Florida State University Libraries

Electronic Theses, Treatises and Dissertations

The Graduate School

2007

Electron Transport in Strongly Correlated Nanostructures

Khaled Al-Hassanieh

THE FLORIDA STATE UNIVERSITY

COLLEGE OF ARTS AND SCIENCES

ELECTRON TRANSPORT IN STRONGLY CORRELATED
NANOSTRUCTURES

By

KHALED A. AL-HASSANIEH

A Dissertation submitted to the
Department of Physics
in partial fulfillment of the
requirements for the degree of
Doctor of Philosophy

Degree Awarded:
Summer Semester, 2007

The members of the Committee approve the Dissertation of Khaled A. Al-Hassanieh defended on May 30, 2007.

Nicholas Bonesteel
Professor Directing Dissertation

Naresh Dalal
Outside Committee Member

Jorge Piekarewicz
Committee Member

David Lind
Committee Member

Jianming Cao
Committee Member

Approved:

David Van Winkle, Chair
Department of Physics

Joseph Travis, Dean, College of Arts and Sciences

The Office of Graduate Studies has verified and approved the above named committee members.

ACKNOWLEDGEMENTS

I will always look back to the years I spent at FSU and remember the friends I met and the rich experience I had. I would like to express my deep gratitude to my advisor, Professor Elbio Dagotto, for his constant support and encouragement. I am indebted to the invaluable guidance and the great work environment Elbio provided us with. I would also like to thank professors Adriana Moreo, Jorge Piekarewicz, Nick Bonesteel, and Naresh Dalal for all their help and support throughout my graduate years.

During my thesis work, I had the privilege of working with several great people. I want to thank my friends and collaborators, Carlos Büsser, Viacheslav Dobrovitski, Adrian Feiguin, Ivan Gonzalez, Fabian Heidrich-Meisner, George Martins, Roger Melko, Florentin Popescu, Jose Riera, Fernando Reboredo, Cengiz Sen, and Yucel Yildirim, for all the fruitful interaction and collaboration.

Most of all, I want to express my deep appreciation to my parents, my sister Haifa, and my brothers, Salah, Jalal and Naser, whose help, support and encouragement were beyond any limits. And last but not least, thanks to all my friends and cousins for all the companionship and encouragement. Special thanks to Mollie who has been a wonderful friend. Mollie, your dedication and sacrifice are a true inspiration.

— Khaled.

TABLE OF CONTENTS

List of Figures	vi
Abstract	xvii
1. INTRODUCTION	1
1.1 Motivation	1
1.2 Transport in Nanostructures: Resonant Tunneling and Coulomb Blockade	3
1.3 The Kondo Effect in Nanostructures	4
1.4 Key Experiments On Nanotransport	7
1.5 Overview of the Works Presented in this Dissertation	10
2. INTERFERENCE EFFECTS IN THE CONDUCTANCE OF MULTILEVEL QUANTUM DOTS	13
2.1 Introduction	13
2.2 One Dot with two levels: Formalism	16
2.3 Conductance Cancellations in Multilevel Dots	18
2.4 Integer-Spin Kondo Effect	26
2.5 Understanding the interference in the conductance	28
2.6 Conclusions	34
3. PREDICTION OF FERROMAGNETIC COUPLING IN COUPLED DOUBLE-LEVEL QUANTUM DOTS	36
3.1 Introduction	36
3.2 Model	37
3.3 Numerical Results	39
3.4 Conclusion	44
4. TRANSPORT PROPERTIES OF STRONGLY CORRELATED ELECTRONS IN QUANTUM DOTS USING A SIMPLE CIRCUIT MODEL	46
4.1 Introduction	46
4.2 Model Hamiltonian	47
4.3 Numerical Results	48
4.4 Circuit Model	50
4.5 Discussion	52
4.6 Conclusion	54

5. ELECTRON TRANSPORT THROUGH A MOLECULAR CONDUCTOR WITH CENTER-OF-MASS MOTION	56
5.1 Introduction	56
5.2 Model and Numerical Technique	57
5.3 Results	58
5.4 Conclusion	64
6. ADAPTIVE TIME-DEPENDENT DENSITY-MATRIX RENORMALIZATION GROUP TECHNIQUE FOR CALCULATING THE CONDUCTANCE OF STRONGLY CORRELATED NANOSTRUCTURES	66
6.1 Introduction	66
6.2 Model and Conductance Calculation	70
6.3 Non-Interacting Case	74
6.4 One Quantum Dot	81
6.5 Two Coupled Quantum Dots	89
6.6 Conclusions	90
7. CONCLUSION	92
A. THE EMBEDDED CLUSTER APPROXIMATION	94
REFERENCES	96
BIOGRAPHICAL SKETCH	106

LIST OF FIGURES

- 1.1 A schematic depiction of the two main nanosystems studied in this dissertation. Left: A lithographic semiconductor quantum dot (figure taken from Ref. [1]). The system consists of a two dimensional electron gas in a semiconducting heterostructure. On top of the heterostructure, metallic electrodes are deposited (grey color). By applying a negative voltage in these electrodes, electrons underneath are depleted and the system is divided into a quantum dot (labeled α) and two leads (labeled A and B). The voltage in gates I controls the tunneling between the dot and the leads, while the voltage in gate II controls the energy of the dot orbitals. Right: A schematic representation of the setup used for studying transport in single molecules (figure taken from Ref. [2]). The system consists of a molecule connected to two metallic electrodes, a source and a drain. A voltage difference is applied to these electrodes and the current through the molecule is measured. A third gate electrode is used to change the energy of the molecular orbitals. Note that tuning the energy levels and tunnel barriers is easier in quantum dots than in molecules. 3
- 1.2 (a) Energy diagram of the quantum dot in the regime $-U < \epsilon_d < 0$, that is, one electron occupies the dot (figure taken from Ref. [3]). The red areas represent the Fermi sea of the leads. Adding an extra electron to the dot is penalized an energy U , while removing the electron costs an energy $|\epsilon_d|$. However, an electron can tunnel into (out of) the dot, creating a virtual state with two (zero) electrons in the dot, and then tunnel back. This can result in a net spin flip of the electron in the dot. The coherent superposition of all such processes produces the Kondo effect which creates an extra resonance at the Fermi level as shown in (b). The resonance width is the energy scale of the Kondo effect, defined as the Kondo temperature T_K . Electrons can use this resonance as a transport channel thus dramatically enhancing the conductance. Note that the Kondo resonance is pinned to the Fermi level even if the energy of the dot orbital is varied using the gate voltage. 6

1.3	A schematic of the gate voltage dependence of the QD charge and conductance. The left panel shows the regime $T > T_K$. The conductance is dominated by Coulomb blockade: The charge is well defined due to the strong Coulomb interaction. This suppression of charge fluctuation results in a low conductance except at charge degeneracy points. The right panel shows the conductance when temperature is lowered below the Kondo temperature, $T < T_K$. In the gate voltage intervals where the dot occupation is odd, the plot shows plateaus of enhanced conductance due to the Kondo effect.	7
1.4	The main results of two important experiments reporting the observation of the Kondo effect in QDs. The left panel shows the conductance as a function of gate voltage at different temperatures (figure taken from Ref. [4]). At high temperature, the results show Coulomb blockade behavior (orange line). As temperature is decreased, the conductance in the even occupation regimes is suppressed (due to the suppression of thermal fluctuations), while in the odd occupation regimes, the conductance is enhanced due to the Kondo effect until it reaches the unitary limit ($2e^2/h$) for low enough temperatures. This was the first experiment to report the unitary conductance limit in the Kondo regime [4]. The right panel shows the differential conductance versus bias voltage for different temperatures and magnetic fields (figure taken from Ref. [1]). As the temperature is lowered, a peak is observed at zero bias (zero-bias anomaly). This peak is split upon the application of a magnetic field due to Zeeman effect. These results are the “smoking gun” for the Kondo effect in nanotransport. This experiment was the first observation of the Kondo effect in a QD [1].	8
1.5	Main results of the experiment by Craig <i>et al.</i> on nonlocal spin control in quantum dots [5]. The inset shows a schematic of the system studied (see also Fig. 4.1). The two quantum dots are connected by an open central region. Current is measured through QD1 as indicated by the thin arrow for different occupations of the other dot (QD2), and different couplings between QD2 and the open region as indicated by the thick arrow. The left figure shows the results for odd occupation of QD2. As the coupling is increased, the zero-bias anomaly in QD1 is split. The right figure shows the results for even occupation where increasing the coupling strength slightly reduces the conductance of QD1.	9
2.1	Schematic representation of the main system studied in this chapter, consisting of one quantum dot with two levels α and β . The two possible paths carrying different phases Φ_α and Φ_β are shown.	16

2.2	Conductance for five different values of ΔV . The conductance (in units of $2e^2/h$) is shown in solid lines and the phase difference (divided by π) in dotted lines. When ΔV is the largest energy ($\Delta V=2t$ (e)), two structures similar to those found in a single-level QD, separated by ΔV , appear in the conductance. Case (d) is similar, but since ΔV is not large enough, an extra thin peak appears around $V_g = -1$ (this peak is shown with more detail in Fig. 2.8). When ΔV decreases, these structures begin to interact giving rise to conductance dips, as shown in (b) and (c) for the cases $\Delta V=0.2t$ and $\Delta V=0.5t$ ($\Delta V=0.2t$ is studied in more detail in Fig. 2.4). Finally, in the limit $\Delta V=0$, the conductance dips transform into discontinuities.	19
2.3	Conductance (in units of $2e^2/h$) in a narrow range of V_g and ΔV . The discontinuity at $\Delta V=0$ is transformed into a dip for finite values of ΔV	21
2.4	Conductance (in units of $2e^2/h$) (a), phase difference (divided by π) (b), and partial conductances (c) for $\Delta V=0.2t$. The dips, indicated by “a” and “b” in (a), are in regions where the phase difference is π producing exact cancellations when $\sigma_\alpha=\sigma_\beta$	22
2.5	Conductance (in units of $2e^2/h$), phase difference, and partial conductances for $\Delta V=0.2t$ around the “b” dip of Fig. 2.4(a). Thick arrows show the gate potential where the conductance vanishes, $\Delta\Phi = \pi$, and $\sigma_\alpha = \sigma_\beta$	22
2.6	(a) Conductance (in units of $2e^2/h$), (b) total charge of the QD, and (c) charge of the individual levels for the case $\Delta V=0.2t$. The central peak emerges in the regime where two electrons occupy the dot, one in each level.	23
2.7	Conductance (in units of $2e^2/h$) and charges as a function of the gate potential for $\Delta V=t$. These results should be compared to those in Fig. 2.6	23
2.8	Conductance (in units of $2e^2/h$), phase difference and partial conductances for $\Delta V=t$ around the central peak shown in Fig. 2.2(d). The partial contributions to the conductance (σ_α and σ_β) have the same shape as in Fig. 2.4 but now the phase difference between them is zero. This phase difference creates a peak whereas in the other case a dip appears.	24
2.9	Conductance convergence with the size of the exactly-solved cluster L for the case $\Delta V=0.2t$. Dotted lines are for $L = 4$, continuous thin lines for $L = 8$, and dashed lines for $L = 12$. Extra points for $L = 16$ and $L = 20$ are shown with crosses and diamonds, respectively.	25
2.10	Conductance and total charge when U' and J are not active. The results are shown are results for $U'=J=0$ (solid line) compared to the previous results for $U'=2/3U$ and $J=U - U'$. $\Delta V=0.35$ for both plots. The behavior is qualitatively the same in both cases.	25

2.11	Conductance (in units of $2e^2/h$) for large $U'=2t$. (a) shows the case of ferromagnetic Hund coupling $J= U - U' $ while (b) shows the case of anti-ferromagnetic $J=U - U'$. $\Delta V=0$ in both cases.	26
2.12	Schematic representation of the different scenarios that can occur in a two-level QD as the gate voltage changes. See the text for more details.	27
2.13	Conductance (in units of $2e^2/h$) for different values of J . (a) corresponds to $J=U - U'$, (b) to $J=0$, and (c) to a large negative $J=-0.5t$. The central peak disappears when the ferromagnetic spin correlations are eliminated at even more negative J . More details can be found in the text.	29
2.14	Density-of-states of the bare $L = 4$ cluster, for $\Delta V=0.05t$ and $V_g=-1.082t$. The Fermi level lies at $\omega=0.0$. Note the relative change of the weight <i>signs</i> of the poles near $\omega = 0$, when comparing the Green functions $g_{\alpha r}$ and $g_{\beta r}$	30
2.15	Comparison between the imaginary parts of $g_{\alpha\alpha}$ and $g_{\alpha r}$, for $\Delta V=0$ and $\Delta V=0.05t$. The pole in $g_{\alpha N}$ near the Fermi level disappears (i.e. carries no weight) at $\Delta V=0$. ($V_g = -1.082t$ and $L = 4$ in both cases.)	31
2.16	Asymmetric α - β case, where the Coulomb repulsions in the two levels are different i.e. $U_\beta=U_\alpha + 0.1t$. Dips in the conductance are observed even for $\Delta V=0$, due to the explicitly broken symmetry. More details can be found in the text.	32
3.1	Schematic representation of the two double-level QDs system and the hopping amplitudes of Eqs. (1) and (3).	37
3.2	Results for the strong inter-dot tunneling regime ($t''/t' \gg 1$) showing the presence of a <i>ferromagnetic</i> state at 1/4-filling and concomitant Kondo effect: (a) Conductance (solid line), charge occupancy per level and per spin (dashed line), and total spin S_T in the four levels of the dots (dotted line) vs. gate potential V_g for two coupled double-level QDs. The values for the parameters are indicated. Note that the maximum in the conductance corresponds to a spin 1 Kondo peak that occurs when there is exactly one electron per dot and the total spin $S_T \approx 1.0$. The other two peaks occur when there are 1 and 3 electrons in the two-dots system, corresponding to $S_T = 1/2$ Kondo resonances. (b) DOS indicating the presence of a Kondo resonance associated with the $S_T = 1$ Kondo peak in (a). The numbers on the right side indicate the total number of electrons inside the two quantum dots. V_g takes the values 0.0, -0.5 , -0.83 , -1.12 , -1.5 from the top to the bottom curve. The Fermi level is located at $\omega/t = 0.0$	39

3.3	(a) Conductance G vs. gate potential V_g for different values of inter-dot tunneling ($t'' = 1.0, 0.6, 0.4, 0.2$, and 0.08 , see convention in (b)). V_g varies across the region where the fourth electron is charged into the double-dot system. Note that the conductance at $V_g = -5.0$ gradually increases from zero (at strong tunneling, $t'' = 1.0$) to the maximum value (at weak tunneling, $t'' = 0.08$). This variation indicates a transition from two $S = 1$ spins (in each dot) forming a global singlet to two uncorrelated $S = 1$ spins, each forming a Kondo resonance with the lead conduction electrons. The inset shows the variation with t'' of the total spin inside the dots. (b) Variation with t'' of the spin-spin correlation between the total spin in the double-dot and a conduction electron located in the first site of the leads.	41
3.4	Variation of the conductance with level separation ΔV . The level separation is indicated for each curve. (a) $t'' = 1.0$: The gate potential decreases down to the particle-hole symmetric value, to highlight how the $S_T = 1$ Kondo peak varies with ΔV . Note that all graphs have the same horizontal axis scale, except for the upper one (scale indicated on top). (b) $t'' = 0.08$: The gate potential varies down to the lowest value (total charging of the dots), to highlight the variation of the central peak (at the particle-hole symmetric point). Here also the upper graph has a different horizontal scale. A discussion of how the data interpolate between double- and single-level dots is given in the text.	42
3.5	Qualitative phase diagram for the strong inter-dot tunneling regime ($t''/t' \gg 1$). Horizontal axis indicates the level occupancy (controlled by V_g) and vertical axis indicates the level separation (controlled by ΔV). In the $1/2$ -filling region, displayed in the left side, one goes from an AF coupling between two $S = 1$ spins (for $\Delta V < V_g$) to a situation where the two lower levels are completely occupied (for $\Delta V > V_g$). In both cases there is no Kondo effect. On the other hand, in the $1/4$ -filling region (right side), for $\Delta V < t''$ one has the novel $S_T = 1$ FM Kondo region, which gives way to an AF region (with no Kondo effect) once $\Delta V > t''$. The regime with 3 electrons in the two dots is very narrow as a function of V_g and it is not shown.	43
3.6	Schematic representation of the main result in this chapter.	44
4.1	(a) Experimental setup used in Ref. [5]. (b) Illustration of the model studied in this work (see text for details).	47

4.2	(a) Variation of G with V_{g1} in QD1 (solid curves) and variation of $\langle n_2 \rangle$ (occupancy of QD2 per spin orientation - dashed curves) for $t'' = 0.2$ and three different values of V_{g2} . For $V_{g2} = -2.0$ (red), QD2 is occupied by 2 electrons ($\langle n_2 \rangle = 1$) for any V_{g1} and the conductance through QD1 is essentially the same as if QD2 was not present. For higher values of V_{g2} , the average value of $\langle n_2 \rangle$ decreases and becomes dependent on V_{g1} (decreasing for lower values of V_{g1}). This is accompanied by a suppression of the ZBA (for -0.35 (green)) and also by a splitting of the ZBA (for $V_{g2} = -0.3$ (blue)). (b) Variation of G and $\langle n_2 \rangle$ with t'' (0.2, 0.4, 0.5) at a fixed value of $V_{g2} = -0.5$. As the value of t'' increases, the average value of $\langle n_2 \rangle$ decreases and this is again accompanied by a suppression of the ZBA. (c) Same as in (b), but now for $V_{g2} = -0.25$ (particle-hole symmetric point) and $t'' = 0.0, 0.1$, and 0.2 . Note that G vanishes at $V_{g1} = -0.25$, where $\langle n_2 \rangle = 0.5$, for all finite values of t'' .	49
4.3	Schematic representation of the main ideas behind the “circuit model”. In (a), the system represented in Fig. 4.1b is divided into its constituent elements: QD1 is modeled as a QD connected in series with the leads and QD2 is modeled as a side-connected QD. The curves in (b) represent the conductance and occupancy of each separate circuit element vs. the applied gate potential. (c) Schematic representation of how the two individual elements are connected to form the final ‘circuit’: Incident and reflected wave amplitudes are represented in the right side of QD2 by black arrows. A transmitted wave through QD2 undergoes multiple reflections between the two quantum dots until it is finally transmitted past QD1. The superposition of all these processes results in the final conductance for the ‘circuit’.	50
4.4	Same as in Fig. 4.2, but now using the ‘circuit model’ for the calculations.	52
4.5	(a) Spin correlation $\mathbf{S}_1 \cdot \mathbf{S}_2$ between QD1 and QD2 for the same parameters as in Fig. 4.2c. For $t'' = 0.0$ (red curve), the two QDs are uncorrelated ($\mathbf{S}_1 \cdot \mathbf{S}_2 = 0$). For finite t'' (0.1 (green) and 0.2 (blue)), $\mathbf{S}_1 \cdot \mathbf{S}_2$ is FM and reaches its maximum value in the region where G is maximum. (b) Kondo correlations $\mathbf{S}_1 \cdot \mathbf{S}_c$ between QD1 and the central site for the same parameters as in (a). All values are AF and they decrease in amplitude as t'' increases, underscoring the decrease of the Kondo effect as the FM correlation between QD1 and QD2 increases (compare with (a)). (c) Variation of G as U_1 (Hubbard interaction in QD1) assumes the values 0.4, 0.2 and 0.0. Note that the dip in G becomes slightly narrower as U_1 decreases, however it does not disappear.	53
5.1	A schematic of the system studied in this chapter. The molecule can oscillate between the two electrodes around the equilibrium position l_0 , thus modulating the tunnel barriers.	57

5.2	(a) NRG and (b) ECA results for G as a function of V_g in the Kondo Regime for $\alpha = 0$ and increasing the electron-phonon coupling strength ($\lambda/\omega_0 = 0.0, 0.4, 0.8$, and 1.2). For small λ/ω_0 , the standard Kondo resonance is obtained with renormalized parameters. In the limit of large λ/ω_0 , the charge Kondo effect is obtained.	59
5.3	(a) G as a function of V_g in the Kondo regime for $\alpha = 0$ (solid line) and $\alpha = 0.4$ (dashed line), $\lambda = 0.2$ in both cases. In the first case, the usual Kondo peak with reduced width is obtained. In the second case, a conductance dip is obtained. (b) Average occupation of the molecular level for the same set of parameters. Note that the charging behavior is almost the same in both cases. (c) G vs. V_g in the absence of Coulomb repulsion ($U = 0$), for $\alpha = 0$ (solid line) and $\alpha = 0.4$ (dashed line), $\lambda = 0.1$ in both cases. The conductance dip is also obtained in this case, thus the physical mechanism behind this effect does not depend on the electron-electron interaction.	60
5.4	G as a function of V_g in the Kondo regime for $\alpha = 0.4$ and $\lambda = 0.15, 0.20$, and 0.25 (dashed, dotted and solid lines respectively). The dip becomes more pronounced as λ increases. The inset shows the convergence of the results with the maximum number of phonons (N_{ph}). Note that the qualitative effect of conductance cancellation is preserved all the way down to $N_{\text{ph}} = 1$	61
5.5	(a) A schematic representation of the two conductance channels, the purely electronic tunneling represented by the two upper arrows and the “phonon-assisted tunneling” channel represented by the two lower arrows. (b) Partial conductance when only one of the channels is active. The dotted line shows the conductance of the purely electronic tunneling channel G_E , while the dashed line shows the conductance of the “phonon-assisted tunneling” channel G_I . (c) Conductance when both channels are active. (d) Phase difference $\Delta\Phi$ of the two channels. Note that $\Delta\Phi$ for all values of V_g is close to π thus leading to destructive interference. In particular, for $V_g = -U/2$, $\Delta\Phi = \pi$ and $G_E = G_I = 2e^2/h$, thus leading to a perfect cancellation in the overall conductance. ($\lambda = 0.2$, $\alpha = 0.4$).	62
5.6	The solid line shows the conductance of the molecule when a breathing vibrational mode is active (no CM motion). The particle-hole symmetry is broken as expected and no conductance dip is obtained. The dashed line shows the conductance when both breathing and CM vibrational modes are active. The combined effects of the two modes lead to a Fano-like interference. The breathing mode parameters used are $\lambda' = 0.2$, $\omega'_0 = 0.3$ and $\alpha' = 0.3$	63

5.7	(a) Convergence of the conductance with the size of the exactly-solved cluster L . The solid line, dashed line, and the circles show the results obtained using $L = 3, 7$, and 11 respectively. In the three cases, a maximum number of three phonons was used. (b) The real and (c) imaginary parts of the isolated cluster Green function g_{lr} that propagates an electron from the left to the right ends of the cluster ($L = 3$ and $V_g = -U/2$). Note that both parts are equal to zero at the Fermi level (located at $\omega = 0.0$). Thus, the origin of the conductance dip can be traced back to the exactly-solved cluster. $\lambda = 0.2$ and $\alpha = 0.4$ in the three figures.	64
6.1	Schematic representation of the geometry used in our study. The leads are modeled by tight-binding Hamiltonians. The ground state at time zero is calculated at zero bias. Then, a finite bias ΔV is applied between the two leads (without ramping time for simplicity, but this can be changed in future studies) and the resulting current is measured.	71
6.2	Exact results for $J(t)/\Delta V$ (in units of e^2/h) vs. time (in units of $\hbar/t_{\text{leads}}$), for the non-interacting 1QD case obtained with clusters of different lengths (L) and $\Delta V=0.001$. (a) $J(t)/\Delta V$ obtained with a large cluster ($L=402$). $J(t)/\Delta V$ shows clear steady-state plateaus at $\pm 2e^2/h$. The periodic changes in the current direction are caused by its reflection at the open boundaries of the cluster. (b) $J(t)/\Delta V$ obtained with decreasing L . The steady-state plateau is obtained even with $L = 32$. The current is quasi-periodic with a period proportional to L . The parameters used are $V_g=U=0$ and $t'=0.4$. . .	75
6.3	This figure shows the propagation of the current in the cluster after the bias $\Delta V=0.001$ is applied at $t=0$. The different panels show the current as a function of position x along the chain, at different times t . The size is $L = 402$ and the dot is at the site 201. The results were obtained exactly, since the Hubbard couplings are zero.	76
6.4	DMRG results compared to the exact results for $J(t)/\Delta V$ obtained using different clusters L and number of states M , with $\Delta V=0.001$. (a) $L = 96$, (b) $L = 64$, (c) $L = 32$, and (d) $L = 16$. Note that for $L = 96$ and 64 , $M = 200$ shows good qualitative agreement and $M \geq 300$ even shows good quantitative agreement with the exact results. For $L = 32$ and 16 , $M = 200$ and 100 already show excellent quantitative agreement with the exact results. . .	77

6.5	(a) DMRG and exact results for G vs. V_g in the case of one non-interacting QD, and $t' = 0.4$ (with $\Delta V=0.001$). The DMRG results are obtained with $L = 64$ and $M = 300$. In this case, G is obtained from the value of the steady-state current plateau. The exact results are for infinite leads. The results show a resonant tunneling peak of $\text{FWHM} = 4t'^2$ at $V_g = 0$. (b) $J(t)/\Delta V$ for $V_g = \pm 0.3$ and $\Delta V = 0.01, 0.001$. Note that decreasing the bias voltage reduces the asymmetry between positive and negative V_g and gives a better steady-state plateau.	78
6.6	(a) DMRG results compared to the exact results in the case of one non-interacting QD for several intermediate and large values of ΔV , namely exploring the influence of a finite bias in the calculations. The parameters used are $V_g = U = 0$, and $t' = 0.4$. Both DMRG and exact results are obtained with a cluster $L = 32$, using $M = 200$ states, for the DMRG results.	79
6.7	The results obtained with the Suzuki-Trotter approach and the static Runge-Kutta method compared to the exact results for (a) $\Delta V = 0.001$ and (b) $\Delta V = 0.5$. The parameters used are $V_g = U = 0.0$, $t' = 0.4$, and $L = 64$	80
6.8	DMRG and exact results for G vs. V_g in the case of 2 coupled non-interacting QD's. The DMRG results are obtained with $L = 64$ and $M = 300$ ($\Delta V=0.001$). The exact results are for infinite leads. The cases (a) $t'' = 0.5$ and (b) $t'' = 0.2$ are investigated, with $t' = 0.4$ in both cases. The results present the bonding and anti-bonding resonant tunneling peaks at $\pm t''$	80
6.9	DMRG local density-of-states results for (a) 1QD and (b) 2QD's compared to the exact (bulk) results. The parameters used are $V_g = U = 0$, $t' = 0.4$ in both cases and $t'' = 0.5$ in (b). In (a), a broadening imaginary component $\eta = 0.1$ was introduced. In (b), $\eta = 0.15$. Smaller values of η would reveal the discrete nature of the LDOS obtained with DMRG on a finite $L=64$ system.	81
6.10	$J(t)/\Delta V$ in the case of one quantum dot for different cluster lengths L . The parameters used are $U = 1.0$, $t' = 0.4$, $\Delta V=0.005$, and $V_g = -0.5$. As L increases, the conductance approaches the unitarity limit ($2e^2/h$) due to the Kondo screening effect.	83
6.11	$J(t)/\Delta V$ in the case of one interacting QD for different values of V_g , and with $\Delta V=0.005$. The value of the conductance is obtained by averaging the current over an interval of time, corresponding to the steady state. The solid horizontal lines represent this time interval over which the average of the current is taken, and the value of the average. The parameters used are $U = 1.0$, $t' = 0.4$, $L = 128$, and $M = 300$	83

6.12	$J(t)/\Delta V$ in the case of one interacting QD for $V_g/U = -1.5$, $U = 1.0$, $t' = 0.4$, $L = 128$, and different values of M . Note that as M increases, the oscillations at long times tend to decrease.	84
6.13	Conductance G and the dot occupation $\langle n_d \rangle$ for one interacting QD. The circles show G obtained by averaging the current over an interval of time corresponding to the steady state, as shown in Fig.6.11. The squares show G obtained from $\langle n_d \rangle$ using the Friedel sum rule (FSR). G has the shape of the expected Kondo or mixed-valence plateau centered at $V_g = -U/2$. The feature would be sharper reducing t' . Results are shown for: (a) $U = 1.0$, $t' = 0.4$, and (b) $U = 2.0$, $t' = 0.5$. In both cases $L = 128$ and $M = 300$. Note that the DMRG conductance results in (a) show a slightly better agreement with the FSR results. This is expected since the finite size effects are stronger for larger U	85
6.14	Conductance G ($\Delta V=0.001$) and charge at the dot $\langle n_d \rangle$, for one interacting QD in the case of large U . The circles show G obtained from the DMRG procedure outlined in this paper, while the squares show G obtained from $\langle n_d \rangle$ using the Friedel sum-rule. The finite-size effects are obvious here, since the results are halfway between the expected Kondo plateau (properly reproduced by the FSR procedure) and the Coulomb blockade peaks. This case is shown as an illustration of important size effects in some limits. The parameters used are $U = 4.0$, $t' = 0.4$, $L = 128$, and $M = 300$	86
6.15	Finite-size scaling of the conductance G at $V_g = -U/2$ for the odd-1QD-even (circles) and even-1QD-even (triangles) clusters. Note that in both cases G converges to 1 in the bulk limit. However, the odd-1QD-even cluster converges faster, which makes it the most useful for practical calculations. The parameters used are $U = 1.0$ and $t' = 0.4$	88
6.16	Finite size scaling of G for odd-QD-even clusters using OBC (circles) and DBC (triangles). $d = 0.5$ was used. Note that $L = 64$ cluster with DBC gives better results than a $L=128$ cluster with OBC. The same parameters are used as in Fig.6.15.	88
6.17	Conductance G vs. gate voltage V_g in the case of one interacting QD ($U = 2.0$, $t' = 0.5$) for different values of the magnetic field B (and $\Delta V=0.001$). For $B = 0$, a Kondo plateau is obtained, centered at $V_g = -U/2$. As B increases, the Kondo effect is suppressed, and for moderate B , two Coulomb blockade peaks are observed at $V_g = -U$ and $V_g = 0$, as expected.	89
6.18	$J(t)/\Delta V$ for two coupled QDs at $V_g = -U/2$ for different values of t''/t'^2 (and $\Delta V=0.005$). As in the case of one interacting QD, the conductance is obtained by averaging the steady-state current over the indicated intervals. The parameters used are $U = 1.0$, $t' = 0.5$, $L = 127$, and $M = 300$	90

6.19	Conductance G as a function of t''/t'^2 , at $V_g = -U/2$ ($\Delta V=0.005$). In this regime, G is determined by the competition between the Kondo correlation of each dot with the neighboring leads and the antiferromagnetic correlation between the two dots. The circles represent our DMRG results obtained with $L = 127$ and $M = 300$. The solid line is the plot of the functional form obtained by Georges and Meir using SBMFT [6].	91
------	--	----

ABSTRACT

We present the results of our numerical studies on the transport properties of strongly correlated nanostructures, particularly quantum dots and single molecules. The main focus is on correlation, interference and phononic effects. Interesting interferences are observed in multilevel quantum dots, and under the appropriate conditions, a novel ferromagnetic phase is observed in coupled double-level quantum dots at quarter filling. Our simulations of experiments involving nonlocal spin control provide more insight of the experimentally observed results. In the case of single molecules, our study of phonon effects reveals that the center-of-mass motion opens a new channel for transport. This channel can interfere destructively with the purely electronic channel leading to a conductance dip. Finally, we propose a new technique to study nanotransport based on the adaptive time-dependent density-matrix renormalization group. The technique is tested for different cases and is very promising particularly in the nonequilibrium case where most other techniques cannot be applied.

