Revealing strong correlations in higher order transport statistics: a noncrossing approximation approach

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We present a method for calculating the full counting statistics of a nonequilibrium quantum system based on the propagator noncrossing approximation (NCA). This numerically inexpensive method can provide higher order cumulants for extended parameter regimes, rendering it attractive for a wide variety of purposes. We compare NCA results to Born–Markov quantum master equations (QME) results to show that they can access different physics, and to numerically exact inchworm quantum Monte-Carlo data to assess their validity. As a demonstration of its power, the NCA method is employed to study the impact of correlations on higher order cumulants in the nonequilibrium Anderson impurity model. The four lowest order cumulants are examined, allowing us to establish that correlation effects have a profound influence on the underlying transport distributions. Higher order cumulants are therefore demonstrated to be a proxy for the presence of Kondo correlations in a way that cannot be captured by simple QME methods.

I. INTRODUCTION

A. Background and overview

Electron transport through mesoscopic and nanoscale junctions is a complex phenomenon where nonequilibrium statistical mechanics is entwined with quantum many-body effects.11 Systems are driven out of equilibrium by, e.g., an external bias voltage or a temperature gradient, and their response is measured. Perhaps the simplest response observable in many experimental setups is the electronic current. Increasingly, however, is has become both possible and desirable to access so-called higher order transport characteristics. This includes the current’s fluctuations and its higher moment,54 as well as the statistics of individual electron transfer events.8 Interestingly, ultracold atom experiments can simulate electronic transport and allow for directly extracting statistical distributions of populations in different parts of the system, marking another path towards detailed characterization of transport.

Theoretically, all such information can be obtained from the full counting statistics (FCS) approach pioneered by Levitov and Lesovik11,12 where all moments and cumulants of transport events are efficiently represented by a single generating functional. Since its inception, this idea has attracted a great deal of attention.13–26

Many experimental studies concentrate on the current noise and the current-to-noise ratio, also known as the Fano factor. In both classical and quantum systems, these quantities already contain information not present in the mean current.27,28 For example, they enable probing of effective quasiparticle charges.29–31 Moreover, noise measurements have allowed researchers to, e.g., identify electron bunching and anti-bunching during transport.72,91–94

time distributions and determine the number and transmission probabilities of active levels contributing to transport.42,29 Other studies reported the measurement of higher-order cumulants that further elucidate the mechanisms underlying electronic transport.52

Given sufficient cumulants, it is in principle possible to reconstruct the full FCS. Much of the motivation for this comes from insights regarding noninteracting systems, where the exact FCS is given by the Levitov–Lesovik formula.12,13 There, the ability to measure the FCS could provide indirect access to theoretically intuitive but experimentally unattainable properties like channel coherence and entanglement entropy.53 This scheme holds also true for interacting systems, where the FCS provides insight onto many-body quantum effects. For example, even though the role of electronic correlations is not yet well understood, it is known that correlation-driven physics like the Kondo effect modify the current noise and its higher order cumulants.54

Still, the theoretical prediction of the FCS for interacting systems is generally non-straightforward and a variety of theoretical approaches has been applied. Among the approximate approaches used are quantum master equations (QME),4,38,68–76 the approximative approaches used are quantum master equations (QME),4,38,68–76 the approximative approaches used are quantum master equations (QME),4,38,68–76 the approximative approaches used are quantum master equations (QME),4,38,68–76 the approximate approaches used are quantum master equations (QME),4,38,68–76 the approximate approaches used are quantum master equations (QME),4,38,68–76 the approximate approaches used are quantum master equations (QME),4,38,68–76 the approximate approaches used are quantum master equations (QME),4,38,68–76 Numerically exact approaches to FCS include the Inchworm quantum Monte Carlo (iQMC) method24,77 the hierarchical equations of motion technique (HEOM)20,23 the density matrix renormalization group approach29,31 and the iterative path integral method.24,25,78 A variety of ongoing research programs are aimed at extending exact approaches to new experimentally relevant regimes, and at developing new exact and approximate methodologies.
B. Noncrossing approximations

