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**To cite this version:**

Minchul Lee, Rosa Lopez, Mahn-Soo Choi, Thibaut Jonckheere, Thierry Martin. Many-body Correlation Effect on Mesoscopic Charge Relaxation. Physical Review B: Condensed Matter and Materials Physics (1998-2015), 2011, 83, pp.201304. 10.1103/PhysRevB.83.201304 . hal-00735474

**HAL Id: hal-00735474**

**https://hal.science/hal-00735474v1**

Submitted on 9 Jun 2023

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arXiv:1101.0468v1 [cond-mat.mes-hall] 3 Jan 2011

# Many-body Correlation Effect on Mesoscopic Charge Relaxation

Minchul Lee,[1] Rosa López,[2,3] Mahn-Soo Choi,[4] Thibaut Jonckheere,[5] and Thierry Martin[5,6]

[1]*Department of Applied Physics, College of Applied Science, Kyung Hee University, Yongin 446-701, Korea*
[2]*Departament de Física, Universitat de les Illes Balears, E-07122 Palma de Mallorca, Spain*
[3]*Institut de Física Interdisciplinar i de Sistemes Complexos IFISC (CSIC-UIB), E-07122 Palma de Mallorca, Spain*
[4]*Department of Physics, Korea University, Seoul 136-701, Korea*
[5]*Centre de Physique Théorique, UMR6207, Case 907, Luminy, 13288 Marseille Cedex 9, France*
[6]*Université de la Méditerranée, 13288 Marseille Cedex 9, France*

We investigate in a nonperturbative way the dynamics of a correlated quantum capacitor. We find that the many-body correlations do not disturb the universal low-frequency relaxation resistance per channel, $R_q(\omega = 0) = h/4e^2$ ensured by the Korringa-Shiba rule whereas the interpretation of the quantum capacitance $C_q$ in terms of the density of states fails when strong correlations are present. The AC resistance $R_q(\omega)$ shows huge peaks (with values larger than $h/4e^2$) at $\hbar\omega \approx \pm\Gamma^*$, where $\Gamma^*$ is the renormalized level broadening. These peaks are merged to a single one at $\omega = 0$ when a finite Zeeman field is applied comparable to $\Gamma^*$. The observed features of $R_q$, being most evident in the Kondo regime, are attributed to the generation of particle-hole excitations in the contacts accomplished by spin-flip processes in the dot.

PACS numbers: 73.63.-b, 74.50.+r, 72.15.Qm, 73.63.Kv

*Introduction.—* Optoelectronic devices such as light sources emitting single photons on demand are of an enormous interest in quantum information. Recently, its solid-state analogue, a *quantum capacitor* (QC), was created by Fève *et al.* [1], in which a quantum dot (QD) was coupled to a single reservoir via a quantum point contact. Fast time-controlled variations of fractions of nanoseconds on the dot gate potential produced the single-electron source of emitting electrons in a desirable quantum state. Previously, Gabelli *et al.* [2] showed that the QC could act as a RC circuit with a quantized resistance as predicted in a series of seminal works [3–5]. The experimental interest in the AC properties of mesoscopic conductors [6–11] has been revived recently due to the experimental confirmation of the quantization of both the AC current [1] and the quantum resistance [2]. In a coherent conductor the AC transport is highly sensitive to the internal distribution of charges and potentials that need to be calculated in a self-consistently manner to ensure a gauge invariant and current conserving description. For a macroscopic capacitor the low-frequency dynamical conductance depends on two elements: the geometrical capacitance $C$, and the resistance $R$. Classically, the electric field on the surface of the metallic plates is completely screened and $C$ is characterized solely via Coulomb forces. However, Büttiker *et al.* pointed out that for a coherent nanoscale system that rule statement is not longer valid, since electric fields penetrate at distances of the order of the Thomas-Fermi screening length [3, 4, 12, 13]. Consequently, the capacitance for a coherent conductor, termed as *electrochemical capacitance*, depends on the geometry, on its physical properties and particularly on its density of states (DOS) through the quantum capacitance $C_q$. Even more surprisingly, the resistance becomes quantized independently of the value of the transmission through the mesoscopic conductor [14–17]. To distinguish it from the DC resistance, $R_q$ is called *charge relaxation resistance*, and together with $C_\mu = (C^{-1} + C_q^{-1})^{-1}$ defines the RC time for a QC, that is, the charge relaxation time upon the action of time-dependent potential.