CHAPTER 1

INTRODUCTION

1.1 Motivation

1.1.1 Many-body Physics in a Tunable Environment

Understanding the behavior of strongly correlated electronic systems and the consequent utilization of their properties in technological applications lie at the heart of condensed matter physics. For several decades, there has been a vast experimental and theoretical effort to unravel the complicated behavior of such systems. In most materials, many effects compete in a highly nontrivial way, resulting in a very rich physical behavior. This includes phase diagrams with a plethora of phases and interesting electronic, transport and magnetic properties. Such effects include kinetic energy, Coulombic interactions, lattice disorder, and electron-lattice interaction to mention a few.

Several major difficulties make the study of strongly correlated systems extremely challenging and interesting. From the theoretical point of view, the strongly correlated nature of the systems renders the standard perturbation treatments and first-principle calculations inapplicable. In addition, the presence of several competing effects makes the theoretical investigations rather complicated. From the experimental point of view, such bulk systems are not well controlled. These materials spontaneously grow in a specific lattice, magnetic, charge, and orbital order dictated by the competing effects. Changing the parameters of the system in this case is very difficult and can be done in discrete steps only. In other words, these bulk systems cannot be well tuned. In addition, the system parameters can not always be determined, making the quantitative comparison with theory rather difficult.

For all these reasons, the first experimental observation of a celebrated many-body effect, the Kondo effect, in a well-controlled mesoscopic system, a lithographic semiconductor quantum dot (QD), captured the attention of the condensed matter community; although the

Kondo effect in metals had been experimentally observed and well understood for decades. Among many, some of the advantages of studying mesoscopic systems, as opposed to bulk systems, are: (a) Many of the parameters are easily tuned. (b) The study is performed using a well-isolated and controlled sample, as opposed to a large number of randomly located magnetic impurities, as in the example of the Kondo effect in metals. (c) The impurities can be located at well controlled distances with well-tuned interactions. And (d) the measurements can be done under non-equilibrium conditions.

Following the observation of the Kondo effect in a QD [1], there have been several interesting studies on more elaborate systems that show competing tendencies or more exotic types of the Kondo effect. A competition between the Kondo effect and antiferromagnetic correlations is observed in coupled QDs [7], an integer-spin Kondo effect is observed in multilevel dots [8], the orbital Kondo effect is observed in carbon nanotube QDs [9], and recently a two-channel Kondo effect was observed in semiconductor QDs [10].

1.1.2 Quantum Computing

Quantum dots are very promising candidates for the implementation of quantum computing: a spin in a QD is a natural two-state quantum system. In the Loss-DiVincenzo proposal [11], a system composed of two coupled QDs has been proposed to carry out quantum computing. The qubits are encoded by the state of the electron spins localized in the dots. The single-qubit quantum gates are realized by rotating the spins using external magnetic fields; and the two-qubit gates are implemented by tuning the voltage in the gate connecting the two dots. The nuclear spins in the dots are proposed as a long-term memory, the read and write processes are executed by mapping the coherence of the electron spin onto the collective state of the nuclear spins [12, 13].

However, due to the interaction of the electronic spin with the environment, the electron loses its coherence (decoherence) leading to the loss of encoded information. At low temperature, the main source of decoherence is the hyperfine interaction of the electron spin with the surrounding spin bath. This has been the subject of numerous studies [14, 15, 16, 17, 18, 19].

Figure 1.1: A schematic depiction of the two main nanosystems studied in this dissertation. Left: A lithographic semiconductor quantum dot (figure taken from Ref. [1]). The system consists of a two dimensional electron gas in a semiconducting heterostructure. On top of the heterostructure, metallic electrodes are deposited (grey color). By applying a negative voltage in these electrodes, electrons underneath are depleted and the system is divided into a quantum dot (labeled α) and two leads (labeled A and B). The voltage in gates I controls the tunneling between the dot and the leads, while the voltage in gate II controls the energy of the dot orbitals. Right: A schematic representation of the setup used for studying transport in single molecules (figure taken from Ref. [2]). The system consists of a molecule connected to two metallic electrodes, a source and a drain. A voltage difference is applied to these electrodes and the current through the molecule is measured. A third gate electrode is used to change the energy of the molecular orbitals. Note that tuning the energy levels and tunnel barriers is easier in quantum dots than in molecules.

1.2 Transport in Nanostructures: Resonant Tunneling and Coulomb Blockade

In many nanostructures, such as single-molecule conductors and quantum dots, the system consists basically of a central interacting region, usually the dot or molecule, weakly coupled to ideal noninteracting leads. Due to the confined nature of the nanostructures, the energy spectrum is discrete with several energy levels or quasilocal states. The hybridization of these levels with the continuum states of the leads gives the levels a finite width which is proportional to the rate of electron escape to the leads. A rough model of the dot, for example, is a quantum well separated from the leads by two tunnel barriers. The position of the dot energy levels can be controlled by applying a voltage in the gate electrode. The

conductance of the dot is suppressed except when one of the levels crosses the Fermi level of the leads. In other words, the conductance versus gate voltage shows a collection of peaks each corresponding to a state on the dot crossing the Fermi level. The width of these peaks is proportional to the width of the levels. This is known as *resonant tunneling*. In the rest of the introduction, only one energy level is assumed to be relevant. The other levels are assumed to be of either too high or too low energy to contribute to transport.

In addition to the discrete spectrum, in many nanosystems, the Coulombic interaction is dominant. This causes the dot to have a well defined number of electrons. Adding an electron to the dot is prevented by this strong interaction. By changing the gate voltage, the energy of the dot with N electrons changes. At specific degeneracy points, the dot states having N and $N + 1$ electrons are degenerate. This allows for charge fluctuations and, thus, electron transport. The conductance versus gate voltage shows a very low conductance except at charge degeneracy points. This is known as the *Coulomb blockade*.

1.3 The Kondo Effect in Nanostructures

Before explaining the Kondo effect, I will first describe briefly the Anderson impurity model,

$$H = \epsilon_d n_d + U n_{d\uparrow} n_{d\downarrow} + \sum_{k\sigma, \alpha=L,R} (V d_{\sigma}^{\dagger} c_{k\sigma, \alpha} + H.c.) + \sum_{k\sigma, \alpha=L,R} \epsilon(k) c_{k\sigma, \alpha}^{\dagger} c_{k\sigma, \alpha}. \quad (1.1)$$

The first term is the energy of the dot level, the second term is the Coulomb interaction between two electrons of opposite spins occupying the dot, the third term is the hybridization between the dot levels and the continuum states in the leads, and the fourth term is the Hamiltonian of the leads.

This model is particle-hole symmetric at $\epsilon_d = -U/2$. Due to the hybridization of the dot with the leads, the energy levels gain a width $\Gamma = 2\pi V^2 \rho(\epsilon_F)$ (the factor 2 corresponds to the presence of two leads), where $\rho(\epsilon_F)$ is the density of states of the leads at the Fermi energy, ϵ_F . In the following and the rest of the dissertation, we set ϵ_F to be zero. In general, the Coulomb interaction is the dominant energy scale, i.e. $U \gg \Gamma$. This causes the charge on the dot to be well defined: Zero, one, and two electrons occupy the dot for $\epsilon_d > 0$, $-U < \epsilon_d < 0$, and $\epsilon_d < -U$ respectively. The charge transition occurs at $\epsilon_d = -U, 0$ with a width Γ . Around these transition points, the dot is in the so-called mixed valence regime, that is, a combination of two charge states. Note also that for $-U < \epsilon_d < 0$, one electron

with a net spin occupies the dot, thus the dot has a magnetic moment. This is called the local moment regime.

Based on the reasoning presented before, the conductance of the Anderson impurity model should show a Coulomb blockade behavior. That is, the conductance should be suppressed for all values of gate voltage except for the charge transition points which occur at $-U$ and 0 , where two peaks of width Γ are observed. This does in fact occur at low temperatures ($T \ll \Gamma$). However, below a characteristic temperature T_K , called the Kondo temperature, the conductance is enhanced. This enhancement of conductance at low temperatures is called the Kondo effect.

The low energy physics of the Andersen impurity model can be better understood by mapping it into the Kondo model. This can be done, in the local moment regime with charge fluctuations suppressed, using the Schrieffer-Wolff transformation [20]. The resulting effective model, the Kondo model, is given by

$$H_K = J \vec{S}_d \cdot \vec{s}(0) + \sum_{k\sigma, \alpha=L,R} \epsilon(k) c_{k\sigma, \alpha}^\dagger c_{k\sigma, \alpha}. \quad (1.2)$$

$\vec{S}_d$ is the spin operator of the electron localized on the quantum dot, and $\vec{s}(0)$ is the conduction electron spin density operator at the position of the dot. J is the effective spin coupling between the localized and conduction spins, given by:

$$J = 2V^2 \left[\frac{1}{\epsilon_d + U} - \frac{1}{\epsilon_d} \right]. \quad (1.3)$$

Note that $J > 0$, the coupling is antiferromagnetic, for $-U < \epsilon_d < 0$ which is the regime of interest. At the particle-hole symmetry point, $\epsilon_d = -U/2$, $J = 8V^2/U$.

Therefore, in the local moment regime, there is an effective antiferromagnetic coupling between localized and conduction spins. This coupling leads to the screening of the local moment by the conduction electrons.

Figure 1.2 shows a schematic explanation of the Kondo effect in a quantum dot. Real charge fluctuations are suppressed, but spin fluctuations can occur through virtual intermediate states where zero or two electrons occupy the dot. At low temperatures, the coherent superposition of all such processes creates a narrow resonance (the Kondo resonance) in the density-of-states of the dot. Electrons can use this resonance as a conduction channel, thus conductance is enhanced in the Kondo regime. A very important

Figure 1.2: (a) Energy diagram of the quantum dot in the regime $-U < \epsilon_d < 0$, that is, one electron occupies the dot (figure taken from Ref. [3]). The red areas represent the Fermi sea of the leads. Adding an extra electron to the dot is penalized an energy U , while removing the electron costs an energy $|\epsilon_d|$. However, an electron can tunnel into (out of) the dot, creating a virtual state with two (zero) electrons in the dot, and then tunnel back. This can result in a net spin flip of the electron in the dot. The coherent superposition of all such processes produces the Kondo effect which creates an extra resonance at the Fermi level as shown in (b). The resonance width is the energy scale of the Kondo effect, defined as the Kondo temperature T_K . Electrons can use this resonance as a transport channel thus dramatically enhancing the conductance. Note that the Kondo resonance is pinned to the Fermi level even if the energy of the dot orbital is varied using the gate voltage.

property of this resonance is that it is always pinned to the Fermi level of the leads as ϵ_d is varied (by changing the gate voltage) in the range $[-U, 0]$. Thus, looking at the conductance versus the gate voltage, one obtains a plateau in that range. (See Fig. 1.3).

The above discussion is valid in the equilibrium case, that is, the Fermi levels of the left and right leads are the same. The Kondo peak in the density-of-states is split upon the application of a bias voltage [21]. The split peaks, one corresponding to the Fermi level of each lead, are suppressed by non equilibrium dissipation. Thus the differential conductance shows a peak at zero bias, known as the zero-bias anomaly. As a magnetic field is applied, the Kondo peaks in the density of states shift away from the Fermi levels by the Zeeman energy. This causes a peak in the differential conductance when the the bias voltage is equal to the Zeeman energy. This was originally proposed as the "smoking gun" for the presence of Kondo physics in transport through a quantum dot [21].

Figure 1.3: A schematic of the gate voltage dependence of the QD charge and conductance. The left panel shows the regime $T > T_K$. The conductance is dominated by Coulomb blockade: The charge is well defined due to the strong Coulomb interaction. This suppression of charge fluctuation results in a low conductance except at charge degeneracy points. The right panel shows the conductance when temperature is lowered below the Kondo temperature, $T < T_K$. In the gate voltage intervals where the dot occupation is odd, the plot shows plateaus of enhanced conductance due to the Kondo effect.

1.4 Key Experiments On Nanotransport

The Kondo effect in a QD was first observed experimentally by Goldhaber-Gordon *et al.* [1]. The results show a zero-bias peak in the differential conductance versus bias voltage. The height of this peak decays as temperature increases. The conductance peak is split upon applying a magnetic field. In this case the differential conductance shows two peaks separated by a bias equal to the Zeeman splitting. This confirmed the prediction of Meir *et al.* [21]. In a later experiment [4], the unitary conductance limit ($2e^2/h$) was reached. As temperature is decreased, the results of zero bias conductance versus gate voltage show a transition from the Coulomb blockade to the Kondo regime. For low enough temperature, the conductance reaches the unitary limit.

More elaborate systems were studied afterwards. Jeong *et al.* [7] studied the conductance of two neighboring QDs coupled through a potential barrier. The results show the formation of bonding and antibonding dimer states. A competition between the Kondo effect and the antiferromagnetic correlations between the dots is also observed [22, 6].

The integer-spin Kondo effect was observed by Sasaki *et al.* [8] in a multilevel QD. When

Figure 1.4: The main results of two important experiments reporting the observation of the Kondo effect in QDs. The left panel shows the conductance as a function of gate voltage at different temperatures (figure taken from Ref. [4]). At high temperature, the results show Coulomb blockade behavior (orange line). As temperature is decreased, the conductance in the even occupation regimes is suppressed (due to the suppression of thermal fluctuations), while in the odd occupation regimes, the conductance is enhanced due to the Kondo effect until it reaches the unitary limit ($2e^2/h$) for low enough temperatures. This was the first experiment to report the unitary conductance limit in the Kondo regime [4]. The right panel shows the differential conductance versus bias voltage for different temperatures and magnetic fields (figure taken from Ref. [1]). As the temperature is lowered, a peak is observed at zero bias (zero-bias anomaly). This peak is split upon the application of a magnetic field due to Zeeman effect. These results are the “smoking gun” for the Kondo effect in nanotransport. This experiment was the first observation of the Kondo effect in a QD [1].

Figure 1.5: Main results of the experiment by Craig *et al.* on nonlocal spin control in quantum dots [5]. The inset shows a schematic of the system studied (see also Fig. 4.1). The two quantum dots are connected by an open central region. Current is measured through QD1 as indicated by the thin arrow for different occupations of the other dot (QD2), and different couplings between QD2 and the open region as indicated by the thick arrow. The left figure shows the results for odd occupation of QD2. As the coupling is increased, the zero-bias anomaly in QD1 is split. The right figure shows the results for even occupation where increasing the coupling strength slightly reduces the conductance of QD1.

an even number of electrons occupies the dot, the energy difference between the singlet and triplet states was tuned using a magnetic field. At the singlet-triplet degeneracy point, the spin-1 Kondo effect was observed. The characteristic energy scale is much larger than the ordinary spin-1/2 case, and was observed at a much higher temperature.

An orbital Kondo effect, in addition to the ordinary spin Kondo effect, was observed in carbon nanotube QDs [9]. The conduction electrons screen both the spin and orbital degrees of freedom of the localized electrons. When orbital and spin degeneracies are present simultaneously, a strongly enhanced Kondo effect obeying the $SU(4)$ symmetry is observed.

A possible QD realization of the RKKY interaction between two magnetic impurities was claimed by Craig *et al.* [5]. The system studied consists of two similar QDs separated by an open region. Current through one of the dots, labeled QD1, was measured for different charge states (even or odd occupation) on the other quantum dot (QD2); and for different couplings between QD2 and the open region. The main result was the splitting of the

zero-bias anomaly in QD1 when an odd number of electrons occupies QD2. This splitting increases as the coupling between QD2 and the open region is increased. The observed results are explained as the effect of RKKY interaction between spins localized on the QDs, mediated by electrons in the open region.

Correlation effects were also observed in single-molecule conductors. Park *et al.* [2] and Liang *et al.* [23] reported the observation of Coulomb blockade as well as the Kondo effect in coordination compounds containing Co and V_2 ions respectively. Yu *et al.* observed the Kondo effect in C_{60} molecules [24]. In addition to correlation effects, electron-phonon coupling plays an important role in the transport properties of single-molecule conductors. This was observed experimentally [25, 24] and has been the subject of extensive theoretical investigation [26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36].

1.5 Overview of the Works Presented in this Dissertation

In this dissertation, I present the results of our numerical studies on electron transport in quantum dots and single-molecule conductors. The main focus is on correlation and electron-phonon coupling effects. The organization is as follows: In chapter 2, we study the transport properties of *multilevel* QDs in the Kondo regime. The conductance can be decomposed into the contributions of each level. It is shown that these channels can carry a different phase, and destructive interference processes are observed when the phase difference between them is $\pm\pi$. The effect is different from previous studies of interference processes in dots. In particular, no external magnetic field is applied, and the dot-leads hopping amplitude is the same for all levels. However, conductance cancellations are still observed. Another interesting effect observed is the formation of localized states that do not participate in transport. When one of these states crosses the Fermi level, the electron occupation of the dot changes, modifying the many-body physics of the system and indirectly affecting the transport properties.

In chapter 3, we study the transport properties of two coupled double-level quantum dots. The results strongly suggest that, under the appropriate conditions, the dots develop a novel “ferromagnetic” regime at quarter filling (one electron per dot). For large Coulomb repulsion, and interdot tunneling larger than tunneling to the leads, a spin-1 Kondo resonance develops in the density of states leading to a conductance peak. This effect is proposed as a

QD realization of the double exchange mechanism and can be observed experimentally.

Chapter 4 shows the main results of our numerical simulations of an important experimental effort by Craig *et al.* [5] involving nonlocal spin control. Our results reproduce the main features obtained in the experiment. To understand better the numerical and consequently the experimental results, we introduce a simple “circuit model” that treats the two dots as circuit elements connected in series. This model also reproduces the main results of the experiment. Our main conclusion is that the results of the experiment can also be explained as the effect of Fano antiresonance, alternative to the RKKY explanation proposed before [5, 37, 38].

In chapter 5, we study the effect of electron-phonon coupling on the linear transport properties of a molecule, mainly in the Kondo regime. We study first the effect of the Holstein coupling, i.e. the vibrational excitation coupled to the charge of the molecule. We reproduce previous results obtained using the Numerical Renormalization Group technique. In the weak electron-phonon coupling regime, the standard spin Kondo effect is obtained but with renormalized parameters; while in the strong electron-phonon coupling regime, the charge Kondo effect is obtained. We then proceeded to study the effect of center-of-mass motion, i.e. the molecule can oscillate between the two electrodes thus dynamically modifying the molecule-leads tunnel barriers. The results present an interesting interference that leads to a conductance dip at the particle-hole symmetry point. It is shown that the dip is the result of the destructive interference between the purely electronic and phonon-assisted tunneling channels, which are found to carry opposite phases. When an internal breathing mode is also active, the particle-hole symmetry is broken but a Fano-like interference is still observed.

The studies in chapters 2, 3, 4, and 5 were carried out using the embedded cluster approximation (ECA). The leads are modeled as semi-infinite tight binding chains. A cluster consisting of the central interacting region and a few sites of each lead is isolated. Then, using Lanczos, the ground state and Green functions of the cluster are calculated exactly. The cluster is then connected to the rest of the leads using the Dyson equation. Finally, the conductance is calculated using the embedded-cluster Green functions.

In chapter 6, we present a procedure to calculate the zero temperature conductance of nanostructures based on the recently developed “adaptive” time-dependent density-matrix renormalization group (DMRG) [39, 40, 41]. The leads are modeled using noninteracting

tight-binding Hamiltonians. The ground state at time zero is calculated at zero bias. Then, a small bias is applied between the two leads, the wave function is DMRG evolved in time, and currents are measured as a function of time. Typically, the current is expected to present periodicities over long times, involving intermediate well-defined plateaus that resemble steady states. The conductance can be obtained from those steady-state-like currents. To test this approach, several cases of interacting and noninteracting systems were studied. Our results show excellent agreement with exact results in the noninteracting case. More importantly, the technique also reproduces quantitatively well-established results for the conductance in both the cases of one and two coupled interacting QDs. The technique also works at finite bias voltages, and it can be extended to include interactions in the leads.

CHAPTER 2

INTERFERENCE EFFECTS IN THE CONDUCTANCE OF MULTILEVEL QUANTUM DOTS

2.1 Introduction

The low temperature transport properties of quantum dots (QDs) have received considerable attention in the last decade. In addition to being extremely tunable and well controlled test beds for studying many-body effects, these structures are strong candidates for carrying out quantum computing and spintronics. More specifically, two coupled quantum dots (2QD) have been proposed to represent tunable qubit circuits [42], where the qubit is encoded by the state of the localized spin in the QD. Thus, the study of interacting electrons in nanosystems and its associated quantum transport are very important for progress in condensed matter physics in general.

In several previous studies it has been usually *assumed* that, e.g., the 2QD have only one localized spin per dot, since only one level is assumed active in each dot. For the success of these proposals, it is important to understand the role played by other levels in the QDs (i.e. considering *multilevel* dots). This provides part of the motivation for the work presented here. In addition, in order to properly perform these subtle studies, it is crucial to carry out the calculations using *unbiased* techniques that do not assume properties of familiar states that arise from previous experience with bulk materials. Ideally, the calculations should make the least possible number of assumptions, and in this framework, it is important to develop and use suitable *numerical* techniques to handle these complicated fully-quantum problems accurately. For this reason, the calculations discussed below were carried out with a powerful numerical method that has been recently developed.

Other interesting investigations in this context have proposed the use of quantum dots

as *interferometers* in the Aharonov-Bohm (AB) geometry with external magnetic fields applied [43, 44]. In general, previous efforts have considered a magnetic flux enclosed in two possible paths for the electrons. In these studies, the tunneling matrix-elements are defined with an explicit phase factor that represents the magnetic flux. These investigations have shown that narrow *dips* in the conductance can appear as a consequence of phase interference in the AB circuit. Recent theoretical investigations in AB rings have shown topological charge phase transitions that can induce interference [45, 46]. These transitions are driven by changes in the symmetry of the ground state. Other investigations have also found conductance cancellations for the case of two quantum dots, individually connected to leads and between themselves [47]. This effect is present even using one-body interactions in the formalism, and is caused by interference between two different paths in the geometry of the problem.

One additional important motivation for the analysis discussed in the present chapter is to find alternative sources of interference among wave functions that can also lead to conductance dips, *in the absence of external fields and also in the absence of obviously distinct paths*. Effects of this variety were recently addressed in multilevel dots by Silva *et al.* and Kim and Hershfield [48, 49], where the consequences of having tunneling matrix elements with different signs in a multilevel QD and in an AB circuit were discussed. These authors show that the matrix-element signs are important to determine the global phase of the electrons when they cross the QD.

In the investigations presented here, we consider a system composed of one quantum dot – with two active levels and including the Hubbard many-body interactions – coupled to ideal single-channel leads. In gate-voltage regimes where there is only one electron in the QD, the usual Kondo physics [50, 51, 21] can be found at low temperatures. The coupling between the spin in the QD and the spins in the leads creates a resonance at the Fermi level that contributes to the electron transport. When a second electron enters the QD, more interesting physics is observed because now the total spin of the system (singlet or triplet) plays an important role [8, 52, 53]. One of the main results of our investigations is that the two levels can act as different channels that individually contribute to the overall conductance with their own phase. This phase plays a role similar to that of a magnetic field in the AB interferometer, and it can induce constructive or destructive interferences. Below, it is explicitly shown that a phase difference of π can exist between the two channels, even if

there is no explicit difference between their hopping amplitudes and Coulombic interactions. Only a tiny difference in the energies of the two levels is needed for the effect to develop, as well as a nonzero Hubbard coupling which is required to produce the Kondo physics. It should be remarked that this phase difference is different from the phase measured in some QD experiments [54] since it is *internal* to the QD. A magnetic field is not necessary to observe this interference.

