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Electron Waiting Times in a Strongly Interacting Quantum Dot: Interaction Effects and Higher-Order Tunneling Processes

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Distributions of electron waiting times have been measured in several recent experiments and have been shown to provide complementary information compared with what can be learned from the electric current fluctuations. Existing theories, however, are restricted to either weakly coupled nanostructures or phase-coherent transport in mesoscopic conductors. Here, we consider an interacting quantum dot and develop a real-time diagrammatic theory of waiting time distributions that can treat the interesting regime, in which both interaction effects and higher-order tunneling processes are important. Specifically, we find that our quantum-mechanical theory captures higher-order tunneling processes at low temperatures, which are not included in a classical description, and which dramatically affect the waiting times by allowing fast tunneling processes inside the Coulomb blockade region. Our work paves the way for systematic investigations of temporal fluctuations in interacting quantum systems, for example close to a Kondo resonance or in a Luttinger liquid.

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Introduction.—Electronic waiting time distributions are an important concept in the analysis of quantum transport in nanoscale structures [1–36], and recently they have been measured in several experiments [37–42]. Unlike full counting statistics, which typically considers the time-integrated current fluctuations [43–45], waiting time distributions are concerned with the short time span that passes between the charge transfers in a nanoscale conductor. The waiting time distribution contains a wealth of information about the underlying transport processes. For example, it has been predicted that the waiting time distribution for a quantum point contact should exhibit a crossover from Wigner-Dyson statistics at full transmission to Poisson statistics close to pinch-off [4,9], illustrating a profound connection between free fermions and the eigenvalues of random matrices [46].

Experimentally, waiting time distributions have been used to demonstrate accurate control of the emission time statistics of a dynamic single-electron transistor [22,41] and to characterize the splitting of Cooper pairs in the time domain [26,36,42]. Theoretically, electron waiting time distributions have been considered in two opposite regimes. For sequential tunneling in nanostructures, the waiting time distribution can be obtained from a master-equation description of the charge transport [1,3,35]. On the other hand, for phase-coherent transport of noninteracting electrons, the waiting time distribution can be evaluated using scattering theory [4,7,9], tight-binding calculations [12], or Green’s function methods [11,14]. However, for the interesting intermediate regime, where both interaction effects and higher-order tunneling processes are important,

a systematic theory of electronic waiting time distributions has so far been lacking.

In this Letter, we investigate the temporal fluctuations of charge tunneling in a Coulomb-blockade quantum dot and develop a real-time diagrammatic theory of electron waiting time distributions that can treat both strong interactions and higher-order tunneling processes [47–49]. Cotunneling

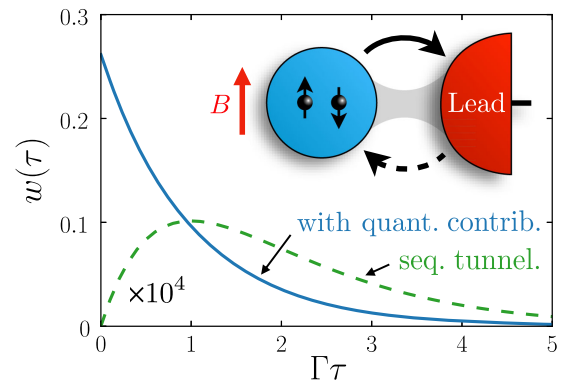


FIG. 1. Electron waiting times for an interacting quantum dot in a magnetic field. Electrons tunnel back and forth (arrows) between the quantum dot and an external reservoir. We consider the waiting time between electrons leaving the quantum dot (full arrow). Waiting time distributions are shown for lowest-order sequential tunneling only (green curve) as well as with higher-order processes included (blue curve). The level position with respect to the chemical potential of the lead is $\epsilon = -0.2U$, where U is the Coulomb interaction, and the magnetic field causes the Zeeman splitting $\Delta = 1.1U$. The temperature and the coupling are $k_B T = U/30$, $\hbar\Gamma = 10^{-7}U$.

processes can be described at the level of mean currents using T -matrix approaches [48–50], which, however, cannot account for non-Markovian effects that influence the charge transport fluctuations beyond average values [51,52]. Our setup is illustrated in Fig. 1, where we also show waiting time distributions for sequential tunneling only as well as with higher-order tunneling processes included. As we will see, the higher-order tunneling processes are predominantly of quantum nature, and they tend to get washed out by an increasing electronic temperature for which we recover a classical description based on sequential tunneling. While we here focus on quantum dots, it will become clear that our approach can be applied to a wide range of interacting nanostructures at low temperatures and bias voltages, where quantum effects are important [53–59].

