Distribution of waiting times between electron cotunnelings

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Single electron transport through quantum dots or molecules that are weakly coupled to macroscopic electrodes is governed by sequential processes in the resonant tunneling regime. In the Coulomb blockade regime, however, cotunneling processes dominate. Cotunneling is an inherently quantum phenomenon, and thus gives rise to interesting observations, such as in increase in the current shot noise. We consider two questions in this paper. Given that an electron has tunneled from the source to the drain via a cotunneling or sequential process, what is the waiting time until another electron cotunnels from the source to the drain? What are the statistical properties of these waiting time intervals? In answering these questions, we extend the existing formalism for waiting time distributions in single electron transport to include cotunneling processes via an n-resolved Markovian master equation. We demonstrate that for a single resonant level the waiting time distribution including cotunneling processes yields information on individual tunneling amplitudes, and that cotunneling induces small correlations in the waiting times.

I. INTRODUCTION

With the ever-present search for smaller transistors and the advent of modern technologies such as quantum computing, the world in recent years has turned its gaze inwards to probe electron transport through nanoscale devices, where a fundamental understanding of the mechanisms of quantum dynamics is required. This has yielded intriguing experimental and theoretical results: for example, single-molecule transistors, quantum heat engines, and spintronics. Of particular interest in quantum nanoscale systems is the potential for encountering transport phenomena that are macroscopically invisible, such as microscopic current fluctuations and individual electron tunneling events, and phenomena that are classically forbidden, such as the existence of electron transport through virtual quantum states that temporarily “violate” energy conservation laws; both of which form the focus of this paper.

Electron transport through quantum systems can display a phenomena known as cotunneling. Inelastic cotunneling was first proposed theoretically by Averin and Odintsov and confirmed experimentally shortly after the theoretical prediction by Geerligs et al. with the introduction of the modern combined inelastic and elastic theory detailed simultaneously by Averin and Nazarov. In contrast to the more commonly studied sequential tunneling, which describes single electron tunneling events and can essentially be described classically, cotunneling is a coherent quantum process that involves the tunneling of an electron from the source to the drain (or vice versa) through an intermediate “virtual” state, which may or may not be classically forbidden. Elastic cotunneling leaves the system with the same energy, while inelastic cotunneling leaves the intermediate quantum system in an excited state. The common explanation is that cotunneling is an example of the uncertainty principle $\Delta t \Delta E \sim \hbar$; energy conservation can be violated only if the electron spends a sufficiently short time in the intermediate virtual state, although in recent years this notion has been challenged by Romito and Gefen.

Cotunneling processes dominate transport in the Coulomb blockade regime, as the electronic energy level is pushed by the Coulomb repulsion outside the voltage bias window and sequential tunneling is exponentially suppressed. Hence, cotunneling manifests experimentally as a small current in the Coulomb blockade regime, and as a small correction to the sequential current in the resonant tunneling regime. Theoretical research into cotunneling has investigated its effect on transport in correlated systems, such as electron-electron and electron-phonon interactions. Additional cotunneling research has focused on heat conductance, transport in double quantum dots, and inelastic cotunneling spectroscopy. Recently multiple authors have studied the noise and full counting statistics (FCS) of cotunneling phenomena in an attempt to explore its effect on current fluctuations. Such investigations have demonstrated that inelastic cotunneling transport induces super-Poissonian shot noise for a variety of systems, which is in agreement with experimental measurements.

Although the zero-frequency noise and FCS are the most common theoretical techniques for describing current fluctuations in quantum nanoscale systems, the waiting time distribution (WTD) has been shown to be a useful method that may contain information complementary to that found in other statistics. Perhaps the recent interest has been spurred onwards in part by the development of real-time single electron detection techniques, which have enabled experimental measurement of microscopic current fluctuations for many different quantum systems. However, there remains experimental difficulties in measuring electron tunnelings via virtual processes due to the collapse of the intermediate state. In the context of this paper the WTD is defined as the conditional probability density that, given an extra electron was counted in the drain electrode at time $t$, another extra electron was counted in the drain at time $t + \tau$, where intermediate tunneling events are allowed. WTDs as a statistical tool have been
used in quantum optics for many years, and since their introduction to single-electron transport have successfully been applied to a diverse range of scenarios, such as systems with electron-electron interactions, electron-phonon interactions, non-Markovian quantum transport, transient dynamics, and spintronics. Furthermore, multiple authors have explored how WTDs can demonstrate whether subsequent electron tunnelings are correlated. However, so far the statistical properties of electron cotunneling events have not been studied using the WTD.

In this paper, we study the WTD for successive tunnelings to the drain, including cotunneling, and compare it to the WTD for successive tunnelings to the drain for only sequential tunneling processes. We first develop a systematic method for extending the current master equation approach for WTDs developed by Brandes to include cotunneling processes, and then demonstrate its use for transport through a single resonant level.

The master equation approach to quantum transport is a powerful tool for analysing quantum electron transport through mesoscopic systems. Although the full master equation is useful for describing quantum effects such as interference and decoherence between double quantum dots, electron transport through quantum dot attached to superconducting leads, and driven quantum transport, in many cases the transport is incoherent and thus is effectively described by rate equations. This is the approach taken in this paper. In order to connect this formalism to waiting times we will in fact have to work with the $n$-resolved master equation.

