consisting of several independent sub-processes factors into a product of the GFs corresponding to each of the sub-processes, in analogy to how the partition function in Statistical Mechanics may be written as a product of the partition functions.

\[\text{We suppose that we start counting at } t = 0.\]
for each independent sub-system. By factorizing the GF one can thus identify elementary transport processes. For noninteracting electrons elementary single-electron processes can be identified [67, 68, 69, 70] (also see Sec. 1.2.5). As an example of factorization for an interacting system let us mention Ref. [71], where the total transport through a multi-level Coulomb blockade quantum dot can be understood as a superposition of events with charge $-ne$, where $-e$ is the electron charge and $n = 1, 2, 3, \ldots$ (also see Secs. 2.3 and 3.3.3).

Another means of describing the statistics are moments and cumulants. Like the probability distribution $P(n, t)$ they are measurable quantities. The statistical moments are defined to be

$$\langle n^m \rangle(t) \equiv \sum_n n^m P(n, t) = \partial_z^m \mathcal{M}(z, t)|_{z \to 0},$$

(1.14)

where we have introduced the moment generating function (MGF)

$$\mathcal{M}(z, t) = G(e^z, t) = \sum_n P(n, t)e^{nz}.$$  

(1.15)

The MGF of a transport process composed of several independent processes factors into a product of the corresponding MGFs. However, the moments of the full process are not related to the moments of the individual sub-processes in a simple way. This motivates the definition of cumulants, also known as irreducible moments. The cumulant generating function (CGF) is defined as the logarithm of the MGF

$$S(z, t) = \log[\mathcal{M}(z, t)] = \log[G(e^z, t)],$$

(1.16)

which again delivers the cumulants of $n$ by differentiation with respect to $z$ at $z = 0$:

$$\langle \langle n^m \rangle \rangle(t) = \partial_z^m S(z, t)|_{z \to 0}.$$  

(1.17)

The first cumulant is the mean of $n$, $\langle \langle n \rangle \rangle = \langle n \rangle$, the second cumulant is the variance, $\langle \langle n \rangle \rangle = \langle n^2 \rangle - \langle n \rangle^2$, and the third is the skewness, $\langle \langle n^3 \rangle \rangle = \langle (n - \langle n \rangle)^3 \rangle$. It is easy to show that the cumulants of a transport process are simply the sum of the cumulants corresponding to each independent sub-process. Moreover, for a Gauss distribution only the first and second cumulants are non-zero, while all higher cumulants vanish. In this respect, one may use cumulants of a distribution as a measure of (non-)gaussianity.

By looking at the probability distribution $P(n, t)$ we suggest counting the charge traversing a given mesoscopic conductor during the measuring time $t$. Thus, the
measured quantity is charge, not current. Alternatively, one can of course consider the current. The zero-frequency cumulants of the current are given by the long-time limit of the cumulants of $n$ as

$$\langle \langle \langle n^m \rangle \rangle \rangle \equiv \lim_{t \to \infty} \frac{\langle \langle n^m \rangle \rangle}{t}. \quad (1.18)$$

The current is treated as a continuous variable and continuous variables are typically characterized by their cumulants. The conventional moments and cumulants, as defined above, have been investigated intensively in the field of FCS [65]. However, another interesting class of statistical quantities exists, which has received much less attention in FCS. These are the factorial moments and the factorial cumulants, which are mostly discussed in the context of discrete variables [72, 73]. The number of counted electrons $n$ is obviously a discrete variable, and it is natural to ask if the current cumulants, as defined in Eq. (1.18), carry signatures of this discreteness?

The factorial moments are again generated by a factorial MGF, which can be defined based on the GF in Eq. (1.13). The factorial MGF is defined as

$$M_F(z, t) = G(z + 1, t) \quad (1.19)$$

and the corresponding factorial moments read

$$\langle n^m \rangle_F(t) \equiv \partial_z^m M_F(z, t)|_{z \to 0} = \langle n(n-1) \cdots (n-m+1) \rangle \quad (1.20)$$

in terms of the ordinary moments. In analogy with the conventional CGF, the factorial CGF is defined as

$$S_F(z, t) = \log[M_F(z, t)] = \log[G(z + 1, t)] \quad (1.21)$$

and the corresponding factorial cumulants read

$$\langle \langle n^m \rangle \rangle_F(t) \equiv \partial_z^m S_F(z, t)|_{z \to 0} = \langle \langle n(n-1) \cdots (n-m+1) \rangle \rangle. \quad (1.22)$$

As mentioned above, factorial moments and factorial cumulants are of particular interest when considering probability distributions of discrete variables. For example, for a Poisson process with rate $\Gamma$, which is the physical limit of rare events,
Figure 1.2: Example of probability distributions. The points correspond to Poisson distributions with increasing mean, which is indicated with $\Gamma t$. Here, $\Gamma$ represents a tunneling rate and its inverse sets the time unit. The mean value of the distributions increases linearly with time and the distributions spread. The full lines are Gaussian distributions with the same mean and variance $\Gamma t$. a and b are the exact same plots except that in b the probabilities are plotted on a logarithmic scale. This points out the relative difference between the two distributions, which is largest in the tails. The (factorial) cumulants characterize this difference.

The FCS is well-known and reads

$$P(n, t) = \frac{(\Gamma t)^n}{n!} e^{-\Gamma t}.$$  \hspace{1cm} (1.23)

The corresponding GF then becomes

$$G(z, t) = e^{\Gamma t(z-1)}, \quad \text{(Poisson process)}$$  \hspace{1cm} (1.24)

from which it is easy to show that the cumulants are

$$\langle \langle n^m \rangle \rangle (t) = \Gamma t, \quad \text{(Poisson process)}$$  \hspace{1cm} (1.25)

for all $m = 1, 2, \ldots$. Often the noise is compared to Poissonian noise corresponding to a process of uncorrelated events. This is usually done in terms of the Fano factor defined as the ratio

$$F = \frac{\langle \langle n^2 \rangle \rangle}{\langle n \rangle}.$$  \hspace{1cm} (1.26)

The Fano factor equals unity, $F = 1$, for a Poisson process, and as we shall see below, $F > 1$ may only be reached in interacting systems.

In contrast to the cumulants for a Poisson process, the first factorial cumulant
reads
\[ \langle \langle n \rangle \rangle_F(t) = \Gamma t, \quad \text{(Poisson process),} \tag{1.27} \]
while all higher factorial cumulants are zero
\[ \langle \langle n^m \rangle \rangle_F(t) = 0, \quad m > 1 \quad \text{(Poisson process).} \tag{1.28} \]

Thus, similarly to how ordinary cumulants are useful as measures of (non-) Gaussianity, we may use factorial cumulants to characterize deviations of a distribution from Poisson statistics. This is illustrated in Fig. 1.2 where Poisson and Gaussian statistics for a random process with rate \( \Gamma \) are compared at three different instants of time. From the central limit theorem we expect the Poissonian to be close to the Gaussian. However the largest relative difference is in the tails of the distributions and can be clearly seen from a logarithmic plot. The fact that the cumulants of a Poissonian \( \langle \langle n \rangle \rangle \) are nonzero for \( m > 2 \) or alternatively the fact that the factorial cumulants for a Gaussian are nonzero for \( m > 1 \) indicate this difference. Finally, let us mention that a factorial cumulant of a given order is the sum of the factorial cumulants of all independent sub-processes, in the same way as for the cumulants.

The cumulants and the factorial cumulants carry the same information, and from their totality we get the GF and can transform back to obtain the probability distribution \( P(n, t) \). They are linked by the Stirling numbers. The factorial cumulants can be expressed in terms of the ordinary cumulants as follows
\[
\langle \langle n^m \rangle \rangle_F = \sum_{j=1}^{m} s(m, j) \langle \langle n^j \rangle \rangle, \tag{1.29}
\]
where \( s(m, j) \) are the Stirling numbers of the first kind. They can be generated from the relation \[ [\ln (1 + x)]^j = j! \sum_{m=j}^{\infty} [s(m, j) x^m/m!] \].

To conclude this section we point out the different ways of going about counting statistics. Theoretically it may be convenient to compute the GF \( \langle \langle \rangle \rangle \), which then delivers any other statistical quantity as the probability distribution or (factorial) moments and (factorial) cumulants. Having an analytical result for the GF has the advantage that symmetries or elementary sub-processes of the transport can be identified. Theoretical proposals for coupling a detector to the mesoscopic conductor so as to measure the GF have also been put forward. For example a Gedanken-experiment using a rotating spin galvanometer has been suggested \( \langle \langle \rangle \rangle \), or a method using qubits instead \( \langle \langle \rangle \rangle \). Experimentally, however, the direct measurement of the GF remains a challenge and the probability distribution \( P(n, t) \) is more
accessible, for example in experiments where real-time counting of the excess charge on the mesoscopic conductor is possible (also see Sec. 1.2.3 and Ch. 3). Thus, a histogram of the transferred charge can be obtained, which directly yields $P(n, t)$. The (factorial) cumulants can then be obtained using $k$-statistics [72]. Finally, one can consider $P(n, t)$ at short, transient or long times $t$. Since the GF and thus the (factorial) cumulants become linear in time at long times, one in the long-time limit typically considers the current statistics instead of the charge statistics, as in (1.18).

In this thesis we mainly consider zero-frequency counting statistics as in the definitions in this section. In analogy to the finite frequency noise [13] one can do finite frequency counting statistics [76, 77, 78] in order to analyze transport also on short time scales. On the other hand, at zero frequency, interaction and correlation effects are averaged over long times [14]. Let us also mention that the waiting time distribution (WTD) yields a complementary description of charge transport [79, 80, 81, 82, 83]. The WTD is the statistical distribution of waiting times between consecutive tunneling events, which is another way of accessing the short time scales.

### 1.2.3 Measurements of Full Counting Statistics

Parts of this section have been taken, in part verbatim, from publication (iii).

Because (factorial) cumulants are quantities characterizing essentially the tails of a distribution (also see Fig. 1.2), measuring them is a challenge. The uncertainties, including the statistical uncertainty, have to be made extremely small in order to measure high order statistics. Several remarkable experiments have achieved this [84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 78].

Voltage fluctuations up to the third cumulant have been measured in tunnel junctions [84, 86, 92]. Fluctuations of the electronic environment play a crucial role [84] and one way to go about this problem is to account for the environment in the model [84]. Another way is to suppress environmental noise. A great advance towards high-quality statistics is on-chip detection. Ref. [92] uses an on-chip Josephson junction to measure non-Gaussian current fluctuations. Moreover, the fourth and fifth current cumulants have been detected in an avalanche diode [95].

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13 The time scales accessed via finite frequency statistics should not be confused with the counting time $t$ in $P(n, t)$, which may be small. In finite frequency statistics one considers correlators of quantities at different times, that is, one can access relevant time scales of the correlations, whereas from $P(n, t)$ one obtains correlators of $n$ where the time difference is averaged out.

14 Long here means larger than the largest relevant time scale.
In measurements of counting statistics, either current or voltage fluctuations are measured, as for example in Refs. [84, 92, 95], or \( P(n, t) \) is measured directly. The latter way has namely been made possible in systems where electrons can be counted in real-time. This also circumvents issues like subtraction of environmental noise. The pioneering experiments are reported on in Refs. [85, 87]. In order to detect electrons in real time, tunneling rates have to be reduced to the radio frequency range or even lower, say, to a few hundreds of Hz. This can for example be achieved due to strong Coulomb interactions, as in Coulomb blockaded systems. The detector is on-chip and typically consists of a single electron transistor (SET) (examples in [99]) or a quantum point contact (QPC) (examples in [94]) capacitively coupled to the system, such that the SET or QPC conductance is highly sensitive to the charge state of the system, which could for example be a small metallic island or a Coulomb blockaded quantum dot. Thus, the detector current takes distinct values, each of which corresponds to a different charge state of the system. The detector thereby monitors charges entering or leaving the system. If the transport is uni-directional this directly yields a histogram for the transferred charge in a given time, and thus \( P(n, t) \). Typical detector current time traces, showing what charge state the system is in, are shown in Figs. 3.4 and 3.5, and in the reviews mentioned above, Refs. [94, 99].

Counting statistics of charge transport in single [89, 90, 91, 94, 96, 97, 98, 78] and double quantum dots [88, 94] have been measured in this way. Following the initial measurements of the third cumulant of transport through quantum dots [88, 89], a series of experiments have addressed the conditional statistics of the QD given a QPC current [91], the time-dependent high-order cumulants [96, 97, 98], and finite-frequency counting statistics [78] in quantum dot systems. The works on transient high-order cumulants have shown that high-order cumulants generically oscillate as functions of basically any system parameter as well as the observation time [96]. Remarkably, cumulants up to the 15th order and higher have been measured in [96, 97, 98].

Very recently the full counting statistics, \( P(n, t) \), in a SINIS structure has been measured [publication (iv) as well as Sec. 1.3 and Ch. 3], where for energies inside the superconducting gap the dominant transport mechanism is Andreev reflection, so that electrons tunnel in pairs.
1.2.4 Counting statistics calculation techniques

In this section we give a brief overview of the different theoretical approaches to counting statistics and then go on to present the Levitov-Lesovik determinant formula, which applies to noninteracting electrons, as well as the master equation formalism, applying to Coulomb blockaded systems. We chose to elaborate these two techniques in a bit more detail since later results of this thesis employ them. The part about the master equation formalism is based on a course by C. Flindt in the context of the CUSO program at the University of Geneva, from the spring 2013. Moreover, parts of this section have been taken, in part verbatim, from publication (i).

**Brief overview**

On the theory side, several techniques have been developed for calculating the statistics of transferred charges. In the seminal work by Levitov and Lesovik [31], the FCS of noninteracting electrons propagating coherently through a conductor was expressed by a determinant formula containing the scattering matrix of the problem [31, 32, 33, 34]. In many-channel conductors the statistics is predominantly classical and can be described using a Langevin–Boltzmann equation [100, 101]. A powerful and elegant formulation in this semiclassical regime is the stochastic path integral approach, which was introduced for FCS in the pioneering works by Pilgram et al. [102, 103, 104]. For interacting systems, the FCS can be related to a generalized master equation describing the charge transport [66, 76, 105, 106] or obtained using Keldysh Greens functions [107, 34].

**Levitov-Lesovik determinant formula**

In systems where transport can be described with noninteracting electrons, the scattering matrix formalism [20, 21, 22, 23, 24] can be applied. The scattering matrix $S$ is typically given in the basis of asymptotic scattering states and is characteristic for a given conductor. The only source of correlations in this case are the fermionic statistics, that is, the Pauli principle. Levitov and Lesovik have shown [31] (also see Levitov or Klich in [65]) that for noninteracting electrons the MGF is given by the functional determinant

$$G(e^z) = \det \left[ \mathbb{I} + \hat{n}(S^t e^{zP} Se^{-zP} - \mathbb{I}) \right],$$

(1.30)
where the operator $\hat{n}$ is diagonal in the space of scattering states with the occupation numbers on the diagonal, which are Fermi distributions for reservoirs at equilibrium. Moreover, $P$ is a projector onto the drain reservoir as we choose to count electrons entering the drain. The determinant (1.30) is readily generalized to multi-terminal systems [109, 110]. We have supposed that the scattering time is very short, that is,

$$\tau_{sc} \sim \hbar \left| \frac{\partial S_{\alpha \beta}}{\partial E} \right|$$

(1.31)

is the shortest time scale so that $S$ does not introduce a delay time in the scattering process. Therefore the time dependence of the MGF in the long-time limit is Markovian and is obtained by integrating on energy according to

$$\ln G(e^{\epsilon}) = t \int \ln \left[ G_E(e^{\epsilon}) \right] \frac{dE}{2\pi\hbar},$$

(1.32)

where the $G_E(e^{\epsilon})$ is given by (1.30). What is more, a time-dependent scattering matrix $S = S(t)$, obtained for example in the presence of time-dependent external fields, bias or gate voltages, can be Fourier-transformed to yield $S(h\omega)$, where we have introduced the quasi-energies $h\omega$. Since time-dependent scattering will mix electron states of different energies [32, 108], the determinant (1.30) has to be taken over the transverse conduction channel indices, as well as over the quasi-energies, which makes (1.30) a functional determinant (see for example Levitov in [65]). Therefore relation (1.30) is in general an infinite determinant and has to be regularized accordingly if we consider infinite reservoirs. This has been addressed in [108] and [111], where an electron-hole symmetric regularized determinant is proposed.

In order to express the GF instead of the MGF, Eq. (1.30) can be cast into the following form [69, 70], which we shall use in Sec. 1.2.5:

$$G(z) = \det \left[ \left[ I + (z - 1)X \right] z^{-P} \right],$$

(1.33)

---

15 The time dependence in Eq. (1.30) comes in through the scattering matrix $S(t')$ as well as the occupation number operator, which is nonlocal in time [see e.g. Eq. (E8) in [108]]:

$$\hat{n}_{nn}(t, t') = \sum_k e^{iE_k(t-t')} n_F(E_k),$$

where $n$ is the channel index, $n_F(E)$ is the Fermi distribution function [Eq. (1.63)] and $E_k = \frac{\hbar^2 k^2}{2m} + E_n$ with $E_n$ the transverse energy of the $n^{th}$ conduction channel. The matrix product in (1.30) includes a convolution over $t'$, which in Fourier space becomes a product.
with \( X = (1 - \hat{n})P + \hat{n}S^\dagger PS \), and we have substituted \( e^z \rightarrow z \).

**Master equation formalism**

Consider a conductor (system) that is coupled to electronic reservoirs, such that electrons can hop on and off the conductor and transport between the reservoirs, through the conductor, is possible. To simplify matters, let us consider two reservoirs, \( S \) and \( D \), as sketched in Fig. 1.1. Suppose our system is described by its density matrix \( \hat{\rho}(t) \), the evolution of which is governed by a generalized Markovian master equation

\[
\frac{d}{dt} \hat{\rho}(t) = \mathcal{L} \hat{\rho}(t) .
\]  

(1.34)

The operator \( \mathcal{L} \) is called the Liouvillian, which for a typical quantum system takes the following form:

\[
\mathcal{L} \hat{\rho} = \frac{1}{i\hbar} \left[ \hat{H}_S, \hat{\rho} \right] + \sum_{\alpha, j} \Gamma_{\alpha, j} \left( \hat{c}_{\alpha, j}^\dagger \hat{\rho} \hat{c}_{\alpha, j} - \frac{1}{2} \{ \hat{c}_{\alpha, j}^\dagger \hat{c}_{\alpha, j}, \hat{\rho} \} \right),
\]

(1.35)

where \( \hat{H}_S \) is the system Hamiltonian and the first term thus governs the system evolution, whereas the second term describes the quantum jumps in the occupations as well as the evolution of the coherences due to the coupling of the system to the reservoirs. The operators \( \hat{c}_{\alpha, j}^\dagger \) and \( \hat{c}_{\alpha, j} \) are the creation and annihilation operators of an electron in state \( j \) of reservoir \( \alpha \), with \( \alpha = S, D \).

Since at the end of the day we are interested in the probability distribution \( P(n, t) \), we unravel the density matrix with respect to the number \( n \) of electrons transferred through the conductor during \( t \), and write

\[
\hat{\rho}(t) = \sum_n \hat{\rho}(n, t),
\]

(1.36)

where \( \hat{\rho}(n, t) \) is the \( n \)-resolved density matrix [112, 113]. Here, \( t = 0 \) corresponds to the time at which we start counting electrons. For definiteness we shall choose that the positive direction of transport corresponds to electrons tunneling from the source (S) into the drain (D), and we count electrons entering the drain. The next step is to unravel the master equation [133]. We obtain

\[
\frac{d}{dt} \hat{\rho}(n, t) = \mathcal{L}_0 \hat{\rho}(n, t) + \mathcal{J}_+ \hat{\rho}(n - 1, t) + \mathcal{J}_- \hat{\rho}(n + 1, t),
\]

(1.37)

16Markovian means that any memory effects can be neglected and therefore \( \mathcal{L} \) is time-independent here.
supposing that electrons tunnel one by one. We have written

\[ \mathcal{L} = \mathcal{L}_0 + \mathcal{J}_+ + \mathcal{J}_-, \quad (1.38) \]

with \( \mathcal{J}_\pm \) the quantum jump operators to and from the drain reservoir, respectively, and \( \mathcal{L}_0 \) describing the free evolution. Let us remark that if the transport is unidirectional, for example in the presence of a large bias compared to temperature, then we can set \( \mathcal{J}_- \equiv 0 \).

In order to solve Eq. (1.37), let us now introduce an auxiliary counting variable \( z \) through the following transform:

\[ \hat{\rho}(z, t) = \sum_n \hat{\rho}(n, t) z^n. \quad (1.39) \]

Inserting (1.39) into (1.37) yields

\[ \frac{d}{dt} \hat{\rho}(z, t) = \left[ \mathcal{L}_0 + z \mathcal{J}_+ + \frac{1}{z} \mathcal{J}_- \right] \hat{\rho}(z, t) \equiv \mathcal{L}(z) \hat{\rho}(z, t), \quad (1.40) \]

which is readily solved to give

\[ \hat{\rho}(z, t) = e^{\mathcal{L}(z)t} \hat{\rho}(t = 0) = e^{\mathcal{L}(z)t} \hat{\rho}_S. \quad (1.41) \]

For a constant bias voltage there is typically a unique steady state, \( \hat{\rho}_S \). Supposing that we start counting when the system is in its steady state we have \( \hat{\rho}(t = 0) = \hat{\rho}_S \).

Introducing the transform (1.39) turns out to be very convenient since the GF (1.13) is given by

\[ \mathcal{G}(z, t) = \text{Tr} \left[ \hat{\rho}(z, t) \right] = \text{Tr} \left[ e^{\mathcal{L}(z)t} \hat{\rho}_S \right], \quad (1.42) \]

and the (factorial) moments and cumulants can be readily generated, as described in Sec. 1.2.2. Alternatively, we can transform back to the probability picture,

\[ P(n, t) = \int_{-\pi}^{\pi} e^{-inx} \mathcal{G}(e^{ix}, t) \frac{d\chi}{2\pi}. \quad (1.43) \]

For numerical calculations the inversion (1.43) can be done with the method from Ref. [114], an approximation the numerical accuracy of which is readily controlled by a parameter. The result is formulated as a theorem and is given in App. B.2.

If the system can be described with quasiclassical probabilities \( p_j(n, t) \equiv \hat{\rho}_{jj}(n, t) \) to be in state \( j \) given that \( n \) charges have traversed it during \( t \), then the GME (1.40)
becomes a classical master equation

\[ \frac{d}{dt} \langle g(z, t) \rangle = M(z) \langle g(z, t) \rangle \]  

(1.44)

where we have used the notations of publication (i), that is, \( |g(z, t)\rangle \rangle = \sum_n |p(n, t)\rangle \rangle z^n \) with \( |p(n, t)\rangle \rangle = [p_1(n, t), \ldots, p_N(n, t)]^T \) and \( M(z) \) is the \( z \)-dependent kernel of the master equation, in analogy to the Liouvillian operator. At \( z = 1 \), that is without the counting, the kernel \( M(z = 1) \) has a unique zero eigenvalue, and the corresponding eigenvector is the steady state that we denote \( |0\rangle \rangle \). Since by probability conservation \( \sum_j \sum_n p_j(n, t) = G(z = 1, t) = 1 \), the kernel at \( z = 1 \), \( M(z = 1) \), has a unique left eigenvector \( \langle \langle \tilde{0} | = [1, \ldots, 1] ^T \) such that \( \langle \langle \tilde{0} | M(1) = 0 \). Multiplication from the left by \( \langle \langle \tilde{0} \) corresponds to the trace operation in the quantum case. Finally, we normalize the steady state such that \( \langle \langle \tilde{0} | 0 \rangle \rangle = 1 \). Thus, in the quasiclassical case (1.42) becomes

\[ G(z, t) = \langle \langle \tilde{0} | e^{M(z) t} |0\rangle \rangle . \]  

(1.45)

For a given model determining the kernel \( M(z) \) we can in principle obtain the full counting statistics from (1.45), at any time \( t \). However, in practice, taking high order derivatives of the GF (1.45) might be a difficult task, especially if the dimensions of the kernel are large. Therefore, in order to compute high order (factorial) cumulants we propose a method\footnote{Already for \( 3 \times 3 \) matrices the proposed method speeds up computations considerably and helps numerical accuracy.} where derivatives are taken at the level of Eq. (1.44) [App. A.1].

In the long-time limit, \( t \gg \Gamma^{-1} \) with \( \Gamma \) a characteristic tunneling rate, the (factorial) CGF simplifies\footnote{Already for \( 3 \times 3 \) matrices the proposed method speeds up computations considerably and helps numerical accuracy.} according to

\[ S_{(F)}(z, t) \simeq \lambda_0(z) t . \]  

(1.46)

The function \( \lambda_0(z) \) generates the (factorial) current cumulants\footnote{Already for \( 3 \times 3 \) matrices the proposed method speeds up computations considerably and helps numerical accuracy.} and corresponds to the dominant eigenvalue of \( \tilde{M}(z) \equiv M(e^z) \) [or \( \tilde{M}(z) \equiv M(z + 1) \) for the factorial cumulants], which develops adiabatically from \( \lambda_0(0) = 0 \). Eq. (1.46) can be readily understood from (1.45) by observing that \( M(1) = \tilde{M}(0) \) has only eigenvalues with negative real parts in addition to the unique zero eigenvalue. If for nonzero \( z \) the eigenvalues develop continuously, then \( \lambda_0 \) must be the dominant eigenvalue close to \( z = 0 \). Although Eq. (1.46) is formally simple, it might be cumbersome to obtain (factorial) cumulants up to a high order \( m \) by differenti-
ating $\lambda_0(z) m$ times. Instead, it may be computationally much easier to obtain the (factorial) current cumulants recursively order by order \[106\]. The recursive scheme uses the expansion

$$
\lambda_0(z) = \sum_{n=1}^{\infty} \frac{z^n}{n!} \langle \langle I^n \rangle \rangle ,
$$

(1.47)

and is explained in App. \[A.2\] for completeness.

Another way of representing the FCS in the long-time limit is the large deviation function (LDF) $\xi(I)$

$$
\lim_{t \to \infty} \frac{\ln P(n/t, t)}{t} \rightarrow \xi(I) ,
$$

(1.48)

where $I = n/t$ is treated as a continuous variable. As its name already suggests the LDF characterizes large deviations, that is, events far from the mean of the distribution that have a small probability to occur. It is given by

$$
\xi(I) = \lambda_0(z_0) - Iz_0 ,
$$

(1.49)

with $z_0 = z_0(I)$ satisfying

$$
\lambda_0'\left(z_0\right) - I = 0 ,
$$

(1.50)

as explained in App. \[A.4\]. In case it is lengthy to obtain the LDF analytically, the latter can be computed numerically for a discrete set of values of the current $I$. To this end we observe that we have

$$
\lambda_0'(z) = \langle \langle 0 | \frac{d}{dz} M(e^z) | 0(e^z) \rangle \rangle ,
$$

(1.51)

where $| 0(z) \rangle \rangle$ is the eigenvector corresponding to the 0 eigenvalue, i.e. $M(z)| 0(z) \rangle \rangle = 0$, with the normalization condition $\langle \langle 0 | 0(z) \rangle \rangle = 1$ for all $z$. Thus, for a given $I$, Eq. (1.50) yields $z_0(I)$ that is then inserted into (1.49).

The quasiclassical formalism of generalized Markovian master equations can generally be applied to Coulomb blockaded mesoscopic systems in the sequential tunneling limit, such that $eV, k_B T \gg \hbar \Gamma$, where $V$ is the bias voltage, $T$ is the temperature and $\Gamma$ is a characteristic tunneling rate \[66\]. The main advantage of the master equation formalism is that the interaction effects are treated non-perturbatively. We have applied the formalism to several concrete examples of Coulomb blockaded systems and refer the reader to publications (i) - (iv), where the kernels $M(z)$ are obtained from the corresponding Hamiltonians.

\[18\] Here $\lambda_0$ is the long-time limit CGF, that is, the dominant eigenvalue of $M(e^z)$.
The formalism can be extended to non-Markovian dynamics \cite{105, 106}, which may for example come about if the system is described by a reduced density matrix, where the environment degrees of freedom have been traced out. For example, coupling to a dissipative phonon bath may introduce memory effects when the bath degrees of freedom are traced out \cite{105, 106}. What is more, the finite frequency counting statistics can be accessed using the formalism from Refs. \cite{76, 77, 78}. Also, instead of applying a constant bias voltage such that there is a steady state \( \hat{\rho}_S \), one can consider time-dependent driving of the system. If, for example, the driving is periodic, we can define a probability distribution per period and then consider the limit of a large number of periods \cite{108}. However, in the examples in this thesis we deal with Markovian master equations and a constant bias.

1.2.5 Factorization of the statistics for non-interacting electrons

Parts of this section have been taken, in part verbatim, from publication (i).

Abanov and Ivanov have shown that the counting statistics of noninteracting electrons
\footnote{Noninteracting means that the system can be described by a quadratic fermionic Hamiltonian.} underlies a constraint \cite{69, 70}: The GF must take a generalized binomial form

\[
G(z, t)_{\text{generalized binomial}} = z^{-Q} \prod_i (1 - p_i + p_i z),
\]

(1.52)

with \( 0 \leq p_i \leq 1 \). In other words, the zeros of the GF, \(-(1 - p_i)/p_i\), must lie on the real negative axis [Fig. 1.3 a]. The factor \( z^{-Q} \) corresponds to a deterministic background charge transfer \( Q = \sum_i p_i - \langle n \rangle \geq 0 \) opposite to the positive direction, where \( \langle n \rangle \) is the mean value of the total transferred charge. For unidirectional transport, we have \( Q = 0 \). A GF of the form (1.52) has been dubbed generalized binomial in \cite{115} since the factors \( (1 - p_i + p_i z) \) correspond to the binomial GF for a single attempt with success probability \( p_i \). In fact, the GF for a process with \( N \) trial events and with success probability \( p \) is

\[
G_b(z) = (1 - p + pz)^N,
\]

(1.53)

yielding the binomial distribution for the probability of \( n \) successful events,

\[
P_b(n) = \binom{N}{n} p^n (1 - p)^{N-n}.
\]

(1.54)
Figure 1.3: Schematics of two zeros of the GF in the complex plane. a Real and negative zeros corresponding to generalized binomial statistics. b The zeros have split into a pair of complex conjugate zeros due to interactions.

Eq. (1.52) states that the statistics can be factorized into independent single-electron events of binomial form with different success probabilities $p_i$. The $p_i$, however, depend on system parameters as well as time in a nontrivial way and can only be interpreted as true attempt probabilities in the limit where the wave packets of consecutive electrons have no overlap, such that their transport stays uncorrelated. Eq. (1.52) holds in the limit of instant scattering [Eq. (1.31)] of noninteracting electrons, for an arbitrary scattering matrix $S = S(E, t)$ and for any temperature [70]. It can be obtained from (1.33) by observing that the eigenvalues of the operator $X$ are real and confined to the interval $[0, 1]$. This result generalizes previous results for a generic scatterer driven by a time-dependent voltage bias [67, 68] and for a finite number of wave-packets in the presence of energy-dependent scattering [115]. We remark that for a multi-terminal setup the constraint on the GF does not take as simple a form [110].

Importantly, the result of Abanov and Ivanov implies that if a GF cannot be factorized as in Eq. (1.52) with real probabilities $p_i$, it cannot be describing noninteracting electrons. Interactions, on the other hand, may cause the zeros of the GF to move into the complex plane [Fig. 1.3b] so that the statistics is no longer generalized binomial. We note, however, that the statement cannot be turned around: even in the presence of interactions the statistics may still be generalized binomial [70].

1.2.6 Generic oscillating behavior of high order cumulants

Parts of this section have been taken, in part verbatim, from publications (i) and (iii). We also adopt the notation of the latter.

In this section we discuss an approximation for high derivatives [116, 117] and
apply it to high order (factorial) cumulants. The approximation gives us an understanding of the generic behavior of high order (factorial) cumulants and suggests that cumulants generically oscillate as functions of any system parameter and of the counting time \[96\]. As we shall find, the factorial cumulants behave differently.

From (1.16) and (1.21) respectively, we see that the (factorial) CGF at finite times has logarithmic singularities at the zeros of the corresponding MGF, namely \( \mathcal{G}(e^z, t) \) or \( \mathcal{G}(z + 1, t) \). Close to the singularity \( z_j \) we can write

\[
S_{(F)}(z, t) \simeq \alpha_j \ln(z_j - z), \quad z \text{ close to } z_j,
\]

where \( \alpha_j \) is the degeneracy of \( z_j \). Next, we take consecutive derivatives

\[
\partial_z^m S_{(F)}(z, t) \simeq -\alpha_j \frac{(m - 1)!}{(z_j - z)^m}, \quad z \text{ close to } z_j.
\]

According to the Darboux approximation \[116, 117\] we can approximate high order derivatives by summing over all the singularities \( \{z_j\} \). Specifically, at \( z = 0 \) this yields an approximation for the (factorial) cumulants

\[
\langle \langle n^m \rangle \rangle_{(F)} = \partial_z^m S_{(F)}(z, t)|_{z \to 0} \\
\simeq - (m - 1)! \sum_j \alpha_j \frac{e^{-im \arg[z_j]}}{|z_j|^m}.
\]

Here we have introduced the polar notation \( z_j = |z_j|e^{i\arg z_j} \). The above approximation (1.57) is of practical use if the sum can be truncated. We notice that the high-order (factorial) cumulants are determined by the singularities closest to \( z = 0 \), which dominate the sum for large \( m \). Contributions from other terms are suppressed with the relative distance to \( z = 0 \) and the power \( m \) \[96\].

Let us remark that in the long-time limit the (factorial) CGF typically does not exhibit logarithmic but other types of singularities. For example in the master equation formalism, the long-time limit CGF \( \lambda_0(z) \) [Eq. (1.46)] typically has square root singularities. Following \[96\] we have therefore used a general notation in publication (iii) and wrote, instead of (1.55),

\[
S_{(F)}(z, t) \simeq \frac{A_j}{(z_j - z)^{\mu_j}}, \quad z \text{ close to } z_j,
\]

where \( A_j, \mu_j \) are constants and \( \mu_j \) is determined by the nature of the singularity. For example \( \mu_j = -1/2 \) for a square root branch point. In analogy to (1.57) we
then write an approximation for the (factorial) cumulants

\[ \langle \langle n^m \rangle \rangle_F \simeq \sum_j A_j B_{m,\mu_j} |z_j|^{m+\mu_j} e^{-i(m+\mu_j)\arg z_j}, \]  

(1.59)

with

\[ B_{m,\mu_j} \equiv \mu_j (\mu_j + 1) \cdots (\mu_j + m - 1). \]  

(1.60)

To recover (1.57) from (1.59) we need to formally set \( \mu_j = 0, A_j = -1 \) as well as \( B_{m,\mu_j} \to (m - 1)! \). From either relation, however, we observe that the cumulants of the transferred charge will oscillate as a function of any parameter that changes the arguments of the singularities of the (factorial) CGF. Moreover, the (factorial) cumulants grow factorially with their order \( m \). We also observe trigonometric oscillations as a function of the order \( m \).

Let us now look at two special cases where we can truncate the sum in (1.59). For example, if a single complex conjugate pair of singularities \( z_0, z_0^* \), are closest to \( z = 0 \), the high-order (factorial) cumulants can be approximated as

\[ \langle \langle n^m \rangle \rangle_F \simeq 2|A_0| B_{m,\mu_0} |z_0|^{m+\mu_0} \cos [(m + \mu_0) \arg z_0 - \arg A_0]. \]  

(1.61)

This result emphasizes once more that the absolute value of the (factorial) cumulants generically grows factorially with the cumulant order \( m \), due to the factors \( B_{m,\mu_0} \), and that they tend to oscillate as a function of any parameter, including time \( t \), that changes \( \arg z_0 \). Such universal oscillations have been observed experimentally in electron transport through a quantum dot [96, 97, 98].

In contrast, in the particular situation where there is just a single dominant singularity \( z_0 \) on the negative real axis, the high-order (factorial) cumulants can be approximated as

\[ \langle \langle n^m \rangle \rangle_F \simeq (-1)^{m+\mu_0} \frac{A_0 B_{m,\mu_0}}{|z_0|^{m+\mu_0}}. \]  

(1.62)

In this case, the factorial growth with the order persists, but no oscillations are expected as long as the dominant singularity \( z_0 \) stays on the negative real axis.

\[ ^2\text{We remark that since the GF [Eq. (1.13)] is a transform of the probability distribution } P(n, t), \text{ which is real, the zeros of the GF always come in complex conjugate pairs. It follows directly that the singularities of the (factorial) CGF must come in complex conjugate pairs as well.} \]
1.3 Counting statistics in N-S hybrid systems

Before starting this section, the author would like to recommend the comprehensive textbook by Tinkham [118] as well as to point out that an excellent summary on the basic properties of BCS superconductors can be found in Ch. 2 of Ref. [119]. The author has also profited from the thesis draft by V. F. Maisi from Aalto University School of Science, Aalto, Finland and Centre for Metrology and Accreditation (MIKES), Espoo, Finland, as well as from Ref. [120].

