Fine structure in the off-resonance conductance of small Coulomb-blockade systems

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We show how a fine, multiple-peak structure can arise in the off-resonance, zero-bias conductance of Coulomb-blockade systems. In order to understand how this effect comes about one must abandon the orthodox, mean-field understanding of the Coulomb-blockade phenomenon and consider quantum fluctuations in the occupation of the single-particle electronic levels. We illustrate such an effect with a spinless Anderson-like model for multilevel systems and an equation-of-motion method for calculating Green's functions that combines two simple decoupling schemes. [S0163-1829(97)02723-9]

Resonant tunneling through small, isolated, multilevel systems such as a quantum dot[1] causes a peak in the zero-bias conductance whenever a single-electron level coincides with the chemical potential (μ) in the leads. Without a Coulomb interaction a series of peaks with separation $\Delta\epsilon$ (energy-level spacing) would be observed when, for instance, the energy levels are lowered relative to μ by external gates. At each peak the number of electrons in the system increases by one, filling the corresponding single-electron state. Coulomb interactions, however, have drastic effects on the conductance. (i) On the one hand, the current can be suppressed over a large range of gate voltages since incoming electrons may be strongly repelled by those already present in the system. This phenomenon is generally known as Coulomb blockade[1] (CB). A mean-field-type picture[2] suffices to describe such an effect: Each time an additional electron is added to a single-particle state in the system all the other energy levels are shifted with respect to their previous values by an amount U that is related to the Coulomb repulsion. Therefore, if $U \gg \Delta\epsilon$, a sparse series of conductance peaks with separation U is expected instead of that with separation $\Delta\epsilon$. (ii) A more exotic phenomenon can take place when, at very low temperatures, the conductance between CB peaks (off-resonance conductance) is enhanced assisted by quantum fluctuations in the single-electron degenerate (usually spin-degenerate) levels.[3,5,4] This effect is closely related to the Kondo effect, which is well known in the literature.[6] Below the Kondo temperature[6] what determines which one of the above-mentioned phenomena dominates the conductance properties of the system is the ratio $\Delta\epsilon/\Gamma$, where Γ is the coupling strength to the leads. For $\Delta\epsilon/\Gamma \gg 1$, the mean-field picture is basically correct and CB physics dominates.[3] For $\Delta\epsilon/\Gamma \ll 1$, fluctuations can take over and so can Kondo-type physics.[3–5]

Although the CB phenomenon has been experimentally well established in quantum dots,[1] to our knowledge, the latter is yet to be observed in such systems, in part, due to the extremely low temperatures required. In this work we show that the presence of quantum fluctuations in the occupation of the electronic levels can also play a significant role close to the limit where CB dominates. From our results we conclude that there is a possibility of observing an enhancement of the off-resonance current through small quantum dots that carries a direct ''fingerprint'' of the discrete single-particle levels. Specifically, we address the problem on how the mean-field picture breaks down as $\Delta\epsilon/\Gamma$ crosses over from $\gg 1$ to $\gtrsim 1$. We find that in the regime $\Delta\epsilon/\Gamma \gtrsim 1$ quantum fluctuations in the single-particle levels create ''dynamical'' channels that are available for transport. In this limit, these new channels give rise to a multiple-peak structure in the off-resonance conductance in addition to the smooth signature of virtual tunneling processes, which is known as elastic cotunneling.[7]

We begin by considering a multilevel system connected to left and right leads described by the Hamiltonian

$$H=\sum_{i=1}^{N_L}\epsilon_i d_i^\dagger d_i+\sum_{k\in R,L}E_k c_k^\dagger c_k+\sum_{j>i=1}^{N_L}U_{ij}d_i^\dagger d_i d_j^\dagger d_j+\sum_{i=1}^{N_L}\sum_{k\in R,L}V_i(k)[c_k^\dagger d_i+d_i^\dagger c_k], \quad (1)$$

where $d_i^\dagger$ (d_i) are the creation (annihilation) operators associated with the N_L single-particle levels in the system with energies ϵ_i, $c_k^\dagger$ (c_k) are the ones for the levels in the left and right leads with energies E_k, and $V_i(k)$ are the hopping matrix elements between them. The third term in the Hamiltonian takes care of the electronic correlations within the system. Such a term contains the necessary contributions (those of the type density-density interaction) to study the most fundamental aspects of transport through quantum dots. (Additional terms might be added if one is interested in more detailed correlation effects,[8] but this lies beyond the scope of this work.) Degeneracies like those due to the spin degree of freedom are not considered either (a high magnetic field may be implicitly assumed).

