



HAL
open science

Transport through an interacting quantum dot driven out-of-equilibrium

M. Lavagna

► **To cite this version:**

M. Lavagna. Transport through an interacting quantum dot driven out-of-equilibrium. Journal of Physics: Conference Series, 2015, 592, pp.012141. 10.1088/1742-6596/592/1/012141 . hal-01899462

HAL Id: hal-01899462

<https://hal.science/hal-01899462>

Submitted on 19 Oct 2018

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Transport through an interacting quantum dot driven out-of-equilibrium

Mireille Lavagna*

Université Grenoble Alpes, CEA, INAC-SPSMS, Grenoble, France

E-mail: mireille.lavagna@cea.fr

Abstract. We study a single-level quantum dot in the presence of strong Coulomb interaction under nonequilibrium condition. By extending the equation of motion method to nonequilibrium, we study the transport behavior of the system when a dc bias voltage is applied to the leads. The spectral density exhibits two broad peaks centered around the normalized dot level energies and a split Kondo resonance at low temperature with two peaks pinned at the Fermi level of each lead. The approach allows one to recover the unitary condition for the density of states at the Fermi levels and by the way to cure the long-standing problem about the presence of spurious peak in the density of states at equilibrium. Finally we discuss the consequences for the linear and differential conductances of the quantum dot in its steady state.

1. Introduction

Understanding the mechanisms involved in interacting systems far from equilibrium is one of the major problems in condensed matter physics. We study in this paper quantum dots under nonequilibrium conditions. Quantum dots are realized by confining electrons in a small spatial region weakly coupled to two leads. They give rise to a Kondo effect at low temperature [1] when the dot is occupied by a single electron, and hence acquires a spin which is antiferromagnetically coupled to the spin of the conduction electrons in the leads. Predicted at a theoretical level at the end of the 80ies [2, 3], the Kondo effect in quantum dots has been observed experimentally since the end of the 90ies [4-6]. Out of equilibrium the Kondo effect in quantum dots has been studied by a variety of analytical and numerical techniques. In most cases, these techniques combine many-body methods to treat strong interactions with non-equilibrium Green function techniques to take non-equilibrium conditions into account. The system is driven out of equilibrium either by the application of a dc bias voltage between the two leads or by irradiation with an electromagnetic field. Driving the system out of equilibrium leads to a decoherence of the Kondo many-body singlet state and induces a crossover from the Fermi liquid strong coupling regime (local Fermi liquid) to the weak coupling regime. Despite all the effort developed these last years along this direction, many open questions remain about how electron correlations and nonequilibrium effects interfere in those mesoscopic systems.

In this paper we present how the equation of motion (EOM) technique can be generalized to the study of quantum dots out of equilibrium when a dc bias voltage is applied to the two leads. The EOM technique had been applied to the original Anderson model in equilibrium a long time ago [7-9] in the context of dilute magnetic alloys and later on in that of quantum dots [10-20]. The strength of the method is to allow the description of both the high temperature regime



with logarithmic dependence of either resistivity for dilute alloys (or conductance for quantum dots respectively) and the low temperature regime with the formation of a Kondo resonance in the spectral density in addition to broad peaks. However the method is known to present some serious drawbacks as the presence of a spurious peak in the density of states at low temperature. This spurious peak corresponds to an antiresonance in the density of states which occurs just at the position of the Kondo resonance peak at the particle-hole symmetric limit, letting the Kondo physics paradoxically not recovered in that limit. By carefully extending the EOM method to nonequilibrium, it turns out that we solved this long-standing problem and cure the presence of this spurious peak in the spectral density at equilibrium.

The organization of the paper is the following. In section 2 we present the results for the retarded electronic Green function in the dot obtained by solving the set of equations of motion of successive order Green functions when truncated at the second order in $t_{\alpha\sigma}$ by performing all possible decouplings preserving the spin: $\langle n_{\sigma} \rangle$, $\langle c_{k'\alpha\bar{\sigma}}^{\dagger} c_{k\alpha\bar{\sigma}} \rangle$, and the mixed parameter $\langle f_{\bar{\sigma}}^{\dagger} c_{k\alpha\bar{\sigma}} \rangle$. In section 3 we show how, provided that the system is in its steady state, the various decoupling parameters considered can be determined self-consistently from the knowledge of the retarded dot Green function only, even if the fluctuation-dissipation theorem does not hold any longer when the system is out of equilibrium. The derivation is exact and does not require the use of any ansatz as was often used in the past. In Section 4 we stress the importance of incorporating the normalization of the dot level energy and of the inverse lifetime broadening of the various excited states involved in the expression of the retarded dot Green function. The former corrections correspond to the dressing of the bare propagators by interactions and are obtained by taking the real part of the self energy term, whereas the latter corrections are evaluated by means of the generalized Fermi golden rules. In section 5 we discuss the results for the spectral density and the linear and differential conductances when the system driven out of equilibrium is in its steady state.