At the present time, methods able to address Kondo physics remain computationally expensive. Here, we propose a simple and inexpensive approximate scheme for evaluating FCS that is based on one variation of the noncrossing approximation (NCA). The NCA and its extensions have long been a successful qualitative approach to several aspects of nonequilibrium Kondo physics in quantum transport.\textsuperscript{35,36} The approximation has multiple, inequivalent formulations, most of which are unsuitable to the evaluation of FCS due to the introduction of an auxiliary pseudoparticle space.\textsuperscript{37,38} It can easily be used to obtain high order cumulants or the complete FCS generating functional. To verify the method and map out its regime of applicability, we compare our NCA results with numerically exact generalizations of the NCA (QME) to be presented below in Sec. IV, the QME approach is based on a second order expansion in the dot–leads coupling. In contrast to the NCA, the QME does not employ a Dyson-like diagrammatic resummation scheme. Rather, it uses a Liouville-space resummation based on the Nakajima–Zwanzig equation.\textsuperscript{103–106} This results in an analytically solvable and intuitive equation of motion for the reduced density matrix, which is in the bath subspaces comprising the left and right lead, and the dot–leads coupling. The dot Hamiltonian is given by

\[ H_D = \sum_{\sigma = \uparrow, \downarrow} \epsilon_{0\sigma} a_{\sigma}^\dagger a_{\sigma} + U d_{\uparrow}^\dagger d_{\uparrow} d_{\downarrow}^\dagger d_{\downarrow}. \]  

Here, the \( a_{\sigma}^\dagger \) and \( a_{\sigma} \) denote creation/annihilation operators for an electron of spin \( \sigma \) on the dot, \( \epsilon_{0\sigma} \) is the single particle occupation energy, and \( U \) determines the strength of the Coulomb interaction. Experimentally, the single particle occupation energy can be tuned by an external gate voltage \( \Phi_{\text{gate}} \). We model the influence of such a gate voltage by setting \( \epsilon_0 = \Phi_{\text{gate}} - \frac{U}{2} \).

The leads are assumed to be a noninteracting continuum,

\[ H_B = \sum_{\sigma = \uparrow, \downarrow} \sum_{\ell \in \{L,R\}} \sum_{k \in \ell} \epsilon_k a_{k\sigma}^\dagger a_{k\sigma}, \]  

where the \( a_{k\sigma}^\dagger \) and \( a_{k\sigma} \) denote creation/annihilation operators on a lead level with index \( k \), spin \( \sigma \) and energy \( \epsilon_k \). The indices \( L \) and \( R \) denote the “left” and “right” lead, respectively.

Finally, the coupling between the dot and leads is assumed to take the form

\[ H_{DB} = \sum_{\sigma \in \{\uparrow, \downarrow\}} \sum_{\ell \in \{L,R\}} \sum_{k \in \ell} \left( V_{\ell} a_{k\sigma}^\dagger d_{\sigma} + \text{h.c.} \right), \]

where \( V_{\ell} \) represents the coupling interaction.
with coupling parameters $V_k$ that can be parameterized in terms of a coupling strength function
\[ \Gamma_\ell(\epsilon) = \pi \sum_{k \in \ell} |V_k|^2 \delta(\epsilon - \epsilon_k). \tag{5} \]

We explicitly consider symmetric coupling to the two leads, each of which is taken to be a flat band with a soft cutoff: \[
\Gamma_L(\epsilon) = \Gamma_R(\epsilon) = \frac{\Gamma}{2} \frac{1}{(1 + e^{\nu(\epsilon - \epsilon_c)})(1 + e^{-\nu(\epsilon + \epsilon_c)})}, \tag{6} \]

The overall strength of the dot–lead coupling is set by the constant $\Gamma$, which is used as our unit of energy. The coupling strength defines the hybridization functions,
\[ \Delta^<(\epsilon) = \frac{1}{\pi} \int d\epsilon e^{\epsilon - i\epsilon t} \Gamma(\epsilon) f(\epsilon), \tag{7} \]
\[ \Delta^>(\epsilon) = \frac{1}{\pi} \int d\epsilon e^{\epsilon + i\epsilon t} \Gamma(\epsilon)(1 - f(\epsilon)). \tag{8} \]

Here $f(\epsilon) \equiv \frac{1}{1 + e^{\epsilon(\epsilon_e - \epsilon_c)}}$, where $\mu_{L/R} = \pm V/2$ are chemical potentials set by a symmetrically applied bias voltage $V$, and $\beta$ is the inverse temperature in the leads. Moreover, we set $\nu = 1/\Gamma$ and $\epsilon_c = 50\Gamma$ – much larger than all other energy scales in the problem – such that we are effectively working in the wide band limit. With our choice of parameters, particle-hole symmetry is obeyed for $\Phi_{\text{gate}} = 0$.