The transition rates in the master equation are calculated using the T-matrix approach: a perturbation expansion around the tunnelling coupling $H_T$. Sequential tunneling corresponds to the lowest order of this expansion, and cotunneling processes correspond to next-to-leading order in $H_T$: first and second order in the tunneling coupling strength $\gamma$, respectively. Cotunneling has previously been explored via a comprehensive real-time diagrammatic method, however, the T-matrix approach is a suitable approximation for this more rigorous method when $k_BT,eV \gg \gamma$, as in this regime the dynamics does not exhibit non-Markovian phenomena such as broadening and renormalisation of the energy level. Similarly to Thomas and Flindt’s approach, we start with an $n$-resolved master equation, then derive the WTD from the idle time probability and show that for forward tunneling only it reduces to the method introduced by Brandes, albeit with a non-intuitive Liouvillian splitting.

We demonstrate that, likewise to the WTD for sequential tunneling through a single resonant level, the WTD including cotunneling offers information on the individual electrode coupling parameters. Furthermore, elastic cotunneling phenomena introduce small correlations into the transport, which is formally zero when only sequential processes are included. However, the use of the method presents difficulties in two key areas: when the level is inside the voltage bias window and when backward tunneling processes are included.

The paper is organised as follows. Section II outlines the construction of the master equation and the derivation of the WTD including cotunneling. Section III details analytic results for cotunneling through a single resonant level. Section IV outlines the main results and discusses future work. The appendix details calculations and derivations used throughout the paper.

Throughout this paper we use natural units: $\hbar = k_B = e = 1$.

## II. METHODS

### A. Quantum rates for cotunneling processes

Let us consider a nanoscale quantum system weakly coupled to two macroscopic metal electrodes: the source and drain. The source and drain are held at different chemical potentials to cause a voltage bias across the system and induce a non-equilibrium state. For such a setup, the Hamiltonian is

$$H = H_S + H_D + H_M + H_T. \quad (1)$$

The source and drain are modelled as a sea of non-interacting electrons with the Hamiltonians

$$H_S = \sum_s \varepsilon_s a_s^\dagger a_s, \quad H_D = \sum_d \varepsilon_d a_d^\dagger a_d. \quad (2)$$

The operators $a_s^\dagger$ (a) represent creation (annihilation) of an electron in the single-particle state $s/d$ with free energy $\varepsilon_{s/d}$, where $a_{s/d}$ is the corresponding annihilation operator. In this paper we examine transport through a molecule or quantum dot represented by a single resonant level, which is described by the Hamiltonian

$$H_M = \varepsilon a^\dagger a, \quad (3)$$

where the operator $a^\dagger$ (a) creates (annihilates) an electron on the single particle level with energy $\varepsilon$. The interaction between the nanoscale quantum system and the macroscopic electrodes is described by the Hamiltonian

$$H_T = t_S \sum_s (a_s^\dagger a + a_s^\dagger a_s) + t_D \sum_d (a_d^\dagger a + a_d^\dagger a_d), \quad (4)$$

where $t_S$ and $t_D$ are tunneling amplitudes between the molecule and source and drain electrode, respectively. The electrons are considered to be spinless fermions in our derivations.

The quantum system has two states; it can either be empty ($\langle 0 \rangle$) or occupied by a single electron ($\langle 1 \rangle$), with associated probabilities $P_0 = \langle 0 | \rho | 0 \rangle$ and $P_1 = \langle 1 | \rho | 1 \rangle$, where $\rho$ is the density matrix of the single molecular level.
The dynamics of the system is defined by a quantum master equation, which is constructed from quantum rates associated with electron tunneling processes. The rate of transforming from a state occupied by \( m \) electrons to one occupied by \( n \) electrons is denoted \( \Gamma_{nm} \). To calculate the \( \Gamma_{nm} \) we use the T-matrix approach, which is suitable as it provides a direct method for calculating transition rates between eigenstates of quantum many-body systems. Here, we briefly summarise the method outlined by Bruus and Flensberg. First, the Hamiltonian is reformulated as

\[
H(t) = H_S + H_D + H_M + H_T e^{\eta t}, \tag{5}
\]

where the time-independent part \( H_0 = H_S + H_D + H_M \) has a trivial but fast time-evolution \( e^{-iH_0 t} \), and the complex but slow time-evolution is due to the interaction \( H_T e^{\eta t} \), which is treated as a perturbation. The time factor \( e^{\eta t} \) ensures that the perturbation is turned on adiabatically at \( t = -\infty \) by assuming that \( \eta \) is an infinitesimal positive real number.