2. Equation of motion approach for the Anderson model out-of-equilibrium

We consider a quantum dot with two leads driven out of equilibrium by the effect of a finite bias voltage V and if appropriate placed in a magnetic field B . We model the quantum dot by the single-level Anderson model [21]

$$H = \sum_{k,\alpha \in (L,R),\sigma} \varepsilon_{k\alpha\sigma} c_{k\alpha\sigma}^{\dagger} c_{k\alpha\sigma} + \sum_{\sigma} \varepsilon_{\sigma} f_{\sigma}^{\dagger} f_{\sigma} + U n_{\uparrow} n_{\downarrow} + \sum_{k,\alpha \in (L,R),\sigma} (t_{\alpha\sigma} c_{k\alpha\sigma}^{\dagger} f_{\sigma} + h.c.). \quad (1)$$

where $c_{k\alpha\sigma}^{\dagger}$ ($c_{k\alpha\sigma}$) creates (destroys) an electron in the α (left and right) lead with momentum k , spin σ ($\sigma = \pm 1$) and energy $\varepsilon_{k\alpha\sigma}$; the equilibrium momentum distribution in the α lead is $n_F(\omega - \mu_{\alpha}) = [\exp[(\omega - \mu_{\alpha})/k_B T] + 1]^{-1}$ where μ_{α} is the chemical potential in the α lead and $eV = \mu_L - \mu_R$ the bias voltage applied between the two leads; f_{σ}^{\dagger} (f_{σ}) creates (destroys) an electron in the dot with energy $\varepsilon_{\sigma} = \varepsilon_0 + \sigma\Delta/2$, where $\Delta = \mu_B B$ is the Zeeman splitting due to the magnetic field B ; $n_{\sigma} = f_{\sigma}^{\dagger} f_{\sigma}$; U is the on-site Coulomb interaction -charging energy- in the dot; and $t_{\alpha\sigma}$ is the tunneling matrix element between the states $|k\alpha\sigma\rangle$ and $|\sigma\rangle$ in the leads and the dot respectively.

The equation of motion technique has been extensively used in the past to study quantum impurities at equilibrium. More recently there have been works to extend its applicability to nonequilibrium [10-20]. Briefly speaking the equation of motion method consists in differentiating the Green function $G_{\sigma}^r(t, t')$ with respect to time t , thereby generating higher-order Green's functions. Writing the equations of motion for those series of Green-functions leads to a set of linear equations that can be closed -truncated- at a given order by performing the proper decouplings. Using the set of EOM truncated at the second order in $t_{\alpha\sigma}$ by performing a decoupling in terms of all possible two-operator correlation functions with equal-spin, $\langle f_{\bar{\sigma}}^{\dagger} c_{\alpha k \bar{\sigma}} \rangle$,

$\langle c_{\alpha k' \bar{\sigma}}^\dagger c_{\alpha k \bar{\sigma}} \rangle$ and $\langle n_{\bar{\sigma}} \rangle$, we obtain the following expression for $G_\sigma^r(\omega)$ [20]

$$G_\sigma^r(\omega) = \frac{1 - \langle n_{\bar{\sigma}} \rangle}{\omega - \varepsilon_\sigma - \Sigma_\sigma^0(\omega) - \Pi_{\sigma 1}(\omega)} + \frac{\langle n_{\bar{\sigma}} \rangle}{\omega - \varepsilon_\sigma - U - \Sigma_\sigma^0(\omega) - \Pi_{\sigma 2}(\omega)}. \quad (2)$$

where in the wide band limit $\Sigma_\sigma^0(\omega) = -i\Gamma_\sigma(\omega)$ with $\Gamma_\sigma(\omega) = \sum_{\alpha=L,R} \Gamma_{\alpha\sigma}(\omega)$ and $\Gamma_{\alpha\sigma}(\omega) = \pi \sum_k |t_{\alpha\sigma}|^2 \delta(\omega - \varepsilon_{k\alpha\sigma}) = \pi |t_{\alpha\sigma}|^2 \rho_{\alpha\sigma}^0(\omega)$ (where $\rho_{\alpha\sigma}^0(\omega)$ is the unrenormalized density of states in the lead α with spin σ). In the case when the band is flat of infinite width (i.e. in the wide flat band WFB limit), $\Sigma_\sigma^0(\omega)$ is independent of ω , taking the value $\Sigma_\sigma^0 = -i\Gamma_\sigma$. $\Pi_{\sigma 1}(\omega)$ and $\Pi_{\sigma 2}(\omega)$ are defined as

$$\Pi_{\sigma 1}(\omega) = -U \frac{\Sigma_{\sigma 1}(\omega) - (\omega - \varepsilon_\sigma) \Sigma_{\sigma 4}(\omega)}{\omega - \varepsilon_\sigma - U - \Sigma_\sigma^0(\omega) - \Sigma_{\sigma 3}(\omega) + U \Sigma_{\sigma 4}(\omega)} = \frac{-\Sigma_{\sigma 1}(\omega) + (\omega - \varepsilon_\sigma) \Sigma_{\sigma 4}(\omega)}{[I_\sigma(\omega)]^{-1} + \Sigma_{\sigma 4}(\omega)}, \quad (3)$$

$$\Pi_{\sigma 2}(\omega) = U \frac{\Sigma_{\sigma 2}(\omega) + (\omega - \varepsilon_\sigma - U) \Sigma_{\sigma 4}(\omega)}{\omega - \varepsilon_\sigma - \Sigma_\sigma^0(\omega) - \Sigma_{\sigma 3}(\omega) + U \Sigma_{\sigma 4}(\omega)} = \frac{\Sigma_{\sigma 2}(\omega) + (\omega - \varepsilon_\sigma - U) \Sigma_{\sigma 4}(\omega)}{(1 + [I_\sigma(\omega)]^{-1}) + \Sigma_{\sigma 4}(\omega)}. \quad (4)$$