The conductance g through the interacting system can be calculated with the formula[9]

$$g=\frac{2e^2}{h}\int_{-\infty}^{\infty}d\omega\ f'_{\rm FD}(\omega)\,{\rm Im}\{{\rm tr}[\,\gamma_{ij}(\omega)G_{ij}(\omega)]\},\qquad(2)$$

where $f'_{\rm FD}(\omega)$ is the derivative of the Fermi-Dirac distribution function, $\gamma(\omega)$ is the hopping matrix defined by $\gamma^R(\omega)\gamma^L(\omega)/[\,\gamma^R(\omega)+\gamma^L(\omega)]$, where

$$\gamma_{ij}^{R(L)}(\omega)\equiv -2\,{\rm Im}[\Sigma_{ij}^{R(L)}(\omega)]$$
$$=-2\,{\rm Im}\left[\lim_{\delta\to 0}\sum_{k\in R(L)}V_i(k)V_j(k)/(\omega-E_k+i\,\delta)\right],$$

$\boldsymbol{G}(\omega)$ is the retarded Green's function that must be calculated in equilibrium, and tr denotes the trace over the levels of the interacting region.

Equation-of-motion (EOM) techniques for calculating Green's functions have been used in the past in the context of the Anderson model[10] and recently also in the context of quantum dots.[5,11,12] According to such a technique one may write

$$\omega\langle\langle d_i;d_j^\dagger\rangle\rangle=\langle\{d_i,d_j^\dagger\}\rangle+\langle\langle[d_i,H];d_j^\dagger\rangle\rangle,\qquad(3)$$

where $G_{ij}(\omega)\equiv\langle\langle d_i;d_j^\dagger\rangle\rangle$. The higher-order Green's functions generated by the last term in Eq. (3) must be approximated at some stage of the calculation to obtain a closed set of equations. The simplest way to do that is in the Hartree-Fock approximation (HFA), where the higher-order Green's functions generated in the first EOM cycle are decoupled in the following way: $\langle\langle d_i n_j;d_i^\dagger\rangle\rangle\approx\langle n_j\rangle\langle\langle d_i;d_i^\dagger\rangle\rangle$, with $\langle n_j\rangle\equiv\langle d_j^\dagger d_j\rangle$. In this approximation the Green's function takes the simple form $\mathbf{G}(\omega)=[(\mathbf{G}^0(\omega))^{-1}-\mathbf{\Sigma}(\omega)]^{-1}$, where $\mathbf{G}^0(\omega)$ is the diagonal Hartree-Fock Green's function for the isolated system

$$G_{ii}^0(\omega)=\lim_{\delta\to 0}\frac{1}{\omega-\epsilon_i-\Sigma_j U_{ij}\langle n_j\rangle+i\,\delta},\qquad(4)$$

and $\mathbf{\Sigma}(\omega)=\mathbf{\Sigma}^R(\omega)+\mathbf{\Sigma}^L(\omega)$ is the total coupling self-energy. The Green's function projected on a level i depends on the occupation numbers $\langle n_j\rangle$ of all the other levels that are usually calculated self-consistently. Now, in order to include dynamical processes (which will turn out to be relevant in certain limits) in the Green's function, one has to improve upon the HFA. The way to do that in the framework of the EOM method consists in generating higher-order Green's functions from $\langle\langle d_i n_j;d_i^\dagger\rangle\rangle$, namely, $\langle\langle c_k d_j^\dagger d_i;d_i^\dagger\rangle\rangle$, $\langle\langle c_k^\dagger d_i d_j;d_i^\dagger\rangle\rangle$, $\langle\langle d_i n_j n_l;d_i^\dagger\rangle\rangle$, and $\langle\langle c_k n_j;d_i^\dagger\rangle\rangle$. Following Hewson and Zuckerman,[13] we neglect Green's functions such as $\langle\langle c_k d_j^\dagger d_i;d_i^\dagger\rangle\rangle$ and $\langle\langle c_k^\dagger d_i d_j;d_i^\dagger\rangle\rangle$, which contain unpaired operators. Physically, this corresponds to considering only one-electron processes. By generating new Green's functions from the remaining terms and successive decouplings similar to those mentioned in the HFA, a closed set of equations is obtained. We will refer to this approximation as the Hewson-Zuckerman approximation (HZA) from now on. After some lengthy algebra and excluding off-diagonal terms.[14] we find the following expression for the retarded Green's function:

$$\begin{aligned}G_{ii}(\omega)=\frac{1}{\omega-\epsilon_i-\Sigma_{ii}(\omega)}\Bigg[&1+\sum_{j=1,\neq i}^{N_L}\frac{U_{ij}\langle n_j\rangle}{\omega-\epsilon_i-U_{ij}-\Sigma_{ii}(\omega)}+\sum_{k>j=1,\neq i}^{N_L}\frac{U_{ij}U_{ik}\langle n_j n_k\rangle}{\omega-\epsilon_i-U_{ij}-U_{ik}-\Sigma_{ii}(\omega)}\\
&\times\left(\frac{1}{\omega-\epsilon_i-U_{ij}-\Sigma_{ii}(\omega)}+\frac{1}{\omega-\epsilon_i-U_{ik}-\Sigma_{ii}(\omega)}\right)+\sum_{l>k>j=1,\neq i}^{N_L}\frac{U_{ij}U_{ik}U_{il}\langle n_j n_k n_l\rangle}{\omega-\epsilon_i-U_{ij}-U_{ik}-U_{il}-\Sigma_{ii}(\omega)}\\
&\times\left(\frac{1}{[\omega-\epsilon_i-U_{ij}-\Sigma_{ii}(\omega)][\omega-\epsilon_i-U_{ij}-U_{ik}-\Sigma_{ii}(\omega)]}+\frac{1}{[\omega-\epsilon_i-U_{ij}-\Sigma_{ii}(\omega)][\omega-\epsilon_i-U_{ij}-U_{il}-\Sigma_{ii}(\omega)]}\right.\\
&+\frac{1}{[\omega-\epsilon_i-U_{ik}-\Sigma_{ii}(\omega)][\omega-\epsilon_i-U_{ik}-U_{ij}-\Sigma_{ii}(\omega)]}+\frac{1}{[\omega-\epsilon_i-U_{ik}-\Sigma_{ii}(\omega)][\omega-\epsilon_i-U_{ik}-U_{il}-\Sigma_{ii}(\omega)]}\\
&\left.+\frac{1}{[\omega-\epsilon_i-U_{il}-\Sigma_{ii}(\omega)][\omega-\epsilon_i-U_{il}-U_{ij}-\Sigma_{ii}(\omega)]}+\frac{1}{[\omega-\epsilon_i-U_{il}-\Sigma_{ii}(\omega)][\omega-\epsilon_i-U_{il}-U_{ik}-\Sigma_{ii}(\omega)]}\right)\\
&+\cdots\Bigg],\end{aligned}\qquad(5)$$

where additional terms containing products in U up to U^{N_L-1} are present, but are not shown here. As in the HFA, the occupancies $\langle n_j\rangle$ and correlation functions $\langle n_j\ldots n_l\rangle$ may be calculated self-consistently.