Throughout this work, the on-site Coulomb repulsion is set to $U = 8\Gamma$. This determines the Kondo temperature consistent with our parameters, $T_K \approx 0.8\Gamma$. Three representative lead temperatures will therefore be considered in this work: $T = 0.25\Gamma < T_K$, $T = 0.5\Gamma \lesssim T_K$ and $1.0\Gamma \gtrsim T_K$.

III. FCS AND COUNTING FIELDS

Determining the FCS of an observable means evaluating the generating function of its underlying probability distribution, from which cumulants and moments can be extracted. We provide a brief overview this approach and the main concepts here, and recommend Refs.~\cite{19} and \cite{128} for more details.

Consider an experiment where at time zero the system is prepared in a known initial density matrix where, e.g., the number of electrons in the left lead $L$ is known. The system is allowed to evolve freely until time $t$, when the total number of electrons in lead $L$ is measured. Let $P_L(t, n)$ be the probability that $n$ electrons are found in this measurement. The generating function is then defined as
\[ Z_L(t, \lambda) = \sum_n P_L(t, n) e^{\lambda n} \equiv \text{Tr}_{D+B} \{ \rho_L(t) \}, \tag{9} \]
where $\lambda$ is known as the counting field. This defines $\rho_L(t) = e^{-iH_L t} \rho(0) e^{iH_L t}$, a counting-field-modified (or, for brevity, simply “modified”) density matrix; which in turn defines $H_\lambda \equiv e^{i\lambda/2N_L} H e^{-i\lambda/2N_L}$, a modified Hamiltonian. $N_L = \sum_{\alpha \in \{\uparrow, \downarrow\}} \sum_{k \in L} a_{\alpha k}^\dagger a_{\alpha k}$ is the particle number operator in the left lead $L$. Modifying the Hamiltonian by the counting field corresponds to transforming the dot–bath coupling strength of the lead under consideration according to
\[ V_k(t_\pm) \rightarrow V_k e^{i\lambda\Gamma/2}, \tag{10} \]

Where $t_\pm$ is a time variable on either the backward (+) or forward (−) branch of the Keldysh contour. This idea can be generalized to other observables and counting fields.

Normally, the generating function $Z_L(t, \lambda)$ itself cannot be directly accessed in experiments. However, experiments can measure its moments and cumulants, or sometimes the probabilities $P_L(t, n)$. In particular, the cumulants $C^L_\alpha(t)$ of the generating function are given by its logarithmic derivatives:
\[ C^L_\alpha(t) = (-i)^\alpha \frac{\partial^\alpha}{\partial \lambda^\alpha} \ln \{ Z_L(t, \lambda) \} \big|_{\lambda=0}. \tag{11} \]

The first few cumulants have simple physical interpretations. The time derivative of the first cumulant corresponds to the electronic current $I_L(t)$ exiting lead $L$:
\[ C^L_1(t) = \langle N_L(t) \rangle, \tag{12} \]
\[ \frac{\partial}{\partial t} C^L_1(t) = I_L(t). \tag{13} \]

The second cumulant is related to the variance of the population in the lead,
\[ C^L_2(t) = \langle N_L^2(t) \rangle - \langle N_L(t) \rangle^2. \tag{14} \]

At steady state, its time derivative is the noise $S_L$:
\[ \lim_{t \to \infty} \frac{\partial}{\partial t} C^L_2(t) = S_L. \tag{15} \]

Higher order population cumulants and the full probability distributions $P_L(t, n)$ can also be obtained from the generating functional. These have a more complicated relationship with the statistics of the current, but are arguably more straightforward than the latter to describe theoretically. For the scope of this work, we will also consider the steady state time derivatives of the third and fourth cumulants, $\lim_{t \to \infty} \frac{\partial}{\partial t} C^L_3(t) = S_{L2}$ and $\lim_{t \to \infty} \frac{\partial}{\partial t} C^L_4(t) = S_{L3}$. These quantities express the skewness and the bifurcation of the underlying probability distribution, respectively, and are of interest in a variety of contexts.