where

$$I_\sigma(\omega) = \frac{U}{\omega - \varepsilon_\sigma - U - \Sigma_\sigma^0(\omega) - \Sigma_{\sigma 3}(\omega)}, \quad (5)$$

$$\Sigma_{\sigma i}(\omega) = \sum_{k,\alpha} |t_{\alpha\bar{\sigma}}|^2 \left[\frac{A_{k\alpha\sigma i}}{\omega + \tilde{\varepsilon}_{\bar{\sigma}} - \tilde{\varepsilon}_\sigma - \varepsilon_{k\alpha\bar{\sigma}} + i\tilde{\gamma}_\sigma} + \frac{A'_{k\alpha\sigma i}}{\omega + \tilde{\varepsilon}_{k\alpha\bar{\sigma}} - \tilde{\varepsilon}_\sigma - \tilde{\varepsilon}_{\bar{\sigma}} - U + i\tilde{\gamma}_D} \right]. \quad (6)$$

with $A_{k\alpha\sigma 1} = \sum_{k'} \langle c_{k'\alpha\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$, $A_{k\alpha\sigma 2} = 1 - \sum_{k'} \langle c_{k'\alpha\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$, $A_{k\alpha\sigma 3} = 1$, and $A_{k\alpha\sigma 4} = \langle f_{\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle / t_{\alpha\bar{\sigma}}$; $A'_{k\alpha\sigma i} = A_{k\alpha\sigma i}^*$ for $i = 1, 2, 3$, and $A'_{k\alpha\sigma 4} = -A_{k\alpha\sigma 4}^*$.

As a starting point, $\tilde{\gamma}_\sigma$ and $\tilde{\gamma}_D$ are both an infinitesimal positive ($\gamma_\sigma = \gamma_D = +i\delta$), and $\tilde{\varepsilon}_\sigma$ is the bare level energy in the dot ε_σ . A major improvement that we will discuss below consists in replacing respectively (i) the bare level energy ε_σ by the normalized level energy $\tilde{\varepsilon}_\sigma$; and (ii) γ_σ, γ_D by the relaxation rates $\tilde{\gamma}_\sigma, \tilde{\gamma}_D$ determined from the use of Fermi golden rules.

Eq.(2) can be rewritten in the following way [20]

$$G_\sigma^r(\omega) = \frac{1 + I_\sigma(\omega) [\langle n_{\bar{\sigma}} \rangle + \Sigma_{\sigma 4}(\omega)]}{[\omega - \varepsilon_\sigma - \Sigma_\sigma^0(\omega)] + I_\sigma(\omega) [\Sigma_{\sigma 1}(\omega) - \Sigma_\sigma^0(\omega) \Sigma_{\sigma 4}(\omega)]}. \quad (7)$$

Both of these expressions Eqs.(2,7) constitute a generalization of Meir, Wingreen, Lee (MWL) [10, 11] and Lacroix [9] results to out-of-equilibrium situation for any value of U and temperature. But whereas the domain of validity of MWL theory is restricted to high temperature, the approximation scheme that we propose extends its validity to any temperature range. The differences essentially lies in the term proportional to $\Sigma_{\sigma 4}(\omega)$; this latter contribution is brought by the additional decoupling that is considered which provides an extra mean field parameter $\langle f_{\bar{\sigma}}^\dagger c_{\alpha k \bar{\sigma}} \rangle$ that can be viewed as a pseudo-order parameter which is different from zero in the strong coupling regime reminding of the slave-boson introduced in auxiliary-field approaches. We will show below that, provided that the system is in its steady state, these 3 parameters can be determined self-consistently even out-of-equilibrium when the fluctuation-dissipation theorem does not hold any longer. We argue that taking this additional term into account is crucial to describe the strong coupling regime reached at low temperature, weak bias voltage and Zeeman field. $\Sigma_{\sigma i}(\omega)$ ($i = 1, 2$) show logarithmic singularities at the Fermi level at low temperatures defining a temperature scale T_K below which a Kondo peak resonance forms in the density of states. Below T_K , $\Sigma_{\sigma 4}(\omega)$ contribution plays a key role to compensate those singularities and make $G_\sigma^r(\omega)$ fulfill the unitary condition at the Fermi level and hence the conductance reach the unitary limit at zero temperature. We have checked that the above expressions for $G_\sigma^r(\omega)$ given in Eqs.(2,7) give back both Lacroix' result in the infinite U limit and $V = 0$, and MWL result in the high temperature limit, so when $\langle f_{\bar{\sigma}}^\dagger c_{\alpha k \bar{\sigma}} \rangle = 0$ (and hence $\Sigma_{\sigma 4}(\omega) = 0$ and $\sum_{k'} \langle c_{\alpha k' \bar{\sigma}}^\dagger c_{\alpha k \bar{\sigma}} \rangle = \delta_{kk'} n_F(\varepsilon_{k\bar{\sigma}} - \mu_\alpha)$).

