Effect of many-body correlations on mesoscopic charge relaxation

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Introduction. For a macroscopic capacitor coupled to a reservoir [Fig. 1(a)], the low-frequency dynamical conductance is simple and determined solely by the geometrical capacitance and the resistance. As the capacitor scales down to nanometers size, the transport properties are significantly modified due to quantum coherence. First of all, the capacitance now depends strongly on the density of states (DOS) as well as the geometrical capacitance. More interestingly, the dynamical resistance becomes quantized, universal, and independent of the transmission through the mesoscopic conductor. The capacitance and resistance are called electrochemical capacitance and charge relaxation resistance, respectively, to be distinguished from the macroscopic counterparts. So far, such a coherent RC circuit has been described with various theoretical methods: the mean-field approach, the spin-polarized configurations, the perturbative treatment, and the semi-classical limit. However, a proper description of the many-body correlation effects is still missing. In this Rapid Communication, we attempt to fill this gap by treating the many-body interaction in a nonperturbative way. We find that the many-body correlations break significantly the universality of the charge relaxation resistance and that the deviation is maximal at certain energy scale $\Gamma^*$, which we interpret as an effective level broadening. Namely, (i) The charge relaxation resistance has peaks at finite frequencies $\Gamma^*/h$, where $\Gamma^*$ is an effective level broadening, and (ii) the zero-frequency charge relaxation resistance deviates from the universal value when the Zeeman splitting is comparable to $\Gamma^*$. This behavior becomes even more prominent in the Kondo regime. The observed features are ascribed to the generation of particle-hole excitations in the contacts accomplished by spin-flip processes in the dot.

We investigate nonperturbatively the charge relaxation resistance and quantum capacitance in a coherent RC circuit in the strong-coupling regime. We find that the many-body correlations break the universality in the charge relaxation resistance: (i) The charge relaxation resistance has peaks at finite frequencies $\Gamma^*/h$, where $\Gamma^*$ is an effective level broadening, and (ii) the zero-frequency charge relaxation resistance deviates from the universal value when the Zeeman splitting is comparable to $\Gamma^*$. This behavior becomes even more prominent in the Kondo regime. The observed features are ascribed to the generation of particle-hole excitations in the contacts accomplished by spin-flip processes in the dot.

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The coherent RC circuit is an interesting and promising electron analog of the on-demand single-photon source, which is an essential part of photon-based quantum information processing. The on-demand single-electron generation has been successfully demonstrated in a recent experiment, not long after the quantized charge relaxation resistance was observed experimentally. Encouraged by the two pioneering experiments and motivated by the potential applications, there are a number of experimental studies of coherent RC circuits.

Model and Methods. We consider a nanoscale capacitor (quantum dot) coupled to a single reservoir. A weak time-dependent external gate voltage $V(t) = V_0 \cos \omega t$ is applied on the quantum dot. In such a coherent RC circuit, the ac transport is highly sensitive to the internal distribution of charges and potentials, which needs to be calculated in a self-consistently manner to ensure the gauge invariance and current conservation. To achieve this accurately, we use the numerical renormalization group (NRG) method and calculate the charge relaxation resistance $R_q(\omega)$ and the electrochemical capacitance $C_q(\omega)$. Note that $C_q(\omega)$ is related to the geometrical capacitance $C$ and quantum capacitance $C_q(\omega)$ by $C_q = (C^{-1} + C_q^{-1})^{-1}$.

The essential features of the system in the presence of correlations can be well captured in the Anderson model, where the dot is described by an interacting single level. The Hamiltonian $\mathcal{H} = \mathcal{H}_L + \mathcal{H}_D + \mathcal{H}_T$ consists of the lead part

$$\mathcal{H}_L = \sum_{k\mu} (\epsilon_k + eV(t)) c_{k\mu}^\dagger c_{k\mu},$$

the dot part

$$\mathcal{H}_D = \sum_{\mu} (\epsilon_\mu + eU(t)) d^\dagger_\mu d_\mu + 2E\bar{d}^\dagger_* d_\dagger_* ,$$