The starting point for the T-matrix approach is the probability \( P_f(t) \) that the system is in state \( |f⟩ \) at time \( t \) given that time \( t = 0 \) it was in state \( |i⟩ \), which is just the square of their overlap; and from here, the transition rate between the two states is the time derivative of \( P_f(t) \):

\[
\Gamma_{fi} = \frac{d}{dt} |⟨f|i(t)⟩|^2. \tag{6}
\]

Using the interaction picture, Eq.\ref{5} is transformed to

\[
\Gamma_{fi} = 2\pi |⟨f|T|i⟩|^2 \delta(E_i - E_f), \tag{7}
\]

where the T-matrix is

\[
T = H_T + H_T \frac{1}{E_i - H_0 + i\eta} H_T
+ H_T \frac{1}{E_i - H_0 + i\eta} H_T \frac{1}{E_i - H_0 + i\eta} H_T + ... \tag{8}
\]

The sequential tunneling regime corresponds to second order in \( H_T \) in the transition rates: the first linear term in the T-matrix. So the sequential rates are

\[
\Gamma_{fi} = 2\pi |⟨f|H_T|i⟩|^2 \delta(E_i - E_f), \tag{9}
\]

which is just the standard Fermi’s Golden Rule.

In the many-body configuration in the full Fock space, the initial and final states are tensor products of the discrete system states (molecular or quantum dot) and continuous electrode states: \( |m⟩ \otimes |i_{S/D}⟩ \) and \( |n⟩ \otimes |f_{S/D}⟩ \), with eigen-energies \( E_m + \varepsilon_{i_{S/D}} \) and \( E_n + \varepsilon_{f_{S/D}} \) respectively. Consequently, there are multiple final and initial states that correspond to a system state of \( |m/n⟩ \); they must be summed over, and the initial states weighted with a thermal distribution function \( W_{im}^{S/D} \):

\[
\Gamma_{im}^{S/D} = 2\pi \sum_{f_{S/D} i_{S/D}} |⟨f_{S/D}|⟨n|H_T^{S/D} |i_{S/D}⟩|^2 W_{im}^{S/D}
\times \delta(E_m - E_n + \varepsilon_{i_{S/D}} - \varepsilon_{f_{S/D}}). \tag{10}
\]

At this point we can now calculate the sequential rates for electron tunneling between the electrodes and the system:

\[
\Gamma_{10}^{S/D} = \gamma^{S/D} n_F(\varepsilon - \mu_{S/D}) \quad \text{and} \quad \tag{11}
\]

\[
\Gamma_{01}^{S/D} = \gamma^{S/D} (1 - n_F(\varepsilon - \mu_{S/D})), \tag{12}
\]

where \( \gamma^{S/D} = 2\pi |t_{S/D}|^2 \rho(\varepsilon_{S/D}) \) and \( \rho(\varepsilon_{S/D}) \) is the density of states for the source and drain electrodes, which is assumed to be constant. Throughout the paper we use a symmetric coupling, such that \( \gamma^S = \gamma^D = \gamma \). The \( n_F(\varepsilon - \mu_{S/D}) \) are the Fermi-Dirac distributions for the source and drain electrodes:

\[
n_F(\varepsilon - \mu_{S/D}) = \frac{1}{1 + e^{(\varepsilon - \mu_{S/D})/T_{S/D}}}. \tag{13}
\]

When the electronic level is within the bias window and in the limit of infinite source-drain bias, which is achieved by making the voltage \( \mu_S - \mu_D \) large, the configuration undergoes forward tunneling only: that is, from the source to the molecule or from the molecule to the drain. However, in the Coulomb blockade regime the electronic level is outside the bias window, regardless of the large voltage. To reconcile the two scenarios we note that their combined processes are tunneling from the source to the molecule, from the molecule to the source, and from the molecule to the drain. In effect, the total sequential rates can be reduced to \( \Gamma_{10} = \Gamma_{10}^S + \Gamma_{01}^S + \Gamma_{10}^D \), where we have adopted the shorthand \( \Gamma^D = \Gamma_{01}^D \).

The next-to-leading term in the T-matrix expansion is the second order in the tunneling interaction \( H_T \), which is fourth order in \( H_T \) in the rate expression, and describes cotunneling effects. Since there are two scatterings on the perturbation and the system is in the infinite bias limit, there are only two cotunneling pathways. Either an electron tunnels into the empty level from the source and then tunnels out again in the same quantum process, or an electron tunnels out from the level into the drain and is replaced by an electron from the source in the same quantum process. The two processes have transition rates \( \Gamma_{00}^{(2)} \) and \( \Gamma_{11}^{(2)} \) respectively. Since the same molecular energy level is filled and emptied, both processes are elastic. To fourth order in \( H_T \) Eq.\ref{11} becomes
\[ \Gamma_{nn}^{(2)} = 2\pi \lim_{\eta \to 0^+} \sum_{i,f} \frac{1}{E_{i,n} - H_0 + i\eta} \langle f | H_T^{S/D} | n \rangle \langle n | H_T^{S/D} | i \rangle W_{i,m} W_{m,n} \delta(\varepsilon_i - \varepsilon_f). \] (14)

It is assumed that the thermal probabilities for the source and drain are independent and so can be factored: \( W_{i,m} W_{m,n} \). Additionally, we assume weak coupling, so that the electrode thermal probabilities are independent of the state of the quantum system at time \( t = t_0 \). Evaluating Eq. (14) and taking the appropriate limits, one obtains firstly that \( \Gamma_{00}^{(2)} = \Gamma_{11}^{(2)} = \Gamma^{(2)} \) and secondly that

\[ \Gamma^{(2)} = \beta \frac{\Gamma_S \Gamma_D}{4\pi^2} n_B(\mu_D - \mu_S) \times \Im \{ \psi^{(1)} \left( \frac{1}{2} + \frac{i\beta}{2\pi}(\varepsilon - \mu_S) \right) - \psi^{(1)} \left( \frac{1}{2} + \frac{i\beta}{2\pi}(\varepsilon - \mu_D) \right) \}. \] (15)

Here \( \beta = \frac{1}{2\pi} \), \( \psi^{(1)} \) is the digamma function, and \( n_B(\mu_D - \mu_S) \) is the Bose-Einstein distribution function:

\[ n_B(\mu_D - \mu_S) = \frac{1}{e^{(\mu_D - \mu_S)\beta} - 1}. \] (16)

The details for deriving Eq. (13) are in Appendix A.