So far several aspects of AC conductance for RC quantum circuits have been addressed [18–25] focusing mostly on the spin polarized case. Therefore the understanding on the effect of many-body correlations in RC circuits is still missing. This Letter attempts to fill this gap, offering a physical picture of the influence of many-body interactions on the dynamics of QCs. We consider a QC formed by an ultrasmall QD and calculate its $C_q$ and $R_q$ by using their relations with the charge susceptibility in the linear response regime. We perform a thorough study by tuning the QD from the resonant tunneling regime to the Kondo regime. In the Kondo regime virtual tunneling transitions between the dot and the reservoir flip efficiently the dot spin resulting in a many-body singlet spin state with binding energy given by the Kondo energy $k_B T_K$. First, we find that the Korringa-Shiba (KS) relation [26], valid only in the Fermi-liquid regime, ensures the quantization of $R_q$ even in the Kondo regime as long as the frequency is low enough, $\hbar\omega \ll k_B T_K$. However, it is pointed out that the relation of $C_q$ with the localized DOS should be revised when the charge dynamics is frozen due to Kondo cor-

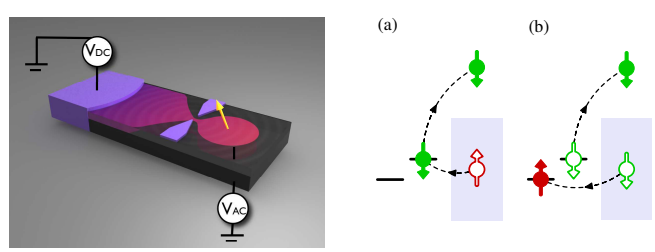


FIG. 1: (color online) (LEFT) Illustration of a quantum capacitor and (RIGHT) second-order tunneling processes that generate a single particle-hole pair in the conduction band without [(a)] and with [(b)] a spin flip in the dot. Filled and empty arrows in the contact indicates particles and holes, respectively. Here we assume $\Delta_Z > 0$.

relations. More importantly, the AC resistance $R_q(\omega)$ displays peaks at $\hbar\omega \sim \pm\Gamma^*$, not present in a mean-field description for the Coulomb interaction. Here $\Gamma^*$ is the renormalized level broadening of the major tunneling dot level and $\Gamma^* = k_B T_K$ in the Kondo regime. Third, a departure from the universal value for $R_q(\omega = 0)$ is achieved when an applied Zeeman field $\Delta_Z$ is comparable to $\Gamma^*$. The enhancement of $R_q$ at zero and finite frequencies is attributed to the creation of particle-hole ($p$-$h$) excitations accompanying a spin-flip in the dot due to the strong Coulomb interaction, see Fig. 1(b).

