Quench dynamics in interacting and superconducting nanojunctions

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Doctoral thesis submitted for the degree of
Doctor of Philosophy

Universidad Autónoma de Madrid

Departamento de Física Teórica de la Materia Condensada

Condensed Matter Physics Center (IFIMAC)

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Madrid, junio de 2018

*Nothing in this world is to be feared,
it is only to be understood.*

Marie Curie

Agradecimientos

Resumir los agradecimientos a las personas que me han ayudado y apoyado a lo largo del largo camino que es una tesis doctoral constituye una ardua tarea. Sin duda han sido muchas personas las que han hecho que, en mayor o menor medida, esta tesis llegue a buen puerto. Por ello pido disculpas anticipadas a todos aquellos a los que puedo olvidar mencionar en estas páginas.

En primer lugar me gustaría agradecer a mis padres científicos, Alfredo Levy y Álvaro Martín por su guía, su apoyo y, sobre todo, por su paciencia. Con ellos no solamente he aprendido las habilidades necesarias para finalizar con éxito esta tesis, sino que también me han transmitido su pasión por el conocimiento. De mi tiempo bajo su supervisión me llevo importantes lecciones a cerca de la constancia, honestidad y rigurosidad, entre otros temas.

I would like to mention specially the support from our collaborators during this thesis. Firstly, I would like to thank prof. Carmina Monreal, who was really involved on my learning during the earliest stages. From her I have learned a lot of physics and many of the tools used along the thesis. I am also grateful to Dr. Rémi Avriller, who was my hosting researcher during my research stay in Bordeaux. From him I have learned to be always precise and to do thorough works, essential skills to be a good scientist.

I am also grateful to profs. Cristian Urbina, Rémi Avriller, Ramón Aguado, Carlos Tejedor and Juan Carlos Cuevas for agreeing to take part in the defense of this thesis as jury members. I thank also Francisco Rodríguez-Adame and Pablo San-José for their availability as reserve members of this jury. I am also in debt with profs. Reinhold Egger and Marcelo Goffman for their fast reports for obtaining the doctorate international mention.

Me gustaría agradecer también a todos aquellos que, fuera de mi entorno científico, han colaborado en el buen desarrollo de mi tesis doctoral. En primer lugar me gustaría mostrar mi gratitud con todos aquellos que colaboran a crear un muy buen ambiente en nuestro módulo. En este apartado entraría un listado casi interminable de gente que he ido conociendo a lo largo de los años en el departamento, motivo por el cual me tomo la licencia de no incluir explícitamente nombres. Sin embargo sí que me gustaría mencionar a algunas personas que han sido especialmente importantes para mí a lo largo de estos años. En primer lugar me gustaría agradecer mis compañeros de comida, con los que tanto he disfrutado de las sobremesas y que tanto me han apoyado a lo largo de la tesis. Me gustaría mencionar a Carlos (“loosing the control of my life”), Javi Galego (y su fino sentido del humor) y Álvaro (la persona con un conocimiento más interdisciplinar que conozco) y el inolvidable Rui-Qi. Asimismo, tampoco quería olvidarme de Paloma, Carlos Sánchez, Miguel, Raj y Ville que han contribuido a que mi estancia en el departamento fuera agradable. Especial mención merecen Laura y Stefan sin los que el camino de la tesis habría resultado más complicado. También me gustaría agradecer al grupo de biólogos que tan bien me han acogido y que tanto me han enseñado sobre biología: Filip, Hernán, Predes, Silvia, Rocío, María y Guille. I am also grateful to Gianluca, Anna, Rémi, Sergey, Doru and Jonathan, who made my stay research stay in Bordeaux a nice time.

Me gustaría reconocer especialmente el apoyo de mis sucesivos compañeros y amigos, que han ido compartiendo conmigo entorno de trabajo. Por orden de antigüedad, me gustaría comenzar agradeciendo a Javier del pino. En él encontré un amigo siempre dispuesto a escuchar y ayudar cualquiera que fuera el problema. También me gustaría agradecer a Diego, quien ha sabido estar a mi lado a pesar de todas las dificultades. Asimismo me gustaría mostrar mi gratitud hacia María, quien ha puesto la sensatez al despacho ayudando además a crear un ambiente increíble. También me gustaría agradecer a Sergio (el “canishe”), cuya alegría y sentido del humor es capaz de animar cualquier situación. Tampoco me puedo olvidar de Víctor, una persona de sonrisa imborrable (a pesar de las bromas que ha ido sufriendo a lo largo de los años) que ha contribuido al buen ambiente del despacho. Me gustaría agradecer además a Raúl por su amabilidad y por toda la ayuda técnica que me ha prestado sin la que el camino de la tesis habría sido más tortuoso. A Alberto, de quien espero que su ilusión por la ciencia perdure en el tiempo. Me gustaría concluir este breve repaso mencionando a una nueva incorporación de un viejo amigo: Guilherme. A él y a Laura les agradezco la experiencia que han sabido transmitirme a lo largo de estos años.

Tampoco quiero olvidarme de mis compañeros de la universidad Complutense, con los que comparto una intensa amistad de más de 10 años (¡y los que quedan!). Así, me gustaría mencionar a Leandro, Gallego, Laura, Irene, Ikki, Daniel (“barriles”), Vicky, Bravo, Escalan, Benjamón, Capote, Miguelón, Alberto (aeronáutico)... También me gustaría agradecer el apoyo de algunos compañeros doctorandos con los que compartí camino y, a los que guardo un enorme aprecio. Me gustaría en primer lugar reconocer la labor de Marta, quien ha ejercido de psicóloga y entrenadora a partes iguales. Tampoco me quería olvidar mencionar a Laura, Mónica, Gallego, Leandro, Irene y Rebe, quienes han convertido el campus de Cantoblanco en un lugar agradable al que considero mi segundo hogar.

Tampoco me puedo olvidar de mis compañeros de entrenamiento. De ellos, a parte de una buena amistad, me llevo lecciones de humildad y de constancia. En primer lugar me gustaría agradecer la paciencia de Diego (TAI), quien me lleva soportando durante casi 5 años. Asimismo, me gustaría agradecer a mis compañeros de triatlón Cantoblanco, con quienes me he divertido y he sufrido a partes iguales: Alex, Ramos, Eva, Manso, Jesús, Pedro, Marta Ernesto, Oscar y Curro. Tampoco me puedo olvidar de Alberto, Cris, Bea y Desi, que siempre son capaces de sacarte una sonrisa.

No me gustaría finalizar sin agradecer a las personas que llevan apoyándome toda la vida en el camino que he ido andando: mi familia. En primer lugar me gustaría agradecer a mis primos, que desde siempre tan bien me han tratado y tanto me han ayudado. Además, me gustaría agradecer a mis tíos todo su cariño a lo largo de estos años. Por supuesto no me puedo olvidar de mis abuelos, quienes siempre han estado a mi lado independientemente de las circunstancias. Mención especial merecen mis padres, quienes me han respaldado en todas y cada una de las decisiones que he ido tomando y sin los que este camino hubiera sido imposible. Tampoco me olvido de Tania, la canija de la casa, que siempre es capaz de sacar una sonrisa a los que la rodean. Me gustaría terminar esta sección agradeciendo a Sara, cuyo apoyo ha sido fundamental a lo largo de estos años.

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Abstract

English

This thesis is devoted to the analysis of the electronic transport properties through nanoscopic junctions. It is focused on the time response of the nanojunctions, analyzing not only the mean current but also its fluctuations, which contain valuable information on the underlying microscopic processes. In particular, the thesis analyzes in detail the coherent regime, where the quantum nature of the electrons manifests itself more clearly. This study can be of relevance in connection to the increasing demand of fabricating smaller and faster devices. One of the aims of this thesis, is to analyze the effects of electron interactions and superconducting correlations in the time evolution of the system.

The first part of the thesis is devoted to the analysis of the role of interactions in the transient evolution of nanoscopic devices coupled to normal metal electrodes. The system is modeled by a single electronic level coupled to fermionic reservoirs. The non-interacting regime is analyzed in detail, finding exact expressions for the charge and current cumulants. Importantly, these expressions describe the short time oscillating behavior of the high order charge cumulants measured experimentally. Situations involving localized electron-phonon and electron-electron interactions are also analyzed. To address these situations a self-consistent method in the time domain is developed and tested using perturbative approximations, providing accurate results in the weak to moderate coupling regime. For both interactions, it is found that correlation effects tend to destroy the charge bistable behavior predicted by mean-field approximations. Moreover, for addressing the polaronic regime of strong electron-phonon interaction, an analytic approximation is developed, finding that interactions tend to exponentially increase the system relaxation times.

The second part of the thesis is devoted to the analysis of the dynamics of super-

conducting nanojunctions. The transport properties of these devices are determined by multiple Andreev reflection processes which induce the appearance of subgap states named Andreev bound states. It is found that the system dynamics becomes frozen after their formation, estimated to appear after ~ 3 Andreev reflections, leading to a metastable non-equilibrium population. By analyzing the charge transfer statistics, the non-equilibrium population of the system many-body states can be inferred. It is also shown that, for a single channel nanojunction, the system state can be determined by the mean current and the shot noise, which constitutes a less invasive measurement than the one performed in some recent experiments consisting on coupling the junction to a bosonic mode. This last situation is also analyzed finding that for a well transmitted channel the probability of the system to get trapped in an odd state is ~ 0.5 even for a weakly coupled mode. This is in qualitative agreement with recent experimental observations. Finally, the analogy between the charge counting statistics and the theory of phase transitions in equilibrium statistical mechanics is also analyzed.

Castellano

Esta tesis está dedicada al análisis del transporte electrónico a través de uniones nanoscópicas. La tesis está centrada en la respuesta temporal de dichas uniones, estudiando no sólo la corriente promedio, sino también sus fluctuaciones que contienen información importante a cerca de los procesos microscópicos. En particular, la tesis se centra en el estudio del régimen coherente, donde los efectos cuánticos de los electrones se manifiestan más claramente. Este estudio es relevante en conexión con la creciente demanda de desarrollar componentes electrónicos más pequeños y rápidos. En la tesis se analizan los efectos de interacciones y correlaciones superconductoras.

La primera parte de la tesis está dedicada al estudio de los efectos de interacciones localizadas en el régimen transitorio de sistemas nanoscópicos acoplados a electrodos normales. Por simplicidad, el sistema es modelizado por un nivel electrónico acoplado a reservorios electrónicos. El caso no interactuante es estudiado en detalle, desarrollando expresiones exactas para describir los cumulantes de la carga y la corriente a través del sistema. Es importante remarcar que estas expresiones describen las oscilaciones universales a tiempos cortos en los cumulantes de orden alto, medidas experimentalmente. Además, se estudian situaciones que involucran interacciones electrón-fonón y electrón-electrón localizadas en el sistema. Para ello se desarrolla un método autoconsistente que, en combinación con aproximaciones perturbativas, proporciona resultados precisos en el régimen de acoplo débil e intermedio. Para ambos tipos de interacción, la correlación electrónica tiende a destruir la biestabilidad en la carga, predicha por las aproximaciones de campo medio. Además, el régimen polarónico para la interacción electrón-fonón es estudiado a través de una aproximación analítica desarrollada a lo largo de la tesis, encontrando que la interacción tiende a alargar exponencialmente los tiempos de relajación del sistema.

En la segunda parte de la tesis se analiza el caso superconductor. El transporte electrónico a través del sistema acoplado a electrodos superconductores está controlado por las reflexiones de Andreev que generan estados dentro del gap, llamados estados de Andreev. La aparición de dichos estados, que se produce para un número aproximado de ~ 3 de reflexiones de Andreev, induce una congelación en la dinámica del sistema que adquiere una población de no equilibrio a tiempos largos. Analizando la estadística de las cargas transferidas a través de la unión, el estado del sistema puede ser inferido. En el caso más simple de una unión con un solo canal, la corriente y el ruido pueden ser usados

para determinar el estado del sistema, lo que constituiría una medida menos invasiva con respecto a experimentos recientes en los que acoplan un modo bosónico al sistema. Esta última situación también es analizada en la tesis, encontrando que la probabilidad del sistema de permanecer atrapado en un estado con una cuasipartícula atrapada es ~ 0.5 incluso para un modo muy débilmente acoplado. Este resultado está en concordancia con las observaciones experimentales. Finalmente, la conexión entre la estadística de cargas transferidas y la teoría de transiciones de fase en equilibrio es también discutida.

List of acronyms

This is a list of the acronyms used in the text (in alphabetical order).

ABS	andreev Bound state
CGF	Cumulant Generating function
DTA	Dressed Tunneling Approximation
DYLZ	Dynamical Yang-Lee zero
FCS	Full counting statistics
FGF	Factorial generating function
FSR	Friedel sum rule
GF	Generating function
HF	Hartree Fock
MAR	Multiple Andreev reflections
MC	Monte Carlo
NRG	Numerical renormalization group
PTA	polaron tunneling approximation
QD	Quantum dot
QPC	Quantum point contact
RPA	Random phase approximation
RWA	Rotating wave approximation
SPA	Single particle approximation
WTD	Waiting time distribution

1 General introduction

The main topic of this thesis is the electron transport dynamics through nanoscale systems, paying special attention to the role of electron interactions and superconducting correlations. In order to better understand the transport mechanisms through the system, I make use of the full counting statistics (FCS) analysis, which provides a complete information about the charge and current distributions. The interest of the thesis can be summarized on the following two questions: *What is the effect of interactions on the electron transport dynamics?* and *What kind of information can be obtained from a charge fluctuations analysis?*

In 1975 Gordon Moore, Intel cofounder, observed that the transistors were shrinking so fast that every year its density onto a silicon chip was doubled. The Moore's law has described the evolution of the silicon based electronics for almost half a century. Although the semiconducting devices are relatively small (7-10 nanometers), they are still bigger than the electron mean free path: the electrons suffer several elastic and inelastic collisions in the transport process, losing their phase coherence. In this regime, the electron transport can be described by the classical Ohm's law, which states that the conductance is proportional to the device section (S) and inversely proportional to its length (L),

$$G = \sigma \frac{S}{L},$$

where σ is the material conductivity.

However, if the Moore's law continues to be valid, it predicts that the industry will reach systems of the order of the electron mean free path in the next decade. These nanoscopic systems are usually also referred to as *mesoscopic*, from the Greek prefix

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“meso” which means “in between” the macroscopic and microscopic (atomic) world. In samples smaller than the mean free path, the electrons are not scattered in the device, reaching the so-called *ballistic* transport regime. In this regime, the wave nature of the electrons manifests itself and a quantum treatment of the system becomes necessary. Advances in nanofabrication enabled to reach this regime in different systems. One example which is widely used is the two dimensional electron gas created at the interface of GaAs and AlGaAs. These systems exhibit a relatively large mean free path, allowing to study different geometries such as small constrictions known as quantum point contacts (QPCs) [1, 2] or small electron boxes, known as quantum dots (QDs) [3]. In metallic systems, where the electron coherence length is much shorter, the ballistic regime can be achieved for small contacts or atomic wires between the two electrodes [4]. Another important examples are the so-called molecular junctions, where single molecules are trapped in a small gap created in between two metallic electrodes. The phenomenology of these systems is very rich, since the transport properties depend on the electronic structure of the molecule and internal degrees of freedom [5].

The size of a device is not the only important aspect to characterize its performance. It is also fundamental to know its time response: A faster device is able to make more operations in the same time. During the past decades, the operation frequency (commonly known as cutoff frequency) of the semiconducting-based components has been increased exponentially with time (about a factor 10 every 10 years). However, in the middle of the 2000s decade, the cutoff frequency has saturated to few GHz, indicating the limitations of the silicon based technology. In this context, nanodevices can be a way of overpass these limitations. There has been a lot of effort on realizing fast nanostructures operating in the range of hundreds of GHz [6] or even at a few THz [7]. This may constitute an improvement of several orders of magnitude with respect to the traditional devices.

Another important aspect is the energy efficiency of the devices: It is predicted that, in the 2030s decade, the energy consumption of computing will reach the actual world energy production [8]. Bennet and Landauer showed in Ref. [9] that the lower limit for performing an irreversible computation (usually also referred as switch) is $k_B T \ln 2$ (approximately $10^{-21} J$ at room temperature), about 7 orders of magnitude smaller than the actual energy consumption. Nanodevices could be a way to approach this limit [10, 11].

1.1 Theory of the quantum transport

The basic theory of quantum transport through coherent nanoscopic devices in the absence of interaction have been developed by R. Landauer and M. Büttiker [12, 13]. In their theory the electron phase coherence is preserved when moving inside the system. However, it considers that the phase coherence is broken by the environment. The theory treats the electrons from a macroscopic electrode in thermal equilibrium as incoming waves which are scattered by the system (reason why it is usually also referred as scattering theory). The different incoming electrons have probabilities τ_n to be transmitted through the system (and $1 - \tau_n$ to be reflected back). τ_n is usually known as the channel transmission coefficient. The system conductance is described by the Landauer formula [14], given by

$$G = G_0 \sum_{n=1}^N \tau_n, \quad (1.1)$$

where the sum is performed over the all the possible conduction channels and $G_0 = e^2/h$ is the quantum of conductance (an additional 2 factor is usually included to account for the spin electron degeneracy). The Landauer expression states that every totally open channel contributes to the conductance by G_0 . As an illustration of the validity of Eq. (1.1), the conductance of a QPC exhibits steps of amplitude G_0 with the gate voltage when a new channel is opened [1, 2].