1.3.1 Densities of states in a normal metal vs. superconductor

In a normal metal (N) the density of states can typically be assumed constant around the Fermi energy. The occupation number at equilibrium is given by the Fermi distribution function

\[ n_F(E) = \frac{1}{e^{(E - \mu)/k_BT} + 1}, \]  

(1.63)

where \( k_B \) is Boltzmann’s constant and \( T \) is the temperature. The carriers contributing to transport have energies close to the chemical potential \( \mu \), which coincides with the Fermi energy \( E_F \) at zero temperature. In contrast, in a standard superconductor (S) described by BCS theory, the single-particle density of states is gapped around \( E_F \).

Below a characteristic temperature \( T_C \), BCS superconductors have the property to conduct dissipationless electrical current, and this even without an applied bias. This is due to the pairing of electrons into Cooper-pairs as a consequence of the weak effective binding force mediated through electron-phonon interactions. Below \( T_C \) the ground state corresponds to a condensate of Cooper-pairs, which can be written in a single macroscopic wave function called the BCS ground state. The superconducting phase transition is described by a complex order parameter \( \tilde{\Delta} = \Delta e^{i\phi(\vec{r})} \). Its magnitude \( \Delta \) yields the superconducting energy gap of \( 2\Delta \), being maximal at \( T = 0 \) and vanishing for \( T > T_C \). A gradient in the superconducting phase \( \phi(\vec{r}) \), on the other hand, drives a supercurrent. Two electrons forming a Cooper-pair are highly correlated, having exactly opposite spin and momentum.

\(^{21}\)BCS stands for Bardeen-Cooper-Schrieffer [121, 122]. For an introduction to the theory, see any introductory textbook on superconductivity, e.g. [118].

\(^{22}\)They form a spin singlet.
Remarkably, the spatial extension of a Cooper-pair can be on the order of \( \xi \sim 1 \mu m \) \(^{23}\), the superconducting correlation length, which is much larger than the mean distance between free electrons.

Inside the superconducting gap of \( 2\Delta \) there are no single-particle states. The BCS single-particle density of states around the Fermi energy (i.e. we set \( E_F = 0 \)) reads

\[
N_S(E) = N_0 \frac{|E|}{\sqrt{E^2 - \Delta^2}} \Theta(E^2 - \Delta^2),
\]

where \( N_0 \) is the constant density of states in the normal metallic state. Dissipative transport where the carriers are single electron quasiparticles therefore only occurs at energies \( E > \Delta \), for example when the applied bias voltage \( eV \) exceeds \( \Delta \). Also, temperature induces quasiparticle excitations above the gap and therefore empty states below, according to the Fermi function \((1.63)\).

### 1.3.2 N-S contacts, the proximity effect and Andreev reflection

When a normal metal and a superconductor are brought into good contact, then superconducting pair correlations penetrate into the N part to a distance of the order of the correlation length \( \xi \). This is called the proximity effect \(^{123}\) \(^{124}\), the key mechanism of which is Andreev reflection \(^{125}\) \(^{124}\) \(^{118}\) \(^{126}\). At energies inside the superconducting gap, \( |E| < \Delta \), single-electron transport between the normal metal and the superconductor is forbidden. In the process of Andreev reflection, two electrons are transferred across the N-S interface, as described in Sec. 3.3.1. In short, an incident electron in the N part is retroreflected as a hole such that a Cooper pair is formed in the superconductor. The retroreflected hole has opposite spin and momentum than the incident electron.

The superconducting proximity effect has diverse consequences \(^{119}\), for example that an SNS weak link can support a supercurrent \(^{127}\) \(^{128}\) \(^{129}\).

### 1.3.3 Counting statistics in N-S hybrid systems

Counting statistics in N-S hybrid structures has become a topic of several theoretical investigations \(^{130}\) \(^{131}\) \(^{132}\) \(^{133}\) \(^{134}\) \(^{135}\) \(^{136}\) \(^{137}\) \(^{138}\) \(^{139}\). Using circuit theory and Keldysh Green’s functions \(^{65}\), a general formalism to calculate the FCS

\(^{23}\)The size of a Cooper pair can be estimated \(^{2}\) using the Heisenberg uncertainty relation: \( \xi \equiv \delta r \sim h/\delta p \sim hv_F/\Delta \), where \( v_F \) is the Fermi velocity. Using values for aluminum \(^{119}\) we get \( \xi \sim 7 \mu m \).
in N-S hybrid structures has been developed \[130, 134\]. The FCS of a SNS weak link has been investigated, where the N part is shorter than the superconducting coherence length \[131, 133, 136\]. Moreover, the correlations due to the breaking of Cooper pairs in interferometers or beam splitters have been addressed \[132, 138\], as well as a superconductor-ferromagnet entangler \[140\]. If in such a way one could produce entangled electron pairs, many interesting applications would open up, for example in the context of quantum computing. Moreover, the FCS in a chaotic cavity connected to a normal and a superconducting lead have been investigated \[139, 137\]. Also, the energy dependent FCS in a NINIS junction indicates that the current is transported via the tunneling of correlated pairs of electrons \[135\]. The I here stands for a thin insulating layer forming a tunnel barrier. What is more, the proximity effect as a function of the level detuning has been investigated in a superconductor-QD-normal metal structure by analyzing the FCS \[141\]. Finally, let us remark that the FCS in superconducting point contacts has been found to follow a multinomial distribution, showing that charge is transferred in large quanta, which is due to multiple Andreev reflection \[142, 143\].

Experiments investigating the FCS of electrons transported in pairs have, however, so far been lacking. In publication (iv) and Ch. 3 we present such an experiment, where Andreev tunneling events have been detected in real-time. The experiment has been performed in a symmetric hybrid structure with a tunnel barrier (I) separating the N and S parts. The so-called SINIS structure is shown in Fig. 3.1.
Chapter 2

Factorial cumulants reveal interactions in counting statistics

In this chapter we shall first present our main result and then reprint publications (i) through (iii), which contain model calculations illustrating our result. In publication (i) [Sec. 2.2] a QD with a single spin degenerate level is considered. When two electrons can occupy the dot, indications of the Coulomb interaction are seen in the counting statistics. In publication (ii) [Sec. 2.3.2] a model of a two-level QD is considered, such that the QD can at most be occupied by one additional electron. This strong Coulomb blockade also strongly affects the behavior of the factorial cumulants. Finally, in publication (iii) [Sec. 2.4.3] we consider a double quantum dot (DQD), where the two dots are in series and there are coherent oscillations between the two dots. When there is one additional electron on each dot there is again Coulomb repulsion, which leaves a signature in the counting statistics. Moreover, we consider the case where the DQD is capacitively coupled to a QPC detecting the charge state of each of the dots, and study its influence on the counting statistics. In an experiment the DQD would typically be coupled to a QPC, with which the counting statistics are measured [see Sec. 1.2.3].

2.1 Main result

Parts of this section have been taken, in part verbatim, from publications (i) and (iii).

Our main result is based on the results from Secs. 1.2.5 and 1.2.6. First, the statistics for noninteracting electrons takes a generalized binomial form such that the zeros of the GF are real and negative [69, 70]. Second, high order cumulants
generally oscillate as functions of basically any parameter, including the counting time \[96\]. This might suggest to the reader that any information contained in high order statistics is masked by these oscillations. However, the factorial cumulants behave differently. For noninteracting electrons the GF is of the form (1.52). The cumulants are complicated functions of the \( \{ p_i \} \), whereas the factorial cumulants take a simple form:

\[
\langle \langle n^m \rangle \rangle_F \overset{\text{generalized binomial}}{=} (-1)^{m-1}(m-1)! \left[ \sum_i p_i^m - Q \right].
\] (2.1)

If the transport is uni-directional, then \( Q = 0 \) and two factorial cumulants of consecutive order have a different sign that is otherwise fixed. This means that for noninteracting electrons the factorial cumulants do not oscillate. In fact, even if system parameters or time are varied, the zeros \( z_j = (p_j - 1)/p_j \) of the GF [Eq. (1.52)] must stay on the real negative axis. Thus, also Eqs. (1.57) or (1.59) yield that the factorial cumulants do not oscillate, since the argument of the \( \{ z_j \} \) is fixed to \( \arg[z_j] = \pi \), which equally gives the alternating sign for consecutive orders \( m \). We remark that it is straightforward to extend this analysis to bi-directional transport. From the expression \( Q = \sum_i p_i - \langle n \rangle \) we see that in that case the quantity

\[
\langle \langle n^m \rangle \rangle_F - (-1)^{m-1}(m-1)! \langle n \rangle = (-1)^{m-1}(m-1)! \sum_i (p_i^m - p_i)
\] (2.2)

has alternating sign as a function of \( m \) for generalized binomial statistics.

For uni-directional transport \( (Q = 0) \), the largest probability \( p_{\text{max}} \) will dominate the high factorial cumulants, which can be approximated as

\[
\langle \langle n^m \rangle \rangle_F \simeq (-1)^{m-1}(m-1)! p_{\text{max}}^m.
\] (2.3)

This expression can also be understood from Eq. (1.57) by noting that the zero corresponding to the largest probability \( p_{\text{max}} \) is closest to \( z = 0 \) and will dominate the high factorial cumulants as seen in Eq. (2.3).

As a result, if oscillating factorial cumulants are observed, this must be due to interactions. The test may be made for high order factorial cumulants as functions of a system parameter or time. Alternatively, we can fix parameters and look at the factorial cumulants as functions of their order \( m \). If we obtain a simple sign-flip for consecutive orders, then the statistics is generalized binomial. However, if the
factorial cumulants follow Eq. (1.61) with a cosine oscillation, then electrons must be interacting. This kind of analysis has been applied to three different models of Coulomb blockaded QDs in publications (i) through (iii). Importantly, factorial cumulants are measurable and the test is consequently also immediately applicable to experimental data. The first experiment measuring high order factorial cumulants uses a \( p \)-type GaAs QD [144]. The factorial cumulants are non-oscillating, which is consistent with the modelling as a two-state system.

Finally, we remark that our test using factorial cumulants is stronger than what can be deduced from the Fano factor [Eq. (1.26)]. In fact, we deduce from (1.52) that for uni-directional transport of noninteracting electrons, the (time-dependent) Fano factor reads

\[ F(t) \equiv \frac{\langle n^2 \rangle(t)}{\langle n \rangle(t)} = \frac{\sum_i p_i (1 - p_i)}{\sum_i p_i}, \]

which is always smaller than unity, corresponding to a Poisson process. Following this reasoning, a super-Poissonian Fano factor, \( F > 1 \), can be taken as a sign of interactions. Still, the noise may also be sub-Poissonian, \( F < 1 \), in the presence of interactions. Such an example is given in publication (iii) [Sec. 2.4.3] where for some parameter range we find a Fano factor smaller than one, however the factorial cumulants oscillate and thus clearly indicate the presence of interactions.
2.2 Factorial cumulants reveal interactions in counting statistics: publication (i)

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Factorial cumulants reveal interactions in counting statistics

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Full counting statistics concerns the stochastic transport of electrons in mesoscopic structures. Recently it has been shown that the charge transport statistics for noninteracting electrons in a two-terminal system is always generalized binomial: it can be decomposed into independent single-particle events, and the zeros of the generating function are real and negative. Here we investigate how the zeros of the generating function move into the complex plane due to interactions and demonstrate that the positions of the zeros can be detected using high-order factorial cumulants. As an illustrative example we consider electron transport through a Coulomb blockade quantum dot for which we show that the interactions on the quantum dot are clearly visible in the high-order factorial cumulants. Our findings are important for understanding the influence of interactions on counting statistics, and the characterization in terms of zeros of the generating function provides us with a simple interpretation of recent experiments, where high-order statistics have been measured.

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1. INTRODUCTION

Full counting statistics (FCS) has been a topic of active research for nearly two decades.1–4 FCS describes the statistics of charge transport through mesoscopic conductors [Fig. 1(a)] and is expected to provide more information about the physical processes inside a conductor compared to what is available from the mean current and the shot noise only. Within this framework it is natural to ask how such additional information can be extracted from the high-order statistics and what quantities or measures are most suitable to this end. This core problem of FCS constitutes the main focus of the present work.

On the experimental side, FCS has recently gained considerable impetus due to a number of measurements of high-order statistics in nanoscale systems. While earlier experiments were restricted to the first few moments or cumulants of the current, high-order cumulants of the charge transport are now becoming experimentally accessible. The fourth and fifth cumulants of the current have recently been measured both in Coulomb blockade quantum dots3 and in avalanche diodes.4 Quantum dots, in particular, have emerged as useful sources of high-accuracy counting statistics. In these systems, the low (kiloherertz) tunneling rates allow for detection of individual electrons in real-time using a nearby quantum point contact whose conductance is sensitive to the charge occupations of the dots.3,5,6 Although quantum effects are typically suppressed at the long time scales characterizing the charge transport, quantum dots provide a unique setting to experimentally test theoretical predictions for high-order statistics. Remarkably, time-dependent cumulants of the transferred charge beyond the 15th order have recently been measured in single-electron transport through a Coulomb blockade quantum dot7–9 and a wealth of statistical data is now available.

On the theory side, several techniques have been developed for calculating the statistics of transferred charges. In the seminal work by Levitov and Lesovik,1 the FCS of noninteracting electrons propagating coherently through a conductor was expressed by a determinant formula containing the scattering matrix of the problem.1,10–12 In many-channel conductors the statistics is predominantly classical and can be described using a Langevin-Boltzmann equation.13,14 A powerful and elegant formulation in this semiclassical regime is the stochastic path integral approach, which was introduced for FCS in the pioneering works by Pilgram et al.15–17 For interacting systems, the FCS can be related to a generalized master equation describing the charge transport18–21 or obtained using Keldysh Greens functions.12,22

The central object in the theory of FCS is the generating function (GF) for the probability distribution \( P(n,t) \) of the number of transferred electrons \( n \). Its analytic properties as well as its symmetries as a function of voltage, temperature, and counting fields, which, for example, lead to fluctuation relations,23–26 are therefore of fundamental interest. If the transport process consists of independent, elementary events, the GF may be factorized according to these. This has been

FIG. 1. Nanoscale conductors. (a) Generic transport setup consisting of a nanoscale conductor (gray) connected to source and drain electrodes. Electron flow in both directions (arrows) is allowed. The probability distribution for the number of electrons \( n \) that have been collected in the drain electrode during the time span \([0,t]\) is denoted \( P(n,t) \). (b) Single-level quantum dot coupled via tunnel barriers to source (S) and drain (D) electrodes, kept at temperatures \( T_S \) and \( T_D \), respectively, and chemical potentials \( \mu_S \) and \( \mu_D \). The single-level energy of the quantum dot is \( \epsilon_d \) and the on-site Coulomb interaction is \( U \) (see Sec. III A).
pointed out by Vanević, Nazarov and Belzig, who identified the elementary transport processes in a quantum conductor driven by a time-dependent voltage.\textsuperscript{27,28} Importantly, Abanov and Ivanov have shown recently that for noninteracting electrons in a two-terminal setup, the GF can always be factorized into single-particle events and the zeros of the GF correspondingly lie on the negative real axis.\textsuperscript{29,30} This form of the counting statistics has been dubbed generalized binomial by Hassler et al.\textsuperscript{31} Interactions, however, can change these properties and cause the zeros to move into the complex plane.\textsuperscript{30} As evident from this discussion, the zeros of the GF are of crucial importance for understanding the physical mechanisms that determine the transport statistics. However, given a complicated GF or even actual experimental data, it is not clear how one can tell whether or not the statistics are generalized binomial and if the charges interacted inside the conductor.

Traditionally within FCS, the probability distribution $P(n,t)$ has been characterized by its cumulants, from which one could hope to extract information about the physical processes inside the conductor.\textsuperscript{7} The cumulants are related to derivatives of the GF. Several theoretical studies, however, have found that the high-order cumulants tend to oscillate strongly as some system parameter is varied.\textsuperscript{20,32–37} Recently, this phenomenon has been explained on general grounds and it has been shown that the high-order cumulants for almost any non-Gaussian distribution should oscillate as functions of basically any system parameter, following work by Berry on high derivatives of smooth functions.\textsuperscript{38} The fact that these oscillations are so generic could indicate that the high-order cumulants are fragile objects from which it is difficult to extract information about a particular system: The information in the high-order cumulants seems to be masked by the oscillatory behavior that should be present in almost any system. However, as we explain below, such oscillations do not prevent high-order statistics from bearing information.

It is the purpose of this paper to argue that factorial cumulants provide an alternative characterization of the probability distribution $P(n,t)$ that is particularly useful for describing the high-order statistics of nanoscale transport. In mesoscopic physics, factorial cumulants have so far received only limited attention, except in studies of photons emitted from a quantum point contact\textsuperscript{39} and, very recently, in connection with current fluctuations and entanglement entropy.\textsuperscript{40} However, as we show, the high-order factorial cumulants provide a simple description of generalized binomial statistics and they directly reflect the zeros of the GF. In fact, for noninteracting systems, the high-order factorial cumulants do not oscillate (unlike the ordinary cumulants), no matter what parameter is varied. In contrast, if a high-order factorial cumulant oscillates as a function of some parameter, it cannot be describing generalized binomial statistics and the electrons must have interacted inside the conductor. We illustrate these points with a model of transport through a quantum dot [Fig. 1(b)], for which we show how interactions on the dot cause the zeros of the GF to move into the complex plane as clearly seen in the factorial cumulants.

The paper is organized as follows: In Sec. II we introduce the general framework of FCS, including the GF and the moments and cumulants, as well as their factorial counterparts. We show that the factorial cumulants are particularly simple for generalized binomial statistics, and we give an asymptotic expression for the high-order factorial cumulants, which is directly related to the zeros of the GF. We show that the behavior of the high-order factorial cumulants changes drastically as the zeros move into the complex plane due to interactions. In Sec. III we illustrate our findings using a model of electron transport through a quantum dot, which is weakly coupled to source and drain electrodes. At low bias voltages, the quantum dot can only be empty or singly occupied. This corresponds to the recent experiment by Fricke et al.,\textsuperscript{27} and we show how the measured oscillations of the 15th cumulant as a function of time and coupling to the leads can be explained by the motion of the zeros of the GF. In contrast, the factorial cumulants do not oscillate. As the voltage difference between the leads is increased, the quantum dot can be occupied also by two electrons at a time, and the on-site Coulomb interaction now strongly affects the FCS. The zeros of the GF move into the complex plane, which is clearly visible in the high-order factorial cumulants. Finally, Sec. IV contains our concluding remarks as well as a number of open questions and directions for future research. Appendices A and B, respectively, describe methods for calculating high-order (factorial) moments and cumulants at finite times from a master equation and for extracting the position of the zeros from the high-order factorial cumulants.

II. FULL COUNTING STATISTICS

We consider a generic transport setup in which a nanoscale conductor is connected to source and drain electrodes [Fig. 1(a)]. A bias voltage (possibly time dependent) between the electrodes drives electrons through the conductor, and we denote by $P(n,t)$ the probability distribution of the number of electrons $n$ that have traversed the nanoscale conductor during the time span $[0,t]$. Depending on the direction of the tunneling events between the source and the drain electrodes, the number of transferred electrons $n$ can be either positive or negative. For concreteness we define electron flow into the drain electrode as the positive direction of the current. Examples of nanoscale conductors used in recent counting statistics experiments include tunnel junctions,\textsuperscript{41,42} quantum point contacts,\textsuperscript{43} double quantum dots,\textsuperscript{44} and nanowires,\textsuperscript{45} but for the discussion in this section it is not necessary to specify the details of the system.

A. Generating function

The object of main concern in FCS is the GF, defined as

$$\mathcal{G}(z,t) = \sum_n P(n,t) z^n. \quad (1)$$

The GF encodes the full information about the probabilities $P(n,t)$ and it allows us to introduce (factorial) moments and (factorial) cumulants in the following. As mentioned in Sec. I, calculations of the GF can be approached with several different techniques, depending on the specific system at hand. Interestingly, however, Abanov and Ivanov have recently shown that the counting statistics for a two-terminal scattering problem involving noninteracting electrons can always be decomposed into independent single-particle events for any
form of the scattering matrix and at any temperature.\textsuperscript{29,30}  

Formally, this can be expressed as a factorization of the GF of the form\textsuperscript{29,30}

\[ G(z,t) = \frac{1}{z-0} \prod_{i} G_i(z,t), \]  

where \( G_i(z,t) = 1 - p_i + p_i z \) is the binomial GF corresponding to a single-particle event occurring with probability \( 0 \leq p_i \leq 1 \), depending on time \( t \) as well as all other parameters of the system. The factor \( z^{-0} \) corresponds to a deterministic background charge transfer \( Q = \sum_{j} p_{j} - (n) \geq 0 \) opposite to the positive direction, where \( (n) \) is the mean value of the total transferred charge. For unidirectional transport, we have \( Q = 0 \).

A statistical distribution given by Eq. (2) has been dubbed generalized binomial statistics.\textsuperscript{31} Importantly, the result of Abanov and Ivanov implies that if a GF cannot be factorized as in Eq. (2) with real probabilities \( p_i \), it cannot be describing noninteracting electrons. We note, however, that the opposite is not true: even in the presence of interactions, the statistics may still be generalized binomial.\textsuperscript{30}

B. Moments and cumulants

We now turn to the moments and cumulants of \( P(n,t) \), which are commonly used to characterize the probability distribution. The moments \( \langle n \rangle \) can be found from the moment generating function, which is obtained from the GF [Eq. (1)] via the substitution \( z \to e^{z} \),

\[ M(z,t) = G(e^{z},t) = \sum_{n} P(n,t) e^{zn}. \]  

The moments of the transferred charge are given by the derivatives of the moment generating function with respect to \( z \), evaluated at \( z = 0 \),

\[ \langle n \rangle(t) = \sum_{n} n^{m} P(n,t) = \partial_{z}^{m} M(z,t)|_{z=0}. \]  

The cumulant generating function (CGF) is defined as

\[ S(z,t) = \ln[M(z,t)] = \ln[G(e^{z},t)] \]

and the cumulants are similarly defined as derivatives of the CGF at \( z = 0 \):

\[ \langle n \rangle(t) = \partial_{z}^{m} S(z,t)|_{z=0}. \]  

The first cumulant \( \langle n \rangle = \langle n \rangle \) is the mean of \( n \), the second \( \langle n^2 \rangle = \langle (n) \rangle^2 - \langle n \rangle \) is the variance, and the third \( \langle n^3 \rangle = \langle (n - \langle n \rangle) \rangle^3 \) is the skewness. For a Poisson distribution all cumulants are equal to the mean \( \langle n \rangle \), while only the first and second cumulants are nonzero for a Gaussian distribution, that is, \( \langle n \rangle = 0 \) for \( m \geq 2 \).

C. Factorial moments and factorial cumulants

A complementary characterization of the probability distribution is provided by the factorial moments and the corresponding factorial cumulants.\textsuperscript{56,61} The factorial moments are defined as follows

\[ \langle n \rangle F = (n(n-1) \cdots (n-m+1)). \]  

It is easy to show that they are generated by the function

\[ M_f(z,t) = G(z+1,t) = \sum_{n} P(n,t)(z+1)^n, \]

obtained from the GF [Eq. (1)] via the substitution \( z \to z + 1 \). Thus

\[ \langle n^m \rangle_F(t) = \partial_{z}^{m} M_f(z,t)|_{z=0}, \]

and similarly to the cumulants, the factorial cumulant generating function (FCGF) is defined as

\[ S_f(z,t) = \ln[M_f(z,t)] = \ln[G(z+1,t)], \]

whose derivatives at \( z = 0 \) deliver the factorial cumulants

\[ \langle n^m \rangle_F(t) = \partial_{z}^{m} S_f(z,t)|_{z=0}. \]

The first two factorial cumulants are \( \langle n \rangle_F = \langle n \rangle \) and \( \langle n^2 \rangle_F = \langle (n) \rangle^2 - \langle n \rangle \). For a Poisson distribution, only the first factorial cumulant is nonzero and \( \langle n^m \rangle_F = 0 \) for \( m > 1 \).

The factorial cumulants can be expressed in terms of the ordinary cumulants via the relations

\[ \langle n \rangle_F = \langle n \rangle, \]

\[ \langle n^2 \rangle_F = \langle (n) \rangle - \langle n \rangle, \]

\[ \langle n^3 \rangle_F = \langle (n) \rangle - 3\langle n \rangle + 2\langle n^2 \rangle, \]

which, for arbitrary order \( m \), read\textsuperscript{57}

\[ \langle n^m \rangle_F = \sum_{j=1}^{m} s(m,j)\langle n^j \rangle, \]

where \( s(m,j) \) are the Stirling numbers of the first kind. They can be generated from the relation \( [\ln(1+x)]^{j} = j! \sum_{m=j}^{\infty} s(m,j) x^m / m! \).\textsuperscript{57}

In the case of generalized binomial statistics [Eq. (2)] the expression for the factorial cumulants becomes particularly simple:

\[ \langle n^m \rangle_F = (1)^{m-1}\langle n \rangle |^m - 1 \sum_{i} p_i^{m} - Q. \]  

This expression provides us with a direct test of whether or not a statistical distribution can be factorized into independent single-particle events as described by Eq. (2). In particular, for unidirectional transport (where \( Q = 0 \)), the factorial cumulants must have alternating signs as functions of the cumulant order \( m \) due to the factor \( (1)^{m-1} \), if the statistics is generalized binomial. We remark that it is straightforward to extend this analysis to bidirectional transport. In that case, the quantity \( \langle n^m \rangle_F = (1)^{m-1}(m-1)!\langle n \rangle = (1)^{m-1}(m-1)! \sum_i p_i^{m} - p_i \) has alternating signs as a function of \( m \) for generalized binomial statistics. In this work we use Eq. (14) to test whether the counting statistics of charge transport through a nanoscale conductor is generalized binomial. Importantly, factorial cumulants are measurable and the test is consequently immediately applicable to experimental data.

D. High-order (factorial) cumulants

As the zeros of the GF move into the complex plane due to interactions, the behavior of the high-order factorial cumulants changes compared to that of generalized binomial statistics
The ordinary and the factorial cumulants, respectively. These singularities determine the high-order asymptotics of the ordinary and the factorial cumulants, respectively.

In this work we consider cases for which the (F)CGF has only logarithmic singularities, as is typical at finite times, but in the long-time limit it may have branch-point singularities, as we discuss at the end of this section.

We first note that the CGF (or the FCGF) close to a logarithmic singularity \( z_j \) with degeneracy \( \alpha_j \) behaves as

\[
S_F(z,t) \simeq \alpha_j \ln(z_j - z), \quad z \text{ close to } z_j,
\]

with corresponding derivatives reading

\[
\partial^m z S_F(z,t) \simeq -\alpha_j \frac{(m-1)!}{(z_j - z)^m}, \quad z \text{ close to } z_j.
\]

The high-order derivatives evaluated at \( z = 0 \), that is, the high-order (factorial) cumulants, can then be approximated as a sum over all singularities

\[
\langle \langle n \rangle \rangle_F = \sum_{m=1}^{\infty} \frac{e^{-im \arg [z_0]}}{|z_0|^m}
\]

according to the first Darboux approximation.

Equation (17) shows that the high-order (factorial) cumulants are determined by the singularities closest to \( z = 0 \), which dominate the sum for large \( m \). Relative contributions from other terms are suppressed with the relative distance to \( z = 0 \) and the power \( m \). If the closest (nondegenerate) singularity, denoted \( z_0 \), lies on the negative real axis such that \( z_0 = |z_0|e^{i\pi} \), Eq. (17) becomes particularly simple for large \( m \) and reduces to

\[
\langle \langle n \rangle \rangle_F \rightarrow (-1)^{m-1}(m-1)!|z_0|^m.
\]

This is the case for high-order factorial cumulants corresponding to generalized binomial statistics [Eq. (2)]. For unidirectional transport (\( Q = 0 \)) the corresponding FCGF is

\[
S_F(z,t) \simeq \sum_i \ln (1 + p_i z),
\]

which has logarithmic singularities at \( z_j = -1/p_i \leq -1 \). The high-order factorial cumulants are then dominated by the singularity \( z_{\text{max}} = -1/p_{\text{max}} \) closest to \( z = 0 \), where \( p_{\text{max}} \) is the largest probability among the \( p_i \)’s. We thus find

\[
\langle \langle n \rangle \rangle_F \rightarrow (-1)^{m-1}(m-1)!p_{\text{max}}^m
\]

for large \( m \). For the above example, this conclusion could also have been reached directly from Eq. (14) for \( Q = 0 \).

The singularities, however, do not always lie on the negative real axis, but in general they come in complex-conjugate pairs, ensuring that the (factorial) cumulants are real. In case only a single complex-conjugate pair of logarithmic singularities, \( z_0 \) and \( z_0^* \), is closest to \( z = 0 \), Eq. (17) simplifies to

\[
\langle \langle n \rangle \rangle_F \rightarrow -\frac{2(m-1)!}{|z_0|^m} \cos(m \arg [z_0])
\]

for large \( m \). In the case where \( \arg [z_0] = \pi \), this reduces to the right-hand side of Eq. (18) multiplied by 2, since the two singularities are then degenerate.

Interestingly, our analysis shows that high-order factorial cumulants corresponding to generalized binomial statistics have a sign that is determined solely by the order \( m \) via the factor \((-1)^{m-1}\). In contrast, if the statistics is not generalized binomial, the factorial cumulant of a given order \( m \) will oscillate as a function of any parameter that changes the position of the singularities due to the factor \( \cos (m \arg [z_0]) \), which also causes trigonometric oscillations as function of the order \( m \).

In this section, we have analyzed the situation where the (F)CGF has logarithmic singularities due to zeros of the GF. It is, however, well known that the (F)CGF can have, for example, branch-point singularities in the long-time limit, and a generalization of the above analysis is necessary to treat such cases. The overall conclusions, however, remain intact also for branch-point singularities, and since we mainly consider finite times in this work, we do not encounter such situations.

Instead, we refer the interested reader to Refs. 7, 38, and 48, and, in particular, Sec. IV in Ref. 21, for a more general analysis of high-order derivatives and (factorial) cumulants for (F)CGFs with branch-point singularities.

In the next section, we illustrate how the statistics of charge transport through a quantum dot due to interactions can change from being generalized binomial, with factorial cumulants given by Eqs. (14), to a different statistical distribution, with high-order factorial cumulants governed by Eq. (21).

### III. COULOMB BLOCKADE QUANTUM DOT

#### A. Model

We consider electron transport through a quantum dot (QD) with a single spin-degenerate level coupled to source and drain electrodes [Fig. 1(b)]. The energy of the level is denoted by \( \varepsilon_d \) and \( U \) is the on-site Coulomb interaction. The Hamiltonian for the coupled system reads

\[
\hat{H} = \hat{H}_d + \hat{H}_T + \hat{H}_R,
\]

where

\[
\hat{H}_d = \varepsilon_d (\hat{n}_1 + \hat{n}_1^+) + U \hat{n}_1 \hat{n}_1^+
\]

is the Hamiltonian of the QD, tunneling between the QD and the leads is given by the term

\[
\hat{H}_T = \sum_{a \in S,D} (\varepsilon_a \hat{c}_a^\dagger \hat{c}_a + \text{H.c.}),
\]

and the source (\( \alpha = S \)) and drain (\( \alpha = D \)) electrodes are described as reservoirs of free electrons with energy \( \varepsilon_{a\alpha} \)

\[
\hat{H}_R = \sum_{a \in S,D} \varepsilon_{a\alpha} \hat{c}_a^\dagger \hat{c}_a + \text{H.c.}
\]

Here we have defined the fermionic operators \( \hat{c}_a^\dagger \) (\( \hat{c}_a \)), which create (annihilate) electrons with spin \( \sigma \) on the QD, and the corresponding spin-resolved occupation number operators are \( \hat{n}_\sigma = \hat{c}_a^\dagger \hat{c}_a \sigma \), \( \sigma = \uparrow, \downarrow \). The operators \( \hat{c}_a^\dagger \) and \( \hat{c}_a \) create and annihilate, respectively, electrons with momentum \( k \), spin \( \sigma \), and energy \( \varepsilon_{a\alpha} \) in the source (\( \alpha = S \)) or drain (\( \alpha = D \)).
electrodes. We assume that the tunneling matrix elements $t_{\sigma k}$ are independent of spin and consider the situation without an applied magnetic field. Both of these assumptions can be lifted, although such extensions of the model are not considered here.

In the following, we consider weak coupling between the QD and the leads. Electron transport through the QD can then be described by a master equation for transitions between different many-body eigenstates of the QD. The eigenstates of the isolated QD corresponding to Eq. (23) are $|0\rangle$, $|\uparrow\rangle$, and $|\downarrow\rangle$, where the first and the last eigenstate correspond to the QD being occupied by zero or two electrons, respectively, while the other two correspond to the QD being occupied by a single electron with spin $\sigma = \uparrow, \downarrow$, respectively. We now define the $n$-resolved probabilities $p_0(n,t), p_1(n,t), p_1(n,t)$, and $p_2(n,t)$ for each of the eigenstates to be occupied, while $n$ electrons have been collected in the drain during the time span $[0,t]$. Since tunneling is spin independent, it is equally probable to occupy each single-electron spin state, $p_1(n,t) = p_1(n,t)$, and we can define $p_2(n,t) = p_1(n,t) + p_1(n,t)$ and collect the probabilities in the vector

$$|\rho(n,t)\rangle = [p_0(n,t), p_1(n,t), p_2(n,t)]^T,$$

where we use double-brackets to avoid confusion with the quantum states of the Hamiltonian. We also define $\langle \rho \rangle = [1,1,1]$, allowing us to express the probability $P(n,t)$ as the inner product $P(n,t) = \langle \rho | \rho(n,t) \rangle$. The GF is then

$$\mathcal{G}(z,t) = \langle \rho | e^{\mathcal{M}(z \tau)} | \rho \rangle,$$

(27)

where $|g(z,t)\rangle = \sum_p e^{i \epsilon_p t} | p(n,t) \rangle$. We note that $|g(1,t)\rangle = |p_0(t), p_1(t), p_2(t)\rangle$ contains the probabilities $p_i$ of occupying the QD with $i = 0, 1, 2$ electrons independently of the number of transferred electrons $n$. As a consequence of probability conservation, we have $\mathcal{G}(1,t) = \langle \rho | g(1,t) \rangle = \sum_p P(n,t) = 1$ for all $t$.

The time dependence of the GF is determined by the dynamics of $|g(z,t)\rangle$. We find the time evolution of $|g(z,t)\rangle$ by setting up a master equation for the transitions between the eigenstates of the QD. In the weak coupling regime, the transition rates can be found using Fermi’s Golden rule, treating the tunneling Hamiltonian in Eq. (24) as the perturbation. Working in the wide-band limit and assuming a constant tunneling density of states in both leads, we define the bare energy-independent tunneling rates $U_\alpha = \frac{\pi}{4} \hbar |a_\alpha|^2 D_\alpha$, where $D_\alpha$ is the density of states in lead $\alpha = S, D$. The master equation for $|g(z,t)\rangle$ then reads

$$\partial_t |g(z,t)\rangle = \mathcal{M}(z) |g(z,t)\rangle,$$

(28)

with the $z$-dependent rate matrix

$$\mathcal{M}(z) = \begin{bmatrix} -2(\Gamma_1 S_0 + \Gamma_2 n_0^{(0)}) & z \Gamma_D (1 - n_0^{(0)}) + \Gamma_3 (1 - n_S^{(0)}) & 0 \\ 2(\Gamma_1 S_0 + z^{-1} \Gamma_D n_0^{(0)}) & -\Gamma_D (1 + n_D^{(0)} - n_0^{(0)}) - \Gamma_3 (1 + n_D^{(0)} - n_S^{(0)}) & 2 \Gamma_3 (1 - n_D^{(0)}) + z \Gamma_D (1 - n_D^{(0)}) \\ 0 & z^{-1} \Gamma_D n_D^{(0)} + \Gamma_3 S_0^{(0)} & -2 \Gamma_3 (1 - n_D^{(0)}) + \Gamma_D (1 - n_D^{(0)}) \end{bmatrix}.$$

Here, we have introduced the Fermi functions of the leads, kept at electron temperature $T_\alpha$ and chemical potential $\mu_\alpha$, evaluated at the energy $e_d + E$

$$n_\alpha^{(E)} = \frac{1}{e^{(e_d + E - \mu_\alpha)/k_B T_\alpha} + 1}, \quad \alpha = S, D.$$  

(29)

In the rate matrix above, only $n_0^{(0)}$ and $n_D^{(0)}$ appear, which we use to parametrize the applied voltage biases and the temperatures of the electrodes in the following subsection. We note that the off-diagonal elements of $\mathcal{M}(z)$ include factors of $z$ and $z^{-1}$ multiplying the rates corresponding to processes that increase or decrease, respectively, by 1 the number of electrons that have been collected in the drain.18

We find the GF by formally solving Eq. (28) for $|g(z,t)\rangle$. To this end, we need to define an appropriate initial condition. We assume that the occupations of the QD eigenstates have reached their steady state at $t = 0$ when counting begins and $P(n,t = 0) = \delta_{n,0}$. We then have $|g(z,t = 0)\rangle = |0\rangle$, where the stationary state $|\rho\rangle$ of the QD is given by the unique solution to $\mathcal{M}(z) |\rho\rangle = 0$. We thereby obtain the following compact expression for the GF:

$$\mathcal{G}(z,t) = \langle \rho | e^{\mathcal{M}(z \tau)} | \rho \rangle.$$

(30)

This is a general and formally exact result, but in practice, given a rate matrix $\mathcal{M}(z)$, it may not be possible to obtain a simple, closed-form expression for the GF. Further complications arise when trying to calculate (factorial) moments and (factorial) cumulants, since derivatives of the GF with respect to $z$ are required. To calculate high-order (factorial) cumulants at finite times we have thus developed the method described in Appendix A.