and the tunneling part

$$\mathcal{H}_T = \sum_{\mu} T c_{k\mu}^\dagger d_\mu + h.c.$$
and the tunneling part

$$H_T = \sum_{k\mu} [\hbar d_{k\mu}^\dagger c_{k\mu} + (\text{H.c.})].$$

The operator $c_{k\mu}$ describes the noninteracting conduction electrons with energy $\epsilon_k$ (measured with respect to the Fermi energy $\epsilon_F = 0$) and spin $\mu$ in the single-channel reservoir and $d_{k\mu}$ the interacting electron on the dot. $E_C = e^2/2C$ is the Coulomb charging energy and $\epsilon_{d\mu} = \epsilon_{d\mu} - \mu \Delta_x/2$ are the dot levels with Zeeman splitting $\Delta_x$ on the dot. $t_k$ is the tunneling amplitude of electrons between the reservoir and the dot. For simplicity, we assume $t_k = t$ and characterize the dot-lead hybridization by $\Gamma = \pi \rho_0 |t|^2$ ($\rho_0$ is the contact DOS at the Fermi energy).

The time-dependent voltage $V(t)$ induces the polarization charges $N_U(t)$ between the dot and the gate, in turn, leads to the time-dependent potential $U(t) = [e|N_U(t)/C$ inside the dot. Consequently, the applied voltage not only generates a current $I(t)$ between the lead and the dot, but also induces a dot-gate displacement current $I_d(t) = e(dN_U/dt) = -C(dU/dt)$. Charge conservation requires $I(t) + I_d(t) = 0$. Assuming that the gate-invariant potential, $V(t) - U(t)$, is sufficiently small, the linear response theory leads to the relation, $I(\omega) = g(\omega)[V(\omega) - U(\omega)]$, where $g(\omega) = (ie/\hbar) \langle \Delta(t)/\Delta(\omega) \rangle \Theta(t)$ is the equilibrium correlation function between the occupation operator $\Delta = \sum_{k\mu} d_{k\mu}^\dagger d_{k\mu}$ and the current operator $\Delta = e(dN_U/dt)$. Note that the current-density correlation function $g(\omega)$ is directly related to the charge susceptibility $\chi_c(t) = -i \langle [\Delta(t), \Delta(\omega)] \Theta(t) \rangle$, which is preferable for numerical computation, via the relation $g(\omega) = i/o(\epsilon/h) \chi_c(\omega)$. Then, with the help of $I(\omega) = -I_d(\omega) = -i/oCU(\omega)$, the dot-lead impedance $Z(\omega) = V(\omega)/I(\omega)$, which is experimentally accessible, is given by $Z(\omega) = 1/(-i/oC) + 1/g(\omega)$. The relaxation resistance and the quantum capacitance are then expressed in terms of the charge susceptibility as

$$R_q(\omega) = \text{Re} \left[ \frac{1}{2\pi io\chi_c(\omega)} \right], \quad \frac{e^2}{h} C_q(\omega) = \text{Im} \left[ \frac{1}{2\pi i \chi_c(\omega)} \right].$$

We first calculate the imaginary part of the susceptibility using the NRG method, and then its real part via the Kramers-Kronig relation. Note that the NRG results for the finite-frequency linear response in the Kondo regime are known to be reliable as long as the time-dependent voltage $V(t)$ is weak enough. We focus on the zero-temperature case and use the contact bandwidth $D$ as the energy unit. We set $k_B = 1$ hereafter unless specified.

No Zeeman splitting, $\Delta_x = 0$. Figure 2(a) shows the zero-frequency limit values of $R_q$ and $C_q$ versus $\epsilon_d$. (b) Typical spectral structure of the real and imaginary parts of $\chi$.

1. The NRG results show the quantization of charge relaxation even in the Kondo regime where many-body correlations are effective. The observed small deviations from the exact value $h/4e^2$, persisting even in the noninteracting case, are attributed to the finiteness of the contact bandwidth $D$, which introduces a frequency-dependent real part into the dot self-energy. The universal value can be restored by setting all the relevant energy scales to be much smaller than $D$.