The HZA, as well as many other self-consistent EOM approximations beyond the HFA,[5,11,12] presents, however, a serious drawback: It gives unphysical values for the occupation numbers. Figure 1 illustrates this shortcoming in the simplest two-level case. As usual,[6] we take the total coupling self-energy independent of ω, equal for both levels, and purely imaginary $\Sigma_{jj}(\omega)=-i\Gamma$. We plot half the difference in the occupation numbers of the two levels as a function of $\Delta\epsilon/\Gamma$ when the chemical potential lies in between the singly and doubly occupied states (notice that this corresponds to plotting the magnetization for the usual symmetric Anderson model[6,10]). It is well known from other methods that the

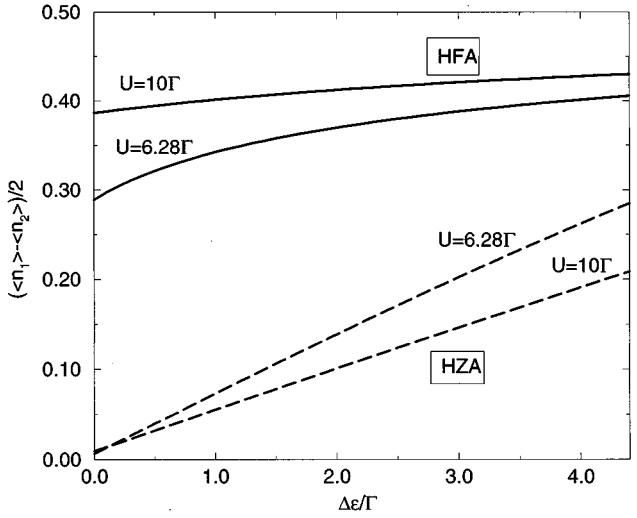

FIG. 1. Difference in the occupation numbers of the two levels in the symmetric case as a function of $\Delta\epsilon/\Gamma$. The HFA and the HZA are shown for two values of U. The HFA gives the correct limit for large values of $\Delta\epsilon/\Gamma$ when compared with, e.g., second-order perturbation theory in U (see the text). In the HZA, however, the difference is too small and decreases with U instead of increasing.

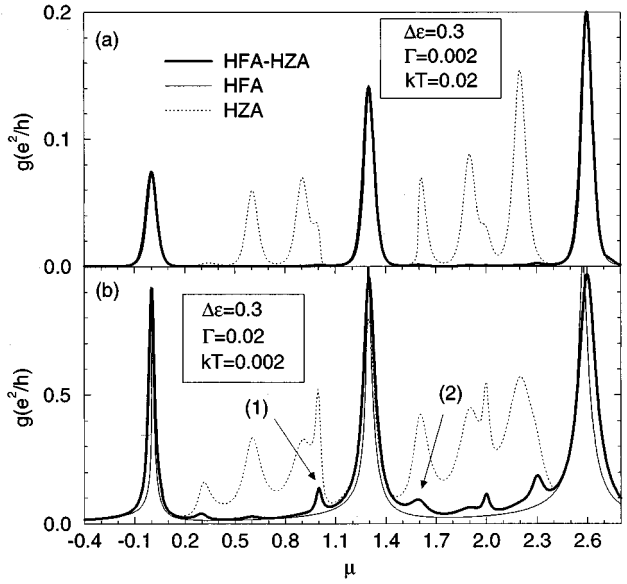

FIG. 2. Conductance in the HFA-HZA (thick solid line), in the HFA (thin solid line), and in the HZA (dotted line) as a function of μ in a five-level system up to $N=3$. We have set $\epsilon_1=0$. The other parameters are $\Delta\epsilon=0.3$, $U_{ij}=1$, (a) $kT=0.02$, $\Gamma=0.002$ and (b) $kT=0.002$, $\Gamma=0.02$ (all the magnitudes are in units of U). The smaller peaks in (b) correspond to dynamical channels opened by the fluctuating occupation numbers of the individual single-particle levels.