B. Results

We concentrate on the situation where the leads are voltage biased such that the energy level of the QD is well above the chemical potential of the drain, $e_d \gg \mu_D$ or $n_D^{(0)} = n_D^{(0)} \approx 0$, and electrons cannot tunnel back into the QD from the drain. At the same time, the level is below the chemical potential of the source electrode, $e_d \ll \mu_S$ or $n_S^{(0)} \approx 1$. Under these voltage conditions, electron transport takes place from source to drain via the QD. The single-occupied state of the QD always participates in transport, but the doubly occupied state only becomes populated for nonzero values of $n_S^{(U)}$. In the following we study the charge transport statistics as a function of $n_S^{(U)}$, that is, we vary the bias in the source electrode such that $n_S^{(U)}$ takes values between 0 and 1.

1. Low bias

We first analyze the low-bias regime $n_S^{(U)} \approx 0$. This corresponds to the situation recently investigated in the experiments
FIG. 2. (Color online) High-order cumulants in the low-bias regime, $n_S^{(3)} \simeq 0$. (a) The cumulant $\langle \langle n^{15} \rangle \rangle$ as a function of the asymmetry $a$ [Eq. (32)] and the dimensionless time $\tau = 2\Gamma_S t$. The high-order cumulant oscillates as a function of both parameters, in agreement with the experimental results of Fricke et al.¹ (Note: We do not normalize our results by $1/39$.) The figures below and next to the contour plot show results along the orange lines $\tau = 3$ and $a = 0.6$ in the contour plot. Solid lines are numerical results, while open circles correspond to approximation (17) taking into account the three pairs of singularities indicated by filled red circles in (b). (b) Singularities of the CGF in the complex plane for $\tau = 3$ and $a = 0.6$. The singularities are $2\pi$-periodical along the imaginary axis. Filled circles indicate numerical results obtained from Eq. (37), while open red and green circles show approximations (38) and (39), respectively. Filled red circles indicate the three pairs of singularities entering Eq. (17), as shown by open circles in (a). With time the singularities move toward the points indicated by filled (black) squares.

having introduced

$$\eta(z) = \sqrt{a^2 + z(1 - a^2)}. \quad (35)$$

In Eq. (34) we have corrected for a missing factor of $4\eta(z)$ in the Supporting Information in Ref. 7. This factor ensures that the GF is an analytic function of $z$, since $-\eta(z)$ is the analytic continuation of $\eta(z)$ across the branch cut and $G(z, \tau)$ is invariant under the substitution $\eta \rightarrow -\eta$. Solving next for the zeros $z_k$ of the GF, we find

$$z_k = -\frac{h_k^2 + a^2}{1 - a^2}, \quad k = 1, 2, \ldots. \quad (36)$$

where $h_k$ is determined by the transcendental equation

$$h_k \tau = (1 + a)(k\pi - 2\arctan h_k), \quad k = 1, 2, \ldots. \quad (37)$$

By graphical inspection of the transcendental equation we find that the solutions $h_k$ are real and positive for all $k = 1, 2, \ldots$. Therefore, all zeros [Eq. (36)] are real and negative, since $0 \leq a^2 \leq 1$. Remarkably, the positions of the zeros agree with the general statements by Abanov and Ivanov for noninteracting electrons. This can be understood by noting that the master equation [Eq. (33)] can also describe noninteracting spinless electrons tunneling through a resonant level for which the statements by Abanov and Ivanov directly apply.

In the following, we solve the transcendental equation (37) numerically. However, in two limiting cases, it can be solved analytically: For any fixed $k$, we can choose a sufficiently
FIG. 3. (Color online) High-order factorial cumulants in the low-bias regime, \( n_k^{(1)} \approx 0 \). (a) The factorial cumulant \( \langle n_{15} \rangle_F \) as a function of the dimensionless time \( \tau = 2\Gamma \Delta \) and the asymmetry \( a \) [Eq. (32)]. Parameters are \( a = 0.6 \) (upper plot) and \( \tau = 3 \) (lower plot), corresponding to the orange lines in Fig. 2(a). Solid lines are numerical results, while open circles correspond to approximation (17) taking into account the three singularities indicated by filled red circles in (b). (b) Singularities of the FCGF in the complex plane for \( \tau = 3 \) and \( a = 0.6 \). Filled red circles indicate the three singularities entering Eq. (17) as shown by open circles in (a). With time the singularities move toward the point indicated by the filled (black) square. Singularities lie on the negative real axis, and the factorial cumulants do not oscillate as functions of either time or asymmetry.

large \( \tau \) such that \( h_k \) must be small and \( \arctan h_k \approx h_k \). We then obtain

\[
h_k \approx \left( \frac{1 + a}{1 + a + \frac{\pi}{2}} \right)^2 \frac{\pi}{2}, \quad k \text{ fixed}, \quad \tau \text{ large}. \tag{38}
\]

In the other limiting case, we fix the time \( \tau \) and consider a large \( k \), such that \( h_k \) must be large and \( \arctan h_k \approx \pi / 2 \). We then find

\[
h_k \approx \left( \frac{1 + a}{\tau} \right)^2 \pi (k - 1), \quad \tau \text{ fixed}, \quad k \text{ large}. \tag{39}
\]

From the zeros of the GF we can understand the behavior of the high-order (factorial) cumulants. Analyzing first the ordinary cumulants, we note that the CGF has logarithmic singularities at \( \ln |z_k| + i \pi (2l + 1), \ l \in \mathbb{Z} \), corresponding to the zeros \( z_k \) of the GF. In Fig. 2(b) we show the positions of these singularities obtained from numerical solutions of the transcendental equation [Eq. (37)]. For comparison, we also show the limiting cases, Eqs. (38) and (39), which are in good agreement with the numerical results. In Fig. 2(a), we show numerically exact results together with the asymptotic expression [Eq. (17)], taking into account the three pairs of complex-conjugate singularities that are closest to \( z = 0 \) [indicated by filled red circles in Fig. 2(b)]. The agreement is good and the analysis provides us with a simple interpretation of the experimental data from Ref. 9: The motion of the singularities in the complex plane as functions of the dimensionless time \( \tau \) and the asymmetry \( a \) cause the oscillations of the high-order cumulants observed in our numerical calculations and in the experiment.

We next turn to the corresponding factorial cumulants. Numerical results for the factorial cumulant of order \( m = 15 \) as a function of the dimensionless time \( \tau \) and the asymmetry \( a \) are shown in Fig. 3(a), and clearly no oscillatory behavior is observed. Again, this can be understood by considering the logarithmic singularities of the FCGF. The FCGF has logarithmic singularities at \( z_k - 1 \) corresponding to the (negative) zeros \( z_k \) of the GF. Since the singularities of the FCGF are real, the factorial cumulants do not oscillate as functions of either \( \tau \) or \( a \) [according to Eq. (18)]. In Fig. 3(b) we show the positions of the singularities, and in Fig. 3(a) we show the asymptotic expression [Eq. (17)], taking into account the three singularities that are closest to \( z = 0 \) [indicated by filled red circles in Fig. 3(b)]. The asymptotic expression fully accounts for the numerical results.

Before closing this part, we briefly discuss the motion of the singularities at long times. As indicated in Figs. 2(b) and 3(b), the singularities of the CGF and the FCGF all move toward the points marked by filled black squares. According to Eq. (36), the zeros of the GF behave as \( z_k \to a^2/(a^2 - 1) \lesssim 0 \) for long times \( \tau \), since then \( h_1 \approx 0 \) in Eq. (38). The points marked by filled black squares in Figs. 2(b) and 3(b) are thus \( \ln [a^2/(a^2 - 1)] \) (for the CGF) and \( a^2/(a^2 - 1) \approx 1/(a^2 - 1) \) (for the FCGF), respectively. Interestingly, the point \( z = a^2/(a^2 - 1) \) corresponds to the square-root branch point of the function \( \eta(z) \) in Eq. (35): In the long-time limit, the logarithm of the GF goes as \( \ln G(z, \tau) \to \tau \eta(z - 1)/(1 + a) \) according to Eq. (34), and the singularities of the CGF and the FCGF are then determined by the branch points of \( \eta(z) \). Thus, in this example, all logarithmic singularities of the CGF and the FCGF move toward particular points in the complex plane, which in the long-time limit become branch-point singularities.

2. Finite bias

In the general case of a finite bias, \( 0 < n_k^{(1)} < 1 \), the on-site Coulomb interaction strongly influences the charge transport statistics and we expect that the statistics will no longer be generalized binomial with all zeros of the GF lying on the
we show results for the factorial cumulants to second order. In Fig. 4, for which the dominant singularity $z_0 \simeq -8.2$ of the FCGF $G(z + 1, t)$ is real and negative. Factorial cumulants have been rescaled as $\langle \langle n^m \rangle \rangle_F \cdot |z_0|^m / (m - 1)!$. Numerical results (filled squares) follow the predicted behavior for high-order factorial cumulants given by Eq. (18) (open circles).

The change of statistics is directly reflected in the high-order factorial cumulants. In Fig. 5 we show the factorial cumulants $\langle \langle n^m \rangle \rangle_F$ as functions of their order $m$ with parameters for which the dominant singularity is still real and negative, such that the high-order factorial cumulants should follow Eq. (18). In Fig. 5 we have rescaled the factorial cumulants as $\langle \langle n^m \rangle \rangle_F \cdot |z_0|^m / (m - 1)!$, where $z_0$ is the singularity closest to $z = 0$, and for high orders they clearly follow the predicted pattern with alternating signs due to the factor $(-1)^{m-1}$ in Eq. (18). The asymptotic behavior of the rescaled high-order factorial cumulants (shown by open circles) agrees well with our numerical results (filled squares). From the fifth factorial cumulant and onward, the asymptotic expression completely accounts for our numerical results.

In Fig. 6 we show results for the factorial cumulants with parameters for which the dominant singularity has moved into the complex plane and the statistics is no longer generalized binomial. The high-order factorial cumulants are now governed by a pair of complex-conjugate singularities and are thus expected to follow Eq. (21). In Fig. 6 we have rescaled the factorial cumulants as $\langle \langle n^m \rangle \rangle_F \cdot |z_0|^m / (m - 1)!$, where $z_0$ and $z_0^*$ are the closest complex-conjugate pair of singularities. In this case, we see clear trigonometric oscillations as a function of the order $m$ due to the factor $\cos(m \arg(z_0))$ with a frequency determined by $\arg(z_0)$. The asymptotic expression (open circles) accounts well for our numerical results for the factorial cumulants (filled squares) already from the third order onward. We stress that this oscillatory behavior of the factorial cumulants would not be possible in a noninteracting system for which all singularities would lie on the negative real axis such that $\arg(z_0) = \pi$ and $\cos(m \arg(z_0)) = (-1)^{m-1}$.

For the results presented in Figs. 4 to 6 we found numerically the logarithmic singularities of the FCGF. In an actual experiment, however, the GF would typically not be known, and it is therefore relevant to ask if the position of the singularities can be deduced from the measured high-order factorial cumulants alone. In Appendix B we describe a simple method for extracting the position of a dominant pair of complex-conjugate singularities from four consecutive high-order factorial cumulants. As illustrated in Fig. 7, we find excellent agreement between our numerical results and the positions obtained directly from the high-order cumulants (up to order $m = 25$) using this method. The figure shows results in the full range of bias voltages from $n_s^{(U)} = 0$ to $n_s^{(U)} = 1$. For low bias voltages $n_s^{(U)} \simeq 0$, the dominant singularity is real and negative, and as $n_s^{(U)}$ is increased, two complex-conjugate singularities move into the complex plane, showing that the statistics is no longer generalized binomial. Finally, in the high-bias limit $n_s^{(U)} = 1$, the singularities move back onto the real axis, and the statistics is again generalized binomial. In the high-bias regime, the interaction strength $U$ drops out of the problem and transport takes place via the two parallel and perpendicular channels.
of the zeros. We expect that the motion of zeros into the complex plane due to interactions will be experimentally detectable using available measurement techniques.

Our work leaves a number of open questions for future research. It would be interesting to understand physically the exact point at which interactions cause the zeros of the GF to become complex. We have considered only two-terminal devices, and it might be possible to generalize our findings to multilead setups.31

IV. CONCLUSIONS

We have shown that factorial cumulants are useful for detecting interactions among electrons passing through a nanoscale device. For noninteracting electrons in a two-terminal conductor, the counting statistics is always generalized binomial, as recently found by Abanov and Ivanov.29,30 The charge transport statistics can be factorized into single-particle transfer events and the zeros of the GF are correspondingly real and negative. Interactions among the electrons, however, can drive the zeros of the GF away from the negative real axis and into the complex plane.30 As we have shown, this change of the statistics is clearly visible in the factorial cumulants. For generalized binomial statistics, the factorial cumulants have a sign that is determined by the cumulant order only. In contrast, as the zeros of the GF move into the complex plane due to interactions, the factorial cumulants oscillate as functions of basically any parameter.

To illustrate our findings, we have considered transport through a quantum dot weakly coupled to source and drain electrodes. At low bias voltages, the dot can only be empty or singly occupied. This corresponds to recent experiments3 for which we reproduce and explain the measured oscillations of the high-order cumulants by considering the zeros of the GF. In this case, the statistics is generalized binomial and the high-order factorial cumulants do not oscillate. As the bias voltage is increased an additional electron can occupy the dot. The on-site interaction now strongly affects the counting statistics, and the zeros of the GF move into the complex plane. As we have shown, this is clearly visible in the high-order factorial cumulants, which also allow us to locate the positions of the zeros. We expect that the motion of zeros into the complex plane will be experimentally detectable using available measurement techniques.

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APPENDIX A: CALCULATION OF (FACTORIAL) CUMULANTS AT FINITE TIMES

Here we describe our method for calculating (factorial) cumulants at finite times without explicitly evaluating the (F)CGF. We begin with a master equation of the form given by Eq. (28),

$$\partial_t |g(z,t)\rangle = M(z) |g(z,t)\rangle,$$

(A1)

for a system with N states. Next, we substitute in the master equation $|g(z,t)\rangle$ by $|\hat{g}(z,t)\rangle$ and $M(z)$ by $\hat{M}(z)$, where $|\hat{g}(z,t)\rangle = |g(e^{z},t)\rangle$ and $\hat{M}(z) = M(e^{z})$ for ordinary cumulants and $|\hat{g}(z,t)\rangle = |g(z+1,t)\rangle$ and $\hat{M}(z) = M(z+1)$ for factorial cumulants, respectively. For $z = 0$, the master
equation then reads
\[ \partial_t |\tilde{z}(0,t)\rangle = \tilde{M}(0)|\tilde{z}(0,t)\rangle, \tag{A2} \]

having defined \(|\tilde{z}(0,t)\rangle = |\tilde{z}(0)\rangle\) and \(\tilde{M}(0) = \tilde{M}(0)\). Taking instead \(m = 1, 2, \ldots, k\) consecutive derivatives with respect to \(z\) (evaluated at \(z = 0\)), we obtain
\[ \partial_z |\tilde{z}(1,t)\rangle = \tilde{M}(1)|\tilde{z}(1,t)\rangle, \]
\[ \partial_z |\tilde{z}(2,t)\rangle = \tilde{M}(2)|\tilde{z}(2,t)\rangle + 2\tilde{M}(1)|\tilde{z}(1,t)\rangle + \tilde{M}(0)|\tilde{z}(0,t)\rangle, \]
\[ \vdots \]
\[ \partial_z |\tilde{z}(k,t)\rangle = \sum_{j=0}^{k} \binom{k}{j} \tilde{M}(j)|\tilde{z}(j,t)\rangle, \tag{A3} \]

where we have introduced the notation \(|\tilde{z}(m,t)\rangle = \partial_z^m |\tilde{z}(z,t)\rangle|_{z=0}\) and \(\tilde{M}(m) = \partial_z^m \tilde{M}(z)|_{z=0}\). The (factorial) moments of order \(m \leq k\) are then
\[ \langle \tilde{z}^m(t) \rangle = \langle \tilde{z}(m,t) \rangle = \langle \tilde{z}(m) \rangle = \langle \tilde{z}(m)|_{z=0} \rangle, \tag{A4} \]

depending on the substitutions made above. The vector \(\tilde{z}(0) = [1, 1, \ldots, 1]\) contains \(N\) elements equal to unity.

We solve the system of coupled equations (A2)-(A3) by introducing the auxiliary vector
\[ \langle G(t) \rangle = [\langle \tilde{z}(t) \rangle, \langle \tilde{z}(1,t) \rangle, \ldots, \langle \tilde{z}(k,t) \rangle]^T, \tag{A5} \]

containing \(N(k+1)\) elements. The equation of motion for \(\langle G(t) \rangle\) reads
\[ \partial_t |G(t)\rangle = \tilde{M} |G(t)\rangle, \tag{A6} \]

where, according to Eqs. (A2) and (A3),
\[ \tilde{M} = \begin{bmatrix} \tilde{M}(0) & 0 & 0 & 0 & 0 \\ \tilde{M}(1) & \tilde{M}(0) & 0 & 0 & 0 \\ \tilde{M}(2) & 2\tilde{M}(1) & \tilde{M}(0) & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \ddots \\ \tilde{M}(k) & n\tilde{M}(k-1) & n\tilde{M}(k-2) & \cdots & \tilde{M}(0) \end{bmatrix} \tag{A7} \]
is a matrix of dimensions \(N(k+1) \times N(k+1)\). We proceed by solving Eq. (A6) as
\[ |G(t)\rangle = e^{\tilde{M}t} |G(t = 0)\rangle. \tag{A8} \]

Here, the initial condition as counting begins reads
\[ |G(t = 0)\rangle = [\langle \tilde{z}(0) \rangle, \langle \tilde{z}(1) \rangle, \ldots, \langle \tilde{z}(k) \rangle]^T, \tag{A9} \]

and contains the stationary state \(|\tilde{z}(0)\rangle\), which solves \(\tilde{M}|\tilde{z}(0)\rangle = 0\), followed by \(N\) elements equal to 0. Even for large dimensions of \(\tilde{M}\) we may calculate numerically the matrix exponentiation \(e^{\tilde{M}t}\) for a given time \(t\) and obtain \(|G(t)\rangle\) via Eq. (A8). Having determined the (factorial) moments using Eq. (A4), the corresponding (factorial) cumulants are obtained via the relation\(^{27}\)
\[ \langle \langle n^m \rangle \rangle_{(F)} = \langle \langle n^m \rangle \rangle_{(F)} - \sum_{k=1}^{m-1} \binom{m-1}{k} \langle \langle n^k \rangle \rangle_{(F)} \langle \langle n^{m-k} \rangle \rangle_{(F)}. \tag{A10} \]

For the particular \(N = 3\) state model studied in this work, we could easily calculate the first \(m = 50\) (factorial) cumulants at finite times.

APPENDIX B: DETERMINATION OF A PAIR OF COMPLEX-CONJUGATE SINGULARITIES

A pair of dominant, complex-conjugate logarithmic singularities \(z_0^*\) and \(z_0^*\) can be extracted from four consecutive high-order factorial cumulants using methods from Refs. 21 and 52, which we repeat here for completeness. Using Eq. (21) we obtain the matrix equation
\[ \begin{bmatrix} 1 & -\frac{1}{m-1} \frac{\langle \langle n^{m-1} \rangle \rangle_{(F)}}{\langle \langle n^1 \rangle \rangle_{(F)}} & \cdots & -\frac{1}{m-1} \frac{\langle \langle n^{m-1} \rangle \rangle_{(F)}}{\langle \langle n^1 \rangle \rangle_{(F)}} \\ \vdots & \ddots & \ddots & \ddots \\ -\frac{1}{m-1} \frac{\langle \langle n^1 \rangle \rangle_{(F)}}{\langle \langle n^{m-1} \rangle \rangle_{(F)}} & \cdots & \cdots & \frac{2\Re(z_0)}{\langle \langle n^1 \rangle \rangle_{(F)}} \end{bmatrix} = \begin{bmatrix} (m-3) \frac{\langle \langle n^{m-3} \rangle \rangle_{(F)}}{\langle \langle n^{m-3} \rangle \rangle_{(F)}} \\ \vdots \\ (m-2) \frac{\langle \langle n^{m-2} \rangle \rangle_{(F)}}{\langle \langle n^{m-2} \rangle \rangle_{(F)}} \end{bmatrix}, \tag{B1} \]

which we solve for \(\Re(z_0)\) and \(|z_0|^2\). Typically the accuracy of the method increases with the cumulant order \(m\).\(^{52}\)

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FACTORIAL CUMULANTS REVEAL INTERACTIONS IN . . .

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2.3 Multi-level Coulomb blockade quantum dot

In this section we consider a model of a Coulomb blockade QD with two levels that was discussed by Belzig in [71]. The situation is readily generalized to the multi-level case with two groups of levels [71]. The model features transport in avalanches of electrons. A similar situation, however for the transport of pairs of electrons, is observed in the experiment in Ch. 3.

2.3.1 Model calculations

![Diagram of Coulomb blockade quantum dot with two energy levels inside the bias window. The situation is readily generalized to the multi-level case with two groups of levels [71].](image)

Figure 2.1: Coulomb blockade quantum dot with two energy levels inside the bias window. The situation is readily generalized to the multi-level case with two groups of levels [71].

The model we look at is sketched in Fig. 2.1. The QD contains two levels inside the bias window $\mu_S - \mu_D$. It is operated close to a charge degeneracy point in order to allow transport and we consider the strong Coulomb blockade regime, such that at most one extra electron can occupy the dot. Since there is no relaxation between the levels, the occupation of one level blocks transport through the other level. Moreover, the chemical potential in the drain reservoir, $\mu_D$, lies in the vicinity of the lower level, $\varepsilon_-$, that is $|\varepsilon_- - \mu_D| \sim k_B T_D$, implying that electrons can tunnel from the lower level to the drain reservoir as well as in the reverse direction. We assume weak coupling and denote the bare tunneling rates on the source (S) and drain (D) sides by $\Gamma_S, \Gamma_D$, respectively. Since at $\varepsilon_-$ the drain reservoir states are partially filled, we define the parameter

$$x = 1 - n_F^{(D)}(\varepsilon_-), \quad (2.5)$$

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Figure 2.2: The factorial cumulants for the two-level QD wildly oscillate for most of the parameter space spanned by \( a \) and \( x \). In (a) we show the 10th factorial cumulant in the long-time limit, rescaled by the mean current, as a function of \( x \) [Eq. (2.5)]. The oscillations are a consequence of the motion of the singularities of the factorial CGF in the complex plane, as shown in (b). We set \( \Gamma_S = 9.5\Gamma_D \), that is, \( a = -0.9 \). In (b) \( x \) is varied in steps of 0.05.

where \( n_F^{(D)}(\varepsilon) = \left[ 1 + e^{(\varepsilon-\mu_D)/k_BT_D} \right]^{-1} \) is the Fermi distribution function of the drain reservoir. With these assumptions we can write a master equation of the form (1.44) with the kernel

\[
M(z) = \begin{bmatrix}
-\{(1 - x)\Gamma_D + 2\Gamma_S\} & x\Gamma_D & \Gamma_D \\
z\Gamma_S + (1 - x)\Gamma_D & -x\Gamma_D & 0 \\
z\Gamma_S & 0 & -\Gamma_D
\end{bmatrix}.
\tag{2.6}
\]

We analyze the transport statistics as a function of \( x \), that is, of bias, and as a function of the asymmetry\(^1\)

\[
a = \frac{\Gamma_D - 2\Gamma_S}{\Gamma_D + 2\Gamma_S}.
\tag{2.7}
\]

The analysis is done at finite times as well as in the long-time limit. Furthermore, we analyze the statistics as a function of time. We have used the methods of App. A.1, A.2, and A.3.

Belzig has shown in Ref. [71] that there is an interesting ringing behavior in this model: for small \( x \) the lower level may be occupied and then block transport

\(^1\)The asymmetry varies between \(-1\) for \( \Gamma_S \gg \Gamma_D \) and \( 1 \) for \( \Gamma_S \ll \Gamma_D \). The factor of two in front of \( \Gamma_S \) has been introduced since at \( x = 1 \) the dot is filled at the rate \( 2\Gamma_S \), but as the occupation of one level blocks the other level, the dot is emptied at the rate \( \Gamma_D \).
through the upper level. However, with a small rate \( x \Gamma_D \) the lower level is emptied and \( n \) consecutive electrons can tunnel through the upper level before the lower level is occupied again and blocks transport. For small \( x \) and \( \Gamma_S \gg \Gamma_D \) one shows [71] that the transport statistics is composed of the transfer of avalanches of electrons through the upper level, of different sizes \( n \), which gives rise to a large Fano factor [Eq. (1.26)] as well as oscillating factorial cumulants. For \( x \)-values that are not small the physical interpretation is less simple, however the interaction effects are as well visible in the statistics.

For \( x = 1 \) the two channels for transport through the upper and lower levels are equivalent and we do not expect to detect the presence of two dot levels from the transport statistics. In fact in this case the system is equivalent to a two-state system, for which the statistics is generalized binomial as discussed in Sec. III B of publication (i) [Sec. 2.2]. On the other hand, for \( x = 0 \) the lower level blocks transport and we also do not expect to see any interaction effects in the statistics. The interesting effects are therefore expected for \( x \in (0,1) \).

Let us now go on to the analysis. First, we consider the situation where the drain bias is varied, that is, \( x \) varies between 0 and 1. Fig. 2.2 a shows the 10\(^{th}\) factorial cumulant in the long-time limit for \( \Gamma_S = 9.5 \Gamma_D \). We observe strong oscillations that are due to the motion of the singularities of the factorial CGF [Eq. (1.21)] in the complex plane. This motion is shown in Fig. 2.2 b where the
dominant pair of complex conjugate singularities is displayed for \( x \) varying between 0 and 1. Fig. 2.2 clearly shows the signatures of the interactions that are present. In his paper [71] Belzig considers the limit of small \( x \) and finds the cumulants for \( x \to 0 \). We remark that this analysis breaks down immediately as \( x \) grows from 0 due to the violent oscillations. The latter are present in the factorial cumulants in most of the parameter space spanned by \( a \) and \( x \), as shown in Fig. 2.3.

Fig. 2.3 shows a “phase diagram” for the parameter space spanned by \( a \) and \( x \) in the long-time limit. In region 1 (pink) we observe oscillations in the factorial cumulants, whereas in region 2 (blue) the statistics is generalized binomial. This illustrates the fact that the statement of our main result cannot be turned around, that is, even in the presence of interactions the statistics can still be generalized binomial. Fig. 2.3 has been obtained by analyzing the long-time limit factorial CGF and its square root branch point singularities. The line \( x = x_C \) with

\[
x_C = \frac{1}{54 \Gamma_S \Gamma_D^2} \left[ -4 \Gamma_S^3 + 24 \Gamma_S^2 \Gamma_D + 33 \Gamma_S \Gamma_D^2 + 5 \Gamma_D^3 - (\Gamma_S + \Gamma_D) \sqrt{(2 \Gamma_S - \Gamma_D)^3 (2 \Gamma_S - 25 \Gamma_D)} \right]
\]  

(2.8)

delimits regions with a pair of complex conjugate singularities and a real singularity, respectively.

We still have a similar situation at finite times. Fig. 2.4 shows the 10th factorial cumulant as a function of time for a given asymmetry, \( a = 1/3 \), and two different parameters \( x \). At \( x = 0.1 \) the 10th factorial cumulant features oscillations whereas at \( x = 0.9 \) it does not, showing that also at finite times there are cases where we do or do not see oscillations in the high-order factorial cumulants, in analogy to the long-time limit situation analyzed in Fig. 2.3.

Finally, for a comparison of high-order factorial cumulants at transient, large and infinite time we refer to Fig. 2 in publication (ii) [Sec. 2.3.2], which shows the factorial cumulants of order 13 through 15 as functions of the parameter \( x \) [Eq. (2.5)] for the three different times. We see that at large finite times the situation is similar as in the long-time limit, reflecting the continuity in time. Also, for large \( x \)-values there are no more oscillations, suggesting that at finite times we have a similar situation as in Fig. 2.3 that is, also at finite times there is a blue region where the statistics is generalized binomial.

To summarize, we have observed that the interactions in this two-level QD clearly leave their signature in the factorial cumulants, which oscillate in most

\footnote{The usual cumulants oscillate for any \( a \) and \( x \).}
Figure 2.4: High-order factorial cumulant as a function of time. The 10th factorial cumulant is rescaled with the mean charge and plotted as a function of time for two different $x$-values. The logarithm is taken so as to make the oscillations more visible. The red parts correspond to positive values of $\langle n^{10} \rangle_F/\langle n \rangle$ and the blue parts to negative values. At $x = 0.1$ the 10th factorial cumulant features oscillations, whereas at $x = 0.9$ it does not.

Moreover, we observe from Fig. 2 in publication (ii) [Sec. 2.3.2] that our analysis can be done equally well at finite times as in the long-time limit. Also, we have seen in Fig. 2.4 that high-order factorial cumulants may oscillate as functions of time for short and transient times, before the long-time limit is reached where the (factorial) cumulants become linear in time. This can be of use experimentally since short and transient time statistics are more accessible.
2.3.2 Oscillating factorial cumulants in counting statistics are due to interactions: publication (ii)

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Abstract. We discuss our recent theoretical proposal to detect interactions in the electron transport through a nano-scale system by measuring high-order factorial cumulants of the full counting statistics. Our proposal is based on theoretical studies which have demonstrated that the zeros of the generating function for the full counting statistics are always real and negative for non-interacting electrons in a two-terminal scattering setup. As we have shown, this implies that the factorial cumulants do not oscillate as functions of any system parameter. Interactions, however, can cause the zeros to move away from the negative real axis into the complex plane. This transition is clearly visible in the factorial cumulants which start oscillating. We illustrate our findings with a model of transport through a two-level Coulomb blockade quantum dot, which we analyze both for finite times and in the long-time limit, and we discuss possible experimental implementations to test our predictions.
so-called factorial cumulants are in fact very useful to extract physical information about the charge transfer process [13]. Briefly, the factorial cumulants do not oscillate for non-interacting electrons in a two-terminal scattering problem. In contrast, if a factorial cumulant oscillates as a function of some parameter it must be due to interactions among the electrons passing through the conductor.

Our finding relies on a deeper mathematical analysis of the generating function for the counting statistics by Abanov and Ivanov who showed that its zeros must always be real and negative for non-interacting electrons in a two-terminal configuration [16, 17]; the multi-terminal problem is analyzed in Ref. [18]. Only in the presence of interactions the zeros may move away from the negative real axis into the complex plane. As we have shown, this transition is clearly visible in the factorial cumulants, which start oscillating when the zeros of the generating function become nonreal. Importantly, factorial cumulants are measurable in Coulomb blockade quantum dots using currently available experimental techniques [7, 8, 9, 10, 11], thus enabling an experimental test of our predictions.

In the following we give a short introduction to FCS, including the basic definitions of moments and cumulants as well as their factorial counterparts. We discuss why the factorial cumulants do not oscillate for non-interacting electrons in a two-terminal scattering problem. As an illustrative example, we then consider the FCS of transport through an interacting two-level quantum dot for which the factorial cumulants clearly oscillate due to interactions. In our recent work we only analyzed the FCS for finite times [13], but here we explicitly show that our findings also hold in the long-time limit. Finally, we present our concluding remarks.

2. Full counting statistics

The central object in FCS is the probability distribution $P(n, t)$ of having transferred $n$ charges through a nano-scale system during the time span $[0, t]$ together with the corresponding generating function

$$G(z, t) = \sum_{n} P(n, t) z^n.$$ (1)

The generating function provides us with a convenient mathematical representation of $P(n, t)$ and it allows us for example to define the moments and cumulants of $n$. The moment generating function is

$$M(z, t) = G(e^z, t) = \sum_{n} P(n, t) e^{nz},$$ (2)

from which the moments follow as derivatives with respect to $z$ evaluated at $z = 0$,

$$\langle n^m \rangle (t) = \partial_z^m M(z, t)|_{z \rightarrow 0} = \sum_{n} n^m P(n, t).$$ (3)

The cumulant generating function is

$$S(z, t) = \ln [M(z, t)] = \ln [G(e^z, t)]$$ (4)

and the cumulants are similarly defined as the derivatives

$$\langle\langle n^m \rangle \rangle (t) = \partial_z^m S(z, t)|_{z \rightarrow 0}.$$ (5)

The first three cumulants expressed in terms of the moments are shown in Table 1. For a Gauss distribution, only the first and second cumulants are nonzero, $\langle\langle n^m \rangle \rangle = 0$ for $m > 2$. Moreover, for large times $t \rightarrow \infty$, the cumulants typically become linear in time such that $\langle\langle n^m \rangle \rangle (t) \rightarrow \langle\langle I^m \rangle \rangle t$, where $\langle\langle I^m \rangle \rangle$ are the (zero-frequency) cumulants of the current. Generally, the high-order cumulants grow factorially with the cumulant order and oscillate as functions of basically any system parameter as it has recently been shown [10].
Table 1. Ordinary $\langle n^m \rangle$ and factorial cumulants $\langle n^m \rangle_F$ in terms of the moments $\langle n^m \rangle$.

<table>
<thead>
<tr>
<th>Cumulant</th>
<th>$m = 1$</th>
<th>$m = 2$</th>
<th>$m = 3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\langle n^m \rangle$</td>
<td>$\langle n \rangle$</td>
<td>$\langle (n - \langle n \rangle)^2 \rangle$</td>
<td>$\langle (n - \langle n \rangle)^3 \rangle$</td>
</tr>
<tr>
<td>$\langle n^m \rangle_F$</td>
<td>$\langle n \rangle$</td>
<td>$\langle (n - \langle n \rangle)^2 - \langle n \rangle \rangle$</td>
<td>$\langle (n - \langle n \rangle)^3 - \langle (n - \langle n \rangle)^2 \rangle + \langle n \rangle \rangle$</td>
</tr>
</tbody>
</table>

3. Factorial moments and factorial cumulants

A complementary characterization of the probability distribution $P(n,t)$ is provided by the factorial moments and the corresponding factorial cumulants. In mesoscopic physics, factorial cumulants have so far received only limited attention, except in studies of photons emitted from a quantum point contact [19], and very recently in connection with current fluctuations and entanglement entropy [20]. The factorial moment generating function is defined in terms of the generating function as [21, 22]

$$M_F(z, t) = G(z + 1, t) = \sum_n P(n, t)(z + 1)^n.$$  \hfill (6)

The factorial moments are now

$$\langle n^m \rangle_F(t) = \partial^m_z M_F(z, t) |_{z \rightarrow 0},$$  \hfill (7)

which can be written in terms of the ordinary moments as [22]

$$\langle n^m \rangle_F = \langle n(n-1)(n-2)\cdots(n-m+1) \rangle = \sum_{j=1}^{m} s(m, j) \langle n^j \rangle,$$  \hfill (8)

where $s(m, j)$ are the Stirling numbers of the first kind.1 The factorial cumulant generating function is

$$S_F(z, t) = \ln [M_F(z, t)] = \ln [G(z + 1, t)]$$  \hfill (9)

whose derivatives at $z = 0$ deliver the factorial cumulants

$$\langle n^m \rangle_F(t) = \partial^m_z S_F(z, t) |_{z \rightarrow 0}.$$  \hfill (10)

The factorial cumulants can also be expressed in terms of the ordinary cumulants as

$$\langle n^m \rangle_F = \langle n(n-1)(n-2)\cdots(n-m+1) \rangle = \sum_{j=1}^{m} s(m, j) \langle n^j \rangle.$$  \hfill (11)

The first three factorial cumulants given in terms of the ordinary moments are shown in Table 1. For a Poisson distribution, only the first factorial cumulant is nonzero, $\langle n^m \rangle_F = 0$ for $m > 1$. For large times $t \rightarrow \infty$, the factorial cumulants typically also become linear in time such that $\langle n^m \rangle_F(t) \rightarrow \langle I^m \rangle_F t$, where $\langle I^m \rangle_F$ are the (zero-frequency) factorial cumulants of the current.