From here it is tempting to construct the standard master equation for probabilities that the system is occupied by 0 or 1 electrons. However, since the cotunneling rates do not change the state of the quantum system, they do not contribute to the rate equation for the system state probabilities. Instead, one must consider the macroscopic parts of the experimental setup that cotunneling does affect.

**B. n-resolved master equation**

The master equation can be resolved upon the number of electrons transferred to the drain; so \( P_0(n,t) \) is the probability that the system is empty at time \( t \) and that \( n \) electrons were transferred to the drain in the interval \( [0, t] \), and similarly for \( P_1(n, t) \). For the infinite bias regime \( n = 0, 1, 2, 3, \ldots, +\infty \). Thus the total probability that \( n \) electrons were transferred by time \( t \) is

\[ P(n, t) = (I, P(n, t)) = P_0(n, t) + P_1(n, t), \] (17)

where \( I \) is the identity vector

\[ I = \begin{bmatrix} 1 & 1 \end{bmatrix} \] (18)

and \( P(n, t) \) is the probability vector

\[ P(n, t) = \begin{bmatrix} P_0(n, t) \\ P_1(n, t) \end{bmatrix}. \] (19)

The \( n \)-resolved Markovian master equation follows the general form

\[ \dot{P}(n, t) = \sum_{n'} L(n - n') P(n, t). \] (20)

For the tunneling interaction in Eq. (14) each \( n \) is connected only to its neighbouring values \( n' = n, n \pm 1 \) and for a single resonant level in the infinite bias regime, including cotunneling processes, the \( n \)-resolved master equation is intuitively

\[
\begin{bmatrix}
\dot{P}_0(n, t) \\
\dot{P}_1(n, t)
\end{bmatrix} = \begin{bmatrix}
-(\Gamma_{10}^{S} + \Gamma^{(2)}) & \Gamma_{01}^{S} \\
\Gamma_{10}^{D} + \Gamma_{01}^{D} + \Gamma^{(2)} & -\Gamma_{10}^{S} - \Gamma^{(2)}
\end{bmatrix} \begin{bmatrix}
P_0(n, t) \\
P_1(n, t)
\end{bmatrix} + \begin{bmatrix}
\Gamma^{(2)} \\
0
\end{bmatrix} \begin{bmatrix}
P_{n-1}(n, t)
\end{bmatrix}.
\] (21)

Evidently, the \( n \)-resolved master equation is an infinite set of coupled equations since \( n = 0, 1, 2, \ldots, +\infty \). To solve, we use the elegant idea, proposed first by Nazarov and extended to master equations by Bagrets and Nazarov, of introducing a continuous counting field \( \chi \), with \( 0 \leq \chi \geq 2\pi \).

\[
P(\chi, t) = \sum_n e^{i\chi n} P(n, t),
\] and

\[
P(n, t) = \frac{1}{2\pi} \int_{0}^{2\pi} e^{-i\chi n} P(\chi, t) d\chi,
\] (23)

Multiplying Eq. (21) by \( e^{i\chi n} \) and transforming \( \sum_n e^{i\chi n} P(n - 1, t) \rightarrow \sum_m e^{i(m+1)\chi} P(m, t) \), one obtains the \( n \)-resolved master in \( \chi \)-space in the form

\[
\dot{P}(\chi, t) = \begin{bmatrix}
\Gamma_{10}^{S} e^{i\chi} + \Gamma_{01}^{S} \\
-\Gamma_{10}^{D} - \Gamma_{01}^{D} - \Gamma^{(2)} e^{i\chi}
\end{bmatrix} \begin{bmatrix}
P_0(\chi, t) \\
P_1(\chi, t)
\end{bmatrix}.
\] (24)

Eq. (24) has the formal solution:

\[
P(\chi, t) = e^{L(\chi)t} P(\chi, 0),
\] (25)

where the initial condition is \( P(\chi, 0) = P(n = 0, 0) \), since
it is assumed that electron counts are monitored after $t = 0$. We also assume that the system was prepared in the steady state at $t = 0$; that means $P(n = 0, 0) = P$ with $P$ being a null vector of the standard Liouvillian
\[ \mathbf{L}(0)\mathbf{P} = 0. \] (26)

Then, the probability that $n$ electrons have been transferred to the drain by time $t$ is
\[ P(n, t) = \frac{1}{2\pi} \int_{0}^{2\pi} e^{-in\chi} (\mathbf{I}, e^{\mathbf{L}(\chi)t}\mathbf{P}) d\chi. \] (27)

At this point one could define a moment-generating function $M(\chi, t) = (\mathbf{I}, e^{\mathbf{L}(\chi)t}\mathbf{P})$ and derive the moments of transferred charge $\langle n^k \rangle = (-i)^k \frac{\partial^k}{\partial \chi^k} M(\chi, t) |_{\chi=0}$ to obtain the full counting statistics. However, we are interested in the WTD.