Other recent investigations have also focused on systems that present interference processes even without magnetic fields or explicitly different paths. For instance, Hofstetter and Schoeller analyzed lateral quantum dots near the *singlet-triplet* degeneracy point, which occurs from the competition of the Hund's rule coupling, that favors the triplet, and the nonzero energy difference between levels, that favors a singlet state [55, 56, 57]. A dip in the conductance was observed in this region. The study discussed in the present chapter includes this singlet-triplet transition regime as a special case, and mainly focuses on a different variety of interference effects that are present even for a very small energy difference between the levels in the quantum dot. Our main picture is based on the notion that charge transport can occur with different phases when electrons cross different levels of a quantum dot, rather than a singlet-triplet competition. Alternatively, our results can also be visualized as interference between two Kondo states: one with $S = 1$ and the other with $S = 1/2$.

The organization of this chapter is the following: In Sec. 2.2, the discussion of the formalism, mainly the model and technique, is presented. In Sec. 2.3, the case of one dot with two levels is studied in detail, showing the novel interferences in transport. The analysis of the integer-spin Kondo effect, that appears here in the interesting region of parameters, is presented in Sec. 2.4. It is well-known that this type of states can enhance the conductance near singlet-triplet transitions [8, 52]. A possible intuitive explanation of the effect is given in Sec. 2.5, and the conclusions are provided in Sec. 2.6. Other effects analyzed in this work include the possibility of observing *localized states* near the dot. One such localized state can cross the Fermi level of the system without contributing to the conductance, but affecting the transport of other states through the Coulomb interaction. This is shown to lead to discontinuities – as opposed to cancellations – in the conductance versus gate voltage.

Figure 2.1: Schematic representation of the main system studied in this chapter, consisting of one quantum dot with two levels α and β . The two possible paths carrying different phases Φ_α and Φ_β are shown.

2.2 One Dot with two levels: Formalism

The main system analyzed in this chapter consists of one quantum dot, with several interacting levels, connected to two ideal leads. It will be argued that – mainly due to the many-body interactions – these levels can carry different phases in transport processes. This is shown schematically in Fig. 2.1. The phase difference gives rise to a complex conductance pattern including destructive interferences (i.e. an “internal” Aharonov-Bohm effect appears to be dynamically generated).

To model this system, the Anderson impurity Hamiltonian with two levels (α and β) is used. The portion corresponding to the isolated dot is

$$\begin{aligned}
 H_{\text{dot}} = & \quad U/2 \sum_{\lambda=\alpha,\beta} \sum_{\sigma} n_{\lambda\sigma} n_{\lambda\bar{\sigma}} + U'/2 \sum_{\sigma\sigma'} n_{\alpha\sigma} n_{\beta\sigma'} - J \sum_{\sigma\sigma'} c_{\alpha\sigma}^\dagger c_{\alpha\sigma'} c_{\beta\sigma'}^\dagger c_{\beta\sigma} + \\
 & \sum_{\sigma} [V_g n_{\alpha\sigma} + (V_g + \Delta V) n_{\beta\sigma}], \quad (2.1)
 \end{aligned}$$

where the first term represents the usual Coulomb repulsion between electrons in the same level, which for simplicity is considered equal for both levels. The second term represents the Coulomb repulsion between electrons in different levels (the U' notation is borrowed from standard many-orbital studies in atomic physics). The third term represents the Hund coupling ($J > 0$) that favors the parallel alignment of spins; and the last term is simply the energy of the levels regulated by the gate voltage V_g . Note that level α is assumed to be

separated from β by ΔV , and by modifying this parameter an interpolation between one- and two-orbital physics can be obtained. The dot is connected to the leads (represented by semi-infinite chains) by a hopping term with amplitude t' , while t is the hopping amplitude in the leads. More specifically,

$$H_{\text{leads}} = -t \sum_{i\sigma} \left[c_{li\sigma}^\dagger c_{li+1\sigma} + c_{ri\sigma}^\dagger c_{ri+1\sigma} + \text{h.c.} \right], \quad (2.2)$$

$$H_{\text{dot-leads}} = -t' \sum_{\sigma, \lambda=\alpha, \beta} \left[c_{\lambda\sigma}^\dagger (c_{l0\sigma} + c_{r0\sigma}) + \text{h.c.} \right], \quad (2.3)$$

where $c_{li\sigma}^\dagger$ ($c_{ri\sigma}^\dagger$) creates an electron with spin σ at site i in the left (right) lead. Site “0” is the first site to the left and right of the dot, in each lead. The total Hamiltonian is

$$H_{\text{T}} = H_{\text{dot}} + H_{\text{leads}} + H_{\text{dot-leads}}. \quad (2.4)$$

Note that for $V_g = -U/2 - U' + J/2 - \Delta V/2$, the Hamiltonian is particle-hole symmetric.

Using the Keldysh formalism [58, 59], the conductance through this system can be written as

$$\sigma = 2 \frac{e^2}{h} |t^2 G_{lr}(\epsilon_F)|^2 [\rho(\epsilon_F)]^2, \quad (2.5)$$

where $G_{lr}(\epsilon_F)$ is the Green function that corresponds to moving an electron from the left lead to the right one, ϵ_F is the Fermi energy, and $\rho(\epsilon_F)$ is the density of states of the leads (assumed the same left and right, for simplicity):

$$\rho(\omega) = -\frac{1}{\pi} \text{Im} \left\{ \frac{(\omega + i\eta) - \sqrt{(\omega + i\eta)^2 - 4t^2}}{2t^2} \right\}. \quad (2.6)$$

To use Eq.2.5, the Green functions are calculated using the ECA technique, i.e. a small cluster is solved exactly using Lanczos then embedded in the rest of the leads using Dyson equation. Note that G_{lr} has two contributions, one corresponding to transport through path “ α ” and the other through path “ β ”. From the equations of motion [60, 61, 21], which correspond to an exact expansion of the Green function, it can be shown that

$$G_{lr} = \tilde{g}_l t' G_{\alpha r} + \tilde{g}_l t' G_{\beta r}, \quad (2.7)$$

where $\tilde{g}_l$ is the Green function of the first left-contact and $G_{\alpha r}$ ($G_{\beta r}$) is the dressed Green function to move from the state α (β) to the right-contact.

The two terms in Eq. 2.7, involving $G_{\alpha r}$ and $G_{\beta r}$, can be evaluated independently. To calculate the phase difference, the following expression is used

$$i \Delta\Phi = \log \left\{ \frac{G_{\alpha r}}{G_{\beta r}} \frac{|G_{\beta r}|}{|G_{\alpha r}|} \right\}. \quad (2.8)$$

The conductance is proportional to $|G_{lr}|^2$ and, using Eqs. 2.5, 2.7 and 2.8, it can be written as

$$\sigma = \sigma_{\alpha} + \sigma_{\beta} + 2\sqrt{\sigma_{\alpha}\sigma_{\beta}} \cos \Delta\Phi, \quad (2.9)$$

where σ_{α} and σ_{β} are the partial conductances given by

$$\sigma_{\alpha} = 2\frac{e^2}{h} [t^2 \tilde{g}_l t' \rho(E_F)]^2 |G_{\alpha r}(E_F)|^2, \quad (2.10)$$

$$\sigma_{\beta} = 2\frac{e^2}{h} [t^2 \tilde{g}_l t' \rho(E_F)]^2 |G_{\beta r}(E_F)|^2. \quad (2.11)$$

An exact cancellation of the conductance can occur when $\sigma_{\alpha}=\sigma_{\beta}$ and $\Delta\Phi=\pm\pi$ as a consequence of a destructive interference process. In the same way, the situation with $\Delta\Phi=0$ and $\sigma_{\alpha}=\sigma_{\beta}$ can be considered as a constructive interference.

One of the important energy scales in this problem is the broadening of the dot levels due to hybridization with the leads, $\Gamma=2\pi t'^2 \rho(\epsilon_F)$. The ratio Γ/U defines the Kondo temperature for the case of a spin-1/2 in the quantum dot [62]. The coupling values used are (unless otherwise stated) $U=0.5t$, $U'=2/3U$, $J=U-U'$, and $t'=0.2t$. This allows for a fast convergence with the cluster size [22]. Fixing the values for the Coulombic interaction couplings is only for simplicity, to avoid exploring a vast parameter space. Considering U' to be smaller than U is reasonable because the latter is expected to be the largest Coulombic interaction. The equation for J is borrowed from atomic physics problems [63, 64, 65]. It gives a reasonably small value for the Hund interaction in units of U . In addition, we select the ratio Γ/U to be close to the experimental values [1].

2.3 Conductance Cancellations in Multilevel Dots

Using the Lanczos method to evaluate the Green functions of a small cluster followed by an embedding process, we calculate the conductance, the charge of the QD, and the phase difference (Eq. 2.8) as a function of V_g . Unless otherwise stated, we used the smallest possible cluster size L . For the case analyzed in this section this smallest size is $L=4$,

Figure 2.2: Conductance for five different values of ΔV . The conductance (in units of $2e^2/h$) is shown in solid lines and the phase difference (divided by π) in dotted lines. When ΔV is the largest energy ($\Delta V=2t$ (e)), two structures similar to those found in a single-level QD, separated by ΔV , appear in the conductance. Case (d) is similar, but since ΔV is not large enough, an extra thin peak appears around $V_g = -1$ (this peak is shown with more detail in Fig. 2.8). When ΔV decreases, these structures begin to interact giving rise to conductance dips, as shown in (b) and (c) for the cases $\Delta V=0.2t$ and $\Delta V=0.5t$ ($\Delta V=0.2t$ is studied in more detail in Fig. 2.4). Finally, in the limit $\Delta V=0$, the conductance dips transform into discontinuities.

which corresponds to two “lead” sites and two levels in the dot. In the last part of this section we analyze the convergence of the results with increasing L . In Fig. 2.2, we show the conductance for several values of the energy separation ΔV between the two levels of the dot. At large ΔV (Fig. 2.2(e)), the levels are well separated and the interaction between them is weak. The conductance shows just two wide peaks associated with the standard Kondo effect for each level. Note that the width of these peaks is U , and with increasing

temperature, these broad peaks will transform into two sharper peaks each, corresponding to the Coulomb blockade. This physics has been discussed at length in previous publications [22, 53], and it basically amounts to the behavior of single-level QDs, which is natural at large ΔV .

Far more interesting results are obtained by reducing ΔV . For instance, in Fig. 2.2(d), a thin central-peak is already observed. This peak seems to be the consequence of a constructive interference between the two possible channels, in a region where a state with total spin $S_D=1$ is formed, as will be discussed more extensively below. Upon further decreasing of ΔV (Figs. 2.2(b) and (c)), the two largest structures begin to interact with the thin central peak, increasing its width, and a more complex pattern emerges. With the levels of the dot getting closer in energy, it is more likely to have them populated at the same time, and due to the Hund coupling, the state with $S_D=1$ has a high probability for the case of two electrons in the dot, giving rise to a spin-1 Kondo effect within the central peak. The other two peaks are associated with $S_D=1/2$ and 1 or 3 electrons in the dot. For $\Delta V \leq 0.5t$, dips in the conductance are clearly observed, and later we show that these cancellations are related to interference processes. Finally, at $\Delta V=0$ (Fig. 2.2(a)), several *discontinuities* appear in the conductance. It will be argued that these discontinuities are a consequence of the existence of *localized* states at the dot. These states are not of direct relevance for the transport of charge, but when the gate voltage moves them below the Fermi level of the leads, they simply increase the charge of the quantum dot, which indirectly affects transport because of the Coulombic interaction. Note that for $\Delta V=0$, the phase difference between levels α and β is zero, since the Green functions $G_{\alpha 2}$ and $G_{\beta 2}$ are equal. Thus, no destructive interferences are observed, but discontinuities can still occur.

Figure 2.3 shows how one of the conductance discontinuities rapidly transforms into a dip as ΔV increases from zero. In Figs. 2.4, 2.5 and 2.6, we can observe in detail the case with $\Delta V=0.2t$. Figure 2.4 shows the conductance together with the phase difference and the partial conductances. Clearly, the zeros of the conductance are in regions where $\Delta\Phi=\pi$. Then, from Eq. 2.9, the dips must occur when the partial conductances σ_α and σ_β take the same value. More details of dip "b" are shown in Fig. 2.5 where thick arrows point to the zero in the conductance and the location where $\sigma_\alpha = \sigma_\beta$. The dot charge for the same ΔV is shown in Fig. 2.6. The central peak in the conductance occurs in the regime where two electrons are in the dot, occupying different levels. It can also be observed that the total

Figure 2.3: Conductance (in units of $2e^2/h$) in a narrow range of V_g and ΔV . The discontinuity at $\Delta V=0$ is transformed into a dip for finite values of ΔV .

charge at the QD ($\sum_{\sigma} [n_{\alpha\sigma} + n_{\beta\sigma}]$) does not show the characteristic sharp plateaus of the Coulomb blockade regime.

Similar results, but for $\Delta V=t$, are shown in Fig. 2.7. In this case, the large ΔV well separates levels α and β , and as a consequence, one of the levels is charged almost completely before some charge starts to occupy the other level (Fig. 2.7(c)). The latter occurs at the gate voltage where the conductance peak is observed.

To better understand the physics associated with the thin central peak for the case $\Delta V=t$ shown in Fig. 2.2, the conductance and the phase difference are presented in more detail in a narrow V_g range in Fig. 2.8. We can observe that the partial conductances σ_{α} and σ_{β} are individually small (< 0.25), but as the phase between the two channels is 0, they contribute in a constructive way, inducing a peak in the total conductance.

Our results have been obtained by solving exactly a relatively small cluster and, then, approximately reaching the bulk limit using the Dyson equation. It is important to verify that our results are robust if the cluster size increases. In Fig. 2.9, the convergence of the conductance with increasing cluster size at $\Delta V=0.2t$ is shown. Remarkably, the difference between $L=8$ and $L=20$ is less than 0.5%, showing that the results presented here are nearly

Figure 2.4: Conductance (in units of $2e^2/h$) (a), phase difference (divided by π) (b), and partial conductances (c) for $\Delta V=0.2t$. The dips, indicated by “a” and “b” in (a), are in regions where the phase difference is π producing exact cancellations when $\sigma_\alpha=\sigma_\beta$.

Figure 2.5: Conductance (in units of $2e^2/h$), phase difference, and partial conductances for $\Delta V=0.2t$ around the “b” dip of Fig. 2.4(a). Thick arrows show the gate potential where the conductance vanishes, $\Delta\Phi = \pi$, and $\sigma_\alpha = \sigma_\beta$.

Figure 2.6: (a) Conductance (in units of $2e^2/h$), (b) total charge of the QD, and (c) charge of the individual levels for the case $\Delta V=0.2t$. The central peak emerges in the regime where two electrons occupy the dot, one in each level.

Figure 2.7: Conductance (in units of $2e^2/h$) and charges as a function of the gate potential for $\Delta V=t$. These results should be compared to those in Fig. 2.6

Figure 2.8: Conductance (in units of $2e^2/h$), phase difference and partial conductances for $\Delta V=t$ around the central peak shown in Fig. 2.2(d). The partial contributions to the conductance (σ_α and σ_β) have the same shape as in Fig. 2.4 but now the phase difference between them is zero. This phase difference creates a peak whereas in the other case a dip appears.

exact. The fast convergence is a consequence of the local characteristics of the dot; namely, all the important effects are contained in the small cluster solved exactly. The fast size convergence shows that even using the smallest cluster, the results are already qualitatively correct.

It is important to remark that the cancellations observed here do not crucially depend on the couplings U' and J , since they are present in our results even for $U'=J=0$, as shown in Fig. 2.10. Also for completeness, the case of a large $U'=2t$ is shown in Fig. 2.11. Even though the situation $U'>U$ is unphysical – since the Coulomb repulsion in the same level should be larger than for electrons in different levels – it is interesting to note that U' plays a role similar to that of ΔV . Since the energy of having a second electron in β is related to $U'-J+\Delta V$, for a large U' (with a J of the same sign as considered before), the system acts the same as in the case of large ΔV . Figure 2.11 contains two different cases for $\Delta V=0$, one with a ferromagnetic Hund coupling $J=|U-U'|>0$ (a) and the other with an antiferromagnetic coupling $J=U-U'<0$ (b). In the first case, since the Hund coupling is proportional to U' ,

Figure 2.9: Conductance convergence with the size of the exactly-solved cluster L for the case $\Delta V=0.2t$. Dotted lines are for $L = 4$, continuous thin lines for $L = 8$, and dashed lines for $L = 12$. Extra points for $L = 16$ and $L = 20$ are shown with crosses and diamonds, respectively.

Figure 2.10: Conductance and total charge when U' and J are not active. The results are shown are results for $U'=J=0$ (solid line) compared to the previous results for $U'=2/3U$ and $J=U - U'$. $\Delta V=0.35$ for both plots. The behavior is qualitatively the same in both cases.

Figure 2.11: Conductance (in units of $2e^2/h$) for large $U'=2t$. (a) shows the case of ferromagnetic Hund coupling $J=|U - U'|$ while (b) shows the case of antiferromagnetic $J=U - U'$. $\Delta V=0$ in both cases.

we observe the three-peak structure in the conductance, with a central-peak associated to the $S_D=1$ states. In the second case, U' and J try to eliminate the $S_D=1$ states, and the conductance has a structure similar to the one shown in the previously studied example with $\Delta V=t$ (see Fig. 2.2(d)).

2.4 Integer-Spin Kondo Effect

Recently, transport through a multilevel QD connected to two leads was experimentally studied as a function of the level separation (ΔV) tuned by an external magnetic field. For a dot with an even number of electrons, it was shown that the Kondo correlations formed between the $S_D=1$ triplet state of the dot and the spins of the leads have a strong influence on the current [8]. This problem was theoretically studied using a mapping onto a two-impurity Kondo model [52, 53].

Figure 2.12 shows schematically the different situations that can arise as the number of electrons in the QD is changed using the gate voltage. Starting from the upper left (a), the gate potential V_g is above the Fermi level and there are no electrons in the QD. Level α

Figure 2.12: Schematic representation of the different scenarios that can occur in a two-level QD as the gate voltage changes. See the text for more details.

is separated from level β by the energy ΔV . Decreasing V_g , as shown in Fig. 2.12(b), one electron populates level α , and the energy to add an extra electron in β is $\Delta V + U' - J$, since an energy U' is paid due to Coulomb repulsion and an energy J is gained due to the spin coupling. The system is now in the Kondo regime for transport if $T < T_K$. Decreasing V_g even further, a second electron can enter the dot. The spin of this second electron defines the transport properties since the total spin of the QD can be $S_D=1$ (with a Kondo resonance for temperatures below $T_{K(S=1)}$) or $S_D=0$ with no Kondo effect. In the right-lower frame (d) the second electron enters the lower energy state α . The energy of this configuration is $E_{S_D=0}=2V_g + U$. On the other hand, if the second electron enters the dot with the same spin as the first one forming a $S_D=1$ state, we have the case shown in the left-lower frame (c). The energy of this state in the Kondo regime is $E_{S_D=1}=2V_g + \Delta V + U' - J - T_{K(S=1)}$ [53]. For $\Delta V > U - U' + T_{K(S=1)}$, we expect a transition from the $S_D = 1$ (right-lower frame) to the $S_D = 0$ state (left-lower frame).

Note that the spin of the dot S_D is not a well-defined quantum number because the QD is not isolated. In the $L = 4$ cluster, we found that the ground state used for the calculations has a spin $S_D - 0.7426$ [calculated from the relation $\langle \vec{S}_D^2 \rangle_{\text{cluster}} = S_D(S_D + 1)$] at $\Delta V = 0.2$ and $V_g = -0.6$ (particle hole symmetry). Since the number of particles at this gate potential is 2, we deduce that the ground state can be expressed as a combination of states with $S_D = 1$ and 0, with a weight of 86% for $S_D = 1$. In the case $\Delta V = t$, we obtain a total spin $S_D = 0.352$ for the particle-hole symmetric gate potential, which corresponds to a 60% chance of having $S_D = 1$.

One way to confirm that the central peak in Fig. 2.4(a) indeed originates in a $S_D = 1$ state is by reducing the value of J , making less probable the ferromagnetic coupling between levels α and β . Figure 2.13 shows the conductance for the case shown in Fig. 2.4(a) plus two different cases with the same values of U , U' and t' but different J 's. In (b), with $J = 0$, we observe a narrow central peak showing that the effect persists due to the $T_{K(S=1)}$ energy. For negative values of J , as shown in (c), the ferromagnetic state is even less probable and the central peak is narrower. This peak must disappear for larger negative values of J [i.e. $|J| \gg T_{K(S=1)}$]. All this suggests that it is the spin-one Kondo effect that is responsible for some of the effects discussed here.

2.5 Understanding the interference in the conductance

The previous analysis shows that the origin of the three peaks in the conductance can be traced back to Kondo effect that occurs for 1, 2, and 3 electrons in the dot. However, it remains to be understood what causes the conductance cancellation between them. It is expected that the two paths – through α and β – are responsible for the interference, but a more detailed explanation would be desirable.

2.5.1 Origin of the π Phase Difference

To understand how the phase difference arises, let us first study the *cluster* Green function. In Fig. 2.14, the real and the imaginary parts of $g_{\alpha r}$ and $g_{\beta r}$ are shown for the case of $\Delta V = 0.05t$ and a gate potential where the relative phase is π ($V_g = 0.108t$). Since the cluster is by definition a finite system, these two Green functions are simply a collection of a large number of poles. For $\Delta V = 0$, level α is equal to β in energy, and both Green functions must

Figure 2.13: Conductance (in units of $2e^2/h$) for different values of J . (a) corresponds to $J=U - U'$, (b) to $J=0$, and (c) to a large negative $J=-0.5t$. The central peak disappears when the ferromagnetic spin correlations are eliminated at even more negative J . More details can be found in the text.

have poles at exactly the same positions. We expect that for a small ΔV their features must still remain similar. The most important information in Fig. 2.14 is that the poles corresponding to $g_{\alpha r}$ and $g_{\beta r}$ at approximately the same energy near the Fermi level ($\omega = 0$) have weights of *different signs*. This means that these poles have a phase difference of π , which causes the interference.

Another special property of the bare (cluster) Green function is shown in Fig. 2.15. In this figure, the imaginary parts of $g_{\alpha\alpha}$ and $g_{\alpha r}$ for $\Delta V=0.05t$ and $\Delta V=0$ are shown. While for $\Delta V=0.05t$ both Green functions have poles at the same positions, at $\Delta V=0$ the weight of the poles near the Fermi level for $g_{\alpha r}$ are *zero*. This is a characteristic of a *localized* state.

To understand these two properties we have to analyze the non-diagonal Green functions $g_{\alpha r}$ and $g_{\beta r}$ in their Lehmann representation:

$$g_{\alpha r}(\omega) = \sum_l \frac{\langle f | c_{\alpha\sigma} | \phi_l \rangle \langle \phi_l | c_{r\sigma}^\dagger | f \rangle}{\omega - (E_F - E_l) + i\eta} + \sum_l \frac{\langle f | c_{r\sigma}^\dagger | \phi_l \rangle \langle \phi_l | c_{\alpha\sigma} | f \rangle}{\omega + (E_F - E_l) + i\eta}, \quad (2.12)$$

$$g_{\beta r}(\omega) = \sum_l \frac{\langle f | c_{\beta\sigma} | \phi_l \rangle \langle \phi_l | c_{r\sigma}^\dagger | f \rangle}{\omega - (E_F - E_l) + i\eta} + \sum_l \frac{\langle f | c_{r\sigma}^\dagger | \phi_l \rangle \langle \phi_l | c_{\beta\sigma} | f \rangle}{\omega + (E_F - E_l) + i\eta}, \quad (2.13)$$

Figure 2.14: Density-of-states of the bare $L = 4$ cluster, for $\Delta V = 0.05t$ and $V_g = -1.082t$. The Fermi level lies at $\omega = 0.0$. Note the relative change of the weight *signs* of the poles near $\omega = 0$, when comparing the Green functions $g_{\alpha r}$ and $g_{\beta r}$.

where $|f\rangle$ and E_F are the ground state and its eigenenergy, $\{|\phi_l\rangle\}$ is a complete basis of Hamiltonian eigenstates with eigenenergies E_l , and η is a small number (typically 10^{-7}).

Both Green functions have poles at the same positions $\omega = \pm(E_F - E_l)$. For simplicity, only the first term in Eqs. 2.12 and 2.13 is analyzed, but the study presented below is qualitatively the same for the second term. As the matrix element $\langle\phi_l|c_{r\sigma}^\dagger|f\rangle$ is the same for both Green functions, it is clear that they can have a π phase difference only if the matrix element $\langle f|c_{\alpha\sigma}|\phi_l\rangle$ has a different sign than $\langle f|c_{\beta\sigma}|\phi_l\rangle$.

This change in the sign of the matrix elements can be understood based on the “reflection” symmetry between levels α and β . For the case $\Delta V = 0$, the transformation $\alpha \rightarrow \beta$ must leave the Hamiltonian invariant. The operator $\hat{O}_r$ associated with this transformation commutes with the total Hamiltonian and has eigenvalues 1 and -1 (if the states are even or odd under this transformation). The complete basis $\{|\phi_l\rangle\}$ used in Eqs. 2.12 and 2.13 can be divided

Figure 2.15: Comparison between the imaginary parts of $g_{\alpha\alpha}$ and $g_{\alpha r}$, for $\Delta V=0$ and $\Delta V=0.05t$. The pole in $g_{\alpha N}$ near the Fermi level disappears (i.e. carries no weight) at $\Delta V=0$. ($V_g = -1.082t$ and $L = 4$ in both cases.)

into two subsets $\{|\phi_l^+\rangle\}$ and $\{|\phi_l^-\rangle\}$ depending on whether the states are even or odd:

$$\hat{O}_r |\phi_l^+\rangle = |\phi_l^+\rangle, \quad (2.14)$$

$$\hat{O}_r |\phi_l^-\rangle = -|\phi_l^-\rangle. \quad (2.15)$$

The destruction operators for levels α and β transform as

$$c_{\alpha\sigma} = \hat{O}_r^\dagger c_{\beta\sigma} \hat{O}_r. \quad (2.16)$$

Suppose that the ground state is an even state $|f^+\rangle$ (we confirmed this numerically). Then, there are two kinds of matrix elements, $\langle f^+ | c_{\alpha\sigma} | \phi_l^+ \rangle$ and $\langle f^+ | c_{\alpha\sigma} | \phi_l^- \rangle$. These two matrix elements must be compared with their corresponding $c_{\beta\sigma}$ matrix elements. Using Eqs. 2.15 and 2.16 we have

$$\langle f^+ | c_{\alpha\sigma} | \phi_l^+ \rangle = \langle f^+ | c_{\beta\sigma} | \phi_l^+ \rangle, \quad (2.17)$$

$$\langle f^+ | c_{\alpha\sigma} | \phi_l^- \rangle = -\langle f^+ | c_{\beta\sigma} | \phi_l^- \rangle. \quad (2.18)$$

Figure 2.16: Asymmetric α - β case, where the Coulomb repulsions in the two levels are different i.e. $U_\beta = U_\alpha + 0.1t$. Dips in the conductance are observed even for $\Delta V = 0$, due to the explicitly broken symmetry. More details can be found in the text.

Then, some of the poles of Eqs. 2.12 and 2.13 carry weights with different signs. When ΔV is different from zero, the weights are different but the sign difference remains.

2.5.2 Interference between $S=1/2$ and $S=1$ Kondo states?

Note that the conductance cancellations mainly occur for values of the gate voltage where the charge of the dot is changing, for example from 1 to 2. This suggests that an alternative way to visualize the interference process is to imagine the ground state in this intermediate regime as a linear combination of two Kondo states, one corresponding to spin-1/2 at the dot and the other to spin-1. In fact, in the important range of gate voltages considered in our study, the charge at the dot is not sharply defined, thus this approximation is reasonable. The interference can arise from a destructive process between the transport corresponding to each of these two nearly-degenerate states. This hypothesis will be analyzed more extensively in future studies.