*Model.—* The essential features of a mesoscopic RC circuit in the presence of correlations can be well captured in the Anderson model where an interacting single-level QD is coupled to a single-channel electron reservoir subject to a weak time-dependent voltage $V(t) = V_{\rm ac}\cos\omega t$. The Hamiltonian of this system is $\mathcal{H} = \mathcal{H}_{\rm L} + \mathcal{H}_{\rm D} + \mathcal{H}_{\rm T}$. The lead part, $\mathcal{H}_{\rm L} = \sum_{\mathbf{k}\mu}[\epsilon_{\mathbf{k}} + eV(t)]c^\dagger_{\mathbf{k}\mu}c_{\mathbf{k}\mu}$, describes the noninteracting conduction electrons with energy $\epsilon_{\mathbf{k}}$ (measured with respect to the Fermi energy $\epsilon_F = 0$) and spin $\mu$ in the reservoir, and the tunneling of electrons between the reservoir and the dot is modeled by $\mathcal{H}_{\rm T} = \sum_{\mathbf{k}\mu}\left[t_{\mathbf{k}} d^\dagger_\mu c_{\mathbf{k}\mu} + (h.c.)\right]$ in terms of energy-independent tunneling matrix element, $t_{\mathbf{k}} = t$. The hybridization between the dot and the lead is characterized by a tunneling amplitude $\Gamma = \pi\rho_0|t|^2$ ($\rho_0$ is the contact DOS at the Fermi energy). The dot Hamiltonian reads $\mathcal{H}_{\rm D} = \sum_\mu[\epsilon_\mu + eV(t)]\,n_\mu + 2E_{\rm C}n_\uparrow n_\downarrow$, where $n_\mu = d^\dagger_\mu d_\mu$ is the dot occupation operator and $E_{\rm C} = e^2/2C$ is the Coulomb charging energy. The orbital level $\epsilon_\mu = \epsilon_d - \mu\Delta_Z/2$ is spin-dependent due to a Zeeman energy $\Delta_Z$. The time-dependent voltage $V(t)$ induces the polarization charges $N_U(t)$ between the dot and the gate, which 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 perturbation $V(t) - U(t)$ is sufficiently small, the linear response theory leads to the relation, $I(\omega) = g(\omega)(V(\omega) - U(\omega))$, where $g(t) = (ie/\hbar)\langle[\mathcal{I}(t), \mathcal{N}]\rangle\,\Theta(t)$ is the equilibrium correlation function between the occupation operator $\mathcal{N} = \sum_\mu n_\mu$ and the current operator $\mathcal{I} = e(d\mathcal{N}/dt)$. Note that the current-density correlation function $g(\omega)$ is directly related to the charge susceptibility $\chi_c(t) = -i\langle[\mathcal{N}(t), \mathcal{N}]\rangle\,\Theta(t)$, which is preferable for numerical computation, via the relation $g(\omega) = i\omega(e^2/\hbar)\chi_c(\omega)$. Then, with the help of $I(\omega) = -I_d(\omega) = -i\omega CU(\omega)$, the dot-lead impedance $Z(\omega) = V(\omega)/I(\omega)$, which is experimentally accessible, is given by $Z(\omega) = 1/(-i\omega C) + 1/g(\omega)$. The relaxation resistance and the quantum capacitance are then expressed in terms of the charge susceptibility as

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

The numerical normalization group (NRG) [27, 28] treats the Coulomb interaction in a nonperturbative way being the most adequate method for computing the charge susceptibility [29].

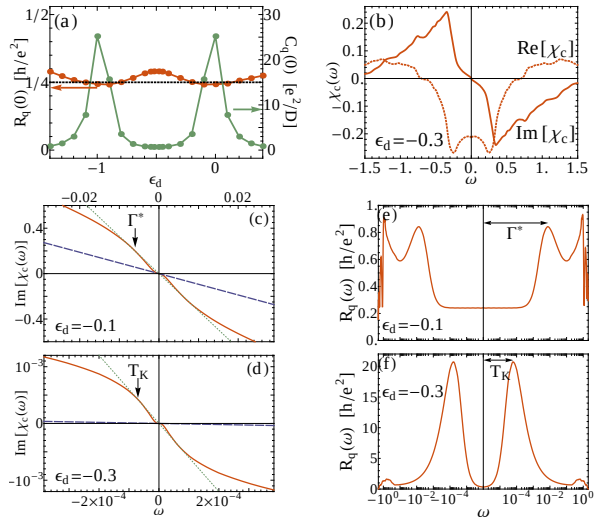


FIG. 2: (color online) (a) Zero-frequency limits of $R_q$ and $C_q$ versus $\epsilon_d$. (b) Typical spectral structure of the real and imaginary parts of the charge susceptibility $\chi_c$. (c,d) Enlarged views of $\mathrm{Im}[\chi_c]$ near $\omega = 0$ for two values of $\epsilon_d$. The dashed (blue) and dotted (green) lines are tangent lines at infinitesimally small $\omega$ and at points (indicated by arrow) where the slope is maximal, respectively. (e, f) $R_q(\omega)$ in the logarithmic scale. Here $\Gamma = 0.04$, and $E_{\rm C} = 0.5$.