Composite observables like the Fano factor can be a problematic quantity for studying Kondo physics, because the low energy features are obscured by the zero bias Nyquist–Johnson singularity. This stems from the different symmetry of $I$ and $S$ with respect to the bias voltage. Deep in the
universal Kondo regime and at very low voltages, this can be rectified by defining a “backscattering” current that must be separated from the unitary linear-response current. Below, we discuss an alternative and more widely applicable approach: to define a set of generalized Fano factors in terms of higher order cumulants, while taking symmetry into account.

IV. METHODOLOGY

We will now describe the propagator hybridization expansion for the FCS generating function $Z(t, \lambda) \equiv Z_L(t, \lambda)$ within the NCA. The approximation is based on a second order expansion of the time evolution operator in the dot–lead coupling, which is treated self consistently within a Dyson resummation scheme.

Using Eq. (9) in the context of Sec. II and assuming an initial condition factorized between the dot and bath spaces, $\rho(t = 0, \lambda) = \rho_B \otimes \rho_D$, we obtain

$$Z(t, \lambda) = \text{Tr}(\varrho(t, \lambda)) = \sum_{\alpha \beta} \langle \beta | \rho_D | \beta \rangle K_{\alpha \beta}(t, t, \lambda). \quad (16)$$

Here, $\alpha$ and $\beta$ are states in the dot subspace, and the modified vertex function $K_{\alpha \beta}(t, t', \lambda)$ takes the form

$$K_{\alpha \beta}(t, t', \lambda) = \text{Tr}_B \left\{ \rho_B \langle \alpha | U^\dagger_{\alpha \lambda}(t) | \beta \rangle \langle \beta | U_{\lambda \lambda}(t') | \alpha \rangle \right\}. \quad (17)$$

$\text{Tr}_B$ denotes tracing over the bath degrees of freedom. We have also made use of a modified time evolution operator, $U_{\alpha \lambda}(t) \equiv \text{T} \exp(-i \int_0^t H_{\alpha \lambda}(\tau) d\tau)$, where $\text{T}$ is the time ordering operator. The vertex function $K_{\alpha \beta}(t, t', \lambda)$ is the central object within the NCA method. In other contexts, without FCS, only the $\lambda = 0$ form appears. This can be used to construct approximate expressions for the expectation values of a variety of observables.

To derive the NCA, one extracts the lowest nonvanishing correction of a perturbative expansion of Eq. (17) in the dot–lead coupling $H_{\alpha \beta \lambda}$. The approximation is obtained by writing a self consistent expression for the vertex function in terms of this correction, resulting in the Dyson equation

$$K_{\alpha \beta}(t, t', \lambda) = k_{\alpha \beta}(t, t') + \sum_{\alpha' \beta'} \int_0^t \int_0^{t'} d\tau_1 d\tau_1' \xi_{\alpha \beta}(t - \tau_1, t' - \tau_1') G_{\alpha' \beta'}(\tau_1, \tau_1', \lambda). \quad (18)$$

This is defined in terms of the the cross-branch hybridization self-energy

$$\xi_{\alpha \beta}(t) = \sum_{\sigma \in \{\uparrow, \downarrow\}} \sum_{\ell \in \{L, R\}} \left( \Delta_{\alpha \beta}^\ell(t) \langle \alpha | d_{\sigma} | \beta \rangle \langle \beta | d_{\sigma}^\dagger | \alpha \rangle + \Delta_{\alpha \beta}^\ell(t) \langle \alpha | d_{\sigma} | \beta \rangle \langle \beta | d_{\sigma}^\dagger | \alpha \rangle \right), \quad (19)$$

and $k_{\alpha \beta}(t, t')$, a term that is independent of the counting field and will be introduced momentarily. The term NCA refers to the fact that there are no crossing hybridization lines in the diagrammatic representation of the terms included in this approach (see, e.g., Ref. 1[2]. Higher order expansions such as the one-crossing approximation employ different forms for the cross-branch self-energy $\rho_{\alpha \beta}(t, t')$.