2.5.3 Localized States

The transport properties of the system are related to the Green function G_{lr} [see Eq. 2.5]. The Lehmann representation of this Green function contains matrix elements such as $\langle f^+ | c_{r\sigma}^\dagger | \phi_l^- \rangle$, with associated poles at $\omega = E_f - E_l$. Note that for $\Delta V = 0$, this matrix element must be zero as the operator $c_{r\sigma}^\dagger$ does not change the symmetry of the state $|\phi_l^- \rangle$. Then, the resulting state $c_{r\sigma}^\dagger |\phi_l^- \rangle$ is odd under the reflection $\hat{O}_r$ and orthogonal to $|f^+ \rangle$. As a consequence of this symmetry, the transport Green function G_{lr} misses the poles at $\omega = E_f - E_l$ for the $|\phi_l^- \rangle$ states. However, these poles still appear in the density-of-states of the dot. In this case, the associated matrix element is of the form $\langle f^+ | c_{\alpha\sigma}^\dagger | \phi_l^- \rangle$. Since the operators $c_{\alpha\sigma}^\dagger$ and $c_{\beta\sigma}^\dagger$ do not commute with $\hat{O}_r$ (only the symmetric or antisymmetric combination of these operators commutes with $\hat{O}_r$), the state $c_{\alpha\sigma}^\dagger |\phi_l^- \rangle$ is not orthogonal to $|f^+ \rangle$.

This fact produces *localized states* in the QD due to the poles associated with the states $|\phi_l^- \rangle$ (located at $\omega = E_f - E_l$) that are present in $g_{\alpha\alpha}$ or $g_{\beta\beta}$ (the element $\langle f^+ | c_{r\sigma}^\dagger | \phi_l^- \rangle$ does not appear in the Green functions), but are not present in the Green function that transports electrons from one lead to the other ($g_{\alpha r}$ or g_{lr}). This fact explains the results observed in the right frame of Fig. 2.15. When ΔV is not zero, this is no longer valid because $\hat{O}_r$ does not commute with H_T .

The important consequence of this effect is that some states do not contribute with their resonances to the conductance. However, when one of these states crosses the Fermi level, the QD is charged abruptly. Then, the many-body properties of the system also change abruptly, producing the conductance discontinuities shown in Fig. 2.3.

One way to eliminate the symmetry between α and β is by using different intralevel Coulomb repulsions for the two levels. Figure 2.16 shows the conductance, phase difference, and partial conductances for $\Delta V = 0$ and $U_\beta = U_\alpha + 0.1t$. Note that the first discontinuity survives because for zero or one electrons in the QD, the Coulomb repulsions are not active and the symmetry is not broken. In this case, the physics related to the poles associated with the $\alpha - \beta$ symmetry discussed above can be applied, and the discontinuities shown in Fig. 2.2(a) appear.

2.6 Conclusions

In this work, we investigated the interference effects that can arise in the conductance of multilevel quantum dots. Although there are no obvious different paths, as in an AB interferometer, transport using different levels inside the QD can produce similar interference. Using an exact diagonalization plus embedding technique, the phase difference between these paths was calculated. When this phase difference is $\pm\pi$, a destructive interference can appear. An important aspect of these results is that the conductance cancellations can occur in the absence of external magnetic fields – usually used in AB experiments – and also without introducing at the Hamiltonian level any relative phase for the hopping amplitudes of the states involved. The Coulombic interaction is the main cause of the effect, which can also be visualized as an interference between $S_D = 1$ and $S_D = 1/2$ Kondo states.

Another novel effect found in this investigation is the possible formation of localized states in QDs. These states do not contribute to the conductance but they modify the many-body physics of the system through Coulombic effects. These localized states induce discontinuities in the conductance versus gate voltage for particular couplings, while in the most general case, conductance cancellations are observed.

The results presented in this work will hopefully provide motivation for experimentalists to refine their measurements to search for conductance cancellations. While our conductance versus gate voltage curves should be cautiously used to compare against experiments – since the details of the results vary as a function of parameters, and in addition most results were obtained using only two active levels – the ideas introduced here are expected to be robust. When multilevel quantum dots with interacting electrons are considered, cancellations are the rule more than the exception.

Experimental realizations of quantum dots with many active levels are possible. Recently, Aikawa *et al.* [66] observed that quantum-dot states can be classified into a small number of states that are strongly coupled to the leads, and a large number of states weakly coupled to those leads. The mixing between these states was found to be of relevance to explain transport experiments. It is clear that many theoretical calculations must be revisited to incorporate the multiplicity of levels in dots. This is also important if transport properties through atoms are studied. This is more challenging experimentally than the study of relatively larger size quantum dots, but possible. In the case of atoms, degeneracies are

natural in open-shell atoms, and a variety of interference processes as those observed here are possible.

CHAPTER 3

PREDICTION OF FERROMAGNETIC COUPLING IN COUPLED DOUBLE-LEVEL QUANTUM DOTS

3.1 Introduction

Strongly correlated electronic systems, such as high- T_c cuprates, heavy fermions, and manganites, display a variety of nontrivial collective states, which are difficult to analyze due to the many-body character of the interactions, and the difficulties in experimentally *controlling* the parameters determining these interactions. These problems are severe in materials that spontaneously grow in particular structures and patterns, with several effects (lattice, spin, charge, orbital) in direct competition. Therefore, the observation of a celebrated many-body effect, the Kondo effect, in a single quantum dot (QD) [1] has captured the attention of the strongly correlated community. It is conceivable that the most interesting states that are spontaneously stabilized in some materials - and are very difficult to control - could instead be artificially created in a man-made structure. In this framework, a natural first-step is to analyze coupled QDs. In fact, the two-impurity Kondo problem - extensively studied since the 80's [67, 68] - can now be realized in a real system. Moreover, recent investigations have reported antiferromagnetic (AF) correlations between two single-level coupled QDs, in competition with Kondo correlations [6, 69, 70, 71, 72, 73, 22]. As a consequence, it is now clear that two of the most remarkable magnetic states known to exist in spontaneously grown materials - the Kondo and AF states - have already found realizations in the context of QDs. However, the other dominant magnetic state of some materials - the ferromagnetic (FM) state - has comparatively received much less attention [5]. For the dream of artificially replicating collective states using QDs to be fulfilled, a realization of FM states must be achieved. The lack of attention to FM states in QDs should not be surprising in view of the physics of FM materials, such as manganites. Here, the

Figure 3.1: Schematic representation of the two double-level QDs system and the hopping amplitudes of Eqs. (1) and (3).

FM state is reached by removing electrons (doping) from an AF state. Under the constraint of having one particle per level (1/2-filling), and only one level active per QD as in most previous investigations, the double-exchange [74] generated FM state *cannot* be realized. To reach a FM state, more levels need to be active, resulting in *less* than one electron per level.

In this chapter, clear evidence is presented for the development of *ferromagnetic* correlations between two *double-level* QDs [75, 48, 55, 56, 49]: at 1/4-filling (one electron per dot), two coupled double-level QDs form a triplet state. Coupling this state to ideal metallic leads produces a Kondo resonance and a peak in the conductance. The results do not appear to be restricted to only a pair of QDs, but they seem valid for larger QD arrays. Basically here *it is reported a realization of the double-exchange mechanism using QDs*. Although the above mentioned effect is stronger if the appropriate intra-dot inter-level many-body interactions are added to the Hamiltonian, it is important to stress that these interactions are not necessary: considering just an intra-level Coulomb repulsion (Hubbard U) is enough to obtain qualitatively the same results, opening the possibility for the FM regime to be experimentally observable.

3.2 Model

Figure 3.1 schematically depicts the system analyzed here and introduces the labelling for the different tunneling parameters t' and t'' . To model this system, the Anderson impurity Hamiltonian that describes the two QDs with two levels (labeled α and β) is given by

$$\begin{aligned}
H_d = & \sum_{\sigma, i=1,2} [U/2(n_{i\alpha\sigma}n_{i\alpha\bar{\sigma}} + n_{i\beta\sigma}n_{i\beta\bar{\sigma}}) + V_g n_{i\alpha\sigma} + (V_g + \Delta V)n_{i\beta\sigma}] + \\
& \sum_{\sigma\sigma', i=1,2} [(U'/2)n_{i\alpha\sigma}n_{i\beta\sigma'} - Jc_{i\alpha\sigma}^\dagger c_{i\alpha\sigma'} c_{i\beta\sigma}^\dagger c_{i\beta\sigma}] - t'' \sum_{\sigma, \lambda=\alpha\beta} (c_{1\lambda\sigma}^\dagger c_{2\lambda\sigma} + \text{h.c.}), \quad (3.1)
\end{aligned}$$

where the first term represents the usual Coulomb repulsion between electrons in the same level (considered equal for both levels). The second term represents the Coulomb repulsion between electrons in different levels (the U' notation is borrowed from standard many-orbital studies in atomic physics). The third term is the energy of the levels regulated by the gate voltage V_g and the fourth term represents the Hund coupling ($J > 0$) that favors the parallel alignment of spins. To decrease the number of free parameters, all the calculations presented here assume the following relations: $U' = 2U/3$ and $J = U - U'$. As discussed later, the main result of this work does not depend on the specific values taken by U' and J . Note that α and β are separated by ΔV , and by modifying this parameter an interpolation between one- and two-orbital physics can be obtained. The last term represents the inter-dot coupling, with matrix element t'' . For simplicity, we assume that there is no hopping between levels α and β . The dots are connected to the leads (represented by semi-infinite chains) by a hopping term with amplitude t' , while $t = 1$ is the hopping amplitude in the leads (and energy scale). More specifically,

$$H_{\text{leads}} = -t \sum_{i\sigma} (c_{li\sigma}^\dagger c_{li+1\sigma} + c_{ri\sigma}^\dagger c_{ri+1\sigma} + \text{h.c.}), \quad (3.2)$$

$$H_{\text{d-leads}} = -t' \sum_{\sigma, \lambda=\alpha, \beta} (c_{1\lambda\sigma}^\dagger c_{l0\sigma} + c_{2\lambda\sigma}^\dagger c_{r0\sigma} + \text{h.c.}), \quad (3.3)$$

where $c_{li\sigma}^\dagger$ ($c_{ri\sigma}^\dagger$) creates an electron with spin σ at site i in the left (right) contact. Site “0” is the first site at the left of the left dot and at the right of the right dot, for each half-chain. The total Hamiltonian is $H = H_d + H_{\text{leads}} + H_{\text{d-leads}}$. Note that for $V_g = -U/2 - U' + J/2 - \Delta V/2$, the Hamiltonian is particle-hole symmetric. Using the Keldysh formalism [58, 59], the conductance through this system can be written as $G = \frac{e^2}{h} |t^2 G_{lr}(E_F)|^2 [\rho(E_F)]^2$. In practice, a cluster containing the interacting dots and a few sites of the leads is solved exactly, the Green functions are calculated, and the leads are incorporated through a Dyson Equation embedding procedure (ECA technique). In Figs. 3.2-3.4, the cluster used involves the two QDs plus one lead site at left and right.

3.3 Numerical Results

Figure 3.2: Results for the strong inter-dot tunneling regime ($t''/t' \gg 1$) showing the presence of a *ferromagnetic* state at 1/4-filling and concomitant Kondo effect: (a) Conductance (solid line), charge occupancy per level and per spin (dashed line), and total spin S_T in the four levels of the dots (dotted line) vs. gate potential V_g for two coupled double-level QDs. The values for the parameters are indicated. Note that the maximum in the conductance corresponds to a spin 1 Kondo peak that occurs when there is exactly one electron per dot and the total spin $S_T \approx 1.0$. The other two peaks occur when there are 1 and 3 electrons in the two-dots system, corresponding to $S_T = 1/2$ Kondo resonances. (b) DOS indicating the presence of a Kondo resonance associated with the $S_T = 1$ Kondo peak in (a). The numbers on the right side indicate the total number of electrons inside the two quantum dots. V_g takes the values 0.0, -0.5 , -0.83 , -1.12 , -1.5 from the top to the bottom curve. The Fermi level is located at $\omega/t = 0.0$.

In Fig. 3.2a, results for conductance (solid line) at strong inter-dot tunneling ($t''/t' \gg 1$) are shown. The main feature displayed is the peak at $V_g \approx -1.0$ (at and near 1/4-filling). The occupancy for this value of V_g is approximately one electron per dot (dashed line $\times 4$) and the total spin S_T of the four levels¹ (dotted line) is ≈ 1.0 . The smooth charging of the

¹Note that the total spin in the four levels in the two quantum dots (denoted as S_T and obtained through

levels as the gate potential decreases (in the peak region) indicates a possible Kondo regime. This is confirmed in Fig. 3.2b, where the density of states (DOS) close to the Fermi level is displayed as the gate potential varies from 0.0 to -1.5 (top to bottom). Through this variation of V_g the two dots are charged with one additional electron (the total mean charge varies from ≈ 1.6 to 2.6). One can clearly see a Kondo resonance pinned to the Fermi level. For lower values of the gate potential (in the region at and near $1/2$ -filling, with 2 electrons per dot) the conductance is drastically reduced and the total spin S_T inside the dots reaches its minimum value, indicating the formation of a global singlet state. Calculations of the total spin in each dot indicate that this singlet state is formed by the AF coupling of two spins $S = 1$. A description of how this picture changes as t'' decreases is shown in Fig. 3.3a, where results are shown for five different values of t'' .

The conductance at the particle-hole symmetric point, $V_g = -5.0$, varies from zero for $t'' = 1.0$, to 1.0 (in units of e^2/h per spin channel) for $t'' = 0.08$. Figure 3.3b shows how the Kondo correlation (between the total spin in the dots and the conduction electrons in the first site of one of the leads) evolves from a negligible value for $t''/t' \gg 1$ to a large value (≈ -0.65) for $t''/t' < 1$. The inset of Fig. 3.3a displays the change of the total spin (from $S_T \approx 0.0$ to $\approx 3/4$) as t'' decreases. The two main peaks in the conductance discussed up to now were the $S_T = 1$ Kondo peak at $1/4$ -filling (relevant in the strong inter-dot tunneling regime) and the peak at $1/2$ -filling (relevant in the weak inter-dot tunneling regime). It is interesting to discuss how these peaks evolve as ΔV increases. Figure 3.4a shows the results for $t''/t' \gg 1$ ($t'' = 1.0$, $U = 5.0$ and $t' = 0.2$). The solid line displays the conductance and the dotted line displays the total spin S_T . Level separation ΔV increases from bottom to top (values for each graph are displayed in the left side). From $\Delta V = 0.0$ up to $\Delta V \approx 0.6$, the width of the conductance peak slowly decreases, and so does the maximum value of S_T . Above $\Delta V \approx 0.7$ (not shown) the narrowing of the peak accelerates (as does the decrease of S_T), until the peak has vanished for $\Delta V = 1.0$. For higher values of ΔV (top graph, $\Delta V = 10.0$), the conductance shows the typical Coulomb blockade profile previously discussed [22] for coupled single-level dots when $t''/t' \gg 1$.

Figure 3.4b shows the corresponding results for $t''/t' < 1$ ($t'' = 0.08$, $U = 5.0$ and $t' = 0.2$).

$\langle \vec{S}_T^2 \rangle_{\text{cluster}} = S_T(S_T + 1)$, where $\vec{S}_T = \sum_{i,\lambda} \vec{S}_{i,\lambda}$, where i labels the dots and λ labels the levels) is not a good quantum number (since the dots are not isolated). However, S_T gives a good indication on the nature of the Kondo effect ($S = 1/2$ or $S = 1$).

Figure 3.3: (a) Conductance G vs. gate potential V_g for different values of inter-dot tunneling ($t'' = 1.0, 0.6, 0.4, 0.2$, and 0.08 , see convention in (b)). V_g varies across the region where the fourth electron is charged into the double-dot system. Note that the conductance at $V_g = -5.0$ gradually increases from zero (at strong tunneling, $t'' = 1.0$) to the maximum value (at weak tunneling, $t'' = 0.08$). This variation indicates a transition from two $S = 1$ spins (in each dot) forming a global singlet to two uncorrelated $S = 1$ spins, each forming a Kondo resonance with the lead conduction electrons. The inset shows the variation with t'' of the total spin inside the dots. (b) Variation with t'' of the spin-spin correlation between the total spin in the double-dot and a conduction electron located in the first site of the leads.

Note that the central peak does not change appreciably from $\Delta V = 0.0$ to $\Delta V = 2.0$. In fact, changes start only above $\Delta V = 3.0$, when the central peak splits in two ($\Delta V \gtrsim 3.2$, not shown). For $\Delta V > 3.4$ the two peaks start moving farther apart from each other and become very narrow. Finally, for $\Delta V = 4.0$ the central peaks have disappeared, and the remaining structures are already similar to the single-level result $t''/t' < 1$. The top graph ($\Delta V = 30.0$) is basically the result reported for single-level QDs at weak interdot tunneling [22] (if one discards the slight shoulders in the internal peaks).

Based on the results displayed in Fig. 3.4a, a qualitative phase diagram for the strong

Figure 3.4: Variation of the conductance with level separation ΔV . The level separation is indicated for each curve. (a) $t'' = 1.0$: The gate potential decreases down to the particle-hole symmetric value, to highlight how the $S_T = 1$ Kondo peak varies with ΔV . Note that all graphs have the same horizontal axis scale, except for the upper one (scale indicated on top). (b) $t'' = 0.08$: The gate potential varies down to the lowest value (total charging of the dots), to highlight the variation of the central peak (at the particle-hole symmetric point). Here also the upper graph has a different horizontal scale. A discussion of how the data interpolate between double- and single-level dots is given in the text.

Figure 3.5: Qualitative phase diagram for the strong inter-dot tunneling regime ($t''/t' \gg 1$). Horizontal axis indicates the level occupancy (controlled by V_g) and vertical axis indicates the level separation (controlled by ΔV). In the 1/2-filling region, displayed in the left side, one goes from an AF coupling between two $S = 1$ spins (for $\Delta V < V_g$) to a situation where the two lower levels are completely occupied (for $\Delta V > V_g$). In both cases there is no Kondo effect. On the other hand, in the 1/4-filling region (right side), for $\Delta V < t''$ one has the novel $S_T = 1$ FM Kondo region, which gives way to an AF region (with no Kondo effect) once $\Delta V > t''$. The regime with 3 electrons in the two dots is very narrow as a function of V_g and it is not shown.

inter-dot tunneling regime can be sketched. In Fig. 3.5, the electron occupancy is in the horizontal axis (controlled by V_g) and ΔV is in the vertical axis. The left side (indicating 1/2-filling) is dominated by antiferromagnetism for all values of $\Delta V \lesssim U$. The singlet formed by the four levels is made of two spins $S \approx 1.0$. For $\Delta V > U$ one recovers the single-level picture. The right side of the phase diagram, which describes the evolution of the central peak in Fig. 3.4a, is more interesting. For $\Delta V < t''$ one has the novel FM region, where $S_T = 1$ Kondo effect is present. For $\Delta V > t''$ an AF region is found, with no Kondo effect.

A finite size scaling analysis was done (results not shown) to verify how our numerical results converge with cluster size. We found very little change in the results with increasing cluster size, giving us confidence that all the qualitative results here discussed are not caused by finite-size effects. It is also important to stress that the calculations presented in Fig. 3.2a were reproduced for $U' = J = 0.0$, with the values of all other parameters kept the same as before ($U = 5.0$, $t'' = 1.0$ and $t' = 0.2$), and the results obtained barely changed. This indicates that the FM correlation and the $S_T = 1$ Kondo should be experimentally observable, since the only requirement is to have two double-level QDs with strong inter-dot tunneling.²

Figure 3.6: Schematic representation of the main result in this chapter.

3.4 Conclusion

Figure 3.6 qualitatively summarizes the main result presented in this chapter: (a) For double-level coupled quantum dots in the strong inter-dot tunneling regime at 1/4-filling, FM correlations develop and conductance through a Kondo channel is allowed. (b) On the other hand, single-level coupled QDs develop AF correlations in the strong inter-dot tunneling regime and conductance is suppressed. The results discussed in this chapter complete the analogy between QD states and magnetic phenomena in bulk materials. Previous investigations had shown that Kondo and AF states were possible in QDs. Now, at least theoretically, a regime with ferromagnetism has also been found, if more than one level per

²One can understand why an *effective* FM J_{eff} is generated (even in the absence of the Hund J and the U' terms) by using the following argument: At 1/4-filling (two electrons in the four levels), the hopping from one lead to the adjacent dot (and vice-versa) is maximized when the spins of the electrons in the levels α and β in the same QD are parallel to each other. A strong inter-dot tunneling then generates an effective FM coupling between the QDs. Therefore, the only restriction for the FM coupling seems to be that the leads should have just one conduction channel.

dot is active. Certainly, it would be important to confirm experimentally this prediction. Our calculations emphasizing multilevel dots present analogies with multi-orbital materials such as manganites, nickelates, cobaltites, and ruthenates. These compounds have a plethora of phases, all of which could find realizations in QDs systems as well.

CHAPTER 4

TRANSPORT PROPERTIES OF STRONGLY CORRELATED ELECTRONS IN QUANTUM DOTS USING A SIMPLE CIRCUIT MODEL

4.1 Introduction

The observation of the Kondo effect in a single quantum dot (QD) [1] and the subsequent theoretical and experimental studies of more complex structures, such as two QDs directly coupled through a tunable potential barrier [73], has provided impetus for the analysis of more elaborate systems. In a recent seminal work, Craig *et al.* [5] report on the possible laboratory realization of the two-impurity Kondo system. Two similar QDs are coupled through an open conducting central region (CR). A finite bias is applied to one of the QDs (QD1 from now on) as well as to the CR, while the other QD (QD2) is kept at constant gate potential. The differential conductance of QD1 is then measured for different charge states of QD2 and different values of its coupling to the CR. The main result was the suppression and splitting of the zero-bias-anomaly (ZBA) in QD1 by changing the occupancy of QD2 from even to odd number of electrons and by increasing its coupling to the CR. A Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between the QDs was suggested as an explanation for the observed effects [76, 37]. The importance of Craig *et al.*'s experiments cannot be overstated: the possibility of performing nonlocal spin control in a system with two lateral QDs has potential applications in QD-based quantum computing [11].

In this work, numerical simulations in good agreement with the experiments are presented. The central conclusion of this work is that our computational data, and as a consequence the experimental results, can be explained using a very simple “circuit model”, where one of the elements is a T-connected QD that has an intrinsic reduction of conductance with varying biases. This proposal is an alternative to the more standard RKKY ideas.

Figure 4.1: (a) Experimental setup used in Ref. [5]. (b) Illustration of the model studied in this work (see text for details).

4.2 Model Hamiltonian

Figure 4.1a depicts the experimental set up used in the measurements of Craig *et al.* [5] with the labeling used in this work. Figure 4.1b is a schematic representation of the system, introducing two different tunneling parameters (hopping matrix elements t' and t'') and the Coulomb repulsion U in each QD (assumed the same for simplicity). To model this system, the Anderson impurity Hamiltonian is used for both QDs:

$$H_d = \sum_{i=1,2;\sigma} [U/2n_{i\sigma}n_{i\bar{\sigma}} + V_{gi}n_{i\sigma}], \quad (4.1)$$

where the first term represents the usual Coulomb repulsion between two electrons in the same QD, and the second term is the effect of the gate potential V_{gi} over each QD. QD1 is directly connected to the left lead and to the CR with hopping amplitude t' , while QD2 is connected only to the CR (with hopping amplitude t''), which itself is connected to the right lead with hopping amplitude t (which is also the hopping amplitude in both leads, and our energy scale). In summary,

$$H_{\text{leads}} = -t \sum_{i\sigma} [c_{li\sigma}^\dagger c_{li+1\sigma} + c_{ri\sigma}^\dagger c_{ri+1\sigma} + \text{h.c.}], \quad (4.2)$$

$$H_{12} = \sum_{\sigma} \left[-t' c_{1\sigma}^\dagger (c_{l0\sigma} + c_{CR\sigma}) - t'' c_{2\sigma}^\dagger c_{CR\sigma} - t c_{CR\sigma}^\dagger c_{r0\sigma} + \text{h.c.} \right], \quad (4.3)$$

where $c_{li\sigma}^\dagger$ ($c_{ri\sigma}^\dagger$) creates an electron with spin σ at site i in the left (right) lead. The CR is composed of one tight-binding site,¹ unless otherwise stated. Site ‘0’ is the first site at the left (right) of QD1 (CR) in the left (right) lead. The total Hamiltonian is $H = H_d + H_{\text{leads}} + H_{12}$. Note that for $V_{g1} = V_{g2} = -U/2$, the Hamiltonian is particle-hole symmetric. To calculate the conductance G , using the Keldysh formalism [58], a cluster containing the interacting dots and a few sites of the leads is solved exactly,² the Green functions are calculated, and the leads are incorporated through a Dyson Equation embedding procedure (ECA technique). All the results shown were obtained for $U = 0.5$, $t' = 0.2$, zero-bias, and zero temperature.

4.3 Numerical Results

In Fig. 4.2, results for the conductance across QD1 (solid curves) and for the occupancy per spin orientation $\langle n_2 \rangle$ of QD2 (dashed curves) are presented. In Fig. 4.2a, $t'' = 0.2$ and V_{g2} varies from -2.0 to -0.3 . For $V_{g2} = -2.0$ (red), QD2 is occupied by two electrons ($\langle n_2 \rangle = 1$) and the conductance of QD1 displays the characteristic Kondo behavior reported before.³ For $V_{g2} = -0.35$ (green) the average value of $\langle n_2 \rangle$ decreases to ≈ 0.7 (≈ 1.4 electrons in QD2) and $\langle n_2 \rangle$ now depends on V_{g1} . In addition, G decreases in comparison with the results obtained for $V_{g2} = -2.0$. Then, these numerical results are qualitatively in agreement with the experimental results shown in Fig. 2 of Craig *et al.* [5], namely, by decreasing the occupancy of QD2, from even to odd number of electrons, the ZBA in QD1 is suppressed. As V_{g2} is further increased (-0.3 (blue)) a *qualitative* change occurs: For values of V_{g1} where $\langle n_2 \rangle \approx 0.5$ (QD2 singly occupied), the conductance of QD1 vanishes and therefore there is a narrow dip in G . This splitting of the ZBA is remarkably similar to that observed in Fig. 3A of the experimental results [5]. For finite-temperature calculations, the dip in G will not reach zero, resembling even better the experiments [77].

To further test the similarities between simulations and experiments, in Figs. 4.2b and 4.2c, results for G and $\langle n_2 \rangle$ are shown for fixed V_{g2} and different t'' values. In Fig. 4.2b, where $V_{g2} = -0.5$, as t'' increases from 0.2 to 0.5 there is only a slight decrease of G . This is accompanied by a slight decrease in the average value of $\langle n_2 \rangle$, from ≈ 0.9 to ≈ 0.7 . A more dramatic change is obtained in Fig. 4.2c, where $V_{g2} = -0.25$, and t'' varies from 0.0

¹Calculations for a CR with a larger number of sites did not present any qualitatively new results.

²Results shown include one site on each side. Calculations with more sites were done, but no significative size-effects were observed.

³Removing QD2 results in essentially the same conductance as the one for $V_{g2} = -2.0$.