While the imaginary part of the susceptibility is obtained by the NRG procedure its real part is calculated 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 perturbation is weak enough [30]. We focus on the zero-temperature case and use the contact bandwidth $D$ as the energy unit. We set $k_B = 1$ hereafter.

*No Zeeman splitting, $\Delta_Z = 0$.—* Figure 2(a) shows our main results for $R_q$ and $C_q$ for the spin-degenerate case. First, the zero-frequency limit of the relaxation resistance, $R_q(\omega \to 0)$ is always close to the universal value $h/4e^2$, regardless of values of $\epsilon_d$ and $E_{\rm C}$. This value can be interpreted as the composite resistance of two parallel resistors of resistance $h/2e^2$, the well-known universal resistance per channel [3–5, 20]. 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 , $\mathrm{Re}[\Sigma(\omega)] = -(\Gamma/2\pi)\ln|(D - \hbar\omega)/(D + \hbar\omega)|$. 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 $\mathrm{Re}[\Sigma(\omega)]$ increases with $\omega$ in magnitude, the deviations are larger as the resonant level becomes far from $\epsilon_F = 0$. The universal value can be restored by setting all the relevant energy scales to be much smaller than $D$ [31]. Second, the quantum capacitance, $C_q$ exhibits two remarkable considerations: *(i)* at the degenerate points, $\epsilon_d \sim \epsilon_F$ and $\epsilon_d + 2E_{\rm C} \sim \epsilon_F$, $C_q$ shows two pronounced peaks [see Fig. 2(a)], which is consistent with

the known understanding that $C_q$ is proportional to the dot DOS $\rho_{\rm dot}(\epsilon_F)$ [3, 4, 12], and *(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 $R_q(\omega)$ and $C_q(\omega)$ is analyzed in Fig. 2 where the the behavior of the real and imaginary part of the charge susceptibility $\chi_c(\omega)$ is shown, see Fig. 2(b) for $\epsilon_d = -0.3$. The imaginary part of $\chi_c(\omega)$ reflects the coupling between the ground state and $p$-$h$ excitations due to the dot-lead hybridization. Since the spectral density of multiple $p$-$h$ excitations increases with energy, $|{\rm Im}[\chi_c]|$ would grow monotonically with $|\omega|$. However, a finite $D$ puts an upper limit to the energy for $p$-$h$ excitations [$|\omega| \gtrsim \mathcal{O}(D)$] resulting in the observed nonmonotonic behavior for ${\rm Im}[\chi_c]$. Moreover, ${\rm Im}[\chi_c]$ has two kinks at $|\omega| = \min(|\epsilon_d|, |\epsilon_d + 2E_{\rm C}|)$ since beyond this frequency $p$-$h$ excitations accompanied with a charge excitation contributes to ${\rm Im}[\chi_c]$ as well. An interesting structure appears in ${\rm Im}[\chi_c]$ near $\omega = 0$, see Figs. 2(c) and (d) for two dot level positions, $\epsilon_d = -0.1$ (corresponding to the fluctuating valence regime) and $\epsilon_d = -0.3$ (Kondo regime). Close to $\omega = 0$, ${\rm Im}[\chi_c]$ depends linearly with $\omega$, mainly due to single $p$-$h$ excitations [see tangent (dashed) lines in Figs. 2(c) and (d)]. However, ${\rm Im}[\chi_c]$ departs from linearity when $\omega$ becomes of the order of the effective hybridization $\Gamma^*$ ($= T_K$ in the Kondo regime). The renormalized hybridization $\Gamma^*(T_K)$ is extracted from the width of the resonance close to(at) $\epsilon_F$ in $\rho_{\rm dot}$. Besides, we found that the slope of ${\rm Im}[\chi_c(\omega)]/\omega$ is the largest at $\hbar\omega = \Gamma^*$, while the change in ${\rm Re}[\chi_c(\omega)]$ is marginal. As a consequence, $R_q(\omega)$ (see Eq. (1)) exhibits two side peaks at $\hbar\omega = \pm\Gamma^*$ as shown in Figs. 2(e) and (f). Notice that in the Kondo regime $R_q(\omega)$ becomes much larger in order of magnitude than the universal value, see Fig. 2(f). Remarkably, such peak structure in $R_q(\omega)$ is absent in the noninteracting case. For a noninteracting system, the analytical expression of $R_q(\omega)$ in the wide-band limit is given by [32] $R_q^{\rm non}(\omega) = (h/e^2)\{[G(\omega) + (\hbar\omega/\Gamma)F(\omega)]/[G(\omega)^2 + F(\omega)^2]\}$ with $G(\omega) = \ln\{[(\epsilon_d + \hbar\omega)^2 + \Gamma^2][(\epsilon_d - \hbar\omega)^2 + \Gamma^2]/[\epsilon_d^2 + \Gamma^2]\}$ and $F(\omega) = 2\{\tan^{-1}[\Gamma/(\epsilon_d - \hbar\omega)] - \tan^{-1}[\Gamma/(\epsilon_d + \hbar\omega)]\}$. $R_q^{\rm non}(\omega)$ increases monotonically with increasing $|\omega|$, and the only characteristic energy scale is $\epsilon_d$. Hence, the peaks seen in Figs. 2(e) and (f) are a genuine many-body effect. In summary many-body correlations, apparently having no impact on the zero-frequency value do affect $R_q(\omega)$ at finite frequencies, by forming a pronounced peak at $\hbar\omega = \Gamma^*(T_K)$. The explanation for these observations will be given later.