3. Self-consistency for $\langle n_\sigma \rangle$, $\langle c_{k'\alpha\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$ and $\langle f_{\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$

Under both equilibrium and nonequilibrium condition, $\langle n_\sigma \rangle$ can be obtained from the lesser Green function $G_\sigma^<(\omega)$ according to: $\langle n_\sigma \rangle = -i \int \frac{d\omega}{2\pi} G_\sigma^<(\omega)$. In equilibrium the lesser Green function is related to the retarded Green function through the fluctuation-dissipation theorem: $G_\sigma^<(\varepsilon) = -2in_F(\omega - \mu_\sigma) \text{Im}G_\sigma^r(\varepsilon)$ and $\langle n_\sigma \rangle$ can be simply expressed as a function of the retarded Green function.

Out of equilibrium the fluctuation-dissipation theorem does not hold any longer and one cannot directly connect the lesser to the retarded Green function. Nevertheless we have shown that in the steady state and when $\Gamma_{L\sigma}(\varepsilon)$ and $\Gamma_{R\sigma}(\varepsilon)$ are both independent of ε , $\langle n_\sigma \rangle$ can still be expressed as a function of the only retarded (or advanced) Green function without requiring any knowledge of the lesser (or greater) Green function. The self-consistent equation for $\langle n_\sigma \rangle$ [20] is

$$\langle n_\sigma \rangle = i \int \frac{d\omega}{2\pi} \frac{\Gamma_{L\sigma} n_F(\omega - \mu_{L\sigma}) + \Gamma_{R\sigma} n_F(\omega - \mu_{R\sigma})}{\Gamma_{L\sigma} + \Gamma_{R\sigma}} [G_\sigma^r(\omega) - G_\sigma^a(\omega)]. \quad (8)$$

This constitutes an exact result which does not require the use of any ansatz (as for instance the Ng ansatz [22]) as often used in the past.

As far as the self-consistent determinations of $\langle c_{k'\alpha\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$ and $\langle f_{\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$ are concerned, it is important to realize that $\langle c_{k'\alpha\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$ and $\langle f_{\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$ come into play only through some related integrals that are denoted as $L_\sigma(\omega)$ and $M_\sigma(\omega)$

$$L_\sigma(\omega + i\delta) = \sum_{k,k',\alpha} |t_\sigma|^2 \frac{\langle c_{k'\alpha\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle}{\omega - \varepsilon_{k\alpha\sigma} + i\delta} \quad ; \quad M_\sigma(\omega + i\delta) = \sum_{k,\alpha} t_{\alpha\sigma} \frac{\langle f_{\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle}{\omega - \varepsilon_{k\alpha\sigma} + i\delta} \quad . \quad (9)$$

Thus $\Sigma_{\sigma 1}(\omega)$ and $\Sigma_{\sigma 4}(\omega)$ can be written in terms of $L_\sigma(\omega)$ and $M_\sigma(\omega)$ according to

$$\Sigma_{\sigma 1}(\omega) = L_{\bar{\sigma}}(\omega + \tilde{\varepsilon}_{\bar{\sigma}} - \tilde{\varepsilon}_\sigma + i\tilde{\gamma}_\sigma) - L_{\bar{\sigma}}^*(-\omega + \tilde{\varepsilon}_{\bar{\sigma}} + \tilde{\varepsilon}_\sigma + U + i\tilde{\gamma}_D), \quad (10)$$

$$\Sigma_{\sigma 4}(\omega) = M_{\bar{\sigma}}(\omega + \tilde{\varepsilon}_{\bar{\sigma}} - \tilde{\varepsilon}_\sigma + i\tilde{\gamma}_\sigma) + M_{\bar{\sigma}}^*(-\omega + \tilde{\varepsilon}_{\bar{\sigma}} + \tilde{\varepsilon}_\sigma + U + i\tilde{\gamma}_D). \quad (11)$$

knowing that $\Sigma_{\sigma 2}(\omega) = \Sigma_{\sigma 3} - \Sigma_{\sigma 1}(\omega)$. Then in the wide band limit, the self-consistent equations for $\langle c_{k'\alpha\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$ and $\langle f_{\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$ result in the following equations (see [13]) for $L_\sigma(\omega)$ and $M_\sigma(\omega)$

$$L_\sigma(\omega + i\delta) = \sum_\alpha \frac{\Gamma_{\alpha\sigma}}{\pi} \int d\varepsilon \frac{n_F(\varepsilon - \mu_\alpha)}{\omega - \varepsilon + i\delta} [1 + i\Gamma_\sigma G_\sigma^a(\varepsilon)], \quad (12)$$

$$M_\sigma(\omega + i\delta) = \sum_\alpha \frac{\Gamma_{\alpha\sigma}}{\pi} \int d\varepsilon \frac{n_F(\varepsilon - \mu_\alpha)}{\omega - \varepsilon + i\delta} G_\sigma^a(\varepsilon). \quad (13)$$

In the wide band limit, it turns out that even out of equilibrium the self-consistent equations for the integrals $L_\sigma(\omega)$ and $M_\sigma(\omega)$ (i.e. for $\langle c_{k'\alpha\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$ and $\langle f_{\bar{\sigma}}^\dagger c_{k\alpha\bar{\sigma}} \rangle$) are written exactly in the same way as at equilibrium and depend only on the advanced Green function $G_\sigma^a(\varepsilon)$ without requiring any knowledge of the corresponding lesser (or greater) Green function. The only change is to have μ_L , μ_R entered in the expression of the momentum distribution functions for the leads L, R respectively.