Figure 4.2: (a) Variation of G with V_{g1} in QD1 (solid curves) and variation of $\langle n_2 \rangle$ (occupancy of QD2 per spin orientation - dashed curves) for $t'' = 0.2$ and three different values of V_{g2} . For $V_{g2} = -2.0$ (red), QD2 is occupied by 2 electrons ($\langle n_2 \rangle = 1$) for any V_{g1} and the conductance through QD1 is essentially the same as if QD2 was not present. For higher values of V_{g2} , the average value of $\langle n_2 \rangle$ decreases and becomes dependent on V_{g1} (decreasing for lower values of V_{g1}). This is accompanied by a suppression of the ZBA (for -0.35 (green)) and also by a splitting of the ZBA (for -0.3 (blue)). (b) Variation of G and $\langle n_2 \rangle$ with t'' (0.2, 0.4, 0.5) at a fixed value of $V_{g2} = -0.5$. As the value of t'' increases, the average value of $\langle n_2 \rangle$ decreases and this is again accompanied by a suppression of the ZBA. (c) Same as in (b), but now for $V_{g2} = -0.25$ (particle-hole symmetric point) and $t'' = 0.0, 0.1$, and 0.2 . Note that G vanishes at $V_{g1} = -0.25$, where $\langle n_2 \rangle = 0.5$, for all finite values of t'' .

to 0.2. By increasing t'' from 0.0 (red curves) to 0.1 (green), the ZBA is now split in two and $\langle n_2 \rangle$ acquires a dependence on V_{g1} . As t'' further increases (0.2 (blue)), the dip becomes wider, the two side-peaks decrease and G still vanishes for $\langle n_2 \rangle = 0.5$ (one electron in QD2). Our calculations show that, if $\langle n_2 \rangle$ varies around 0.5, the dip in G is present for all finite values of t'' , with a width proportional to t'' . Comparing the results in Figs. 3A and 3B of Craig *et al.* [5] with Figs. 4.2c and 4.2b in this chapter, respectively, one notices a striking similarity: The splitting of the ZBA observed in the experimental results (their Fig. 3A), when the number of electrons in the control QD is odd and the coupling to the central region is increased, is very similar to the dip in G for all finite- t'' curves in Fig. 4.2c (as mentioned

Figure 4.3: Schematic representation of the main ideas behind the “circuit model”. In (a), the system represented in Fig. 4.1b is divided into its constituent elements: QD1 is modeled as a QD connected in series with the leads and QD2 is modeled as a side-connected QD. The curves in (b) represent the conductance and occupancy of each separate circuit element vs. the applied gate potential. (c) Schematic representation of how the two individual elements are connected to form the final ‘circuit’: Incident and reflected wave amplitudes are represented in the right side of QD2 by black arrows. A transmitted wave through QD2 undergoes multiple reflections between the two quantum dots until it is finally transmitted past QD1. The superposition of all these processes results in the final conductance for the ‘circuit’.

above, at finite temperatures, one expects that the dip in G will not reach zero). When the occupancy of QD2 is even (Fig. 3B in the experimental results [5] and Fig. 4.2b in this chapter), the G dependence on t'' is much less significant and the splitting of the ZBA does not occur.

4.4 Circuit Model

What is the origin of these results? Below, it is argued that a qualitative description of the results can be achieved by analyzing the two quantum dots through a so-called ‘circuit model’. This model starts with the conductance of each QD calculated separately, as independent elements of a circuit, and then the conductance of the ‘complete circuit’ is obtained by combining the conductances of the two elements connected in series. Figure 4.5 describes schematically the steps involved in this approach. In Fig. 4.5a, the complete system formed

of QD1 and QD2 (shown in Fig. 4.1b) is divided into two components. QD1 is modeled as a QD connected directly to left (L) and right (R) leads, while QD2 is modeled as a side-connected QD [78]. Figure 4.5b shows the respective conductances and occupancies for each independent element vs. gate voltage, and Fig. 4.5c represents the scattering processes (represented by transmission and reflection amplitudes) that an electron undergoes while moving through the complete ‘circuit’. The superposition of all these processes leads to the total transmittance (proportional to the conductance) for the circuit model. This can be calculated in two ways: coherently or incoherently [79]. Since there is no qualitative difference between them, and in order to keep the simplicity of the model, we present the incoherent results. The equation which provides the final transmittance for the processes depicted in Fig. 4.5c is

$$T = \frac{T_1 T_2}{1 - R_1 R_2}, \quad (4.4)$$

where the transmittances T_1 and T_2 are proportional to the conductances of QD1 and QD2, as depicted in Fig. 4.5b, and $R_{1(2)} = 1 - T_{1(2)}$ are the reflectances. To calculate T , one needs to establish how T_2 depends on V_{g1} . The natural way to do that is to use the dependence of $\langle n_2 \rangle$ on V_{g1} , as depicted in Fig. 4.2, and then use the relation between conductance and occupancy, as shown in the red curves in Fig. 4.5b. In other words, the functional relation can be expressed as $T_2 = T_2[\langle n_2 \rangle(V_{g1})]$. It is not surprising that in a strongly correlated system like the one being analyzed here, the variation of the gate potential of QD1 influences the charge occupancy of QD2, and in turn this influences the conductance through QD1.

In Fig. 4.4, conductance results using Eq. 4.4 are shown for the same parameters as in Fig. 4.4. Although the quantitative agreement varies, there is a good overall qualitative agreement. All the trends are correctly reproduced and some of the details are quite similar, such as for example the asymmetric shape of the curves at higher values of V_{g2} (-0.35 and -0.3) in Fig. 4.4a. It is important to notice that there are *no* adjustable parameters in the circuit model presented here. The only input necessary is $\langle n_2 \rangle$ vs. V_{g1} , which is obtained through a calculation for the complete system (values displayed for $\langle n_2 \rangle$ in Fig. 4.2). The success of the circuit model implies that the dip in G arises from the Fano anti-resonance which cancels the conductance of QD2 (red solid curve in Fig. 4.5b). The Fano anti-resonance can be seen as a destructive interference process between two different trajectories an electron can take on its way to QD1: it can cross the CR without passing through QD2;

Figure 4.4: Same as in Fig. 4.2, but now using the ‘circuit model’ for the calculations.

or it can visit QD2, return to the CR and then proceed to QD1 [78].

4.5 Discussion

The similarities between the experimental results and our simulations suggest that our model and numerical technique have captured the essential physics of the experiments. However, these same experiments have also been explained using RKKY ideas [37, 76]. Can our numerical results be also understood in this alternative context? To try to answer this question, several calculations were performed with different parameter values and number of sites in the CR. In Fig. 4.3a, results for spin correlations between QD1 and QD2 (denoted $\mathbf{S}_1 \cdot \mathbf{S}_2$) are presented for the same parameters used in Fig. 4.2c. At $t'' = 0.0$ (red curve) QD1 and QD2 are uncorrelated as expected. As t'' increases to 0.1 (green), and then 0.2 (blue), it is observed that in the region where G reaches its maximum value (see Fig. 4.2c), $\mathbf{S}_1 \cdot \mathbf{S}_2$ also assumes a maximum value and it is positive (ferromagnetic (FM)). For $t'' > 0.2$ (not shown), $\mathbf{S}_1 \cdot \mathbf{S}_2$ saturates and starts decreasing. The maximum of $\mathbf{S}_1 \cdot \mathbf{S}_2$, for all values of t'' , decreases even further as the size of the central region increases (the results in Fig. 4.3a are for a CR with just one site). In addition, the sign of $\mathbf{S}_1 \cdot \mathbf{S}_2$ alternates as the size of the CR

Figure 4.5: (a) Spin correlation $\mathbf{S}_1 \cdot \mathbf{S}_2$ between QD1 and QD2 for the same parameters as in Fig. 4.2c. For $t'' = 0.0$ (red curve), the two QDs are uncorrelated ($\mathbf{S}_1 \cdot \mathbf{S}_2 = 0$). For finite t'' (0.1 (green) and 0.2 (blue)), $\mathbf{S}_1 \cdot \mathbf{S}_2$ is FM and reaches its maximum value in the region where G is maximum. (b) Kondo correlations $\mathbf{S}_1 \cdot \mathbf{S}_c$ between QD1 and the central site for the same parameters as in (a). All values are AF and they decrease in amplitude as t'' increases, underscoring the decrease of the Kondo effect as the FM correlation between QD1 and QD2 increases (compare with (a)). (c) Variation of G as U_1 (Hubbard interaction in QD1) assumes the values 0.4, 0.2 and 0.0. Note that the dip in G becomes slightly narrower as U_1 decreases, however it does not disappear.

increases and the QDs move farther apart from each other. In Fig. 4.3b, results for the spin correlation between QD1 and its neighboring site in the CR (denoted $\mathbf{S}_1 \cdot \mathbf{S}_c$) are shown for the same parameters as in Fig. 4.3a. $\mathbf{S}_1 \cdot \mathbf{S}_c$ is a rough measure of the Kondo correlation in QD1, having a direct connection with the ZBA in Fig. 4.2c. Indeed, for $t'' = 0.0$ (red) when G reaches the unitary limit, a robust antiferromagnetic (AF) correlation develops between QD1 and its neighboring site in the CR. For $t'' = 0.1$ (green), despite the narrow dip in G , the side-peaks are still close to the unitary limit (see Fig. 4.2c) and $\mathbf{S}_1 \cdot \mathbf{S}_c$ is still strongly AF. However, for $t'' = 0.2$ (blue), both G and $\mathbf{S}_1 \cdot \mathbf{S}_c$ are strongly suppressed, in qualitative agreement with a suppressed ZBA due to a weakened Kondo resonance.

The results thus far seem to indicate that the CR could be mediating a long range coupling between QD1 and QD2, with the characteristics of RKKY interaction. However,

the magnitude of the maximum value of $\mathbf{S}_1 \cdot \mathbf{S}_2$ (see scale in Figs. 4.3a-b) is too small to account for all the effects observed in the conductance in Fig. 4.2c. One possible way of increasing $\mathbf{S}_1 \cdot \mathbf{S}_2$ is by coupling QD1 more strongly to the CR than to the left lead. This was exactly the setup chosen in Ref. [5], where those authors performed the measurements with asymmetric couplings to the left (Γ_L) and right (Γ_{CR}) sides of QD1. In fact, the voltages applied to the gates in Fig. 4.1a were such that $\Gamma_{CR} \gg \Gamma_L$. In our model, this is equivalent to having an asymmetric t' , with $t'_{CR} \gg t'_L$. An analysis of the results in this asymmetric regime indicates that the correlation between QD1 and QD2 does indeed increase. However, if one performs the calculations with the sites in the CR at a filling lower than one electron per site (half-filling), it is observed that $\mathbf{S}_1 \cdot \mathbf{S}_2$ is gradually suppressed as the electron filling falls to a more appropriate level to simulate the two-dimensional electron gas in the CR. Although one can argue that some of the dependence of the conductance of QD1 on the charge state of QD2 seen in Fig. 4.2 is associated to the correlations between the two dots, it is apparent that other effects are also present. This is dramatically exemplified by the fact that the cancellation of G presented in Fig. 4.2c occurs for *any* finite value of t'' , and of course for $t'' \approx 0$, one finds that $\mathbf{S}_1 \cdot \mathbf{S}_2 \approx 0$. The fact that the dip seen in the conductance in Fig. 4.2c is not dominantly caused by correlations between the dots can be made more clear by checking the results for the conductance as U_1 (Hubbard interaction in QD1) is reduced to zero. In Fig. 4.3c, results for G are shown for 3 different values of U_1 , for the same parameters as for the blue curve in Fig. 4.2c. As U_1 decreases from 0.4 (blue) to 0.2 (green), and then to 0.0 (red), the dip in the conductance remains, only becoming narrower, indicating that its origin is not associated with many-body interactions, but more likely with cancellations typical of T-geometries [78] that occur even in the non-interacting limit.

4.6 Conclusion

In summary, the numerical results qualitatively reproduce the main aspects of important recent experiments [5] involving nonlocal spin control in nanostructures. The main result is that the splitting observed in the ZBA is caused by a cancellation in the conductance due to a destructive interference. This so-called Fano anti-resonance has its origin in one of the dots being side-connected to the current's path. A simple 'circuit model' qualitatively reproduces the experiments and offers an alternative to a purely RKKY interpretation of the results,

underscoring that a laboratory realization of the two-impurity Kondo system should avoid any geometry susceptible to a Fano anti-resonance.

CHAPTER 5

ELECTRON TRANSPORT THROUGH A MOLECULAR CONDUCTOR WITH CENTER-OF-MASS MOTION

5.1 Introduction

Molecular electronics has received much attention in the past decade, particularly since it became possible to fabricate devices in which the active element is a single molecule [80, 81, 82, 83]. A fundamental property of molecular conductors is their discrete electronic spectrum. Although the weak coupling of the molecule to the two metallic electrodes leads to the broadening of the molecular energy levels, their discrete nature is maintained. Due to the small size of these molecules, electronic correlations are dominant and lead to interesting many-body effects, such as the Coulomb blockade and Kondo resonance [50, 51, 21, 84]. These effects have been observed experimentally in molecular conductors [2, 23, 24] and other nanostructures [1, 85, 86]. Another interesting property of molecules is their flexible nature. They have an intrinsic spectrum of internal vibrational modes and, when coupled to the electrodes, some molecules acquire external vibrational modes as well. The excitation of one or more of these modes leads to the modulation of the electronic energy levels and tunneling barriers between the molecule and the electrodes or between different parts of the molecule, thus changing the transport properties. These vibrational effects have been observed in a number of recent experiments [25, 24], and have been the subject of considerable theoretical investigation [27, 28, 29, 26, 30]. Several efforts have focused on the “shuttle effect”, both in the classical [34] and quantum regimes [32, 31, 33, 35, 36], a molecule’s motion between the two electrodes that can give rise to a new mechanism for electron transport at finite bias.

In this work, we study the low bias conductance of a molecular conductor model with one relevant electronic energy level, both with interacting and noninteracting electrons.

Figure 5.1: A schematic of the system studied in this chapter. The molecule can oscillate between the two electrodes around the equilibrium position l_0 , thus modulating the tunnel barriers.

The molecule is allowed to oscillate between the two electrodes. This center-of-mass (CM) vibrational mode is treated quantum mechanically and leads to an asymmetric modulation of the molecule-electrodes tunneling barriers. The results show an interesting and unexpected conductance cancellation when an odd number of electrons occupy the molecule. It is discussed below that this cancellation is due to the *destructive interference between the purely electronic and phonon-assisted tunneling channels*, which are found to carry opposite phases [87, 88]. In this case both channels are elastic. The phonons are virtual, not thermal.

5.2 Model and Numerical Technique

Figure 5.1 schematically depicts the system analyzed in this work. The molecule can oscillate between the source and drain electrodes, thus modulating the tunneling barriers. In our calculations, this modulation and the electron-vibration coupling are expanded up to the linear term¹ [89]. The electronic part of the system is modeled using the Anderson impurity Hamiltonian. In general, the vibrational excitation also couples to the net charge of the molecule. This is due to charge images, charge traps and impurities in the electrodes, and due to the electric field corresponding to the small bias applied in any transport experiment. The total Hamiltonian can be written as $\hat{H} = \hat{H}_M + \hat{H}_{\text{leads}} + \hat{H}_{M-\text{leads}}$, where $\hat{H}_M$ is the Hamiltonian of the molecule,

¹Some results were gathered keeping up to the second order in the tunneling barriers dependence on the molecule position, and the conclusions are unchanged

$$\hat{H}_M = V_g \hat{n}_d + U \hat{n}_{d\uparrow} \hat{n}_{d\downarrow} + \lambda(1 - \hat{n}_d)(a + a^\dagger) + \omega_0 a^\dagger a. \quad (5.1)$$

The first term represents the energy of the relevant molecular orbital controlled by the gate voltage, the second term represents the Coulomb repulsion between the electrons occupying the molecular orbital, the third term couples the vibrational excitation to the net charge on the molecule ($a^\dagger$ and a are the phonon creation and annihilation operators), and the fourth term represents the vibrational energy. $\hat{H}_{\text{leads}}$ describes the two leads modeled here as semi-infinite ideal chains, $\hat{H}_{\text{leads}} = -t \sum_{i\sigma} (c_{li\sigma}^\dagger c_{li+1\sigma} + c_{ri\sigma}^\dagger c_{ri+1\sigma} + h.c.)$, where $c_{li\sigma}^\dagger$ ($c_{ri\sigma}^\dagger$) creates an electron with spin σ at site i in the left (right) lead. t is the hopping amplitude in the leads and the energy scale ($t = 1$). $\hat{H}_{M-\text{leads}}$ connects the molecule to the leads,

$$\hat{H}_{M-\text{leads}} = -t'[1 - \alpha(a + a^\dagger)] \sum_{\sigma} (d_{\sigma}^\dagger c_{l0\sigma} + h.c.) - t'[1 + \alpha(a + a^\dagger)] \sum_{\sigma} (d_{\sigma}^\dagger c_{r0\sigma} + h.c.), \quad (5.2)$$

where $d_{\sigma}^\dagger$ creates an electron with spin σ in the molecule, t' is the hopping parameter between the molecule and the first site of each lead, and α is a parameter that carries the dependence of t' on the molecule displacement from its equilibrium position $\hat{x}$ (note the opposite signs in this dependence for the two leads). This displacement can be written in terms of the phonon operators as $\hat{x} = (a + a^\dagger)$. The total Hamiltonian is invariant under the particle-hole and ($a \rightarrow -a$) transformation [90]. In the results shown, unless otherwise stated, the following set of parameters was used ($U = 1.0, t' = 0.2, \omega_0 = 0.2$) while λ and α were varied. The value of ω_0 was fixed since its increase or decrease would simply produce the opposite effect of increasing or decreasing λ and/or α .

Using the Keldysh formalism [58, 59, 91], the zero bias and zero temperature conductance can be written as $G = \frac{2e^2}{h} |t^2 G_{lr}(\epsilon_F)|^2 [\rho(\epsilon_F)]^2$, where G_{lr} is the Green function that propagates an electron from the left to the right lead and $\rho(\epsilon_F)$ is the density of states in the leads at the Fermi level. Note that at zero bias only elastic processes can be observed. The Green's functions are calculated using exact diagonalization supplemented by a Dyson equation embedding procedure (Embedded cluster approximation technique).

5.3 Results

It is useful to start by studying briefly the model when the tunnel barriers do not depend on the vibrational excitation ($\alpha = 0$). This corresponds to the Anderson-Holstein model.

Figure 5.2: (a) NRG and (b) ECA results for G as a function of V_g in the Kondo Regime for $\alpha = 0$ and increasing the electron-phonon coupling strength ($\lambda/\omega_0 = 0.0, 0.4, 0.8$, and 1.2). For small λ/ω_0 , the standard Kondo resonance is obtained with renormalized parameters. In the limit of large λ/ω_0 , the charge Kondo effect is obtained.

By applying the canonical transformation $\hat{U} = e^{-\frac{\lambda}{\omega_0}(\hat{n}_d - 1)(a^\dagger - a)}$ [92, 28], this model can be mapped into:

$$\begin{aligned} \hat{U}^\dagger \hat{H} \hat{U} = & (V_g + \lambda^2/\omega_0) \hat{n}_d + (U - 2\lambda^2/\omega_0) \hat{n}_{d\uparrow} \hat{n}_{d\downarrow} + \omega_0 a^\dagger a \\ & - t' \sum_{\sigma, \gamma=l,r} [e^{\frac{\lambda}{\omega_0}(a^\dagger - a)} d_\sigma^\dagger c_{\gamma 0 \sigma} + h.c.] + \hat{H}_{\text{leads}}, \end{aligned} \quad (5.3)$$

that is, an effective electronic model with renormalized parameters: $\tilde{U} = U - 2\lambda^2/\omega_0$, $\tilde{V}_g = V_g + \lambda^2/\omega_0$ and $\tilde{t}' \propto t' \exp(\frac{-\lambda^2}{2\omega_0^2})$. Figure 5.2 shows the conductance G of the molecule as a function of the gate voltage V_g for different values of λ/ω_0 . For weak electron-phonon coupling ($\tilde{U} > 0$), G shows the standard spin Kondo peak with a width that decreases with λ . For strong electron-phonon coupling ($\tilde{U} < 0$), the “charge Kondo effect” is observed. That is, the charge degeneracy between states with zero and two electrons on the molecule gives rise to a narrow conductance peak at $V_g = -U/2$. This model was previously studied using NRG [28], and the agreement between NRG and ECA results is quite good.

We now move to the center-of-mass motion case, i.e. $\alpha \neq 0$. Figure 5.3 contains the main results of this work. Figure 5.3a shows the conductance in the interacting electrons case. For $\alpha = 0$, the conductance simply shows a Kondo resonance peak with reduced width. However, when $\alpha \neq 0$, a conductance *dip* is obtained when an odd number of electrons occupy the

Figure 5.3: (a) G as a function of V_g in the Kondo regime for $\alpha = 0$ (solid line) and $\alpha = 0.4$ (dashed line), $\lambda = 0.2$ in both cases. In the first case, the usual Kondo peak with reduced width is obtained. In the second case, a conductance dip is obtained. (b) Average occupation of the molecular level for the same set of parameters. Note that the charging behavior is almost the same in both cases. (c) G vs. V_g in the absence of Coulomb repulsion ($U = 0$), for $\alpha = 0$ (solid line) and $\alpha = 0.4$ (dashed line), $\lambda = 0.1$ in both cases. The conductance dip is also obtained in this case, thus the physical mechanism behind this effect does not depend on the electron-electron interaction.

molecule. Figure 5.3b provides the average occupation $\langle n_d \rangle$ of the molecular orbital where it can be clearly seen that the charging behavior is almost the same in both cases. Note that for $\alpha \neq 0$, the usual Friedel sum rule [62] $G = \frac{2e^2}{h} \sin^2(\frac{\pi}{2} \langle n_d \rangle)$ is not satisfied and this can be an indication of a non-Fermi-liquid behavior. Figure 5.3c shows the conductance in the absence of Coulomb repulsion ($U = 0$): the same effect is obtained for $\alpha \neq 0$. The conductance cancellation does not depend on the electron-electron interaction, which agrees with the qualitative explanation presented below.

Figure 5.4 shows the conductance as a function of V_g for different values of λ . The dip

Figure 5.4: G as a function of V_g in the Kondo regime for $\alpha = 0.4$ and $\lambda = 0.15, 0.20$, and 0.25 (dashed, dotted and solid lines respectively). The dip becomes more pronounced as λ increases. The inset shows the convergence of the results with the maximum number of phonons (N_{ph}). Note that the qualitative effect of conductance cancellation is preserved all the way down to $N_{\text{ph}} = 1$.

becomes more pronounced as λ increases i.e. as the average number of phonons in the ground state increases. The inset shows the results obtained by truncating the phonon Hilbert space at different maximum number of phonons (N_{ph}). In all the calculations, $N_{\text{ph}} = 7$ was used unless stated otherwise. The qualitative effect of conductance cancellation is obtained all the way down to $N_{\text{ph}} = 1$, allowing us to study larger clusters and reduce size effects to intuitively understand the origin of the dip.

In Fig. 5.5, an explanation of the conductance dip is presented. The reasoning starts by noting that $\hat{H}_{\text{M-leads}}$ in Eq.5.2 can be rewritten as a sum of two channels contributing to the overall molecule-leads connection. The first term, $-t' \sum_{\sigma} (d_{\sigma}^{\dagger} c_{l0\sigma} + d_{\sigma}^{\dagger} c_{r0\sigma} + h.c.)$, represents the purely electronic tunneling between the molecule and the two electrodes. The second term, $-t' \alpha (a + a^{\dagger}) \sum_{\sigma} (d_{\sigma}^{\dagger} c_{r0\sigma} - d_{\sigma}^{\dagger} c_{l0\sigma} + h.c.)$, represents a phonon assisted tunneling channel, i.e. the electron absorbs (emits) a phonon upon entering the molecule and, then, emits (absorbs) a phonon upon leaving. Note that both channels are coherent

Figure 5.5: (a) A schematic representation of the two conductance channels, the purely electronic tunneling represented by the two upper arrows and the “phonon-assisted tunneling” channel represented by the two lower arrows. (b) Partial conductance when only one of the channels is active. The dotted line shows the conductance of the purely electronic tunneling channel G_E , while the dashed line shows the conductance of the “phonon-assisted tunneling” channel G_I . (c) Conductance when both channels are active. (d) Phase difference $\Delta\Phi$ of the two channels. Note that $\Delta\Phi$ for all values of V_g is close to π thus leading to destructive interference. In particular, for $V_g = -U/2$, $\Delta\Phi = \pi$ and $G_E = G_I = 2e^2/h$, thus leading to a perfect cancellation in the overall conductance. ($\lambda = 0.2$, $\alpha = 0.4$).

and elastic. The number of phonons in the system does not change. Figure 5.5a shows a schematic of the two channels. These channels were studied separately by keeping only the relevant term in $\hat{H}_{M-\text{leads}}$. The conductance and the phase carried by each channel were calculated. Figure 5.5b shows the conductance of the separate channels. Figure 5.5c contains the conductance when both channels are active i.e. when both terms are included in $\hat{H}_{M-\text{leads}}$. Figure 5.5d shows the phase difference between the two channels. Note that for $V_g = -U/2$, the conductance of each of the channels is $2e^2/h$ and the phase difference is π , leading to a perfect cancellation in the overall conductance [87]. This interference effect is independent of the electron-electron interaction and, thus, the cancellation should still

Figure 5.6: The solid line shows the conductance of the molecule when a breathing vibrational mode is active (no CM motion). The particle-hole symmetry is broken as expected and no conductance dip is obtained. The dashed line shows the conductance when both breathing and CM vibrational modes are active. The combined effects of the two modes lead to a Fano-like interference. The breathing mode parameters used are $\lambda' = 0.2$, $\omega'_0 = 0.3$ and $\alpha' = 0.3$.

be present for $U = 0$ as already shown. The dip becomes more pronounced as λ increases, increasing the average number of phonons in the ground state.

The stability of the dip is tested by adding an internal vibrational mode which leads to the symmetric modulation of the tunnel barriers to the leads (breathing mode). To account for this mode, the following terms were added to the Hamiltonian:

$$\hat{H}' = \lambda'(1 - \hat{n}_d)(b + b^\dagger) + \omega'_0 b^\dagger b - t'\alpha'(b + b^\dagger) \sum_{\sigma} (d_{\sigma}^\dagger c_{l0\sigma} + d_{\sigma}^\dagger c_{r0\sigma} + h.c.), \quad (5.4)$$

where the first term represents the electron-phonon coupling, the second term represents the breathing vibrational energy, and the third term represents the subsequent modulation of the tunnel barriers. The results are shown in Fig. 5.6. When only the internal mode is active (solid line), the electron-hole symmetry is broken but no dip is observed. This agrees with previous results [29] obtained using NRG calculations. In the case where both vibrational modes are active (dashed line), the dip appears. The combined effect

Figure 5.7: (a) Convergence of the conductance with the size of the exactly-solved cluster L . The solid line, dashed line, and the circles show the results obtained using $L = 3, 7$, and 11 respectively. In the three cases, a maximum number of three phonons was used. (b) The real and (c) imaginary parts of the isolated cluster Green function g_{lr} that propagates an electron from the left to the right ends of the cluster ($L = 3$ and $V_g = -U/2$). Note that both parts are equal to zero at the Fermi level (located at $\omega = 0.0$). Thus, the origin of the conductance dip can be traced back to the exactly-solved cluster. $\lambda = 0.2$ and $\alpha = 0.4$ in the three figures.

of conductance cancellation and electron-hole asymmetry leads to a Fano-like interference.

The finite-size effects on the results are shown in Fig. 5.7a where the convergence of the conductance with the size of the exactly-solved cluster is presented. Note that increasing the cluster does *not* change the qualitative effect of the conductance dip. Moreover, the origin of the conductance dip can be traced back to the exactly-solved cluster by studying Green functions *before* the embedding process. Figures 5.7b and c show the real and imaginary parts of the Green function g_{lr} that propagates an electron from the left to the right end of the cluster for $\alpha \neq 0$ and $V_g = -U/2$. Both parts are zero at the Fermi level ($\omega = 0$). For $\alpha = 0$ (not shown here), g_{lr} has a pole at the Fermi level and the system is perfectly conducting. When α is turned on, the pole splits into two, one below and one above the Fermi level thus causing the zero conductance.