We now return to the final quantity defined in Eq. (18), $k_{\alpha \beta}(t, t')$. This is a zeroth-order approximation for the vertex function that can be written in the form

$$k_{\alpha \beta}(t, t') = \delta_{\alpha \beta} G_{\alpha}(t) G_{\beta}(t'). \quad (20)$$

Here,

$$G_{\alpha}(t) = \langle \alpha | \text{Tr}_B (\rho_B U(t)) | \alpha \rangle \quad (21)$$

is a single-branch propagator that is diagonal in the many-particle basis of the dot due to the structure of the Hamiltonian, Eq. (1). $G$ obeys a set of equations similar to those obeyed by $K$, but on a single branch of the Keldysh contour:

$$G_{\alpha}(t) = g_{\alpha}(t) - \int_0^t \int_0^{\tau_1} d\tau_1 d\tau_2 g_{\alpha}(t - \tau_1) \Sigma_{\alpha}(\tau_1 - \tau_2) G_{\alpha}(\tau_2). \quad (22)$$

The single-contour self-energy $\Sigma_{\alpha}(t)$ depends on the propagator $G_{\alpha}(t)$ and is given within the NCA by

$$\Sigma_{\alpha}(t) = \sum_{\sigma \in \{\uparrow, \downarrow\}} \sum_{\ell \in \{L, R\}} \sum_{\beta} \left( \Delta_{\alpha \beta}^\ell(t) \langle \alpha | d_{\sigma} | \beta \rangle \langle \beta | d_{\sigma}^\dagger | \alpha \rangle + \Delta_{\alpha \beta}^\ell(t) \langle \alpha | d_{\sigma}^\dagger | \beta \rangle \langle \beta | d_{\sigma} | \alpha \rangle \right) G_{\beta}(t). \quad (23)$$

Finally, $g_{\alpha}(t) = e^{-iH_D t}$ is the propagator on the isolated dot. $G(t)$ remains unmodified by the counting field, due to being restricted to one branch of the Keldysh contour.

V. RESULTS

Subsequently, we use the NCA methodology described above to study the four lowest order cumulants, $I_L$, $S_L$, $S_{L2}$, and $S_{L3}$, at steady state. As we are considering the steady state, we henceforth drop the lead index $L$. Further, since these quantities diverge linearly in time, we plot their first time derivative. We will investigate their dependence on bias voltage, gate voltage, and temperature. Since the bias voltage dependence is of primary interest, but can be weak in some regimes, we plot the second derivative of the cumulants with respect to the bias voltage.

A. Signature of correlations in observables associated to higher order cumulants

We begin by exploring the influence of Kondo physics on higher order cumulants. We compare NCA results,
where a qualitative signature of such phenomena is expected, with QME results, where none is expected. Later, in Sec. [V C] we evaluate the accuracy of the NCA predictions by comparing with numerically exact iQMC results.

Figs. [1] and [2] provide an overview of the behavior of four different observables in the NCA and QME approximations, respectively. To facilitate comparison, the figures employ equivalent false color representations of the data. The observables $\partial^2 I / \partial V^2$, $\partial^2 S / \partial V^2$, $\partial^2 S_2 / \partial V^2$, and $\partial^2 S_3 / \partial V^2$, are shown as a function of bias voltage and gate voltage in each panel, at a constant lead temperature. Columns of panels correspond to the different observables, while rows correspond to different temperatures.

Figs. [1] and [2] contain a great deal of information in a rather compact form. To make them easier to understand, it is useful to focus on two particular sets of physical features. First, the transition between resonant and nonresonant transport, which is marked by dashed black lines. The associated behavior is clearly apparent in all QME plots, where the resonance condition matches nodal curves delineating changes in the signs of the observables. However, within the NCA, at several parameter regimes this transition is either shifted or completely missing. This is because the availability of resonant transport channels, which in QME is broadened only by the temperature, can also be modulated or hidden by broadening effects not accounted for within the QME. Second, the emergence of Kondo and mixed-valence physics is clearly visible in the NCA plots, but completely missing from the QME data. The signature of these correlation-driven effects is a pronounced feature centered around zero bias voltage, which disappears at high temperatures. As can be seen by comparing the top and middle panels of Fig. [1], higher cumulants reveal progressively richer and more complex dependencies on the bias and gate voltages. Thus, they provide increasingly detailed modes of characterization.

An interesting point to note is that the temperature at which cumulants exhibit correlated phenomena does not appear to vary significantly with the cumulant order. This is true both in and out of equilibrium, and to some degree supports the idea that the low energy physics is controlled by a few universal energy scales even when a bias voltage is applied. However, a firm statement on this issue requires a more systematic study going beyond the NCA.