2. Second, the quantum capacitance, $C_q$ exhibits two remarkable considerations: (i) at the degenerate points, $\epsilon_d \sim \epsilon_F$ and $\epsilon_d + 2E_C \sim \epsilon_F$, $C_q$ shows two pronounced peaks [see Fig. 2(a)], which is consistent with the known understanding that $C_q$ is proportional to the dot DOS $\rho_{dd}(\epsilon_F)$. (ii) $C_q$ remains quite small in the Kondo regime although the DOS at the Kondo resonant level pinned at the Fermi level achieves its maximum value. It implies that the Kondo resonant level, even though it can open a tunneling channel, is not a real level which can hold real charges and cannot contribute to the capacitance. Hence, in the presence of many-body correlations $C_q$ is not, always, directly related to the DOS.

The frequency dependence of $R_q(\omega)$ is shown in Fig. 2. One can clearly see that $R_q(\omega)$ exhibits two peaks at $\omega = \pm \pi/\hbar$, see Figs. 2(e) and 2(d) for two dot level positions, $\epsilon_d = -0.1$ (fluctuating valence regime) and $\epsilon_d = -0.3$ (Kondo regime). One can interpret this structure in terms of the p-h excitations and the relation in Eq. (4). $\text{Im}[\chi_c(\omega)]$ reflects the coupling between the ground state and p-h excitations via the dot-lead hybridization. Since the spectral density of multiple p-h excitations increases with energy, $\text{Im}[\chi_c]$ would grow monotonically with $|\omega|$. However, a finite $D$ puts an upper limit to the energy for p-h excitations $|\omega| \lesssim O(D)$ resulting in a
nonmonotonic behavior for $\text{Im}[\chi_c]$, see Fig. 2(b). Moreover, $\text{Im}[\chi_c]$ has two kinks at $|\omega| = \min(|\epsilon_d|, |\epsilon_d + 2E_C|)$ since beyond this frequency p-h excitations accompanied with a charge excitation contributes to $\text{Im}[\chi_c]$ as well. An interesting structure appears in $\text{Im}[\chi_c]$ near $\omega = 0$ [not seen in Fig. 2(b) due to logarithmically small energy scale]. Close to $\omega = 0$, $\text{Im}[\chi_c]$ depends linearly with $\omega$, mainly due to single p-h excitations. However, $\text{Im}[\chi_c]$ departs from the linearity when $\omega$ becomes of the order of the effective hybridization $\Gamma^* (= T_K$ in the Kondo regime). Here the effective level broadening $\Gamma^*(T_K)$ is extracted from the width of the resonance close to $\omega = 0$ in $\rho_{\text{dot}}$. Besides, we found that the slope of $\text{Im}[\chi_c(\omega)]$ is the largest at $\omega = \Gamma^*/h$, and hence leading to the peak structure in $R_q(\omega)$ as seen in Figs. 2(c) and 2(d).

In the Kondo regime $R_q(\omega)$ becomes several orders of magnitude larger than the universal value, see Fig. 2(d). Note that such peak structure in $R_q(\omega)$ is absent in the noninteracting case and thus in the usual Fermi liquid picture of the Kondo effect. For a noninteracting system, $R_q(\omega)$ increases monotonically with increasing $|\omega|$, and the only characteristic energy scale is $\epsilon_d$. Hence, the peaks seen in Figs. 2(c) and 2(d) are a genuine many-body effect.