A generalization of the Landauer formula to systems which exhibit local interactions between electrons in the system was developed by Meir and Wingreen [15]. This expression states that the current through a single channel system is given by

$$I = \frac{e}{\hbar} \int d\epsilon \sum_{\sigma} [f_L(\epsilon) - f_R(\epsilon)] \Gamma_{\sigma}(\epsilon) A_{\sigma}(\epsilon), \quad (1.2)$$

where Γ_{σ} describes the tunneling probability to the electrodes (σ being the electron spin), $f_{L(R)}$ are the electrodes density of states, $A_{\sigma}(\epsilon)$ is the density of states of the system coupled to the leads and the integration is performed in energy (ϵ). In the simple wideband approximation, the density of states of the electrodes are described by Fermi-Dirac distributions, the coupling to the electrodes can be considered energy independent and Eq. (1.2) is transformed to a integral of the density of states in the voltage window at zero temperature.

$$I_{T=0} = \frac{e}{\hbar} \Gamma \int_{\mu_R}^{\mu_L} d\epsilon A_{\sigma}(\epsilon), \quad (1.3)$$

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where $\mu_{L,R}$ are the electrodes chemical potentials of each electrode. As an illustration of the validity of Eq. (1.2), the current through a QD, which have localized energy levels, exhibits steps when a new state enters in the voltage window.

1.2 Superconductivity at the nanoscale

Superconductors are materials that undergo a phase transition at low temperatures. Below this critical temperature, the material exhibits a zero resistance and behaves as an ideal diamagnet. This sensitivity to small magnetic fields has been exploited for several applications ranging from medical to geophysical detectors. The first observation of superconductivity was performed in the laboratory of Kamerling Onnes in 1911. However, the first microscopical theory was developed around 40 years later by J. Bardeen, L. Cooper and J. Schieffer. In this theory (known as BCS), the electrons suffer an attractive interaction mediated by the lattice deformation. When this interaction overcomes the strength of the Coulomb repulsion, the electrons with opposite spins and momenta group into pairs (Cooper pairs). The order parameter of the superconducting phase is a complex number, with a modulus Δ , which is the value of the energy gap opened at the Fermi edge, and a phase ψ (superconducting phase). Along the thesis I will restricted myself to the conventional BCS superconductors, without considering situations involving more exotic pairing mechanisms, like the high temperature superconductors.

One important part of this thesis is devoted to the research field called *mesoscopic superconductivity* where superconductors are coupled to regions with spatial dimensions smaller than the superconducting coherence length. This field has recently experienced a huge activity in relation to the advent of the topological materials and, in particular in relation to the detection of Majorana states at the end of a one dimensional topological superconductor [16, 17].

The electronic transport through these regions is dominated by the Andreev reflections: An incoming electron can generate a hole reflected back with opposite momentum, leading to the transfer of a Cooper pair. The multiple Andreev reflections (MAR) generate the so-called Andreev bound states (ABSs) localized at the interface. In the simplest case of a QPC, two ABSs per channel appears at the interface of the two superconductors with energies given by [18]

$$\epsilon_A(\phi) = \pm \Delta \sqrt{1 - \tau \sin(\phi/2)}, \quad (1.4)$$

where $\phi = \psi_L - \psi_R$ is the phase difference between the left and right electrodes. For an arbitrary phase difference the ABSs lead to a non-dissipative current, known as Josephson current, whose value [18] is determined by

$$I_s(\phi) = \frac{e\Delta}{2\hbar} \sum_n \frac{\tau_n \sin(\phi)}{\sqrt{1 - \tau_n \sin(\phi/2)}}. \quad (1.5)$$

For a channel with a high transmission factor, the maximum current, known as critical current (I_c), is achieved at $\phi \approx \pi$. In this regime the expression of the the current simplifies as $I_s(\phi) = I_c \sin(\phi/2)$. It is important to remark that, in a QPC the only contribution to the current is provided by the ABSs. This is not in general the case when studying more complex hybrid structures. For instance, in a QD coupled to superconducting electrodes (a system that will be analyzed along the thesis), quasiparticle excitations provide an additional contribution to the current, with opposite sign to I_s (for a review see Ref. [19]).

Up to this point, I have not commented the role of quasiparticle excitations in the system. It has been experimentally measured [20] and theoretically studied [21–23] that the number of quasiparticles in superconducting nanostructures largely exceeds the expected equilibrium value. These quasiparticles undermine the coherence in the system, setting a limit to the proper functioning of the device. Eventually, they can decay to the ABSs, producing a non-equilibrium population of these states (known as *quasiparticle poisoning*). The population of the ABSs has been measured in some recent experiments in QPCs [24] and nanowires [25], driven by the proposal of using the ABSs as a base for the quantum technologies [26, 27]. In these experiments, the authors demonstrated the possibility of coherently manipulate their occupation by the use of external radiation. However, they also found an almost constant probability of finding the system trapped in a state of opposite parity with respect to the ground state (refereed to as odd state) of $P_{odd} \approx 0.5$.

When a constant finite voltage is applied to the superconducting junction, the phase difference between the two superconductors increases linearly in time as $\phi(t) = (2eV/\hbar)t$, leading to a current which oscillates in time. For voltages smaller than the superconducting gap, the ABSs are able to follow adiabatically the position given by Eq. (1.4), exchanging charge between them with a probability described by a Landau-Zener transition, as described in Ref. [28]. In this situation the transport is dominated by multiple Andreev reflections: processes involving the transfer of more than two electrons at once. For higher voltages, $eV \gtrsim \Delta$, the picture becomes more complex, since the ABSs cannot

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follow the expected position given by Eq. (1.4). In this regime quasiparticles are created in the junction and the system exhibits dissipation.

1.3 Interactions at the nanoscale

Mesoscopic and nanoscopic devices constitute a controlled playground to understand the fundamental processes that give rise to the transport phenomenon. In these spatially confined systems, the electrons are specially sensitive to the interactions with the rest of the electrons and the localized mechanical modes. The importance of the interactions has already been pointed out in the 1930s decade by the observation that, small amount of localized magnetic impurities give raise to an increase in the resistance when decreasing the temperature [29]. This effect, produced by the interactions between the itinerant electrons and the localized (d or f) ones, was explained by Kondo in the 1960s. At the nanoscale the Kondo effect has been observed in several nanoscale devices such as QPCs, where it has been demonstrated to be responsible of the 0.7 anomaly in the conductance steps in QPCs [30], QDs [31, 32], nanotubes [33, 34] or single molecules [35].

The minimal model describing the phenomenology of this interaction is the so-called Anderson model [36], which considers a spin dependent level coupled to electron reservoirs including an on-site electrostatic repulsion term between electrons, which penalizes the double occupancy of the level. The main parameter in the model is the Kondo temperature (T_K), which sets the frontier between the Kondo ($T < T_K$) and the Coulomb blockade ($T > T_K$) regimes. If $T < T_K$ and the Coulomb repulsion is larger than the coupling to the electronic reservoir the charge becomes frozen in the impurity, exhibiting large spin fluctuations. In this regime the model predicts an increasing in the resistance and the creation of a new state at the Fermi level with an exponentially decreasing energy width with increasing interaction strength.

Another kind of interaction that will be analyzed in this thesis is the localized electron-phonon interaction, important since many nanoscale systems are flexible. There several systems which can exhibit this kind of interaction like small molecular junctions [37], suspended nanowires [38, 39] or carbon nanotubes [40]. One of the simplest model which captures the system phenomenology is the spinless Anderson-Holstein model [41], which considers a localized phonon mode linearly coupled to the electrons in the nanoscopic device (described by a spinless electronic level). For a weak electron-phonon coupling

strength the correction in the current exhibits a crossover from negative to positive values when moving from high to low transmission coefficients [39] (the transition happens around half transmission, i.e. $\tau = 0.5$). In the strong electron-phonon coupling, the interaction leads to a suppression of the current at low voltage, known as Frank-Condon blockade. This suppression can be related to the creation of a mixed quasiparticle formed by electrons and phonons, known as polaron, which reduces the mobility of the electrons in the nanostructure. In this regime the transport through the junction occurs in avalanches: many electrons are transferred in a short period of times, followed by a long waiting time between events [42, 43]. This transport mechanism leads to huge shot noise values. In this regime, when the bias voltage is increased, the electrons have energy enough to excite the phonons, opening new conduction channels and leading to the appearance of steps in the current when the the voltage is an even multiple of the phonon frequency [44].

Finally, it is also worth commenting the situation where both, electron-electron and electron-phonon interactions are present. In this situation two regimes can be distinguish depending on the strongest interaction: the Kondo and the phonon dominated regimes. The first regime is characterized by the presence of the Kondo peak at the Fermi edge, which is slightly broadened due to the electron-phonon interaction. In the opposite regime, the Kondo peak is exponentially suppressed, leading to an insulating state [45].

1.4 Time dependent transport

The main part of this thesis is devoted to the study of the time evolution of mesoscopic systems. This analysis provides a more complete information about the electron transport than the conventional stationary one. For instance, information about the typical electron time scales, relaxation time scales or transport mechanisms can be obtained from a time dependent analysis. The first works analyzing the time dependent evolution of this kind of systems were focused on the dynamics of non-interacting systems [46]. In the simplest situation of a non-interacting single level, the electron reservoirs act as baths which make the system relax to the steady state in a time scale controlled by the coupling between the system and the leads [47, 48].

A particularly interesting example is described in Ref. [49], where Levitov and cowork-

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ers analyzed the transport properties of a nanojunction under different time-dependent voltage profiles. In particular, they demonstrated that some Lorentzian pulses minimize the shot noise. These pulses can generate single electron excitations of the Fermi sea (usually known as levitons), without generating holes, which have been recently measured in a two dimensional electron gas [50]. The levitons, together with other advances in condensed matter like the quantum hall effect, have led to the birth of the electron quantum optics field whose purpose is to transfer discoveries from the quantum optics field to the case of ballistic conductors (for a recent review see [51]).

1.4.1 Dynamics of interacting nanojunctions

The situation in the presence of interactions becomes richer and much more complex from a theoretical point view, since even the simple toy models are usually not solvable. There is a large number of works addressing the dynamics of interacting nanojunctions using different techniques like Monte Carlo (MC) [44, 52, 53], time dependent numerical renormalization group (NRG) [54, 55] or diagrammatic techniques [56], among others. Very often interactions increase the system relaxation times. A particular interesting problem that will be treated along the thesis is the dynamics of a system exhibiting electron-phonon interaction. In this situation it is still controversial whether the system exhibits a bi-stable behavior (long time trapping of the system in two different configurations depending on the initial state) or not. There is a series of works discussing this possibility [56–62], looking to different observables and for different parameter regimes.

Much less analyzed is the dynamics of a superconducting nanojunction. This case is important for studying the mechanisms and time scales of quasiparticle trapping and relaxation. In an equilibrium situation it has been observed that, under sudden changes in the system such as voltage pulses, the system current is not able to relax to the expected stationary value [63]. In contrast, if a bias voltage is applied to the device, the current converges to the steady state in times of the order of the inverse of the voltage [64, 65]. What is missing in these works is the analysis of the system state evolution, which provides information about the quasiparticle poisoning and the possible relaxation mechanisms.

1.5 Current fluctuations

While traditionally most of the works are focused on the current or the conductance (which reflects the transmission probability), the noise reveals fundamental aspects of the physical mechanisms. The best known examples are the fluctuation-dissipation relations, where the fluctuations of a system can be used to characterize its response to external fields. The first experimental application was performed by Perrin, who used the motion of a particle in a liquid to measure the Avogadro number [66]. These relations have been later used to determine a large variety of response functions such as the resistance of a device or the magnetic susceptibility. The importance of the system fluctuations is illustrated by Ladauer's famous quotation: *The noise is the signal*.

In mesoscopic devices in an equilibrium situation ($V = 0$), the system exhibits thermal fluctuations also known as Johnson-Nyquist noise, due to the particle agitation. The expression of this kind of noise, $S_{th} = 4k_B T/R$, provides information about the device resistance R (T is the temperature and k_B the Boltzmann constant). In the non-equilibrium situation ($V \neq 0$), the incoming electrons can be either transmitted or reflected back. These two processes lead to current fluctuations, usually known as *shot noise*. The theory of the shot noise for uncorrelated electrons (when the transfer events are independent stochastic processes) was developed by Schottky in 1918. In this regime, the transfer distribution follows a Poissonian distribution, leading to a shot noise with a form $S_{shot} = 2e \langle I \rangle$. It is important to note the fundamental difference between the thermal and the shot noise, since they provide information about the system in equilibrium and non-equilibrium situations, respectively. It is usually useful to define the Fano factor, as $F = S/2e \langle I \rangle$, which provides information about the effective carriers charge in units of the electron charge, e . If $F > 1$ (super-poissonian distribution), some transfer events occur at very close times leading to the bunch phenomenon. In the opposite situation, when $F < 1$ (sub-poissonian distribution), the events are well separated on time and the reflection probability is reduced. Although there are additional sources of noise like the telegraph noise and the $1/f$ noise, they will be not considered along the thesis.

The shot noise provides the demonstration of a large variety of phenomena based on charge, spin, coherence and many-body correlations of electrons. As a valuable example, the shot noise provides evidence of the $1/3$ fractional charge (in units of the electron charge) of the quasiparticle carriers in the fractional quantum Hall effect [67, 68], im-

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possible to detect from the conductance measurements. In a similar way, the shot noise reveals an effective charge of 2 for the carriers in normal-superconducting junctions, reflecting the transference of two electrons at a time [69]. Another interesting example is the case of low transmitting superconducting nanojunctions with a subgap voltage where multiple charges are transfer due to the MAR processes [70]. In a general case, the situation can be more complex exhibiting a richer phenomenology, going from an almost zero Fano factor for perfect transmitting channels (for example in a QPC) to huge values (for example in the electron avalanches [42]).

1.5.1 Full counting statistics

The idea of FCS was originally developed in the quantum optics field, to characterize the state of the light through the probability distribution of the photon absorbed in a detector in a time interval. In the mesoscopic transport field, the main object of the FCS studies are the probability distribution of the number of transfer electrons in a time interval. The detection of these probabilities become involved since there is not an “electron counter”, and they have to be accessed through indirect measurements. As an example, in Refs. [71–74] a QPC is capacitively coupled to the system (usually a QD). Using the current through the QPC, they are able to determine the state of the QD, inferring from it the tunnel events. By performing several independent measurements, the probabilities, $P_n(t)$, of n electrons transfer after a measurement time t can be obtained. It is important to note that, this kind of scheme is only working under rather particular conditions, known as sequential regime. In this regime the electrons are transferred in an uni-directional way, spending a time long enough in the system to be detected. It is still unclear how to address experimentally more general situations beyond this regime. Despite these difficulties, the FCS turns to be a formidable tool to understand the fundamentals of the electron transport in the nanoscale. For instance, it has been proposed as a tool to detect interactions [75, 76] or phase transitions [77, 78].

From the single electron probabilities the generating function (GF) can be defined as

$$Z(\chi, t) = \sum_n P_n(t) e^{in\chi}, \quad (1.6)$$

where the sum is performed over the possible number of electrons transfer in a time interval t . The variable χ is the so-called counting field: a fictitious field used to count the number of electrons transfer. This field simulates the coupling to an ideal detector

Figure 1.1: Left: representation of how a current measurement might look like. In the right panel I show the current probability distribution. The cumulants, $\langle\langle I^n \rangle\rangle$ fully characterizes the curve: The first cumulant is the mean value (mean current), the second cumulant is the width of the distribution (shot noise), the third measures the asymmetry of the curve (Skewness),...

which provides instantaneous projective measurements without influencing the the system dynamics. From the GF, the charge cumulants of the distribution, $\langle\langle q^n(t) \rangle\rangle$, can be obtained by successive derivatives, as

$$\langle\langle q^n(t) \rangle\rangle = \left. \frac{\partial}{\partial i\chi} \log [Z(\chi, t)] \right|_{\chi=0}. \quad (1.7)$$

The corresponding current cumulants, $\langle\langle I^n(t) \rangle\rangle$, can be obtained as time derivative of Eq. (1.7). In the left panel of Fig. 1.1 I show schematically a typical current measurement in time domain. By performing statistics on the time fluctuations of the current, one can arrive to the current distribution shown in the right panel of Fig. 1.1. The first cumulant (mean current) provides the mean value of the distribution, the second cumulant (shot noise) its width, the third cumulant (skewness) measures its asymmetry, and so on. An important part of this thesis is devoted to the study of the high order charge and current cumulants, analyzing the information they provide.