The factorial cumulants are useful to consider in the light of the findings by Abanov and Ivanov who showed that the generating function for non-interacting electrons can always be factorized as

$$G(z, t) \stackrel{\text{generalized binomial}}{=} \left( z - Q \right) \prod_i (1 - p_i + p_i z),$$  \hfill (12)

1 The Stirling numbers of the first kind can be generated from the relation $[\ln (1 + x)]^j = j! \sum_{m=j} \frac{s(m,j)x^m}{m!}$ [22].
The quantum dot is operated close to a charge degeneracy point where only a single (additional) electron at a time can occupy either the upper ($\varepsilon_+$) or the lower ($\varepsilon_-$) level. The bare couplings to the leads are denoted as $\Gamma_S$ and $\Gamma_D$, and $x = 1 - f(\varepsilon_-)$, where $f(\varepsilon_-)$ is the Fermi-Dirac distribution of the drain ($D$) evaluated at the energy of the lower level.

In case of generalized binomial statistics we readily find for the factorial cumulants

$$
\langle\langle n^m \rangle\rangle_F^{\text{generalized binomial}} = (-1)^{m-1}(m-1)! \left[ \sum_i p_i^m - Q \right].
$$

(13)

This expression provides us with a direct test of whether or not a statistical distribution can be factorized into independent single-particle events as described by Eq. (12). In particular, for uni-directional transport (where $Q = 0$), a factorial cumulant of a given order $m$ must have a fixed sign that is determined by the factor $(-1)^{m-1}$ and no oscillations can occur as some parameter is varied. We have tested that this prediction indeed holds for several different non-interacting systems, see e.g. Refs. [13, 23]. However, as we have also shown, once interactions cause the zeros of the generating function to become complex, the factorial cumulants start oscillating as some parameter is varied [13]. Below, we consider one such interacting model, where interactions cause oscillations of the factorial cumulants.

4. Model

We consider the model of a two-level Coulomb blockade quantum dot depicted in Fig. 1 [24]. The quantum dot is weakly coupled to source and drain electrodes and the system is operated close to a charge degeneracy point where only a single (additional) electron at a time can enter and leave the quantum dot from the electrodes. The quantum dot has two single-particle levels which are both below the chemical potential of the source electrode. However, only one of the levels at a time can be occupied due to the strong Coulomb interactions. The upper level is well above the chemical potential of the drain electrode, while the lower level is close to the chemical potential, determined by the parameter $x = 1 - f(\varepsilon_-)$ which depends both on the temperature $T$ and the applied bias voltage. Here, $f$ is the Fermi-Dirac distribution of the drain electrode.

Charge transport through the quantum dot can be described by a master equation of the form

$$
\frac{d}{dt} \mathbf{p}(z) = \mathbf{M}(z)\mathbf{p}(z),
$$

(14)

where $\mathbf{p}(z) = [p_0(z), p_-(z), p_+(z)]^T$ contains the probabilities for the system to be empty (0) or having the lower (−) or upper (+) level occupied, respectively. Here, counting of electrons is effected by the parameter $z$ which is conjugate to the number of electrons that have tunneled.
Figure 2. High-order factorial cumulants. We show from left to right the 13th to 15th factorial cumulants as functions of the parameter $x$ at different times $t$. We plot the logarithm $\log |\langle \langle n^m \rangle_F / \langle \langle n \rangle \rangle|$, $m = 13, 14, 15$, for which dips occur as the factorial cumulants change sign. The colors of the curve segments correspond to the sign of the factorial cumulants (red is positive and blue is negative). Parameters are $\Gamma_S = 1$ and $\Gamma_D = 6$. Results are shown for the transient time $t = 0.5/\Gamma_S$, long time $t = 6/\Gamma_S$, and for infinite time $t \to \infty$, where the ratio $\langle \langle n^m \rangle_F / \langle \langle n \rangle \rangle$ is still well-defined and finite.

from the source electrode onto the quantum dot [25]. The corresponding rate matrix is

$$M(z) = \begin{bmatrix} -\{(1-x)\Gamma_D + 2\Gamma_S \} & x\Gamma_D & \Gamma_D \\ z\Gamma_S + (1-x)\Gamma_D & -x\Gamma_D & 0 \\ z\Gamma_S & 0 & -\Gamma_D \end{bmatrix},$$

(15)

where $\Gamma_S$ and $\Gamma_D$ are the bare couplings to the source and drain electrodes, respectively. The parameter $z$ multiplies the rate $\Gamma_R$ in the off-diagonal elements of $M(z)$, corresponding to charge transfer events across the source barrier. The generating function is then given as $G(z, t) = p_0(z) + p_-(z) + p_+(z)$ [13].

5. Results

We now use the methods of Refs. [13] and [26, 27] to calculate the high-order factorial cumulants for finite times and in the long-time limit, respectively. In Fig. 2 we show the 13th to 15th factorial cumulants as functions of the parameter $x$ at different times $t$. The factorial cumulants vary over many orders of magnitude as a function of $x$ and we therefore show the logarithm of the absolute value of the factorial cumulants. The colors of the curves then correspond to the sign of the factorial cumulants (red is positive and blue is negative). The factorial cumulants are normalized with respect to the mean of $n$, such that the ratio $\langle \langle n^m \rangle_F / \langle \langle n \rangle \rangle$ has a well-defined and finite value also in the long-time limit.
The figure clearly illustrates the oscillations of the high-order factorial cumulants as functions of the parameter $x$ (dips occur when the factorial cumulants cross zero and change sign). This holds both at finite times and in the long-time limit. The latter case was not considered explicitly in our work [13]. We stress that such oscillations cannot occur in a two-terminal scattering problem involving non-interacting electrons. Factorial cumulants are measurable using currently available counting techniques [7, 8, 9, 10, 11] and an experimental test of our predictions should therefore realistically be within reach. Experimentally it might be necessary to resolve the direction of the tunneling events. To this end, a double quantum dot with an asymmetrically coupled quantum point contact used for electron counting could be a promising candidate system [8].

6. Conclusions
We have discussed our recent theoretical proposal to detect interactions in the full counting statistics of charge transport through a nano-scale system by measuring high-order factorial cumulants. In the absence of interactions, the factorial cumulants do not oscillate. In contrast, oscillating factorial cumulants are a clear indication of interactions in the transport. This holds both at finite times and in the long-time limit. Factorial cumulants are measurable using available experimental techniques and we therefore believe that an experimental test of our prediction should be feasible. A more detailed account of our proposal can be found in Ref. [13].

7. Acknowledgements
The work was supported by the Swiss NSF and the NCCR Quantum Science and Technology.

References
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2.4 Some additions to our main result

In this section we address two important issues in connection with our main result. We do this on the example of a DQD composed of two dots in series strongly coupled to the reservoirs. Each of the dots can accommodate zero or one extra electrons, which can oscillate coherently between the dots before tunneling out to the drain electrode. We assume a large voltage bias, such that transport is unidirectional. Details are shown in publication (iii) [Sec. 2.4.3]. On the one hand we show that for an interacting system with Fano factor less than unity the factorial cumulants may still oscillate and thus pinpoint that the system is interacting. This means that the condition using factorial cumulants is stronger than having a Fano factor exceeding unity, which can also only occur in interacting systems (Also see the end of Sec. 2.1). On the other hand we consider capacitive coupling of the DQD to a QPC, which serves as a detector for the excess charge on each of the dots. This situation corresponds to a typical experimental setup [88, 89] and it is therefore crucial to analyze in what way the detector alters our result. We find that the coupling to the detector in our case indeed attenuates the oscillations in the factorial cumulants. However we stress that this statement is not general but specific to our model.

2.4.1 Factorial cumulants can indicate interactions even for sub-Poissonian statistics

In Fig. 2 of publication (iii) [Sec. 2.4.3] we plot the long-time limit Fano factor as a function of the detuning $\epsilon$ of the two dot levels. We notice that with a nonzero Coulomb interaction term in case of double occupancy of the DQD there are ranges of the detuning where the Fano factor exceeds unity. This reflects the fact that the electrons are interacting. However, for values of $\epsilon$ around zero, that is, when the left and right dot levels are roughly aligned, the Fano factor is smaller than one. For parameters fixed in this range we can therefore not conclude from the Fano factor whether the system is interacting. We then go on to show in Figs. 4 and 6 of publication (iii) [Sec. 2.4.3] that even in the range where the Fano factor is smaller than one the high-order factorial cumulants oscillate and thus clearly indicate the presence of the Coulomb interactions. Fig. 4 of publication (iii) [Sec. 2.4.3] shows the factorial cumulants of orders 11 through 14 as a function of time (a) once for a detuning such that $F < 1$ and (b) once for the case $F > 1$. Remarkably, they oscillate in both cases. The same conclusion is drawn from the analysis of the
long-time limit factorial cumulants as a function of their order, as shown in Fig. 6 of publication (iii) [Sec. 2.4.3]. We therefore conclude that our test using factorial cumulants is stronger than what can be deduced from the Fano factor alone.

### 2.4.2 A model for a QPC charge detector coupled to a QD and its effect on the counting statistics

In a next step we consider a QPC that is capacitively coupled to each of the dots. The QPC is modeled as a simple tunnel barrier\(^3\) [146]. If the coupling to the left and right dots is not the same, this asymmetry introduces decoherence. In fact, the measurement localizes the electrons, which without a detector undergo coherent oscillations between the left and right dots. With the asymmetrically coupled detector, however, the electrons inside the DQD are dephased, and the larger the asymmetry in the coupling, the larger is the dephasing rate [146].

Fig. 7 of publication (iii) [Sec. 2.4.3] shows the 10\(^{th}\) factorial cumulant as a function of the level detuning \(\varepsilon\) for different dephasing due to the QPC detector. If the QPC is symmetrically coupled to the two dots there is no dephasing and we observe many oscillations in the corresponding black curve. With increasing dephasing due to asymmetric coupling of the QPC to the left and right dots, the oscillations fade away, until for strong dephasing we no longer observe any oscillations in the 10\(^{th}\) factorial cumulant. We conclude that the detection mechanism can drastically alter the statistics and that it is crucial to choose the detection scheme carefully, or to include it in the modeling. Finally, we remark that this detection scheme is a very specific one, and that our particular result does not allow us to make a general statement about the influence of charge detection on the oscillations in the factorial cumulants.

\(^3\)We do not consider possible charge pile-up inside the QPC here. A more elaborate model for a QPC can for example be found in [145].
2.4.3 Time-dependent factorial cumulants in interacting nano-scale systems: publication (iii)

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Time-dependent factorial cumulants in interacting nano-scale systems

Dania Kambly · Christian Flindt

Received: date / Accepted: date

Abstract We discuss time-dependent factorial cumulants in interacting nano-scale systems. Recent theoretical work has shown that the full counting statistics of non-interacting electrons in a two-terminal conductor is always generalized binomial and the zeros of the generating function are consequently real and negative. However, as interactions are introduced in the transport, the zeros of the generating function may become complex. This has measurable consequences: With the zeros of the generating function moving away from the real-axis, the high-order factorial cumulants of the transport become oscillatory functions of time. Here we demonstrate this phenomenon on a model of charge transport through coherently coupled quantum dots attached to voltage-biased electrodes. Without interactions, the factorial cumulants are monotonic functions of the observation time. In contrast, as interactions are introduced, the factorial cumulants oscillate strongly as functions of time. We comment on possible measurements of oscillating factorial cumulants and outline several avenues for further investigations.

Keywords Full counting statistics · noise · factorial cumulants · interactions · generalized master equations

PACS 02.50.Ey · 72.70.+m · 73.23.Hk

1 Introduction

The full counting statistics (FCS) of charge transfers in sub-micron electrical conductors has become an active field of research [1–3]. Initially, investigations of FCS were primarily of theoretical interest, but several experiments [4–19] have now clearly demonstrated that measurements of FCS are achievable and much progress has been made: Non-Gaussian voltage and current fluctuations have been measured in tunnel junctions [4, 6, 12] and quantum point contacts [13], and the fourth and fifth current cumulants have been detected in an avalanche diode [15]. Additionally, real-time electron detection techniques [5, 7] have paved the way for measurements of the FCS of charge transport in single [9–11, 16–19] and double quantum dots [8, 14]. Following the initial measurements of the third cumulant of transport through quantum dots [8, 9], a series of experiments have addressed the conditional FCS [11], the transient high-order cumulants [16–18], and the finite-frequency FCS [19] in quantum dot systems. The works on transient high-order cumulants showed that high-order cumulants generically oscillate as functions of basically any system parameter as well as the observation time [16].

Investigations of FCS are motivated by the expectation that more information about the fundamental transport mechanisms can be extracted from the full statistical distribution of transferred charges than from the mean current and shot noise only [1–3]. However, the fact that high-order cumulants generically oscillate makes it less clear exactly what information the high-order cumulants contain? In a recent work [20], we have been drawing attention to the use of factorial cumulants to characterize the FCS of charge transport in nano-scale electrical conductors. So far, factorial cumulants have only received limited attention in mesoscopic physics (but see Refs. [21–24]). However, as we have shown, the factorial cumulants never oscillate (unlike the ordinary cumulants) for non-interacting two-terminal scattering problems. This result is based on the recent finding that the FCS for non-interacting electrons in a two-terminal scattering setup is always generalized binomial and the zeros of the generating function for the FCS consequently are real and negative [25, 26]; see Ref. [27] for a discussion of multi-terminal
conductors. In contrast, as interactions are introduced in the transport, the zeros of the generating function may become complex and the factorial cumulants start to oscillate [20]. This indicates that factorial cumulants may be useful to detect interactions among charges passing through a nano-sized electrical conductor. As such we address the fundamental question concerning FCS, namely what we can learn about a physical system by measuring the transport statistics beyond the mean current and the noise.

The purpose of this work is to illustrate these ideas on a model of transport through coherently coupled quantum dots. In previous work [20, 28], we considered systems described by classical master equations. We now turn to a situation, where the quantum coherent coupling between two parts of the conductor is important. The system we consider is a double quantum dot (DQD) attached to external source and drain electrodes. We employ a generalized master equation (GME) approach which allows us to treat strong coupling to the leads together with the coherent evolution of electrons inside the DQD. We treat two cases of particular interest: In the non-interacting regime, the DQD can accommodate zero, one, or two electrons at a time, without additional charging energy required for the second electron. We show that the factorial cumulants in this case do not oscillate as functions of the observation time and from the high-order factorial cumulants we extract the zeros of the generating function which are real and negative. Next, we consider the strongly interacting case, where double-occupation of the DQD is excluded. In this case, the time-dependent factorial cumulants oscillate – a clear signature of interactions in the transport – and the zeros of the generating function are complex.

In the interacting case, we find that the Fano factor $F$, i.e. the ratio of the shot noise over the mean current, may either be super-Poissonian ($F > 1$) or sub-Poissonian ($F < 1$). Super-Poissonian noise is typically taken as a signature of interactions in the transport [2], while no clear conclusion can be drawn from a sub-Poissonian Fano factor. Interestingly, we find that the factorial cumulants may oscillate in both situations, showing that the factorial cumulants can provide a clear signature of interactions even when the current fluctuations are sub-Poissonian. We conclude our theoretical investigations of factorial cumulants by examining the influence of dephasing of electrons passing through the DQD [29, 30], for instance due to a nearby charge detector.

The paper is organized as follows: in Sec. 2 we introduce the essential terminology used in FCS and provide the basic definitions with a special emphasis on factorial cumulants and the concept of generalized binomial statistics. In Sec. 3 we then turn to the asymptotic behavior of high-order cumulants (both ordinary and factorial cumulants) and show why the high-order factorial cumulants do not oscillate for transport of non-interacting electrons in a two-terminal conductor. In Sec. 4 we introduce a model of electron transport through a DQD described by a Markovian GME, while Sec. 5 is devoted to the details of our calculations of time-dependent factorial cumulants. In Sec. 6 we demonstrate how interactions on the DQD give rise to clear oscillations of the high-order factorial cumulants with the zeros of the generating function moving away from the negative real-axis and into the complex plane. Finally, Section 7 is dedicated to a summary of the work as well as our concluding remarks.

2 Full counting statistics & factorial cumulants

Full counting statistics concerns the quantum statistical process of electron transport in mesoscopic conductors [1–3, 20, 25–49]. The full counting statistics (FCS) is the probability $P(n,t)$ that $n$ electrons have traversed a conductor during a time span $[0,t]$ of duration $t$. The information contained in the probability distribution may equally well be encoded in the generating function (GF) defined as

$$\mathcal{G}(z,t) = \sum_{n} P(n,t)z^n. \quad (1)$$

The normalization condition for the probabilities, $\sum_{n} P(n,t) = 1$, implies for the GF that $\mathcal{G}(z=1,t) = 1$. Important information about the charge transport can be obtained from the GF: If the transport process consists of several independent sub-processes, the GF factors into a product of the GFs corresponding to each of these sub-processes, similarly to how the partition function in statistical mechanics may be written as a product of the partition functions for each independent sub-system. Elementary transport processes can thus be identified by factorizing the GF. In the case of transport of noninteracting electrons through a two-terminal conductor, Abanov and Ivanov have shown recently that the GF can be factorized into single-particle events of binomial form [25, 26] (also see [44, 45]). Such distributions have been dubbed generalized binomial statistics [46, 25, 26], which will be of central importance in this work.

Several useful statistical functions and quantities can be obtained from the GF. First, we can define a moment generating function (MGF)

$$\mathcal{M}(z,t) = \mathcal{G}(e^z,t), \quad (2)$$

which generates the statistical moments of $n$ by differentiation with respect to the counting field $z$ at $z = 0$

$$\langle n^m \rangle(t) = \left. \frac{\partial^m}{\partial z^m} \mathcal{M}(z,t) \right|_{z=0} = \sum_{n} n^m P(n,t). \quad (3)$$

The MGF of a transport process composed of several independent processes factors into a product of the corresponding MGFs. However, the moments of the full process are not related to the moments of the individual sub-processes in a simple way. This motivates the definition of cumulants,
also known as irreducible moments. The cumulant generating function (CGF) is defined as the logarithm of the MGF
\[ \mathcal{G}(z,t) = \log[M(z,t)] = \log[\mathcal{G}(e^z,t)], \tag{4} \]
which again delivers the cumulants of \( n \) by differentiation with respect to \( z \) at \( z = 0 \):
\[ \langle n^m \rangle(t) = \partial_z^m \mathcal{G}(z,t)|_{z=0}. \tag{5} \]
The first cumulant is the mean of \( n \), \( \langle n \rangle = \langle n\rangle \), the second cumulant is the variance, \( \langle n^2 \rangle = \langle n\rangle^2 \), and the third is the skewness, \( \langle n^3 \rangle = \langle n^2 \rangle \). It is easy to show that the cumulants of a transport process are simply the sum of the cumulants corresponding to each independent sub-process. Moreover, for a Gaussian distribution only the first and second cumulants are non-zero, while all higher cumulants vanish. In this respect, one may use cumulants of a distribution as a measure of (non-)gaussianity.

The conventional moments and cumulants, as defined above, have been investigated intensively in the field of FCS [3]. The zero-frequency cumulants of the current are given by the long-time limit of the cumulants of \( n \) as
\[ \langle I^m \rangle \equiv \lim_{t \to \infty} \frac{\langle n^m \rangle}{t}. \tag{6} \]
The current is treated as a continuous variable and continuous variables are typically characterized by their cumulants. However, another interesting class of statistical quantities exists, which has received much less attention in FCS. These are the factorial moments and factorial cumulants, which are mostly discussed in the context of discrete variables [50, 51]. The number of counted electrons \( n \) is obviously a discrete variable, and it is natural to ask if the current cumulants, as defined in Eq. (6), carry signatures of this discreteness?

The factorial moments are again generated by a factorial MGF, which can be defined based on the GF in Eq. (1). The factorial MGF is defined as
\[ M_F(z,t) = \mathcal{G}(z+1,t) \tag{7} \]
and the corresponding factorial moments read
\[ \langle n^m \rangle_F(t) = \partial_z^m M_F(z,t)|_{z=0} = \langle n(n-1) \cdots (n-m+1) \rangle \tag{8} \]
in terms of the ordinary moments. In analogy with the conventional CGF, the factorial CGF is defined as
\[ \mathcal{G}_F(z,t) = \log[M_F(z,t)] = \log[\mathcal{G}(z+1,t)] \tag{9} \]
and the corresponding factorial cumulants read
\[ \langle n^m \rangle_F(t) = \partial_z^m \mathcal{G}_F(z,t)|_{z=0} = \langle n(n-1) \cdots (n-m+1) \rangle. \tag{10} \]
As mentioned above, factorial moments and factorial cumulants are of particular interest when considering probability distributions of discrete variables. For example, for a Poisson process with rate \( \Gamma \), which is the physical limit of rare events, the FCS is well-known and reads
\[ P(n,t) = \frac{(\Gamma t)^n}{n!} e^{-\Gamma t}. \tag{11} \]
The corresponding GF then becomes
\[ \mathcal{G}(z,t) = \frac{e^{\Gamma z(t-1)}}{\Gamma z(t-1)}, \quad \text{(Poisson process)} \tag{12} \]
from which it is easy to show that the cumulants are
\[ \langle n^m \rangle_F(t) = \Gamma t, \quad \text{(Poisson process)} \tag{13} \]
for all \( m = 1,2, \ldots \). In contrast, the first factorial cumulant reads
\[ \langle n \rangle_F(t) = \Gamma t, \quad \text{(Poisson process)}, \tag{14} \]
while all higher factorial cumulants are zero
\[ \langle n^m \rangle_F(t) = 0, \quad m > 1 \quad \text{(Poisson process)}. \tag{15} \]
Thus, similarly to how ordinary cumulants are useful as measures of (non-)gaussianity, we may use factorial cumulants to characterize deviations of a distribution from Poisson statistics. It is also clear that a factorial cumulant of a given order is the sum of the factorial cumulants of all independent sub-processes, in the same way as for the cumulants.

Throughout this work we will rely on an important result by Abanov and Ivanov, who have shown that the FCS of non-interacting electrons in a two-terminal scattering problem is always generalized binomial [25, 26]. In this case, the GF takes the special form
\[ \mathcal{G}(z,t) \overset{\text{generalized binomial}}{=} z^{-Q} \prod_i (1 - p_i + p_i z), \tag{16} \]
where the (time-dependent) \( p_i \)'s are real with \( 0 \leq p_i \leq 1 \). Each factor in the product can be interpreted as a single binomial charge transfer event occurring with probability \( p_i \). The factor in front, \( z^{-Q} \), corresponds to a deterministic background charge transfer
\[ Q = \sum_i p_i - \langle n \rangle \geq 0 \tag{17} \]
and the (time-dependent) Fano factor then reads
\[ F(t) \equiv \langle \frac{n^2}{\langle n \rangle} \rangle(t) \overset{\text{generalized binomial}}{=} \frac{\sum_i p_i (1 - p_i)}{\sum_i p_i}, \tag{18} \]
which is always smaller than unity, corresponding to a Poisson process. Following this reasoning, a super-poissionian
Fano factor, $F > 1$, can be taken as a sign of interactions. Super-Poissonian noise was recently measured in an experiment on transport of interacting electrons through a double quantum dot [52]. Still, the noise may also be sub-Poissonian, $F < 1$, in the presence of interactions.

3 High (factorial) cumulants

In this Section we are interested in the generic behavior of high-order (factorial) cumulants. To this end, we first discuss an approximation of high derivatives [53, 54] and then show how these ideas can be applied in the context of FCS.

In the following, we consider a generic (factorial) CGF $S_F(z)$ and assume that it has a number of singularities $z_j$ in the complex plane. Close to each of these singularities, we may approximate the (factorial) CGF as

$$S_F(z) \simeq \frac{A_j}{(z_j - z)^{\mu_j}}, \quad \text{for } z \simeq z_j \tag{19}$$

for some constants $A_j$ and $\mu_j$. Here, the constant $\mu_j$ is determined by the nature of the singularity, for instance $\mu_j = -1/2$ corresponds to a square-root branch point, while an integer value of $\mu_j$ would correspond to the order of a pole at $z = z_j$. Using the first Darboux approximation [53, 54, 16], we may now evaluate the (factorial) cumulant of order $m$ by differentiating the expression in Eq. (19) $m$ times with respect to $z$ at $z = 0$ and sum over the contributions from all singularities as

$$\langle\langle n^m \rangle\rangle_{(F)} \simeq \sum_j \frac{A_j B_{m,\mu_j} e^{-(m+\mu_j) \arg z_j}}{|z_j|^{m+\mu_j}}. \tag{20}$$

Here we have introduced the polar notation $z_j = |z_j| e^{i \arg z_j}$ together with the factors

$$B_{m,\mu_j} \equiv \mu_j (\mu_j + 1) \cdots (\mu_j + m - 1). \tag{21}$$

Equation (20) is particularly useful if the sum can be reduced to only a few terms. For high orders, the singularities closest to $z = 0$ dominate the sum, which leads to a considerable simplification. For example, if a single complex conjugate pair of singularities, $z_0 = |z_0| e^{i \arg z_0}$ and $z_0^* = |z_0| e^{-i \arg z_0}$, are closest to $z = 0$, the high-order (factorial) cumulants can be approximated as

$$\langle\langle n^m \rangle\rangle_{(F)} \simeq \frac{2 |A_0| B_{m,\mu_0}}{|z_0|^{m+\mu_0}} \cos [(m + \mu_0) \arg z_0 - \arg A_0]. \tag{22}$$

This result shows that the absolute value of the (factorial) cumulants generically grows factorially with the cumulant order $m$, due to the factors $B_{m,\mu_0}$, and that they tend to oscillate as a function of any parameter, including time $t$, that changes $\arg z_0$. Such universal oscillations have been observed experimentally in electron transport through a quantum dot [16–18].

In contrast, in the particular situation, where there is just a single dominant singularity $z_0$ on the real-axis, the high-order (factorial) cumulants can be approximated as

$$\langle\langle n^m \rangle\rangle_{(F)} \simeq (-1)^{m+\mu_0} \frac{A_0 B_{m,\mu_0}}{|z_0|^{m+\mu_0}}. \tag{23}$$

In this case, the factorial growth with the order persists, but no oscillations are expected as long as the dominant singularity $z_0$ stays on the real-axis.

Let us now consider non-interacting electrons in a two-terminal conductor. As shown by Abanov and Ivanov [25, 26], the statistics is generalized binomial in this situation and the GF takes on the form given by Eq. (16). The corresponding cumulants are complicated functions of the probabilities $p_i$. In contrast, the factorial cumulants are simply

$$\langle\langle n^m \rangle\rangle_{F \text{ generalized binomial}} \equiv (-1)^{m-1} (m-1)! \left[ \sum_i p_i^m - Q \right]. \tag{24}$$

For uni-directional transport ($Q = 0$), the largest probability $p_{\text{max}}$ will dominate the high factorial cumulants, which can be approximated as

$$\langle\langle n^m \rangle\rangle_F \simeq (-1)^{m-1} (m-1)! p_{\text{max}}^m. \tag{25}$$

This expression can also be understood from Eq. (20) by noting that the factorial CGF has logarithmic singularities at values of the counting field $z$, where the factorial MGF is zero. Combining Eqs. (7) and (16), we easily see that the factorial MGF has zeros at $z_j = -1/p_j \leq -1$. Moreover, the zero corresponding to the largest probability $p_{\text{max}}$ is closest to $z = 0$ and will dominate the high factorial cumulants as seen in Eq. (25).

We have seen above that high-order cumulants tend to oscillate as functions of basically any parameter, with or without interactions. In contrast, as our analysis also shows, the high-order factorial cumulants never oscillate for non-interacting electrons in a two-terminal scattering problem. This behavior can be traced back to the factorization of the GF in Eq. (16), which implies that the singularities of the factorial CGF are always real and negative. This makes factorial cumulants promising candidates for the detection of interactions in FCS. In particular, oscillating factorial cumulants must be due to interactions. In our previous work [20], we employed these ideas to incoherent electron transport through a single quantum dot. We showed how interactions may cause the singularities of the factorial CGF to move away from the real-axis and into the complex plane, making the high-order factorial cumulants oscillate.

In the following we apply these ideas to electron transport through a DQD, where the electrons may oscillate quantum coherently between the two quantum dots. In contrast to our previous work [20], we consider not only the time-dependent factorial cumulants of the transferred charge, but also the zero-frequency factorial cumulants of the current.
4 Coulomb blockade quantum dots

We consider two-terminal nano-scale conductors connected to source and drain electrodes. Charge transport is described using a generalized master equation (GME) for the reduced density matrix \( \hat{\rho} \) of the conductor obtained by tracing out the electronic leads. The GME accounts for the coherent evolution of charges inside the conductor as well as the transfer of electrons between the conductor and the leads. To evaluate the FCS, it is convenient to unravel the reduced density matrix with respect to the number of electrons that have been collected in the drain electrode during the time span \([0,t]\) [55, 56]. With this \( n \)-resolved density matrix \( \hat{\rho}(n,t) \) at hand, the FCS is obtained by tracing over the states of the conductor

\[
P(n,t) = \text{Tr}[\hat{\rho}(n,t)].
\]

(26)

Similarly, we recover the original reduced density matrix by summing over \( n \), i.e.

\[
\hat{\rho}(t) = \sum_n \hat{\rho}(n,t).
\]

(27)

From these definitions, the GF reads

\[
G(z,t) = \sum_n \text{Tr}[\hat{\rho}(n,t)]z^n = \text{Tr}[\hat{\rho}(z,t)],
\]

(28)

where we have introduced the \( z \)-dependent reduced density matrix

\[
\hat{\rho}(z,t) = \sum_n \hat{\rho}(n,t)z^n.
\]

(29)

The particular conductor we now discuss consists of two quantum dots in series attached to source and drain electrodes. A schematic of the system is shown in Fig. 1. The inter-dot Coulomb interaction can be tuned, so that both regimes of noninteracting and interacting electrons can be realized and compared. Disregarding the electronic spin degree of freedom (for example due to a strong magnetic field), the double quantum dot can be occupied by either zero, one, or two (additional) electrons. Experimentally, the charge on the quantum dots can be measured using a nearby quantum point contact (QPC), whose conductance it sensitive to the occupations of the individual quantum dots [9, 14]. This charge detection protocol makes it possible to deduce the number of electrons that have passed through the DQD in a given time span. If the QPC is not sensitive to the charge occupations of the individual quantum dots, but only to the total charge, the measurement is not expected to destroy the coherent oscillations between the quantum dots. On the other hand, if the QPC measures the charge states of the individual quantum dots, it introduces decoherence in the dynamics of electrons inside the DQD [57, 58].

The full many-body Hamiltonian of our system reads

\[
\hat{H} = \hat{H}_{\text{DQD}} + \hat{H}_{\text{leads}} + \hat{H}_T + \hat{H}_{\text{QPC}} + \hat{H}_{\text{DQD-QPC}}.
\]

(30)

It consists of the Hamiltonian of the DQD

\[
\hat{H}_{\text{DQD}} = E_L \hat{\alpha}_L^\dagger \hat{\alpha}_L + E_R \hat{\alpha}_R^\dagger \hat{\alpha}_R + \Omega (\hat{\alpha}_L^\dagger \hat{\alpha}_R + \hat{\alpha}_R^\dagger \hat{\alpha}_L) + U \hat{\alpha}_L \hat{\alpha}_R,
\]

where the operators \( \hat{\alpha}_L^\dagger \) and \( \hat{\alpha}_R^\dagger \) create an electron on the left and right quantum dot level with energy \( E_L \) or \( E_R \), respectively. The tunnel coupling between the levels is denoted as \( \Omega \) and \( \hat{\alpha}_L = \hat{\alpha}_L^\dagger \hat{\alpha}_L = 0,1, \alpha = L,R \) is the occupation number operator of each quantum dot. The inter-dot Coulomb interaction is denoted as \( U \). The electrons in the leads are treated as non-interacting and are given by the Hamiltonian

\[
\hat{H}_{\text{leads}} = \sum_{k,\alpha=L,R} E_{k\alpha} \hat{\alpha}_{k\alpha}^\dagger \hat{\alpha}_{k\alpha},
\]

(31)

where the operators \( \hat{\alpha}_{k\alpha}^\dagger \) create an electron in lead \( \alpha = L,R \) with momentum \( k \) and energy \( E_{k\alpha} \). The coupling between the DQD and the leads is accounted for by the standard Hamiltonian

\[
\hat{H}_T = \sum_{k,\alpha=L,R} (t_{k\alpha} \hat{\alpha}_{k\alpha}^\dagger \hat{\alpha}_L + t_{k\alpha}^\ast \hat{\alpha}_L^\dagger \hat{\alpha}_{k\alpha}),
\]

(32)

which connects the left (right) lead to the left (right) quantum dot. Finally, the QPC is modeled as a tunnel barrier

\[
\hat{H}_{\text{QPC}} = \sum_{k,\alpha=L,R} \tilde{\epsilon}_{k\alpha} \hat{\alpha}_{k\alpha}^\dagger \hat{\alpha}_{k\alpha}^\dagger + \sum_{k,k'} \tilde{\epsilon}_{k\alpha} \hat{\alpha}_{k\alpha}^\dagger \hat{\alpha}_{k\alpha}^\dagger + \tilde{\epsilon}_{k\alpha} \hat{\alpha}_{k\alpha} \hat{\alpha}_{k\alpha},
\]

where the first sum corresponds to the electronic reservoirs on the left (\( \alpha = L \)) and right side (\( \alpha = R \)) of the QPC and the second sum describes the coupling of states in different leads with tunnel coupling \( \tilde{\epsilon}_{k\alpha} \).
If the QPC only couples to the total charge of the DQD, the charge detection is not expected to cause decoherence of the coherent oscillations of electrons inside the DQD. It is, however, interesting to investigate how an asymmetrically coupled QPC will affect the transport in the DQD. To this end, we assume that the QPC, besides the coupling to the total charge, has an additional capacitive coupling to the left quantum dot only. The charge occupation of the left quantum dot modulates the transparency of the QPC according to the Hamiltonian

$$\hat{H}_{\text{DQD-QPC}} = \sum_{k,k'} \left( \delta \hat{t}_{kk'} \hat{c}_{kL}^\dagger \hat{c}_{kR} + \delta \hat{t}_{kk'}^* \hat{c}_{kR}^\dagger \hat{c}_{k'\text{L}} \right). \quad (33)$$

Here $\delta \hat{t}_{kk'}$ is the change of the QPC tunnel coupling in response to an (additional) electron occupying the left quantum dot.

We now follow Gurvitz in deriving a Markovian GME for the reduced density matrix $\hat{\rho}_{\text{DQD}}$ of the DQD obtained by tracing out the electronic leads and the QPC. The details of the derivation can be found in Refs. [57, 58]. We assume that a large bias is applied between the electronic leads, such that electron transport is uni-directional from the left to the right electrode. The electronic reservoirs have a continuous density of states and the discrete levels of the DQD are situated well inside the transport window. Under these assumptions, we may formulate a Markovian GME for the $n$-resolved reduced density matrix $\hat{\rho}_{\text{DQD}}(n,t)$. Its diagonal elements are the probabilities for the DQD to be either empty, having only left or right quantum dot occupied, or to be doubly-occupied, while $n$ electrons have been collected in the right lead during the measuring time $t$.

The diagonal elements of $\hat{\rho}_{\text{DQD}}(n,t)$ are denoted as $\rho_0(n,t)$, $\rho_L(n,t)$, $\rho_R(n,t)$, and $\rho_{LR}(n,t)$. Additionally, we need the coherences between the left and the right quantum dot levels, denoted as $\rho_{LR}(n,t)$ and $\rho_{RL}(n,t)$. Coherences between states with different charge occupations are excluded. Since the off-diagonal elements fulfill $\rho_{LR}(n,t) = \rho_{RL}(n,t)$, it suffices to consider the real and imaginary parts of $\rho_{LR}(n,t)$. The elements of the reduced density matrix can then be collected in the vector

$$|\rho(n,t)\rangle = [\rho_0, \rho_L, \rho_R, \Re[\rho_{RL}], \Im[\rho_{RL}], \rho_{\text{LR}}]^T(n,t). \quad (34)$$

The corresponding $z$-dependent reduced density matrix follows from the definition in Eq. (29) and reads

$$|\rho(z,t)\rangle = \sum_n |\rho(n,t)\rangle |z^n\rangle. \quad (35)$$

The Markovian GME then takes the form

$$\partial_t |\rho(z,t)\rangle = \mathbf{M}(z) |\rho(z,t)\rangle, \quad (36)$$

with the rate matrix reading

$$\mathbf{M}(z) = \begin{bmatrix}
-\Gamma_L & z\Gamma_R & 0 & 0 & 0 \\
\Gamma_L & 0 & 0 & -2\Omega & z\Gamma_R \\
0 & 0 & -\Gamma_L - \Gamma_R & 0 & 2\Omega \\
0 & 0 & -\Gamma & -\varepsilon & 0 \\
0 & \Gamma_L & 0 & 0 & -\Gamma_R
\end{bmatrix}. \quad (37)$$

where $\varepsilon \equiv E_R - E_L$ is the energy detuning of the two quantum dot levels. Additionally, the rate

$$\Gamma = \frac{1}{2} (\Gamma_R + \Gamma_L + \gamma) \quad (38)$$

determines the decay of the off-diagonal elements of $\hat{\rho}_{\text{DQD}}(n,t)$ and the broadening of the electronic levels. The electronic tunneling rates depend on the charge occupation of the DQD and read

$$\Gamma_a = \frac{2\pi}{\hbar} \mathcal{D}_a(E_a) |t_{ka}|^2, \quad \alpha = L, R \quad (39)$$

and

$$\tilde{\Gamma}_a = \frac{2\pi}{\hbar} \mathcal{D}_a(E_a + U) |t_{ka}|^2, \quad \alpha = L, R, \quad (40)$$

where $\mathcal{D}_a$ denotes the density of states in lead $\alpha = L, R$, and the tunneling amplitudes $t_{ka}$ are assumed to be $k$-independent. Here, $\Gamma_a$ is the tunneling rate from the left lead onto the left quantum dot, if the DQD is empty initially. On the other hand, if the right quantum dot is already occupied, electrons tunnel into the left quantum dot at rate $\tilde{\Gamma}_L$. Similarly, electrons tunnel from the right quantum dot into the right lead with rate $\tilde{\Gamma}_R$, if the left quantum dot is empty, and with rate $\Gamma_R$, if the left quantum dot is occupied. Without inter-dot interactions, $U = 0$, we have $\Gamma_{L(R)} = \tilde{\Gamma}_{L(R)}$. Factors of $z$ have been included in the off-diagonal elements of $\mathbf{M}(z)$ corresponding to charge transfers from the right quantum dot to the right lead.