Finite Zeeman splitting, $\Delta_Z \neq 0$. The spin-split case ($\Delta_Z > 0$) in the presence of external magnetic fields is illustrated in Fig. 3. In contrast to the spin-degenerate case, $R_q(\omega \to 0)$ is no longer fixed to the universal value. Interestingly, $R_q(\omega \to 0)$ versus $\Delta_Z$ exhibits a peak structure reaching magnitude larger than the universal value, see Fig. 2(d). For $\Delta_Z$ sufficiently larger than $\Gamma^*$, the peak is exactly located at $\Delta_Z = \Gamma^*(T_K)$ for the noninteracting case ($\Delta_Z = 0$). With increasing $\Delta_Z$, the peak height increases as the effective hybridization decreases so it is the highest in the Kondo regime. In the meanwhile, $C_q(\omega \to 0)$ remains rather constant, except at the resonant tunneling regime ($\epsilon_d \approx 0$) where it displays a small peak, see Fig. 3(b). The evolution of the spectral distribution of $R_q(\omega)$ with $\Delta_Z$ is displayed in Fig. 3(c) for $\epsilon_d = -0.1$ (Kondo regime). As $\Delta_Z$ increases, the low-frequency part of $R_q(\omega)$ for $|\omega| < k_BT_K/h$ keeps going up until $\Delta_Z$ reaches $k_BT_K$; the side peaks are merged into the central peak. With increasing $\Delta_Z$ further, the central peak diminishes gradually and, eventually, together with the side peaks located at $\omega = \pm k_BT_K/h$, disappear completely. We have observed a similar transition of $R_q(\omega)$ with $\Delta_Z$ in the resonant tunneling regime ($\epsilon_d = -0.05, 0$) except that the variation of the central part is smaller.

To clarify the role of the Coulomb interaction, we compare the zero-frequency values of $R_q$ for different values of the Coulomb interaction in the resonant tunneling regime, see Fig. 3(d). In the noninteracting case ($E_C = 0$), there is no peak at all, with $R_q(\omega \to 0) = \text{const} = h/4e^2$. However, as soon as the charging energy $2E_C$ becomes comparable to $\Gamma^* \sim \Gamma$, a peak starts to rise up and manifests itself for $2E_C \gg \Gamma$. It implies that the existence of the peak structure observed in Fig. 3(d) definitely has its origin in the Coulomb interaction.

Discussion. Now we have two questions to be answered:

(1) How do Coulomb interaction affect the relaxation resistance far beyond the universal value, $h/4e^2$ and (2) Why does it take place noticeably at $\omega = \pm \Gamma^*(T_K)/h$ in the fluctuating valence (Kondo) regime for $\Delta_Z = 0$ or at $\omega = 0$ for $\Delta_Z$.

\[ \Delta_Z = \Gamma^*(T_K)? \] The charge relaxation resistance is attributed to p-h pair generation in the conduction band as shown in Fig. 1. Such processes are put in action when the dot-lead tunneling is switched on. The tunneling, in turn, hybridizes dot and conduction band electrons, resulting in the lowering of the ground state by the effective binding energy $\Gamma^*(T_K)$ in the Kondo regime. It means that the p-h generation starts when the energy supplied by the source is larger than $\Gamma^*$. This argument explains the observed peak in $R_q(\omega)$ at $\omega = \pm \Gamma^*/h$ in the absence of the Zeeman splitting. In the presence of finite but small Zeeman splitting, the energy cost can be compensated by the Zeeman splitting. The energy of the p-h pair excitation states shown in Fig. 1(c) are now lowered by $\Delta_Z$ compared to the states in Fig. 1(b), and when $\Delta_Z \approx \Gamma^*$ they become almost degenerate with the ground state, allowing p-h pair generation with negligible energy cost. Hence, $R_q(\omega)$ exhibits a single peak at $\omega = 0$ when $\Delta_Z \approx \Gamma^*$. This argument works solely when $\Delta_Z \lesssim \Gamma^*$ in which the ground state is not yet completely polarized and there exists a finite coupling among spin-down dot states and spin-up dot states accompanying with a p-h pair generation in the reservoir, see Fig. 1. The importance of the spin flip in the boosting of the relaxation resistance also explains why $R_q(\omega)$ can reach higher values in the Kondo regime. The Kondo ground state is built from spin fluctuations due to spin-flip scattering among the localized dot electron and the delocalized electrons in the reservoirs, thus spin-flip processes have large amplitudes in its wave function. Hence the processes as shown in Fig. 1(c) can happen more frequently, leading to a large $R_q$. Similarly, the answer for the first question is now ready. The spectral weight for the charge correlation function is proportional to $|\langle \alpha|N|\gamma\rangle|^2$, where $gs$ and $\alpha$ represent the ground and the excited states. In the second-order perturbation theory, the weight corresponding to the processes in Fig. 1(c) is given by