5.4 Conclusion

In conclusion, the zero temperature electron transport through a molecular conductor with center-of-mass motion was studied numerically for interacting and noninteracting electrons. The results present an interesting conductance dip when an odd number of electrons occupy

the molecule. It is argued that this dip is caused by the destructive interference between the purely electronic and phonon-assisted tunneling channels, which are found to carry opposite phases. When an internal vibrational mode is also active, the particle-hole symmetry is broken but a Fano-like interference is still obtained. The conductance cancellation would best be observed on a broad conductance peak such as the Kondo peak which is broader than the resonant tunneling peak.

CHAPTER 6

ADAPTIVE TIME-DEPENDENT DENSITY-MATRIX RENORMALIZATION GROUP TECHNIQUE FOR CALCULATING THE CONDUCTANCE OF STRONGLY CORRELATED NANOSTRUCTURES

6.1 Introduction

The rapidly developing investigations in the area of nanometer-scale systems and its concomitant potential technological applications in real devices have induced considerable interest in the study of electrical transport through small molecules and quantum dots. In fact, the construction of molecular electronic devices [93, 94, 95, 96, 23] is among the most exciting areas of investigations in physics, and theoretical guidance is needed for the success of this vast effort. Molecules can change their shape and position relative to the leads as electrons enter or leave the molecule, making the study of these systems very challenging. Moreover, Coulomb correlations cannot be neglected in small devices. For a conceptual understanding of these complex systems, it is imperative to develop models and unbiased many-body methods that rely on a minimal number of assumptions, in order to accurately handle both strong Coulombic and electron-phonon couplings. Quantum dots constructed using conventional semiconductor technology also provide an important playground for the analysis of transport properties in nanoscopic systems, and the theoretical challenges in this context are equally important [97].

The conductance of small nanoscopic systems can be theoretically estimated using a variety of techniques. Among the most popular approaches are the ab-initio calculations using density functional theory (DFT). These one-electron self-consistent methods have been

successful in describing various I-V characteristics [98, 99, 100]. However, the applicability of these ideas must be carefully scrutinized, as recently remarked by Muralidharan *et al.* [101]. For example, it is clear that in small molecules charging effects are important, and they effectively act as quantum dots in the Coulomb Blockade regime. Moreover, techniques that do not take into account the strong correlation between electrons cannot capture important effects such as the Kondo resonance (arising from the coupling between localized spins and conduction electrons) which induces a new channel for transport in a variety of small systems [1, 50, 51, 21]. In addition, it is well known that several bulk materials, such as transition metal oxides, cannot be described with ab-initio methods that neglect correlations. The complexity of their behavior, including potentially useful effects such as large magnetoresistances in Mn oxides [63], may manifest in nanoscopic systems as well, and the use of strongly correlated materials in nanodevices may lead to interesting applications. To study all these systems (small molecules, quantum dots, and in general nanodevices that include strongly correlated materials), techniques beyond DFT must be developed.

A similar challenge occurred before in the study of bulk materials, and several years of research have shown that the following two-steps process leads to profound insights. The first step consists of a simple modeling of the material, typically either deducing the relevant degrees of freedom from atomistic considerations when the states are very localized, or borrowing from band structure calculations to isolate the minimal ingredients needed to capture the essence of the problem. The second step, the hardest, is solving the resulting model, which is typically of a tight-binding nature with the addition of Coulombic and phononic couplings. In the strong coupling regime, the use of numerical techniques provides the most reliable unbiased approach for the approximate investigation of tight-binding-like models with Coulombic interactions [102]. As a consequence, a natural path toward the study of transport in strongly correlated nanoscopic systems can also start with simple models and use computational techniques for their analysis. It is the main purpose of this chapter to propose a technique that can be used to study transport in systems described by strongly correlated electronic models.

The method to calculate conductances proposed here relies on the successful Density Matrix Renormalization Group (DMRG) technique [103, 104]. While in principle this method is applicable only to quasi-one-dimensional models, such a geometry is quite acceptable in several nanoscopic important problems including transport in arrays of quantum dots or

using small molecules as bridges between leads. However, a straightforward application of the original DMRG methodology is not immediately useful to study transport, particularly when arbitrarily large external electric and magnetic fields, with potentially complicated time dependences, are switched on and off at particular times. Nevertheless, progress toward a computational tool for this type of problems has been steady in recent years. For instance, important numerical methods for dynamical DMRG studies were presented to handle frequency dependent spectral functions [105, 106, 107]. More directly focusing on real-time investigations, interesting techniques were proposed [108, 109]. While useful for many qualitative applications, these methods are in general not as accurate and stable as needed for the detailed study of finite bias transport in complicated nanosystems. The reason is that the method of Ref. [108] is “static” in the sense that the truncated Hilbert space found to be optimal at time $t=0$, namely before switching on the external fields, is kept at all times. This approach breaks down after relatively short times, since extra states are needed for a proper description of transport at finite times. A further approximation to improve on this first proposal is to enlarge substantially the initial Hilbert space so that it remains suitable for properties calculated at finite times [110]. This technique has the problem that the number of states grows rapidly with the simulation time and eventually it becomes impractical. Nevertheless, the method has been successfully used to study the propagation of a density excitation in an interacting clean system [111].

Recently, important developments have led to the “adaptive” time-dependent version of the DMRG method, that is efficient over long times and, thus, it is suitable to handle the problems we are focusing on. The method to be used here was developed independently by White and Feiguin [39] and Daley *et al.* [41], after the idea of how to do time-evolution to a matrix product was introduced by Vidal [112], and relies on an adaptive optimal Hilbert space that follows the state as time grows. The method is based on a Suzuki-Trotter break-up of the evolution operator, and as a consequence a Trotter truncation error is introduced. Fortunately, this systematic error can be easily estimated and controlled. The adaptive DMRG numerical method will only be briefly reviewed below since our proposal uses the technique to calculate conductances, but does not modify the method itself. The reader should consult the original literature [39, 41] for more details. It is important to remark that the technique is easy to implement once a ground-state DMRG code is prepared and, moreover, the time-evolution is stable, as shown explicitly in our results and in some previous

investigations (further improvements can be added with the time-step targeting method recently proposed by Feiguin and White [40]). The conclusion of our effort documented below is that the adaptive method provides accurate results for the calculation of conductances. The technique has passed the test of noninteracting electrons, as well as the cases of one and two interacting quantum dots, where a subtle Kondo effect occurs. Moreover, the method is not restricted to small biases but it produces reasonable answers at finite bias as well. As a consequence, it has the potential of being the method of choice to study transport under both weak and strong external fields, in small nanostructures of substantial complexity. Multilevel model Hamiltonians, possibly inspired by ab-initio calculations, can be used to describe the “bridge” between leads. In addition, the method is particularly transparent since it relies on the straightforward calculation of a current in the presence of a voltage, rather than relying on other indirect linear-response formulas.

Of course, the reader must be aware that the method is not of unlimited applicability. If the molecules or Kondo clouds are too long in size, eventually not even DMRG can handle the very long chains needed for a proper description. For completeness, and to assure a balanced description of the technique, one of these difficult cases is also presented. But often the qualitative physics can be understood by relaxing parameters, thus we expect that even in very complicated cases the proposed technique will be helpful, at least at the conceptual level. Other limitations of the present technique is that energy dissipation is not incorporated, and the temperature is restricted to be zero. Improving on these issues is a task left for the near future.

It is important to remark that there are other numerical techniques that can also be used to study transport in strongly correlated nano-systems. One of them is the Numerical Renormalization Group, that evolved from Wilson’s original RG ideas. This technique is quite accurate, as exemplified by some recent calculations [28, 113], but it cannot be used for arbitrary problems. Since our goal is to try to develop a method that can handle the fairly complex models that will be used in the near future to represent, e.g., small molecular conductors, this method does not have sufficient flexibility for our purposes. In cases where NRG works, it should be the method of choice, but this occurs in a small subset of problems in the area of transport across correlated systems. A second approach relies on the static DMRG method, using a ring geometry and with a current induced by a flux threading the ring [114, 115, 116].

A recently proposed third method combines linear response Kubo theory with static DMRG and the conductance is calculated based on correlation functions in the ground state [117]. A fourth method relies on the Quantum Monte Carlo technique to calculate Green functions and the conductance of impurity models [118]. A fifth method is the Exact Diagonalization technique followed by a Dyson equation embedding procedure (ED+DE) [87, 119] where the interacting region is solved exactly (including some sites of the leads), and then the rest of the leads are taken into account via a Dyson self-consistent approach. The method directly treats bulk systems, contrary to the DMRG technique that is necessarily limited to a large but finite chains, it is flexible and has led to interesting results for difficult cases, such as center-of-mass phonons in molecular conductors, and multilevel systems [87, 120, 121, 122, 27]. However, the Dyson embedding is somewhat arbitrary and it is difficult to control its accuracy.

The organization of this chapter is as follows. After the present introduction, in Sec. 6.2, the models are defined and the technique is very briefly described. Section 6.3 contains the important test of noninteracting electrons (note that although the Coulombic coupling is zero, there are different hopping amplitudes at different links). Here, the systematic behavior of the method is discussed in detail. Section 6.4 deals with the case of a quantum dot, with a nonzero Hubbard coupling. The value of U is chosen to prevent the Kondo cloud from reaching huge sizes that would render the DMRG method useless (nevertheless, one “large” U case is studied for completeness, to illustrate the limitations of the method). Section 6.18 contains the case of two dots, which themselves can be coupled in a “singlet” preventing conduction or loosely coupled having individually a Kondo effect. The conclusions in Sec. 6.6 briefly summarize our findings.

6.2 Model and Conductance Calculation

In general, the systems studied here consist of a relatively small region where Coulomb interactions are present, weakly coupled to two non-interacting leads (see Fig. 6.1). The interacting region can represent one or several quantum dots (QD’s), a single-molecule conductor, or other nanoscopic regions. In fact, the generality of the method presented in this paper allows for a wide variety of interacting systems.

The leads are modeled as ideal tight-binding chains. As examples, the focus will be on one QD and two QD’s connected in series. The total Hamiltonian of these systems can be

Figure 6.1: Schematic representation of the geometry used in our study. The leads are modeled by tight-binding Hamiltonians. The ground state at time zero is calculated at zero bias. Then, a finite bias ΔV is applied between the two leads (without ramping time for simplicity, but this can be changed in future studies) and the resulting current is measured.

written in general as

$$\hat{H} = \hat{H}_{\text{leads}} + \hat{H}_{\text{cluster}} + \hat{H}_{\text{cluster-leads}}, \quad (6.1)$$

where $\hat{H}_{\text{leads}}$ is the Hamiltonian of the leads, which is

$$\hat{H}_{\text{leads}} = -t_{\text{leads}} \sum_{i\sigma} [c_{li\sigma}^\dagger c_{li+1\sigma} + c_{ri\sigma}^\dagger c_{ri+1\sigma} + h.c.]. \quad (6.2)$$

t_{leads} is the hopping amplitude in the leads, which in the following is taken as the energy scale (i.e. $t_{\text{leads}} = 1$). The operator $c_{li\sigma}^\dagger$ ($c_{ri\sigma}^\dagger$) creates an electron with spin σ at site i in the left (right) lead. $\hat{H}_{\text{cluster}}$ is the Hamiltonian of the cluster where the interactions are present. Finally, $\hat{H}_{\text{cluster-leads}}$ is the Hamiltonian that connects the interacting region to the leads, typically a hopping term. In all the results presented below, the half-filling case $n = 1$ was considered in the entire system.

6.2.1 One Quantum Dot

For the case of one QD, represented simply by one active level, $\hat{H}_{\text{cluster}}$ can be written as

$$\hat{H}_{\text{cluster}} = V_g n_d + U n_{d\uparrow} n_{d\downarrow}, \quad (6.3)$$

where the first term represents the location of the energy level of the QD controlled by the gate voltage V_g . The second term represents the Hubbard repulsion between electrons of opposite spins occupying the QD. $n_d = n_{d\uparrow} + n_{d\downarrow}$ is the number of electrons at the dot. $\hat{H}_{\text{cluster-leads}}$ can be written as

$$\hat{H}_{\text{cluster-leads}} = -t' \sum_{\sigma} [c_{l\sigma}^\dagger c_{d\sigma} + c_{r\sigma}^\dagger c_{d\sigma} + h.c.], \quad (6.4)$$

where t' is the amplitude for the electronic hopping between the QD and the leads. $c_{d\sigma}^\dagger$ creates an electron with spin σ at the dot, while $c_{l\sigma}^\dagger$ creates an electron at the last site of the left lead and $c_{r\sigma}^\dagger$ creates an electron at the first site of the right lead, if sites are numbered from left to right.

6.2.2 Two Coupled Quantum Dots in Series

In the case of two QD's, $\hat{H}_{\text{cluster}}$ can be written as

$$\hat{H}_{\text{cluster}} = \sum_{\alpha=1,2} [V_g n_{d\alpha} + U n_{d\alpha\uparrow} n_{d\alpha\downarrow}] - t'' \sum_{\sigma} [c_{d1\sigma}^\dagger c_{d2\sigma} + h.c.], \quad (6.5)$$

where $n_{d\alpha} = n_{d\alpha\uparrow} + n_{d\alpha\downarrow}$ is the number of electrons at the quantum dot α , and t'' is the hopping between the two dots. $\hat{H}_{\text{cluster-leads}}$ is written as

$$\hat{H}_{\text{cluster-leads}} = -t' \sum_{\sigma} [c_{l\sigma}^\dagger c_{d1\sigma} + c_{r\sigma}^\dagger c_{d2\sigma} + h.c.]. \quad (6.6)$$

6.2.3 Conductance Calculation

The current at any time t between nearest-neighbors sites i and j is calculated as

$$J_{ij}(t) = i \frac{2\pi e}{h} t_{ij} \sum_{\sigma} \langle \Psi(t) | (c_{i\sigma}^\dagger c_{j\sigma} - c_{j\sigma}^\dagger c_{i\sigma}) | \Psi(t) \rangle, \quad (6.7)$$

where $|\Psi(t)\rangle$ is the wave function of the system at time t , which will be calculated with the DMRG method, using a number M of states in the process. $c_{i\sigma}^\dagger$ creates an electron with spin σ at site i , which can be part of the interacting region or the leads. In the results presented below, the current shown without any link or site index corresponds to

$$J(t) = (J_L(t) + J_R(t))/2, \quad (6.8)$$

where $J_L(t)$ is the current between the last site of the left lead and the first dot, and $J_R(t)$ is the current between the last dot and the first site of the right lead, moving from left to right. The conductance G can be obtained by simply dividing the steady-state current by the total bias ΔV . The individual voltages $\pm \Delta V/2$ in the leads are applied uniformly in each one, as indicated in Fig. 6.1. Note that the use of a symmetrized current $J(t)$ is convenient to obtain smoother results particularly when the left and right leads have a different number of sites.

6.2.4 Technique

Closely following Ref. [39], a brief description of the numerical technique is provided here. The basic idea is to incorporate the Suzuki-Trotter (ST) decomposition of the time-evolution operator [112] into the DMRG finite-system algorithm [39, 41]. The second order ST decomposition of the 1D Hamiltonian as employed in Ref. [39] can be written as

$$e^{-i\tau H} \approx e^{-i\tau H_1/2} e^{-i\tau H_2/2} \dots e^{-i\tau H_2/2} e^{-i\tau H_1/2}, \quad (6.9)$$

where H_j is the Hamiltonian of the link j . The DMRG representation of the wavefunction at a particular step j during the finite-system sweep is

$$|\psi\rangle = \sum_{l\alpha_j\alpha_{j+1}r} \psi_{l\alpha_j\alpha_{j+1}r} |l\rangle |\alpha_j\rangle |\alpha_{j+1}\rangle |r\rangle, \quad (6.10)$$

where l and r represent the states of the left and right blocks (in a truncated basis, optimally selected as eigenvectors of a density matrix), while α_j and α_{j+1} represent the states of the two central sites. An operator A acting on sites j and $j+1$ (namely, only involving nearest-neighbors) can be applied to $|\psi\rangle$ exactly, and re-expressed in the same optimal basis as

$$[A\psi]_{l\alpha_j\alpha_{j+1}r} = \sum_{\alpha'_j\alpha'_{j+1}} A_{\alpha_j\alpha_{j+1};\alpha'_j\alpha'_{j+1}} \psi_{l\alpha'_j\alpha'_{j+1}r}. \quad (6.11)$$

Thus, the time evolution operator of the link j can be applied exactly at the DMRG step j . As a consequence, the time evolution is done by applying $e^{-i\tau H_1/2}$ at DMRG step 1, $e^{-i\tau H_2/2}$ at DMRG step 2, and so on, thus forming the usual left-to-right sweep. Then, applying all the reverse terms in the right-to-left sweep. A full sweep evolves the system one time step τ . The error introduced by the second order decomposition is order τ^3 in each time step [39]. Thus, upon evolving the system one time unit ($1/\tau$ steps), an order τ^2 error is introduced. Numerically, the influence of this small systematic error is easy to control. In all the presented results, a time step $\tau = 0.05$ was used. Note that the Trotter decomposition error is dominant at short times, while at longer times, the DMRG truncation error dominates. Decreasing τ would decrease the Trotter error. However; in the case of a very small τ , more DMRG sweeps are required to evolve the system one time unit thus accumulating the DMRG truncation errors. Therefore, for a given system, an optimal τ should be found. (See Ref. [123] for an analysis of this error). This brief summary gives the reader an idea of the technique used here. Details regarding lattice sizes, number of states kept in the DMRG procedure, and influence of other parameters are discussed below.

6.3 Non-Interacting Case

Properties of the method discussed in this chapter are exemplified in Fig. 6.2(a), where the current at the center of the chain is shown (divided by the voltage difference) as a function of time. The current in this figure is *exactly* calculated, not using DMRG, since for non-interacting particles the problem reduces to a single electron problem. A small bias $\Delta V=0.001$ is used, unless otherwise stated. Thus, in this first study the focus will be on trying to reproduce results expected from linear response, but a few results with a finite bias are also included as discussed below. Returning to Fig. 6.2(a), for a bulk system the current would be expected to raise for a small fraction of time, and then reach a steady state. This indeed occurs even in our finite-size systems. In fact, the transition from zero current at $t=0$ to an approximately time-independent current regime is very fast, and can be barely observed in the scale of Fig. 6.2(a). But the existence of a very flat plateau in the current is clear, and its value will be used to extract the conductance below. Note that due to the finite size of the system, the current cannot continue in the same steady state at all times. The Hamiltonian is particle number conserving and, as a consequence, the presence of a current implies a population/depopulation of the leads, which cannot continue forever. In fact, once the front of the charge pulse reaches the end of the chain, it bounces back and eventually generates a current of the opposite sign. This effect will be discussed in more detail later. Here, it is important to remark that in spite of this periodicity present in finite open-end systems, the flat plateaus are clearly defined over an extended period of time for the lattice of 402 sites used, and the value of the conductance can be easily deduced from those individual plateaus, as discussed below. Note that the setup of Fig. 6.1 and the existence of plateaus in the current Fig. 6.2 are natural in the DMRG/transport context and was observed before [111]. Our main contribution will be the use of the adaptive DMRG method for the calculations, as shown below.

To further illustrate the propagation of charge in the cluster after the finite bias is switched on at time $t=0$, in Fig.6.3 the exact current at different positions x is shown, parametric with time. At small times, $t=5$ (in units of $\hbar/t_{\text{leads}}$) only the central portion is affected as expected. At time $t=55$, the affected region is much larger, while at $t=105$, the front has reached the ends of the chain and soon after it starts bouncing back. At times $t=200$ and 205 , the initial condition is approximately recovered, and almost everywhere the

Figure 6.2: Exact results for $J(t)/\Delta V$ (in units of e^2/h) vs. time (in units of $\hbar/t_{\text{leads}}$), for the non-interacting 1QD case obtained with clusters of different lengths (L) and $\Delta V=0.001$. (a) $J(t)/\Delta V$ obtained with a large cluster ($L=402$). $J(t)/\Delta V$ shows clear steady-state plateaus at $\pm 2e^2/h$. The periodic changes in the current direction are caused by its reflection at the open boundaries of the cluster. (b) $J(t)/\Delta V$ obtained with decreasing L . The steady-state plateau is obtained even with $L = 32$. The current is quasi-periodic with a period proportional to L . The parameters used are $V_g=U=0$ and $t'=0.4$.

current to the left and right cancel out nearly exactly. For larger times, a reverse sign current is created. Note that in our studies there are *no* sources of dissipation, and the current will keep on oscillating forever. Adding inelastic processes is a next major challenge in this context, left for the future.

It is also important to show that the existence of the plateaus is not restricted to very long chains of hundreds of sites, but they are visible on much smaller systems, increasing the chances that the numerical DMRG method can be used even for complicated nanosystems. Figure 6.2(b) contains the current vs. time for a variety of lattice sizes, ranging from 402 to systems as small as 16 sites. The time width of the plateaus depends on L , as expected, but the value of the current at the plateaus is approximately L independent even up to systems as small as containing $L=32$ sites. Even the $L=16$ chain has a periodicity with a first plateau in the current which is also in good agreement with the expected value from larger sizes. Thus, this behavior appears to be robust and the plateaus are also expected to be present for the chains that the DMRG method can handle. That this is the case can be shown in

Figure 6.3: This figure shows the propagation of the current in the cluster after the bias $\Delta V=0.001$ is applied at $t=0$. The different panels show the current as a function of position x along the chain, at different times t . The size is $L = 402$ and the dot is at the site 201. The results were obtained exactly, since the Hubbard couplings are zero.

Fig. 6.4, where DMRG results for the current vs. time are shown, compared with exact data. Consider first a sufficiently long chain, as shown in (a), such that a sharp plateau is observed in the exact result. Figure 6.4(a) shows that increasing the number of states M used in the DMRG approximation, a convergence to the exact solution is observed. In fact, for $M=300$ or higher, the DMRG results cannot be distinguished from the exact ones. A similar behavior is found using shorter chains as for the case with $L=64$ sites in Fig. 6.4(b), but in this example the plateau can be observed accurately even with a smaller number of states such as $M=200$. The trend continues for smaller systems (c,d): For $L=32$ and 16, the DMRG method reproduces the exact results with high accuracy using ~ 100 states.

Figure 6.5(a) shows the conductance, deduced from the behavior of the current obtained with the DMRG method, versus the gate voltage V_g for the case of a single “noninteracting” quantum dot, namely one having $U=0$. The hopping amplitude between the dot and the leads is $t'=0.4$. It is expected that the maximum value of the conductance be obtained when the level in the dot is aligned with the Fermi level of the leads, and this occurs in our case at $V_g=0$. The DMRG results beautifully confirm this expectation. As the gate voltage changes

Figure 6.4: DMRG results compared to the exact results for $J(t)/\Delta V$ obtained using different clusters L and number of states M , with $\Delta V=0.001$. (a) $L = 96$, (b) $L = 64$, (c) $L = 32$, and (d) $L = 16$. Note that for $L = 96$ and 64 , $M = 200$ shows good qualitative agreement and $M \geq 300$ even shows good quantitative agreement with the exact results. For $L = 32$ and 16 , $M = 200$ and 100 already show excellent quantitative agreement with the exact results.

away from 0, the conductance is expected to decrease symmetrically and this is indeed shown in Fig. 6.5(a). In fact, the results at nonzero gate voltage are also in excellent quantitative agreement with the exact results.

All the previous results were obtained for a sufficiently small value of the bias voltage $\Delta V=0.001$, as already explained. It is interesting to observe how the results change when larger values of ΔV are employed. Figure 6.5(b) shows the current for a couple of biases. The existence of the plateau is clear in both cases, but for $\Delta V=0.01$, asymmetries between positive and negative gate voltages can be observed, which are not expected in the limit $\Delta V \rightarrow 0$. As a consequence, the rest of the results discussed below were obtained with $\Delta V=0.001-0.005$ unless otherwise noted. An easy criterion to realize if a sufficiently small bias is used to obtain the linear response limit is to repeat the calculations for the same amplitude of gate voltage, but opposite signs, and see if a noticeable difference is obtained.

The method proposed here also works in the case of a finite bias voltage, namely it is not restricted to the linear response regime. To show that the technique can handle even a large

Figure 6.5: (a) DMRG and exact results for G vs. V_g in the case of one non-interacting QD, and $t' = 0.4$ (with $\Delta V = 0.001$). The DMRG results are obtained with $L = 64$ and $M = 300$. In this case, G is obtained from the value of the steady-state current plateau. The exact results are for infinite leads. The results show a resonant tunneling peak of FWHM $= 4t'^2$ at $V_g = 0$. (b) $J(t)/\Delta V$ for $V_g = \pm 0.3$ and $\Delta V = 0.01, 0.001$. Note that decreasing the bias voltage reduces the asymmetry between positive and negative V_g and gives a better steady-state plateau.

bias, in Fig. 6.6 results for the current vs. time are shown at the indicated voltages. It is only at ΔV as large as 1.0 that small differences are visually observed in the figure, between the DMRG and exact results. This can be fixed increasing the number of states M . Thus, overall the method appears to be sufficiently robust to handle arbitrary voltages, showing the generality of the technique here proposed. Nevertheless, further work in the finite bias context will be important to fully test this case, calculating differential conductances and analyzing the regime of very strong bias.

In our investigations, the numerical study was also carried out using a “static” procedure, where the $t=0$ DMRG basis is not adapted with growing time. In this case, the results are obtained by integrating the time-dependent Schrödinger equation using the fourth order Runge-Kutta method, and also using the DMRG ground state as the initial state [108]. This is to be contrasted with the procedure of Refs. [39, 41] where the basis is modified with time. Figure 6.7 shows the results of both procedures: clearly using an adaptive basis

Figure 6.6: (a) DMRG results compared to the exact results in the case of one non-interacting QD for several intermediate and large values of ΔV , namely exploring the influence of a finite bias in the calculations. The parameters used are $V_g = U = 0$, and $t' = 0.4$. Both DMRG and exact results are obtained with a cluster $L = 32$, using $M = 200$ states, for the DMRG results.

provides superior data, reproducing accurately the exact results. In the static procedure a similar accuracy is reached only by increasing substantially the number of states, thus missing its economical CPU-time advantages.

The method discussed here also works nicely for the case of two non-interacting QD's, as shown in Fig. 6.8 for two different values of the hopping amplitude t'' between the dots. The slight difference between the DMRG and the bulk exact results can be improved increasing the number of sites.

Although not directly related with the method to obtain the conductance of an interacting nano-system discussed here, for completeness we have also studied the local density-of-states which are important to guide the intuition and contrast with other methods and scanning tunneling microscopy experiments as well. The local density-of-states at the dot is given by $\rho_d(\omega) = \sum_{\sigma} \rho_{d\sigma}(\omega) = -\frac{1}{\pi} \text{Im}[\sum_{\sigma} G_{d\sigma}^R(\omega)]$, where $G_{d\sigma}^R(\omega)$ (the retarded Green function) can be written as $G_{d\sigma}^R(\omega) = \langle \Psi_0 | c_{d\sigma} \frac{1}{\omega + E_0 - \hat{H} + i\epsilon} c_{d\sigma}^{\dagger} | \Psi_0 \rangle + \langle \Psi_0 | c_{d\sigma}^{\dagger} \frac{1}{\omega - E_0 + \hat{H} + i\epsilon} c_{d\sigma} | \Psi_0 \rangle$. In the results shown, $G_{d\sigma}^R(\omega)$ is evaluated using the continuous fraction expansion technique with

Figure 6.7: The results obtained with the Suzuki-Trotter approach and the static Runge-Kutta method compared to the exact results for (a) $\Delta V = 0.001$ and (b) $\Delta V = 0.5$. The parameters used are $V_g = U = 0.0$, $t' = 0.4$, and $L = 64$.