Hamiltonian.—We start by considering a quantum dot coupled to a single electrode before extending the discussion to a transport setup with both a source and a drain electrode. The full Hamiltonian of the setup reads as

$$\hat{H} = \hat{H}_{\text{qd}} + \hat{H}_{\text{res}} + \hat{H}_{\text{tun}}, \quad (1)$$

where the quantum dot $\hat{H}_{\text{qd}} = \sum_{\sigma} \epsilon_{\sigma} \hat{d}_{\sigma}^{\dagger} \hat{d}_{\sigma} + U \hat{d}_{\uparrow}^{\dagger} \hat{d}_{\uparrow} \hat{d}_{\downarrow}^{\dagger} \hat{d}_{\downarrow}$ can be either empty $|0\rangle$, occupied by a single electron $|\sigma\rangle$ with spin $\sigma = \uparrow, \downarrow$, or doubly occupied $|d\rangle$, with U denoting the on site Coulomb interactions. An applied magnetic field lifts the degeneracy of spin-up, $\epsilon_{\uparrow} = \epsilon + \Delta/2$, and spin-down electrons, $\epsilon_{\downarrow} = \epsilon - \Delta/2$, where Δ is the Zeeman splitting. The orbital energy ϵ relative to the chemical potential μ of the reservoir can be tuned by an external gate voltage. The reservoir contains noninteracting electrons and is described by the Hamiltonian $\hat{H}_{\text{res}} = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma} \hat{a}_{\mathbf{k}\sigma}^{\dagger} \hat{a}_{\mathbf{k}\sigma}$, while electron tunneling between the quantum dot and the reservoir is governed by the Hamiltonian $\hat{H}_{\text{tun}} = \sum_{\mathbf{k}\sigma} (g \hat{a}_{\mathbf{k}\sigma}^{\dagger} \hat{d}_{\sigma} + g^{*} \hat{d}_{\sigma}^{\dagger} \hat{a}_{\mathbf{k}\sigma})$. The tunnel coupling between the quantum dot and the lead is given by $\Gamma = 2\pi |g|^2 \nu / \hbar$, where ν is the density of states in the lead, which is kept at temperature T .

Master equation.—We describe the quantum dot by its density matrix $\hat{\rho}_N(t)$, which is resolved with respect to the number of transferred particles, so that $P_N(t) = \text{Tr}[\hat{\rho}_N(t)]$ is the probability that $N \geq 0$ electrons have tunneled into the reservoir during the time span $[0, t]$ [44,60,61]. The dynamics of the quantum dot is governed by the non-Markovian master equation [51,52,62,63]

$$\frac{d}{dt} \hat{\rho}_N(t) = \sum_{N'} \int_0^t dt' \mathbb{W}_{N-N'}(t-t') \hat{\rho}_{N'}(t') + \hat{\gamma}_N(t), \quad (2)$$

where the kernel \mathbb{W} is obtained by evaluating real-time diagrams on the Keldysh contour up to the desired order in the tunnel coupling Γ [47,64]. The inhomogeneity $\hat{\gamma}_N(t) = \int_{-\infty}^0 dt' \tilde{\mathbb{W}}_N(t, t') \hat{\rho}_{\text{stat}}$ describes correlations that build up between the quantum dot and the lead before the counting

of particles begins at $t = 0$ [6,52,62,68,69], and it is expressed in terms of a modified kernel $\tilde{\mathbb{W}}$, which extends to earlier times. The quantum dot evolves from an arbitrary state in the far past and has reached its stationary state $\hat{\rho}_{\text{stat}}$ well before counting begins.

Solving the non-Markovian master equation is a formidable task. However, we can use an operator-valued generalization of the standard generating function technique in Laplace space by introducing the transformed density matrix $\hat{\rho}_s(z) = \int_0^{\infty} dt e^{-zt} \sum_N s^N \hat{\rho}_N(t)$ together with similar definitions for the kernel and the inhomogeneity. Using these definitions, the solution of Eq. (2) in Laplace space then becomes [6,52]

$$\hat{\rho}_s(z) = \frac{1}{z - \mathbb{W}_s(z)} [\hat{\rho}_{\text{stat}} + \hat{\gamma}_s(z)], \quad (3)$$

which is a powerful formal result that in principle yields the full distribution of transferred charge for any observation time. As we now will show, it also leads to a systematic theory of electron waiting times in nanoscale conductors, which can include both interaction effects and higher-order tunneling processes.