C. WTD definition

The WTD can be defined in different ways. Based on the ideas from quantum optics single photon counting theories\textsuperscript{36,37} Brandes first introduced the concept of a WTD to electron transport with a formalism that used “jump” operators defined from the master equation of the system.\textsuperscript{37} In order to include cotunneling rates, however, we will start with the WTD defined in terms of the idle time probability\textsuperscript{38,39}
\[ \omega(\tau) = \frac{1}{p} \frac{\partial^2}{\partial \tau^2} \Pi(\tau). \] (28)

The idle time probability $\Pi(\tau)$ is the probability that there were no electron tunnelings to the drain in the measurement time $\tau$. Here, $p$ is the initial probability of observing an electron tunneling to the drain, and can be defined in terms of $\Pi(\tau)$ as well: $p = -\frac{\partial}{\partial \tau} \Pi(\tau) |_{\tau=0}$. The key relation is that the idle time probability is the probability for no electrons to be transferred to the drain in time $\tau$, so that $\Pi(\tau) = P(0, \tau)$\textsuperscript{38} The moment-generating function can be written as
\[ M(\chi, \tau) = P(0, \tau) + \sum_{n=1}^{\infty} e^{in\chi} P(n, \tau); \] (29)
hence in the infinite bias regime the idle time distribution is
\[ \Pi(\tau) = \lim_{\chi \to \pm \infty} (\mathbf{I}, e^{\mathbf{L}(\chi)\tau}\mathbf{P}). \] (30)

Combining with the definition of the WTD from Eq (28), we get
\[ w(\tau) = -\lim_{\chi \to \pm \infty} \frac{(\mathbf{I}, \mathbf{L}(\chi)e^{\mathbf{L}(\chi)\tau}\mathbf{L}(\chi)\mathbf{P})}{(\mathbf{I}, \mathbf{L}(\chi)\mathbf{P})}, \] (31)
and in Laplace space
\[ \tilde{w}(z) = -\lim_{\chi \to \pm \infty} \frac{(\mathbf{I}, \mathbf{L}(\chi)(z - \mathbf{L}(\chi))^{-1}\mathbf{L}(\chi)\mathbf{P})}{(\mathbf{I}, \mathbf{L}(\chi)\mathbf{P})}. \] (32)

Similarly to the sequential tunneling case, $\mathbf{L}(\chi)$ is formally split into a quantum jump part $\mathbf{J}(\chi) = \mathbf{J}e^{i\chi}$, containing the $\chi$-dependence, and the $\chi$-independent $\mathbf{L}_0$:
\[ \mathbf{L}(\chi) = \begin{bmatrix} -\left(\begin{array}{cc} \Gamma_S^0 + \Gamma^{(2)}_0 \end{array}\right) & \Gamma_S^1 \\ -\left(\begin{array}{cc} \Gamma_D^0 + \Gamma^{(2)}_0 \end{array}\right) & \Gamma_D^0 \end{bmatrix} \]  
\[ + \begin{bmatrix} \Gamma^{(2)}_0 & 0 \\ 0 & \Gamma^{(2)}_0 \end{bmatrix} e^{i\chi} = \mathbf{L}_0 + \mathbf{J}e^{i\chi}. \] (33)

Using the splitting in Eq. (33) the WTD becomes
\[ w(\tau) = -\lim_{\chi \to \pm \infty} \frac{(\mathbf{I}, (\mathbf{L}_0 + \mathbf{J}e^{i\chi})e^{(\mathbf{L}_0 + \mathbf{J}e^{i\chi})\tau}(\mathbf{L}_0 + \mathbf{J}e^{i\chi})\mathbf{P})}{(\mathbf{I}, (\mathbf{L}_0 + \mathbf{J}e^{i\chi})\mathbf{P})}, \] (34)
which is
\[ w(\tau) = -\frac{(\mathbf{I}, \mathbf{L}_0 e^{\mathbf{L}_0\tau}\mathbf{L}_0\mathbf{P})}{(\mathbf{I}, \mathbf{L}_0\mathbf{P})}. \] (35)

By making note that $\mathbf{L}_0 = \mathbf{L}(0) = \mathbf{J}$, $\mathbf{L}(0)\mathbf{P} = 0$ and $\langle (\mathbf{I}, \mathbf{L}(0)\mathbf{A}) \rangle = 0$ for arbitrary $\mathbf{A}$, we obtain the standard expressions for the WTD in the time domain:
\[ w(\tau) = -\frac{(\mathbf{I}, \mathbf{J}e^{\mathbf{J}\tau}\mathbf{J}\mathbf{P})}{(\mathbf{I}, \mathbf{J}\mathbf{P})}, \] (36)
which in Laplace space becomes
\[ \tilde{w}(z) = -\frac{(\mathbf{I}, \mathbf{J}(z - \mathbf{L}_0)^{-1}\mathbf{J}\mathbf{P})}{(\mathbf{I}, \mathbf{J}\mathbf{P})}. \] (37)

Here, we see that in the case of forward tunneling the WTD reduces to the one calculated using Brandes’ method.\textsuperscript{37} Despite this, the $n$-resolved master equation is still necessary as it tells us how to construct $\mathbf{L}_0$ from the quantum jump operator $\mathbf{J}$. We notice that the method breaks down if backwards tunneling processes are included, as their factor $e^{-i\chi}$ will diverge in the limit $\chi \to i\infty$.