Finally, the decoherence rate introduced by the QPC is given by [58]

$$\gamma = \frac{eV_d}{2\pi\hbar} (\sqrt{T} - \sqrt{T})^2, \quad (41)$$

where $V_d$ is the bias applied across the QPC. The transmission probability for electrons to tunnel through the QPC is

$$T = (2\pi)^2 |\tilde{\mathcal{T}}_{k,k'}|^2 \mathcal{D}_L \mathcal{D}_R, \quad (42)$$

when the left quantum dot is empty. In contrast, when the left quantum dot is occupied, the QPC transmission reads

$$\tilde{T} = (2\pi)^2 |\tilde{\mathcal{T}}_{k,k'} + \delta \mathcal{T}_{k,k'}|^2 \mathcal{D}_L \mathcal{D}_R. \quad (43)$$

Above, the symbols $\mathcal{D}_{L(R)}$ denote the density of states in the left (right) lead of the QPC.
5 Calculations

We now evaluate the FCS by formally solving Eq. (36). We consider fluctuations in the stationary state, which we suppose has been reached at $t = 0$, when we start counting charges. The stationary state is denoted as $|0\rangle$ and is obtained by solving $M(z)|0\rangle = 0$ with the normalization condition $\langle 0|0\rangle = 1$, where $\{1, 1, 1, 0, 0, 1\}$. From Eq. (36), the GF can now be written as

$$G(z, t) = \langle \langle 0|e^{M(z)t}|0\rangle \rangle. \quad (44)$$

It is easy to verify that this expression fulfills the condition $G(z = 1, t) = 1$ for the GF. It is a general, formally exact result, which yields the complete FCS at any time given the matrix $M(z)$. However, in practice the expression may be difficult to evaluate due to the matrix exponentiation, in particular if the aim is to calculate the high (factorial) moments or (factorial) cumulants. In our recent work [20], we developed a simple method to evaluate the high-order, time-dependent statistics for these types of problems and we will also be using this method here. For details of the method, we refer the interested reader to Appendix A of Ref. [20].

In addition to the finite-time FCS, it is interesting to investigate the GF at long times. In this limit, the GF takes on a large-deviation form

$$G(z, t) \sim e^{tδθ(z)}, \quad (45)$$

where the rate of change is determined by the eigenvalue of $M(z)$ with the largest real-part, i.e.

$$θ(z) = \max_j \{λ_j(z)\}. \quad (46)$$

From $θ(z)$ we may obtain the (factorial) CGF for the zero-frequency cumulants of the currents. The zero-frequency cumulants of the current are

$$\langle \langle \langle m^n \rangle \rangle \rangle = \partial e^{θ(z)}|_{z \to 0}, \quad (47)$$

while the corresponding factorial cumulants read

$$\langle \langle m^n \rangle \rangle_{\mathcal{F}} = \partial e^{θ(z + 1)}|_{z \to 0}. \quad (48)$$

In general, we can assume that the matrix $M(z)$ at $z = 1$ has a single eigenvalue equal to zero, i.e. $λ_0(1) = 0$, corresponding to the (unique) stationary state, while all other eigenvalues have negative real-parts, ensuring relaxation toward the stationary state. For values of $z$ close to unity, we expect that $λ_0(z)$ develops adiabatically from 0 and still determines $θ(z)$ such that $θ(z) = λ_0(z)$ for $z \approx 1$. The derivatives of $λ_0(z)$ with respect to $z$ at $z = 1$ then determines the (factorial) cumulants of the current according to Eqs. (47) and (48).

Again, for large matrices $M(z)$, it might not be viable to directly calculate the eigenvalue $λ_0(z)$ and its derivatives with respect to $z$ at $z = 1$. This problem may be circumvented by considering the calculation of $λ_0(z)$ as a perturbation problem around $z = 1$. The matrix $M(z)$ is written as $M(z) = M(1) + δM(z)$, where $M(1)$ is the unperturbed matrix with eigenvalue $λ_0(z = 1) = 0$ and $δM(z) = M(z) - M(1)$ is the perturbation. The eigenvalue $λ_0(z)$ can then be calculated order by order in $z$ using the recursive perturbation method developed in Refs. [39,40,41]. This method yields the (factorial) cumulants of the current and will be used below.

Finally, it is important to understand the connection between the FCS at finite times and in the long-time limit. As discussed in the previous section, the high (factorial) cumulants are related to the singularities of the (factorial) CGF in the complex plane of $z$. At finite times, the (factorial) CGF

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**Fig. 2** Fano factor versus the energy detuning $\varepsilon$. Results are shown with $(U \neq 0)$ and without $(U = 0)$ strong Coulomb interactions on the DQD. With strong Coulomb interactions, only 0 or 1 electrons can occupy the DQD. In contrast, without Coulomb interactions the DQD may also be doubly occupied. The QPC is coupled symmetrically to the DQD ($γ = 0$). The other parameters are $I_R = \frac{1}{4} I_L$ and $Ω = \hbar I_L$. The squares and stars mark $\varepsilon = 0.8Ω$ and $\varepsilon = 2.2Ω$, respectively.

**Fig. 3** Time-dependent factorial cumulants without interactions $(U = 0)$. The factorial cumulants $\langle \langle n^m \rangle \rangle_{\mathcal{F}}$ of order $m = 11$ through $m = 14$ are shown as functions of time. The results correspond to the point marked with a red star in Fig. 2. The factorial cumulants do not oscillate, as expected without interactions. The full lines indicate numerical results, while circles show the approximation given by Eq. (25).
has logarithmic singularities at values of $z$, where the (factorial) MGF is zero. In contrast, in the long-time limit, the singularities of the (factorial) CGF are determined by the singularities of the eigenvalues of $\mathbf{M}(z)$. Typically, the eigenvalues have square-root branch points from the two degeneracy points, where two eigenvalues are equal, i.e. $\lambda_0(z_i) = \lambda_1(z_i)$ for some $z_i$. Considering now the GF at finite times close to such a degeneracy point, we may approximate the GF in Eq. (44) as

$$\mathcal{G}(z,t) = \sum_j c_j(z)e^{\lambda_j(z)t} \simeq c_0(z)e^{\lambda_0(z)t} + c_1(z)e^{\lambda_1(z)t}, \quad (49)$$

where the coefficients $c_j(z)$ depend on the initial condition and only the contributions from the two largest eigenvalues have been included. Solving for the zeros of $\mathcal{G}(z,t)$, we obtain the equations

$$\lambda_0(z) = \lambda_1(z) + \frac{\log\{c_1(z)/c_0(z)\} + i\pi(2n+1)}{t}, \quad (50)$$

where $n$ is an integer. Importantly, we see that the second term on the right-hand side vanishes in the limit $t \to \infty$. This analysis shows that the zeros of the GF as functions of time move towards the solutions of the equation $\lambda_0(z) = \lambda_1(z)$, which also determines the branch-point singularities in the long-time limit cf. the discussion above. This connects the finite-time FCS with its long-time behavior.

6 Results

We are now ready to illustrate the use of factorial cumulants on the concrete example of charge transport through a DQD. We analyze several different parameter regimes of the system which are discussed in turn. To begin with, it is instructive to consider the Fano factor $F$ of the transport in the long-time limit

$$F = \frac{\langle (n^2) \rangle (t)}{\langle n \rangle (t)} \bigg|_{t \to \infty} = \frac{\langle I^2 \rangle}{\langle I \rangle} \quad (51)$$

given as the ratio of the zero-frequency current noise over the mean current. Figure 2 shows the Fano factor as a function of the energy dealignment $\epsilon$ without any decoherence due to the QPC, $\gamma = 0$. We present results with $(U \neq 0)$ and without interactions $(U = 0)$. For the non-interacting case, the Fano factor is never super-Poissonian $(F > 1)$ as expected for uni-directional transport with generalized binomial statistics. In contrast, for the interacting case there are certain ranges of the dealignment, where the noise becomes super-Poissonian. However, there is also a range of dealignments around $\epsilon = 0$, where the noise is sub-Poissonian $(F < 1)$, and in this regime a measurement of the Fano factor would not give any clear evidence of interactions. We note that recent noise measurements on transport through vertically coupled quantum dots are in qualitative agreement with the results shown for the interacting case [60,52,61].

We mark two points on the curves corresponding to values of the dealignment, where the noise in the interacting case is either sub-Poissonian (squares) or super-Poissonian (stars). As we demonstrate now, the factorial cumulants give clear signatures of the interactions even in the cases with sub-Poissonian noise, where no conclusions can be drawn from the Fano factor alone. (We note that we also find oscillating factorial cumulants with symmetric rates $I_L = I_R$, where the noise is always sub-Poissonian.)

In Fig. 3 we show the time-dependent factorial cumulants for the non-interacting case, corresponding to the point marked with a star in Fig. 2. For the point marked with a square similar results are obtained. Without interactions, the FCS is generalized binomial and the factorial cumulants are
expected to follow Eqs. (24) and (25), which predict no oscillations of the factorial cumulants as functions of time or any other parameter. This prediction is confirmed by Fig. 3, where a clearly monotonic behavior is found as a function of time. Moreover, from the calculated factorial cumulants, we may extract \( p_{\text{max}} \) in Eq. (25) as a function of time. Inserting \( p_{\text{max}} \) back into Eq. (25), we can compare this expression with the numerical results for the high-order factorial cumulants. In Fig. 3, the predicted behavior based on Eq. (25) is shown with circles and is seen to be in very good agreement with the full numerics.

Next, we turn to the time-dependent factorial cumulants in the interacting case. In Fig. 4 we show the high-order factorial cumulants corresponding to the two dealignments marked with stars and squares in Fig. 2. In this case, the factorial cumulants oscillate as functions of time in contrast to the non-interacting situation, where no oscillations are observed. To best visualize the oscillations, we show the logarithm of the absolute value of the factorial cumulants. Downwards-pointing spikes then correspond to the factorial cumulants passing through zero and changing sign. The oscillating factorial cumulants are a clear signature of interactions in the transport and they show that the FCS for this system is not generalized binomial, neither when the noise is sub-poissonian (square) nor super-poissonian (star).

Again, we can understand the high-order factorial cumulants using the expressions from Sec. 3. In this case, when the FCS is not generalized binomial, the high-order factorial cumulants are expected to follow Eq. (22), which assumes that the factorial CGF has a complex-conjugate pair of singularities. At finite times, the factorial FCS has logarithmic singularities corresponding to the zeros of the factorial MGF. With only a single dominant pair of singularities, \( z_0 \) and \( z_0' \), the expression for the high-order factorial cumulants, Eq. (22) simplifies for finite times to \[ \langle \langle F \rangle F \rangle_{(F)} \simeq -\frac{2(m-1)!}{|z_0|^m}\cos\left(m\arg[z_0]\right). \] (52)

From four consecutive high-order factorial cumulants, we can solve this relation for the dominant pair of singularities as functions of time using the methods described in Refs. [62, 41, 20] (see e.g. Appendix B of Ref. [20]). Inserting the solution back into Eq. (52), we can benchmark the approximation against the numerical results. The approximation is shown with circles in Fig. 4 and is seen to be in excellent agreement with the full numerics.

Having extracted the dominant singularities as functions of time in the non-interacting and interacting cases, we may investigate their motion in the complex plane. In Fig. 5 we show the dominant singularities as they move with time. In the non-interacting case (\( U = 0 \)), corresponding to the factorial cumulants shown in Fig. 3, the dominant singularity (marked with a red star) moves along the negative real-axis as expected for generalized binomial statistics. This behavior should be contrasted with the interacting case (\( U > 0 \)), corresponding to the factorial cumulants shown in Fig. 4. In this case, the dominant singularities (marked with blue squares and stars) are no longer real and they now move in the complex plane as functions of time. We stress that this behavior cannot occur for a non-interacting system and should thus be taken as a signature of interactions.

In Fig. 5, we also indicate the points in the complex plane to which the dominant singularities move in the long-time limit (encircled points). As discussed in the previous section, these points correspond to the dominant singularities of the factorial CGF for the zero-frequency factorial cumulants of the current. We extract the position of these singularities by calculating the high order factorial cumulants of the current \( \langle \langle F \rangle F \rangle \) using the recursive scheme developed in Ref. [41], here adapted to the calculation of factorial cumulants. The results for the factorial cumulants of the current are shown in Fig. 6 as functions of the order \( m \). Together with the numerical results, we show the approximation in Eq. (22) with full lines. From four consecutive high-order factorial cumulants we have extracted the parameters entering Eq. (22) using the method proposed in Ref. [41]. Typically, in the long-time limit, the singularities are square-root branch points such that \( \mu_j = -1/2 \) in Eq. (22). Figure 6
teractions cause oscillations of the factorial cumulants. This oscillation which allowed us to treat strong coupling to the leads together with the coherent evolution of electrons inside the DQD. Interestingly, we found that even in cases where the Fano factor of the transport is sub-Poissonian, the high-order factorial cumulants still enable us to detect interactions among the charges passing through the DQD. Finally, we discussed the influence of detector-induced dephasing on the FCS and found that, for this model, such dephasing processes may reduce the oscillations of the high order factorial cumulants.

Here, we have illustrated these ideas with a system consisting of two coherently coupled quantum dots attached to voltage-biased electronic leads. The dynamics of the DQD was described using a Markovian generalized master equation which allowed us to treat strong coupling to the leads together with the coherent evolution of electrons inside the DQD. Interestingly, we found that even in cases where the Fano factor of the transport is sub-Poissonian, the high-order factorial cumulants still enable us to detect interactions among the charges passing through the DQD. Finally, we discussed the influence of detector-induced dephasing on the FCS and found that, for this model, such dephasing processes may reduce the oscillations of the high order factorial cumulants.

Our work leaves several open questions for future research. It is still not clear exactly under what conditions interactions cause oscillations of the factorial cumulants. This will require further careful investigations of the singularities of generating functions in FCS, for example as in the recent work on singularities in FCS for molecular junctions [63]. The answer to this question may moreover come from future work on singularities in FCS for molecular junctions [63].

7 Conclusions

We have discussed our recent proposal to detect interactions among electrons passing through a nano-scale conductor by measuring time-dependent high-order factorial cumulants. For non-interacting electrons in a two-terminal scattering problem, the full counting statistics is always generalized binomial, the zeros of the generating function are real and negative, and consequently the factorial cumulants do not oscillate as functions of the observation time or any other system parameter. In contrast, oscillating factorial cumulants must be due to interactions in the charge transport. In cases where the factorial cumulants oscillate, the zeros of the generating function have moved away from the real-axis and into the complex plane. As we have shown, the motion of the dominant zeros of the generating function can be deduced from the oscillations of the high order factorial cumulants.

Finally, we turn to the influence of detector-induced dephasing. In Fig. 7 we consider the situation where the QPC charge detector is asymmetrically coupled to the two quantum dots, thereby causing dephasing of electrons passing through the DQD. Due to strong Coulomb interactions, the DQD can only be either empty or occupied by one (additional) electron at a time. Without detector-dephasing ($\gamma = 0$), clear oscillations of the tenth factorial cumulant of the current are observed as a function of the energy realignment $\varepsilon$. However, as the dephasing rate is increased, the oscillations are gradually washed out and they essentially vanish in the strong dephasing limit with $\gamma = 0.4\hbar/\Omega$. Thus, in this case, dephasing of the coherent oscillations of electrons inside the DQD seems to reduce the signatures of interactions in the FCS.

![Fig. 6](image1.png)

Fig. 6 Factorial cumulants $\langle \langle I^m \rangle \rangle_F$ of the current as functions of their order $m$. We compare numerical results (marked with symbols) and the approximation (full line) given by Eq. (22). Panel (a) corresponds to the point marked with a blue square in Fig. 2. Panel (b) corresponds to the point marked with a blue star. The corresponding dominant singularities extracted from the numerical results are encircled in Fig. 5.

![Fig. 7](image2.png)

Fig. 7 Factorial current cumulants with detector-induced dephasing. The parameters are $I_D = (1/3)I_F$ and $\gamma = 0$ (black), $0.1\hbar/\Omega$ (red), $0.2\hbar/\Omega$ (purple), $0.4\hbar/\Omega$ (blue). The curves are shifted for clarity.
measurements of oscillating factorial cumulants in interacting nano-scale conductors. In this work, we have focused on Markovian master equations, and it would be interesting to investigate similar phenomena for non-Markovian systems [39–43]. Finally, a new and promising direction of research combines the zeros of generating functions and high order statistics with dynamical phase transitions in stochastic many-body systems [64–66].

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References

2.5 Conclusions

In summary, we have shown in this section that factorial cumulants can indicate whether the electrons in a transport process are interacting. If the high-order factorial cumulants oscillate as a function of a system parameter or time, this is a direct consequence of interactions. This statement, however, cannot be turned around, that is, even for some interacting systems the factorial cumulants may not feature any oscillations and the statistics is generalized binomial, with the zeros of the GF on the real negative axis. We have illustrated our result on several examples of Coulomb blockaded QDs [Secs. 2.2, 2.3, and 2.4]. Furthermore, our test using factorial cumulants is stronger than what can be deduced from the Fano factor alone, that is, even if $F < 1$ the factorial cumulants may pinpoint that there are interactions. Finally, we have shown on an example of a QPC coupled to a DQD that a detector may drastically alter the statistics, so that the oscillations in the factorial cumulants can be damped if the coupling is too strong. However, we stress once more that this is only a particular example. Our findings could possibly be tested experimentally, for example in Coulomb blockade QD systems, where real-time electron counting is possible [89, 88, 90, 91, 94, 96, 97, 98, 78]. One first experiment on a $p$-type GaAs QD has produced non-oscillating factorial cumulants, which is consistent with its modelling as a two-state system [144]. Remarkably, having around two million electron counts, the factorial cumulants were computed up to the 12th order.
Chapter 3

Full counting statistics of Andreev tunneling

This chapter is about a collaboration with V. F. Maisi and Prof. J. P. Pekola from Aalto University School of Science, Aalto, Finland. They designed and performed the experiments and provided us with the counting statistics data that we were able to fit to the model described in Sec. 3.3. Parts of this chapter have been taken, in part verbatim, from our common publication (iv).

3.1 SINIS system

Fig. 3.1 shows an electron micrograph of our SINIS structure, where tunneling takes place from superconducting aluminum leads (S) through the tunnel barriers formed by insulating aluminum-oxide layers (I) onto a normal state copper island (N) and vice versa. The aluminum and copper structures were formed by electron beam lithography, as explained in Sec. 3.2.1 The excess charge $N$ on the island is discrete and is monitored in real-time by the current $I_d$ through a single-electron transistor (SET). The latter is capacitively coupled to the island and its conductance is very sensitive to the island charge state $89, 88, 91, 96, 147, 78$. The excess charge $N$ fluctuates due to the finite temperature and we analyze the counting statistics of the transitions on and off the island. All the statistical information is extracted from the detector current time-traces, after appropriate filtering. They show a step-like behavior with each current plateau corresponding to a given charge state of the copper island. Typical time traces are shown in Figs. 3.4b and 3.5b.

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Figure 3.1: SINIS structure: A metallic normal state (N) island (brown) is connected by insulating (I) tunneling barriers to superconducting (S) leads (green). Between the S (aluminum) and N (copper) layers there is an insulating aluminum oxide layer, forming tunnel barriers where the N and S parts overlap. One of the four tunnel barriers is shown in the inset. The two large arms are designed to increase the island capacitance $C_\Sigma$. The current $I_d$ through a separate single-electron transistor (SET) is sensitive to the charge occupation of the island and is used to read out the number $N$ of excess charges on the island. The copper electrode underneath the structure (yellow) increases the capacitive coupling of the normal state island to the SET, thereby increasing the detector signal-to-noise ratio.

The potential landscape for the fluctuating number of excess electrons $N$ on the copper island can be controlled by applying a voltage $V_g$ to a gate electrode below it. The gate voltage polarizes the island and induces the off-set charge $eN_g = C_g V_g$, where $C_g$ is the gate capacitance. The energy required for charging the island with $N$ electrons is

$$E = E_c (N - N_g)^2,$$

where the charging energy $E_c = e^2/2C_\Sigma$ contains the total island capacitance $C_\Sigma$. The structure is designed to have a large capacitance, such that the charging energy is smaller than the superconducting gap $\Delta$ of aluminum, thereby allowing for Andreev processes [also see Sec. 3.3 and Fig. 3.3] to occur between the island and the superconducting leads [148, 147, 149]. In what follows, we do counting statistics on the one hand for the single electron events when $N_g = 1/2$ and on the other hand for the Andreev events when $N_g = 0$. First we briefly describe the
3.2 Experiment

In this section we give a very brief description of the experimental techniques. For more details, we refer the reader to Sec. I.1. in [99] and to Ch. 3 in [119, 120).

3.2.1 Sample fabrication

The metal structures in Fig. 3.1 were patterned on an oxidized silicon chip using multi-step electron beam lithography. In each step, several resist copolymer layers are added to a silicon chip, then the mask is drawn using a scanning electron microscope (SEM), before its development in a solvent. After that the metal structures are formed using the shadow evaporation technique: several layers of metals are evaporated at different angles. When aluminum is deposited a native oxide forms if the sample is exposed to oxygen after the Al evaporation. Where the different metal layers overlap, metallic contacts are formed, or tunnel barriers in the case there is an oxide layer in between. In a first lithographic step, a copper coupling strip was formed, shown yellow in Fig. 3.1 and covered with a 50 nm thick atomic layer deposition (ALD) grown aluminum-oxide layer. Next, gold leads were patterned, making a direct contact between the actual SINIS structure seen in Fig. 3.1 and the external wiring. Finally, the SET/SINIS structures and the gate leads were patterned. Aluminum and copper were evaporated at different angles. This is why we see two copies of the resist mask in Fig. 3.1 that are shifted with respect to each other. Before deposition, the surface was cleaned with Ar plasma and between evaporations, the aluminum surface was thermally oxidized to form the tunnel barriers. In Fig. 3.1 the experimentally relevant tunnel barriers between the aluminum leads (S) and the copper island (N) are where the green and brown parts overlap [see the inset in Fig. 3.1].

3.2.2 Measurements

Measurements were performed in a dilution refrigerator at 50 mK bath temperature. The bias and gate voltages were provided with room-temperature resistive dividers and the current was amplified with a standard low-noise room-temperature preamplifier. The charging energy $E_c = 40 \mu$eV of the copper island, the super-
conducting gap $\Delta = 210 \, \mu\text{eV}$ of the aluminum leads and the tunnel resistance $R_T = 490 \, \text{k}\Omega$ between the two were determined by measuring the current-voltage characteristics of the SINIS structure. $R_T$ is determined from the asymptotic slope of the current-voltage graph, whereas $\Delta$ and $E_C/\Delta$ are obtained by fitting the sub-gap and transient regions of the current-voltage graphs. Typical current-voltage graphs, in this case for SINIS SETs, can be found in Fig. 4 or Ref. [147].

### 3.2.3 Data analysis

The counting statistics data consists in time traces $I_d$ of 3s duration, where the current is measured every 0.05ms. Examples are shown in Figs. 3.4b, 3.5b, and in the inset of Fig. 3.6. The traces were read off by a computer and numerically filtered with a sharp cut-off at 1 kHz, that is, all the harmonics with a frequency higher than 1 kHz are cut off. This rounds the shape of the signal and makes it less noisy. The choice of the cutoff is a tradeoff between the signal to noise ratio and the time resolution of the measurement. The gate off-set charge $N_g$ was compensated for after each time trace to keep the SET/SINIS structures at the operation points, say, for example at $N_g = 0$ as in Fig. 3.5a.

The SET current time traces $I_d$ feature plateaus, each corresponding to a given charge state of the copper island. The time traces were analyzed with a counting algorithm: when the current crosses a threshold midway between two plateaus a transition out of the given state is detected. Thus, all the statistics $p(n, t)$ in the form of probability histograms, as well as the tunneling rates can be determined, the latter with the help of the following relation [see App. B.1]:

$$\Gamma_{i \rightarrow j} = \frac{N_{i \rightarrow j}}{\sum t_i}, \quad (3.2)$$

where $\Gamma_{i \rightarrow j}$ is the tunneling rate from state $i$ to state $j$, $N_{i \rightarrow j}$ is the total number of transitions from state $i$ to state $j$, and $\sum t_i$ is the total time spent in state $i$. We remark that the result is the better, the longer the time trace, given that there is no significant drift in the rates along the time trace.

Let us consider the $N_g = 0$ case. The charging diagram along with possible transitions are shown in Fig. 3.5a, and will be studied in more detail in the following section. Before we determine the rates with Eq. (3.2), we analyze the waiting time distribution $w(\tau)$ of the $N = 0$ charge state in Fig. 3.2. The theory yields

$$w(\tau) = 2\Gamma_u e^{-2\Gamma_u \tau}, \quad (3.3)$$

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Figure 3.2: Waiting-time distribution of the $N = 0$ charge state for $N_g = 0$. 

a The blue dots show the data obtained after 1 kHz filtering. The time bin size is 0.05 ms. The red line shows the theoretical fit, Eq. (3.3), where the rate $\Gamma_u$ was obtained from Eq. (3.2). We notice that we get too many counts for very short $\tau$ and deduce that these are mis-interpreted transitions between the $N = \pm 1$ states. We therefore exclude the first 25 time bins in b. Plot b is obtained from a by excluding the first 25 bins and integrating over every 25 bins, such that the bin size becomes 1.25 ms. The red line again corresponds to Eq. (3.3) and the error bars are estimated according to Eq. (3.11).
which describes the distribution of the time $\tau$ that it takes for the system to leave the $N = 0$ state. Eq. (3.3) can be understood by noting that the total tunneling rate out of the state $N = 0$ is $2\Gamma_u$ and that in the long-time limit the system will for sure have made the transition, $\int_0^\infty w(\tau)d\tau \xrightarrow{t \to \infty} 1$. In Fig. 3.2 we show the experimental data (blue dots) together with the theoretical fit (red line), Eq. (3.3), where $\Gamma_u$ was determined from (3.2). Fig. 3.2a displays the waiting time data obtained after the 1kHz filtering. The histogram has been rescaled with its numerical integral in order to compare to Eq. (3.3). We notice that for very small $\tau$, that is $\tau \lesssim 1.2\text{ ms}$, we get far too many counts. In fact, about 20% of the counts in Fig. 3.2a fall in the peak around $\tau \lesssim 1.2\text{ ms}$. We interpret this as follows: when the system is in one of the $N = \pm 1$ states [see Fig. 3.5a], it makes transitions $-1 \leftrightarrow 1$ with a large rate, which is comparable to the 1kHz cutoff. If two such Andreev events happen within less than about 1ms, then the counting algorithm might detect that as a transition to the 0-state and back, because of the filtering. Therefore, we add the criterion to the counting algorithm that if we detect the system to prevail less than 1ms in the 0-state, then actually this corresponds to two Andreev events and the system was not in the 0-state at all. With this correction of the counting algorithm we no longer get the hump in the waiting time distribution for small $\tau$, and we only miss about 3% of the single electron events. We still miss some Andreev events, namely the ones followed almost immediately by a single electron event or vice versa, such that the filtered time trace does not capture it. In fact, as discussed below in Sec. 3.3, avalanches of Andreev events occur [see Figs. 3.5b and 3.6] and there is a finite probability that the first or last events in the avalanche are missed by the detector due to the filtering. Finally, after the above correction, we find the tunneling rates as given in Sec. 3.3 using Eq. (3.2) above. In Fig. 3.2b we exclude the first 25 time bins, corresponding to the data for $\tau < 1.25\text{ ms}$, to remove the hump and then integrate over every 25 time bins, which improves the statistics.

Finally, we remark that in the $N_g = 0$ case, very fast sequences of single-electron tunneling events $N = -1 \to 0 \to 1$ or $N = 1 \to 0 \to -1$ could be misinterpreted as Andreev events. That these events can only make up a very small fraction of the counted Andreev events has been shown experimentally in [147]. The percentage of these events can be estimated as follows: the rise time of the detector current corresponding to the 1kHz cutoff is $t_{\text{rise}} \sim 1\text{ ms}$. The probability to observe a dwell time in the zero state shorter than the rise time is computed from (3.3)

$$
\int_0^{t_{\text{rise}}} w(\tau)d\tau \simeq 2\Gamma_u t_{\text{rise}} \simeq 0.02. \quad (3.4)
$$
Figure 3.3: Andreev processes: An electron above the Fermi level of the normal-state island is reflected as a hole and a Cooper pair is formed in one of the superconductors. In the absence of a voltage across the SINIS, the Fermi energy $E_F$ of the normal-state material lies in the middle of the superconducting gap $2\Delta$.

To estimate the probability for the above sequences we additionally have to multiply with the probability that starting from state $N = -1$ the next transition is to $N = 0$, namely $\Gamma_d/(\Gamma_A + \Gamma_d) \simeq 0.3$, as well as the probability that starting from $N = 0$ the next transition is to $N = 1$, namely $\Gamma_u/2\Gamma_u = 1/2$. Thus a fraction of $2 \cdot 0.02 \cdot 0.3 \cdot 0.5 = 0.006$ of the measured Andreev events could be such “false counts”, which can therefore be neglected. This statement is also supported by the good fits we obtain.

### 3.3 Theory and data fit

#### 3.3.1 Andreev reflection

Electronic transport through an interface between a normal state metal (N) and a superconductor (S) is nontrivial. In the N part the charge carriers are electron quasi-particles whereas in the S part, for energies inside the superconducting gap of $2\Delta$, charge carriers are Cooper pairs. When no bias is applied to the SINIS structure the dominant process in the transport is Andreev reflection [125, 118, 126], where an incident electron in the N part is retroreflected as a hole such that a Cooper pair is formed in the superconductor. Electrons thus tunnel in pairs that we shall refer to as Andreev pairs in what follows. A sketch of a simple Andreev reflection is shown in Fig. 3.3. Without a bias, the Fermi energy $E_F$ of the normal-state material lies in the middle of the superconducting gap $2\Delta$. The retroreflected hole has opposite spin and momentum than the incident electron. By time reversal
Figure 3.4: Statistics of single-electron events. a Charging diagram showing Eq. (3.1) with \( N_g = 0.5 \). The charge states with \( N = 0 \) or \( N = 1 \) excess charges on the island are degenerate. The transitions \( 0 \leftrightarrow 1 \) occur with rate \( \Gamma = 49 \text{ Hz} \). Other charge states are energetically unfavorable. b Time trace of the current \( I_d \) in the SET-detector, which switches between two levels corresponding to \( N = 0 \) and \( N = 1 \), respectively. c Measured probability \( p(n, t) \) of single-electron events for different observation times \( t = 10, 100, \) and \( 1000 \text{ ms} \). Poisson distributions given by Eq. (3.5) are shown with full lines.

Symmetry the reverse process can as well take place, where a Cooper pair from the S part tunnels and an incident hole in the N part is retroreflected as an electron of opposite spin and momentum. Let us remark that the process of Andreev reflection is highly spin dependent. However, this does not play a role in our experiment since there are no external magnetic fields applied nor is there a preferred spin direction in the N part. Moreover, with several normal state electrodes coupled to the same superconductor, crossed Andreev reflections may occur where two electrons coming from different electrodes combine into a Cooper pair. This would play a role in, say, a NISIN structure, but not in a SINIS.

In our experiment we have been able to monitor single-electron and Andreev events in real-time. This has also been discussed in [147]. Here, we analyze the counting statistics as shown in this section.

### 3.3.2 Model

When the gate offset charge is tuned to \( N_g = 1/2 \), then the charge states with \( N = 0 \) and \( N = 1 \) are degenerate. The charging diagram corresponding to (3.1)
with \(N_g = 1/2\) is shown in Fig. 3.4a. When electrons tunnel on and off the island, transitions between the 0 and 1 states take place, with the same rate \(\Gamma\) for either direction [Fig. 3.4a]. The states with \(N = -1\) and \(N = 2\) lie much higher in energy and are hardly accessible. The probability \(p(n, t)\) that \(n\) single-electron events have taken place within a counting time \(t\) corresponds to a Poisson process with rate \(\Gamma\), namely

\[
p(n, t) = \frac{(\Gamma t)^n}{n!} e^{-\Gamma t}.
\]  

(3.5)

Fig. 3.4c shows the experimental result for \(p(n, t)\) (dots) along with the Poissonian (3.5) (lines) for three different times \(t\). The rate \(\Gamma\) has been extracted from the detector current time traces according to (3.2) and there are no fitting parameters. We see that the data fit is good and that the process is indeed Poissonian.

The mean \(\langle n \rangle = \Gamma t\) increases linearly with time and the width grows with time according to \(\sqrt{\Gamma t}\).

When the gate offset charge is made smaller, the state \(N = -1\) becomes accessible (also see Fig. 3b in [147]), until for \(N_g = 0\) the states \(N = -1\) and \(N = 1\) are degenerate. The charging diagram corresponding to this situation is depicted in Fig. 3.5a. The \(N = 0\) state is energetically the most favorable, so that the system spends most of the time in it. However, when at some point a single-electron event promotes the system to one of the excited states \(N = \pm 1\), Andreev pairs can tunnel to or from the island, such that its charge state switches between -1 and 1. The possible transitions along with the corresponding rates are shown in Fig. 3.5a. The energy cost for a single-electron event is higher than for an Andreev event, since for a single-electron event an energy of the order of the superconducting gap \(\Delta\) is needed to break a Cooper pair inside the superconductor, in addition to the charging energy \(\pm E_C\). Here, the sign depends on the event, positive for tunneling from 0 to \(\pm 1\) and negative for the opposite direction. Since our structure is designed such that \(E_C < \Delta\) (in fact \(E_C/\Delta \simeq 0.19\)) this explains why we have \(\Gamma_u \ll \Gamma_d \ll \Gamma_A\). In our fits in Fig. 3.5c we used the rates \(\Gamma_u = 11.75\) Hz, \(\Gamma_d = 252\) Hz and \(\Gamma_A = 614.5\) Hz that we obtained from Eq. (3.2).

\(^2\)That the rates be the same within 10% was the criterion to select the data. This check had to be done all along the experiment, after each 3s time trace, in order to compensate for drifts or jumps in the gate voltage.

\(^3\)The data has been selected such that \(\Gamma_{-1 \to 1}\) equals \(\Gamma_{1 \to -1}\) to within 10%. Again, this had to be done all along the experiment to ascertain that \(N_g = 0\). What is more, the values for the rates used in the symmetric model of Fig. 3.5c correspond to the mean values of the electron and hole tunneling rates, that is, we measured \(\Gamma_u = 12.6\) (10.9) Hz, \(\Gamma_d = 254\) (250) Hz and \(\Gamma_A = 617\) (612) Hz. The electron tunneling rates are higher than the hole tunneling rates since the detector noise can assist electron tunneling. The capacitance between the SET and the
Figure 3.5: Statistics of Andreev events. a Charging diagram showing Eq. (3.1) with \( N_g = 0 \). In the ground state, the island is occupied by \( N = 0 \) excess charges. A single-electron event may however bring the island to a state with \( N = \pm 1 \) excess charges. The excitations \( 0 \rightarrow \pm 1 \) occur with the rate \( \Gamma_u = 11.75 \) Hz. Relaxation back to the ground state \( \pm 1 \rightarrow 0 \) happens with the rate \( \Gamma_d = 252.0 \) Hz. In the excited states, the transitions \( -1 \leftrightarrow 1 \) correspond to Andreev events with the rate \( \Gamma_A = 614.5 \) Hz. b Time trace of the current \( I_d \) in the SET-detector, which switches between three levels corresponding to \( N = -1, N = 0, \) and \( N = 1 \), respectively. c Measured probability of Andreev events \( p(n, t) \) for different observation times \( t = 10, 100, \) and 1000 ms. Full lines are theoretical calculations based on Eqs. (1.44) and (3.6). For comparison a Poisson distribution with the same mean as the 1000ms data is indicted with a dashed line.
With these ingredients we can now write a master equation for $|g(z, t)\rangle = \sum_n |p(n, t)\rangle z^n$ of the form (1.44) where the probability vector reads $|p(n, t)\rangle = [p_0(n, t), p_{-1}(n, t), p_1(n, t)]^T$ and with the kernel

$$M(z) = \begin{pmatrix} -2\Gamma_u & \Gamma_d & \Gamma_d \\ \Gamma_u & -(\Gamma_d + \Gamma_A) & z\Gamma_A \\ \Gamma_u & z\Gamma_A & -(\Gamma_d + \Gamma_A) \end{pmatrix}.$$  \hspace{0.5cm} (3.6)

Here, $p_j(n, t)$ is the probability that the island is in charge state $j$ and $n$ Andreev events have occurred during $t$. Supposing that the counting experiment starts when the system is in its steady state $|0\rangle$ and writing $\langle \langle 0 | = [1, 1, 1]$, the GF is given by (1.45). We finally obtain the probability $p(n, t)$ by transforming back according to Eq. (1.43), for which we have used the numerical method introduced in [114] and presented in App. B.2. These are the full lines in Fig. 3.5c, yielding a good fit to the experimental data (dots). Moreover, we observe that the distributions are much broader than a Poissonian. This is indicated in the figure by showing a Poissonian with the same mean $\langle n \rangle$ as the data for $t = 1000\text{ms}$.