At the nanoscale, the simplest example of a DC current through a two terminal non-interacting junction was analyzed by Levitov in middle of the 1990s decade. The long time statistics of the transfer charges are dependent on a single parameter, which is the energy-dependent transmission probability, $\tau(\epsilon)$. In this situation, the GF has a compact

Figure 1.2: Left: schematic representation of an open system of volume V surrounded by particles. In the top right panel I show an scheme of a possible distribution of Yang-Lee zeros in the complex fugacity plane, y . In the bottom right panel I show schematically how the particle density (first derivative of the partition function, Z) might look like, exhibiting two phase transition points where the Yang-lee zeros approach the real fugacity axis (marked with stars in the upper panel).

expression [79]

$$\begin{aligned} \log [Z(\chi, t)] &= \frac{t}{2\pi} \int d\epsilon \log \left\{ 1 + \tau(\epsilon) \left[(e^{i\chi} - 1) f_L(\epsilon) (1 - f_R(\epsilon)) \right. \right. \\ &\quad \left. \left. + (e^{-i\chi} - 1) (1 - f_L(\epsilon)) f_R(\epsilon) \right] \right\}. \end{aligned} \quad (1.8)$$

In the presence of interactions, the general expression of the GF remains unknown, although some progress have been done for calculating the current cumulants [80].

1.5.2 Analogy to equilibrium statistical mechanics: Yang-Lee zeros

In equilibrium statistical mechanics, all the physical properties of an open system are fully determined by the partition function. Following the example in Ref. [81] for a monoatomic gas (see left panel of Fig. 1.2), the partition function can be written as

$$Z(y, V) = \sum_{n=0}^N \frac{Q_n}{n!} y^n, \quad (1.9)$$

where the sum is performed over the number of particles in the system volume V . In this expression y is the fugacity and $Q_n(V)/n!$ is the probability of finding n particles

in the system. At the beginning of the 1950s decade, Yang and Lee realized that, in the thermodynamical limit, the zeros of Eq. (1.9) form branches in the complex fugacity plane. These branches can eventually define regions in the plane (marked by the colors in the top right panel of Fig. 1.2), which can be related to the different phases. The points where the branches approach the real axis are the phase transition points (marked with stars), where two or more phases coexist. The order of the phase transition could be inferred from the density of zeros and the angle with which the branches approach the real axis [82]. At the coexistence points, the density of each phase can be determined by the density of zeros close to the real axis. The Yang-Lee zeros have been used to analyze the phase transition phenomena in a large variety of systems, such as the Ising model [83] or van der Waals gases [84] (for a review see [85]). In the lower part of the right panel of Fig. 1.2 the particle number (first derivative of the partition function) is depicted, which exhibits jumps at the phase transition points, signature of a first order phase transition. Although for the non-equilibrium situation an equivalent general theory is still lacking, some recent works have demonstrated that the original Yang-Lee theory holds, at least for some systems [82, 86]. In this theory, the equivalent quantity to the equilibrium partition function is the sum of the steady state weights (also known as un-normalized probabilities).

Following the extensions for the out of equilibrium situation, the function that plays the role of the partition function (1.9) in the FCS theory is the counting field dependent GF (2.96), which has its same functional form. In this analogy, the role of the extensive variable of the volume is played by the measurement time, the complex exponential of the counting field ($z = e^{i\chi}$) plays the role of the fugacity and the number of particles in the system the number of electrons transfer. Following the reasoning of Yang and Lee for equilibrium statistical mechanics, the zeros of the GF, named as dynamical Yang-Lee zeros (DYLZs) in analogy to the equilibrium situation, can also define regions in the complex counting field plane, related to the non-equilibrium phases. The DYLZs have already been analyzed for non-interacting electrons flowing between two normal electrodes in a series of works [87, 88]. In these systems, where the GF has the Levitov form (1.8), the zeros are located in the real negative z -axis, suggesting that singularities off the negative real axis would characterize electronic correlations. Deviations from this behavior have been observed in mesoscopic systems involving superconductivity [89], electron-electron [75] and electron-phonon interactions [90].

1.6 Outline

This thesis is divided into eight chapters, including this introduction to the field. While the second is devoted to introduce the basic theoretical formalism, the main results of the thesis are discussed in the chapters 3-7, which are organized into two groups. In the first one, composed by the chapters 3-5, the quantum transport through nanoscopic system coupled to normal metallic electrodes is discussed.

In chapter 3 the non-interacting situation is analyzed, paying special attention to the charge transfer and current statistics. In this chapter, analytic expressions relating the charge cumulants and the roots of the GF are developed, which explain the short time oscillatory behavior observed in the high order cumulants. The role of the electron coherence and directionality is also discussed. In chapter 4 the transient transport properties through a molecular junction in presence of localized electron-phonon interaction in the system are analyzed. This chapter is focused on the polaronic regime, characterized by a strong coupling between electrons and phonons, making use of the dressed tunneling approximation developed during the thesis and in quantitative agreement with the exact numerical calculations. Special attention is paid to the high order cumulants and to the conductance and differential noise evolution, which contain clear signatures of the interaction. In 5 an efficient algorithm to perform self-consistent calculations in the time domain is discussed. As an illustration of the accuracy of the method, the electron-electron and electron-phonon interactions are analyzed by means of perturbative self-consistent approximations, leading to a remarkable agreement with exact numerical results for a wide range of parameters ranging from weak to rather strong interactions. In both cases it is found that the electron correlation effects tend to destroy the charge bistable behavior predicted by the mean-field approach.

The second part of the thesis is devoted to the analysis of the transient evolution of superconducting nanojunctions. In chapter 6 the single electron properties, population, current and density of states are studied. Special attention is paid to the formation of the Andreev bound states. In this chapter both situations, the phase and voltage biased situations are analyzed. In chapter 7 the charge statistics of superconducting nanojunctions is analyzed, showing that it contains valuable information about the system state. Moreover, the relation between the theory of equilibrium phase transitions in statistical mechanics is related to the system GF. The possibility of using voltage pulses to

coherently control the quantum state of the system is also discussed. Furthermore, the situation of a superconducting nanojunction coupled to a bosonic mode is also explored.

In chapter 8 the main conclusions of the thesis are summarized, analyzing the open question and possible future research lines. In addition, some appendix are included to discuss numerical and analytic details. Appendix A is used to introduce the NRG technique implemented to study the electron-phonon interaction. The discarded states method is also discussed, which is used to obtain the finite frequency properties. In appendix A.2 the Toeplitz theory is introduced, which provides the link between the time dependent formalism and the asymptotic limit. Appendix B is used to discuss the mathematical details about the calculation of the inverse phonon propagator developed during the thesis. Appendix C introduces a simplified rate equation to understand the dynamics of a superconducting nanojunction. Finally, appendix D is used to discuss the numerical calculation details of the bidirectional Poisson distribution.

Bibliography

- [1] B. J. van Wees, H. van Houten, C. W. J. Beenakker, J. G. Williamson, L. P. Kouwenhoven, D. van der Marel, and C. T. Foxon. Quantized conductance of point contacts in a two-dimensional electron gas. *Phys. Rev. Lett.* **60**, 848 (1988).
- [2] D. A. Wharam, T. J. Thornton, R. Newbury, M. Pepper, H. Ahmed, J. E. F. Frost, D. G. Hasko, D. C. Peacock, D. A. Ritchie, and G. A. C. Jones. One-dimensional transport and the quantisation of the ballistic resistance. *Journal of Physics C: Solid State Physics* **21**, L209 (1988).
- [3] L. P. Kouwenhoven, C. M. Marcus, P. L. McEuen, S. Tarucha, R. M. Westervelt, and N. S. Wingreen. *Electron Transport in Quantum Dots* (Springer Netherlands, Dordrecht, 1997).
- [4] N. Agrait, A. L. Yeyati, and J. M. van Ruitenbeek. Quantum properties of atomic-sized conductors. *Physics Reports* **377**, 81 (2003).
- [5] J. C. Cuevas and E. Scheer. *Molecular electronics: an introduction to theory and experiment*. World scientific series in nanoscience and nanotechnology (New Jersey: World Scientific, 2010).
- [6] Y.-C. Liao, Leiand Lin, M. Bao, R. Cheng, Y. Q. Y. Bai, Jingweiand Liu, K. L. Wang,

BIBLIOGRAPHY

- Y. Huang, and X. Duan. High-speed graphene transistors with a self-aligned nanowire gate. *Nature* **467**, 305 EP (2010).
- [7] L. C. Castro, D. L. John, D. L. Pulfrey, M. Pourfath, A. Gehring, and H. Kosina. Method for predicting fT for carbon nanotube FETs. *IEEE Transactions on Nanotechnology* **4**, 699 (2005).
- [8] Rebooting the it revolution: A call to action. Semiconducting industry association (2015).
- [9] R. Landauer. Irreversibility and heat generation in the computing process. *IBM Journal of Research and Development* **5**, 183 (1961).
- [10] M. Lopez-Suarez, I. Neri, and L. Gammaitoni. Sub- $k_b t$ micro-electromechanical irreversible logic gate. *Nat. Commun.* **7**, 12068.
- [11] J. Hong, B. Lambson, S. Dhuey, and J. Bokor. Experimental test of Landauer’s principle in single-bit operations on nanomagnetic memory bits. *Science Advances* **2** (2016).
- [12] R. Landauer. Spatial variation of currents and fields due to localized scatterers in metallic conduction. *IBM Journal of Research and Development* **32**, 306 (1988).
- [13] M. Buttiker. Symmetry of electrical conduction. *IBM Journal of Research and Development* **32**, 317 (1988).
- [14] R. Landauer. Spatial variation of currents and fields due to localized scatterers in metallic conduction. *IBM Journal of Research and Development* **1**, 223 (1957).
- [15] Y. Meir and N. S. Wingreen. Landauer formula for the current through an interacting electron region. *Phys. Rev. Lett.* **68**, 2512 (1992).
- [16] J. Alicea. New directions in the pursuit of Majorana fermions in solid state systems. *Reports on Progress in Physics* **75**, 076501 (2012).
- [17] C. W. J. Beenakker. Random-matrix theory of Majorana fermions and topological superconductors. *Rev. Mod. Phys.* **87**, 1037 (2015).
- [18] Y. Nazarov and Y. Blanter. *Quantum Transport: Introduction to Nanoscience* (Cambridge University Press, 2009).
- [19] A. Martín-Rodero and A. L. Yeyati. Josephson and Andreev transport through quantum dots. *Advances in Physics* **60**, 899 (2011).

- [20] M. Zgirski, L. Bretheau, Q. Le Masne, H. Pothier, D. Esteve, and C. Urbina. Evidence for long-lived quasiparticles trapped in superconducting point contacts. *Phys. Rev. Lett.* **106**, 257003 (2011).
- [21] J. Aumentado, M. W. Keller, J. M. Martinis, and M. H. Devoret. Nonequilibrium quasiparticles and $2e$ periodicity in single-Cooper-pair transistors. *Phys. Rev. Lett.* **92**, 066802 (2004).
- [22] D. G. Olivares, A. L. Yeyati, L. Bretheau, Ç. Ö. Girit, H. Pothier, and C. Urbina. Dynamics of quasiparticle trapping in Andreev levels. *Phys. Rev. B* **89**, 104504 (2014).
- [23] A. Zazunov, A. Brunetti, A. L. Yeyati, and R. Egger. Quasiparticle trapping, Andreev level population dynamics, and charge imbalance in superconducting weak links. *Phys. Rev. B* **90**, 104508 (2014).
- [24] C. Janvier, L. Tosi, L. Bretheau, Ç. Ö. Girit, M. Stern, P. Bertet, P. Joyez, D. Vion, D. Esteve, M. F. Goffman, H. Pothier, and C. Urbina. Coherent manipulation of Andreev states in superconducting atomic contacts. *Science* **349**, 1199 (2015).
- [25] M. Hays, G. de Lange, K. Serniak, D. J. van Woerkom, D. Bouman, P. Krogstrup, J. Nygård, A. Geresdi, and M. H. Devoret. Direct microwave measurement of Andreev-bound-state dynamics in a proximitized semiconducting nanowire. *arXiv:1711.01645* (2017).
- [26] A. Zazunov, V. S. Shumeiko, E. N. Bratus, J. Lantz, and G. Wendin. Andreev level qubit. *Phys. Rev. Lett.* **90**, 087003 (2003).
- [27] N. M. Chtchelkatchev and Y. V. Nazarov. Andreev quantum dots for spin manipulation. *Phys. Rev. Lett.* **90**, 226806 (2003).
- [28] A. L. Yeyati, A. Martín-Rodero, and E. Vecino. Nonequilibrium dynamics of Andreev states in the Kondo regime. *Phys. Rev. Lett.* **91**, 266802 (2003).
- [29] L. Kouwenhoven and L. Glazman. Revival of the Kondo effect. *Physics World* **14**, 33 (2001).
- [30] B. Brun, F. Martins, S. Faniel, B. Hackens, A. Cavanna, C. Ulysse, A. Ouerghi, U. Gennser, D. Mailly, P. Simon, S. Huant, V. Bayot, M. Sanquer, and H. Sellier. Electron phase shift at the zero-bias anomaly of quantum point contacts. *Phys. Rev. Lett.* **116**, 136801 (2016).
- [31] D. Goldhaber-Gordon, H. Shtrikman, D. Mahalu, D. Abusch-Magder, U. Meirav, and

BIBLIOGRAPHY

- M. A. Kastner. Kondo effect in a single-electron transistor. *Nature* **391**, 156 EP (1998).
- [32] S. M. Cronenwett, T. H. Oosterkamp, and L. P. Kouwenhoven. A tunable Kondo effect in quantum dots. *Science* **281**, 540 (1998).
- [33] J. Nygård, D. H. Cobden, and P. E. Lindelof. Kondo physics in carbon nanotubes. *Nature* **408**, 342 EP (2000).
- [34] P. Jarillo-Herrero, J. Kong, H. S. J. van der Zant, C. Dekker, L. P. Kouwenhoven, and S. De Franceschi. Orbital Kondo effect in carbon nanotubes. *Nature* **434**, 484 EP (2005).
- [35] W. Liang, M. P. Shores, M. Bockrath, J. R. Long, and H. Park. Kondo resonance in a single-molecule transistor. *Nature* **417**, 725 EP (2002).
- [36] P. W. Anderson. Localized magnetic states in metals. *Phys. Rev.* **124**, 41 (1961).
- [37] H. Park, J. Park, A. K. L. Lim, E. H. Anderson, A. P. Alivisatos, and P. L. McEuen. Nanomechanical oscillations in a single-C60 transistor. *Nature* **407**, 57 EP (2000).
- [38] N. Agraït, C. Untiedt, G. Rubio-Bollinger, and S. Vieira. Onset of energy dissipation in ballistic atomic wires. *Phys. Rev. Lett.* **88**, 216803 (2002).
- [39] L. de la Vega, A. Martín-Rodero, N. Agraït, and A. L. Yeyati. Universal features of electron-phonon interactions in atomic wires. *Phys. Rev. B* **73**, 075428 (2006).
- [40] R. Leturcq, C. Stampfer, K. Inderbitzin, L. Durrer, C. Hierold, E. Mariani, M. G. Schultz, F. von Oppen, and K. Ensslin. Franck-Condon blockade in suspended carbon nanotube quantum dots. *Nature Physics* **5**, 327 EP (2009).
- [41] T. Holstein. Studies of polaron motion: Part i. the molecular-crystal model. *Annals of Physics* **8**, 325 (1959).
- [42] J. Koch and F. von Oppen. Franck-Condon blockade and giant Fano factors in transport through single molecules. *Phys. Rev. Lett.* **94**, 206804 (2005).
- [43] C. S. Lau, H. Sadeghi, G. Rogers, S. Sangtarash, P. Dallas, K. Porfyrakis, J. Warner, C. J. Lambert, G. A. D. Briggs, and J. A. Mol. Redox-dependent Franck-Condon blockade and avalanche transport in a graphene-fullerene single-molecule transistor. *Nano Letters* **16**, 170 (2016).
- [44] L. Mühlbacher and E. Rabani. Real-time path integral approach to nonequilibrium many-body quantum systems. *Phys. Rev. Lett.* **100**, 176403 (2008).