fluctuations in both levels are asymptotically suppressed as one increases $\Delta\epsilon/\Gamma$ and/or U/Γ and that eventually the HFA occupation numbers are basically correct.[3,6,15,16] For instance, second-order perturbation theories in U on top of the Hartree-Fock solution[15] give $(\langle n_2\rangle-\langle n_1\rangle)/2\approx 0.4$ for $\Delta\epsilon/\Gamma\approx 4$ and $U\approx 2\pi\Gamma$, which is what we obtain in the HFA. By contrast, the self-consistent HZA gives a much smaller value. Moreover, this value, instead of increasing with U/Γ, *decreases*. One way to get around this difficulty and to obtain realistic results out of the dynamical expression (5) is to avoid the fully self-consistent procedure. This can be effectively done by using the HFA static results for the occupation numbers and multiple-particle correlation functions in the expression (5) for the Green's function (we will hereafter call this approximation HFA-HZA). In this way, as we approach the isolated-system limit $\Gamma\to 0$, the correct occupancies are guaranteed while keeping open the possibility for fluctuations at finite hopping. We expect this approximation to give reliable qualitative results in the limit $\Delta\epsilon/\Gamma\gtrsim 1$. It cannot be valid, however, for $\Delta\epsilon/\Gamma\lesssim 1$ for several reasons: (i) The HFA breaks spontaneously the local symmetry when $U/\Gamma>1$ (see Fig. 1), (ii) off-diagonal elements have been ignored, and (iii) in order to reliably account for the strong fluctuation effects, which occur in that limit, one should have gone beyond the HZA in the EOM method.[10,11]

We now calculate the conductance of a five-level system where an increasing coupling of the levels to the leads $\Sigma_{jj}(\omega)=-i(\Gamma j)$ simulates a realistic situation for quantum dots. All the interaction terms U_{ij} are set to one. Figure 2 shows g vs μ in two different limits. When $\Delta\epsilon/\Gamma\gg 1$ [Fig. 2(a)] both the HFA and HFA-HZA give similar results: There is a peak in the conductance whenever $\mu\approx E(N)-E(N-1)\equiv\mu_{\rm dot}(N)$, where $E(N)$ is the ground-state total energy of N particles in the dot. The single-particle states are successively filled and remain fully occupied as μ moves up between the renormalized single-particle levels. As expected, due to the derivative of the Fermi-Dirac distribution function in Eq. (2), the heights and widths of the peaks are proportional to $1/T$ and T, respectively. In contrast, the level occupancies obtained from the HZA when off resonance [away from the charge-degeneracy points where $\mu\approx\mu_{\rm dot}(N)$] are noninteger numbers and the total charge in the dot is not perfectly quantized. (Detailed analysis will be given elsewhere.[17])

As Γ increases [Fig. 2(b)] the occupation of the single-particle levels is no longer either strictly zero or one as a function of μ. The mean-field picture rendered by the HFA is only approximately valid and quantum-mechanical fluctuations mediated by the interaction play their role now. In fact, within the HFA-HZA, new peaks appear in the off-resonance conductance as a consequence of these fluctuations. The presence of the peak labeled (1) at $\mu=1.0$ can be understood in the following way. There is a finite probability for the electron to be in any of the levels 1, 2, 3, 4, and 5. It spends most of its time in level 1, but if it happens to be in levels 2, 3, 4, or 5 a second, external electron can enter the system through level 1. Schematically these processes can be represented like $|\circ\bullet\circ\cdots\rangle\to|\bullet\bullet\circ\cdots\rangle\to|\circ\bullet\circ\cdots\rangle$, $|\circ\circ\bullet\cdots\rangle\to|\bullet\circ\bullet\cdots\rangle\to|\circ\circ\bullet\cdots\rangle$, and so on, where empty and filled dots represent empty and occupied states at a given time, respectively. Although the cost in energy for these processes is the same, $\epsilon_j+\epsilon_1+U_{1j}-\epsilon_j=1.0$, their likelihood decreases with j. Peak (2) at $\mu=1.6$ can be understood in a similar way. There is a small but finite probability for the system to have the level 2 empty even when $\mu>\mu_{\rm dot}(2)$. This is taken advantage of by an electron in the lead to sneak through via level 3. Schematically,

$|\bullet\circ\circ\cdots\rangle\rightarrow|\bullet\circ\bullet\cdots\rangle\rightarrow|\bullet\circ\circ\cdots\rangle$. The major peaks lie basically where the HFA predicts and all the other minor peaks can be associated with dynamical processes like those described above. [As can be seen, the self-consistent HZA overestimates the importance of these types of processes and gives rise to the spurious off-resonance structure seen in Fig. 2(a).]