4. Normalization of the dot level energy and of the relaxation rates

Initially the dot level energy $\tilde{\varepsilon}_\sigma$ entering the expressions of the self-energies (cf. Eq.6) is the bare dot level energy ε_σ and the relaxation rates $\tilde{\gamma}_\sigma$, $\tilde{\gamma}_D$ are both infinitesimal positive. As was already mentioned at the end of section 2, a major improvement consists in considering normalization effects on those quantities. The normalized dot level energy $\tilde{\varepsilon}_\sigma$ is identified as the

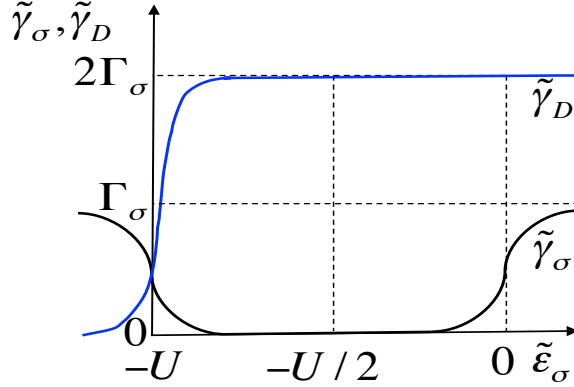


Figure 1. Schematic dependence of relaxation rates $\tilde{\gamma}_\sigma$ and $\tilde{\gamma}_D$ versus normalized dot level energy $\tilde{\epsilon}_\sigma$ for $V = B = 0$ in the limit $T \ll U$.

lower energy pole of the retarded dot electron Green function $G_\sigma^r(\omega)$ (i.e. $\tilde{\epsilon}_\sigma = \epsilon_\sigma + Re\Pi_{\sigma 1}(\tilde{\epsilon}_\sigma)$), and obtained from

$$\tilde{\epsilon}_\sigma = \epsilon_\sigma + Re\Sigma_{\sigma 1}(\omega = \tilde{\epsilon}_\sigma). \quad (14)$$

since the contribution brought by $Re\Sigma_{\sigma 4}(\omega = \tilde{\epsilon}_\sigma)$ is negligible. In the regime when $|\epsilon_\sigma - \mu| \gg \Gamma$, we get

$$\tilde{\epsilon}_\sigma = \epsilon_\sigma - \sum_\alpha \frac{\Gamma_{\alpha\bar{\sigma}}}{\pi} \log \frac{|\epsilon_{\bar{\sigma}} - \mu_\alpha|}{|\epsilon_{\bar{\sigma}} + U - \mu_\alpha|}. \quad (15)$$

The latter result is in agreement with Haldane's result [23] obtained by using scaling theory.

Besides the relaxation rates $\tilde{\gamma}_\sigma, \tilde{\gamma}_D$ are the inverse lifetime broadening or decay rates of the corresponding excited states with 1 and 2 electrons in the dot respectively. Those decay rates can be determined by using perturbation theory in $t_{\alpha\sigma}$, leading to the generalized Fermi golden rules, the excited state playing the role of the initial state $|i\rangle$ in the perturbation theory. Up to the fourth order in $t_{\alpha\sigma}$, $\gamma_{\sigma,D} = \gamma_{\sigma,D}^{(2)} + \gamma_{\sigma,D}^{(4)}$ where $\gamma_{\sigma,D}^{(2)} = 2\pi \sum_{|f\rangle} |\langle f|H_T|i\rangle|^2 \delta(E_i - E_f)$ and $\gamma_{\sigma,D}^{(4)} = 2\pi \sum_{|f\rangle} |\sum_{|int\rangle} \langle f|H_T|int\rangle \frac{1}{E_i - E_{int}} \langle int|H_T|i\rangle|^2 \delta(E_i - E_f)$. H_T is the tunneling part of the hamiltonian, $|i\rangle$ is the initial state with energy E_i , $|f\rangle$ and $|int\rangle$ are all the possible final and intermediate states with energies E_f and E_{int} respectively. The calculation is straightforward. The only complication comes from the fact that one must systematically consider all the possible processes by which the initial state can desintegrate (all possible final and intermediate states) provided that $E_i = E_f$ while letting the possibility to pass through distinct intermediate states in back and forth paths respectively as taken into account by the summation over the intermediate states $|int\rangle$ in the analytical expression of $\gamma_{\sigma,D}^{(4)}$ given above. Incorporating the normalization of the dot level energy, we obtain the following results

$$\tilde{\gamma}_\sigma^{(2)} = \sum_\alpha \left[\Gamma_{\alpha\sigma} [1 - n_F(\tilde{\epsilon}_\sigma - \mu_\alpha)] + \Gamma_{\alpha\bar{\sigma}} n_F(\tilde{\epsilon}_{\bar{\sigma}} + U - \mu_\alpha) \right], \quad (16)$$

$$\tilde{\gamma}_D^{(2)} = \sum_{\alpha\sigma} \left[\Gamma_{\alpha\sigma} [1 - n_F(\tilde{\epsilon}_\sigma + U - \mu_\alpha)] \right], \quad (17)$$

$$\tilde{\gamma}_\sigma^{(4)} = \frac{1}{2\pi} \sum_{\alpha\beta} \int d\epsilon [1 - n_F(\epsilon - \mu_\beta)] \cdot \left[n_F(\epsilon - \mu_\alpha) \left\{ \frac{\Gamma_{\alpha\sigma}\Gamma_{\beta\sigma}}{(\epsilon - \tilde{\epsilon}_\sigma)^2} + \frac{\Gamma_{\alpha\bar{\sigma}}\Gamma_{\beta\bar{\sigma}}}{(\epsilon - \tilde{\epsilon}_{\bar{\sigma}} - U)^2} \right\} + n_F(\epsilon + \tilde{\epsilon}_{\bar{\sigma}} - \tilde{\epsilon}_\sigma - \mu_\alpha) \Gamma_{\alpha\bar{\sigma}}\Gamma_{\beta\sigma} \left\{ \frac{1}{\epsilon - \tilde{\epsilon}_{\bar{\sigma}} - U} - \frac{1}{\epsilon - \tilde{\epsilon}_\sigma} \right\}^2 \right],$$