Electron waiting times.—The waiting time distribution is the probability density that two consecutive tunneling events are separated by the time τ [1]. It can be expressed as $w(\tau) = \langle \tau \rangle \partial_{\tau}^2 \Pi(\tau)$, where $\langle \tau \rangle = -1 / [\partial_{\tau} \Pi(0)]$ is the mean waiting time, and $\Pi(\tau) = P_{N=0}(\tau)$ is the idle-time probability that no transfers have occurred during the time span $[0, \tau]$ [4,9]. Importantly, the idle-time probability can be obtained from Eq. (3) using $\Pi(z) = \text{Tr}\{\hat{\rho}_{s=0}(z)\}$. By expanding the kernel around $z = 0$, we can return to the time domain and find [64]

$$w(\tau) = \sum_{m=0}^{\infty} \frac{\langle \tau \rangle}{m!} \partial_z^m \text{Tr}\{\mathcal{J}(z) \mathbb{W}_0^m(z) e^{\mathbb{W}_0(z)\tau} \tilde{\mathcal{J}}(z) \hat{\rho}_{\text{stat}}\}_{z=0}, \quad (4)$$

which is a general result that enables a systematic analysis of how tunneling processes of different orders contribute to the distribution of waiting times. Here, we have defined the jump operators $\mathcal{J}(z) = \mathbb{W}_1(z) - \mathbb{W}_0(z)$ and $\tilde{\mathcal{J}}(z) = \mathcal{J}(z) + \int_0^{\infty} dt z e^{-zt} \int_{-\infty}^0 dt' [\tilde{\mathbb{W}}_1(t, t') - \tilde{\mathbb{W}}_0(t, t')]$, and we note that memory effects are encoded in nonzero derivatives with respect to z . Thus, only the lowest-order term ($m = 0$) remains in the Markovian limit, where we recover the well-known result [1]

$$w_M(\tau) = \langle \tau \rangle \text{Tr}[\mathcal{J} e^{\mathbb{W}_0 \tau} \tilde{\mathcal{J}} \hat{\rho}_{\text{stat}}], \quad (5)$$

since the kernel in that case does not depend on z . Equation (5) is useful to evaluate waiting time distributions in the sequential tunneling regime, where the charge transport can be described by a classical rate equation. By contrast, Eq. (4) allows us to consider lower temperatures, where quantum effects become important.

Tunneling processes.—To evaluate the waiting times between electrons tunneling out of the quantum dot, we expand the kernel order-by-order in the tunnel coupling Γ as $\mathbb{W}_s(z) = \sum_{n=1}^{\infty} \mathbb{W}_s^{(n)}(z)$, with similar expressions for the jump operators and the density matrix. Each term is evaluated using the real-time diagrammatic technique for quantum transport in nanostructures [47–49,64]. Within this framework, we analytically evaluate the waiting time distribution in Eq. (4) up to second order in the coupling and compare our results with the expression in Eq. (5) using sequential tunneling rates [64]. The expansion is controlled by the coupling over the interaction $\hbar\Gamma/U$. In addition, some sequential tunneling rates get exponentially suppressed at low temperatures and are comparable to second-order processes. Higher-order processes, by contrast, can safely be ignored.

Figure 2 shows distributions of waiting times between electrons tunneling out of the quantum dot as indicated by the full arrow in Fig. 1. Our quantum-mechanical calculations are presented with solid lines, while results based on sequential tunneling are indicated with dashed lines. In panel (a), we first focus on waiting time distributions for different electronic temperatures. The state of a single spin-down electron is situated well below the chemical potential of the lead, while the state of a spin-up electron is positioned slightly above the chemical potential. The doubly occupied state has a larger energy.

At low temperatures, we observe marked differences between the quantum-mechanical results that include second-order processes and the classical description based on sequential tunneling only. For the classical description, the quantum dot is likely occupied by a spin-down electron, which energetically is situated below the chemical potential of the lead. It is unlikely that the quantum dot is occupied by a spin-up electron or even doubly occupied. It only

rarely happens that the spin-down electron tunnels out of the quantum dot, which is then quickly refilled by a new spin-down electron, which then tunnels out again much later. Thus, there is a long waiting time between electrons tunneling out of the dot as seen in the corresponding waiting time distribution.

The quantum-mechanical results reveal a completely different physical picture with a large peak at short waiting times (solid blue line). The higher-order processes that are now included lead to a renormalized tunnel coupling as well as an increased tunneling rate that makes it possible for the quantum dot to be doubly occupied [64]. From the doubly occupied state there are two decay paths with very different dynamics. If the spin-up electron tunnels out first, the quantum dot is left with a spin-down electron, and there will be a long waiting time until this electron leaves the quantum dot. On the other hand, if the spin-down electron first tunnels out, the quantum dot is left in an excited state with the spin-up electron, which then quickly leaves the quantum dot, giving rise to the large peak at short waiting times. Thus, higher-order processes lift the blockade of the tunneling events that occurs when only sequential tunneling is considered.