- [45] A. C. Hewson and D. Meyer. Numerical renormalization group study of the Anderson-Holstein impurity model. *Journal of Physics: Condensed Matter* **14**, 427 (2002).
- [46] A. Blandin, A. Nourtier, and D. W. Hone. Localized time-dependent perturbations in metals: formalism and simple examples. *J. Phys. France* **37**, 369 (1976).
- [47] A.-P. Jauho, N. S. Wingreen, and Y. Meir. Time-dependent transport in interacting and noninteracting resonant-tunneling systems. *Phys. Rev. B* **50**, 5528 (1994).
- [48] T. L. Schmidt, P. Werner, L. Mühlbacher, and A. Komnik. Transient dynamics of the Anderson impurity model out of equilibrium. *Phys. Rev. B* **78**, 235110 (2008).
- [49] L. S. Levitov, H. Lee, and G. B. Lesovik. Electron counting statistics and coherent states of electric current. *Journal of Mathematical Physics* **37**, 4845 (1996).
- [50] J. Dubois, T. Jullien, F. Portier, P. Roche, A. Cavanna, Y. Jin, W. Wegscheider, P. Roulleau, and D. C. Glattli. Minimal-excitation states for electron quantum optics using levitons. *Nature* **502**, 659 EP (2013).
- [51] B. Roussel, C. Cabart, G. Fève, E. Thibierge, and P. Degiovanni. Electron quantum optics as quantum signal processing. *physica status solidi (b)* **254**, 1600621 (2017). 1600621.
- [52] E. Gull, D. R. Reichman, and A. J. Millis. Numerically exact long-time behavior of nonequilibrium quantum impurity models. *Phys. Rev. B* **84**, 085134 (2011).
- [53] G. Cohen, E. Gull, D. R. Reichman, A. J. Millis, and E. Rabani. Numerically exact long-time magnetization dynamics at the nonequilibrium Kondo crossover of the anderson impurity model. *Phys. Rev. B* **87**, 195108 (2013).
- [54] F. B. Anders and A. Schiller. Spin precession and real-time dynamics in the Kondo model: time-dependent numerical renormalization-group study. *Phys. Rev. B* **74**, 245113 (2006).
- [55] F. Güttge, F. B. Anders, U. Schollwöck, E. Eidelstein, and A. Schiller. Hybrid NRG-DMRG approach to real-time dynamics of quantum impurity systems. *Phys. Rev. B* **87**, 115115 (2013).
- [56] K. F. Albrecht, A. Martín-Rodero, R. C. Monreal, L. Mühlbacher, and A. Levy Yeyati. Long transient dynamics in the Anderson-Holstein model out of equilibrium. *Phys. Rev. B* **87**, 085127 (2013).

BIBLIOGRAPHY

- [57] A. S. Alexandrov, A. M. Bratkovsky, and R. S. Williams. Bistable tunneling current through a molecular quantum dot. *Phys. Rev. B* **67**, 075301 (2003).
- [58] M. Galperin, M. A. Ratner, and A. Nitzan. Hysteresis, switching, and negative differential resistance in molecular junctions: a polaron model. *Nano Letters* **5**, 125 (2005). PMID: 15792425.
- [59] R.-P. Riwar and T. L. Schmidt. Transient dynamics of a molecular quantum dot with a vibrational degree of freedom. *Phys. Rev. B* **80**, 125109 (2009).
- [60] P. D’Amico, D. A. Ryndyk, G. Cuniberti, and K. Richter. Charge-memory effect in a polaron model: equation-of-motion method for Green functions. *New Journal of Physics* **10**, 085002 (2008).
- [61] K. F. Albrecht, H. Wang, L. Mühlbacher, M. Thoss, and A. Komnik. Bistability signatures in nonequilibrium charge transport through molecular quantum dots. *Phys. Rev. B* **86**, 081412 (2012).
- [62] J. Klatt, L. Mühlbacher, and A. Komnik. Kondo effect and the fate of bistability in molecular quantum dots with strong electron-phonon coupling. *Phys. Rev. B* **91**, 155306 (2015).
- [63] G. Stefanucci, E. Perfetto, and M. Cini. Time-dependent quantum transport with superconducting leads: A discrete-basis Kohn-Sham formulation and propagation scheme. *Phys. Rev. B* **81**, 115446 (2010).
- [64] E. Perfetto, G. Stefanucci, and M. Cini. Equilibrium and time-dependent Josephson current in one-dimensional superconducting junctions. *Phys. Rev. B* **80**, 205408 (2009).
- [65] J. Weston and X. Waintal. Linear-scaling source-sink algorithm for simulating time resolved quantum transport and superconductivity. *Phys. Rev. B* **93**, 134506 (2016).
- [66] J. Perrin. *Brownian Movement and Molecular Reality* (Taylor and Francis, London., 1910).
- [67] R. de Picciotto, M. Reznikov, M. Heiblum, V. Umansky, G. Bunin, and D. Mahalu. Direct observation of a fractional charge. *Nature* **389**, 162 EP (1997).
- [68] L. Saminadayar, D. C. Glatthi, Y. Jin, and B. Etienne. Observation of the $e/3$ fractionally charged Laughlin quasiparticle. *Phys. Rev. Lett.* **79**, 2526 (1997).
- [69] X. Jehl, M. Sanquer, R. Calemczuk, and D. Mailly. Detection of doubled shot noise in

- short normal-metal superconductor junctions. *Nature* **405**, 50 (2000).
- [70] J. C. Cuevas and W. Belzig. Full counting statistics of multiple Andreev reflections. *Phys. Rev. Lett.* **91**, 187001 (2003).
 - [71] T. Fujisawa, T. Hayashi, R. Tomita, and Y. Hirayama. Bidirectional counting of single electrons. *Science* **312**, 1634 (2006).
 - [72] S. Gustavsson, R. Leturcq, B. Simović, R. Schleser, T. Ihn, P. Studerus, K. Ensslin, D. C. Driscoll, and A. C. Gossard. Counting statistics of single electron transport in a quantum dot. *Phys. Rev. Lett.* **96**, 076605 (2006).
 - [73] E. V. Sukhorukov, A. N. Jordan, S. Gustavsson, R. Leturcq, T. Ihn, and K. Ensslin. Conditional statistics of electron transport in interacting nanoscale conductors. *Nature Physics* **3**, 243 EP (2007).
 - [74] C. Flindt, C. Fricke, F. Hohls, T. Novotný, K. Netočný, T. Brandes, and R. J. Haug. Universal oscillations in counting statistics. *Proceedings of the National Academy of Sciences* **106**, 10116 (2009).
 - [75] D. Kambly, C. Flindt, and M. Büttiker. Factorial cumulants reveal interactions in counting statistics. *Phys. Rev. B* **83**, 075432 (2011).
 - [76] P. Stegmann, B. Sothmann, A. Hucht, and J. König. Detection of interactions via generalized factorial cumulants in systems in and out of equilibrium. *Phys. Rev. B* **92**, 155413 (2015).
 - [77] C. Flindt and J. P. Garrahan. Trajectory phase transitions, Lee-Yang zeros, and high-order cumulants in full counting statistics. *Phys. Rev. Lett.* **110**, 050601 (2013).
 - [78] J. M. Hickey, S. Genway, I. Lesanovsky, and J. P. Garrahan. Time-integrated observables as order parameters for full counting statistics transitions in closed quantum systems. *Phys. Rev. B* **87**, 184303 (2013).
 - [79] L. S. Levitov and G. B. Lesovik. Charge distribution in quantum shot noise. *JETP Lett.* **58**, 230 (1993).
 - [80] A. O. Gogolin and A. Komnik. Towards full counting statistics for the Anderson impurity model. *Phys. Rev. B* **73**, 195301 (2006).
 - [81] C. N. Yang and T. D. Lee. Statistical theory of equations of state and phase transitions. i. theory of condensation. *Phys. Rev.* **87**, 404 (1952).

BIBLIOGRAPHY

- [82] R. A. Blythe and M. R. Evans. Lee-Yang zeros and phase transitions in nonequilibrium steady states. *Phys. Rev. Lett.* **89**, 080601 (2002).
- [83] T. D. Lee and C. N. Yang. Statistical theory of equations of state and phase transitions. ii. lattice gas and ising model. *Phys. Rev.* **87**, 410 (1952).
- [84] P. C. Hemmer and E. H. Hauge. Yang-Lee distribution of zeros for a van der Waals gas. *Phys. Rev.* **133**, A1010 (1964).
- [85] I. Bena, M. Droz, and A. Lipowski. Statistical mechanics of equilibrium and nonequilibrium phase transitions: The Yang-Lee formalism. *International Journal of Modern Physics B* **19**, 4269 (2005).
- [86] R. A. Blythe. An introduction to phase transitions in stochastic dynamical systems. *Journal of Physics: Conference Series* **40**, 1 (2006).
- [87] A. G. Abanov and D. A. Ivanov. Allowed charge transfers between coherent conductors driven by a time-dependent scatterer. *Phys. Rev. Lett.* **100**, 086602 (2008).
- [88] D. A. Ivanov and A. G. Abanov. Phase transitions in full counting statistics for periodic pumping. *EPL (Europhysics Letters)* **92**, 37008 (2010).
- [89] K. Brandner, V. F. Maisi, J. P. Pekola, J. P. Garrahan, and C. Flindt. Experimental determination of dynamical Lee-Yang zeros. *Phys. Rev. Lett.* **118**, 180601 (2017).
- [90] Y. Utsumi, O. Entin-Wohlman, A. Ueda, and A. Aharony. Full-counting statistics for molecular junctions: Fluctuation theorem and singularities. *Phys. Rev. B* **87**, 115407 (2013).

2 Theoretical framework in the stationary regime

In this chapter I will introduce the theoretical framework that will be used along the thesis. Firstly, I present a minimal model to analyze the quantum transport through non-interacting mesoscopic systems. After that, I will introduce the non-equilibrium Green functions technique, the main theoretical tool used along the thesis, and its relation to physical observables. After that, the non-interacting model will then be generalized to include electron-electron interactions, electron-phonon interaction and superconducting correlations at the nanoscale. The specific methods for treating interactions in different regimes are also discussed along the thesis. The final part of the chapter is devoted to the full counting statistics analysis in both the interacting and non-interacting situations.

2.1 Impurity level Hamiltonian

Mesoscopic systems are usually composed by a nanoscale region, which will be referred simply as system in the following, connected to macroscopic electrodes which act as electron reservoirs. These devices usually include some gate electrodes used to control the junction parameters, such as the transmission coefficient. The problem is described by the Hamiltonian

$$H = H_{leads} + H_S + H_T \quad (2.1)$$

where $H_{leads} = \sum_{k\sigma,\nu} \epsilon_{k\sigma,\nu} c_{k\sigma,\nu}^\dagger c_{k\sigma,\nu}$ describe the non-interacting electron reservoirs, $c_{k\sigma,\nu}$ ($c_{k\sigma,\nu}^\dagger$) being the electron annihilation (creation) operator of electrons with momentum k and spin $\sigma = \uparrow, \downarrow$ in the electrode ν . For simplicity I will concentrate in the situation

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Figure 2.1: Left panel: schematic representation of a nanojunction coupled to two leads (blue regions). The system and the electrodes are separated by a small insulating region (green). Right panel: I show the energy scheme of the system where the system levels are separated by the electrostatic repulsion, U . The parameter ϵ_0 is the energy difference between the Fermi edge and its closest system energy level. A finite bias voltage can be added to the system, which leads to a symmetric shift of the electrodes' chemical potential with respect to the Fermi edge, ϵ_F .

when the system is only coupled to two electrodes (see the left panel of Fig. 2.1 for a schematic representation), labeled as left and right, i.e. $\nu = L, R$. The density of states of the electrodes will be considered flat with the bandwidth, D , taken as the largest energy in the model (this regime is also known as the *wideband approximation*). A finite voltage bias, V can be applied to the junction, which shifts the electrodes' Fermi level. For simplicity I will consider that the voltage drop is symmetric and the electrodes' chemical potential $\mu_L = -\mu_R = V/2$. From now on I will use the natural units $e = \hbar = k_B = 1$.

The nanoscopic system exhibits discrete energy levels due to the electron confinement (right panel of 2.1), where the level position can be usually controlled by using gate electrodes. The multiple levels give rise to the so-called Coulomb diamonds in the stability diagram: sharp conductance increasing when the levels enter the voltage window. In the limit where the levels are well separated the current is dominated by the level closer to the Fermi level. In this regime and in the absence of local interactions the system Hamiltonian is well approximated by

$$H_s = \sum_{\sigma} \epsilon_0 d_{\sigma}^{\dagger} d_{\sigma}, \quad (2.2)$$

where $d_{\sigma}^{\dagger}$ is the creation operator in the system and ϵ_0 is the energy level position.

Finally, the last term in Eq. (2.1) describes the coupling term between the electrodes and the system. I will consider situations when the electrodes and the system are in close

contact. The overlap of the electron wavefunctions in the electrodes and the system gives rise to a tunneling Hamiltonian given by

$$H_T = \sum_{k\sigma,\nu} \left(t_{k\sigma,\nu} c_{k\sigma,\nu}^\dagger d_\sigma + t_{k\sigma,\nu}^* d_\sigma^\dagger c_{k\sigma,\nu} \right), \quad (2.3)$$

which allows the exchange of particles between the different parts of the device. The $t_{k\sigma,\nu}$ are the tunneling amplitudes which in the wideband approximation can be considered energy independent. In this regime it becomes useful to define the tunneling rates as $\Gamma_\nu \approx \pi |t_\nu|^2 \rho_F$, being ρ_F the density of states at the Fermi energy. Hereafter I will denote $\Gamma = \Gamma_L + \Gamma_R$. An important parameter in the model is the transmission coefficient at the Fermi energy

$$\tau = \frac{4\Gamma_L\Gamma_R}{\Gamma^2 + \epsilon_0^2}, \quad (2.4)$$

which describes the probability of an incoming electron to be transmitted from one to the other electrode.

In this simple model the electron spin does not play any important role (it will only give a factor 2 contribution in the electron transport properties). For this reason it will be omitted in what follows and re-introduced when needed. This simple model, called the spinless Anderson model, describes many situations of interest in the absence of interactions ranging from QDs or molecular junctions to QPCs in the $\Gamma_\nu \rightarrow \infty$ limit. In the next sections generalizations of this simple Hamiltonian will be discussed to introduce electron-electron, electron-phonon and superconducting correlations. However, it is illustrative to introduce first the Green functions formalism and to show how this simple problem can be solved.

2.2 Green function formalism

The Green functions are powerful tools to solve differential equations, used in a wide range of problems. In the case of mesoscopic systems, they are particularly useful to study the quantum transport through interacting systems, as it will be illustrated in the following sections of the chapter. This section is, however, focused on the non-interacting situation described by the Hamiltonian of Eq. (2.1). I will show how the non-equilibrium Green functions (NGFs) can be obtained in this simple example, discussing their relations to the physical observables.

2.2.1 Equilibrium Green functions

I will discuss first the simpler equilibrium situation when no bias is applied to the system. The problem is fully described by the retarded (r) and the advanced (a) Green functions. In the energy representation they are given by

$$[(\epsilon \pm i\eta)\mathbb{I} - H] g^{r,a}(\epsilon) = \mathbb{I}, \quad (2.5)$$

where η is an infinitesimium included to avoid the divergencies in the real energy axis and $\mathbb{I}$ is the unit operator. The general expression for the single particle retarded and advanced Green functions can be obtained by inverting Eq. (2.5), i.e.

$$g^{r,a}(\epsilon) = [(\epsilon \pm i\eta)\mathbb{I} - H]^{-1}. \quad (2.6)$$

As an illustration, the Green function of the uncoupled system described the Hamiltonian (2.2) can be written as

$$g_S^{a,r}(\epsilon) = \frac{1}{\epsilon \pm i\eta - \epsilon_0}. \quad (2.7)$$

Similarly, the uncoupled electrodes Green functions can be written as

$$g_\nu^{a,r}(k) = \sum_k \frac{1}{\epsilon \pm i\eta - \epsilon_k}, \quad (2.8)$$

where the sum is performed over all the energy range and the spin degree of freedom is omitted for simplicity. In the wideband approximation the contribution from the principal parts can be neglected on the previous equation. Averaging over the electron momenta, I find

$$g_\nu^{a,r} \approx -i\pi \sum_k \delta(\epsilon \pm i\eta - \epsilon_k) \approx \pm i\pi \rho_F \quad (2.9)$$

where δ is the Dirac functional and ρ_F is the density of states at the Fermi energy, containing the sum of all Delta functionals.

