

to the line width  $\Gamma$  of the single level state. The effect of Coulomb interaction is manifest already in lowest order in the tunnel coupling, introducing a dependence on the level position of the relaxation times. We find that charge and spin have *independent* dynamics with a charge relaxation time that is reduced and spin relaxation time that is enhanced by interaction. Higher-order tunneling contributions lead to a difference between the  $RC$  time extracted from the ac admittance and the relaxation time extracted from the exponential decay after a fast switch. This is in contrast to the behavior of a classical  $RC$  circuit and signals that care must be taken in extracting the characteristic time-scales of the system.

## II. MODEL AND FORMALISM

We consider a quantum dot attached to a single lead by a tunneling contact, cf. Fig. 1, described by the Hamiltonian  $H = H_{\text{dot}} + H_{\text{tunnel}} + H_{\text{lead}}$ . The single-particle level spacing in the dot is assumed to be larger than any other energy scale (temperature, Coulomb interaction, gate voltage and the highest frequency in the time-dependent signals) such that only one energy level is accessible. The dot can be described by the single-level Anderson model

$$H_{\text{dot}} = \sum_{\sigma=\uparrow,\downarrow} \epsilon(t) d_{\sigma}^{\dagger} d_{\sigma} + U n_{\uparrow} n_{\downarrow}, \quad (1)$$

with  $d_{\sigma}^{\dagger} (d_{\sigma})$  being the creation (annihilation) operator for an electron with spin  $\sigma$  on the dot, and  $n_{\sigma} = d_{\sigma}^{\dagger} d_{\sigma}$  the corresponding number operator. The level position is assumed to depend on time via a gate potential. The onsite repulsion  $U$  describes the energy cost for double occupation and stems from Coulomb interaction (within the constant interaction model<sup>12</sup>). Tunneling of electrons between the dot and the lead is taken into account by  $H_{\text{tunnel}} = \sum_{k,\sigma} V c_{k,\sigma}^{\dagger} d_{\sigma} + \text{h.c.}$ , where we assume a momentum-independent tunnel matrix element  $V$  and define the creation (annihilation) operators  $c_{k,\sigma}^{\dagger} (c_{k,\sigma})$  for electrons with spin  $\sigma$  and momentum  $k$  in the lead. The lead Hamiltonian is given by  $H_{\text{lead}} = \sum_{k,\sigma} \epsilon_k c_{k,\sigma}^{\dagger} c_{k,\sigma}$ . We assume a constant density of states  $\rho$  in the leads and define the tunnel coupling strength  $\Gamma$  as  $\Gamma = 2\pi\rho|V|^2$ . Since we are not interested in the dynamics of the non-interacting leads' degrees of freedom, we trace them out obtaining an effective description of the dot in terms of its reduced density matrix. We choose as basis of the four-dimensional Hilbert space of the reduced system the eigenstates,  $|\chi\rangle$ , of the isolated dot Hamiltonian  $H_{\text{dot}}$ :  $|0\rangle$  for an empty dot,  $|\sigma\rangle$  for a singly occupied dot with spin  $\sigma = \uparrow, \downarrow$  and  $|d\rangle$  for a doubly occupied dot. Assuming spin-independent tunneling, we can restrict our interest to the time evolution of the diagonal elements of the reduced density matrix,  $p_{\chi}$ . These probabilities are arranged in the vector  $\mathbf{P} = (p_0, p_{\uparrow}, p_{\downarrow}, p_d)$ , whose time

evolution is given by the generalized master equation

$$\frac{d\mathbf{P}(t)}{dt} = \int_{t_0}^t dt' \mathbf{W}(t, t') \mathbf{P}(t'). \quad (2)$$

The elements  $W_{\chi, \chi'}(t, t')$  of the matrix  $\mathbf{W}(t, t')$  describe transitions from state  $\chi'$  at time  $t'$  to state  $\chi$  at time  $t$ .

## III. FAST SWITCHING

We first consider the time evolution after a fast switching event at  $t = t_0$ , as shown in Fig. 1 (a). The amplitude of the switching is only limited insofar as we assume that only the single level of the dot is accessible, i.e. the amplitude is smaller than the level spacing. The initial value, i.e., the equilibrium distribution before changing the gate voltage, is denoted by  $\mathbf{P}^{\text{in}}$ . In order to identify the relaxation time, we perform the following steps. First, we expand the probability vector  $\mathbf{P}(t')$  on the right-hand side of Eq. (2) around the final time  $t$ , i.e.  $\mathbf{P}(t') = \sum_{n=0}^{\infty} \frac{1}{n!} (t' - t)^n \frac{d^n \mathbf{P}(t')}{dt'^n} |_{t'=t}$ . Second, we realize that for  $t' > t_0$ , the Hamiltonian is time independent, and hence the transition matrix elements depend only on the difference of the time arguments,  $\mathbf{W}(t, t') \rightarrow \mathbf{W}(t - t')$ . Third, we replace the lower integration bound  $t_0$  by  $-\infty$ . This is justified for times  $t$  that are larger than the characteristic time scale over which the kernel  $\mathbf{W}(t)$  decays. The regime of shorter times  $t$  is not the subject of the present work. As a result of these three steps, we obtain

$$\frac{d\mathbf{P}(t)}{dt} = \sum_{n=0}^{\infty} \frac{1}{n!} \partial^n \mathbf{W} \cdot \frac{d^n \mathbf{P}(t)}{dt^n}, \quad (3)$$

introducing the derivatives of the Laplace transforms,  $\partial^n \mathbf{W} := \frac{\partial^n}{\partial z^n} \left[ \int_{-\infty}^t dt' e^{-z(t-t')} \mathbf{W}(t-t') \right]_{z=0+}$ . The formal solution of Eq. (3) can be written as

$$\mathbf{P}(t) = \exp(\mathbf{A}t) \mathbf{P}^{\text{in}}. \quad (4)$$

The matrix  $\mathbf{A}$  always possesses the eigenvalue 0, which guarantees the existence of a stationary probability distribution. The other eigenvalues of  $\mathbf{A}$  define the relaxation rates. In the following, we concentrate on the limit of weak tunnel coupling between quantum dot and lead. This motivates an expansion of  $\mathbf{A} = \mathbf{A}^{(1)} + \mathbf{A}^{(2)} + \dots$  in powers of the tunnel-coupling strength  $\Gamma$ , indicated by the superscript  $(i)$ . Substituting the formal solution for  $\mathbf{P}(t)$ , Eq. (4), into Eq. (3) iteratively yields  $\mathbf{A}^{(1)} = \mathbf{W}^{(1)}$  and  $\mathbf{A}^{(2)} = \mathbf{W}^{(2)} + \partial \mathbf{W}^{(1)} \cdot \mathbf{W}^{(1)}$ . Note that non-Markovian contributions, i.e. contributions proportional to  $\partial^n \mathbf{W}$ , enter only starting from second order in  $\Gamma$ . The kernel  $\mathbf{W}(z)$  can be computed by the real-time diagrammatic approach developed in Ref. 13.

We are interested in the inverse relaxation times of charge  $\gamma_c = 1/\tau_c$  and spin  $\gamma_s = 1/\tau_s$ . They are found as the eigenvalues of  $\mathbf{A}$  related to the left eigenvectors  $\mathbf{v}_c = (0, 1, 1, 2) - \langle n_c \rangle^{eq} (1, 1, 1, 1)$  and  $\mathbf{v}_s = (0, 1, -1, 0)$ ,