*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. Interestingly, $R_q(\omega \to 0)$ versus $\Delta_Z$ exhibits a peak structure reaching values much larger than the quantized resistance in the spin-degenerate case: for example,

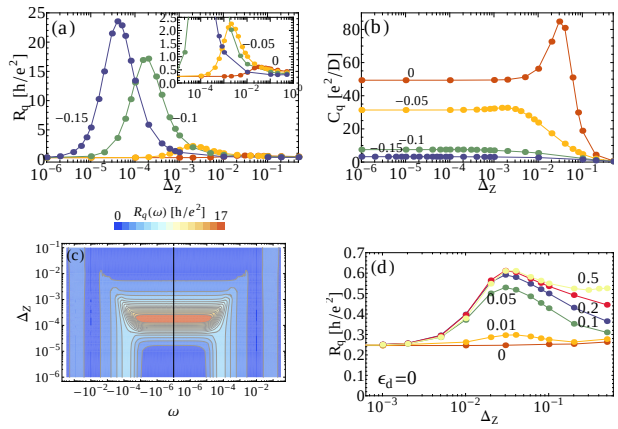


FIG. 3: (color online) (a,b) $R_q(\omega = 0)$ and $C_q(\omega = 0)$ versus $\Delta_Z$ for annotated values of $\epsilon_d$ with $E_{\rm C} = 0.2$. The enlarged view of $R_q(\omega = 0)$ for large $\Delta_Z$ is shown in the inset. (c) Contour plot of $R_q$ as a function of $\omega$ and $\Delta_Z$ in the logarithmic scale in the Kondo regime ($\epsilon_d = -0.1$). (d) $R_q(\omega = 0)$ versus $\Delta_Z$ for different values of $E_{\rm C}$ as annotated. Here $\Gamma = 0.02$ is used.