Although the single WTD is itself an interesting quantity, in order to compute higher-order expectation values and analyse microscopic fluctuations we must also generalise it to two or more consecutive waiting times. For example, the WTD for two waiting times, $w_2(\tau_2, \tau_1)$, is defined as the joint probability distribution that the first electron waits time $\tau_1$ and the next electron waits time $\tau_2$ before tunneling to the drain\textsuperscript{38,39}
\[ w_2(\tau_2, \tau_1) = -\frac{(\mathbf{I}, \mathbf{J}e^{\mathbf{J}\tau_2}\mathbf{J}e^{\mathbf{J}\tau_1}\mathbf{J}\mathbf{P})}{(\mathbf{I}, \mathbf{J}\mathbf{P})}. \] (38)

Moments of the single WTD are easily calculable by introducing a moment-generating function over $\tau$:
\[ K(x) = \int_{0}^{\infty} d\tau e^{i\tau x} w(\tau) = \frac{(\mathbf{I}, \mathbf{J}G(x)\mathbf{J}\mathbf{P})}{(\mathbf{I}, \mathbf{J}\mathbf{P})}, \] (39)
where $x$ is a real number and
\[ G(x) = (L_0 + ix)^{-1}. \]  

We obtain all possible moments by direct differentiation with respect to $x$, such that
\[ \langle \tau^n \rangle = \int_0^\infty d\tau \tau^n w(\tau) = n!(-1)^{n+1}(I, JG(0)^{n+1}J\bar{P})/(I, J\bar{P}). \]  

The second-order expectation value is calculated similarly:
\[ \langle \tau_2 \tau_1 \rangle = \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \tau_1 \tau_2 w_2(\tau_2, \tau_1) = (I, JG(0)^2JG(0)^2J\bar{P})/(I, J\bar{P}). \]

III. RESULTS

In this section we investigate, numerically and analytically, statistics of waiting time intervals between successive electron cotunneling events. Using Eq. (37) and the splitting outlined in Eq. (38), the WTD in Laplace space is
\[ \tilde{w}(z) = \frac{a + bz}{(z + z_+)(z + z_-)} \]  
and the corresponding WTD in time domain is
\[ w(\tau) = \frac{a - bz_+}{z_+ - z_-} e^{-z_+ \tau} - \frac{a - bz_-}{z_+ - z_-} e^{-z_- \tau}, \]  

where the coefficients of the linear function in the numerator are
\[ a = \left\{ \left( \Gamma_D^S \Gamma_0^S \right)^2 + \Gamma^{(2)} \left( \Gamma_D^S + \Gamma_0^S + \Gamma_1^S \right) \right\} \left( \Gamma^{(2)} \right)^2 + 2\Gamma_D^S \Gamma_0^S \]  
\[ + \frac{(\Gamma^{(2)})^2 (\Gamma_D^S)^2 + (\Gamma_0^S + \Gamma_1^S)^2 + \Gamma_D^S (2\Gamma_0^S + 3\Gamma_1^S)}{\left\{ \Gamma_D^S \Gamma_0^S + \Gamma^{(2)} \left( \Gamma_D^S + \Gamma_0^S + \Gamma_1^S \right) \right\}}, \]  
\[ b = \frac{\Gamma^{(2)} (2\Gamma_D^S \Gamma_0^S + \Gamma^{(2)} \left( \Gamma_D^S + \Gamma_0^S + \Gamma_1^S \right))}{\Gamma_D^S \Gamma_0^S + \Gamma^{(2)} \left( \Gamma_D^S + \Gamma_0^S + \Gamma_1^S \right)} \]

The poles of the Laplace space WTD, which are also the exponents in the time-space WTD, are
\[ z_\pm = \frac{1}{2} \left[ 2\Gamma^{(2)} + \Gamma_D^S + \Gamma_0^S + \Gamma_1^S \right] \]  
\[ \pm \sqrt{(\Gamma^{(2)})^2 + 2\Gamma^D \left( \Gamma_0^S - \Gamma_1^S \right) + \left( \Gamma_0^S + \Gamma_1^S \right)^2}. \]

Interestingly, the position of the poles yield information on the individual source-drain couplings, similarly to the results Brandes found for sequential tunneling through a single resonant level. In Fig. (1) the poles and the prefactors from Eq. (13) and Eq. (14) have been plotted over a range of voltages. The evolution of the sequential and cotunneling WTD is shown in Fig. (2). The level starts just above the bias window, where sequential processes can still contribute to the transport. Hence, in (a) of Fig. (2) the cotunneling distribution has the same shape as the sequential with modified parameters. The average waiting times are of the same order of magnitude, so cotunneling processes have not yet begun to dominate the transport. As the level moves further away from the bias, the distributions are modified as sequential processes disappear and cotunneling processes dominate. When the level is deep in the Coulomb blockade regime the poles are different but the prefactors are the same, so one of the exponents dominates the other and it produces essentially a single decaying exponential distribution, which is shown in (d) of Fig. (2).