(18)

$$\tilde{\gamma}_D^{(4)} = \frac{1}{2\pi} \sum_{\alpha\beta,\sigma} \int d\varepsilon [1 - n_F(\varepsilon - \mu_\beta)] \frac{1}{(\varepsilon - \tilde{\varepsilon}_\sigma - U)^2}.$$

$$\left[n_F(\varepsilon - \mu_\alpha) \Gamma_{\alpha\sigma} \Gamma_{\beta\sigma} + [1 - n_F(\tilde{\varepsilon}_\sigma + \tilde{\varepsilon}_{\bar{\sigma}} + U - \varepsilon - \mu_\alpha)] \Gamma_{\alpha\sigma} \Gamma_{\beta\bar{\sigma}} \right]. \quad (19)$$

This result constitutes a generalization of the results of [11, 24] to any value of U . Indeed in [11, 24], since only the infinite U limit is considered, $\tilde{\gamma}_D$ is not accounted for and the result for $\tilde{\gamma}_\sigma$ contains the infinite U contribution only. Moreover these expressions can be seen as an extension of the results of [13] obtained by pushing the truncation of the set of EOM up to following orders. At equilibrium and in the absence of magnetic field, the 4th order contributions in t_σ to both $\tilde{\gamma}_\sigma$ and $\tilde{\gamma}_D$ are zero; $\tilde{\gamma}_\sigma$ and $\tilde{\gamma}_D$ at $V = B = 0$ are then simply given by their second order terms, the dependence of which as a function of $\tilde{\varepsilon}_\sigma$ are represented in Figure 1. Notice that $\tilde{\gamma}_\sigma$ at $V = B = 0$ is zero at the particle-hole symmetric point whereas it equals $\Gamma_{\bar{\sigma}} = \sum_\alpha \Gamma_{\alpha\bar{\sigma}}$ and $\Gamma_\sigma = \sum_\alpha \Gamma_{\alpha\sigma}$ respectively in the empty and doubly occupied dot regimes. Moreover $\tilde{\gamma}_D$ at $V = B = 0$ shows an abrupt increase around the doubly-occupied dot regime before saturating to $(\Gamma_\sigma + \Gamma_{\bar{\sigma}})$. At finite bias or finite magnetic field, the fourth order contributions $\tilde{\gamma}_\sigma^{(4)}$ and $\tilde{\gamma}_D^{(4)}$ becomes nonzero. The respective deviations towards the values obtained at $V = B = 0$ are found to be linear in either V or B .

5. Results about the spectral density and the linear/ differential conductances out of equilibrium

In this section we restrict our discussion to the situation where the magnetic field is zero. The spin-dependent spectral density in the dot is obtained from the retarded Green function: $A_\sigma(\omega) = -ImG_\sigma^r(\omega)/\pi$. The spectral density exhibits a series of peaks identified as the poles of the retarded Green function. We list them below:

- the lower and upper energy poles already discussed in section 4, located at $\tilde{\varepsilon}_\sigma$ and $\tilde{\varepsilon}_\sigma + U$, leading to the formation of the two broad peaks in the spectral density centered around these values;

- two additional poles arising at μ_α for temperature lower than the Kondo temperature T_K resulting from the logarithmic singularities of $\Sigma_{\sigma 1}(\omega)$ and $\Sigma_{\sigma 4}(\omega)$ around these values leading to the formation of a split Kondo resonance with two peaks pinned at the chemical potential of each lead. The splitting of the Kondo peak is equal to the bias voltage energy eV .

To be exhaustive, we will mention the presence of another pole at $\mu_\alpha + \tilde{\varepsilon}_\sigma + \tilde{\varepsilon}_{\bar{\sigma}} + U$ due to the logarithmic singularities of $\Sigma_{\sigma 1}(\omega)$ and $\Sigma_{\sigma 4}(\omega)$ at these values, brought by the second term in the r.h.s. of the expressions given in Eq.(6) for the self-energies $\Sigma_{\sigma i}(\omega)$. This pole leads to the formation of a spurious peak of negative intensity (antiresonance) in the spectral density. However we point out that these latter logarithmic singularities are washed out by the large value of the inverse lifetime broadening $\tilde{\gamma}_D^{(4)}$ as evaluated in section 4. As a result, this spurious peak is no longer present in our theory. This solves a long-standing unsolved problem about the existence of this spurious peak pointed out in previous works and which was all the more undesirable that it just compensates and destroys the Kondo peak at the particle-hole symmetric point and makes the Kondo effect paradoxically disappear at that point. We end up by showing that the Green function in the dot at the energies μ_α fulfills the unitary condition: $|G_\sigma^r(\mu_\alpha)|^2 = -ImG_\sigma^r(\mu_\alpha)/\Gamma_\sigma$. To prove this, we evaluate $L_\sigma(\omega)$ and $M_\sigma(\omega)$ in the vicinity of μ_α . Noticing that at low temperature $\frac{n_F(\varepsilon - \mu_\alpha)}{\omega - \varepsilon + i\delta}$ varies strongly at $\varepsilon \sim \mu_\alpha$ whereas $G_\sigma^a(\varepsilon)$ varies smoothly, one can write for $\omega \sim \mu_\alpha$