To corroborate this physical picture, it is instructive to investigate the effects of an increased electronic temperature, where the doubly occupied state comes into play because of thermal excitations. In this case, the sequential tunneling processes become dominant, and the classical and quantum descriptions in panel (a) coincide as one would expect at high temperatures. A similar behavior is observed in panel (b), where we gradually shift the level upward, so that the energy of a spin-down electron comes close to the chemical potential, and the blockade is lifted. In this case, the spin-up state and the doubly occupied state are energetically out of reach, and the classical and quantum-mechanical results again agree.

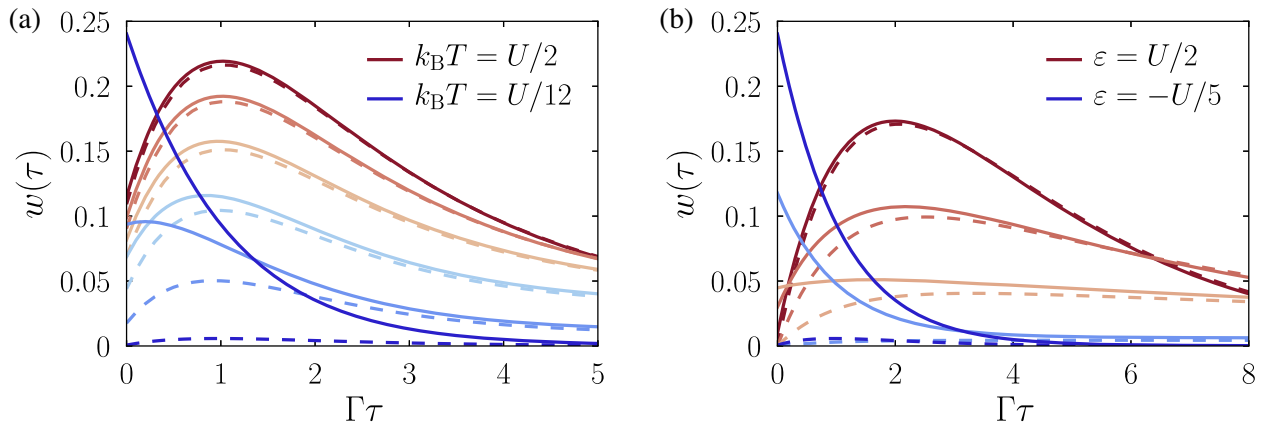


FIG. 2. Distribution of electron waiting times. The quantum-mechanical results are based on Eq. (4) evaluated up to second order in the tunnel coupling $\hbar\Gamma = U/60$ (solid lines), while the results for sequential tunneling using Eq. (5) are evaluated up to first order (dashed lines). (a) The temperature is increased in equidistant steps from $k_B T = U/12$ to $k_B T = U/2$, and we use $\varepsilon = -0.2U$ and $\Delta = 1.1U$ for the level position and the Zeeman splitting as in Fig. 1. (b) The level position is $\varepsilon/U = -0.2, 0.1, 0.3, 0.4$, and 0.5 , and the temperature and Zeeman splitting are $k_B T = U/12$ and $\Delta = 1.1U$.