### 3.3.3 Counting statistics and avalanches of Andreev events

Fig. 3.5b shows a detector current time trace, where a single-electron transition first promotes the system to the $N = 1$ state and then 7 Andreev events take place before the system relaxes back to the $N = 0$ state. This triggering of an avalanche of Andreev events by an initial single-electron event is typical because $\Gamma_A \gg \Gamma_d$.

We propose to analyze the probability $q(m)$ of the avalanche featuring $m$ Andreev events once the system is promoted to one of the excited charge states:

$$q(m) = \frac{\Gamma_d}{\Gamma_A + \Gamma_d} \left( \frac{\Gamma_A}{\Gamma_A + \Gamma_d} \right)^m.$$

Eq. (3.7) is readily obtained by noticing that initially the system is in an excited state and then $m$ Andreev events take place with probability $\Gamma_A/(\Gamma_A + \Gamma_d)$ before it relaxes back to the $N = 0$ state with probability $\Gamma_d/(\Gamma_A + \Gamma_d)$. A formal way to obtain Eq. (3.7) is given in App. B.3. In Fig. 3.6 we plot the experimental copper island needs to be tuned such that the signal to noise ratio is good, however the detector backaction can be neglected.

\[4\] Alternatively, we can use $p_A(n, t) = p_{-1}(n, t) + p_1(n, t)$, so that the problem becomes 2 by 2. This has been done in publication (iv) [Sec. 3.4] as well as Apps. B.3 and B.4.

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Figure 3.6: Number of Andreev events per avalanche. Experimental results for the distribution of the number of Andreev events in an avalanche. The full line indicates the theoretical prediction given by Eq. (3.7) using $\Gamma_d = 252.0$ Hz and $\Gamma_A = 614.5$ Hz. The inset shows a time trace of the SET-detector current $I_d$ during an avalanche with 16 Andreev events.

result for the statistics $q(m)$ and compare it to Eq. (3.7). We notice once more that we get a good fit. We remark that $q(m)$ only probes two of the rates, namely $\Gamma_A$ and $\Gamma_d$. The shift of the first two points, $m = 0$ and $m = 1$, with respect to the theory can be explained by the fact that the counting algorithm may miss the first or last events in an avalanche, as explained in Sec. 3.2.3. Thus the counting algorithm still misses some Andreev events, despite the correction we introduced (see Sec. 3.2.3).

An instructive interpretation of the physical processes is obtained when we expand the long-time limit CGF to lowest order in $\Gamma_u$. This is justified since $\Gamma_u \ll \Gamma_d, \Gamma_A$. The detailed calculation is given in App. B.4 and follows the idea of [71]. We obtain

$$S(z, t) = 2\Gamma_u t \sum_{m=1}^{\infty} q(m) (e^{mz} - 1) + \mathcal{O}(\Gamma_u^2).$$

Thus, the tunneling processes can be viewed as a superposition of Poisson processes generating an avalanche of $m$ Andreev events. The rate with which the processes
are triggered is $2\Gamma_u$ since the system can enter either the $N = -1$ or the $N = 1$ excited state. Moreover, each Poisson process is weighted by the probability $q(m)$ of observing an avalanche with $m$ Andreev events [Eq. (3.7)]. We see that to lowest order in $\Gamma_u$ the avalanches are immediate, that is, the duration of the avalanches is neglected. Obviously, physically at most one avalanche at a time can take place. For small $\Gamma_u$ avalanches are rare, and in (3.8) correlations among subsequent avalanches are entirely neglected. For larger $\Gamma_u$ these two effects would play a role, however (3.8) delivers a very neat interpretation of what is happening.

To get a feeling for the validity of the approximation (3.8) we propose to look at the LDF for $\Gamma_u \ll \Gamma_d, \Gamma_A$ [Fig. 3.7]. Using the approximation

$$\lambda_0^{(1)}(z) \approx 2\Gamma_u \frac{\Gamma_A (e^z - 1)}{\Gamma_d - \Gamma_A (e^z - 1)}$$

for $\lambda_0(z)$, as derived in App. B.4 we solve Eq. (1.50) for $z_0(I)$. At that point we re-insert $z_0(I)$ into Eq. (1.49) using the exact $\lambda_0(z)$. This yields the green curve in Fig. 3.7. The blue curve is the numerically exact LDF, as explained at the end of Sec. 1.2.4. In Fig. 3.7a we show the numerically exact as well as the approximated LDF for the experimental parameters. Here we have experimental data, which intrinsically always corresponds to a finite time. We display data for a counting time of $t = 1s$ (red) as well as for $t = 3s$ (purple). We see that as the counting time gets longer, the finite time probability curves approach the LDF, that is, the long-time limit is reached on the time scale of a few seconds.

In Fig. 3.7b and 3.7c we show the numerically exact as well as the approximated LDF for half and double the rates $\Gamma_d, \Gamma_A$ with respect to the experiment, respectively. We remark that if the rates $\Gamma_d$ and $\Gamma_A$ are changed by the same factor, then the statistics to get $m$ Andreev events in one avalanche [Eq. (3.7)] stays unchanged. We observe that for very rare events of large $I$ we do get a deviation of the approximation to the exact curve that gets smaller as $\Gamma_u$ gets smaller relatively to the two other rates. However, the approximation captures well the qualitative behavior of the numerically exact LDF, supporting the interpretation based on Eq. (3.8).

\footnote{In order to estimate the deviation of the approximated and exact LDF in Fig. 3.7a, we suggest comparing it to the experimental uncertainties.}
Figure 3.7: LDF for the SINIS system. The blue curve corresponds to the numerically exact LDF and the green curve is the approximation for $\Gamma_u \ll \Gamma_d, \Gamma_A$, as explained in the text. a shows the LDF corresponding to the experimental rates, $\Gamma_u = 11.75\text{Hz}$, $\Gamma_d = 252\text{Hz}$, and $\Gamma_A = 614.5\text{Hz}$, along with two (finite time) experimental plots. Dots correspond to the experimental result, whereas full lines are the fit to the model, similarly as in Fig. 3.5. The red plot is for 1s of counting time and the purple plot corresponds to 3s. The data is plotted in LDF form, as on the l.h.s. of Eq. (1.48). In b we show the curves for unchanged $\Gamma_u$, but half the other rates, that is $\Gamma_d = 126\text{Hz}$ and $\Gamma_A = 307\text{Hz}$. In c the latter two rates are doubled with respect to the experiment, that is $\Gamma_d = 504\text{Hz}$ and $\Gamma_A = 1229\text{Hz}$. We remark that if the rates $\Gamma_d$ and $\Gamma_A$ are changed by the same factor, then the statistics to get $m$ Andreev events in one avalanche [Eq. (3.7)] stays unchanged.
3.3.4 Uncertainties calculation

The error bars in Fig. 3.2 as well as Figs. 3.4 through 3.7 represent the statistical scatter due to the finite number of time bins of size $t$. In general, the uncertainties in a histogram due to the finite statistics can be estimated as follows\cite{150}: Suppose that several statistical samples of a stochastic quantity are measured and the outcome is displayed in a histogram\cite{6}. As the total number of samples, $N_{\text{total}}$, is growing larger and larger the shape of the histogram approaches the parental distribution of our measured quantity\cite{7}. However, in an experiment the sample size $N_{\text{total}}$ is finite and hence there is statistical scatter, corresponding to the purely statistical uncertainty. If $N_n$ denotes the number of counts in bin $n$, then $N_n$ is distributed according to a Poissonian around its expectation value $\langle N_n \rangle$. This can be understood since different counts within the experiment are uncorrelated and the $N_n$ are discrete stochastic variables. We stress that these Poissonians (black dots in Fig. 3.8) have nothing to do with the parental distribution represented in the histogram itself. Knowing the variance of a Poissonian [Eq. (1.25)] and using $N_n$ as an estimate for $\langle N_n \rangle$, we deduce that the error bars in Fig. 3.8 can be estimated by\cite{150}

$$\sigma_{N_n} = \sqrt{N_n}. \quad (3.10)$$

For the probabilities $p_n$ estimated experimentally by $N_n/N_{\text{total}}\xrightarrow{N_{\text{total}} \to \infty} p_n$ we thus estimate the statistical scatter as follows:

$$\sigma_{\text{stat},n} = \sqrt{\frac{N_n}{N_{\text{total}}}} = \sqrt{\frac{p_n N_{\text{total}}}{N_{\text{total}}}} = \sqrt{\frac{p_n}{N_{\text{total}}}}. \quad (3.11)$$

The error bars in Figs. 3.4 through 3.7 correspond to the statistical scatter, Eq. (3.11), using the experimental results $p_n \equiv p(n, t)$. We remark that we have compared the thus obtained uncertainties with the ones estimated using the probabilities from the model. The difference is insignificant. What is more, we notice that the data fits well our model, within these error bars. One could alternatively also take a 10\% drift in the tunneling rates into account and compute a contribution to the total uncertainty $\sigma_n = \sqrt{\sigma_{\text{stat},n}^2 + \sigma_{\text{rates},n}^2}$, using the model expressions for the probabilities:

$$\sigma_{\text{rates},n} = \frac{1}{\sqrt{2}} \left[ \left( \frac{\partial p(n, t)}{\partial \Gamma_u} \sigma_{\Gamma_u} \right)^2 + \left( \frac{\partial p(n, t)}{\partial \Gamma_d} \sigma_{\Gamma_d} \right)^2 + \left( \frac{\partial p(n, t)}{\partial \Gamma_A} \sigma_{\Gamma_A} \right)^2 \right]^\frac{1}{2}, \quad (3.12)$$

\footnote{In our case the statistical samples correspond to the number of counts in time slices of length $t$.}

\footnote{Rescaling with $N_{\text{total}}$ yields the parental distribution.}
Figure 3.8: Uncertainties estimate for a histogram. Due to the finite total number of samples (or counts, not to confuse with the electron counts in the experiment), the number of samples $N_n$ in bin $n$ fluctuates according to a Poisson distribution around the expectation value $\langle N_n \rangle$, which is indicated schematically by the black dots. This statistical scatter can thus be estimated by Eq. (3.10).
where $\sigma_{\alpha} = 0.1\Gamma_{\alpha}/\sqrt{3}$, the factor $0.1/\sqrt{3}$ coming from the supposition that the rates are distributed uniformly within 10% deviation from their mean values. A factor $1/\sqrt{2}$ enters Eq. (3.12) because there are actually three pairs of rates, namely $\Gamma_u(0 \to \pm 1)$, and $\Gamma_d(\pm 1 \to 0)$, and $\Gamma_A(\pm 1 \to \mp 1)$, and the model is symmetric for each pair [see Fig. 3.5a]. Finally, we remark that Eq. (3.12) overestimates the uncertainty due to the fluctuations in the rates. In fact, Eq. (3.12) neglects all correlations in the variation of $\Gamma_u(0 \to 1)$ and $\Gamma_u(0 \to -1)$, and similarly for the other pairs of rates. The rates fluctuate due to fluctuations in the gate voltage $V_g$, or equivalently, in the gate offset charge, since $eN_g = C_g V_g$. Hence, the minimum (orange point) in the Coulomb parabola in Fig. 3.5a is shifted, inducing the energy to promote the system to the $N = -1$ state (green point) to rise by $\Delta E$ and the energy to promote the system to the $N = +1$ state (blue point) to decrease by $\Delta E$, or vice versa. Therefore, at a given time, if $\Gamma_u(0 \to -1)$ varies by $-\Delta \Gamma_u$, then $\Gamma_u(0 \to +1)$ varies by $\Delta \Gamma_u$, and vice versa. An analogous statement holds for the two other pairs of rates. Eq. (3.12) neglects this correlation in time. Since it is difficult to quantify this correlation in an uncertainties estimate and since the fits are good considering the statistical uncertainty [Eq. (3.11)] only, we have not considered the uncertainty (3.12) in our plots.

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8Here, we linearize the parabola around $N = \pm 1$, that is, at $E = E_C$. 89
3.4  Full counting statistics of Andreev tunneling: publication (iv)
Full Counting Statistics of Andreev Tunneling

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We employ a single-charge counting technique to measure the full counting statistics of Andreev events in which Cooper pairs are either produced from electrons that are reflected as holes at a superconductor–normal-metal interface or annihilated in the reverse process. The full counting statistics consist of quiet periods with no Andreev processes, interrupted by the tunneling of a single electron that triggers an avalanche of Andreev events giving rise to strongly super-Poissonian distributions.

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Superconductors are materials that below a critical temperature lose their electrical resistance and thereby allow a supercurrent to flow [1]. Inside the superconducting gap electrons combine into Cooper pairs that carry electrical charge through the superconductor without dissipation. The conversion of a Cooper pair into normal-state electrons (or vice versa) is known as an Andreev process [2]. In a direct Andreev process, an electron in a normal-state material is reflected as a hole at the interface with a superconductor where a Cooper pair is formed. Moreover, with several normal-state electrodes coupled to the same superconductor, crossed Andreev reflections may occur where electrons coming from different electrodes combine into a Cooper pair.

Cooper pairs consist of highly quantum-correlated electrons and may thus serve as a source of entanglement when split into different normal-state electrodes [3–5]. The entanglement of the spatially separated electrons can be detected through current noise measurements [5]. Experiments on superconductor–normal-metal junctions have also revealed a doubling of the shot noise due to the conversion of Cooper pairs into normal-state electrons [6]. However, a complete understanding of the fundamental tunneling processes at a superconductor–normal-metal interface requires measurements beyond the average current and the noise only. Higher-order correlation functions are encoded in the full counting statistics (FCS), which quantifies the probability $p(n, t)$ of observing $n$ charge transfer events during the time span $[0, t]$. The FCS of normal-state electrons has been addressed both theoretically [7–9] and experimentally [10–21]. In contrast, measurements of the FCS of charge transfer into superconductors have so far been lacking despite great theoretical interest [22–31].

In this Letter we report measurements of the FCS of Andreev events occurring between a normal-metal island and two superconducting leads. Our measurements of the FCS allow us to develop a detailed understanding of the elementary tunneling processes at the superconductor–normal-metal interfaces. Figure 1(a) shows our SINIS structure consisting of a normal-state copper island (N) connected by insulating (I) aluminum-oxide tunnel barriers (of a few nanometers thickness [32]) to a pair of superconducting (S) aluminum leads. The structure was

FIG. 1 (color online). SINIS structure and Andreev processes. (a) A metallic normal-state (N) island (brown) is connected by insulating (I) tunneling barriers to superconducting (S) leads (green). The current $I_d$ through a separate single-electron transistor (SET) is sensitive to the charge occupation of the island and is used to read out the number $N$ of excess charges on the island. A copper electrode (yellow) increases the capacitive coupling of the normal-state island to the SET and improves the detector signal-to-noise ratio. (b) An electron above the Fermi level of the normal-state island is reflected as a hole and a Cooper pair is formed in one of the superconductors. Without a voltage across the SINIS, the Fermi energy $E_F$ of the normal-state material lies in the middle of the superconducting gap $2\Delta$. 
patterned on an oxidized silicon chip using standard e-beam lithography techniques. A copper coupling strip was first formed and covered with a 50 nm thick aluminum-oxide layer grown by atomic layer deposition. Gold leads (not shown) were then patterned, making a direct metallic contact to the superconducting leads. Finally, the SINIS structure and the gate leads were formed by e-beam evaporation at different angles. Tunnel barriers were created by thermal oxidation in between.

The number of excess electrons \( N \) on the island is discrete and can be controlled by applying a voltage \( V_g \) to a gate electrode below it. We parametrize the offset voltage by the variable \( N_g = C_g V_g / e \), where \( C_g \) is the gate capacitance and \( e \) the electronic charge. The energy required for charging the island with \( N \) electrons is [33]

\[
E = E_c(N - N_g)^2,
\]

where the charging energy \( E_c = e^2 / 2C_\Sigma \) contains the total island capacitance \( C_\Sigma \). The structure was designed to have a large capacitance, such that the charging energy is smaller than the superconducting gap \( \Delta \) of aluminum, thereby allowing for Andreev processes to occur between the island and the superconducting leads, see Fig. 1(b). The charging energy \( E_c = 40 \mu eV \), the superconducting gap \( \Delta = 210 \mu eV \), and the tunnel resistance \( R_T = 490 \) k\( \Omega \) were determined by measuring the current-voltage characteristics of the SINIS structure. Measurements were performed in a dilution refrigerator at 50 mK bath temperature. The charge state of the island was monitored using a nearby single-electron transistor (SET), whose conductance depends strongly on the number of excess charges on the island [12,13,15,17,19,21,34,35].

To illustrate the basic operating principle of our device we first tuned the offset voltage to \( N_g = 0.5 \). Figure 2(a) shows the energy for different numbers of excess charges. The states \( N = 0 \) and \( N = 1 \) are degenerate, while all other charge states are energetically unfavorable. In this case, single electrons may tunnel on and off the island from the aluminum leads with rate \( \Gamma \). The origin of the single-electron tunneling is addressed in Ref. [36]. Figure 2(b) shows a measured time trace of the current \( I_d \) in the SET detector, which switches between two values corresponding to \( N = 0 \) and \( N = 1 \). We count the number of single-electron tunneling events on and off the island. No voltage bias is applied. Figure 2(c) displays the measured distribution \( p(n, t) \) of the number \( n \) of single-electron events that have occurred during the time span \([0,t] \). The mean number of events increases with the observation time \( t \) and the distribution grows wider. The single-electron events are uncorrelated and should be distributed according to a Poisson distribution

\[
p(n, t) = \frac{(\Gamma t)^n}{n!} e^{-\Gamma t}
\]

with mean \( \langle n \rangle = \Gamma t \). From this mean value we can extract the tunneling rate \( \Gamma \). Figure 2(c) then shows that the FCS of single-electron events indeed is well captured by the Poisson distribution above.

We are now ready to measure the FCS of Andreev events. To this end, we tuned the offset voltage to \( N_g = 0 \). In this case, the charging diagram in Fig. 3(a) is slightly more involved: The lowest-energy state of the system is the configuration with \( N = 0 \) excess charges. However, a
single-electron event may bring the system to one of the excited states with \( N = \pm 1 \) excess charges. The excited states are energetically degenerate and the island can make transitions between \( N = -1 \) and \( N = 1 \) through Andreev processes, where two electrons at a time are converted into a Cooper pair in one of the superconductors or vice versa. 

The Andreev events occur with an average rate \( \Gamma_A \) until the system relaxes back to the ground state through a single-electron event. The current \( I_d \) in the SET detector now switches between three different values corresponding to \( N = -1, 0, \) or \( 1 \), see Fig. 3(b). A fast sequence of single-electron events, \( -1 \rightarrow 0 \rightarrow 1 \), may be mistaken for an Andreev process, \( -1 \rightarrow 1 \), although it is unlikely.) We count the number of Andreev tunneling events to and from the island. Figure 3(c) shows the measured FCS of Andreev events obtained from around 640,000 Andreev processes. Again, the mean value of Andreev events grows with time; however, compared to the FCS of single-electron events, the width of the distributions is surprisingly large and the FCS is strongly super-Poissonian.

To understand quantitatively the FCS of Andreev events, we consider the probabilities \( p_0(n,t) \) and \( p_A(n,t) \) of the island being in the ground state or in one of the excited states, where Andreev events are possible. Both probabilities are resolved with respect to the number \( n \) of Andreev events that have occurred during the time span \([0,t]\). The FCS of Andreev events is \( p(n,t) = p_0(n,t) + p_A(n,t) \), which can be conveniently expressed as the inner product \( p(n,t) = \langle 0 | p(n,t) \rangle \) of the vectors \( \langle 0 \rangle = [1,1] \) and \( | p(n,t) \rangle = [p_A(n,t), p_0(n,t)]^T \) [37,38]. We also introduce the moment generating function \( \mathcal{M}(\chi,t) = \sum_{n=0}^{\infty} p(n,t) e^{i\chi n} = \langle 0 | p(\chi,t) \rangle \) with \( | p(\chi,t) \rangle = \sum_{n=0}^{\infty} e^{i\chi n} | p(n,t) \rangle \). The master equation for \( | p(\chi,t) \rangle \) reads

\[
\frac{d}{dt} | p(\chi,t) \rangle = \mathcal{H}(\chi) | p(\chi,t) \rangle
\]

with the rate matrix (see also Ref. [39])

\[
\mathcal{H}(\chi) = \begin{bmatrix} \mathcal{H}_A(\chi) - \Gamma_d & 2\Gamma_u \\ -2\Gamma_u & -\Gamma_d \end{bmatrix}.
\]

Here \( \mathcal{H}_A(\chi) = \Gamma_A (e^{i\chi} - 1) \) is the generator of uncorrelated Andreev events occurring in the excited states with rate \( \Gamma_A \). The rate for exciting the system is \( 2\Gamma_u \) and \( \Gamma_d \) is the relaxation rate back to the ground state, see Fig. 3(a). The tunneling rates are extracted from the time traces of the SET-detector current [12,13,15,17,19,21,34,35]. Solving Eq. (3), we find \( | p(\chi,t) \rangle = e^{\mathcal{H}(\chi)t} | 0 \rangle \), where \( | 0 \rangle = [2\Gamma_u, \Gamma_d]^T / (2\Gamma_u + \Gamma_d) \) is the stationary probability vector defined by \( \mathcal{H}(0) | 0 \rangle = 0 \) and \( \langle 0 | 0 \rangle = 1 \). The moment generating function is then \( \mathcal{M}(\chi,t) = \langle 0 | e^{\mathcal{H}(\chi)t} | 0 \rangle \). Finally, by inverting the moment generating function for \( p(n,t) \) we can evaluate the FCS of Andreev events for different observation times \( t \).

The theoretical predictions agree well with the measurements in Fig. 3(c) using no fitting parameters. Moreover, a physical interpretation of the nontrivial FCS follows from an expansion of the cumulant generating function \( S(\chi,t) = \log \{ \mathcal{M}(\chi,t) \} \) in the smallest tunneling rate \( \Gamma_u \ll \Gamma_d, \Gamma_A \). At long times, the cumulant generating function is determined by the eigenvalue of \( \mathcal{H}(\chi) \) with the largest real part [40,41]. Importantly, the cumulant generating function for
FIG. 4 (color online). Number of Andreev events per avalanche. The full line indicates the theoretical prediction given by Eq. (6) using $\Gamma_d = 252 \text{ Hz}$ and $\Gamma_A = 615 \text{ Hz}$. The inset shows a time trace of the SET-detector current $I_d$ during an avalanche with $m = 16$ Andreev events.

independent processes is the sum of the cumulant generating functions for the individual processes. To lowest order in $\Gamma_u$, we find at long times

$$ S(\chi, t) = 2\Gamma_u t \sum_{m=1}^{\infty} q(m) (e^{im\chi} - 1) + \mathcal{O}(\Gamma_u^2) \quad (5) $$

with

$$ q(m) = \left( \frac{\Gamma_d}{\Gamma_A + \Gamma_d} \right)^m \frac{\Gamma_A}{\Gamma_A + \Gamma_d}. \quad (6) $$

This shows that the FCS can be approximated as a sum of independent Poisson processes that with rate $2\Gamma_u$ generate avalanches of $m$ Andreev events. Each Poisson process is weighted by the probability $q(m)$ of observing an avalanche with $m$ Andreev events. In this approximation, correlations between subsequent avalanches are neglected together with the duration of the individual avalanches. These correlations would enter in Eq. (5) as higher-order terms in $\Gamma_u$, but would not affect the probabilities in Eq. (6). We note that similar single-electron avalanches have been predicted in molecular quantum transport [42].

To corroborate this physical picture, we turn to the number of Andreev events per avalanche. Figure 4 shows experimental results for the statistics of Andreev events within a single avalanche. The figure illustrates that avalanches with more than ten consecutive Andreev events are possible. This is also evident from the inset showing a time trace of the detector current $I_d$, which switches 16 times between the two levels corresponding to $N = -1$ and $N = 1$ excess charges, respectively. The agreement between the experimental results and the probabilities $q(m)$ in Eq. (6) supports the interpretation that avalanches of Andreev events, triggered by the tunneling of single electrons, give rise to the strongly super-Poissonian FCS.

In summary, we have measured the FCS of Andreev events in an SINIS structure which exhibits super-Poissonian distributions due to avalanches triggered by individual single-electron tunneling events. Our experiment opens a number of directions for future research on charge fluctuations in superconductors. These include experimental investigations of the statistics of entangled electron pairs produced in crossed Andreev reflections as well as controllable Cooper pair production and detection for quantum metrological purposes [43].

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3.5 Conclusions

This chapter treats the first counting statistics experiment of Andreev events, where in each event two electrons tunnel through an interface between a normal metal and a superconductor. We counted Andreev pairs tunneling on and off a normal metallic (i.e. copper) island from superconducting aluminum leads, as shown in Fig. 3.1. The experiment was designed and performed by V. F. Maisi, J. P. Pekola and coworkers at Aalto University School of Science, Aalto, Finland. We have been able to describe the time-dependent counting statistics with a master equation, without any fitting parameters, and obtained an excellent fit. We observe experimentally from the detector current time traces [Fig. 3.5b and inset of Fig. 3.6] that the Andreev events occur in avalanches. The reason is that the rate for the Andreev events is the largest of all, while a single-electron event has to promote the system from the ground state for Andreev processes to occur. The Andreev rate is the largest since no Cooper pairs have to be broken in the superconductor in this process, as opposed to single-electron tunneling events. As a consequence the probability distribution is much broader than a simple Poisson distribution. Expanding the cumulant generating function in the smallest rate, we can interprete the process as a superposition of Poisson processes of avalanches of all possible lengths, weighted with the probability for the given avalanche length. We remark that similar avalanches, however for single-electron events, have been predicted in molecular quantum transport [71]. The real-time detection of individual Andreev events opens up several directions for future research. These include the production and study of entangled electron pairs from crossed Andreev reflection as well as the controllable Cooper pair production and detection for quantum metrological purposes.
Chapter 4

Charge and heat current noise in the presence of rectification

For small voltages, in a two-terminal conductor the electrical and thermal conductance are insensitive to the direction of current flow. At higher voltages and temperatures the energy dependence of the transmission plays a role and the conduction is sensitive to the direction of current flow. Interestingly in the fluctuation spectra the rectification of a conductor is seen already in linear order. For such rectifying conductors the charge (or heat) current noise has a component linear in voltage (or in the temperature difference), which is related to temperature times the rectification coefficient through the fluctuation relation (1.12). In this chapter, we investigate the frequency dependent charge (heat) current fluctuation spectra for a conductor subject to a small voltage (temperature difference). Due to charge pile-up (heat accumulation) in the sample, fluctuation spectra measured in the left and right reservoirs can be different. The correlation between currents in the right and left reservoirs is related to the autocorrelations only if the charge (heat) pile-up in the interior of the sample are taken into account. We illustrate these statements for a one-channel wire treated as a simple one-dimensional double barrier problem.

4.1 Noise correlation spectra are in general asymmetric in bias

Quite generally a semiconductor can distinguish right from left [51]. There are notable exceptions to this rule. In particular, in linear response, both the charge and the thermal conductance are insensitive to whether current flows from left to
right or vice versa. However, as we allow for larger voltages or bigger temperature differences, there are corrections which are quadratic in the applied voltage difference and quadratic in the temperature difference which exist across the conductor. Such rectification coefficients exist generally whenever the transmission probability is energy dependent. Below we shall explain that there are two sources to get a finite rectification coefficient. The first one is an asymmetrically applied bias. This generally entails nonzero rectification even in dc driven noninteracting conductors. However, strictly speaking, we may not call the conductor rectifying unless it produces a current with a dc component in response to a pure ac bias.\footnote{If the bias is applied asymmetrically we will always have a dc component. However, we oversimplified things here a little. In dc transport, the transmission probability for electrons from left to right is the same as the transmission from right to left. This is not the case in ac transport due to the dwell time. Therefore a spatially asymmetric conductor may be rectifying, even in a noninteracting formalism with a symmetrically applied ac bias.} The second source of rectification requires interactions. A rectifying conductor distinguishes left from right, that is, it responds differently to a bias applied in the left than in the right reservoir. What is important in both cases, is that an energy dependent transmission probability generates different transmission behavior for the electrons and holes. Interestingly, the charge and heat current noise reflects the presence of rectification already to linear order in voltage and to linear order in the temperature drop. That this has to be so is required by second order fluctuation relations [Eqs. (1.12) and (1.2)], which is an extension of the fluctuation dissipation relation to the non-equilibrium transport regime. These relations link the second order response coefficients to the derivatives with respect to voltage (temperature difference) of the noise. We are interested in the frequency-dependent fluctuation spectra of charge and heat current noise in the linear voltage range and use the fluctuation relations as a benchmark to which the noise spectra converge in the zero-frequency limit.

The above discussion leads to the prediction that the noise of a conductor can be lowered below its Nyquist-Johnson value, Eq. (1.9), by application of a voltage! There exists a range of values of $V$ where the noise is smaller than the Johnson-Nyquist value. We illustrate the behavior of the noise as a function of voltage and temperature in Fig. 4.3.

The rectification coefficient of mesoscopic structures $G^{(2)}$ has been theoretically and experimentally investigated in a series of works for nearly a decade. Fluctuation relations for rectifying mesoscopic conductors have been the subject of several theoretical works [58, 59, 60, 61] treating especially the role of interaction and time-reversal symmetry breaking in the presence of a magnetic field [60, 152]. However,
to date, there is seemingly only one single experiment (on a mesoscopic ring pierced by an Aharonov-Bohm flux) which investigates the near-equilibrium noise \[62\]. In the present work we are interested in the frequency-dependence of near equilibrium fluctuations. The frequency-dependence of these spectra is relevant for instance in the mesoscopic drag effect in which the charge fluctuations of one conductor are rectified by another nearby conductor \[153, 154\].

The introduction given thus far has treated current fluctuations. It can be repeated for heat transport and heat fluctuations. The heat current fluctuation spectrum is defined as

\[
S_{\alpha\beta}^{th}(\omega) = \frac{1}{2} \int dt e^{i\omega t} \langle \Delta \hat{J}_\alpha(t) \Delta \hat{J}_\beta(0) + \Delta \hat{J}_\beta(0) \Delta \hat{J}_\alpha(t) \rangle,
\]

(4.1)

where \( \hat{J}_\alpha \) is the heat current operator in reservoir \( \alpha \), and \( \Delta \hat{J}_\alpha = \hat{J}_\alpha - \langle \hat{J}_\alpha \rangle \). There exists an analogous fluctuation relation to Eq. (1.12) connecting the second order heat conductance, \( G_{th}^{(2)} \), to the derivative of the zero-frequency heat fluctuation:

\[
S_{th}^{(1)} = -2k_BT \cdot G_{th}^{(2)},
\]

(4.2)

with \( S_{th}^{(1)} \equiv (-\beta)\partial_{\Delta T} S_{th}^{th}\big|_{\Delta T=0,\omega=0} \), where \( \beta \) is the inverse temperature \( \beta = 1/k_BT \) and \( k_B \) is Boltzmann’s constant. Here, there is no bias and we have expanded the heat current according to

\[
J = G_{th} \left( \frac{\Delta T}{T} \right) + \frac{1}{2} G_{th}^{(2)} \left( \frac{\Delta T}{T} \right)^2 + \ldots
\]

(4.3)

In App. C.3 we consider expanding the current and heat current in the bias \( V \) and the temperature difference \( \Delta T/T \).

In this work we use scattering matrix theory to compute the current and heat current noise [Eqs. (1.8) and (4.1)] in a two-terminal conductor. The relations we present here have been derived in Ref. [23] and Refs. [155, 156, 157], respectively. Let

\[
s \equiv \begin{bmatrix}
  r_L & t_R \\
  t_L & r_R
\end{bmatrix}
\]

(4.4)

denote the scattering matrix relating the outgoing scattering states to the ingoing

\footnote{Expanding in \( \Delta T/T \) the higher order fluctuation relations for the current and heat current are analogous [App. C.3].}
ones. Here, the block $r_L$ has dimensions $N \times N$ where $N$ is the number of channels in the left reservoir. Similarly for the other blocks, so that if $M$ is the number of channels in the right reservoir then the dimensions of $s$ are $(N+M) \times (N+M)$. The noise and cross-correlation spectra (1.8) and (4.1) for noninteracting electrons and small frequencies can then be expressed using the scattering matrix, Eq. (4.4), and the Fermi distributions in the reservoirs [22, 23, 155, 156]. The noise correlation spectrum in the left reservoir reads

$$S_{LL}(\omega) = \frac{e^2}{\hbar} \int dE \text{Tr} \left\{ \left[ I - r_L^\dagger(E) r_L(E + \hbar \omega) \right] \left[ I - r_L^\dagger(E + \hbar \omega) r_L(E) \right] F_{LL}(E, \omega) + r_L^\dagger(E) t_R(E + \hbar \omega) t_R^\dagger(E + \hbar \omega) r_L(E) F_{LR}(E, \omega) + t_R^\dagger(E) r_L(E + \hbar \omega) r_L^\dagger(E + \hbar \omega) t_R(E) F_{RL}(E, \omega) + t_R^\dagger(E) t_R(E + \hbar \omega) t_R^\dagger(E + \hbar \omega) t_R(E) F_{RR}(E, \omega) \right\}, \quad (4.5)$$

where

$$F_{\alpha\beta}(E, \omega) = f_\alpha(E) [1 - f_\beta(E + \hbar \omega)] + f_\beta(E + \hbar \omega) [1 - f_\alpha(E)]. \quad (4.6)$$

If $V_\alpha$ denotes the bias and $T_\alpha$ the temperature in reservoir $\alpha$, and $\mu$ is the equilibrium chemical potential, the Fermi distribution function reads

$$f_\alpha(E) = \frac{1}{e^{(E - \mu - eV_\alpha)/k_B T_\alpha} + 1}. \quad (4.7)$$

For small bias $V_L, V_R$, we can evaluate the scattering matrix elements at $V_L = V_R = 0$. Thus, (4.5) will only depend on bias through the Fermi functions. The expression for the heat current noise spectra has a similar structure, however, as electrons of different energy carry different amounts of heat, we find an additional
factor of \((E + \hbar \omega / 2 - \mu)^2\) in the integrand \[155, 156, 157\]:

\[
S_{th}^{LL}(\omega) = \frac{e^2}{\hbar} \int dE \left( E + \frac{\hbar \omega}{2} - \mu \right)^2 \times \quad
tr \left\{ \left[ \mathbb{1} - r_{L}^{\dag}(E) r_{L}(E + \hbar \omega) \right] \left[ \mathbb{1} - r_{L}^{\dag}(E + \hbar \omega) r_{L}(E) \right] F_{LL}(E, \omega) \right.
\]
\[
+ r_{L}^{\dag}(E) t_{R}(E + \hbar \omega) t_{R}^{\dag}(E + \hbar \omega) r_{L}(E) F_{LR}(E, \omega) \right.
\]
\[
+ t_{R}^{\dag}(E) r_{L}(E + \hbar \omega) r_{L}^{\dag}(E + \hbar \omega) t_{R}(E) F_{RL}(E, \omega) \right.
\]
\[
+ t_{R}^{\dag}(E) t_{R}(E + \hbar \omega) t_{R}^{\dag}(E + \hbar \omega) t_{R}(E) F_{RR}(E, \omega) \right\}. \quad (4.8)
\]

Eqs. (4.5) and (4.8) suggest that the energy dependent phases in the scattering amplitudes matter, and not only the transmission coefficient. The remaining noise correlation spectra are given in App. C.1.

The charge on the considered mesoscopic conductor fluctuates. In response to the fluctuating reservoir potentials there is momentary charge pileup in the conductor. Therefore, at finite frequency there is charge noise,

\[
S_{QQ}(\omega) = \frac{1}{2} \int dt e^{i \omega t} \langle \hat{Q}(t) \hat{Q}(0) + \hat{Q}(0) \hat{Q}(t) \rangle, \quad (4.9)
\]

which by the continuity equation is given by (see App. C.2)

\[
\sum_{\alpha, \beta} S_{\alpha \beta} = \omega^2 S_{QQ}(\omega). \quad (4.10)
\]

Here, \(\hat{Q}\) is the charge operator in the volume defined by the mesoscopic conductor.