In the case of the system coupled to electrodes the (retarded/advanced) Green functions become matrices whose dimension depends on the number of different regions to describe in the device. In the simple case described by Eq. (2.1), the Green functions can be obtained through

$$\begin{pmatrix} (\epsilon \pm i\eta) - H_L & -H_{LS} & 0 \\ -H_{SL} & (\epsilon \pm i\eta) - H_S & -H_{SR} \\ 0 & -H_{RS} & (\epsilon \pm i\eta) - H_R \end{pmatrix} \begin{pmatrix} G_L^{r,a} & G_{LS}^{r,a} & 0 \\ G_{SL}^{r,a} & G_S^{r,a} & G_{SR}^{r,a} \\ 0 & G_{RS}^{r,a} & G_R^{r,a} \end{pmatrix} = \mathbb{I}, \quad (2.10)$$

where $H_{S\nu}$ is the tunneling part of the Hamiltonian connecting the system to the electrode ν and G are the many-body Green functions. From now on the unperturbed Green functions will be denoted by lower case letters, being the full ones denoted with capital letters. I will concentrate on the solution of the system Green function, since it describes all the interesting transport properties, although similar expressions can be obtained for the other components. Solving the matrix equation it is found

$$G_S^{r,a}(\epsilon) = \{[g_S^{a,r}(\epsilon)]^{-1} - \Sigma_T^{r,a}(\epsilon)\}^{-1}, \quad (2.11)$$

where $\Sigma_T^{r,a}(\epsilon) = \sum_{\nu=L,R} H_{S\nu}[(\epsilon \pm i\eta)\mathbb{I} - H_L]^{-1} H_{\nu S}$ is the tunneling self-energy due to the coupling to the electrodes. In the wideband approximation, this self-energy becomes energy independent and simplifies as $\Sigma_T^{r,a} = \pm i(\Gamma_L + \Gamma_R)$. As it will be shown in the next sections of this chapter, the self-energy will play an important role in the theory, since the many-body interactions can be introduced as a self-energy term. Eq. (2.11) is the so-called Dyson equation which includes the tunneling events to all orders. Another usual way of writing the Dyson equation is

$$G_S^{r,a}(\epsilon) = g_S^{a,r}(\epsilon) + g_S^{a,r}(\epsilon) \Sigma_T^{r,a}(\epsilon) G_S^{r,a}(\epsilon), \quad (2.12)$$

deduced by expanding the Dyson Eq. (2.11) in powers of the tunneling self-energy. The solution to this equation is

$$G_S^{r,a}(\epsilon) = \mp i \frac{1}{\epsilon - \epsilon_0 \pm i\Gamma}, \quad (2.13)$$

where the resonance at the level position becomes broadened due to the coupling to the electrodes. The local density of states (DOS) can be obtained from this Green function as

$$A(\epsilon) = \pm \frac{1}{\pi} \text{Im}[G_S^{a,r}(\epsilon)]. \quad (2.14)$$

In the simple case I was analyzing of a single level coupled to metallic electrodes, the DOS is given by

$$A(\epsilon) = \frac{1}{\pi} \frac{\Gamma}{(\epsilon - \epsilon_0)^2 + \Gamma^2}, \quad (2.15)$$

which is normalized, i.e. $\int d\epsilon A(\epsilon) = 1$. Moreover, it is important to note that the height of the resonance is independent of the level position and becomes $A(\epsilon = \epsilon_0) = 1/\pi\Gamma$. This result is a consequence of the Friedel's sum rule (FSR), which relates the local spectral density to the phase shift on the electrons wavefunctions due to the presence of the system that acts as an scatter region. This is an important result since it has been demonstrated to hold even in some interacting situations [1].

2.2.2 Time-dependent Green functions

The retarded and advanced Greens functions described in the previous subsection can be considered as the Fourier transform of a time-dependent Green functions. These Green functions are useful since they describe the system dynamics and are suitable for describing interacting systems. For defining the Green functions I will use the Heissenberg picture, where the system eigenstates do not evolve with time, and the operators evolve under the action of a Hamiltonian as

$$A^H(t) = e^{iHt} A(0) e^{-iHt}. \quad (2.16)$$

The mean value of a given operators can be written as

$$\langle A^H(t) \rangle = \text{Tr} [A^H(t) \rho^H], \quad (2.17)$$

where ρ^H is the density matrix, which describes the system state. Considering the base formed by the Hamiltonian eigenstates, $H |\Psi_n^H\rangle = \epsilon_n |\Psi_n^H\rangle$, the density matrix can be written as

$$\rho = \sum_n \rho_n |\Psi_n^H\rangle \langle \Psi_n^H|. \quad (2.18)$$

where the coefficients $\rho_n = \exp[(\epsilon_n - \epsilon_0)/T]$ depend on the temperature T . Note that at zero temperature only the ground state will contribute to the density matrix. Now, the retarded Green function of two given quantum operators A and B can be written as

$$G_{A,B}^r(t, t') = -i\theta(t - t') \sum_n \langle \Psi_n^H | [A(t), B(t')]_+ | \Psi_n^H \rangle, \quad (2.19)$$

where Ψ_H is the equilibrium state wavefunction in the Heisenberg picture, the brackets represent the anti-commutation operation and $G_{A,B}^a(t, t') = [G_{A,B}^r(t', t)]^*$. it is worth noticing that the definition of Eq. (2.19) is also valid in the interacting situation. From the retarded Green function of the uncoupled system described it is found

$$g_S^r(t - t') = -i\theta(t' - t) e^{-i\epsilon_0(t - t')}, \quad (2.20)$$

which is as expected the Fourier transform of Eq. (2.7). Another useful way of writing the Green functions is through the Lehman representation. Using the fact that the full set of eigenstates of the Hamiltonian is a complete basis, the identity operator can be written as

$$\mathbb{I} = \sum_m |\Psi_m^H\rangle \langle \Psi_m^H|, \quad (2.21)$$

Figure 2.2: Keldyshs contour where the arrows represent the time ordering in the causal (−) and anti-causal (+) branches.

where n is an index to label the Hamiltonian eigenstates. Using this relation it is possible to write Eq. (2.19) as

$$G_{A,B}^r(t, t') = -i\theta(t - t') \sum_{n,m} \rho_n \left[\langle \Psi_n^H | A(t) | \Psi_m^H \rangle \langle \Psi_m^H | B(t') | \Psi_n^H \rangle + \langle \Psi_n^H | B(t') | \Psi_m^H \rangle \langle \Psi_m^H | A(t) | \Psi_n^H \rangle \right]. \quad (2.22)$$

In the simple situation where the operators are related through $B = A^\dagger$, the Green function in the time representation simplifies as

$$G_{A,A^\dagger}^r(t, t') = -i\theta(t - t') \sum_{n,m} \rho_m e^{-i(\epsilon_m - \epsilon_n)(t - t')} \left(|\langle \Psi_n^H | A | \Psi_m^H \rangle|^2 + |\langle \Psi_n^H | A^\dagger | \Psi_m^H \rangle|^2 \right) \quad (2.23)$$

Finally, taking the Fourier transform of the retarded Green function it is found

$$G_{A,A^\dagger}^r(\epsilon) = \sum_{n,m} \left[\frac{|\langle \Psi_n^H | A | \Psi_m^H \rangle|^2}{\epsilon - (\epsilon_m - \epsilon_n) + i\eta} + \frac{|\langle \Psi_n^H | A^\dagger | \Psi_m^H \rangle|^2}{\epsilon + (\epsilon_m - \epsilon_n) + i\eta} \right], \quad (2.24)$$

which is the so-called Lehmann representation.

2.2.3 Non-equilibrium Green functions

In this subsection I will introduce the non-equilibrium Green functions (NGFs) formalism which provides a general framework for the non-equilibrium statistical mechanics. The main difficulty in applying the Green functions definitions showed previously in this chapter is that they depend on the stationary system wavefunction (2.19). In the non-equilibrium situation the state can evolve in time, where the initial and final states are in principle different. In order to solve this problem, Keldysh in his original work [2] proposed the use of a time contour as depicted in Fig. 2.2. The time contour goes from the initial time to the final one through the anti-causal branch (+) and returns back to the initial time through the causal branch (−). In both cases the evolution happens along the real

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time axis and the imaginary part of the line in Fig. 2.2 has been only included for clarity.

Using these definitions, there exist four different kind of Keldysh Green functions, depending on the branch where the time arguments are. The Green functions can then be written as

$$G_{AB}^{\alpha\beta}(t^\alpha, t^\beta) = -i \langle T_c A(t^\alpha) B(t^\beta) \rangle, \quad (2.25)$$

where the average value is taken over the initial state and will be omitted in the following for simplicity. The superscripts $\alpha\beta = \pm$ denote the Keldyshs branch where the times t, t' are and T_c is the time ordering in the Keldysh contour indicated by the arrows in Fig. 2.2. It is often more convenient to write the Green functions in a matrix way as

$$\hat{G}_{AB}(t, t') = \begin{pmatrix} G_{AB}^{++}(t, t') & G_{AB}^{+-}(t, t') \\ G_{AB}^{-+}(t, t') & G_{AB}^{--}(t, t') \end{pmatrix} \quad (2.26)$$

where the symbol $\hat{}$ will be used in the following to denote the implicit Keldysh structure. There are some fundamental relations between the diagonal and off-diagonal components

$$\begin{aligned} G_{AB}^{++}(t, t') &= \theta(t - t') G_{AB}^{-+}(t, t') + \theta(t' - t) G_{AB}^{+-}(t, t'), \\ G_{AB}^{--}(t, t') &= \theta(t - t') G_{AB}^{+-}(t, t') + \theta(t' - t) G_{AB}^{-+}(t, t'). \end{aligned} \quad (2.27)$$

By performing a rotation of $-\pi/4$ (usually known as Keldysh rotation) the NGF matrix (2.26) becomes a lower triangular matrix given by

$$\hat{G}_{AB}(t, t') = \begin{pmatrix} 0 & G_{AB}^a(t, t') \\ G_{AB}^r(t, t') & G_{AB}^k(t, t') \end{pmatrix}, \quad (2.28)$$

from where the relations between the Keldysh components and the retarded and advanced Green functions can be found

$$\begin{aligned} G_{AB}^r(t - t') &= -\theta(t - t') [G_{AB}^{+-}(t, t') - G_{AB}^{-+}(t, t')] , \\ G_{AB}^a(t - t') &= \theta(t' - t) [G_{AB}^{+-}(t, t') - G_{AB}^{-+}(t, t')] . \end{aligned} \quad (2.29)$$

Finally, the Diagonal component of Eq. (2.28) can be written as $G_{AB}^k(t, t') = G_{AB}^{+-}(t, t') + G_{AB}^{-+}(t, t')$. Thus, only two components of the NGF are independent and the remaining ones can be computed by using the fundamental Keldysh relations. It is also worth commenting that in the stationary regime (long time limit), the NGFs become time translational invariant, depending only on the time difference, $t - t'$.

As an illustration, I will summarize the off-diagonal components of the uncoupled system and electrodes given by Eq. (2.1). For instance, the system NGFs are

$$g_S^{+-}(t-t') = in_d(0)e^{-i\epsilon_0(t-t')}, \quad g_S^{-+}(t-t') = i[n_d(0)-1]e^{-i\epsilon_0(t-t')}, \quad (2.30)$$

where $n_d(0)$ represents the initial system occupation. Note that these components provide information about the occupied and unoccupied part of the system respectively. The uncoupled leads NGFs can be obtained in a rather similar way. Using the energy representation, where their expressions become specially compact it is possible to find

$$g_\nu^{+-}(\epsilon) = 2\pi i \rho_{F,\nu} f_\nu(\epsilon), \quad g_\nu^{-+}(\epsilon) = 2\pi i \rho_{F,\nu} [f_\nu(\epsilon) - 1], \quad (2.31)$$

where f_ν is the Fermi distribution function and Eqs. (2.27) and (2.29) can be used to obtain the remaining components. It turns out [2] that the NGFs satisfy a matrix Dyson equation, similarly to Eq. (2.12) in Keldysh space. In the next subsection I summarize the main relations between the NGFs and the physical observables.

2.2.4 Transport properties

In this subsection I summarize the main relations between the NGFs and the system transport properties. As mentioned above, the off-diagonal Green functions provide information about the occupied and unoccupied part of the level spectral density. More precisely, the DOS can be factorized as

$$A_{o(u)}(\epsilon) = \frac{1}{\pi} \text{Im} \left[\int_0^\infty d(t-t') e^{-i\epsilon(t-t')} G_S^{+-(-+)}(t-t') \right], \quad (2.32)$$

recovering the complete DOS through $A = A_o + A_u$. Moreover, the charge in the system can be computed by integrating A_o over all the energy range (which is equivalent to integrate the full DOS up to the Fermi energy).

The current between one of the electrodes and the system can be computed from the non-local Green functions

$$I_\nu(t) = t_\nu [G_{S\nu}^{+-}(t, t) - G_{\nu S}^{+-}(t, t)]. \quad (2.33)$$

Manipulating this expression the current can be rewritten as a function of the local Green functions as

$$I_\nu(t) = 2t_\nu \int_{t_0}^t [G_S^{+-}(t, t_1) g_L^{-+}(t_1, t) - G_S^{-+}(t, t_1) g_L^{+-}(t_1, t)]. \quad (2.34)$$

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In the stationary regime, the upper and lower integration limits can be taken as $\pm\infty$ respectively. Symmetrizing the current ($\langle I \rangle \equiv (I_L + I_R)/2$) and transforming to the energy representation I arrive to the well known Meir-Wingreen formula [3] for the current

$$\langle I \rangle = 2\Gamma \int_{-\infty}^{\infty} d\epsilon [f_L(\epsilon) - f_R(\epsilon)] A(\epsilon), \quad (2.35)$$

which also holds in the interacting situation. This expression constitutes a generalization of the Landauer formula, applicable to systems exhibiting localized interactions. In the next section I will introduce the minimal models to include electron-electron and electron-phonon interactions. I will introduce first the general framework for treating the interactions.

2.2.5 Interaction picture

In the previous section I have defined the system NGFs and its relation to the physical observables such as the current, the system population and the DOS. In the absence of interactions, the self-energy can be easily determined and the NGFs can be computed by a simple inversion (2.11). However, when interactions are included, it is not always possible to determine analytically the self-energy since it may depend on the system Green function, leading to non-linear coupled equations for the NGFs and the self-energy. There exist some numerical ways of solving these coupled equations based on NRG [4] or MC techniques [5] (MC), among others. In general these codes are computationally demanding and limited to a given parameter regime due to convergence issues. Another strategy for obtaining the system Green functions, followed along the thesis, consists on restricting the calculation to only the leading processes, represented by the corresponding Feynman diagrams. A widely used technique is the so-called *perturbation* expansion, where the interaction strength is considered the smallest energy parameter and the NGFs are expanded at the lowest orders. Let me begin with a Hamiltonian that can be written as

$$H = H_0 + V, \quad (2.36)$$

where H_0 represents the non-interacting part analytically solvable and V represents the interaction term. The non-interacting and the interacting Greens functions can be written as

$$\begin{aligned} g^{a,r}(\epsilon) &= [(\epsilon \pm i\eta) - H_0]^{-1} \\ G^{a,r}(\epsilon) &= [(\epsilon \pm i\eta) - H_0 - \Sigma_I^{a,r}]^{-1}, \end{aligned} \quad (2.37)$$

being Σ_I the interaction self-energy, whose expression will be derived below. By expanding the interacting Green function in powers of Σ_I , the relation between the interacting and the non-interacting Green function is found

$$G^{a,r}(\epsilon) = g^{a,r}(\epsilon) + g^{a,r}(\epsilon) \Sigma_I^{a,r} G^{a,r}(\epsilon), \quad (2.38)$$

which is a generalization of the Eq. (2.12), obtained for the tunneling Hamiltonian. In the interaction representation, the average value of the product of time-dependent operators, A and B can be written as

$$\langle A(t)B(t') \rangle = \frac{\langle T_c [S_c^I A^I(t) B^I(t')] \rangle}{\langle S_c \rangle}, \quad (2.39)$$

where the superindex I is used to denote the interaction picture, T_c is the time ordering in the Keldysh contour and S^I is the time evolution in the contour, defined as

$$S_c^I = T_c \left[-i \int_c dt_1 V^I(t_1) \right]. \quad (2.40)$$

The time evolution can be expanded in powers of the interaction term as

$$S_c^I = 1 - i \int dt_1 V(t_1) - \frac{1}{2} \int dt_1 \int dt_2 V(t_1) V(t_2) + \dots \quad (2.41)$$

For small interaction strengths, a truncation of the perturbation expansion is expected to lead to accurate results. The constant term describes the non-interacting situation while the higher orders are corrections due to interactions. The linear term in V leads to the mean-field approximation which locally renormalizes the system energy and provides the leading term in the weakly interacting situation.

For an increasing interaction strength, higher orders have to be included to achieve a good accuracy. In particular, when the interaction dominates, all the higher order terms in Eq. (2.41) have to be included and the problem cannot be addressed using the simple perturbation theory. A way of increasing the range of validity of a perturbative approximation consists on summing all the diagrams of a family up to infinite order through the Dyson Eq. (2.11). The self-energy up to a finite order can be written as

$$\hat{\Sigma}_I = \left\langle 1 - i \int dt_1 V(t_1) - \frac{1}{2} \int dt_1 \int dt_2 V(t_1) V(t_2) + \dots \right\rangle. \quad (2.42)$$

The calculation of the successive terms of the perturbation expansion usually involves the evaluation of the mean value of a product of multiple operators. To compute them the Wick's theorem can be used, which states that the many operators momentum are

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determined by the two operator moments. For instance, the product of four different operators can be written as

$$\langle ABCD \rangle = \langle AB \rangle \langle CD \rangle \pm \langle AC \rangle \langle BD \rangle + \langle AD \rangle \langle BC \rangle, \quad (2.43)$$

where the sign in the second term depends on the (bosonic/fermionic) nature character of the particle operators. This implies that the many operators Green functions can always be determined by the two operator ones, drastically reducing the number of variables in the problem. The different terms in the previous equation describe the different processes that can be represented by Feynman diagrams. It can be demonstrated that the diagrams written in Eq. (2.43) vanishes when they do not share time arguments, i.e. if they are *disconnected* diagrams.