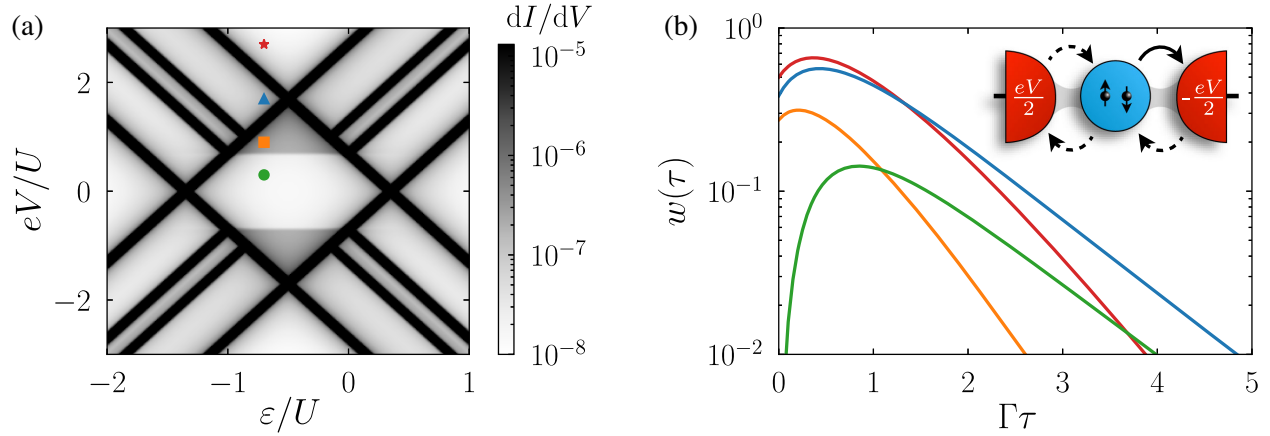


FIG. 3. Waiting time distributions for a transport setup. A bias between two electrodes drives a current through the quantum dot. (a) Differential conductance as a function of the bias voltage and the gate voltage. Parameters are $k_B T = U/200$, $\Delta = 0.7U$, and $\Gamma_S = \Gamma_D \equiv \Gamma = 10^{-5}U/\hbar$. (b) Waiting time distributions corresponding to the points in the conductance map.

Transport setup.—Having understood the importance of higher-order tunneling processes, we go on to consider a transport setup, where a bias voltage between two electrodes drives a current through the quantum dot. We consider the waiting time between electrons that tunnel into the drain electrode and evaluate the waiting time distribution up to second order in the equal couplings to the source and the drain electrodes $\Gamma_S = \Gamma_D$ [64]. Figure 3(a) shows a conductance map as a function of the bias voltage V and the gate voltage, which controls the energy level ϵ of the quantum dot. In the low-bias regime (green circle), Coulomb blockade suppresses the current in the quantum dot, and elastic cotunneling is the dominant transport mechanism. With an increasing voltage, additional transport processes are activated; first inelastic cotunneling (orange square), then sequential tunneling (blue triangle), and eventually all excitations are inside the bias window such that the Coulomb interactions effectively become irrelevant (red star).

Figure 3(b) shows waiting time distributions corresponding to the four points in the conductance map. At low voltages, the waiting time distribution is suppressed at short times, showing that elastic cotunneling processes rarely occur simultaneously. In this case, the quantum dot mainly makes transitions between being occupied by a spin-down electron and being doubly occupied. As the voltage is increased, the suppression at short times is lifted as inelastic cotunneling gets activated, and the quantum dot can now make transitions from the doubly occupied state to being occupied by a spin-up electron, which quickly leaves via the drain electron before the quantum dot is refilled again. These fast events give rise to the peak at short waiting times, while the long waiting times occur when the quantum dot is occupied by a spin-down electron. As the voltage is further increased, sequential tunneling becomes the main transport mechanism, which can be captured by a classical description as we have checked.

Thus, we find that the waiting time distributions can be explained with a classical theory for large bias voltages, while our quantum theory is needed to capture the temporal fluctuations inside the Coulomb blockade region for small voltages and temperatures.

Experimental perspectives.—Finally, we comment on realistic experimental parameters and the perspectives for measuring the waiting time distributions found here. Taking an interacting strength of about $U \simeq 50 \mu\text{eV}$, the tunneling rate in Fig. 1 would be about $\Gamma \simeq 10 \text{ kHz}$, and the temperature around $T \simeq 30 \text{ mK}$. These parameters are compatible with recent measurements of waiting time distributions [41] and the real-time detection of single-electron tunneling involving virtual processes [70]. Moreover, state-of-the-art setups can detect single electrons on a microsecond timescale [71], corresponding to tunneling rates on the order of $\Gamma \simeq 1 \text{ MHz}$ as in Fig. 3 for $U \simeq 50 \mu\text{eV}$ and $T \simeq 10 \text{ mK}$. In addition, we expect further improvements of charge detectors with bandwidths that are 1 or 2 orders of magnitude larger.

Conclusions.—We have investigated the temporal fluctuations of charge tunneling in a quantum dot and developed a real-time diagrammatic theory of electron waiting time distributions that enables a systematic description of interaction effects and higher-order tunneling processes. Our theoretical framework bridges the gap between non-interacting theories of electron waiting times, valid for phase-coherent transport in mesoscopic conductors, and master-equation descriptions, which apply to sequential tunneling in nanostructures with strong interactions. We have found that our quantum-mechanical theory captures higher-order tunneling processes at low temperatures, which are not included in a classical description, and which dramatically affect the distribution of waiting times, for example by allowing fast tunneling processes inside the Coulomb blockade region of a quantum dot. Our work paves the way for future investigations of waiting time distributions

in systems with strong correlations, for example close to a Kondo resonance [72,73] or in a Luttinger liquid [74,75].

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