It is worth mentioning that the tunneling processes described above seem to contribute to the off-resonance conductance in a way different from the usual elastic cotunneling,[7] which also anticipates a finite value of the off-resonance conductance. In such a theory, the contribution to the conductance comes from second- or higher-order (virtual) tunneling processes. Virtual transport is already included in the HFA, where, in addition, divergencies close to the resonances are automatically taken care of. As can be seen in Fig. 2(b), the elastic cotunneling is latent, for instance, in the slight asymmetry of the first major resonance. In agreement with previous work,[7] the $\Delta\epsilon^2/(\Delta\epsilon+U)^2$ dependence of the off-resonance conductance can also be obtained in the HFA. In Fig. 2(b), however, we see that the off-resonance value in the HFA is always a lower limit of the one obtained in the HFA-HZA.

Finally, we would like to stress the limits of validity of our results. At low enough temperatures, the off-resonance fine structure relies on the existence of small quantum fluctuations in the occupation of the electronic levels. As we see from Fig. 1, such fluctuations are suppressed when $U\gg\Delta\epsilon$ or $\Delta\epsilon\gg\Gamma$, but they survive if $U\gtrsim\Delta\epsilon$ and $\Delta\epsilon\gtrsim\Gamma$. Whereas the latter condition can be easily obtained by tuning gate voltages, the former one can only be generically found in relatively small quantum dots (diameter typically $\lesssim 0.1\ \mu$m). (Such quantum dots exist at present, but their transport properties have not been fully analyzed to date.) When $\Gamma\approx\Delta\epsilon$ one expects not only the fine structure to disappear, but the CB phenomenon altogether[18] (at least for a system with an infinite number of levels). This can be understood very easily. The CB disappears when the conductance of the insulating barriers, which keep the dot isolated from the leads, becomes of the order e^2/h. This is equivalent to saying that $\Gamma\rho_{\rm dot}\approx 1$, where $\rho_{\rm dot}$ is the density of states of the dot. Since, to a first approximation, $\rho_{\rm dot}\approx 1/\Delta\epsilon$, we obtain $\Gamma/\Delta\epsilon\approx 1$ as the condition for the disappearance of the CB peaks. This trend is also confirmed by our calculations in the HFA-HZA for five levels, as can be seen in Fig. 3. As the broadening of the single-particle levels approaches $\Delta\epsilon$, the charge quantization is lost progressively and, consequently, the distinctive CB peaks change into a smooth oscillation as a function of μ. However, as we discussed before, a quantitative analysis of this regime is beyond the scope of our present work.

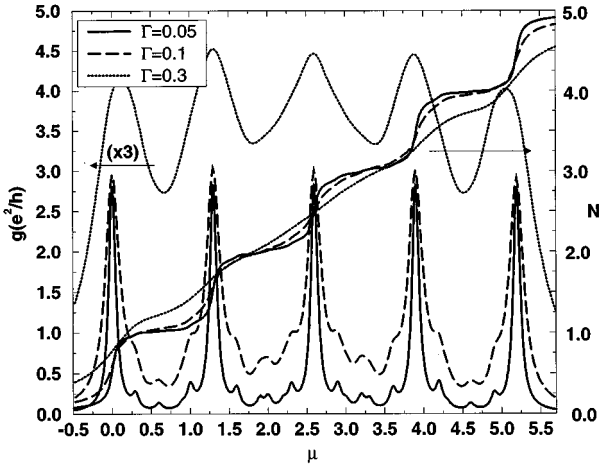

FIG. 3. Conductance in the HFA-HZA and total charge as a function of μ in a five-level system for different values of Γ (no dependence with the single-particle level has been considered now). $kT=0.005$ and all the other parameters are as in Fig. 2. As Γ increases, the fine structure starts disappearing as well as the overall CB effect. One can also see how the charge quantization is lost progressively.

In conclusion, an EOM that combines two different decoupling schemes has been used to calculate the conductance through a general multilevel system with strong interactions. A multiple-peak structure is obtained in certain limits. Whereas the main peaks are easily understood in terms of the orthodox Coulomb-blockade theory, quantum fluctuations must be invoked to explain the smaller peaks. The structure created by these small peaks in the off-resonance conductance lies on top of the contribution coming from virtual elastic tunneling.

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