$R_q(\omega = 0)|_{\rm max} \sim 100 \times h/4e^2$ for $\epsilon_d = -0.15$. Furthermore, the peak is exactly located at $\Delta_Z = \Gamma^*(T_K)$ for the fluctuating valence (Kondo) regime. The peak height increases as the 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 $\epsilon_d = -0.3$ (Kondo regime). As $\Delta_Z$ increases, the low-frequency part of $R_q(\omega)$ for $|\hbar\omega| < k_B T_K$ keeps going up until $\Delta_Z$ reaches $k_B T_K$; 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 $\hbar\omega = \pm k_B T_K$, 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. Finally, Fig. 3(d) compares the zero-frequency values of $R_q$ for different values of the Coulomb interaction in the resonant tunneling regime. In the noninteracting case, there is no peak at all, with $R_q(\omega \to 0)$ equal to $h/4e^2$. However, as soon as the charging energy $2E_{\rm C}$ becomes comparable to $\Gamma^* \sim \Gamma$, a peak starts to rise up and manifests itself for $2E_{\rm C} \gg \Gamma$. 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 can Coulomb interaction increase the relaxation resistance far beyond the universal value, $h/4e^2$ and *(2)* Why does it take place noticeably at $\hbar\omega = \pm\Gamma^*(T_K)$ in the fluctuating valence (Kondo) regime for $\Delta_Z = 0$ or at $\hbar\omega = 0$ for $\Delta_Z = \Gamma^*(T_K)$? 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 low-

ering 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 $\hbar\omega = \pm\Gamma^*$ 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 $p$-$h$ pair excitation states shown in Fig. 1(b) are now lowered by $\Delta_Z$, 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 $\hbar\omega = 0$ when $\Delta_Z = \Gamma^*$. 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 reservoirs, 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 wavefunction. Hence the processes as shown in Fig. 1(b) 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 $|\langle\alpha|\mathcal{N}|\mathrm{gs}\rangle|^2$, where $\alpha$ represent the excited states. In the second-order perturbation theory, this weight corresponding to the processes in Fig. 1(b) is given by

$$|\langle\alpha|\mathcal{N}|\mathrm{gs}\rangle|^2 = t^4 \left| \frac{2}{E_\mu E_{\bar{\mu}}} - \sum_\mu \frac{\mu}{\Delta_Z} \left( \frac{1}{E_\mu} + \frac{1}{\epsilon_\mu} \right) \right|^2 \qquad (2)$$

in the $\omega \to 0$ limit with $E_\mu = 2E_{\mathrm{C}} + \epsilon_\mu$. Interestingly, this weight vanishes for $E_{\mathrm{C}} = 0$ 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 $E_{\mathrm{C}}$, the weight is finite [see Eq. (2)] and for $E_{\mathrm{C}} \to \infty$, it becomes $t^4/(\epsilon_\uparrow\epsilon_\downarrow)^2$. 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 $\Delta_Z \sim \Gamma^*$ 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. Using the relation of charge relaxation resistance and quantum capacitance with the charge susceptibility, we find that in the deep Kondo regime the KS rule ensures a quantized $R_q(\omega = 0) = h/4e^2$. Besides we show that the interpretation of $C_q$ in terms of the DOS fails when many-body effects are present. Here, $C_q(\omega = 0)$ becomes very small even when $\rho_{\mathrm{dot}}(\epsilon_F)$ becomes large due to a completely frozen charge dynamics in the presence of Kondo correlations. Finally, we find that $R_q(\omega)$ is built by dot-lead tunneling events connecting $p$-$h$ excitations in the reservoirs with spin-flip processes in the dot . This interpretation explains our results for $R_q(\Delta_Z = 0, \omega)$ showing peaks at $\hbar\omega = \pm\Gamma^*(T_K)$ and for $R(\Delta_Z = \Gamma^*(T_K), \omega)$ exhibiting a peak at $\hbar\omega = 0$.

M.L. was supported by the NRF Grants (2009-0069554 and 2010-0015416). R. L. was supported by MEC-Spain (Grant No. FIS2008-00781). T.M. and T.J. acknowledge support from ANR 2010 BLANC 0412 02. M.-S.C. was supported by the NRF Grants (2009-0080453 and 2010-0025880).

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