$$L_\sigma(\omega) = - \sum_\alpha \frac{\Gamma_{\alpha\sigma}}{\pi} G_\sigma^a(\omega) \left[\ln \frac{|\mu_\alpha - \omega|}{D} + i\pi n_F(\omega) \right], \quad (20)$$

$$M_\sigma(\omega) = -\sum_\alpha \frac{\Gamma_{\alpha\sigma}}{\pi} [1 + i\Gamma_\sigma G_\sigma^a(\omega)] \left[\ln \left| \frac{\mu_\alpha - \omega}{D} \right| + i\pi n_F(\omega) \right]. \quad (21)$$

Both of these functions show logarithmic singularities around $\omega = \mu_\alpha$. Hence the self-energies $\Sigma_{\sigma 1}(\omega)$ and $\Sigma_{\sigma 4}(\omega)$ at $T = 0$ takes the following form at $\omega \sim \mu_\alpha$

$$\Sigma_{\sigma 1}(\omega) = -\sum_\alpha \frac{\Gamma_{\alpha\sigma}}{\pi} G_\sigma^a(\omega) \ln \left| \frac{\mu_\alpha - \omega}{D} \right|, \quad (22)$$

$$\Sigma_{\sigma 4}(\omega) = -\sum_\alpha \frac{\Gamma_{\alpha\sigma}}{\pi} [1 + i\Gamma_\sigma G_\sigma^a(\omega)] \ln \left| \frac{\mu_\alpha - \omega}{D} \right|. \quad (23)$$

Incorporating the last two expressions into Eq.(2), we show that the dot electron Green function at $\omega = \mu_\alpha$ writes

$$G_\sigma^r(\mu_\alpha) = \frac{G_\sigma^a(\mu_\alpha)}{1 + 2i\Gamma_\sigma G_\sigma^a(\mu_\alpha)}. \quad (24)$$

The solution to this equation is of the form: $G_\sigma^r(\mu_\alpha) = \exp^{-i\delta_\sigma(\mu_\alpha)} \sin \delta_\sigma(\mu_\alpha) / \Gamma_\sigma$. At the value of the chemical potential in the leads $\omega = \mu_\alpha$, the dot electron Green function fulfills the following relation: $|G_\sigma^r(\mu_\alpha)|^2 = -\text{Im} G_\sigma^r(\mu_\alpha) / \Gamma_\sigma$ usually referred to as the unitary condition. As a result: $A_\sigma(\mu_\alpha) = \sin^2 \delta_\sigma(\mu_\alpha) / (\pi \Gamma_\sigma)$. Then in the case when the dot is symmetrically coupled to the leads ($\Gamma_{L\sigma} = \Gamma_{R\sigma}$), the S-matrix at the chemical potential μ_α obtained from $S_\sigma(\omega) = \mathbf{1} - iT_\sigma(\omega)$ with $T_\sigma^{\alpha\beta}(\omega) = 2\sqrt{\Gamma_{\alpha\sigma}\Gamma_{\beta\sigma}} G_\sigma^r(\omega)$ simply writes

$$S_\sigma(\omega) = \exp^{-i\delta_\sigma(\mu_\alpha)} \begin{pmatrix} \cos \delta_\sigma(\mu_\alpha) & -i \sin \delta_\sigma(\mu_\alpha) \\ -i \sin \delta_\sigma(\mu_\alpha) & \cos \delta_\sigma(\mu_\alpha) \end{pmatrix}. \quad (25)$$

Determining the precise value of $\delta_\sigma(\mu_\alpha)$ supposes to solve the self-consistency Eq.8 for $\langle n_\sigma \rangle$ which requires the knowledge of the Green function over the whole energy range and not only at the values of the chemical potentials. At equilibrium the phase shift at the chemical potential obeys the Friedel sum rule $\delta_\alpha(\mu_\alpha) = \pi n_\sigma$, corresponding to the complete screening of the spin in the dot by the spin of the conduction electrons in the leads and $A_\sigma(\mu_\alpha) = \sin^2 \pi n_\sigma / (\pi \Gamma_\sigma)$. At the particle-hole symmetric point $\delta_\sigma(\mu_\alpha) = \pi/2$ leading to the unitary limit with $A_\sigma(\mu) = 1/(\pi \Gamma_\sigma)$.

In the steady state, the dc-current can be calculated by using Meir and Wingreen's formula [25] generalizing Landauer formula to the interacting case. In the case when $\Gamma_{L\sigma}(\omega)$ and $\Gamma_{R\sigma}(\omega)$ are proportional to each other, i.e. $\Gamma_{L\sigma}(\omega) = \lambda \Gamma_{R\sigma}(\omega)$, the dc-current for spin σ is expressed as a function of the retarded Green function in the dot. The linear conductance dI_σ/dV in the limit V going to zero, is found to reach the unitary limit at the particle-hole symmetric point and the differential conductance dI_σ/dV at finite V decreases with increasing V giving rise to a zero bias peak of width T_K around $V = 0$ followed by a broad peak at higher bias. The results will be presented in more details in a forthcoming paper [20].