It is important to note that the self-energy given in Eq. (2.42) may depend generally on the dressed NGFs leading to non-linear coupled equations. These equations can be solved numerically by using self-consistent methods, consisting on compute the NGFs and the self-energy iteratively until reaching the convergence condition [6]. This method requires, however, from a *good* initial condition to convergence to the appropriate result [7]. In chapter 5 I will discuss more about this method, providing an efficient algorithm to solve the self-consistent loop in time domain which provides accurate results in the weak to moderate coupling strengths. The goal of this chapter is, however, to introduce the minimal models to describe interactions at the nanoscale and the approximations used along the thesis to study them.

2.3 Electron-electron interaction: Anderson model

The first example of interaction that is going to be discussed is the localized electron-electron interaction. Electrons confined in nanoscopic regions suffer a strong electrostatic repulsion, limiting its number in the device. This interaction gives raise to the Coulomb blockade and the Kondo effect in nanostructures. I will concentrate on the simplest situation of a single orbital, where the electrons are characterized by the momentum and the spin degrees of freedom. The minimal model which includes this kind of interaction is the so-called Anderson model [8] which supposes a spin degenerate model with on-site

Figure 2.3: Overview of the different regimes of the Anderson model. The shadow region indicates the regime studied in the thesis.

Coulomb repulsion, coupled to metallic electrodes

$$H = H_0 + V_{e-e}, \quad (2.44)$$

where the non-interacting part Hamiltonian is given by Eq. (2.1). The Coulomb repulsion term is given by $V_{e-e} = U n_{\uparrow} n_{\downarrow}$ where $n_{\sigma} = d_{\sigma}^{\dagger} d_{\sigma}$ and U is the interaction strength. The main parameter of the model is the Kondo temperature

$$T_K = \sqrt{\frac{U\Gamma}{2}} e^{-\frac{\pi}{2U\Gamma} |\epsilon_0 + U| |\epsilon_0|}, \quad (2.45)$$

which sets the frontier between the classical and the quantum regime [9]. In the classical regime ($T > T_K$) where the Coulomb blockade effect dominates, a classical rate equation provide accurate results. In this thesis I am mostly interested on the $T < T_K$ regime, where a quantum treatment of the problem becomes necessary. For $U/\Gamma < 1$, a perturbative expansion is valid, while for $U/\Gamma \gg 1$ quantum fluctuations dominate and non-perturbative effects become in principle important. In the thesis I focus on the weakly coupled regime showing that, by using an appropriate perturbation expansion, the range of validity of the approximation can be extended up to rather strong electron-electron interactions ($U/\Gamma \sim 10$). In Fig. 2.3 an schematic representation of the different regimes is presented, where the colored region represents the regime of interest of this thesis.

An exact solution to the problem is given by the Bethe ansatz, which predicts that the ground state at low temperatures is a singlet [10–12]. It becomes specially simple in the atomic limit ($\Gamma \rightarrow 0$) where the self-energy can be written as [13]

$$\Sigma_{\Gamma \rightarrow 0}^r(\epsilon) = U^2 \frac{n_{\bar{\sigma}} (1 - n_{\bar{\sigma}})}{\epsilon - \epsilon_0 - U (1 - n_{\bar{\sigma}})}, \quad (2.46)$$

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and the retarded Green function can be obtained by inverting the Dyson equation (2.11), finding

$$G_{\Gamma \rightarrow 0}^r(\epsilon) = \frac{1}{2\pi} \left[\frac{n_{\bar{\sigma}}}{\epsilon - \epsilon_0 - U} + \frac{1 - n_{\bar{\sigma}}}{\epsilon - \epsilon_0} \right], \quad (2.47)$$

which predicts the appearance of two bands separated in energy by U .

The other limit that can be easily addressed is the weakly coupled regime where $U/\Gamma \ll 1$. In this regime it is expected that a simple perturbation expansion up to finite order will provide accurate results. By expanding the evolution operator in powers of the electron-electron interaction Hamiltonian, as described in Eq. (2.41). In the next subsections I will discuss different level of approximations by including the leading terms in the expansion

2.3.1 Mean field approximation

The simplest way of treating the interaction consists on just including the interaction up to the first order in perturbation expansion, i.e. linear term in Eq. (2.41). Using the definition of the NGFs, the self-energy is given by

$$\Sigma_{HF,\sigma}^{\alpha\alpha}(t, t') = \alpha U \hat{n}_{\bar{\sigma}}(t) \delta(t - t'), \quad \Sigma_{HF,\sigma}^{+-}(t, t') = \Sigma_{HF,\sigma}^{-+}(t, t') = 0, \quad (2.48)$$

where $\bar{\sigma}$ is used to denote the opposite spin to σ and n_{σ} is the charge per spin. It is worth noticing that the charge in the previous equation has to be determine self-consistently. The dot Green function can be obtained by inverting the Dyson equation in Keldysh space,

$$\hat{G}_{HF,\sigma} = [\hat{g}_S^{-1} - \hat{\Sigma}_{HF,\sigma}]^{-1}. \quad (2.49)$$

This approach, known as Hartree-Fock or tadpole approximation, renormalizes the system energy depending on the instantaneous charge in the system as $\tilde{\epsilon}_{\sigma}(t) = \epsilon_0 + U n_{\bar{\sigma}}(t)$. In particular the electron-hole symmetric case is recovered when the level is set at $\epsilon_0 = -U/2$, below the Fermi edge. This approximation is able to reproduce the system observables in the $U/\Gamma < 1$ regime. However, it does not provide an accurate description of the transport properties for $U/\Gamma \gtrsim 1$ due to the absence of electron correlation effects. Moreover it predicts the appearance of a magnetic solution, known to be unphysical (see chapter 5 for more details). In order to include the correlation effects, higher orders beyond the mean-field approximation have to be included.

2.3.2 Effects beyond the mean field

The minimal approximation that include correlation effects is the second order perturbation expansion in the coupling parameter, U^2 term in Eq. (2.41). The off-diagonal Keldysh component for this perturbative approximation can be written as

$$\Sigma_{X,\sigma}^{\alpha\beta}(t, t') = \alpha\beta U^2 G_{S,\sigma}^{\alpha\beta}(t, t') G_{S,\bar{\sigma}}^{\alpha\beta}(t, t') G_{S,\bar{\sigma}}^{\beta\alpha}(t', t). \quad (2.50)$$

Note that this term, known as exchange term (Σ_X), depends on the system dressed Green functions denoted by the capital letters. The equation for the self-energy, together with the Dyson equation constitute two non-linear coupled equations, whose analytic solution is unknown. The numerical solution of the equations can be obtained by means of the self-consistent method [14]. However, this self-consistent method tends to destroy the good analytic properties of the second order perturbation mentioned above, leading to less accurate results. A better strategy consists on perturb over the HF solution given in Eq. (5.13). Thus, the self energy can be written as

$$\Sigma_{\sigma}^{\alpha\beta}(t - t_1) = \alpha\beta U^2 G_{HF,\sigma}^{\alpha\beta}(t - t') G_{HF,\bar{\sigma}}^{\alpha\beta}(t - t') G_{HF,\bar{\sigma}}^{\beta\alpha}(t' - t), \quad (2.51)$$

and the system NGFs can be written by inverting the Dyson equation (2.11). It can be demonstrated that this self-energy exhibit good analytic behavior, recovering also the atomic limit given by Eq. (2.46) [15]. This approximation predicts the appearance of two shoulders at $\pm U/2$, related to the charge side bands, in quantitative agreement with the available numerical results (see Fig. 2.4) for a wide range of parameters. However, the width of the central resonance is somewhat wider in the $U/\Gamma \gg 1$ regime, leading to an overestimation of T_K . The good analytic properties of the second order self-energy has been exploited to develop better approximations which interpolate between the atomic and the atomic limit [15–18].

It is worth commenting that there are some features that are not well captured by this simple approach. For instance, the second order perturbation expansion is unable to describe the splitting of the central resonance at voltages $V < T_K$. This shortcoming is corrected by including higher order terms in the perturbation expansion over the HF solution. For instance, in Ref. [20], the authors demonstrated that the fourth order term is sufficient to describe this effect in the electron-hole symmetric case.

Figure 2.4: Equilibrium DOS of the Anderson model for $U = 8$ and $\epsilon_0 = -U/2$ showing an overall agreement between the second order self-energy (2.51) and the NRG calculations from Ref. [19]. In the inset I show a zoom on small frequency range.

2.4 Electron-phonon interaction: the spinless Anderson-Holstein model

Many nanoscale junctions are flexible, exhibiting localized mechanical modes in the system. These vibrational modes couple to the electrons in the system, leading to possible elastic and inelastic transitions, which play a fundamental role in the transport. Signatures of the electron-phonon interaction have been observed in the transport properties of nanowires [21, 22], carbon nanotubes [23] or small molecular junctions [24], among others. An accurate description of these systems becomes challenging since it requires methods able to describe the mechanical and the electronic degrees of freedom simultaneously. A minimal model to describe this interaction is the so-called spinless Anderson-Holstein model [25] (also known simply as Holstein model). In this model the electrons in the nanojunction are linearly coupled to a phonon mode. Its Hamiltonian is given by

$$H = H_0 + H_{phon} + V_{e-p}, \quad (2.52)$$

Figure 2.5: overview of the different regimes of the Holstein model. The shadow region indicates the regimes studied in the thesis.

where the non-interacting part of the Hamiltonian is given by Eq. (2.1) and $H_{phon} = \omega_0 a^\dagger a$ describes a non-interacting bosonic mode of frequency ω_0 (a and $a^\dagger$ are the boson annihilation and creation operators, respectively). The term $V_{e-p} = \lambda(a + a^\dagger)d^\dagger d$ describes the localized electron-phonon interaction in the system, considered to be linear for simplicity. Note that the spin degree of freedom has been dropped for simplicity.

This simple model contains a very rich phenomenology. The different parameter regimes are determined by two dimensionless parameters [26]. The first one is the ratio between the temperature and the tunneling rate (T/Γ). For large values of T/Γ a classical rate equation has been shown to provide accurate results, while for small T/Γ values a quantum treatment of the problem becomes necessary. This last regime will be studied in detail in the thesis. The second important parameter is the ratio between the electron-phonon coupling with the tunneling rate and the phonon frequency ($\lambda^2/\omega_0\Gamma$). For $\lambda^2/\omega_0\Gamma < 1$ perturbative approximations are expected to provide accurate results, while for $\lambda^2/\omega_0\Gamma > 1$ the system enters into the strong coupling regime (also known as polaronic regime). The different regimes are depicted in Fig. 2.5, where the colored region denotes the parameter range of interest of this thesis. In the remaining of this section I will introduce the approximations to study both the weakly and the strongly coupling regimes.

2.4.1 Second order perturbation expansion

In the weak electron-phonon coupling regime ($\lambda^2/\omega_0\Gamma \ll 1$), a perturbative treatment of the problem provides an accurate description of the system observables. At integer values of $V/2\omega_0$ the conductance exhibits a series steps, due to the opening of new scattering

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channels. The competition between the elastic and inelastic scattering events between electrons and phonons lead to a change on sign of the first conductance step from negative to positive when the transmission factor changes from a nearly perfect situation to a low value [22]. Expanding the self-energy it is possible to find a perturbative approximation for the interaction self-energy

$$\hat{\Sigma}_{e-p} = \left\langle 1 - i \int dt_1 V_{e-p}(t_1) - \int dt_1 \int dt_2 V_{e-p}(t_1) V_{e-p}(t_2) + \dots \right\rangle. \quad (2.53)$$

Differently to the previously discussed electron-electron interaction, the linear term in the interaction parameter vanishes.

The second order term in Eq. (2.53) can be divided into two different contributions. The first one is the Hartree approximation which depends on the system charge as

$$\Sigma_H^{\alpha\beta}(t, t') = \alpha \lambda^2 \delta_{\alpha\beta} \delta(t - t') \int dt' D^r(t, t') n(t'), \quad (2.54)$$

where $\hat{D}(t, t') = -i \langle T_c \varphi(t) \varphi(t') \rangle$ is the phonon Green function dressed with the interactions with the electrons, with $\varphi(t) = a(t) + a^\dagger(t)$ and $n = d^\dagger d$. It is also important to note that the phonon Green function is dressed with the electronic degrees of freedom due to the coupling parameter. The simplest approximation is the Born approximation, which considers that the electrons are much faster and the free phonon propagator can be considered as bare. The expression of the free phonon propagator is

$$\hat{d}(t, t') = -i \begin{pmatrix} 2n_p \cos \omega_0(t - t') + e^{-i\omega_0|t-t'|} & n_p e^{-i\omega_0(t-t')} + (n_p + 1) e^{i\omega_0(t-t')} \\ n_p e^{i\omega_0(t-t')} + (n_p + 1) e^{-i\omega_0(t-t')} & 2n_p \cos \omega_0(t - t') + e^{i\omega_0|t-t'|} \end{pmatrix}, \quad (2.55)$$

where n_p is the free phonon population, described in a thermal equilibrium situation by the Bose-Einstein distribution $n_p = 1/[\exp(\omega_0/T) - 1]$. Thus, the retarded component can be written as $d^r(t, t') = -2\theta(t) \sin[\omega_0(t - t')]$. In the stationary regime, this term renormalizes the system energy as $\tilde{\epsilon} = \epsilon_0 + \lambda^2/2\omega_0$. At variance to the case of electron-electron interaction discussed in the previous section, the electron-phonon interaction introduces retardation effects already at the mean-field level. As it will shown in chapter 5, these effects are important in the system time evolution, except in the regime where the phonon dynamics is much faster than the electron dynamics ($\omega_0 \gg \epsilon_0, \Gamma$) [27]. In this limit the Hartree self-energy tends to

$$\hat{\Sigma}_{H, \omega_0 \gg}^{\alpha\beta}(t, t') = -\alpha \delta_{\alpha\beta} \delta(t - t') \frac{2\lambda^2}{\omega_0} n(t). \quad (2.56)$$

Figure 2.6: Equilibrium DOS of the spinless Holstein model for $\lambda = \Gamma$, $\omega_0 = 2\Gamma$ and $\epsilon_0 = \lambda^2/\omega_0$, showing an overall agreement between the Born approximation (solid line) (5.16,2.58) and NRG calculation (dashed line). The inset show the convergence to the FSR at low frequencies.

The other term in the second order perturbation expansion is the exchange term, which introduces electron correlation effects mediated by the phonon mode. The self-energy expression is

$$\Sigma_X^{\alpha\beta}(t, t') = \alpha\beta\lambda^2 G_S^{\alpha\beta}(t, t') D^{\alpha\beta}(t, t'), \quad (2.57)$$

where both, electron and phonon propagators appearing in the equation, are fully dressed with the interaction. Similarly to the Hartree term discussed previously in the section, the simplest approach consists on substituting the dressed by the free propagators ($\hat{d}$ and $\hat{g}$) in Eq. (2.57). In this simple approach, the exchange term can be written as

$$\Sigma_{X,Born}^{\alpha\beta}(t, t') = \alpha\beta\lambda^2 g_S^{\alpha\beta}(t, t') d^{\alpha\beta}(t, t'), \quad (2.58)$$

and by inverting Eq. (2.11) the the dressed electronic Green function can be obtained. This simple approximation only considers processes involving the adsorption or emission of a single virtual phonon, disregarding the effects from the non-equilibrium phonon dynamics. These processes lead to the appearance of two phonon sidebands in the DOS at energies $\epsilon \approx \pm\omega_0$, as shown in Fig. 2.6. This result is in quantitative agreement with exact results obtained from the NRG method (see appendix A for details). However,

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the Born approximation predicts the appearance of a dip at energies $\epsilon = \pm\omega_0$ due to a divergence in the exchange term of the self-energy given in Eq. (2.58), where the DOS vanishes. This pathological feature is smoothed when including a phonon lifetime, as it will be commented in detail in chapter 5.

This approximation is expected to provide accurate results in the $\lambda^2/\omega_0\Gamma \ll 1$ regime. However, it progressively deviates from the expected result for increasing λ , where a single electron can absorb or emit a high number of phonons. Those processes lead to the appearance of a series of side bands at energies $\sim \pm k\omega_0$, absent in the Born approximation. Moreover, the Born approximation does not describe the effects of non-equilibrium phonon dynamics, generated by the coupling to the electrons. All these shortcomings can be improved, increasing also the range of validity of the perturbative approximations, by means of the self-consistent method, introduce in the next subsection.