Further details are revealed by considering parameters below the resonance condition, at a constant nonzero gate voltage and a range of bias voltages. A cut of this kind across the data of Fig. [1] is shown in Fig. [3] and the parameters chosen for the cut are marked in Fig. [1] by solid red lines. As even(odd) cumulants are symmetric(antisymmetric) with respect to bias voltage, it is instructive to directly compare $I$ with $S_2$; and re-
FIG. 2. QME results. The second derivative with respect to bias voltage is shown for the current (a), the noise (b), $S_2$ (c) and $S_3$ (d). From top to bottom, the temperature increases from $T = 0.25\Gamma$ to $T = 0.5\Gamma$ and finally $T = \Gamma$. The black dashed lines, which serve as a guide for the eye, indicate the conditions $\epsilon_0 = \mu_{L/R}$ and $2\epsilon_0 + U = \mu_{L/R}$ that separate resonant from nonresonant transport.

FIG. 3. NCA results. The second derivative with respect to bias voltage is shown for the current (upper left), the noise (upper right), and the higher order cumulants $S_2$ (lower left) and $S_3$ (lower right). The gate voltage is set to $\Phi_{\text{gate}} = 2\Gamma$. These are horizontal cuts across the data in Fig. 4 as marked by the red solid lines.

spectively $S$ with $S_3$. While $\partial^2 I/\partial V^2$ exhibits a single peak–dip structure, an additional shoulder appears at low temperature in $\partial^2 S_2/\partial V^2$ at a bias voltage of about $V \sim 1.5\Gamma$. Similarly, $\partial^2 S/\partial V^2$ shows a single pronounced peak, whereas at low temperature, $\partial^2 S_3/\partial V^2$ develops distinctive side peaks at a bias voltage $V \sim \Gamma$. This once gain suggests that correlations modify higher order cumulants at correspondingly higher energy scales and bias voltages. Potentially, therefore, access to higher order cumulants could make it easier to identify the signature of correlations, even without fully mapping their dependence on both the bias and gate voltages.

**B. Generalized Fano factors and their implications**

As noted in Sec. III, the Fano factor $F = S/I$ manifests a singularity at zero voltage, where the current (odd with respect to the bias voltage) disappears while the noise (even with respect to the bias voltage) does not. $F$ will be revisited in Sec. [**V**] where we benchmark the NCA method against numerically exact results. In the following, we consider the generalized Fano factors $F' \equiv S_2/I$ and $F'' \equiv S_3/S$. These are the lowest order ratios comprising only odd and even cumulants, respectively. They are therefore free of singular behavior at zero voltage, making them potentially useful for exploring Kondo physics.

For both observables, it is once again more convenient to plot the second derivative with respect to bias voltage.
In Fig. 4 these are shown at the same parameter ranges used in Figs. 1 and 2. $F'$ and $F''$, respectively, are shown in the left and right panels, temperature increases as we go to lower panels. Both generalized Fano factors exhibit sharp, well defined Kondo features at low temperatures. As before, these correlation driven features disappear at higher temperatures.

The separate cumulants in Fig. 4 are dominated by the signature of the transition between off-resonant and resonant transport. Remarkably, however, in Fig. 4 $F'$ exhibits Kondo features of comparable scale to those delineating the resonant transport edge, and $F''$ is dominated by the Kondo features. This suggests that symmetry-corrected higher order Fano factors contain detailed information regarding correlation effects, and may be a more sensitive probe of such physics than lower order quantities.

As the temperature is lowered and the Kondo effect develops, the value of $F'$ and $F''$ at low bias voltages increases, except near the resonance condition. Since the Kondo effect enhances the current $I$, an increase in $F''$ implies that $S_2$ is more strongly enhanced than $I$. Correspondingly, the underlying probability distribution describing electron transfer becomes increasingly skewed. Similarly, while the behavior of the noise is more complicated, $S$ is mostly suppressed by Kondo physics, and the same is true for $S_3$. An increase in $F''$ therefore implies a weaker suppression of $S_2$ than that of $S$, and an increasingly bifurcated probability distribution. A more detailed analysis of the probabilities $P_L(t, n)$ would be interesting in this regard, but is beyond the scope of the present work.