\[ |\langle \alpha|N|\gamma\rangle|^2 = r^4 \frac{2}{E_{\mu}E_{\bar{\mu}}} \sum_{\mu} \frac{\mu}{\Delta_Z} \left( \frac{1}{E_{\mu}} + \frac{1}{E_{\bar{\mu}}} \right)^2. \]
in the $\omega \to 0$ limit with $E_Z = 2E_C + \epsilon_\mu$. Interestingly, this weight vanishes completely for $E_C = 0$ for any value of $\Delta_Z$. Thus, for the noninteracting case there exists no p-h pair generation process accompanying spin flip in the dot, and no boosting of the relaxation resistance can happen. For finite values of $E_C$, the weight is finite [see Eq. (5)] and for $E_C \to \infty$, it becomes $t^\dagger/(\epsilon_\mu \epsilon_d)^2$. This value can be substantial depending on the level position. Note that this analysis is not correct quantitatively because high-order events should be considerably involved in the observed phenomena. The observed boosting of $R_q$ at $\Delta_Z \sim \Gamma^\ast$ indicates that the perturbation in the dot-lead tunneling or $\Gamma$ is risky. A more general theoretical analysis that treats $\Delta_Z$ and $\Gamma$ on equal footing could provide more quantitatively reliable interpretation. Besides, this perturbative analysis does not work in the Kondo regime where the strong dot-lead coupling is important. One may want to study the Kondo regime by an effective single-particle Hamiltonian with a dot level at the Fermi energy with the effective hybridization $T_K$. However, this picture is only suitable in the Fermi-liquid regime in which p-h excitations accomplished by spin-flip events in the dot are not allowed. Besides, this effective model predicts an enhanced mesoscopic capacitance $C_q(\omega \to 0)$ due to the presence of the resonant level at the Fermi level. As noted before, however, the Kondo resonant level cannot contribute to the charging of real charges.

Conclusion. In closing, we have investigated the dynamics of a many-body quantum capacitor, focusing on the strong-coupling regime. We observe the breakdown of the Fermi-liquid features such as the quantized low-frequency relaxation resistance. We find that $R_q(\omega)$ shows peaks at $\omega = \pm \Gamma^\ast(T_K)/h$ for $\Delta_Z = 0$ and that $R(\omega \to 0)$ is enhanced over the quantized value when $\Delta_Z = \Gamma^\ast(T_K)$. It can be understood by the fact that a part of $R_q(\omega)$ is built by dot-lead tunneling events connecting p-h excitations in the reservoirs with spin-flip processes in the dot. The boosted low-frequency (and high-frequency) relaxation resistance, which is much stronger in the Kondo regime, is expected to be experimentally observable by considering the current advances in RC-circuit experiments.8,9

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18For better accuracy, we adopt Hofstetter’s algorithm (Ref. 25) accompanied with the improved z-averaging method (Ref. 26).
21Finite-width-band self-energy is given by $\text{Re}[\Sigma(\omega)] = -(\Gamma/2\pi) \ln(D - \hbar \omega)/(D + \hbar \omega)$. Its presence slightly violates the Fermi-liquid assumptions and, consequently, the KS relation is not exactly fulfilled so that the universal value is not recovered: Since $\text{Re}[\Sigma(\omega)]$ increases with $\omega$, the deviations are larger as the resonant level becomes far from $\epsilon_F = 0$.
24The analytical expression of $R_q(\omega)$ in the wide band limit of the noninteracting system is given by Ref. 27: $R_q(\omega) = (\hbar/e^2)|[G(\omega) + (\hbar \omega/H)(F(\omega))][G(\omega)^2 + F(\omega)^2]|^2$ with $G(\omega) = \ln[(\epsilon_d + \hbar \omega)^2 + \Gamma^2][h \delta - \hbar \omega]^2 + \Gamma^2] and F(\omega) = 2[tan^{-1}[\Gamma/(\epsilon_d - \hbar \omega)] - tan^{-1}[\Gamma/(\epsilon_d + \hbar \omega)]].$