6. Conclusion

In this paper we have generalized the equation of motion approach to the situation of a quantum dot driven out of equilibrium by the application of a dc bias voltage. The main ingredients that we have incorporated are the following: (i) We have considered all possible decoupling parameters with equal spin: $\langle n_\sigma \rangle$, $\langle c_{k'\alpha\sigma}^\dagger c_{k\alpha\sigma} \rangle$ and the mixed parameter $\langle f_\sigma^\dagger c_{k\alpha\sigma} \rangle$ in order to truncate the set of equations of motion of the Green functions which has then been solved; (ii) We have shown that, provided that the system is its steady state, the various decoupling parameters can be obtained self-consistently from the knowledge of the retarded Green functions only, even if the fluctuation-dissipation theorem does not hold any longer; (iii) We have systematically considered the normalization of the bare dot level energy introduced by

the self-energy corrections corresponding to the dressing of the bare propagators, and (iv) We have taken into account the normalized inverse lifetime broadening of the various excited states by using the Fermi golden rules. We have thus derived the retarded electronic Green function in the dot and deduced the spectral density. The spectral density exhibits two broad peaks centered around the normalized dot level energies. The Kondo resonance formed at low temperature is split by the bias voltage with the two peaks pinned at the chemical potentials of each of the two leads. The merit of the approach developed is to enable us both to recover the unitary condition for the density of states at the split resonance peak and to cure the long-standing problem about the presence of spurious peak in the density of states. In this way the quantum dot driven out of equilibrium feels a strong Kondo effect at the particle-hole symmetric point. Finally we have discussed the evolution of the linear and the differential conductances as a function of gate and bias voltages when the quantum dot is driven in its steady state.

Acknowledgments

We wish to thank Shiue-yuan Shiao and Raphaël Van Roermund for helpful discussions at the early stage of the work, Adeline Crépieux, Harold Baranger, Soumya Bera and Shaon Sahoo for stimulating discussions. The work has been funded by the Indo-French Centre for the Promotion of Advanced Research (IFCPAR) under Contract No.4704 and the Nanosciences Foundation of Grenoble under RTRA Contract CORTANO.

* Also at the Centre National de la Recherche Scientifique (CNRS)

References

- [1] Hewson A P 1993 *The Kondo Problem to Heavy Fermions* (Cambridge University Press) and references within
- [2] Ng T K and Lee P A 1988 *Phys. Rev. Lett.* **47** 452
- [3] Glazman L and Raikh M 1988 *JETP Lett.* **11** 2389
- [4] Goldhaber-Gordon D, Shtrikman H, Mahalu D, Abusch-Magder D, Meirav U, and Kastner M 1998 *Nature* **391** 156
- [5] Cronenwett S M, Oosterkamp, and Kouwenhoven L P 1998 *Science* **281** 165115
- [6] van der Wiel W, De Franceschi S, Fujisawa T, Elzerman J, Tarucha S, and Kouwenhoven L P 2000 *Science* **289** 2105
- [7] Appelbaum J A, and Penn D R 1969 *Phys. Rev.* **188** 874
- [8] Theumann A 1969 *Phys. Rev.* **178** 978
- [9] Lacroix C 1981 *J. Phys. F: Met.Phys.* **11** 2389; Lacroix C 1981 *J. Appl. Phys.* **53** 2131
- [10] Meir Y, Wingreen N S, and Lee P A 1991 *Phys. Rev. Lett.* **66** 3048
- [11] Meir Y, Wingreen N S, and Lee P A 1993 *Phys. Rev. Lett.* **70** 260
- [12] Kashcheyevs V, Aharony A, and Entin-Wohlman O 2006 *Phys. Rev. B* **73** 125338
- [13] Van Roermund R, Shiao S Y, and Lavagna M 2010 *Phys. Rev. B* **81** 165115
- [14] Krawiec M and Wysokinski K I 2002 *Phys. Rev. B* **66** 165408
- [15] Martinek J, Utsumi Y, Imamura H, Barnaś J, Maekawa S, König J, and Schön G 2003 *Phys. Rev. Lett.* **91** 127203
- [16] Monreal R C and Flores F 2005 *Phys. Rev. B* **72** 195105
- [17] Entin-Wohlman O, Aharony A, and Meir Y 2005 *Phys. Rev. B* **71** 035333
- [18] Qi Y, Zhu J X, and Ting C S 2009 *Phys. Rev. B* **79** 205110
- [19] Balseiro C A, Usaj G, and Sánchez M J 2010 *J. Phys.: Condens. Matter* **22** 425602
- [20] Lavagna M *to be published*
- [21] Anderson P W 1961 *Phys. Rev.* **124** 41
- [22] Ng T K 1996 *J. Phys. F* *textit*Phys. Rev. Lett. **76** 487
- [23] Haldane F D M 1978 *Phys. Rev. Lett* **40** 416
- [24] Wingreen N S, Meir Y 1994 *Phys. Rev. B* **49** 11040
- [25] Meir Y, Wingreen N S 1992 *Phys. Rev. Lett* **68** 2512