Figure 6.8: DMRG and exact results for G vs. V_g in the case of 2 coupled non-interacting QD's. The DMRG results are obtained with $L = 64$ and $M = 300$ ($\Delta V = 0.001$). The exact results are for infinite leads. The cases (a) $t'' = 0.5$ and (b) $t'' = 0.2$ are investigated, with $t' = 0.4$ in both cases. The results present the bonding and anti-bonding resonant tunneling peaks at $\pm t''$.

Figure 6.9: DMRG local density-of-states results for (a) 1QD and (b) 2QD's compared to the exact (bulk) results. The parameters used are $V_g = U = 0$, $t' = 0.4$ in both cases and $t'' = 0.5$ in (b). In (a), a broadening imaginary component $\eta = 0.1$ was introduced. In (b), $\eta = 0.15$. Smaller values of η would reveal the discrete nature of the LDOS obtained with DMRG on a finite $L=64$ system.

coefficients calculated with the DMRG method [105]. In Fig. 6.9, results for non-interacting quantum dots are shown (namely, dots where the Hubbard repulsion is 0). Clearly, both the exact results (which are shown already in the bulk limit) and the DMRG results, slightly smeared by shifting, using a small imaginary component η , the pole locations in the continued fraction expansion, are in excellent agreement in both cases. A smaller η would have revealed the many δ -functions in the DMRG case obtained using a finite chain with 64 sites.

6.4 One Quantum Dot

In the previous sections, the method was introduced and tested for the case of noninteracting $U=0$ electrons. But the main application of the technique is envisioned to occur in the presence of nontrivial Coulombic interactions (and eventually also adding phononic degrees of freedom). In this section, the case of a nonzero Hubbard coupling will be considered,

focusing on the special case of one quantum dot. The Hamiltonian used was already discussed in previous sections.

6.4.1 Results at intermediate values of U

Figure 6.10 contains our DMRG results for the current vs. time, for the case of $U=1.0$. Similar values of this coupling were extensively used in previous investigations [124, 53, 87, 120, 121, 122, 27], and it is believed to lead to a Kondo cloud of a size amenable to numerical investigations (note that if U is very large, the effective J between localized and mobile spins is reduced, and it is known that the cloud’s size rapidly grows with decreasing J). The figure shows that the systematic behavior found in the noninteracting case survives the presence of a Coulomb interaction, namely the current develops plateaus that can be used to determine the conductance. For instance, this effect is clearly present for $L=96$ and 128, although for smaller sizes (shown for completeness) the maximum current is 10% to 20% less than expected and one must be cautious with size effects. The value of the gate voltage is $-U/2$, which in the absence of the Kondo effect would locate the system in the conductance “valley” (implying a near zero conductance) between the Coulomb blockade peaks at $-U$ and 0. The figure shows that the method introduced in this paper is able to reproduce the existence of a Kondo effect, since the conductance is actually very close to the ideal limit $2e^2/h$ [50, 51, 21, 87], rather than being negligible. This is a highly nontrivial test that the proposed technique has passed.

Results for other values of the gate voltage are shown in Fig. 6.11. Moving away from $V_g=-U/2$, the current is reduced, as discussed below in more detail. Note that the plateaus contain small oscillations as a function of time [125]. The size of the oscillations gives an indication of the errors in the numerical determination of the conductance, for a given lattice size. The procedure followed here to extract the current needed for the calculation of the conductance is to carry out averages over time, as shown in the figure, in the plateau region between the short-time intrinsic oscillations and the long-time oscillations caused by numerical errors. For a given L , as M increases, the oscillations tend to disappear. This is shown in Fig. 6.12.

Following the procedure sketched in Fig. 6.11, the full conductance vs. V_g was prepared for U equal to 1 and 2. The results are shown in Fig. 6.13. The results are compared to the conductance obtained using the Friedel sum rule (FSR), $G = \frac{2e^2}{h} \sin^2(\frac{\pi}{2}\langle n_d \rangle)$. The shape

Figure 6.10: $J(t)/\Delta V$ in the case of one quantum dot for different cluster lengths L . The parameters used are $U = 1.0$, $t' = 0.4$, $\Delta V = 0.005$, and $V_g = -0.5$. As L increases, the conductance approaches the unitarity limit ($2e^2/h$) due to the Kondo screening effect.

Figure 6.11: $J(t)/\Delta V$ in the case of one interacting QD for different values of V_g , and with $\Delta V = 0.005$. The value of the conductance is obtained by averaging the current over an interval of time, corresponding to the steady state. The solid horizontal lines represent this time interval over which the average of the current is taken, and the value of the average. The parameters used are $U = 1.0$, $t' = 0.4$, $L = 128$, and $M = 300$.

Figure 6.12: $J(t)/\Delta V$ in the case of one interacting QD for $V_g/U = -1.5$, $U = 1.0$, $t' = 0.4$, $L = 128$, and different values of M . Note that as M increases, the oscillations at long times tend to decrease.

of the curve is the expected one for the regime considered here: the intermediate values of U do not locate our investigation deep in the Kondo regime, with sharply defined integer charge at the dot, but more into the mixed-valence region. This can be deduced from the value of the dot charge vs. V_g , also shown in Fig. 6.13. With increasing U and/or decreasing t' , sharper charge steps are formed, but the Kondo cloud size increases, as discussed later.

6.4.2 Results at large values of U

There are cases where the technique gives results that are only qualitatively correct. While clearly further increases of the number of states and lattice sizes will improve the accuracy, it is important to judge if at least the essence of the physics has been captured by our proposed method. In Fig. 6.14, results for $U=4$ are shown. This is a representative of the “large” U regime, since it must be compared with t' (as opposed to $t_{\text{leads}}=1$) that is only 0.4 in this figure. Another indication that this U is large is in Fig. 6.14(b) where a clear sharp quantization of the charge inside the dot is observed. In this large- U regime, the DMRG conductance is shown in (a). Clearly, there is a substantial difference between the Friedel sum-rule estimation (which has the correct “box” shape in the gate voltage range

Figure 6.13: Conductance G and the dot occupation $\langle n_d \rangle$ for one interacting QD. The circles show G obtained by averaging the current over an interval of time corresponding to the steady state, as shown in Fig.6.11. The squares show G obtained from $\langle n_d \rangle$ using the Friedel sum rule (FSR). G has the shape of the expected Kondo or mixed-valence plateau centered at $V_g = -U/2$. The feature would be sharper reducing t' . Results are shown for: (a) $U = 1.0$, $t' = 0.4$, and (b) $U = 2.0$, $t' = 0.5$. In both cases $L = 128$ and $M = 300$. Note that the DMRG conductance results in (a) show a slightly better agreement with the FSR results. This is expected since the finite size effects are stronger for larger U .

$[-U, 0]$) and the DMRG numbers. However, at least the fact that there must be a nonzero conductance at $V_g = -U/2$ was properly captured by the method. This example illustrates a case where size effects are important, due to the subtle rapid increase of the Kondo cloud with increasing U . A rough estimate of the Kondo cloud for these parameters gives a Kondo cloud of the order of thousands of sites. The results in this case show a slow convergence with L towards the FSR results. However, the qualitative results were captured, in spite of the fact that to reach a quantitative conclusion much larger sizes must be considered, or a careful finite-size scaling should be done. This case is shown as a cautionary example to the readers, that must be alert of the limitations of the numerical methods. Note that while FSR results are excellent, for other arbitrary cases it would be unclear whether the Friedel

Figure 6.14: Conductance G ($\Delta V=0.001$) and charge at the dot $\langle n_d \rangle$, for one interacting QD in the case of large U . The circles show G obtained from the DMRG procedure outlined in this paper, while the squares show G obtained from $\langle n_d \rangle$ using the Friedel sum-rule. The finite-size effects are obvious here, since the results are halfway between the expected Kondo plateau (properly reproduced by the FSR procedure) and the Coulomb blockade peaks. This case is shown as an illustration of important size effects in some limits. The parameters used are $U = 4.0$, $t' = 0.4$, $L = 128$, and $M = 300$.

sum-rule method is valid and, as a consequence, not always this procedure can be used.

6.4.3 Improving the Convergence

To carry out the investigations presented thus far in this and the previous section, the number of sites in the leads at left and right of the dot region have been chosen such that one lead has an *even* number and the other an *odd* number of sites (to refer to this case, the notation used below will be “odd-QDs-even”). While this should be irrelevant for very long chains, in practice this is important for the speed of convergence of the conductance calculations with increasing cluster size. For example, in the interacting case $U \neq 0$, a Kondo or mixed-valence regime is expected where the spin at the dot couples with electrons at the Fermi level of the leads. This formation of the Kondo cloud occurs more efficiently on a finite-size lattice if already a zero energy level is available, as it occurs when one of the leads has an odd number of sites. That this improves the rate of convergence with increasing lattice size is shown in

Fig. 6.15, where the odd-1QD-even case is contrasted to the even-1QD-even case, where in both leads the number of sites is even. Clearly, the odd-1QD-even case approaches the ideal limit $2e^2/h$ faster than using leads with an even number of sites, and it is recommended to be used in future investigations. The figure also shows that eventually with sufficiently large systems, both combinations will reach the same ideal result, as expected. The remaining possibility (odd-1QD-odd) was also investigated (not shown). For reasons that remain to be analyzed further, the convergence in this case is not as good as in the odd-1QD-even case. As a consequence, empirically it is clear that the combination even-QDs-odd is the most optimal to speed up the size convergence of the calculation.

Another method to improve the size convergence was tested. Following Refs. [126, 117], the finite-size effects can be reduced by using “damped boundary conditions” (DBC). The hoppings in the M_D links at the boundaries are reduced using the formulas $-td, -td^2, \dots, -td^{M_D}$, where $d < 1$. M_D has to be chosen such that the damping occurs far enough from the central region. Figure 6.16 shows the finite size scaling using odd-QD-even clusters and the same parameters as in Fig. 6.15, both with DBC and the standard open boundary conditions (OBC) used in the rest of the work. The latter indeed improves the convergence. While in the present study, OBC were used in most of the manuscript to keep the simplicity in the presentation and reduce the number of parameters, the use of DBC is recommended for cases where size effects are strong.

6.4.4 Influence of Magnetic Fields

To fully confirm that our investigations in the Kondo/mixed valence regime have captured the essence of the problem, namely the formation of a Kondo cloud with antiferromagnetic coupling between the spins at the leads and the dot, investigations including magnetic fields are necessary. In Fig. 6.17, it is shown how the Kondo plateau in the conductance evolves with increasing magnetic field. As expected from previous investigations, including results obtained using very different techniques such as the Lanczos method followed by a Dyson-equation embedding procedure (ED+DE) [119], the conductance broad peak splits with increasing magnetic field B . At large B , two peaks are observed at $-U$ and 0 , as it occurs also in the high temperature regime where only Coulomb blockade effects are present.

Figure 6.15: Finite-size scaling of the conductance G at $V_g = -U/2$ for the odd-1QD-even (circles) and even-1QD-even (triangles) clusters. Note that in both cases G converges to 1 in the bulk limit. However, the odd-1QD-even cluster converges faster, which makes it the most useful for practical calculations. The parameters used are $U = 1.0$ and $t' = 0.4$.

Figure 6.16: Finite size scaling of G for odd-QD-even clusters using OBC (circles) and DBC (triangles). $d = 0.5$ was used. Note that $L = 64$ cluster with DBC gives better results than a $L=128$ cluster with OBC. The same parameters are used as in Fig. 6.15.

Figure 6.17: Conductance G vs. gate voltage V_g in the case of one interacting QD ($U = 2.0$, $t' = 0.5$) for different values of the magnetic field B (and $\Delta V = 0.001$). For $B = 0$, a Kondo plateau is obtained, centered at $V_g = -U/2$. As B increases, the Kondo effect is suppressed, and for moderate B , two Coulomb blockade peaks are observed at $V_g = -U$ and $V_g = 0$, as expected.

6.5 Two Coupled Quantum Dots

The method discussed in this chapter is general, and in principle it can be implemented for a variety of complicated geometries and couplings in the interacting region between the leads. Thus, it is important to confirm that the method will keep its validity going beyond the one quantum dot case. In this section, the case of two dots is studied. Systems with two quantum dots are believed to be understood theoretically and, as a consequence, our numerical data can be contrasted against robust results in the literature. Cases involving more dots [120] are still not fully understood, and their analysis will be postponed for future investigations. In Fig. 6.18, the current vs. time is shown for two dots. The Hamiltonian for this case was already defined in previous sections. For a fixed t' , increasing the amplitude of the direct hopping between the dots t'' amounts to isolating the two dots system from the rest. As a consequence, the current is expected to decrease, and the method indeed reproduces this effect, as shown in the figure. The same physics is obtained reducing t' , at fixed t'' . In fact,

Figure 6.18: $J(t)/\Delta V$ for two coupled QDs at $V_g = -U/2$ for different values of t''/t'^2 (and $\Delta V=0.005$). As in the case of one interacting QD, the conductance is obtained by averaging the steady-state current over the indicated intervals. The parameters used are $U = 1.0$, $t' = 0.5$, $L = 127$, and $M = 300$.

previous studies [6] have shown that the conductance only depends on t''/t'^2 , and this has been verified using our method.

The conductance of a system with two dots in series will decrease with increasing t''/t'^2 (at large t''/t'^2) due to the decoupling of the two-dots system into a small two-sites molecule, as already discussed. But this conductance will also be very small at small t'' when the tunneling from one dot to the next is nearly cutoff. As a consequence, the conductance vs. t''/t'^2 is known to present a peak at intermediate values. In Fig. 6.19, the slave-boson mean-field technique (SBMFT) predictions for this case obtained in previous investigations [6] are shown together with our results. The agreement is fairly reasonable, providing further support that the method discussed here can handle systems where there are competing tendencies, beyond the one quantum-dot case.

6.6 Conclusions

In this chapter, a method was proposed and tested to calculate the conductance of small (nanoscale) strongly correlated systems modeled by tight-binding Hamiltonians. The

Figure 6.19: Conductance G as a function of t''/t^2 , at $V_g = -U/2$ ($\Delta V=0.005$). In this regime, G is determined by the competition between the Kondo correlation of each dot with the neighboring leads and the antiferromagnetic correlation between the two dots. The circles represent our DMRG results obtained with $L = 127$ and $M = 300$. The solid line is the plot of the functional form obtained by Georges and Meir using SBMFT [6].

approach is based on the adaptive time-dependent DMRG method, and it was shown to work properly for non-interacting systems and also in the cases of one and two quantum dots. Besides the finite size effects, discussed in the text as well, there are no other severe limitations to handle complex interacting models with arbitrary couplings. The method is a complement to DFT calculations in the nanoscopic context. Further improvements of the technique must consider temperature and inelastic effects.

It is concluded that the semi-quantitative analysis of transport in models of strongly correlated nanosystems appears under reach, and much progress is expected from the application of the technique presented here to realistic Hamiltonians for small molecules and arrays of quantum dots. The remarkable cross-fertilization between modeling, simulation, and experiments existing in bulk strongly correlated materials, such as transition-metal oxides, can be repeated in a variety of interesting systems at the nanoscale.

CHAPTER 7

CONCLUSION

We studied numerically the transport properties of several interesting nanosystems, focusing primarily on the correlation, interference and phononic effects. In our study of multilevel QDs presented in chapter 2, we observed interesting interference effects even in the absence of applied magnetic fields and different paths in real space for the electrons to take. We showed that the overall conductance of the dot can be decomposed into the contributions of different levels that can carry different phases. When the phase difference is $\pm\pi$, destructive interference leads to conductance cancellations. An integer spin ($S = 1$) Kondo effect was also observed in the even occupation regime. In addition, our studies revealed the possibility of novel conductance discontinuities as the gate voltage is varied. These discontinuities are found to be caused by localized states. Although they do not contribute directly to the conductance, such states get charged abruptly as they cross the Fermi level thus affecting the transport of other states through Coulomb interaction. Some of the above effects have been observed experimentally. In particular, the $S = 1$ Kondo effect was observed in multilevel QDs [8].

In an effort to make further connection between the physics of strongly correlated bulk materials and quantum dots, we proposed a possible realization of the double exchange mechanism in a QD system. As described in chapter 3, the system investigated consists of two coupled double-level QDs. A novel “ferromagnetic” phase is observed in the quarter filling regime. In particular, in the strong coupling regime, and when the interdot hopping is larger than the hopping to the leads, the two dots form an overall spin triplet. Kondo screening of the triplet gives a conductance peak. The mechanism of the ferromagnetic phase formation is analogous to the double exchange in bulk materials.

Another important result is presented in chapter 4: the simulation of a seminal experi-

mental effort involving nonlocal spin control in quantum dots. The authors of Ref. [5] report a possible realization of the RKKY interaction in a QD system. The importance of such achievement lies in the potential utilization of such long-range interaction to couple distant qubits in the quantum-dot architecture of a quantum computer. We reproduced numerically the main features of the experimental results. However, our investigations indicate the possibility of a Fano antiresonance, in the experimental setup, that can produce the same effects observed in the experiment. Thus, more studies have to be done in order to confirm the experimental realization of nonlocal spin control in quantum dots.

In addition to electronic correlations, electron-phonon coupling in single-molecule conductors can have interesting effects on the transport properties. Our investigation, presented in chapter 5, shows that the center-of-mass motion opens a new channel for spin and charge fluctuations. This channel can interfere destructively with the purely-electronic channel leading to a conductance cancellation.

Finally, in chapter 6, we proposed a procedure to study transport in strongly correlated nanosystems based on the adaptive time-dependent density-matrix renormalization group. We tested the technique for several cases involving both interacting and noninteracting systems. We were able to reproduce previous exact and well-established results. We also show that the technique works in the nonequilibrium case, thus providing a powerful numerical tool for nanotransport studies. The proposed procedure is already being used, by different groups, to study several interesting problems particularly nonequilibrium transport where other techniques are inapplicable.

APPENDIX A

THE EMBEDDED CLUSTER APPROXIMATION

The “embedded cluster approximation” (ECA) is a numerical technique to calculate the zero-temperature and zero-bias conductance of nanostructures. In general, such systems consist of a small interacting region, one or more quantum dots or a small molecule, connected to noninteracting semi-infinite leads which can be modeled as semi-infinite tight-binding chains.

The system is then divided into two parts: a small cluster consisting of the interacting region and a few sites of the leads, and the rest of the leads. The cluster, containing the many-body interactions, is treated exactly using the exact-diagonalization (Lanczos) method [102]. The ground state is calculated as well as a set of Green functions g_{ij} where i, j are the site indices. Since Kondo physics is important in all the studies presented, treating exactly not only the dot but also some sites belonging to the leads allows for a proper quantitative consideration of the spin-singlet Kondo “cloud.” The rest of the system is a one-body problem and an analytic expression for the Green functions can be found.

In order to reestablish the connection between the two parts of the system, a Dyson equation “embedding” procedure is used. The embedded cluster Green functions G_{ij} are calculated using the Dyson equation:

$$\hat{G} = \hat{g} + \hat{g}\hat{t}\hat{G}, \quad (\text{A.1})$$

where the matrices $\hat{g}$ and $\hat{G}$ have the elements g_{ij} and G_{ij} respectively, that is, the isolated and embedded cluster Green functions. The matrix $\hat{t}$ contains the hopping amplitudes between the cluster and the rest of the leads.

To take into account possible charge fluctuations inside the cluster, a linear combination of $\hat{g}$ ’s calculated with different number of particles is considered. For this purpose, $\hat{g}^p = (1 - p)\hat{g}_n + p \hat{g}_{n+1}$ is defined, where $\hat{g}_m$ is the Green function for m particles. With

the dressed Green function $\hat{G}^p$, the total charge inside the cluster after the embedding is calculated using $Q^p = -1/\pi \int_{-\infty}^{E_F} \sum_i \text{Im } G_{ii}^p(\omega) d\omega$. On the other hand, the charge in the cluster before the embedding process is $q^p = (1-p)n + p(n+1)$. These two quantities must be equal ($Q^p = q^p$), defining a self-consistent equation for p .

Finally the conductance G can be calculated using the equation

$$G = 2 \frac{e^2}{h} |t^2 G_{lr}(\epsilon_F)|^2 [\rho(\epsilon_F)]^2, \quad (\text{A.2})$$

where $2e^2/h$ is the quantum unit of conductance for the two spin species. G_{lr} is the Green function that propagates an electron from a site in the left lead to a site in the right one. $\rho(\epsilon_F)$ is the density of states of the leads at the Fermi level ϵ_F . For an ideal semi-infinite chain with hopping t , the density of states is given by

$$\rho(\omega) = -\frac{1}{\pi} \text{Im} \left\{ \frac{(\omega + i\eta) - \sqrt{(\omega + i\eta)^2 - 4t^2}}{2t^2} \right\}, \quad (\text{A.3})$$

where $i\eta$ is a small imaginary part introduced to shift the poles of the Green function to the complex plane. In general, the Fermi level is set at $\omega = 0$. In this case the density of states reduces to $\rho(\epsilon_F \equiv 0) = 1/\pi t$.

REFERENCES

- [1] D. Goldhaber-Gordon, Hadas Shtrikman, D. Mahalu, David Abusch-Magder, U. Meirav, and M. A. Kastner. Kondo effect in a single-electron transistor. *Nature*, 391(6663):156–159, Jan 1998. ([document](#)), [1.1.1](#), [1.1](#), [1.4](#), [1.4](#), [2.2](#), [3.1](#), [4.1](#), [5.1](#), [6.1](#)
- [2] Jiwoong Park, Abhay N. Pasupathy, Jonas I. Goldsmith, Connie Chang, Yuval Yaish, Jason R. Petta, Marie Rinkoski, James P. Sethna, Hector D. Abrua, Paul L. McEuen, and Daniel C. Ralph. Coulomb blockade and the kondo effect in single-atom transistors. *Nature*, 417(6890):722–725, Jun 2002. ([document](#)), [1.1](#), [1.4](#), [5.1](#)
- [3] Leo Kouwenhoven and Leonid Glazman. Revival of the kondo effect. *Physics World*, 14(1):33–38, Jan 2001. ([document](#)), [1.2](#)
- [4] W. G. van der Wiel, S. De Franceschi, T. Fujisawa, J. M. Elzerman, S. Tarucha, and L. P. Kouwenhoven. The kondo effect in the unitary limit. *Science*, 289(5487):2105–2108, Sep 200. ([document](#)), [1.4](#), [1.4](#)
- [5] N. J. Craig, J. M. Taylor, E. A. Lester, C. M. Marcus, M. P. Hanson, and A. C. Gossard. Tunable nonlocal spin control in a coupled-quantum dot system. *Science*, 304(5670):565–567, Apr 2004. ([document](#)), [1.5](#), [1.4](#), [1.5](#), [3.1](#), [4.1](#), [4.1](#), [4.2](#), [4.3](#), [4.5](#), [4.6](#), [7](#)
- [6] Antoine Georges and Yigal Meir. Electronic correlations in transport through coupled quantum dots. *Phys. Rev. Lett.*, 82(17):3508–3511, Apr 1999. ([document](#)), [1.4](#), [3.1](#), [6.5](#), [6.5](#), [6.19](#)
- [7] H. Jeong, A. M. Chang, and M. R. Melloch. The kondo effect in an artificial quantum dot molecule. *Science*, 293(5538):2221–2223, Sep 2001. [1.1.1](#), [1.4](#)
- [8] S. Sasaki, S. De Franceschi, J. M. Elzerman, W. G. van der Wiel, M. Eto, S. Tarucha, and L. P. Kouwenhoven. Kondo effect in an integer-spin quantum dot. *Nature*, 405(6788):764–767, Jun 2000. [1.1.1](#), [1.4](#), [2.1](#), [2.4](#), [7](#)
- [9] Pablo Jarillo-Herrero, Jing Kong, Herre S.J. van der Zant, Cees Dekker, Leo P. Kouwenhoven, and Silvano De Franceschi. Orbital kondo effect in carbon nanotubes. *Nature*, 434(7032):484–488, Mar 2005. [1.1.1](#), [1.4](#)
- [10] R. M. Potok, I. G. Rau, Hadas Shtrikman, Yuval Oreg, and D. Goldhaber-Gordon. Observation of the two-channel kondo effect. *Nature*, 446(7132):167–171, Mar 2007. [1.1.1](#)

- [11] Daniel Loss and David P. DiVincenzo. Quantum computation with quantum dots. *Phys. Rev. A*, 57(1):120–126, Jan 1998. [1.1.2](#), [4.1](#)
- [12] J. M. Taylor, C. M. Marcus, and M. D. Lukin. Long-lived memory for mesoscopic quantum bits. *Phys. Rev. Lett.*, 90(20):206803, May 2003. [1.1.2](#)
- [13] V. V. Dobrovitski, J. M. Taylor, and M. D. Lukin. Long-lived memory for electronic spin in a quantum dot: Numerical analysis. *Physical Review B (Condensed Matter and Materials Physics)*, 73(24):245318, 2006. [1.1.2](#)
- [14] K. A. Al-Hassanieh, V. V. Dobrovitski, E. Dagotto, and B. N. Harmon. Numerical modeling of the central spin problem using the spin-coherent-state p representation. *Physical Review Letters*, 97(3):037204, 2006. [1.1.2](#)
- [15] Wenxian Zhang, V. V. Dobrovitski, K. A. Al-Hassanieh, E. Dagotto, and B. N. Harmon. Hyperfine interaction induced decoherence of electron spins in quantum dots. *Physical Review B (Condensed Matter and Materials Physics)*, 74(20):205313, 2006. [1.1.2](#)
- [16] Wenxian Zhang, N. Konstantinidis, K. A. Al-Hassanieh, and V. V. Dobrovitski. Modelling decoherence in quantum spin systems. *Journal of Physics: Condensed Matter*, 19(8):083202 (28pp), 2007. [1.1.2](#)
- [17] I. A. Merkulov, Al. L. Efros, and M. Rosen. Electron spin relaxation by nuclei in semiconductor quantum dots. *Phys. Rev. B*, 65(20):205309, Apr 2002. [1.1.2](#)
- [18] Alexander V. Khaetskii, Daniel Loss, and Leonid Glazman. Electron spin decoherence in quantum dots due to interaction with nuclei. *Phys. Rev. Lett.*, 88(18):186802, Apr 2002. [1.1.2](#)
- [19] Changxue Deng and Xuedong Hu. Analytical solution of electron spin decoherence through hyperfine interaction in a quantum dot. *Physical Review B (Condensed Matter and Materials Physics)*, 73(24):241303, 2006. [1.1.2](#)
- [20] J. R. Schrieffer and P. A. Wolff. Relation between the anderson and kondo hamiltonians. *Phys. Rev.*, 149(2):491–492, Sep 1966. [1.3](#)
- [21] Yigal Meir, Ned S. Wingreen, and Patrick A. Lee. Low-temperature transport through a quantum dot: The anderson model out of equilibrium. *Phys. Rev. Lett.*, 70(17):2601–2604, Apr 1993. [1.3](#), [1.4](#), [2.1](#), [2.2](#), [5.1](#), [6.1](#), [6.4.1](#)
- [22] C. A. Büsser, E. V. Anda, A. L. Lima, Maria A. Davidovich, and G. Chiappe. Transport in coupled quantum dots: Kondo effect versus antiferromagnetic correlation. *Phys. Rev. B*, 62(15):9907–9910, Oct 2000. [1.4](#), [2.2](#), [2.3](#), [3.1](#), [3.3](#), [3.3](#)
- [23] Wenjie Liang, Matthew P. Shores, Marc Bockrath, Jeffrey R. Long, and Hongkun Park. Kondo resonance in a single-molecule transistor. *Nature*, 417(6890):725–729, Jun 2002. [1.4](#), [5.1](#), [6.1](#)