2.4.2 Self-consistent approximations

In this subsection I introduce different self-consistent approximations to improve the range of validity of the Born second order perturbation expansion in the interaction self-energy. The starting point of the subsection is the second order self-energies written in the previous subsection in Eqs. (5.16) and (2.57). The simplest extension to the Born approximation is the self-consistent Born approximation (SCBA), where the electron propagator is considered to be fully dressed with the interaction while keeping bare the phonon one. Thus, the exchange term can be written as

$$\Sigma_{X,SCA}^{\alpha\beta}(t, t') = \alpha\beta\lambda^2 G_S^{\alpha\beta}(t, t') d^{\alpha\beta}(t, t'). \quad (2.59)$$

At variance to the Anderson case discussed in the previous section, perturbations over the Hartree Green function provide a worse qualitative agreement to the exact calculations than using the full dressed propagators in the exchange term. This is due to the fact that both Hartree and exchange self-energies are of the same order in λ , suggesting that they have to be treated in a similar way. The self-consistent procedure generate higher order sidebands, originated by processes involving the emission or absorption of more than a virtual phonon. However, this approach still does not describe the non-equilibrium phonon dynamics. In order to include those effects the phonon propagator has to be dressed with the electronic degrees of freedom. Similarly to the electronic case, the phonon Green function satisfies a Dyson equation

$$\hat{D}(t, t') = \hat{d}(t, t') + \int dt_1 \int dt_2 \hat{d}(t, t_1) \hat{\Pi}(t_1, t_2) \hat{D}(t_2, t'), \quad (2.60)$$

which describes processes involving more than a phonon, including also the possibility of emitting and absorbing real phonons [28]. The corresponding self-energy given by

$$\Pi^{\alpha\beta}(t, t') = -i\alpha\beta\lambda^2 G_S^{\alpha\beta}(t, t') G_S^{\beta\alpha}(t', t). \quad (2.61)$$

I will consider two different schemes for dressing the phonon propagator: the random phase approximation (RPA) [29] and the self-consistent MIGDAL approximation [30]. In the RPA scheme the system Green function in the polarization loop (2.61) is only dressed with the electrodes, while in the MIGDAL the full system Green function is considered. Both approximations will be discussed in more detail in chapter 5 of this thesis. It is worth noticing that the approximations discussed up to this point are perturbative and expected to provide accurate results only in the weak to moderate electron-phonon coupling regimes. In the next subsection I will comment about the exact atomic limit of the model, introducing a family of approximations valid in the $\Gamma \ll \lambda, \omega_0$ regime, known as polaron regime.

In the regime of sufficiently small λ/ω_0 or sufficiently detuned system energy ($\tilde{\epsilon}_0 \gg \Gamma$) processes involving several phonons become unlikely and the terms “ $a a$ ” and “ $a^\dagger a^\dagger$ ” can be removed from Eq. (2.60). In this regime, the phonon propagator dressing the electron self-energy in Eq. (2.57) is $\hat{D}_{aa^\dagger}(t, t') = \langle a(t) a^\dagger(t') \rangle$, whose Dyson equation is

$$\hat{D}_{aa^\dagger} = \hat{d}_{aa^\dagger} + \hat{d}_{aa^\dagger} \hat{\Pi} \hat{D}_{aa^\dagger}, \quad (2.62)$$

where time arguments and integration over intermediate times are considered implicitly. This approximation is known in the literature as the rotating wave approximation (RWA), since it consists on removing the stronger oscillating terms in the phonon dynamics.

2.4.3 Polaron-like approximations

The perturbative approximations introduced up to this point for the electron-phonon interaction are expected to be valid in the weak to moderate coupling regime. The problem can be approached from the polaronic side by using the so-called Lang-Firsov transformation [31], which solves exactly the atomic limit ($\Gamma \rightarrow 0$). It predicts the appearance of the polaron quasiparticles, which are formed by dressing the electron with the vibrational degree of freedom. In the following I will derive the exact atomic limit, showing that the tunneling Hamiltonian enters as a perturbation. The unitary Lang-Firsov transformation

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of the Hamiltonian (2.52) is defined as

$$\tilde{H} = e^{-L} H e^L, \quad (2.63)$$

being $L = \lambda/\omega_0 n_d (a^\dagger - a)$. The transformed Hamiltonian is given by

$$\tilde{H} = \tilde{H}_0 + H_{ph}, \quad (2.64)$$

where the interaction effects appear in the system Hamiltonian, described by $\tilde{H}_0 = H_{leads} + \tilde{H}_S + \tilde{H}_T$. The transformed system Hamiltonian is given by $\tilde{H}_S = \tilde{\epsilon} n_d$ with the renormalized level position $\tilde{\epsilon} = \epsilon_0 - \lambda^2/\omega_0$. The many-body effects are encoded in the tunneling Hamiltonian, written as

$$\tilde{H}_T = \sum_{k,\nu} (t_\nu c_{k,\nu}^\dagger X d + \text{H.C.}), \quad (2.65)$$

being $X = \exp[\lambda/\omega_0 (a - a^\dagger)]$ the polaron cloud operators which mixes electronic and vibrational degrees of freedom. It is useful to define the bare polaron propagator as $\hat{\Lambda}(t, t') = \langle X(t) X(t') \rangle$, whose off-diagonal components are given by

$$\Lambda^{+-(-+)}(t, t') = \sum_{k=-\infty}^{\infty} \alpha_k e^{\mp i k \omega_0 (t-t')}. \quad (2.66)$$

The prefactors α_k are the polaron weights given at finite temperature by

$$\alpha_k = e^{-\lambda^2(2n_p+1)/\omega_0^2} I_k \left[2 \frac{\lambda^2}{\omega_0^2} \sqrt{n_p(1+n_p)} \right] e^{k\omega_0/2T}, \quad (2.67)$$

being I_k the modified Bessel function of the first kind. At zero temperature the coefficients are simplified as

$$\alpha_k = \begin{cases} e^{-g^2 \frac{(\lambda/\omega_0)^{2k}}{k!}} & \text{if } k \geq 0 \\ 0 & \text{if } k < 0 \end{cases} \quad (2.68)$$

In the atomic limit the problem is exactly solvable and the retarded Green function is given by $G^r(t, t') = g^r(t, t') \Lambda^r(t, t')$, which in the frequency domain can be written as

$$G_{\Gamma \rightarrow 0}^r(\epsilon) = \sum_{k=-\infty}^{\infty} \frac{\alpha_k n + \alpha_{-k}(1-n)}{\epsilon - \tilde{\epsilon} + k\omega_0 + i\eta}, \quad (2.69)$$

where η is an infinitesimium used to avoid the divergence of the function. This retarded Green function (and thus, the DOS) exhibits resonances at integers values of the phonon frequency and with a weight given by the coefficients α_k . In the $\lambda/\omega_0 \ll 1$ regime the central resonance (α_0) dominates, consistent with the perturbative result. However, when

$\lambda/\omega_0 \gtrsim 1$ the side bands in the spectral density become important, dominating the transport properties in the $\lambda^2/\omega_0\Gamma \gg 1$ regime, where the central peak is exponentially suppressed.

When the coupling to the electrodes become finite, the system phenomenology becomes richer and the problem cannot be exactly solved. There is a series of approximations studying the problem from the polaron transformation. There are two approximations discussed in the literature that provide accurate description in different parameter regime: the polaron tunneling approximation (PTA) [32] and the single particle approximation (SPA) [33]. The PTA describes accurately the low energy spectrum at low bias $V < \omega_0$, recovering the FSR in the electron-hole symmetric case. However it predicts an exponential narrowing of the high energy phonon sidebands which is a pathological behavior of the approach. In contrast, the SPA provides the exact high energy limit ($\tilde{\epsilon}, V, \epsilon_0 \gg \omega_0$) and in the equilibrated phonon regime, although it does not recover the expected FSR in the equilibrium situation. In this subsection I will concentrate, however, on the dressed tunneling approximation (DTA) developed during the thesis [34]. This approach preserves the good low energy features of the PTA and the high energy ones of the SPA, curing their pathological behavior at high and low energies, respectively. Finally, it is worth mentioning that those three approximations do not consider non-equilibrium effects on the phonon dynamics.

The starting point is the perturbation expansion in the transformed Hamiltonian (2.64). The system Green function can be written as

$$\tilde{G}(t, t') = \langle T_c [X(t)d(t)X(t')d^\dagger(t')\tilde{S}] \rangle, \quad (2.70)$$

where the transformed evolution operator is written as $\tilde{S} = \exp[-i \int_c dt \tilde{H}_T(t)]$. Expanding in powers of the tunneling rates, (Γ) it is found

$$\tilde{G}(t, t') = \left\langle T_c [X(t)d(t)X(t')d^\dagger(t')] \left[1 - \int dt_1 \int dt_2 \tilde{H}_T(t_1)\tilde{H}_T t_2 + \dots \right] \right\rangle. \quad (2.71)$$

Note that the first term in the previous equation corresponds to the exact atomic limit, while the higher orders are perturbations due to the coupling to the electrodes. Differently from the previous situations, the Wick's theorem cannot be applied in this simple model due to the fact that the X operators do not obey the usual commutation relations. Note that the X operators contain information about all possible excitations of the polaronic cloud when tunneling events happen. This is illustrated in the top panel of Fig. 2.7 where

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Figure 2.7: Top panel: On the left part I show a second order expansion and its approximation within the DTA scheme. In the bottom panel I show the first diagrams for the DTA approach.

the second order term in the interaction of Eq. (2.71) is schematically represented. The solid and dashed lines represent the system and the electrodes propagators, respectively, and the crosses the tunneling events. The wavy lines are the polaron propagators where lines represented in the upper part represent $\hat{\Lambda}$, while the lower ones denote the $\hat{\Lambda}^{-1}$ inverse propagator. In the regime where the system is weakly coupled to the electrodes ($\Gamma \ll \lambda, \omega_0$), the electrode self-energies are not significantly modified by the presence of the system phonon. In this regime and in the wideband approximation, the retarded electrodes Green function is local in time, i.e. $\Sigma_\nu^r(t, t') \propto \delta(t - t')$ and the time the electrons spend in the electrodes can be considered the smallest timescale. Thus, the intermediate times at the vertex of the leads lines (dashed lines in the figure) can be approximated as similar ($t_1 \approx t_2$, $t_3 \approx t_4, \dots$) and the $\hat{\Lambda}^{-1}(t_i, t_j)$ terms simplify with the $\hat{\Lambda}(t_i, t_{j-1})$. In the top right part of Fig. 2.7 I show the approximation for the second order term, which only describes the possibility of two tunneling events. To include higher order terms in the tunneling amplitudes, the Dyson equation can be used, finding the approximation depicted in the lower panel of Fig. (2.7) where the electron self-energy is dressed with the polaron

$$\tilde{\Sigma}_{T,\nu}^{+-(-+)}(\omega) = \sum_{k=-\infty}^{\infty} \alpha_{\pm k} \Sigma_{T,\nu}^{+-(-+)}(\omega + k\omega_0). \quad (2.72)$$

Figure 2.8: DOS for the DTA approximation (solid lines) for increasing voltage, $V = 5$ (blue), 15 (green) and 45Γ (red). I show the convergence to the PTA result (dotted line) at low voltages and frequencies and to the SPA at high voltages (symbols). The remaining parameters are $\lambda = \omega_0 = 5\Gamma$ and $\tilde{\epsilon} = 0$.

Then, the off-diagonal components of the DTA Green function is given by [34]

$$\tilde{G}_{DTA}^{+-(-+)} = - \sum_{k=-\infty}^{\infty} \alpha_{\pm k} \frac{\tilde{\Sigma}_T^{+-(-+)}(\omega + k\omega_0)}{\tilde{D}(\omega + k\omega_0)}, \quad (2.73)$$

where $\tilde{D}$ is the determinant of the NGF (2.26) dressed with the polaronic cloud

$$\tilde{D} = |\omega - \tilde{\epsilon} - \tilde{\Sigma}_T^r(\omega)|^2. \quad (2.74)$$

It is worth mentioning that a similar result was derived in parallel by means of the equation of motion technique in Ref. [35].

This Green function can be approximated around the phonon satellites finding the approximated-DTA (ADTA) Green function

$$\tilde{G}_{ADTA}(\omega) = \sum_{k=-\infty}^{\infty} \frac{\alpha_k n_d + \alpha_{-k}(1 - n_d)}{\omega + k\omega_0 - \tilde{\epsilon} - \tilde{\Sigma}_T^r(\omega + k\omega_0)}. \quad (2.75)$$

From this expression it becomes clear that the DTA diagrammatic approximation dresses the leads, inducing a broadening of the polaronic resonances equal to $\text{Im}(\tilde{\Sigma}^r)$. At large

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frequencies, this broadening tends to Γ , recovering the SPA limit. Importantly, the width of the central resonance at low bias is exponentially reduced as $e^{-\lambda^2/\omega_0^2}$, recovering the expected limit [36].

In Fig. 2.8 I represent the DOS of the DTA scheme in the electron-hole symmetric situation and for increasing voltage values. For small voltages ($V < \omega_0$) the central peak recovers the FSR. Moreover, note that the height of the higher order sidebands decrease with the order, which is the expected feature according to the exact calculations (see appendix A). For increasing bias voltages the DTA tends to converge to the SPA result, represented by the points in Fig. 2.8, which correctly describes the high energy limit for the equilibrated phonon number situation. Comparison with the exact NRG calculations in the equilibrium situation are provided in the appendix A.

2.5 Superconducting nanojunctions

An important part of the thesis is in the mesoscopic superconductivity framework, considering situations involving superconducting electrodes coupled to regions smaller than the superconducting coherence length. The theory of the transport through these kind of junctions has been developed around the central concept of Andreev reflections: an incident electron in a superconductor is reflected back as a hole with opposite spin and momentum, leading to the transference of a Cooper pair. The multiple reflections at the interface of a superconductor lead to the appearance of the ABSs, due to the superconductivity frustration. These states can lead to a current through the system in the equilibrium situation driven by a phase difference, at variance to the normal metal situations. In the past decades, several mesoscopic superconducting junctions have been studied like the normal electrodes case [37] or the superconducting QPCs [38–42]. Another interesting situations are the ones involving nanoscopic devices coupled to superconducting electrodes, which will be analyzed in this thesis [43, 44].

The situation of a system coupled to two BCS electrodes can be described by the Hamiltonian written in Eq. (2.1), where the Hamiltonian of the electrodes is given by

$$H_{leads} = \sum_{k\sigma,\nu} \epsilon_{k\sigma,\nu} c_{k\sigma,\nu}^\dagger c_{k\sigma} + \sum_{k\sigma,\nu} \Delta_\nu (c_{k\sigma,\nu}^\dagger c_{k\sigma}^\dagger + \text{h.c.}) . \quad (2.76)$$

The first term of the Hamiltonian describes a non-interacting Fermi sea. For simplicity a constant density of states is considered around the Fermi edge, ρ_F . The second term is

the pairing term, which describes the effective attractive interaction between electrons with opposite spins and momenta. The factor $\Delta_\nu = |\Delta_\nu|e^{i\phi_\nu}$ is the superconducting order parameter, where $|\Delta_\nu|$ is the superconducting gap and ϕ_ν the superconducting phase. For simplicity I will consider in the following $|\Delta_L| = |\Delta_R| \equiv \Delta$. It is usually useful to introduce the Nambu spinor representation. Using this notation, the field operators are written as vectors

$$\bar{\Psi}_k = \begin{pmatrix} \hat{c}_{k\uparrow} \\ \hat{c}_{k\downarrow}^\dagger \end{pmatrix} \quad \bar{\Psi}_k^\dagger = (\hat{c}_{k\uparrow}^\dagger \hat{c}_{k\downarrow}), \quad (2.77)$$

and the problem becomes a 2×2 matrix problem. From now on I will use the symbol $\bar{\Psi}$ over the field operators and the Green functions to denote the Nambu structure. In this notation, the Hamiltonian of Eq. (2.1) can be written in a compact way as

$$\bar{H} = \bar{H}_{leads}^{BCS} + \bar{H}_S + \bar{H}_T, \quad (2.78)$$

where

$$\bar{H}_{leads}^{BCS}(\phi) = \sum_{k\nu} \bar{\Psi}_{k\nu}^\dagger \bar{h}_{k\nu} \bar{\Psi}_{k\nu}, \quad (2.79)$$

where $\bar{h}_k = \epsilon_k \bar{\sigma}_z + \Delta \bar{\sigma}_x$, being $\bar{\sigma}_{x,y,z}$ the Pauli matrices in the Nambu space. As in the normal case discussed before, I will consider that the separation between the system energy levels is the largest energy scale and the system is described by a single spin degenerated level

$$\bar{H}_S = \bar{\Psi}_0^\dagger \bar{h}_0 \bar{\Psi}_0, \quad (2.80)$$

with $\bar{h}_0 = \epsilon_0 \bar{\sigma}_z$. Finally, the tunneling Hamiltonian can be written as

$$\bar{H}_T = \sum_{k\nu} t_\nu \bar{\Psi}_{k\nu}^\dagger \bar{h}_t \bar{\Psi}_0, \quad (2.81)$$

where $\bar{h}_t = \bar{\sigma}_z e^{i\phi_\nu \bar{\sigma}_z}$. A gauge transformation has been used to remove the superconducting phase from the leads Hamiltonian (2.79), appearing now in the tunneling part (2.81). This Hamiltonian is 2π periodic in the phase difference, $\phi = \phi_L - \phi_R$. For this reason I will concentrate in the range of $\phi \in [0, 2\pi]$, without loss of generality. As in the normal situations, I will consider the bandwidth to be in such a way that the rates can be defined in a similar way as $\Gamma \approx \pi |t_\nu|^2 \rho_F$. In the limit $\Gamma \gg \Delta, \epsilon_0$ the typical tunneling timescale is the shortest timescale and the influence of the dot in the system dynamics becomes negligible, recovering the QPC result. For this reason, this regime will be referred in the following as the QPC regime. The opposite situation, $\Gamma \lesssim \Delta$, will be referred to as the

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QD regime. Finally, it is also possible to define the normal transmission using Eq. (2.4).