In a similar way, the heat fluctuation spectrum \(S_{th}^{HH}\) on the mesoscopic conductor is given by

\[
\sum_{\alpha, \beta} S_{\alpha \beta}^{th} = \omega^2 S_{th}^{HH}(\omega). \quad (4.11)
\]

The density of states in the conductor, that is, the states accommodating the piled up charges, can be described using the Wigner-Smith type matrix \(\mathcal{N}\). In the zero-frequency limit we have \[30\]

\[
\mathcal{N}_{\alpha \beta} = \frac{1}{2\pi i} \sum_{\gamma} s_{\gamma \alpha}^{\dagger} \frac{ds_{\gamma \beta}}{dE}. \quad (4.12)
\]
The density of states is determined by the diagonal entries of $\mathcal{N}$ as follows

$$N(E) = \text{Tr} [\mathcal{N}(E)].$$

(4.13)

Also, the entries of $\mathcal{N}$ can be used to express the charge noise spectrum $S_{QQ}$ (also see App. C.2).

In general the current noise spectrum $S_{\alpha\alpha}(\omega, V)$ is asymmetric in bias, and only in the zero temperature and zero frequency limit do we find the symmetric and well-known shot noise expression (1.7). An example of an asymmetric noise spectrum is given in Fig. 4.3. For a small bias and $T_L = T_R \equiv T$, the asymmetry in bias of the current noise spectrum is described by its first order expansion in $V \equiv V_L - V_R$ around $V = 0$. At zero frequency this coefficient, $S^{(1)} \equiv \partial V S_{LL}^{(1)}|_{V=0, \omega=0}$, is related to the current rectification coefficient $G^{(2)} \equiv \partial^2 V I|_{V=0}$ by the fluctuation relation (1.12). The rectification coefficient can be readily found from the Landauer-Büttiker formula for the current, Eq. (1.5) [see App. C.3]. Writing

$$f(E) = \frac{1}{e^{(E-\mu)/k_B T} + 1},$$

(4.14)

as well as $V_L = \eta V$ and $V_R = (\eta - 1)V$ with $0 \leq \eta \leq 1$ we find

$$G^{(2)} = \frac{e^3}{h} \left(2\eta - 1\right) \int_0^\infty dE \mathcal{T}(E) \left. \frac{\partial^2 f(E)}{\partial E^2} \right.,$$

(4.15)

where $\mathcal{T}(E) = \text{Tr}[t^L_L t_L] = \text{Tr}[t^R_R t_R]$ is the transmission coefficient of the conductor. The zero of energy corresponds to the band bottom in the reservoirs. We remark that for a symmetrically applied bias, $\eta = \frac{1}{2}$, electron-hole symmetry is restored and the rectification is zero. Therefore, in noninteracting theory and dc driven conductors we can only get a finite rectification coefficient if the bias is applied asymmetrically around the chemical potential $\mu$. This is because the Landauer-Büttiker formula for two terminals, Eq. (C.9), from which Eq. (4.15) is derived, yields an anti-symmetric current, $I(V) = -I(-V)$ at $\eta = \frac{1}{2}$.

Using Eqs. (1.12) and (4.15) we find the zero temperature limit for $S^{(1)}$:

$$S^{(1)} \xrightarrow{T \to 0} 2 k_B T \frac{e^3}{h} \left(2\eta - 1\right) \left. \frac{d\mathcal{T}}{dE} \right|_{E_F},$$

(4.16)

---

3We remark that with this parametrization, in order to interchange $V_L$ and $V_R$ we need to change $\eta \to (1-\eta)$ and $V \to -V$. 

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where $E_F$ is the Fermi energy.

For zero bias but finite temperature gradient between the left and right reservoirs the maths are similar. The situation, however, is different in the following way: The Fermi energy is a property of the conductor, and we can apply a small bias around it, either symmetrically or asymmetrically. The equilibrium temperature, on the other hand, is not a system property, unless some large heat bath provides a reference temperature $T$. If not, we suggest that we should assume a symmetrically applied temperature gradient. This will lead to zero heat current rectification and a symmetric heat current noise spectrum in the temperature gradient, in the noninteracting formalism. Let us now suppose, however, that there is a reference temperature $T$. The affinity corresponding to energy current is $1/k_B T \equiv \beta$. Therefore, derivatives are taken with respect to $\Delta \beta$. However, expansions are most often written in terms of powers of $\Delta T/T$, where $\Delta T = T_L - T_R$ and $T$ is the equilibrium temperature. Thus the obtained fluctuation relations for the heat transport are analogous to the finite bias and charge current case, except for a difference in sign at every other order since $\Delta (1/T) = (-1/T) \Delta (T)/T$. We also stress at this point that we are doing finite temperature expansions. When zero temperature limits are taken, we must take care to first take the limit $\Delta T \to 0$. The heat current rectification, $G^{(2)}_{\text{th}} \equiv \beta^2 \partial^2_{\Delta \beta} J|_{\Delta \beta = 0}$, then reads

$$G^{(2)}_{\text{th}} = \frac{1}{\hbar} (2\alpha - 1) \int_{0}^{\infty} dE \, T^3 E (E - \mu)^3 \frac{\partial^2 f(E)}{\partial E^2},$$  \hspace{1cm} (4.17)$$

where $1/T_L = 1/T + \alpha \Delta (1/T)$ and $1/T_R = 1/T + (\alpha - 1) \Delta (1/T)$ and $T$ is the equilibrium temperature. Again, if the temperature gradient is applied symmetrically, $\alpha = \frac{1}{2}$, there is no heat current rectification. The second fluctuation relation for the heat current is then given by Eq. (4.2).

We now proceed to enquire where the factor $(2\eta - 1)$ in Eq. (4.16) comes from. For small voltages $V_L$ and $V_R$, the energy dependent transmission can be approxi-

\footnote{We remark that our result (4.16) only agrees with the result by Levchenko and Kamenev, Eq. (6) in Ref. [154], for $\eta = 0$. It is unclear to the author what the difference in their development is. They might just have kept the gauge invariant part in their result, which would not be correct.}

\footnote{To lowest order in $\Delta T/T$ this is equivalent to writing $T_L = T + \alpha \Delta T$ and $T_R = T + (\alpha - 1) \Delta T$, since}

$$\frac{1}{T + \alpha \Delta T} \simeq \frac{1}{T} \left( 1 - \alpha \frac{\Delta T}{T} \right)$$

and

$$\frac{1}{T + \alpha \Delta \left( \frac{1}{T} \right)} = \frac{1}{T} - \alpha \frac{\Delta T}{T^2}.$$
Figure 4.1: For small bias voltages $V_L$ and $V_R$ the ratio between electron and hole currents can be deduced by linearizing the energy dependent transmission $T(E)$ around the chemical potential $\mu$. This is indicated with the red triangles. For an energy-independent transmission, electron and hole currents will always be balanced and there is no rectification.

\[ T(E) \simeq T(\mu) + \frac{dT}{dE}|_{E=\mu} \cdot (E - \mu). \tag{4.18} \]

The difference between the electron and hole currents, which are indicated in red in Fig. 4.1, is proportional to $V_L^2 - V_R^2$, which in our parametrization reads

\[ V_L^2 - V_R^2 = (2\eta - 1)V. \tag{4.19} \]

Higher orders in the $I - V$ expansion, that is $G^{(3)}$ and onwards, correspond to higher orders in the $T(E)$ expansion \[\text{[4.18]},\] beyond linear order.

Since noninteracting theory does not take Coulomb interactions into account, its results are not (always) gauge invariant. Any result should only depend on voltage differences. We clearly see that $V_L^2 - V_R^2$ is not gauge invariant.

Potential fluctuations in the reservoirs, that is, electron-hole excitations of different frequencies, generate momentary charge pileup in the mesoscopic conductor. The finite frequency noise captures this effect, whereas the zero frequency (dc) limit describes fluctuations averaged over long times. The accumulated charges on the conductor create a local screening potential redistributing charges in the ca-

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capacitively coupled close neighborhood. This response can clearly only be described by a formalism that includes Coulomb interactions. In order to express current conservation, also these displacement currents must be taken into account. A non-interacting theory will therefore not yield current conservation at finite frequency, nor gauge invariance, which is of course unphysical. In scattering theory there is a way to introduce Coulomb interactions self-consistently so as to restore current conservation. Typically, the density of states on the conductor [Eq. [143]] is calculated in the noninteracting framework and then capacitive coupling to a nearby gate is considered [28, 30]. This means that the Laplacian in the Poisson equation is replaced by at least one capacitance. The potential drop inside the conductor, from \( V_L \) to \( V_R \), is continuous. Simplifying the problem, we can approximate the potential drop by steps, in the most simple case by one step. Thus we include one gate with gate voltage \( V_g \). In order to compute second order conductances, we need to include the dependence on voltages of the internal potential \( U \) of the conductor up to linear order:

\[
U = U_0 + u_L V_L + u_R V_R + u_g V_g .
\]

By gauge invariance \( u_L + u_R + u_g = 1 \). The transmission coefficient now also depends on the internal potential. Thus, the voltage dependence of the current [Eq. (C.9)] does not only come in through the Fermi functions, but also through \( T(E, U) \). Writing

\[
I = G_L V_L + G_R V_R + \frac{1}{2} G_{LL} V_L^2 + \frac{1}{2} G_{RR} V_R^2 + G_{LR} V_L V_R + \text{h.o.t.} ,
\]

and assuming\[158\]

\[
\frac{\delta T}{\delta e U} \bigg|_{V_L=V_R=0} \simeq - \frac{\partial T}{\partial E} \bigg|_{V_L=V_R=0} ,
\]

6This assumption corresponds to the random phase approximation where the screening charge becomes proportional to the Lindhard function. Moreover the long wavelength limit or quasiclassical approximation is taken. See \[158\].
we find for the first and second order conductances

\[
G_L = \frac{e^2}{h} \int dE \mathcal{T}(E, U_0) \left( -\frac{\partial f}{\partial E} \right) = -G_R, \tag{4.23}
\]

\[
G_{LL}^{(2)} = \frac{e^3}{h} \int dE \left[ \mathcal{T}(E, U_0) \frac{\partial^2 f}{\partial E^2} + 2 u_L \left( -\frac{\partial T}{\partial E} \right) \left( -\frac{\partial f}{\partial E} \right) \right], \tag{4.24}
\]

\[
G_{RR}^{(2)} = -\frac{e^3}{h} \int dE \left[ \mathcal{T}(E, U_0) \frac{\partial^2 f}{\partial E^2} + 2 u_R \left( -\frac{\partial T}{\partial E} \right) \left( -\frac{\partial f}{\partial E} \right) \right], \tag{4.25}
\]

\[
G_{LR}^{(2)} = \frac{e^3}{h} \int dE \left( -\frac{\partial T}{\partial E} \right) \left( -\frac{\partial f}{\partial E} \right) (u_R - u_L). \tag{4.26}
\]

If in the modelling of the internal potential we include one gate that is capacitively coupled to our conductor via a capacitance \( C \), this capacitance suffices to determine the parameters \( u_{L,R} \) and \( u_g \) in (4.20). This is done by the charge balance of the piled up charge on the conductor in response to each of the voltages, \( V_L \) and \( V_R \) [159]:

\[
Cu_{L,R} = e^2 N_{L,R}(\mu) - e^2 N(\mu) u_{L,R} \quad \Rightarrow \quad u_{L,R} = \frac{e^2 N_{L,R}(\mu)}{C + e^2 N(\mu)}. \tag{4.27}
\]

Above, the quantities \( N_\alpha \) are the injectivities [28] from reservoir \( \alpha \), namely

\[
N_\alpha \equiv N_{\alpha\alpha}, \tag{4.28}
\]

where \( N_{\alpha\alpha} \) is given by (4.12). Similarly we find \( u_g \):

\[
Cu_g = C - e^2 N(\mu) u_g \quad \Rightarrow \quad u_g = \frac{C}{C + e^2 N(\mu)}. \tag{4.29}
\]

Thus, in order to compute the rectification coefficients in the self-consistent formalism, we need to estimate the capacitance \( C \) of the conductor, or equivalently, its charging energy \( E_C = e^2 / 2C \). Finally, let us remark that here we suggested to evaluate the density of states, \( N \), as well as the injectivities \( N_{L,R} \) at the equilibrium chemical potential \( \mu \). If the density of states is strongly varying, one would have to solve the equation

\[
Cu_{L,R} = e^2 N_{L,R}(U) - e^2 N(U) u_{L,R}, \tag{4.30}
\]

instead of (4.27), with \( U \) given by (4.20) and \( U_0 = \mu \). Similarly we would have to replace (4.29).

In the presence of perfect screening, \( C \to \infty \) in Eqs. (4.27) and (4.29) above, we
again find the noninteracting result \(4.15\) for the rectification. Let us now consider
the example of strong Coulomb interactions, \(C \to 0\) in Eqs. \(4.27\) and \(4.29\), such
that the charging energy of the conductor becomes infinite and no accumulation of
charge is possible. We write \(u_L = u\), \(u_R = 1 - u\), and \(u_g = 0\) in Eq. \(4.20\). The
second order in bias voltage contribution becomes

\[
\frac{e^3}{2h} \int dE \mathcal{T}(E, U_0) \frac{\partial^2 f}{\partial E^2} (V_L^2 - V_R^2)
\]

\[
+ \frac{e^3}{h} \int dE \left( -\frac{\partial T}{\partial E} \right) \left( -\frac{\partial f}{\partial E} \right) [uV_L^2 + (1 - u)V_R^2 + (1 - 2u)V_LV_R]
\]

\[
= \frac{e^3}{h} \int dE \left( -\frac{\partial T}{\partial E} \right) \left( -\frac{\partial f}{\partial E} \right) (1 - 2u)(V_L - V_R)^2,
\]

where the second line is obtained by partial integration. We see that \(4.31\) does not
depend at all on how the bias is applied between the left and right reservoirs, that is,
there is no dependence at all on our \(\eta\) parameter. However, if the screening is close
to perfect and we introduce at least one parameter \(u_g \neq 0\), then the rectification
will depend on how the bias is applied. We thus see that Coulomb interactions
counteract the dependence of the rectification on \(\eta\). Moreover, Eq. \(4.31\) shows
that if the conductor distinguishes left from right, that is \(u \neq \frac{1}{2}\), we get a finite
rectification, and this even if the bias is applied symmetrically. We call such a
conductor rectifying.

In the presence of a temperature gradient across the conductor we need to
consider a dependence of the internal potential \(U\) on voltages and temperature
gradients. The treatment is analogous except that there is no gauge invariance
constraint for the coefficients of the temperature gradients. We refer the reader to
Ref. \[158\].

4.2 Ballistic quantum wire

To illustrate our results we consider a wire (see Fig. \[4.2a\]), which we model as a
simple double barrier problem (see Fig. \[4.2b\]). The barriers describing the connec-
tion of the wire to bulk reservoirs are taken to be energy independent. The energy
dependence of the total scattering matrix of the wire arises through the traversal
and multiple reflections inside the wire. For scatterers which are partially reflect-
ing, the density of states as a function of the chemical potential exhibits peaks at
the energies of the isolated quantum wire. This oscillatory structure is reflected
in the second order conductance and is reflected in the Fermi energy dependence
Figure 4.2: A single channel quantum wire of length $l$ connected to left and right reservoirs via energy independent scattering matrices $s_1$ and $s_2$, respectively. Thus, the only energy dependence of the total scattering matrix of the wire arises through the traversal and multiple reflections inside the wire. (a) is a cartoon of the system whereas (b) schematically shows the energy landscape. For simplicity we consider the transverse energy in the wire to coincide with the band bottom in the reservoirs. However, if the chemical potential $\mu$ is large compared to the level spacing $\Delta(\mu)$ we expect that this assumption should not considerably influence our results. The small bias $V$ is asymmetrically applied whenever $\eta \neq \frac{1}{2}$.

of the fluctuation spectra. We neglect self-consistent effects (interactions) both in the dc-transport and in the calculation of the fluctuation spectra. Our main purpose here is only to emphasize that the noise spectra at finite temperature are in general asymmetric in the applied bias and that there are three different noise spectra corresponding to fluctuations in the left and right contact and the correlations between these currents. This should be contrasted with theoretical work which represents the frequency dependent noise by a single quantity.

The reason that even a two terminal conductor has three different spectra is that at non-zero frequency there is charge and heat pile-up inside the sample as soon as transmission and reflection amplitudes depend on energy. We remark that energy dependent scattering is associated with a time delay, which leads to the charge pileup and energy storage inside the sample.
4.2.1 Model

We consider a single channel ballistic quantum wire connected to two reservoirs. Fig. 4.2 shows its schematic. We describe the non-perfect contacts to the left (L) and right (R) reservoirs by two scattering matrices, $s_1$ and $s_2$, respectively. The total scattering matrix describing transport through the whole wire, $s_{\text{total}}$, depends on $s_1$ and $s_2$. Moreover, we have to sum over all the possible multiple reflections inside the wire. To keep our model as simple and instructive as possible, we suppose that $s_1$ and $s_2$ do not depend on the energy of the incident electrons. Thus, the whole energy dependence of the total scattering matrix comes through the traversal and multiple reflections of the electrons inside the wire. For a ballistic wire the transverse energy is quantized and the longitudinal motion is free, that is, the longitudinal energy dispersion is quadratic,

$$E = \frac{\hbar^2 k^2}{2m},$$

(4.32)

where $k$ is the longitudinal momentum of the electron and $m$ is its effective mass. For simplicity we suppose that the transverse energy in the wire is the same as the band bottom in the reservoirs. We expect that this assumption does not consider-
Figure 4.4: Density of states, Eq. (4.13), for identical and symmetric contacts, Eq. (4.36). The length of the wire sets the length unit, $l = 1$, and we also set $m = 1$. The energy, $E$, is measured in units of $E_0$, Eq. (4.35). The envelope of the peak heights goes like $\sqrt{E}$. The broadening of the peaks is due to the finite transmission.

ably influence our results for large enough chemical potential $\mu$. Writing

$$s_j = \begin{bmatrix} r_{jL} & t_{jR} \\ t_{jL} & r_{jR} \end{bmatrix}, \quad j = 1, 2,$$

the expression for $s_{\text{total}}$ reads

$$s_{\text{total}} = \begin{bmatrix} r_{1L} + \frac{t_{1L} r_{2L} t_{1R} e^{2ikl}}{1 - r_{1L} r_{2L} e^{2ikl}} & \frac{t_{2R} t_{1R} e^{ikl}}{1 - r_{1L} r_{2L} e^{2ikl}} \\ \frac{t_{1L} t_{2L} e^{ikl}}{1 - r_{1L} r_{2L} e^{2ikl}} & r_{2R} + \frac{t_{2R} r_{1R} t_{2L} e^{2ikl}}{1 - r_{1L} r_{2L} e^{2ikl}} \end{bmatrix},$$

where $l$ is the wire length.

The density of states in the wire is given by Eq. (4.13). If the transmissions $t_{j\alpha}$, $j = 1, 2$, $\alpha = L, R$, all tend to zero, then the wire becomes an infinite square well. At finite transmissions of the reservoir contacts the delta peaks in the density of states broaden. The resonant $k$-values are given by the real parts of the poles of the total scattering matrix and are given by $k_n = (n\pi - \theta/2)/l$, with $n$ an integer.
and $\theta = \arg [r_1 r_2 l]$. The resonant energies are therefore

$$E_n(\theta, l) = \frac{\hbar^2}{2m} \left( \frac{n\pi - \frac{\theta}{2}}{l} \right)^2. \quad (4.33)$$

An example for a density of states in the wire is shown in Fig. 4.4. The longer the wire the closer the peaks in the density of states become. The level spacing $\Delta(E)$ depends on energy and can be written as follows:

$$\Delta(E) = \frac{\partial E}{\partial k} \Delta k = \hbar \sqrt{\frac{2E}{m \pi l}}. \quad (4.34)$$

Finally, in the following subsection we will be using

$$E_0 \equiv E_1(\theta = 0, l = 1) = \frac{\hbar^2}{2m} \left( \frac{\pi}{l} \right)^2\quad (4.35)$$
as an energy unit.

### 4.2.2 Results

Below we look at some specific results for our ballistic wire model. We expect to see interesting features mainly in the quantum limit $\Delta(\mu) > \hbar \omega > k_B T > eV$.

In the following specific examples we look at a symmetric wire with $s_1 = s_2 = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & 1 \\ 1 & i \end{bmatrix}$.\quad (4.36)

Fig. 4.5 explicitly verifies the fluctuation relation (1.12) showing $S^{(1)}$ as well as temperature times the rectification, $2k_B T \cdot G^{(2)}$, both in units of $e^3/\hbar$ and as functions of $\mu/E_0$. Importantly, close to the resonances (4.33) the rectification is zero, changing from negative to positive values, that is, from hole-like to electron-like transport. At the zeros transport is electron-hole symmetric, however already slightly off resonance $S^{(1)}$ is nonzero, meaning that the noise spectra are asymmetric in bias voltage. In the middle between two resonances electron-hole symmetry is again restored and there the slope is smaller. This structure can be understood by performing one partial integration in Eq. (4.15): for small temperature and a large enough chemical potential such that there is no boundary term the rectification is essentially proportional to the energy derivative of the transmission,
Figure 4.5: Fluctuation relations. (a) The red line shows $S^{(1)}$ and the black dots correspond to temperature times the rectification, $2k_B T \cdot G^{(2)}$, both in units of $\frac{e^3}{h}$ and as functions of $\mu/E_0$. The fluctuation relation (4.12) is verified. (b) The red line shows $S^{(1)}_{\text{th}}$ and the black dots correspond to $-2k_B T \cdot G^{(2)}_{\text{th}}$, that is, the r.h.s. of Eq. (4.17), which is also verified. Units are $(k_B T)^2/h = \frac{1}{h} \left( \frac{2E_b}{\pi^2} \right)^2$. In both cases, (a) and (b), we consider symmetric contacts, Eq. (4.36). Parameter values are $l = 1$, $m = 1$, $k_B T = 1$ and $\eta = \alpha = 0.8$. We remark that for $\eta = \frac{1}{2}$ or $\alpha = \frac{1}{2}$ there is no rectification. The main difference between the two cases, (a) and (b), is that the current rectification changes sign as a function of the chemical potential in the reservoirs, whereas the heat current rectification does not. This is because electrons and holes have opposite charge but carry heat of the same sign.
Thus we see that the observed oscillations are a consequence of the energy-dependent phase acquired by electrons traversing the wire. The zeros of the rectification are a bit shifted away from the resonances since $-\partial E f$ is equal to a delta function in the zero temperature limit only but is broadened at finite $k_B T$. Finally, we remark that to break electron-hole symmetry we need $\eta \neq \frac{1}{2}$ [see Eq. (4.15)]. The example of Fig. 4.5a suggests that by tuning the equilibrium chemical potential in the reservoirs we can change between electron-like and hole-like transport. Even at zero frequency, by Eq. (1.12), this translates into a noise spectrum in the reservoirs that is asymmetric in bias voltage. This can be seen in Fig. 4.3.

In Fig. 4.5b we verify the analogous fluctuation relation for the heat current, Eq. (4.2). The relation states that to first order in $\Delta T/T$ the asymmetry in the heat current noise at zero frequency is given by the heat current rectification, Eq. (4.17). At the lower resonances we observe double peaks, whereas we see simple peaks close to the higher resonances. The double peaks appear if the width of the transmission peak at a given resonance is smaller than a few $k_BT$. Thus, for smaller transmissions of the wire contacts the double peaks can be seen up to higher values of $\mu/E_0$. In the heat current rectification we observe no zeros as opposed to the charge current rectification case since electrons and holes carry opposite charge but heat of the same sign. Formally this can be seen by considering that the integrand in (4.17) when $\mu$ coincides with a resonant energy [Eq. (4.33)] is locally symmetric. Let us remark here that if there is no well-defined reference temperature $T$, then we need to consider a symmetrically applied temperature gradient, yielding zero heat current rectification in the noninteracting formalism.

In Fig. 4.6 we plot $S_{LL}^{(th)}(\omega) + S_{LR}^{(th)}(\omega)$ as functions of frequency. We again consider identical contacts to the reservoirs, [Eq. (4.33)], so that $S_{LL}^{(th)}(\omega) = S_{RR}^{(th)}(\omega)$. Also, in the one-channel case we have $S_{LR}^{(th)}(\omega) = S_{RL}^{(th)}(\omega)$ and we therefore find that the fluctuation spectrum of the accumulated charge in the wire is $S_{QQ} = 2(S_{LL} + S_{LR})/\omega^2$. Similarly we get for the heat fluctuations $S_{HH}^{th} = 2(S_{LL}^{th} + S_{LR}^{th})/\omega^2$. At zero frequency our non-interacting theory is current conserving [App. C.2] and we have $S_{LL}^{(th)}(0) = S_{RR}^{(th)}(0) = -S_{LR}^{(th)}(0) = -S_{RL}^{(th)}(0)$. Here we clearly see that this ceases to be true at finite frequencies. In Fig. 4.6 we consider frequencies smaller than the level spacing at the chemical potential, $\hbar \omega/\Delta(\mu) < 1$, and thus avoid strong effects due to the next resonance.

The noise spectra themselves oscillate as functions of the chemical potential $\mu$. This can be seen in Fig. 4.7 where we compare the charge or heat fluctuations, respectively, for three different frequencies. The periodicity of the oscillations is
Figure 4.6: Charge and heat pileup: (a) shows $S_{LL} + S_{LR}$ in units of $\frac{e^2 k_B T}{\hbar}$ as a function of frequency. $\hbar \omega$ is in units of the level spacing [Eq. (4.34)] at the chemical potential. At zero frequency we have $S_{LL}(\omega = 0) + S_{LR}(\omega = 0) = 0$, which expresses charge conservation. Due to potential fluctuations of frequency $\omega$ in the reservoirs charge is momentarily piled up in the wire, which can be seen in the charge noise of the same frequency [Eq. (4.10)]. In this specific case, where we consider a symmetric wire [Eq. (4.36)] such that $S_{LL} = S_{RR}$ and the one-channel case where $S_{LR} = S_{RL}$, we have $S_{QQ} = 2(S_{LL} + S_{LR})/\omega^2$. Parameter values are $\mu = 100k_B T$, which is at the fifth resonance, $eV = 0.2k_B T$, $\eta = 0.8$, $\Delta T = 0$, $m = 1$, and $l = 1$. (b) In the same way as for charge, heat is momentarily piled up in the wire, the fluctuations of which can be expressed through $S_{HH}$, Eq. (4.11). Again here, in the symmetric [Eq. (4.36)] and one-channel case, we have $S_{th}_{HH} = 2(S_{th}^{LL} + S_{th}^{LR})/\omega^2$. At zero frequency there is no heat pileup, that is, heat is conserved. Parameter values are $\mu = 100k_B T$, $eV = 0$, $\Delta T = 0.2T$, $\alpha = 0.8$, $m = 1$, and $l = 1$.

Figure 4.7: Charge and heat noise spectra as functions of $\mu/E_0$ for three different frequencies: $\hbar \omega = 1k_B T$ (blue line), $\hbar \omega = 2k_B T$ (purple line) and $\hbar \omega = 4k_B T$ (red line). (a) Charge noise [Eq. (4.10)] for $eV = 0.2k_B T$, $\eta = 0.8$ and $\Delta T = 0$. (b) Heat noise [Eq. (4.11)] for $\Delta T = 0.2T$, $\alpha = 0.8$ and $eV = 0$. We observe clear oscillations in the noise spectra in the range $\Delta(\mu) > \hbar \omega$, reflecting the resonance condition of the wire. We have considered symmetric contacts, Eq. (4.36), and we have set $m = 1$ and $l = 1$. 
again given by the resonance condition (4.33), where the peaks in the charge and
heat fluctuations are close to a resonance. For the set of parameters we have used,
the charge and heat noise increases with frequency. However, we remark that this
behavior is not generic. Finally, let us stress that the leftmost part of the plots, for
chemical potentials close to the band bottom, $\mu \simeq 0$, is not representative since we
have considered the transverse energy in the wire to equal the band bottom, which
is a very specific assumption made for simplicity, and since we want $\Delta(\mu) > \hbar \omega$.
However, this only concerns the very leftmost part of the plots, $\mu/E_0 < 1$.

4.3 Conclusions

We have considered charge current fluctuations for a current driven by a finite bias
voltage $V$ as well as heat current fluctuations for a heat current driven by a finite
temperature gradient $\Delta T$. We have pointed out that at finite temperature the
noise spectrum in the reservoirs is in general asymmetric in the bias voltage $V$.
Therefore, expanding the noise spectra around $V = 0$ there is a term linear in $V$.
At zero frequency this term is related to the rectification coefficient by Eq. (4.12).
In a similar way we can derive a fluctuation relation [Eq. (4.2)], linking the first
order expansion coefficient in $\Delta T/T$ of the heat current noise to the heat current
rectification. This shows that also the heat current fluctuations are asymmetric
in the applied temperature gradient $\Delta T$. When there is a temperature gradient,
however, there is in general no well-defined reference temperature, like the Fermi
energy for the electrical bias. Therefore we need to consider a symmetrically applied
temperature gradient, which in a noninteracting formalism will yield a symmetric
heat current noise spectrum. The asymmetry in the noise spectrum is essential in
Coulomb drag setups where a current flowing through one conductor entails a drag
current in a nearby conductor that is only capacitively coupled to the first [153, 154].
It is only when electron-hole symmetry is broken, that is, for a finite rectification
that such a drag current can be generated.

We have shown how an energy-dependent transmission entails rectification, even
for a spatially symmetric, noninteracting and dc biased conductor. However, using
scattering matrix theory we have shown that in order to get finite rectification
the bias needs to be applied asymmetrically. Coulomb interactions counteract
this effect, however the effect is not fully compensated for, but is still there if
the screening is nearly perfect. On the other hand, an interacting conductor may
distinguish left from right. If interactions are strong and if this inherent chirality
is very marked, then this is typically the most important source of rectification.

Energy dependent scattering causes charge pileup in the mesoscopic conductor and thus a delay time. The caused density of states in the conductor, or equivalently (multiplying by $\hbar$) the delay time, is given by relation (4.13). Since in a noninteracting theory screening is completely ignored, it does not capture charge conservation at finite frequency. However, it yields the fluctuation spectra of the accumulated charge or heat inside the mesoscopic conductor [Eqs. (4.10) and (4.11)]. Starting from the density of states [Eq. (4.13)], interactions can be included self-consistently [28, 30, 160].

Our findings are essential in the description of Coulomb drag phenomena where the drag current relies on energy dependent tunneling and typically depends on the finite frequency excess noise in the drive conductor. For the response to be finite, electron-hole symmetry needs to be broken so that electron and hole drag currents do not cancel [153, 154]. This requires, by Eq. (1.12), that the noise spectra in the leads be asymmetric in bias. Also, tuning energy dependent phases, as could be envisaged in a ballistic wire by simply tuning the equilibrium chemical potential in the reservoirs, would enable us to tune the drag current along with its sign in a Coulomb drag setup.

Regarding the analogies in the different thermoelectric relations as for example Eqs. (1.12) and (1.2), we understand that we can produce a charge or heat drag current by using a charge or heat drive current by capacitive coupling of two mesoscopic conductors. This opens a large field of potential applications.
Summary and outlook

In this thesis we have looked at some aspects of interactions influencing the full counting statistics or the finite frequency noise. In Ch. 2 we have shown that oscillating factorial cumulants, as functions of any system parameter or of time, are a clear indication of interactions. This applies to any generic two-terminal conductor. We stress that the above statement cannot be turned around, that is, even for some interacting systems the factorial cumulants may not feature any oscillations and the statistics is generalized binomial, with the zeros of the generating function on the real negative axis. Importantly, our test using factorial cumulants is stronger than what can be inferred from the Fano factor, that is, even if $F < 1$ the factorial cumulants may oscillate and indicate that the system is interacting. We mention that a detector may considerably alter the statistics. This is what a particular example of a QPC detector coupled to a DQD has shown. Our findings could possibly be tested experimentally, for example in Coulomb blockade QD systems, where real-time electron counting is possible [89, 88, 90, 91, 94, 96, 97, 98, 78]. We remark that one first experiment, where the factorial cumulants have been measured up to the 12th order, has produced non-oscillating factorial cumulants, consistently with its modeling as a two-state system [144]. Further theoretical investigations could address multi-terminal conductors and cross-correlations, where the condition on the zeros of the generating function is less simple [110]. Moreover, we point out that oscillating factorial cumulants are a sufficient condition to show the presence of effective electron-electron interactions. An important question to ask is what a necessary and sufficient condition would correspond to, or in other words, exactly what kind of interactions are needed to make the zeros of the generating function move out from the real negative axis into the complex plane, thus causing the factorial cumulants to oscillate. Qualitatively speaking, we can say that if two real negative zeros move towards each other and become degenerate when a parameter is varied, they will split into a pair of complex conjugate zeros. We therefore suggest that the origin of degeneracy points of zeros could be inves-
tigated. However we point out that the zeros of the generating function depend nontrivially on system parameters or time, so that it is not straightforward to infer what structure the Hamiltonian would need to have in order to produce oscillating factorial cumulants. Finally, the connection to the waiting time distribution could be inquired. Strong correlations due to interactions are expected to affect the waiting times between subsequent electrons transferred through a conductor. Thus, it could be instructive to investigate what type of waiting time distributions characterize systems featuring oscillating factorial cumulants.

In Ch. 3 we describe an experiment where the full counting statistics of Andreev events have been measured. In each Andreev event two electrons tunnelled through a thin insulating layer separating a normal metal island from superconducting leads. Andreev events could occur when a single-electron tunneling event promoted the metallic island to an excited charge state. Such a single-electron event could trigger an avalanche of Andreev events of variable length before another single-electron event brought the island back into the ground charge state. Because the statistics features these avalanches it turns out to be strongly super-Poissonian. We remark that we counted each pair of electrons that tunnelled either way as one event. Thus we point out that the broadening of the probability distribution has nothing to do with the doubling of shot noise that is observed in the context of Andreev reflection when a pair of electrons is counted as two electrons [161]. The tunneling rates entering our master equation calculation were measured and we obtain an excellent fit without any fitting parameters. Two electrons forming a Cooper pair are highly correlated. In fact, they are orbitally and spin entangled. This makes them interesting candidates to be used for the production and controlled manipulation of entangled electron pairs. The real-time detection of individual Andreev events as presented in our experiment is an important step in this direction.

For small voltages, in a two-terminal conductor the electrical and thermal conductance are insensitive to the direction of current flow. At higher voltages and temperatures the energy dependence of the transmission plays a role and the conduction is sensitive to the direction of current flow. In Ch. 4 we are interested in rectification properties, that is, the second order response of the current to the applied voltage, or of the heat current to the applied temperature gradient, respectively. We show that there are two distinct sources of rectification for stationary transport. Starting from the Landauer-Büttiker formula for the current [Eq. (1.5)] for two terminal conductors, we show that the rectification is in general finite even for a dc biased noninteracting and spatially symmetric conductor. This is the case if the bias is applied asymmetrically around the equilibrium chemical potential.
Coulomb interactions counteract this effect, however if screening is nearly perfect there will in general still be a term in the rectification that depends on how the bias is applied. Another source of rectification is an intrinsic asymmetry of the conductor, such that it responds differently to a bias applied in the left than in the right reservoir. What is important in both cases, is that an energy dependent transmission probability generates different transmission behavior for the electrons and holes. Interestingly in the fluctuation spectra the rectification of a conductor is seen already in linear order. For such rectifying conductors the charge (or heat) current noise has a component linear in voltage (or in the temperature difference), which is related to temperature times the rectification coefficient through a higher order fluctuation relation. This shows that in general the noise spectrum in the reservoirs is asymmetric in the applied bias. The above discussion leads to the prediction that the noise of a conductor can be lowered below its Nyquist-Johnson value by application of a voltage! Our discussions were made in the context of stationary transport. Even though a noninteracting and dc biased conductor may have a finite rectification coefficient if the bias is applied asymmetrically, this does not mean that we can call the conductor rectifying, since an asymmetrically applied bias is not an inherent property of the conductor. We suggest to call a conductor rectifying if it generates a finite rectification due to its inherent chirality. Such a conductor is expected to generate a current with a dc component in response to a pure ac bias. In this context it would be interesting to make the analogous investigations to our Ch. 4 but for ac driving, for example using Floquet scattering matrix theory [24]. Our findings could be applied to Coulomb drag setups, where the excess current noise in one conductor generates a drag current in a capacitively coupled nearby conductor [153, 154]. What is more, time-dependent rectification effects are the underlying principle of quantum pumping [2].
Appendix A

Appendices for Ch. 2

A.1 Calculation of (factorial) cumulants at finite times

This appendix corresponds verbatim to App. A of publication (i) and we repeat it here for completeness.