C. Validation and benchmarks

It is clear from the data that we have presented so far that, when considering higher order transport cumulants, the NCA method captures physics not accounted for by the QME method. This is not entirely surprising, since it is known to do so for single-particle correlation functions and for the current. However, since both these techniques are approximate, it is not at all obvious that the NCA actually provides higher accuracy as well. We will therefore also compare the NCA and QME results to numerically exact benchmarks obtained from the iQMC method.

Fig. 5 depicts the Fano factor $F$ and its generalizations $F'$ and $F''$ as functions of the bias voltage, once again for three different temperatures. Solid lines represent NCA data and dashed lines represent QME data. Dots indicate iQMC results converged with respect to all numerical parameters. Error bars and shading on these dots correspond to confidence intervals (see Apps. A and B for details regarding how these are obtained). We do not consider second derivatives with respect to the bias voltage here, since obtaining these accurately in iQMC involves further technical challenges. Similarly, we refrain from discussing data below a bias voltage of 0.5. We note that in general, lower voltages and higher order cumulants are more difficult to access in iQMC (see Apps. A and B).

The left panel of Fig. 5 shows the Fano factor $F$. As noted in Sec. V B, at low bias voltages $F$ is dominated by the Nyquist–Johnson singularity and the isolation of Kondo-related features is difficult, but here we focus on the accuracy of the different methods. Generally speaking, reasonable agreement can be observed between the NCA, QME and the iQMC results for all temperatures, both qualitatively and quantitatively. At high temperatures and low voltages, NCA and QME results are almost indistinguishable from each other and accurately capture the trends in the exact result. Importantly, however, the QME always predicts Poisson statistics with a Fano factor of 1 at large bias voltages. The NCA method captures physics not accounted for by the QME method.

Results for the generalized Fano factor $F'$ are presented in the middle panel of Fig. 5. Overall, the three methods predict a qualitatively similar dependence of $F'$ on bias voltage and temperature, still there are qualitative differences. The QME method predicts larger values than the NCA approach, whereby the outcome of the NCA calculations is in better agreement with the iQMC data.
Despite the increased errors associated with the iQMC results for $F'$, it is possible to establish that the NCA method provides more accurate results than the QMC approach. However, a concise observation of the Kondo effect is beyond the iQMC data at hand.

For the second generalized Fano factor $F''$ depicted in the right panel of Fig. 5, the error associated with the iQMC scheme dominates the exact data to the extent that trends in the bias and temperature dependence are non obvious. For this Fano factor, the iQMC method in its current implementation breaks down, indicating an area where the usage of approximate schemes is more favorable. When comparing the QME and the NCA results, the QME approach again predicts larger values for $F''$ than the NCA method. As before, the NCA data is in better agreement with the iQMC results, hinting towards a higher accuracy of the NCA method. For a more detailed analysis, better iQMC data is required.

\section*{VI. SUMMARY}

We developed a simple theoretical approach based on the noncrossing approximation (NCA) to the study of full counting statistics (FCS) in nonequilibrium transport, and implemented it for the Anderson impurity model. The approach can be easily generalized to more generic models. Its accuracy can be improved by diagrammatic means, for example by considering one-crossing and vertex corrections. The NCA method requires substantially more modest computational resources than its numerically exact counterpart, the inchworm Monte Carlo (iQMC) method; and is almost as easy to use as the commonly employed quantum master equations (QMEs). Despite this simplicity, it captures some physics not present in the QME approximation.

To showcase the advantages of the NCA approach to FCS, we compared it against the QME method for the first few transport cumulants. Unsurprisingly, this illustrated that the first shows signatures of the Kondo effect while the latter does not. More interestingly, it showed that the NCA predicts a rich and detailed set of features in the higher order cumulants.

Experimentally, it is often advantageous to consider ratios between transport cumulants, like the Fano factor. However, at low bias voltages the Fano factor is dominated by a Nyquist–Johnson singularity that obstructs one’s view of Kondo-related features. We explored a set of generalized, symmetry-motivated Fano factors constructed from higher order cumulants that are designed to remove this singularity. Within the NCA method, we showed that these quantities embody excellent probes of Kondo physics.

Finally, we established the accuracy of the method upon comparison with numerically exact benchmarks obtained from the iQMC scheme. We showed that the predictability of approximate NCA method is superior to data provided by the QME approach. For the Fano factor, we demonstrated that the NCA can even provide qualitative results.