In the Coulomb blockade regime cotunneling provides a quantum pathway for electrons to tunnel through the system that is not visible in the sequential physics. To second order then, the waiting time probability density for subsequent tunnelings to the drain is no longer zero as it is in the sequential regime. In (d) of Fig. (2) the sequential WTD appears to be nonzero; however, this is only for exceptionally small times on the scale of the exceptionally large $\langle \tau \rangle_{SEQ}$. Deep in the Coulomb blockade the shape of the cotunneling WTD is that of a negative exponential, which implies that there is a nonzero probability for electron jumps to the drain to happen simultaneously. From the point of view of the theory this is plausible, as the quantum process that moves an electron from the source to the drain is macroscopically instantaneous.

The moments of the WTD can be derived analytically for a single resonant level model, using Eq. (39). The
FIG. 2: Colour online. Sequential and cotunneling WTDs for four voltages when the level is in the Coulomb blockade regime. The y-axis represents $w_{\text{seq}}(\tau)/\langle \tau \rangle_{\text{seq}}$ and $w_{\text{co}}(\tau)/\langle \tau \rangle_{\text{co}}$ for the sequential and cotunneling WTDs, respectively. Additionally, the sequential and cotunneling WTDs have been plotted over the same relative time: $\tau_{\text{co}}/\langle \tau \rangle_{\text{co}} = \tau_{\text{seq}}/\langle \tau \rangle_{\text{seq}} = \tau/\langle \tau \rangle$. The electronic level is $\varepsilon = 2\text{meV}$, $k_B T = 75\mu\text{eV}$, and $\gamma = k_B T$. Parameters for each plot are: (a) $\mu_S = -\mu_D = 1.9\text{meV}$, $\langle \tau \rangle_{\text{seq}} = 169\text{ps}$, and $\langle \tau \rangle_{\text{co}} = 129\text{ps}$; (b) $\mu_S = -\mu_D = 1.8\text{meV}$, $\langle \tau \rangle_{\text{seq}} = 541\text{ps}$ and $\langle \tau \rangle_{\text{co}} = 275\text{ps}$; (c) $\mu_S = -\mu_D = 1.7\text{meV}$, $\langle \tau \rangle_{\text{seq}} = 1.96\text{ns}$, and $\langle \tau \rangle_{\text{co}} = 561\text{ps}$; (d) $\mu_S = -\mu_D = 1.8\text{meV}$, $\langle \tau \rangle_{\text{seq}} = 7.33\text{ns}$, and $\langle \tau \rangle_{\text{co}} = 976\text{ps}$.

FIG. 3: Pearson correlation coefficient over a range of voltages in the Coulomb blockade regime, and for multiple $\gamma$. The electronic level is $\varepsilon = 2\text{meV}$ and the temperature is $k_B T = 75\mu\text{eV}$.

FIG. 4: Relative dispersion over a range of voltages in the Coulomb blockade regime. The electronic level is $\varepsilon = 2\text{meV}$, the temperature is $k_B T = 75\mu\text{eV}$, and $\gamma = k_B T$.

The average waiting time is

$$\langle \tau \rangle = \frac{\Gamma_D + \Gamma_S^{10} + \Gamma_{10}^{S}}{\Gamma_D \Gamma_{10}^{S} + \Gamma_{10}^{S} \Gamma_{10}^{S} + \Gamma_{10}^{S}} = \frac{1}{\langle I \rangle^{(2)}}.$$  

where $\langle I \rangle^{(2)}$ is the current including cotunneling processes. The pearson correlation coefficient can be calculated from

$$p = \left( \frac{\tau_1 \tau_2 - \langle \tau \rangle^2}{\langle \tau_1 \rangle \langle \tau_2 \rangle - \langle \tau \rangle^2} \right) - \langle \tau \rangle^2,$$

where the components are

$$A = (\Gamma^{(2)})^2 \Gamma_D \Gamma_{10}^{S},$$  

$$B = (\Gamma^{(2)})^2 + \Gamma_D \Gamma_{10}^{S} + (\Gamma^{(2)}) \left( \Gamma_D + \Gamma_{10}^{S} + \Gamma_{10}^{S} \right),$$

and

$$C = (\langle \tau \rangle^2).$$
\[ C = (\Gamma^{(2)}_0 (\Gamma^D + \Gamma^S_{01} + \Gamma^S_{10}))^2 + \Gamma^{(2)}_0 (\Gamma^D + \Gamma^S_{01} + \Gamma^S_{10})^3 + \Gamma^D \Gamma^S_{01} (\Gamma^D)^2 + 2\Gamma^D \Gamma^S_{01} (\Gamma^S_{01} + \Gamma^S_{10})^2. \] (52)

For sequential tunneling through a single resonant level, transport is completely uncorrelated and the Pearson correlation coefficient is formally zero. Fig. (43) shows that, contrary to sequential tunneling, electron waiting times for cotunneling through a single resonant level are negatively correlated. However, the correlation is negligibly small, as expected from the small perturbative changes that cotunneling brings.