Here we describe our method for calculating (factorial) cumulants at finite times without explicitly evaluating the (F)CGF. We begin with a master equation of the form given by Eq. (1.44),

\[ \partial_t |g(z, t)\rangle = M(z) |g(z, t)\rangle, \]

(A.1)

for a system with \( N \) states. Next, we substitute in the master equation \(|g(z, t)\rangle\) by \(|\tilde{g}(z, t)\rangle\) and \(M(z)\) by \(\tilde{M}(z)\), where \(|\tilde{g}(z, t)\rangle = |g(e^z, t)\rangle\) and \(\tilde{M}(z) = M(e^z)\) for ordinary cumulants, and \(|\tilde{g}(z, t)\rangle = |g(z+1, t)\rangle\) and \(\tilde{M}(z) = M(z+1)\) for factorial cumulants, respectively. For \( z = 0 \), the master equation then reads

\[ \partial_t |\tilde{g}^{(0)}(t)\rangle = \tilde{M}^{(0)} |\tilde{g}^{(0)}(t)\rangle, \]

(A.2)

having defined \(|\tilde{g}^{(0)}(t)\rangle = |\tilde{g}(0, t)\rangle\) and \(\tilde{M}^{(0)} = \tilde{M}(0)\). Taking instead \( m = \)
1, 2, . . . , \kappa \text{ consecutive derivatives with respect to } z \text{ (evaluated at } z = 0), \text{ we obtain}

\begin{align*}
\partial_t \langle \tilde{g}^{(1)}(t) \rangle &= \tilde{M}^{(1)} \langle \tilde{g}^{(0)}(t) \rangle + \tilde{M}^{(0)} \langle \tilde{g}^{(1)}(t) \rangle, \\
\partial_t \langle \tilde{g}^{(2)}(t) \rangle &= \tilde{M}^{(2)} \langle \tilde{g}^{(0)}(t) \rangle + 2\tilde{M}^{(1)} \langle \tilde{g}^{(1)}(t) \rangle + \tilde{M}^{(0)} \langle \tilde{g}^{(2)}(t) \rangle, \\
\vdots \\
\partial_t \langle \tilde{g}^{(k)}(t) \rangle &= \sum_{j=0}^{k} \binom{k}{j} \tilde{M}^{(j)} \langle \tilde{g}^{(k-j)}(t) \rangle, \tag{A.3}
\end{align*}

where we have introduced the notation \( \langle \tilde{g}^{(m)}(t) \rangle = \partial_z^m \langle \tilde{g}(z, t) \rangle \big|_{z \to 0} \) and \( \tilde{M}^{(m)} = \partial_z^m \tilde{M}(z) \big|_{z \to 0} \). The (factorial) moments of order \( m \leq k \) are then

\begin{align*}
\langle n^m \rangle_{\langle F \rangle}(t) &= \langle \langle \tilde{0} | \tilde{g}^{(m)}(t) \rangle \rangle, \tag{A.4}
\end{align*}

depending on the substitutions made above. The vector \( \langle \tilde{0} \rangle = [1, 1, \ldots, 1] \) contains \( N \) elements equal to unity.

We solve the system of coupled equations (A.2, A.3) by introducing the auxiliary vector

\begin{align*}
|G(t)\rangle = [\tilde{g}(t), \tilde{g}^{(1)}(t), \ldots, \tilde{g}^{(k)}(t)]^T \tag{A.5}
\end{align*}

containing \( N(k + 1) \) elements. The equation of motion for \( |G(t)\rangle \) reads

\begin{align*}
\partial_t |G(t)\rangle = \hat{M} |G(t)\rangle \tag{A.6}
\end{align*}

where according to Eqs. (A.2, A.3)

\begin{align*}
\hat{M} = \begin{bmatrix}
\tilde{M}^{(0)} & 0 & 0 & 0 & 0 \\
\tilde{M}^{(1)} & \tilde{M}^{(0)} & 0 & 0 & 0 \\
\tilde{M}^{(2)} & 2\tilde{M}^{(1)} & \tilde{M}^{(0)} & 0 & 0 \\
\vdots & \vdots & \vdots & \ddots & \ddots \\
\tilde{M}^{(k)} & n\tilde{M}^{(k-1)} & \tilde{M}^{(0)} & \tilde{M}^{(0)} & \tilde{M}^{(0)}
\end{bmatrix}. \tag{A.7}
\end{align*}

is a matrix of dimensions \( N(k + 1) \times N(k + 1) \). We proceed by solving Eq. (A.6) as

\begin{align*}
|G(t)\rangle = e^{\hat{M}t} |G(t = 0)\rangle. \tag{A.8}
\end{align*}
Here, the initial condition as counting begins reads

\[ |G(t = 0)\rangle = |0\rangle, 0, 0, \ldots, 0]^T, \quad (A.9) \]

and contains the stationary state \( |0\rangle \), which solves \( \tilde{M}(0)|0\rangle = 0 \), followed by \( Nk \) elements equal to 0. Even for large dimensions of \( M \) we may calculate numerically the matrix exponentiation \( e^{M_t} \) for a given time \( t \) and obtain \( |G(t)\rangle \) via Eq. (A.8).

Having determined the (factorial) moments using Eq. (A.4), the corresponding (factorial) cumulants are obtained via the relation

\[ \langle \langle n \rangle \rangle^m_F = \langle n \rangle^m_F - \sum_{k=1}^{m-1} \binom{m-1}{k-1} \langle \langle n \rangle^k_F \rangle \langle \langle n^{m-k} \rangle_F \rangle. \quad (A.10) \]

For the particular \( N = 3 \) state model studied in this work [publication (i)], we could easily calculate the first \( m = 50 \) (factorial) cumulants at finite times.

### A.2 Recursive scheme

In this appendix we present the recursive scheme developed in Sec. III A of Ref. [106] for completeness, as we have employed it in several calculations entering this thesis. Let \( \tilde{M}(z) \equiv M(e^z) \) for the calculation of the usual cumulants or \( \tilde{M}(z) \equiv M(z + 1) \) for the factorial cumulants. We use the fact that \( \tilde{M}(0)|0\rangle = 0 \) to define \( \Delta \tilde{M}(z) \) such that

\[ \tilde{M}(z) = \tilde{M}(0) + \Delta \tilde{M}(z). \quad (A.11) \]

Moreover, we define the projectors \( P = |0\rangle \rangle \langle \langle 0| \) and \( Q = I - P \). With these definitions \( P \) projects onto the null space of \( \tilde{M}(0) \), whereas \( Q \) projects away from the null space. Therefore the operator \( Q \tilde{M}(0)Q \) is invertible and we write the pseudoinverse as

\[ R = Q \tilde{M}(0)^{-1} Q. \quad (A.12) \]

Above, in Sec. [124] we have defined \( \lambda_0(z) \) as the eigenvalue of \( \tilde{M}(z) \) such that \( \lambda_0(0) = 0 \), and we shall denote the corresponding eigenvector \( |0(z)\rangle \), namely

\[ \tilde{M}(z)|0(z)\rangle = \lambda_0(z)|0(z)\rangle, \quad (A.13) \]
with $|0(z = 0)\rangle = |0\rangle$. Now, the two important relations follow, on which the recursive scheme is then based:

\[
\lambda_0(z) = \langle \langle 0 | \Delta \widetilde{M}(z) | 0(z) \rangle \rangle, \quad (A.14)
\]

\[
|0(z)\rangle = |0\rangle + R \left[ \lambda_0(z) I - \Delta \widetilde{M}(z) \right] |0(z)\rangle. \quad (A.15)
\]

Expanding the quantities in the game in powers of the counting variable $z$, we recognize that only the eigenvector has a zeroth order component:

\[
\lambda_0(z) = \sum_{n=1}^{\infty} \frac{z^n}{n!} \langle \langle I^n \rangle \rangle, \quad (A.16)
\]

\[
|0(z)\rangle = \sum_{n=0}^{\infty} \frac{z^n}{n!} |0^{(n)}\rangle, \quad (A.17)
\]

\[
\tilde{M}(z) = \sum_{n=1}^{\infty} \frac{z^n}{n!} \tilde{M}^{(n)}. \quad (A.18)
\]

This is the trick how we can now compute the (factorial) current cumulants order by order following the recursive scheme, from left to right and top to bottom:

| $n$ | $\lambda_0(z)$ | $|0(z)\rangle$ |
|-----|----------------|----------------|
| 0   | 0              | $|0\rangle$    |
| 1   | $\langle \langle I \rangle \rangle$ | $|0^{(1)}\rangle$ |
| 2   | $\langle \langle I^2 \rangle \rangle$ | $|0^{(2)}\rangle$ |
| $\vdots$ | $\vdots$ | $\vdots$ |

Formally,

\[
\langle \langle I^n \rangle \rangle = \sum_{m=1}^{n} \binom{n}{m} \langle \langle 0 | \tilde{M}^{(m)} | 0^{(n-m)} \rangle \rangle, \quad (A.19)
\]

\[
|0^{(n)}\rangle = R \sum_{m=1}^{n} \binom{n}{m} \left[ \langle \langle I^n \rangle \rangle - \tilde{M}^{(m)} \right] |0^{(n-m)}\rangle. \quad (A.20)
\]
A.3 Determination of the dominant pair of complex conjugate singularities

Using the notation of Eqs. (1.58) and (1.59) we derive a method to approximately determine the position of the dominant pair of complex conjugate singularities. For this, we need four consecutive (factorial) cumulants, of order \( m - 3 \) through \( m \).

Approximating the high order (factorial) cumulants using one pair of complex conjugate singularities \(|z_0| e^{\pm i \arg z_0}\), we arrive at Eq. (1.61), namely
\[
\langle \langle n^m \rangle \rangle_F \simeq \frac{2 |A_0| B_{m, \mu_0}}{|z_0|^{m+\mu_0}} \cos [(m + \mu_0) \arg z_0 - \arg A_0].
\] (A.21)

Thus
\[
\langle \langle n^{m+1} \rangle \rangle \simeq \frac{2 |A_0| B_{m+1, \mu_0}}{|z_0|^{m+1+\mu_0}} \cos [(m + 1 + \mu_0) \arg z_0 - \arg A_0]
= \frac{2 |A_0| B_{m+1, \mu_0}}{|z_0|^{m+1+\mu_0}} \left\{ \cos [(m + \mu_0) \arg z_0 - \arg A_0] \cos [\arg z_0]
- \sin [(m + \mu_0) \arg z_0 - \arg A_0] \sin [\arg z_0] \right\},
\] (A.22)

and
\[
\langle \langle n^{m-1} \rangle \rangle \simeq \frac{2 |A_0| B_{m-1, \mu_0}}{|z_0|^{m-1+\mu_0}} \cos [(m - 1 + \mu_0) \arg z_0 - \arg A_0]
= \frac{2 |A_0| B_{m-1, \mu_0}}{|z_0|^{m-1+\mu_0}} \left\{ \cos [(m + \mu_0) \arg z_0 - \arg A_0] \cos [\arg z_0]
+ \sin [(m + \mu_0) \arg z_0 - \arg A_0] \sin [\arg z_0] \right\}.
\] (A.23)

Thus
\[
\frac{\langle \langle n^{m-1} \rangle \rangle}{\langle \langle n^m \rangle \rangle} \simeq \frac{|z_0|}{\mu_0 + m - 1} \left\{ \cos [\arg z_0]
+ \tan [(m + \mu_0) \arg z_0 - \arg A_0] \sin [\arg z_0] \right\},
\] (A.24)

and
\[
\frac{\langle \langle n^{m+1} \rangle \rangle}{\langle \langle n^m \rangle \rangle} \simeq \frac{\mu_0 + m}{|z_0|} \left\{ \cos [\arg z_0]
- \tan [(m + \mu_0) \arg z_0 - \arg A_0] \sin [\arg z_0] \right\},
\] (A.25)
and therefore we get

\[ (\mu_0 + m - 1) \langle \langle n^{m-1} \rangle \rangle + \frac{|z_0|^2}{\mu_0 + m} \langle \langle n^{m+1} \rangle \rangle = 2 \Re [z_0]. \]  \hspace{1cm} (A.26)

Increasing \( m \rightarrow m + 1 \) we get

\[ (\mu_0 + m) \langle \langle n^m \rangle \rangle + \frac{|z_0|^2}{\mu_0 + m + 1} \langle \langle n^{m+1} \rangle \rangle = 2 \Re [z_0]. \]  \hspace{1cm} (A.27)

Let us now write \( m + 2 \rightarrow m, m + 1 \rightarrow m - 1, \ldots \). We need to solve, for some high order \( m \),

\[
\begin{bmatrix}
1 - \frac{1}{\mu_0 + m - 2} \langle \langle n^{m-1} \rangle \rangle \\
1 - \frac{1}{\mu_0 + m - 1} \langle \langle n^m \rangle \rangle
\end{bmatrix}
\begin{bmatrix}
2 \Re [z_0] \\
|z_0|^2
\end{bmatrix}
= \begin{bmatrix}
(\mu_0 + m - 3) \langle \langle n^{m-3} \rangle \rangle \\
(\mu_0 + m - 2) \langle \langle n^{m-2} \rangle \rangle
\end{bmatrix}.
\]  \hspace{1cm} (A.28)

Finally,

\[ \Im [z_0] = \sqrt{|z_0|^2 - \Re [z_0]^2}. \]  \hspace{1cm} (A.29)

Once we have found the dominant pair of complex conjugate singularities, \( |z_0| e^{\pm i \arg z_0} \), we need to find \( A_0 \) in order to approximate the (factorial) cumulants with Eq. \( \text{[A.21]} \).

\[
\langle \langle n^m \rangle \rangle \simeq \frac{2 |A_0| B_{m, \mu_0}}{|z_0|^{m+\mu_0}} \cos [(m + \mu_0) \arg z_0 - \arg A_0]
= \frac{2 |A_0| B_{m, \mu_0}}{|z_0|^{m+\mu_0}} \left\{ \cos [m \arg z_0] \cos [\arg A_0 - \mu_0 \arg z_0]
+ \sin [m \arg z_0] \sin [\arg A_0 - \mu_0 \arg z_0] \right\}
= 2 B_{m, \mu_0} \left\{ \Re \left[ \frac{A_0}{z_0^{m}} \right] \Re \left[ \frac{A_0}{z_0^{\mu_0}} \right] - \Im \left[ \frac{z_0^{-m}}{z_0^{-\mu_0}} \right] \Im \left[ \frac{A_0}{z_0^{\mu_0}} \right] \right\}.
\]  \hspace{1cm} (A.30)

We need to solve, for some high order \( m \),

\[
\begin{bmatrix}
2 B_{m, \mu_0} \Re \left[ \frac{z_0^{-m}}{z_0^{-\mu_0}} \right] & -2 B_{m, \mu_0} \Im \left[ \frac{z_0^{-m}}{z_0^{-\mu_0}} \right] \\
2 B_{m-1, \mu_0} \Re \left[ \frac{z_0^{-(m-1)}}{z_0^{-(\mu_0)}} \right] & -2 B_{m-1, \mu_0} \Im \left[ \frac{z_0^{-(m-1)}}{z_0^{-(\mu_0)}} \right]
\end{bmatrix}
\begin{bmatrix}
\Re \left[ \frac{A_0}{z_0^{m}} \right] \\
\Im \left[ \frac{A_0}{z_0^{\mu_0}} \right]
\end{bmatrix}
= \begin{bmatrix}
\langle \langle n^m \rangle \rangle \\
\langle \langle n^{m-1} \rangle \rangle
\end{bmatrix}.
\]  \hspace{1cm} (A.31)
A.4 Large deviation function

In this appendix we shall derive Eq. (1.49). To this end, we first use the variable $\chi$ such that $z = e^{i\chi}$. Suppose that at long times $G(e^{i\chi}, t) \sim e^{\lambda_0(i\chi)t}$ with $\lambda_0$ the CGF for the current cumulants (1.18). We then have

$$P(n, t) = \frac{1}{2\pi} \int_{-\pi}^{\pi} d\chi G(e^{i\chi}, t) e^{-in\chi} \sim \frac{1}{2\pi} \int_{-\pi}^{\pi} d\chi e^{\lambda_0(i\chi)t} e^{-in\chi}. \quad (A.32)$$

We now write $I = n/t$ and insert it. This yields

$$P(I, t) \sim \frac{1}{2\pi} \int_{-\pi}^{\pi} d\chi e^{[\lambda_0(i\chi)-iI\chi]t} \sim \frac{1}{2\pi} \int_{-\infty}^{\infty} d\chi e^{[\lambda_0(i\chi)-iI\chi]t}, \quad (A.33)$$

where in the second step we have extended the integration domain to infinity, assuming that the leading contribution to the integral comes from a finite region around a saddle point in the complex plane. The saddle point condition reads

$$\frac{d\lambda_0}{d\chi} - iI = 0, \quad (A.34)$$

the solution to which we denote $\chi_0(I)$. Thus

$$P(I, t) \sim e^{[\lambda_0(i\chi_0)-iI\chi_0]t} \int_{-\infty}^{\infty} \frac{d\chi e^{\frac{i}{2}\lambda_0''(i\chi_0)\chi^2t}}{2\pi}$$

$$\sim e^{[\lambda_0(i\chi_0)-iI\chi_0]t} \sqrt{\frac{1}{2\pi|\lambda_0''(i\chi_0)|t}}, \quad (A.35)$$

and finally

$$\ln \frac{P(I, t)}{t} \xrightarrow{t \to \infty} \lambda_0(i\chi_0) - iI\chi_0 = \lambda_0(z_0) - I\chi_0 \equiv \xi(I), \quad (A.36)$$

which is Eq. (1.49).
Appendix B

Appendices for Ch. 3

B.1 Obtaining the tunneling rates

Eq. (3.2) is obtained by combining the expression for the mean time spent in state \( i \),

\[
\langle t_i \rangle = \frac{1}{\sum_j \Gamma_{i \rightarrow j}},
\]

(B.1)

with the expression for the probability that the system makes the next transition to state \( j \), given that it is in state \( i \),

\[
P_{i \rightarrow j} = \frac{\Gamma_{i \rightarrow j}}{\sum_{j'} \Gamma_{i \rightarrow j'}} = \frac{N_{i \rightarrow j}}{\sum_{j'} N_{i \rightarrow j'}}.
\]

(B.2)

Thus

\[
\Gamma_{i \rightarrow j} = \frac{N_{i \rightarrow j}}{\left( \sum_{j'} N_{i \rightarrow j'} \right) \langle t_i \rangle} = \frac{N_{i \rightarrow j}}{\sum t_i},
\]

(B.3)

which is Eq. (3.2).

B.2 Numerical inversion of the GF

In this appendix we provide the formula that we have used to numerically invert the GF in order to obtain the probabilities \( P(n, t) \). It has been taken from Ref. [114], to which we refer the reader for more details. Using this formula is advantageous, since the integrand in Eq. (1.43) is oscillatory. Thus, applying usual techniques for numerical integration may take much longer than the method from [114] and might turn out to be much less accurate or even to break down.
The theorem is formulated as follows: For $0 < r < 1$ and $n \geq 1$,

$$\left| P_n - \tilde{P}_n \right| \leq \frac{r^{2n}}{1 - r^{2n}}, \quad (B.4)$$

where

$$\tilde{P}_n = \frac{1}{2nr^n} \sum_{j=1}^{2n} (-1)^j \Re \left[ G(re^{\frac{\pi i j}{n}}) \right]$$

$$= \frac{1}{2nr^n} \left\{ G(r) + (-1)^n G(-r) + 2 \sum_{j=1}^{n-1} (-1)^j \Re \left[ G(re^{\frac{\pi i j}{n}}) \right] \right\}. \quad (B.5)$$

Above, $\Re$ denotes the real part, $G(z)$ is the GF [Eq. (1.13)] and $i = \sqrt{-1}$. We have denoted the probabilities $P(n, t)$ with the shorthand notation $P_n$ since all the parameters, including time, are fixed here.

Remarks:

- Theorem (B.5) relies on $0 \leq P_n \leq 1$, which is the case for true probabilities.
- The theorem applies to $n \geq 1$. When $n = 0$ is important, we can apply the theorem to $zG(z)$ instead of $G(z)$.
- The accuracy of the method can be determined by a parameter that we shall denote by $a$. The error bound is given by Eq. (B.4). We have

$$\left| P_n - \tilde{P}_n \right| \leq \frac{r^{2n}}{1 - r^{2n}} \simeq r^{2n} \quad (B.6)$$

for small $r$. Thus, to have an accuracy of $10^{-a}$ we need to set

$$r = 10^{-\frac{a}{2n}}. \quad (B.7)$$

B.3 Number of Andreev events per avalanche

We want to find the probability $q(m)$ that $m$ Andreev events occur within one avalanche. The system is described with a master equation of the form (1.44) with the kernel (3.6). Alternatively, we can write $p_A = p_{-1} + p_1$ and consider

$$\mathbf{M}(z) = \begin{bmatrix} -2\Gamma_u & \Gamma_d \\ 2\Gamma_u & -\Gamma_d + \Gamma_A(z - 1) \end{bmatrix}. \quad (B.8)$$
We make the following considerations:

- We want to start counting at the beginning of an avalanche, that is, the initial state is \([0, 1]^T\).
- We allow the state to decay to the \(N = 0\) state but we do not allow the system to go to the next avalanche: \(\Gamma_u \to 0\).
- We need to take the long-time limit since we need to include avalanches of all possible lengths.

Using the transform \(q(z) = \sum_n q(n)z^n\) we see that \(q(z)\) must be the long-time limit of

\[
q(z, t) = [1, 1] e^{\tilde{M}(z)|_{\Gamma_u \to 0} t} \begin{bmatrix} 0 \\ 1 \end{bmatrix}
= e^{-(\Gamma_d - \Gamma_A (z-1)) t} \left[ 1 - \frac{\Gamma_d}{\Gamma_d - \Gamma_A (z-1)} \right] + \frac{\Gamma_d}{\Gamma_d + \Gamma_A} \sum_{n=0}^{\infty} \left( \frac{\Gamma_A}{\Gamma_d + \Gamma_A} \right)^n z^n,
\]

(B.9)

yielding Eq. (3.7).

**B.4 Approximation of the CGF for the SINIS system to lowest order in \(\Gamma_u\)**

We want to find the dominant eigenvalue \(\lambda_0(z)\) of the kernel (3.6) to first order in \(\Gamma_u\). Alternatively, we can write \(p_A = p_{-1} + p_1\) and consider the kernel (B.8). We use the approach of the recursive scheme of App. A.2, but this time expand in the variable \(\Gamma_u\), and write

\[
\lambda_0(z, \Gamma_u) = \sum_{n=1}^{\infty} \lambda_0^{(n)}(z) \frac{\Gamma_u^n}{n!};
\]

(B.10)

\[
|0(z, \Gamma_u)\rangle = \sum_{n=0}^{\infty} |0^{(n)}(z)\rangle \frac{\Gamma_u^n}{n!},
\]

(B.11)

\[
\tilde{M}(z, \Gamma_u) = \tilde{M}_0(z) + \Gamma_u \tilde{M}_1,
\]

(B.12)
with \( \widetilde{M}(z, \Gamma_u)|0(z, \Gamma_u)\rangle \rangle = \lambda_0(z, \Gamma_u)|0(z, \Gamma_u)\rangle \rangle \) and

\[
\widetilde{M}_0(z) = \begin{bmatrix}
0 & \Gamma_d \\
0 & -\Gamma_d + \Gamma_A(z - 1)
\end{bmatrix}
\quad \text{and} \quad
\widetilde{M}_1 = \begin{bmatrix}
-2 & 0 \\
2 & 0
\end{bmatrix}.
\] (B.13)

We thus find \( \lambda_0^{(0)} = 0 \) and the vector \(|0^{(0)}\rangle \rangle\) is the right eigenvector of \( \widetilde{M}_0 \) corresponding to the zero eigenvalue, that is, \(|0^{(0)}\rangle \rangle = [1, 0]^T \). The corresponding left eigenvector that is normalized such that \( \langle \langle \tilde{0}(z)|0^{(0)}\rangle \rangle = 1 \) reads

\[
\langle \langle \tilde{0}(z)| = [1, \frac{\Gamma_d}{\Gamma_d - \Gamma_A(z - 1)}].
\] (B.14)

Using the recursive scheme formulas (A.19) and (A.20) we find

\[
\lambda_0^{(1)}(z) = \langle \langle \tilde{0}(z)|\Gamma_u\widetilde{M}_1|0^{(0)}\rangle \rangle
\]

\[
= 2\Gamma_u \left[ 1, \frac{\Gamma_d}{\Gamma_d - \Gamma_A(z - 1)} \right] \begin{bmatrix}
-1 & 0 \\
1 & 0
\end{bmatrix} \begin{bmatrix}
1 \\
0
\end{bmatrix}
\]

\[
= 2\Gamma_u \left( -1 + \frac{\Gamma_d}{\Gamma_d - \Gamma_A(z - 1)} \right)
\]

\[
= 2\Gamma_u \left( -1 + \frac{\Gamma_d}{\Gamma_d + \Gamma_A} \frac{1}{1 - z \frac{\Gamma_A}{\Gamma_d + \Gamma_A}} \right)
\]

\[
= 2\Gamma_u \left( \frac{\Gamma_d}{\Gamma_d + \Gamma_A} \sum_{n=0}^{\infty} \left( \frac{\Gamma_A}{\Gamma_d + \Gamma_A} \right)^n (z^n - 1) \right).
\] (B.15)

We have used that

\[
1 = \left( \frac{\Gamma_d}{\Gamma_d + \Gamma_A} \right) \sum_{n=0}^{\infty} \left( \frac{\Gamma_A}{\Gamma_d + \Gamma_A} \right)^n.
\] (B.16)

Considering that the long-time limit CGF reads \( S(z, t) = \lambda_0(e^z) t \), and using Eq. (3.7), this directly yields (3.8).
Appendix C

Appendices for Ch. 4

C.1 Noise Formulas

Here we give the remaining noise spectra\textsuperscript{22, 23} to complete relation (4.5).

\[ S_{RR}(\omega) = \frac{e^2}{\hbar} \int dE \text{Tr}\left\{ t^\dagger_L(E) t_L(E + \hbar \omega) t^\dagger_L(E + \hbar \omega) t_L(E) F_{LL}(E, \omega) \right. \]
\[ + t^\dagger_L(E) r_R(E + \hbar \omega) r^\dagger_R(E + \hbar \omega) t_L(E) F_{LR}(E, \omega) \]
\[ + r^\dagger_R(E) t_L(E + \hbar \omega) t^\dagger_L(E + \hbar \omega) r_R(E) F_{RL}(E, \omega) \]
\[ + \left[ \mathbb{I} - r^\dagger_R(E) r_R(E + \hbar \omega) \right] \]
\[ \times \left[ \mathbb{I} - t^\dagger_L(E) t_L(E + \hbar \omega) \right] F_{RR}(E, \omega) \left\} \right., \quad (C.1) \]

\[ S_{LR}(\omega) = \frac{e^2}{\hbar} \int dE \text{Tr}\left\{ - \left[ \mathbb{I} - t^\dagger_L(E) r_L(E + \hbar \omega) \right] t^\dagger_L(E + \hbar \omega) t_L(E) F_{LL}(E, \omega) \right. \]
\[ + r^\dagger_L(E) t_R(E + \hbar \omega) r^\dagger_R(E + \hbar \omega) t_L(E) F_{LR}(E, \omega) \]
\[ + t^\dagger_R(E) r_L(E + \hbar \omega) t^\dagger_L(E + \hbar \omega) r_R(E) F_{RL}(E, \omega) \]
\[ - t^\dagger_R(E) t_R(E + \hbar \omega) \left[ \mathbb{I} - r^\dagger_R(E + \hbar \omega) r_R(E) \right] F_{RR}(E, \omega) \left\} \right. \]
\[ \]
The expressions for the heat current noise spectra have a similar structure except that there is an additional factor of \((E + \hbar \omega/2 - \mu)^2\) in the integrand, as in Eq. (4.8).

From the definitions (1.8) and (4.1) we see that for stationary transport \(S_{\alpha\beta}^{(th)}(\omega) = S_{\beta\alpha}^{(th)}(-\omega)\) and therefore we immediately find \(S_{RL}^{(th)}(\omega) = S_{LR}^{(th)}(-\omega)\). Finally, \(S_{LL}^{(th)}\) as well as \(S_{RR}^{(th)}\) are symmetric in \(\omega\). We remark that in the single channel case we also have \(S_{LR}^{(th)}(\omega) = S_{RL}^{(th)}(\omega)\), which implies that also the cross-correlations are symmetric in frequency.

### C.2 Continuity Equation and Charge Noise

Let \(\hat{Q}\) be the charge operator in the mesoscopic conductor. Charge conservation imposes

\[
\frac{\partial \hat{Q}}{\partial t} + \sum_\alpha \Delta \hat{I}_\alpha(t) = 0. \tag{C.3}
\]

In frequency space this reads

\[
i \omega \hat{Q} + \sum_\alpha \Delta \hat{I}_\alpha(\omega) = 0. \tag{C.4}
\]

For stationary transport considered here we have

\[
\frac{1}{2} \left\langle \Delta \hat{I}_\alpha(\omega) \Delta \hat{I}_\beta(\omega') + \Delta \hat{I}_\beta(\omega') \Delta \hat{I}_\alpha(\omega) \rightangle = S_{\alpha\beta}(\omega) 2\pi \delta(\omega + \omega'), \tag{C.5}
\]

and

\[
\frac{1}{2} \left\langle \hat{Q}(\omega) \hat{Q}(\omega') + \hat{Q}(\omega') \hat{Q}(\omega) \right\rangle = S_{QQ}(\omega) 2\pi \delta(\omega + \omega'). \tag{C.6}
\]

Thus, we immediately find

\[
\sum_{\alpha,\beta} S_{\alpha\beta}(\omega) 2\pi \delta(\omega + \omega')
\]

\[
= \frac{1}{2} \left\langle \sum_{\alpha,\beta} \left[ \Delta \hat{I}_\alpha(\omega) \Delta \hat{I}_\beta(\omega') + \Delta \hat{I}_\beta(\omega') \Delta \hat{I}_\alpha(\omega) \right] \rightangle
\]

\[
= -\omega \omega' \frac{1}{2} \left\langle \hat{Q}(\omega) \hat{Q}(\omega') + \hat{Q}(\omega') \hat{Q}(\omega) \right\rangle
\]

\[
= \omega^2 S_{QQ}(\omega) 2\pi \delta(\omega + \omega') \tag{C.7}
\]
Integrating on $\omega'$ we find relation 4.10.

In particular, at zero frequency the charge noise in terms of the Wigner-Smith type matrix (4.12) is given by

$$S_{QQ}(\omega) = e^2\hbar \sum_{\alpha,\beta} \int dE \text{Tr} \left[ N_{\alpha\beta}^\dagger(E, E + \hbar\omega) N_{\beta\alpha}(E + \hbar\omega, E) \right] F_{\alpha\beta}(E, \omega).$$

(C.8)

If we consider Coulomb interactions and in our mesoscopic region we include any capacitively coupled gates, then charge conservation imposes that charge fluctuations consist of multipole (typically mainly dipole) fluctuations [28, 30]. Therefore, the accumulated charge on this total mesoscopic region is zero. Eq. (C.3) then implies that $\sum_{\alpha} \Delta \hat{I}_\alpha(t) = 0$ and therefore $\sum_{\alpha} S_{\alpha\beta}(\omega) = \sum_{\beta} S_{\alpha\beta}(\omega) = 0$, where the sums are now also over gate reservoirs.

### C.3 Rectification coefficients in a noninteracting two-terminal conductor

The current in a noninteracting two-terminal conductor can be obtained from the Landauer-Büttiker formula, Eq. (1.5), by observing that due to charge conservation we have $R_{\alpha\alpha} + \sum_{\beta} T_{\alpha\beta} = M_\alpha$. For single-channel terminals, $M_{L,R} = 1$, we get

$$I = \frac{e}{\hbar} \int dE \mathcal{T}(E) \left[ f_L(E) - f_R(E) \right],$$

where $\mathcal{T}(E)$ is the energy-dependent transmission coefficient and $f_{L,R}$ are the Fermi functions in the left and right reservoirs [Eq. (4.7)], respectively. Moreover, let us denote the Fermi function at zero bias by $f(E)$ [Eq. (4.14)].

The heat current, on the other hand, is given by

$$J = \frac{1}{\hbar} \int dE \mathcal{T}(E)(E - \mu) \left[ f_L(E) - f_R(E) \right].$$

(C.10)

Expanding the current and the heat current in both, bias and temperature.
Gradient, we write

\[ I(V, \Delta T) = \sum_{n,m} \frac{G^{(n,m)}}{n! m!} V^n \left( \frac{\Delta T}{T} \right)^m, \]  

\[ J(V, \Delta T) = \sum_{n,m} \frac{G_{\text{th}}^{(n,m)}}{n! m!} V^n \left( \frac{\Delta T}{T} \right)^m. \]  

We have \( G^{(0,0)} = 0 \) and \( G_{\text{th}}^{(0,0)} = 0 \). Thus, the expansion coefficients in \( \text{(C.11)} \) are obtained as \( G^{(n,m)} = (-\beta)^m \partial^{(n+m)} I/\partial V^n \partial (\Delta \beta)^m |_{V=0, \Delta \beta=0} \) and analogously for the coefficients in \( \text{(C.12)} \). Here, \( \beta = 1/k_B T \) is the inverse temperature, with \( k_B \) Boltzmann’s constant, and we have observed that

\[ \Delta \beta = \Delta \left( \frac{1}{k_B T} \right) = \frac{1}{k_B T} \frac{\Delta T}{T}. \]  

We now write \( V_L = \eta V \) and \( V_R = (\eta - 1)V \) and analogously \( \beta_L = \beta + \alpha \Delta \beta \) and \( \beta_R = \beta + (\alpha - 1) \Delta \beta \), with \( 0 \leq \alpha, \eta \leq 1 \). Going up to second order we thus obtain

\[ G^{(1,0)} = \frac{e^2}{h} \int dE \mathcal{T}(E) \left( -\frac{\partial f}{\partial E} \right), \]

\[ G^{(0,1)} = \frac{e}{h} \int dE \mathcal{T}(E)(E - \mu) \left( -\frac{\partial f}{\partial E} \right), \]

\[ G^{(2,0)} = \frac{e^3}{h} (2\eta - 1) \int dE \mathcal{T}(E) \left( \frac{\partial^2 f}{\partial E^2} \right), \]

\[ G^{(0,2)} = \frac{e}{h} (2\alpha - 1) \int dE \mathcal{T}(E)(E - \mu)^2 \left( \frac{\partial^2 f}{\partial E^2} \right), \]

\[ G^{(1,1)} = \frac{e^2}{h} (\alpha + \eta - 1) \int dE \mathcal{T}(E)(E - \mu) \left( \frac{\partial^2 f}{\partial E^2} \right), \]
and for the thermal conductances

\[ G_{\text{th}}^{(1,0)} = \frac{e}{\hbar} \int dE \mathcal{T}(E)(E - \mu) \left( -\frac{\partial f}{\partial E} \right), \]

\[ G_{\text{th}}^{(0,1)} = \frac{1}{\hbar} \int dE \mathcal{T}(E)(E - \mu)^2 \left( -\frac{\partial f}{\partial E} \right), \]

\[ G_{\text{th}}^{(2,0)} = \frac{e^2}{\hbar} (2\eta - 1) \int dE \mathcal{T}(E)(E - \mu) \left( \frac{\partial^2 f}{\partial E^2} \right), \]

\[ G_{\text{th}}^{(0,2)} = \frac{1}{\hbar} (2\alpha - 1) \int dE \mathcal{T}(E)(E - \mu)^3 \left( \frac{\partial^2 f}{\partial E^2} \right), \]

\[ G_{\text{th}}^{(1,1)} = \frac{e}{\hbar} (\alpha + \eta - 1) \int dE \mathcal{T}(E)(E - \mu)^2 \left( \frac{\partial^2 f}{\partial E^2} \right). \]

We see that if we expand in \((\Delta T/T)\) we get analogous results as for the coefficients in the \(V\)-expansion.

Eqs. (1.9) and (1.12) are readily verified explicitly.

To summarize

- \(G^{(1,0)}\) is related to the zero frequency equilibrium current-current correlator according to Eq. (1.9).

- \(G_{\text{th}}^{(0,1)}\) is related to the zero frequency equilibrium heat current-heat current correlator in an analogous manner.

- \(G^{(0,1)} = G_{\text{th}}^{(1,0)}\) are equal by Onsager symmetry and are related to the zero frequency correlator of \(\Delta \hat{I}\) and \(\Delta \hat{J}\).

- The thermal conductivity \(\kappa\) is defined as the first order expansion coefficient of \(J\) for \(I = 0\). It is thus given by

\[ \kappa = G_{\text{th}}^{(0,1)} - \frac{G^{(0,1)}}{G^{(1,0)}} G_{\text{th}}^{(1,0)}. \]  

(\text{C.14})

- For \(G^{(2,0)}\) the fluctuation relation (1.12) applies and an analogous relation for \(G_{\text{th}}^{(0,2)}\) [Eq. (1.2)].

- The generalization of the above for higher order fluctuation relations can be derived from the CGF and its symmetries [59, 60, 61].
Bibliography


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<th>Abbreviation</th>
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