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\section*{Appendix A: Calculating observables associated with higher order cumulants within the iQMC scheme}

In this appendix, we describe how the observables $I = \frac{\partial}{\partial \tau} C^1$, $S = \frac{\partial}{\partial \tau} C^2$, $S_2 = \frac{\partial}{\partial \tau} C^3$, and $S_3 = \frac{\partial}{\partial \tau} C^4$ were calculated for the steady state within the iQMC framework. We also provide an account of how confidence intervals are estimated. Whereas the main goal of
this paper is to introduce the NCA methodology and its advantages to theorists and experimentalists interested in transport counting statistics, this appendix is aimed at more specialized readers interested in numerically exact methodologies like the iQMC.

In steady state, all cumulants increase linearly with time. We simulate time propagation using the iQMC method until the cumulants display a linear behavior with time. Then, we perform a linear fit to this part of the data. Using the outcome of the fitting routine and averaging over all possible initial conditions, we determine the observables. This procedure is visualized in Fig. 6 for a representative data set. Fig. 6 also allows for an assessment of the intrinsic noise of the iQMC method and the requirement of a linear behavior of the respective cumulants. The error for the iQMC data is then estimated upon considering extremal values for lines connecting two points within the time range where the cumulants display a linear dependence with time (different colored dashed lines in Fig. 6). Again, we average over all initial conditions, whereby we perform a Gaussian error propagation. Strictly speaking, this approach suffers from the fact that the different initial conditions are not statistically independent. Still, the scheme provides a reasonable estimate for the underlying error.

In contrast to the iQMC method, the approximate NCA and QME approaches employed in this work do not exhibit a statistical error. As such, the derivatives entering the expressions for the observables can be calculated by means of finite differences and no fitting procedure is required. Again, the system is propagated until the steady state establishes.

The cumulants within the FCS framework are given by the derivatives of the logarithm of the generating function \( Z(t, \lambda) \) with respect to the counting field \( \lambda \) at \( \lambda = 0 \) (see Eq. (11)). For numerical applications, the generating function is determined for finite values of the counting field and the derivative with respect to \( \lambda \) is calculated, for example, by means of finite differences.

For the iQMC scheme, the statistical error associated with the different cumulants depends on the finite value employed for the counting field. Within this work, we calculate the derivative by means of symmetric finite differences,

\[
C^1(t) = -i \frac{\partial}{\partial \lambda} \ln(Z(t, \lambda)) \bigg|_{\lambda=0} \approx \frac{\ln(Z(t, \lambda_f)) - \ln(Z(t, -\lambda_f))}{2\lambda_f}, \tag{B1a}
\]

\[
C^2(t) = - \frac{\partial^2}{\partial \lambda^2} \ln(Z(t, \lambda)) \bigg|_{\lambda=0} \approx \frac{\ln(Z(t, \lambda_f)) - \ln(Z(t, 0)) + \ln(Z(t, -\lambda_f))}{\lambda_f^2}, \tag{B1b}
\]

etc. for the finite counting field \( \lambda_f \). The dependence of

### Appendix B: Counting field dependence of iQMC data

![Visualization of the dependence of the iQMC data on the finite counting field \( \lambda_f \) used for the numerical calculation for different bias voltages \( V \). The representation data set shown here corresponds to \( T = 0.25 \) and \( V = 1 \). The finite counting field used for the calculation is \( \lambda = 0.75 \). The individual panels show the behavior of the observables and the correspond errors from iQMC data. The individual panels show the behavior of the different cumulants as a function of time. The different lines correspond to different initial conditions, the fits are marked by dashed lines.](image-url)
the (generalized) Fano factors studied in Sec. [VC] on the finite counting field $\lambda_f$ used for calculating the derivative is visualized in Fig. [7]. For large values of $\lambda_f$, the error is small but the estimate for the derivative provided by the numerical derivative deviates from the true value. With decreasing $\lambda_f$, the error associated with the iQMC data increases. Moreover, as higher order cumulants depend on higher order derivatives with respect to the counting field, the numerical error increases and determining accurate data for quantities depending on higher order cumulants becomes increasingly challenging.

We mention that there are also other possible approaches to determine the derivative with respect to the counting field. As such, an alternative but more expensive route is to obtain the full FCS and calculate the generating function $Z(t, \lambda)$ for various different values of $\lambda$. In this case, the derivatives can be determined analytically for a polynomial fit for the counting field dependence of the generating function. This approach was employed, for example, in Ref. [23].

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