Additionally, the cotunneling alters the relative dispersion

\[ \frac{\sqrt{\langle \tau^2 \rangle - \langle \tau \rangle^2}}{\langle \tau \rangle} \] (53)

in comparison to sequential tunneling. Fig. (44) shows that when the level is deep in the Coulomb blockade regime that the relative dispersion is 1 for both sequential tunneling and cotunneling. For cotunneling this implies that the cotunneling “channel” is fully open, while the sequential tunneling “channel” is fully closed. As the voltage gets larger and sequential processes start to contribute, the cotunneling processes keep the relative dispersion higher, due to them still majorly contributing in the Coulomb blockade regime.

The other interesting regime is when the electronic level is inside the bias window, such that \( \varepsilon < eV \), where second order contributions actually reduce the total current, which amounts to negative regularised cotunneling rates. From the point of view of the theory the total transition rate \( \Gamma_00 = \frac{1}{2} n_F(\varepsilon - \mu_S)(1 - n_F(\varepsilon - \mu_D)) + \Gamma^{(2)}_0 \) is still positive, which is physically necessary, but the regularised cotunneling part \( \Gamma^{(2)}_0 \) can be negative. In such a regime these negative rates cause the WTD itself to be unphysically negative for small \( \tau \) (\( \sim 10^3 f_s \)).

\[ w(0) = \frac{\Gamma^{(2)}_0 (2\Gamma^D \Gamma^S_{10} + \Gamma^{(2)}_0 (\Gamma^D + \Gamma^S_{01} + \Gamma^S_{10}))}{\Gamma^D \Gamma^S_{10} + \Gamma^{(2)}_0 (\Gamma^D + \Gamma^S_{01} + \Gamma^S_{10})} \] (54)

Evidently in such a scenario the positivity condition of the WTD is violated. It is not yet clear how to resolve this small pathology; evidently there should be a well-defined WTD for the cotunneling assisted sequential regime.

\[ \Gamma^{(2)}_00 = 2\pi \lim_{\eta \to 0^+} \sum_{i,f} \left| \langle f \mid 0 \rangle \langle 0 \mid V_D \frac{1}{E_{i,n} - H_0 + i\eta} V_S \mid 0 \rangle \langle 0 \rangle \right|^2 W^D_{i0} W^S_{i0} \times \delta(\varepsilon_{i,0} - \varepsilon_{f,0}). \] (A1)

The initial state of the dot is \( |0\rangle \otimes |i\rangle \) and the final state is \( |0\rangle \otimes a^\dagger_{\nu} a_{\nu,s} |i\rangle \). To span the possible configurations

IV. CONCLUSION

In this paper we have successfully extended Markovian master equation based techniques for calculating WTDs in quantum electron transport to include cotunneling effects, and demonstrated the method for a single resonant level. Additionally, we have demonstrated that, similarly to the WTD for sequential tunneling through a single resonant level, the cotunneling WTD in Laplace space provides information on the individual source-drain couplings. Cotunneling processes also increase the variance in waiting times for successive tunnelings to the drain, as is shown in Fig. (44). This also illustrates the difference between sequential tunneling and cotunneling in the Coulomb blockade regime as sequential channels are fully closed while cotunneling channels are fully open, yet both have the same relative dispersion. It is only when both processes contribute to the current that the relative dispersion differs. Furthermore, we have shown that cotunneling induces a non-zero correlation between subsequent electron tunneling times to the drain. However, these correlations are negligibly small and the system can still not be said to be ‘correlated’, even with cotunneling effects included. Further work in an interacting system such as an Anderson impurity would highlight the relationship that cotunneling has on correlations and interactions.

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Appendix A: Cotunneling rates

In this appendix we explicitly derive the cotunneling rates shown in Eq. (44), from the starting point of Eq. (43). We note that the derivation generally follows the regularisation procedure detailed by Koch et al. [12, 13].

When the dot is initially empty, the tunneling rate is
after the cotunneling rate is summed over the electrode states $\nu_S$ and $\nu_D$. Additionally, it is assumed that the metal electrodes have a constant and continuous density of states. With these assumptions in mind, the rate reduces to

\[
\Gamma_{nn}^{(2)} = \frac{S\gamma^D}{2\pi} \lim_{\eta \to 0} \int d\varepsilon \frac{1}{(\varepsilon - \varepsilon_M)^2 + \eta^2} n_F(\varepsilon - \mu_S)\left[1 - n_F(\varepsilon - \mu_D)\right].
\] (A2)

Numerically with Cauchy’s principal value. However, we follow the approach of Koch et al. and evaluate the integral analytically by transforming it to a contour integral over a semicircle in the upper half-plane of complex space and using residue theory. The rate is then expanded in a power series about $\eta$ and the term proportional to $\eta^{-1}$ is discarded, as with the $\gamma^D$ prefactor its overall order is $\sim \gamma$ and hence corresponds to a sequential process. The final expression is Eq.\(^{15}\) where the digamma functions $\psi^{(1)}\left(\frac{1}{2} + \frac{i\pi}{2\eta}(\varepsilon - \mu_{S/D})\right)$ originate from the complex poles of the Fermi-Dirac distributions $n_F(\varepsilon - \mu_{S/D})$, known as the Matsubara frequencies.

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