- [24] L.H. Yu and D. Natelson. The kondo effect in c_{60} single-molecule transistors. *Nano Lett.*, 4(1):79–83, Jan 2004. [1.4](#), [5.1](#)
- [25] Hongkun Park, Jiwoong Park, Andrew K. L. Lim, Erik H. Anderson, A. Paul Alivisatos, and Paul L. McEuen. Nanomechanical oscillations in a single- c_{60} transistor. *Nature*, 407(6800):57–60, Sep 2000. [1.4](#), [5.1](#)
- [26] A. Mitra, I. Aleiner, and A. J. Millis. Phonon effects in molecular transistors: Quantal and classical treatment. *Physical Review B (Condensed Matter and Materials Physics)*, 69(24):245302, 2004. [1.4](#), [5.1](#)
- [27] K. A. Al-Hassanieh, C. A. Busser, G. B. Martins, and E. Dagotto. Electron transport through a molecular conductor with center-of-mass motion. *Physical Review Letters*, 95(25):256807, 2005. [1.4](#), [5.1](#), [6.1](#), [6.4.1](#)
- [28] P. S. Cornaglia, H. Ness, and D. R. Grempel. Many-body effects on the transport properties of single-molecule devices. *Physical Review Letters*, 93(14):147201, 2004. [1.4](#), [5.1](#), [5.3](#), [5.3](#), [6.1](#)
- [29] P. S. Cornaglia, D. R. Grempel, and H. Ness. Quantum transport through a deformable molecular transistor. *Physical Review B (Condensed Matter and Materials Physics)*, 71(7):075320, 2005. [1.4](#), [5.1](#), [5.3](#)
- [30] E. Vernek, E. V. Anda, S. E. Ulloa, and N. Sandler. Phonon rabi-assisted tunneling in diatomic molecules. *Physical Review B (Condensed Matter and Materials Physics)*, 72(12):121405, 2005. [1.4](#), [5.1](#)
- [31] D. Fedorets. Quantum description of shuttle instability in a nanoelectromechanical single-electron transistor. *Phys. Rev. B*, 68(3):033106, Jul 2003. [1.4](#), [5.1](#)
- [32] D. Fedorets, L. Y. Gorelik, R. I. Shekhter, and M. Jonson. Vibrational instability due to coherent tunneling of electrons. *Europhysics Letters (EPL)*, 58(1):99–104, 2002. [1.4](#), [5.1](#)
- [33] D. Fedorets, L. Y. Gorelik, R. I. Shekhter, and M. Jonson. Quantum shuttle phenomena in a nanoelectromechanical single-electron transistor. *Physical Review Letters*, 92(16):166801, 2004. [1.4](#), [5.1](#)
- [34] L. Y. Gorelik, A. Isacsson, M. V. Voinova, B. Kasemo, R. I. Shekhter, and M. Jonson. Shuttle mechanism for charge transfer in coulomb blockade nanostructures. *Phys. Rev. Lett.*, 80(20):4526–4529, May 1998. [1.4](#), [5.1](#)
- [35] Tomáš Novotný, Andrea Donarini, and Antti-Pekka Jauho. Quantum shuttle in phase space. *Phys. Rev. Lett.*, 90(25):256801, Jun 2003. [1.4](#), [5.1](#)
- [36] Tomas Novotny, Andrea Donarini, Christian Flindt, and Antti-Pekka Jauho. Shot noise of a quantum shuttle. *Physical Review Letters*, 92(24):248302, 2004. [1.4](#), [5.1](#)

- [37] Maxim G. Vavilov and Leonid I. Glazman. Transport spectroscopy of kondo quantum dots coupled by rky interaction. *Physical Review Letters*, 94(8):086805, 2005. [1.5](#), [4.1](#), [4.5](#)
- [38] Rosa Lopez, David Sanchez, Minchul Lee, Mahn-Soo Choi, Pascal Simon, and Karyn Le Hur. Probing spin and orbital kondo effects with a mesoscopic interferometer. *Physical Review B (Condensed Matter and Materials Physics)*, 71(11):115312, 2005. [1.5](#)
- [39] Steven R. White and Adrian E. Feiguin. Real-time evolution using the density matrix renormalization group. *Physical Review Letters*, 93(7):076401, 2004. [1.5](#), [6.1](#), [6.2.4](#), [6.2.4](#), [6.3](#)
- [40] Adrian E. Feiguin and Steven R. White. Time-step targeting methods for real-time dynamics using the density matrix renormalization group. *Physical Review B (Condensed Matter and Materials Physics)*, 72(2):020404, 2005. [1.5](#), [6.1](#)
- [41] A J Daley, C Kollath, U Schollwöck, and G Vidal. Time-dependent density-matrix renormalization-group using adaptive effective hilbert spaces. *Journal of Statistical Mechanics: Theory and Experiment*, 2004(04):P04005, 2004. [1.5](#), [6.1](#), [6.2.4](#), [6.3](#)
- [42] Daniel Loss and Eugene V. Sukhorukov. Probing entanglement and nonlocality of electrons in a double-dot via transport and noise. *Phys. Rev. Lett.*, 84(5):1035–1038, Jan 2000. [2.1](#)
- [43] Björn Kubala and Jürgen König. Flux-dependent level attraction in double-dot aharonov-bohm interferometers. *Phys. Rev. B*, 65(24):245301, May 2002. [2.1](#)
- [44] Daniel Boese, Walter Hofstetter, and Herbert Schoeller. Interference and interaction effects in multilevel quantum dots. *Phys. Rev. B*, 64(12):125309, Sep 2001. [2.1](#)
- [45] A. A. Aligia, K. Hallberg, B. Normand, and A. P. Kampf. Detection of topological transitions by transport through molecules and nanodevices. *Physical Review Letters*, 93(7):076801, 2004. [2.1](#)
- [46] K. Hallberg, A. A. Aligia, A. P. Kampf, and B. Normand. Spin-charge separation in aharonov-bohm rings of interacting electrons. *Physical Review Letters*, 93(6):067203, 2004. [2.1](#)
- [47] M. L. Ladrón de Guevara, F. Claro, and Pedro A. Orellana. Ghost fano resonance in a double quantum dot molecule attached to leads. *Phys. Rev. B*, 67(19):195335, May 2003. [2.1](#)
- [48] Alessandro Silva, Yuval Oreg, and Yuval Gefen. Signs of quantum dot[∗]lead matrix elements: The effect on transport versus spectral properties. *Phys. Rev. B*, 66(19):195316, Nov 2002. [2.1](#), [3.1](#)
- [49] Tae-Suk Kim and S. Hershfield. Effects of signs in tunneling matrix elements on transmission zeros and phase. *Phys. Rev. B*, 67(23):235330, Jun 2003. [2.1](#), [3.1](#)

- [50] L. I. Glazman and M. E. Raikh. Resonant kondo transparency of a barrier with quasilocal impurity states. *JETP Letters*, 47(8):452–455, Apr 1988. [2.1](#), [5.1](#), [6.1](#), [6.4.1](#)
- [51] Tai Kai Ng and Patrick A. Lee. On-site coulomb repulsion and resonant tunneling. *Phys. Rev. Lett.*, 61(15):1768–1771, Oct 1988. [2.1](#), [5.1](#), [6.1](#), [6.4.1](#)
- [52] M. Pustilnik and L. I. Glazman. Conduction through a quantum dot near a singlet-triplet transition. *Phys. Rev. Lett.*, 85(14):2993–2996, Oct 2000. [2.1](#), [2.4](#)
- [53] Maria A. Davidovich, E. V. Anda, C. A. Büsser, and G. Chiappe. Quantum dot spin effect on the conductance of a quantum wire. *Phys. Rev. B*, 65(23):233310, May 2002. [2.1](#), [2.3](#), [2.4](#), [2.4](#), [6.4.1](#)
- [54] A. Yacoby, M. Heiblum, D. Mahalu, and Hadas Shtrikman. Coherence and phase sensitive measurements in a quantum dot. *Phys. Rev. Lett.*, 74(20):4047–4050, May 1995. [2.1](#)
- [55] Walter Hofstetter and Herbert Schoeller. Quantum phase transition in a multilevel dot. *Phys. Rev. Lett.*, 88(1):016803, Dec 2001. [2.1](#), [3.1](#)
- [56] Daniel Boese, Walter Hofstetter, and Herbert Schoeller. Interference in interacting quantum dots with spin. *Phys. Rev. B*, 66(12):125315, Sep 2002. [2.1](#), [3.1](#)
- [57] Shi-Jie Xiong and Yue Yin. Asymmetric line shape and fano interference in the transport of electrons through a multilevel quantum dot in the coulomb blockade regime. *Phys. Rev. B*, 66(15):153315, Oct 2002. [2.1](#)
- [58] Yigal Meir and Ned S. Wingreen. Landauer formula for the current through an interacting electron region. *Phys. Rev. Lett.*, 68(16):2512–2515, Apr 1992. [2.2](#), [3.2](#), [4.2](#), [5.2](#)
- [59] E. V. Anda and F. Flores. The role of inelastic scattering in resonant tunnelling heterostructures. *Journal of Physics: Condensed Matter*, 3(46):9087–9101, 1991. [2.2](#), [3.2](#), [5.2](#)
- [60] Joel A. Appelbaum and David R. Penn. Localized correlations in narrow conduction bands. i. *Phys. Rev.*, 188(2):874–887, Dec 1969. [2.2](#)
- [61] C. Lacroix. Density of states for the anderson model. *Journal of Physics F: Metal Physics*, 11(11):2389–2397, 1981. [2.2](#)
- [62] A. C. Hewson. *The Kondo Problem to Heavy Fermions*. Cambridge University Press, Cambridge, UK, 1993. [2.2](#), [5.3](#)
- [63] Elbio Dagotto, Takashi Hotta, and Adriana Moreo. Colossal magnetoresistant materials: the key role of phase separation. *Physics Reports*, 344(1-3):1–153, Apr 2001. [2.2](#), [6.1](#)
- [64] J. Kuei and R. T. Scalettar. Ferromagnetism in an orbitally degenerate hubbard model. *Phys. Rev. B*, 55(22):14968–14974, Jun 1997. [2.2](#)

- [65] André Luiz Malvezzi and Elbio Dagotto. Origin of spin incommensurability in hole-doped $s = 1y_{2-x}ca_xbanio_5$ chains. *Phys. Rev. B*, 63(14):140409, Mar 2001. [2.2](#)
- [66] Hisashi Aikawa, Kensuke Kobayashi, Akira Sano, Shingo Katsumoto, and Yasuhiro Iye. Interference effect in multi-level transport through a quantum dot. *Journal of the Physical Society of Japan*, 73(12):3235–3238, Dec 2004. [2.6](#)
- [67] C. Jayaprakash, H. R. Krishna-murthy, and J. W. Wilkins. Two-impurity kondo problem. *Phys. Rev. Lett.*, 47(10):737–740, Sep 1981. [3.1](#)
- [68] Kevin Ingersent, Barbara A. Jones, and John W. Wilkins. Study of the two-impurity, two-channel kondo hamiltonian. *Phys. Rev. Lett.*, 69(17):2594–2597, Oct 1992. [3.1](#)
- [69] Tomosuke Aono and Mikio Eto. Kondo resonant spectra in coupled quantum dots. *Phys. Rev. B*, 63(12):125327, Mar 2001. [3.1](#)
- [70] Rosa López, Ramón Aguado, and Gloria Platero. Nonequilibrium transport through double quantum dots: Kondo effect versus antiferromagnetic coupling. *Phys. Rev. Lett.*, 89(13):136802, Sep 2002. [3.1](#)
- [71] Ramón Aguado and David C. Langreth. Kondo effect in coupled quantum dots: A noncrossing approximation study. *Phys. Rev. B*, 67(24):245307, Jun 2003. [3.1](#)
- [72] M. N. Kiselev, K. Kikoin, and L. W. Molenkamp. Resonance kondo tunneling through a double quantum dot at finite bias. *Phys. Rev. B*, 68(15):155323, Oct 2003. [3.1](#)
- [73] W. G. van der Wiel, S. De Franceschi, J. M. Elzerman, T. Fujisawa, S. Tarucha, and L. P. Kouwenhoven. Electron transport through double quantum dots. *Rev. Mod. Phys.*, 75(1):1–22, Dec 2002. [3.1](#), [4.1](#)
- [74] C. Zener. Interaction between the d shells in the transition metals. *Phys. Rev.*, 81(3):440–444, Feb 1951. [3.1](#)
- [75] A. Levy Yeyati, F. Flores, and A. Martín-Rodero. Transport in multilevel quantum dots: From the kondo effect to the coulomb blockade regime. *Phys. Rev. Lett.*, 83(3):600–603, Jul 1999. [3.1](#)
- [76] Pascal Simon, Rosa Lopez, and Yuval Oreg. Ruderman-kittel-kasuya-yosida and magnetic-field interactions in coupled kondo quantum dots. *Physical Review Letters*, 94(8):086602, 2005. [4.1](#), [4.5](#)
- [77] R. Franco, M. S. Figueira, and E. V. Anda. Fano resonance in electronic transport through a quantum wire with a side-coupled quantum dot: X-boson treatment. *Phys. Rev. B*, 67(15):155301, Apr 2003. [4.3](#)
- [78] Masahiro Sato, Hisashi Aikawa, Kensuke Kobayashi, Shingo Katsumoto, and Yasuhiro Iye. Observation of the fano-kondo antiresonance in a quantum wire with a side-coupled quantum dot. *Physical Review Letters*, 95(6):066801, 2005. [4.4](#), [4.4](#), [4.5](#)

- [79] Supriyo Datta. *Electronic Transport in Mesoscopic Systems*. Cambridge University Press, Cambridge, UK, 1995. [4.4](#)
- [80] Christian Joachim, James K. Gimzewski, Reto R. Schlittler, and Corinne Chavy. Electronic transparency of a single c_{60} molecule. *Phys. Rev. Lett.*, 74(11):2102–2105, Mar 1995. [5.1](#)
- [81] M. A. Reed, C. Zhou, C. J. Muller, T. P. Burgin, and J. M. Tour. Conductance of a molecular junction. *Science*, 276(5336):252–254, Oct 1997. [5.1](#)
- [82] R. H. M. Smit, Y. Noat, C. Untiedt, N. D. Lang, M. C. van Hemert, and J. M. van Ruitenbeek. Measurement of the conductance of a hydrogen molecule. *Nature*, 419(6910):906–909, Oct 2002. [5.1](#)
- [83] J. Reichert, R. Ochs, D. Beckmann, H. B. Weber, M. Mayor, and H. v. Löhneysen. Driving current through single organic molecules. *Phys. Rev. Lett.*, 88(17):176804, Apr 2002. [5.1](#)
- [84] J. Paaske, A. Rosch, J. Kroha, and P. Wolfle. Nonequilibrium transport through a kondo dot: Decoherence effects. *Physical Review B (Condensed Matter and Materials Physics)*, 70(15):155301, 2004. [5.1](#)
- [85] Sara M. Cronenwett, Tjerk H. Oosterkamp, and Leo P. Kouwenhoven. A tunable kondo effect in quantum dots. *Science*, 281(5376):540–544, Jul 1998. [5.1](#)
- [86] J. Göres, D. Goldhaber-Gordon, S. Heemeyer, M. A. Kastner, Hadas Shtrikman, D. Mahalu, and U. Meirav. Fano resonances in electronic transport through a single-electron transistor. *Phys. Rev. B*, 62(3):2188–2194, Jul 2000. [5.1](#)
- [87] C. A. Busser, G. B. Martins, K. A. Al-Hassanieh, Adriana Moreo, and Elbio Dagotto. Interference effects in the conductance of multilevel quantum dots. *Physical Review B*, 70(24):245303, 2004. [5.1](#), [5.3](#), [6.1](#), [6.4.1](#)
- [88] Jorge L. D’Amato, Horacio M. Pastawski, and Juan F. Weisz. Half-integer and integer quantum-flux periods in the magnetoresistance of one-dimensional rings. *Phys. Rev. B*, 39(6):3554–3562, Feb 1989. [5.1](#)
- [89] L. Arrachea, A. A. Aligia, and G. E. Santoro. Microscopic theory of vibronic dynamics in linear polyenes. *Phys. Rev. B*, 67(13):134307, Apr 2003. [5.2](#)
- [90] William Barford, Robert J. Bursill, and David Yaron. Dynamical model of the dielectric screening of conjugated polymers. *Physical Review B (Condensed Matter and Materials Physics)*, 69(15):155203, 2004. [5.2](#)
- [91] Horacio M. Pastawski. Classical and quantum transport from generalized landauer-büttiker equations. ii. time-dependent resonant tunneling. *Phys. Rev. B*, 46(7):4053–4070, Aug 1992. [5.2](#)

- [92] A. C. Hewson and D. Meyer. Numerical renormalization group study of the anderson-holstein impurity model. *Journal of Physics: Condensed Matter*, 14(3):427–445, 2002. [5.3](#)
- [93] Arie Aviram and Mark A. Ratner. Molecular rectifiers. *Chem. Phys. Lett.*, 29(2):277–283, Nov 1974. [6.1](#)
- [94] C. Joachim, J. K. Gimzewski, and A. Aviram. Electronics using hybrid-molecular and mono-molecular devices. *Nature*, 408(6812):541–548, Nov 2000. [6.1](#)
- [95] Abraham Nitzan and Mark A. Ratner. Electron transport in molecular wire junctions. *Science*, 300(5624):1384–1389, May 2003. [6.1](#)
- [96] J. Chen, M. A. Reed, A. M. Rawlett, and J. M. Tour. Large on-off ratios and negative differential resistance in a molecular electronic device. *Science*, 286(5444):1550–1552, Nov 1999. [6.1](#)
- [97] R. C. Ashoori. Electrons in artificial atoms. *Nature*, 379(6564):413–419, Feb 1996. [6.1](#)
- [98] Supriyo Datta, Weidong Tian, Seunghun Hong, R. Reifengerger, Jason I. Henderson, and Clifford P. Kubiak. Current-voltage characteristics of self-assembled monolayers by scanning tunneling microscopy. *Phys. Rev. Lett.*, 79(13):2530–2533, Sep 1997. [6.1](#)
- [99] M. Di Ventra, S. T. Pantelides, and N. D. Lang. First-principles calculation of transport properties of a molecular device. *Phys. Rev. Lett.*, 84(5):979–982, Jan 2000. [6.1](#)
- [100] San-Huang Ke, Harold U. Baranger, and Weitao Yang. Addition energies of fullerenes and carbon nanotubes as quantum dots: The role of symmetry. *Phys. Rev. Lett.*, 91(11):116803, Sep 2003. [6.1](#)
- [101] B. Muralidharan, A. W. Ghosh, and S. Datta. Probing electronic excitations in molecular conduction. *Physical Review B (Condensed Matter and Materials Physics)*, 73(15):155410, 2006. [6.1](#)
- [102] Elbio Dagotto. Correlated electrons in high-temperature superconductors. *Rev. Mod. Phys.*, 66(3):763–840, Jul 1994. [6.1](#), [A](#)
- [103] Steven R. White. Density matrix formulation for quantum renormalization groups. *Phys. Rev. Lett.*, 69(19):2863–2866, Nov 1992. [6.1](#)
- [104] U. Schollwock. The density-matrix renormalization group. *Reviews of Modern Physics*, 77(1):259, 2005. [6.1](#)
- [105] Karen A. Hallberg. Density-matrix algorithm for the calculation of dynamical properties of low-dimensional systems. *Phys. Rev. B*, 52(14):R9827–R9830, Oct 1995. [6.1](#), [6.3](#)
- [106] Till D. Kühner and Steven R. White. Dynamical correlation functions using the density matrix renormalization group. *Phys. Rev. B*, 60(1):335–343, Jul 1999. [6.1](#)

- [107] Eric Jeckelmann. Dynamical density-matrix renormalization-group method. *Phys. Rev. B*, 66(4):045114, Jul 2002. [6.1](#)
- [108] M. A. Cazalilla and J. B. Marston. Time-dependent density-matrix renormalization group: A systematic method for the study of quantum many-body out-of-equilibrium systems. *Phys. Rev. Lett.*, 88(25):256403, Jun 2002. [6.1](#), [6.3](#)
- [109] M. A. Cazalilla and J. B. Marston. Cazalilla and marston reply:. *Phys. Rev. Lett.*, 91(4):049702, Jul 2003. [6.1](#)
- [110] H. G. Luo, T. Xiang, and X. Q. Wang. Comment on itime-dependent density-matrix renormalization group: A systematic method for the study of quantum many-body out-of-equilibrium systemsj. *Phys. Rev. Lett.*, 91(4):049701, Jul 2003. [6.1](#)
- [111] Peter Schmitteckert. Nonequilibrium electron transport using the density matrix renormalization group method. *Physical Review B (Condensed Matter and Materials Physics)*, 70(12):121302, 2004. [6.1](#), [6.3](#)
- [112] Guifré Vidal. Efficient classical simulation of slightly entangled quantum computations. *Phys. Rev. Lett.*, 91(14):147902, Oct 2003. [6.1](#), [6.2.4](#)
- [113] Frithjof B. Anders and Avraham Schiller. Real-time dynamics in quantum-impurity systems: A time-dependent numerical renormalization-group approach. *Physical Review Letters*, 95(19):196801, 2005. [6.1](#)
- [114] R. A. Molina, P. Schmitteckert, D. Weinmann, R. A. Jalabert, G.-L. Ingold, and J.-L. Pichard. Residual conductance of correlated one-dimensional nanosystems: A numerical approach. *Eur. Phys. J. B*, 39(1):107–120, May 2004. [6.1](#)
- [115] V. Meden and U. Schollwöck. Conductance of interacting nanowires. *Phys. Rev. B*, 67(19):193303, May 2003. [6.1](#)
- [116] V. Meden and U. Schollwöck. Persistent currents in mesoscopic rings: A numerical and renormalization group study. *Phys. Rev. B*, 67(3):035106, Jan 2003. [6.1](#)
- [117] D. Bohr, P. Schmitteckert, and P. Wlfle. Dmrg evaluation of the kubo formula - conductance of strongly interacting quantum systems. *Europhys. Lett.*, 73(2):246–252, Jan 2006. [6.1](#), [6.4.3](#)
- [118] Liliana Arrachea and Marcelo J. Rozenberg. Quantum monte carlo method for models of molecular nanodevices. *Physical Review B (Condensed Matter and Materials Physics)*, 72(4):041301, 2005. [6.1](#)
- [119] M. E. Torio, K. Hallberg, A. H. Ceccatto, and C. R. Proetto. Kondo resonances and fano antiresonances in transport through quantum dots. *Phys. Rev. B*, 65(8):085302, Jan 2002. [6.1](#), [6.4.4](#)
- [120] C. A. Busser, Adriana Moreo, and Elbio Dagotto. Conductance dip in the kondo regime of linear arrays of quantum dots. *Physical Review B (Condensed Matter and Materials Physics)*, 70(3):035402, 2004. [6.1](#), [6.4.1](#), [6.5](#)

- [121] G. B. Martins, C. A. Busser, K. A. Al-Hassanieh, A. Moreo, and E. Dagotto. Prediction of ferromagnetic correlations in coupled double-level quantum dots. *Physical Review Letters*, 94(2):026804, 2005. [6.1](#), [6.4.1](#)
- [122] G. B. Martins, C. A. Busser, K. A. Al-Hassanieh, E. V. Anda, A. Moreo, and E. Dagotto. Transport properties of strongly correlated electrons in quantum dots studied with a simple circuit model. *Physical Review Letters*, 96(6):066802, 2006. [6.1](#), [6.4.1](#)
- [123] Dominique Gobert, Corinna Kollath, Ulrich Schollwöck, and Gunter Schutz. Real-time dynamics in spin-(1/2) chains with adaptive time-dependent density matrix renormalization group. *Physical Review E (Statistical, Nonlinear, and Soft Matter Physics)*, 71(3):036102, 2005. [6.2.4](#)
- [124] V. Ferrari, G. Chiappe, E. V. Anda, and Maria A. Davidovich. Kondo resonance effect on persistent currents through a quantum dot in a mesoscopic ring. *Phys. Rev. Lett.*, 82(25):5088–5091, Jun 1999. [6.4.1](#)
- [125] Ned S. Wingreen, Antti-Pekka Jauho, and Yigal Meir. Time-dependent transport through a mesoscopic structure. *Phys. Rev. B*, 48(11):8487–8490, Sep 1993. [6.4.1](#)
- [126] M. Vekić and S. R. White. Smooth boundary conditions for quantum lattice systems. *Phys. Rev. Lett.*, 71(26):4283–4286, Dec 1993. [6.4.3](#)

BIOGRAPHICAL SKETCH

Khaled A. Al-Hassanieh

Personal Information

Place of birth	Ayn Jarfa, Lebanon.
Date of birth	March 4, 1980.
Nationality	Lebanese.
Languages	Arabic (native tongue) and English (fluent).

Education

2007	PhD in Physics, Florida State University.
2004	MSc in Physics, Florida State University.
2001	BSc in Chemistry, American University of Beirut.

Publications

“Interference Effects in the Conductance of Multi-Level Quantum Dots,”
C. A. Büsser, G. B. Martins, K. A. Al-Hassanieh, A. Moreo, and E. Dagotto,
Phys. Rev. B **70**, 245303 (2004).

“Prediction of Ferromagnetic Correlations in Coupled Double-Level Quantum Dots,”
G. B. Martins, C. A. Büsser, K. A. Al-Hassanieh, A. Moreo, and E. Dagotto,
Phys. Rev. Lett. **94**, 026804 (2005).

“Electron Transport through a Molecular Conductor with Center-of-Mass Motion,”
K. A. Al-Hassanieh, C. A. Büsser, G. B. Martins, and E. Dagotto,
Phys. Rev. Lett. **95**, 256807 (2005).

“Transport Properties of Strongly Correlated Electrons in Quantum Dots Studied with a Simple Circuit Model,”

G. B. Martins, C. A. Büsser, K. A. Al-Hassanieh, E. V. Anda, A. Moreo, and E. Dagotto, **Phys. Rev. Lett.** **96**, 066802 (2006).

“Numerical Modeling of the Central Spin Problem Using the Spin Coherent States P-representation,”

K. A. Al-Hassanieh, V. V. Dobrovitski, E. Dagotto, and B. N. Harmon, **Phys. Rev. Lett.** **97** 037204 (2006).

“Adaptive Time-Dependent Density-Matrix Renormalization Group Technique for Calculating the Conductance of Strongly Correlated Nanostructures,”

K. A. Al-Hassanieh, A. E. Feiguin, J. A. Riera, C. A. Büsser, and E. Dagotto, **Phys. Rev. B** **73**, 195304 (2006).

“Hyperfine Interaction Induced Decoherence of Electron Spins in Quantum Dots,”

Wenxian Zhang, V. V. Dobrovitski, K. A. Al-Hassanieh, E. Dagotto, and B. N. Harmon, **Phys. Rev. B** **74**, 205313 (2006).

“Modelling decoherence in quantum spin systems,”

Wenxian Zhang, N. Konstantinidis, K. A. Al-Hassanieh, and V. V. Dobrovitski, **J. Phys.: Condens. Matter** **19**, 083202 (2007). (Topical review).

“Study of Strongly Correlated Electrons in Nanoscopic and Bulk Forms,”

G. Alvarez, K. A. Al-Hassanieh, F. Popescu, C. Sen, A. Moreo, and E. Dagotto **J. Phys.: Condens. Matter** **19**, 125213 (2007). (Topical review).