In the Nambu representation, the NGFs can be written in terms of the spinor operators as

$$\hat{\bar{G}}(t, t') = -i \langle T_c \bar{\Psi}(t) \bar{\Psi}^\dagger(t') \rangle. \quad (2.82)$$

Note that, by using this definition, the dimension of the NGFs has been doubled to incorporate the spin degree of freedom, i.e.

$$\hat{\bar{G}}(t, t') = -i \begin{pmatrix} \langle T_c c_\uparrow^\dagger(t) c_\uparrow(t') \rangle & \langle T_c c_\uparrow^\dagger(t) c_\downarrow^\dagger(t') \rangle \\ \langle T_c c_\downarrow(t) c_\uparrow(t') \rangle & \langle T_c c_\downarrow(t) c_\downarrow^\dagger(t') \rangle \end{pmatrix}. \quad (2.83)$$

In the equilibrium situation, the problem becomes significantly simpler, since the Keldysh components can be expressed in terms of the advanced and retarded components. For example, the $+-$ component can be written in the frequency domain as

$$\bar{G}^{+-}(\omega) = f(\omega) [\bar{G}^a(\omega) - \bar{G}^r(\omega)]. \quad (2.84)$$

The retarded/advanced bare Green functions of the superconducting electrodes are given by

$$\bar{g}_\nu^{a,r}(\omega) = \begin{pmatrix} g_\nu^{a,r}(\omega) & -f_\nu^{a,r}(\omega) \\ -f_\nu^{a,r}(\omega) & g_\nu^{a,r}(\omega) \end{pmatrix} \quad (2.85)$$

where

$$f_\nu^{a,r}(\omega) = \Delta / \sqrt{\Delta^2 - (\omega \mp i\eta)^2}, \quad g_\nu^{a,r}(\omega) = -(\omega \mp i\eta)^2 / \sqrt{\Delta^2 - (\omega \mp i\eta)^2}. \quad (2.86)$$

In the equilibrium situation the retarded dot Green function can be written in a particularly simple way in frequency domain as

$$\hat{G}^{a,r}(\omega) = \begin{pmatrix} \omega - \epsilon_0 - \sum_\nu \Gamma_\nu g_\nu^{a,r}(\omega) & \sum_\nu \Gamma_\nu e^{i\phi_\nu} f_\nu^{a,r}(\omega) \\ \sum_\nu \Gamma_\nu e^{-i\phi_\nu} f_\nu^{a,r}(\omega) & \omega + \epsilon_0 - \sum_\nu \Gamma_\nu g_\nu^{a,r}(\omega) \end{pmatrix}^{-1} \quad (2.87)$$

which becomes singular (i.e. $\det(\hat{G}^{a,r})^{-1} = 0$) at energies $\omega = \pm \epsilon_A$. At these energies the ABSs appear due to the MAR. These states are roughly located at

$$\epsilon_A = \pm \tilde{\Delta} \sqrt{1 - \tau \sin^2(\phi/2)}, \quad (2.88)$$

where $\tilde{\Delta}$ is the induced gap, which varies from $\tilde{\Delta} \approx \Gamma$ for $\Gamma \ll \Delta$, to $\tilde{\Delta} \approx \Delta$ for $\Gamma \gg \Delta$ [45]. In Fig. 2.9 I show the dispersion relation for increasing values of Γ at a given transmission factor. At a phase difference $\phi = \pi$ the ABSs get closer to the Fermi level. The gap between the ABS position depend on the normal transmission coefficient as

Figure 2.9: ABSs for $\Gamma/\Delta = 10$ (blue), 4 (green) and 2 (red). The left panel corresponds to a perfect transmitted situation ($\Gamma_L = \Gamma_R$ and $\epsilon_0 = 0$), while the right one to cases with $\epsilon_0 = 1$. The arrow in the right panel is used to mark the gap created between the ABSs in a non-perfect transmitted situation.

$$\Delta_A \sim \Delta \sqrt{1 - \tau}.$$

The current through the ν electrode can be written using the Nambu representation as

$$I_\nu(t) = t_\nu \text{Tr} \left\{ \bar{\sigma}_z \left[\bar{G}_{\nu S}(t, t) - \bar{G}_{S\nu}(t, t) \right] \right\}, \quad (2.89)$$

where the trace is performed in the Nambu space. Using the relation between the keldysh and retarded/advanced components (2.84) the current expression in the stationary regime simplifies as

$$I_\nu = t_\nu \int d\omega f_\nu(\omega) \text{Tr} \left\{ \text{Re} [\bar{G}_{\nu S}^a(\omega) - \bar{G}_{\nu S}^r(\omega)] \right\}. \quad (2.90)$$

The current through these superconducting devices can be divided into two different contributions. Firstly, there is a contribution from the ABSs, which dominate the transport through the system, given by

$$I_{ABS} = -\frac{\partial \epsilon_A(\phi)}{\partial \phi} = \frac{\tau \sin(\phi)}{2\sqrt{1 - \tau \sin^2(\phi/2)}}, \quad (2.91)$$

with a maximum at $\phi \approx \pi$, as shown in the left panel of Fig. 2.10. This term describes the contribution from the Andreev reflections at the interface: transmitted electrodes which are reflected as holes. There is a second contribution coming from the electrodes

Figure 2.10: Current through a mesoscopic system coupled to superconducting electrodes as a function of the phase difference for $\Gamma = \Delta$. In the left panel the current for three different transmission factors are shown: $\tau = 1$ (blue), 0.8 (green) and 0.5 (red). The left panel show the decomposition of the current into the continuum (dashed line) and the ABSs contribution (dotted line).

continuum of states, with opposite sign to I_{ABS} . This contribution vanishes in the QPC and the $\Gamma \rightarrow 0$ limit, and becomes maximal for $\Gamma \approx \Delta$.

2.5.1 AC-Josephson effect

Another situation of interest is the case of a voltage biased superconducting nanojunction. By using a gauge transformation, the problem becomes similar to the phase driven case with a linearly increasing phase difference in time, $\phi(t) = \phi_0 + 2eVt$. This increase gives raise to the ac-Josephson effect, which leads to an oscillating current in time. When $V \ll \Delta$, the ABSs are able to follow adiabatically the time-dependent phase. However, when the bias voltage becomes larger or comparable to the superconducting gap, the ABSs are not able to follow adiabatically their expected position in time and the system exhibits dissipation due to the generation o quasiparticles above the superconducting gap.

Formally, by introducing the effect of the bias voltage in the superconducting phase, the problem becomes time-dependent. In the stationary state the problem can be treated

Figure 2.11: Current through a superconducting QPC for different normal transmission factors $\tau = 1, 0.9, 0.7, 0.5, 0.3$ and 0.1 from top to bottom. In the bottom panel I show the dominant transport process. At high voltages $V > 2\Delta$ the single quasiparticle process dominate (right panel) while at lower voltages the Andreev reflections (middle panel) and MAR (left panel) become dominant.

in terms of the fundamental frequency, $\omega_V = 2V$, and the current can be expanded as

$$I = \sum_{k=-\infty}^{\infty} e^{ik\omega_V} I_k. \quad (2.92)$$

The solution of the problem involves the calculation of the components of the Green functions dependent on two frequencies, $G(j\omega_V, k\omega_V)$. It can be demonstrated that the problem becomes formally equivalent to a tight-binding problem where the j, k components represent the sites in the chain (see Refs. [40, 46] for details).

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The current-voltage relation is plotted in the main panel of Fig. 2.11 in the QPC regime ($\Gamma/\Delta \rightarrow \infty$) for different transmission factors. For low transmission factors a series of kinks are observed in the current at $V = 2\Delta/n$, being n a natural number. When increasing transmission factor, these features become smoother, disappearing in the perfect transmission situation. These features are related to the MAR at the interface. At $V = 2\Delta$ a jump in the current is observed due to the quasiparticle tunneling: an electron of the left lead can be promoted from the states below the gap to states in the right lead above the gap. This process dominates the current at high voltages. At $V = \Delta$ an Andreev reflection takes place: an electron is reflected back as a hole (represented as dotted line in the bottom middle panel of Fig. 2.11). This process takes only place when the energy of the reflected hole is such that it can reach the available states above the superconducting gap ($V \geq \Delta$). The Andreev reflections dominate the current for voltages $\Delta \leq V < 2\Delta$. In a more general situation, for $V = 2\Delta/n$ the MAR take place at the interface of the two superconductors, leading to the transference of n electrons from the left to the right electrode. The signatures observed at low voltages are related to the opening of these conduction channels when moving from low to high voltages. The subgap structure has been measured in experiments involving few channels in Ref. [41].

2.6 Full counting statistics

A complete understanding of quantum transport at the nanoscale implies the analysis of, not only the mean quantities like the mean current, charge occupation or the DOS, but also of the system fluctuations. Quantum measurements at the nanoscale exhibit fluctuations even if they are performed under the same conditions. Some sources of randomness may be originated from imperfectness of a sample or factors beyond the experimental control. The more important sources of randomness are the ones related to the quantum measurement. An example of this last type is the number of electrons transferred through a tunnel junction during a time t , which may vary from one measurement to another even under the same conditions. Then, the number of electrons transferred through the junction n can be considered as a random variable, $P_n(t)$ being the probability distribution of detecting n electrons after a time t which fulfills the normalization condition $\sum_n P_n(t) = 1$. The study of this probability distribution started as a theoretical research in the 1990s decade [47–51]. More recently some experimental studies appeared studying the electron transferred statistics through nanodevices [52–58]. These experiments make

use of a quantum point contact close to the system, whose current is sensitive to the number of electrons in the device. These schemes allows to detect single electron events, addressing the single electron probabilities in the sequential tunneling regime. From an experimental point of view, it becomes challenging to address a more general regime involving electron coherent effects.

In the stationary regime, where the system conditions does not change in time, the long time statistics can be obtained by performing a measurement during a long enough time. However, in a more general situation it becomes necessary to resolve the charge statistics in time by performing several independent measurements and averaging over them. Along this thesis I will consider this second kind of measurement protocol, which makes possible to study the system fluctuations dynamics. The mean number of electrons transferred until a time t can be expressed in terms of the probabilities as

$$\langle c(t) \rangle = \sum_n n P_n(t). \quad (2.93)$$

The second cumulant of the distribution, which measures the width of the distribution (also know as charge noise) is determined by

$$\langle c^2(t) \rangle = \langle c^2(t) \rangle - \langle c(t) \rangle^2 = \sum_n n^2 P_n(t) - \left[\sum_n n P_n(t) \right]^2. \quad (2.94)$$

In general the cumulants can be obtained by a recursion formula

$$\langle c^j(t) \rangle = c^j(t) - \sum_{k=1}^{j-1} \binom{j-1}{k-1} c^k(t) \langle c^{j-k}(t) \rangle, \quad (2.95)$$

where $c^j(t) = \sum_n n^j P_n(t)$ are the moments of the distribution. The higher order cumulants describe the higher momenta distribution. It becomes convenient to define the characteristic function as

$$Z(\chi, t) = \sum_n P_n(t) e^{i n \chi}, \quad (2.96)$$

which contains a complete information about the possible tunneling events. The variable χ is the so-called counting field which plays a fundamental role in the theory and whose physical meaning will be commented below. The function given in Eq. (2.96) is also known GF and all the cumulants can be obtained by successive derivatives as

$$\langle c^n(t) \rangle = \frac{\partial^n}{\partial (i\chi)^n} S(\chi, t) \Big|_{\chi=0}, \quad (2.97)$$

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where the function

$$S(\chi, t) = \text{Log}[Z(\chi, t)] \quad (2.98)$$

is usually known as cumulant generating function (CGF). The instantaneous current cumulants are then defined as $\langle I^n(t) \rangle = \partial \langle c^n(t) \rangle / \partial t$. Thus, the main problem constitutes on the determination of the system GF, which is in general a difficult task.

The problem of quantum measurement is in general complex since it may require the description of the detector-system coupling. In this thesis I am analyzing the simplest case of an ideal detector to the system. This detector is considered to provide instantaneous measurements without affecting the electron dynamics. Moreover, these measurements are projective: the system wavefunction is fully projected onto a base with a well defined number of particles. This kind of detector is equivalent to include phase factors to the tunneling Hamiltonian as [49, 51]

$$H_{T,\chi} = \sum_{k,\nu} \left[t_{\nu,k} c_{k\nu}^\dagger d e^{i\chi_\nu/2} + t_{\nu,k} d^\dagger c_{k\nu} e^{-i\chi_\nu/2} \right]. \quad (2.99)$$

Using this simple definition, when an electron tunnels from the ν electron to the system it induces an extra phase $e^{i\chi_\nu}$. With this method it becomes possible to study the charge and current statistics resolved in spin for each electrode. Note that in the stationary regime the statistics on the left and right part are similar for conserving approximations. For this reason, in the following of this chapter I will concentrate on the left tunneling statistics without loss of generality (i.e. $\chi_R = 0$ and $\chi_L \equiv \chi$).

Finally, I would like to remark other possible choices of the counting field. Firstly, the symmetrized current can be addressed by choosing $\chi_L = -\chi_R = \chi/2$. Another interesting possibility is to take $\chi_L = \chi_R = \chi/2$ which can be used to study the system population statistics. A final relevant example is to take $\chi_\nu = \chi \epsilon_{k\nu}$, which can be used to address the energy statistics at the ν interface [59]. It has been shown that in the time domain this is equivalent to a time shift in the off-diagonal Keldysh components of the self-energy [60, 61].

2.6.1 Non-interacting system

Firstly, I will analyze the case of a non-interacting nanojunction, described by the Hamiltonian given in Eq. (2.1), with the tunneling term described by Eq. (2.99). In this system, electron tunnel events are independent, except for the Pauli principle, which avoids two

electron occupying the same energy state at the same time. The GF is then given by the mean value of the counting field dependent evolution operator [62]

$$Z(\chi, t) = \langle T_c e^{\int_c dt_1 H_{T,\chi}(t_1)} \rangle. \quad (2.100)$$

This very simple case can be integrated exactly [63] reaching the central result of this subsection

$$S(\chi, t) = \int_0^t dt_1 \text{Log} \{ \det [\hat{g}_S^{-1} - \hat{\Sigma}_{T,\chi}] \}, \quad (2.101)$$

where the determinant is a functional determinant on the system NGFs dressed with the counting field dependent tunneling Hamiltonian part, described in Eq. (2.99). In the stationary regime the CGF can be written in frequency domain, finding

$$S(\chi, t) = t \int \frac{d\omega}{2\pi} \text{Log} \{ \det [\hat{g}_S^{-1}(\omega) - \hat{\Sigma}_{T,\chi}(\omega)] \}. \quad (2.102)$$

The components of the counting field dependent tunneling self-energy in Keldysh space can be written as [49, 51]

$$\Sigma_{T,\chi}^{+-}(\omega) = e^{i\chi/2} \Sigma_T^{+-}(\omega), \quad \Sigma_{T,\chi}^{-+}(\omega) = e^{-i\chi/2} \Sigma_T^{-+}(\omega), \quad (2.103)$$

while the Keldysh diagonal components, $\Sigma_{T,\chi}^{++}(\omega) = \Sigma_T^{++}(\omega)$ and $\Sigma_{T,\chi}^{--}(\omega) = \Sigma_T^{--}(\omega)$, remain unchanged. Expanding the determinant and using the equilibrium expressions for the bare dot Green function and the tunneling self-energy, I arrive to the well-known Levitov formula [47]

$$S(\chi, t) = t \int \frac{d\omega}{2\pi} \text{Log} \{ 1 + \tau(\omega) [f_R(f_L - 1)(e^{i\chi/2} - 1) + f_L(f_R - 1)(e^{-i\chi/2} - 1)] \}. \quad (2.104)$$

This expression describes the charge fluctuations of a non-interacting junction (2.1), characterized by a single parameter: the energy-dependent transmission probability, $\tau(\omega) = 4\Gamma_L\Gamma_R/[(\omega - \epsilon_0)^2 + \Gamma^2]$. A factor 2 is usually included in Eq. (2.104) to account from the spin degeneracy. This expression can be extended to many channels in the situation where charge fluctuations in each of the channels are independent. In that case, the CGF will be a sum of contributions of the form of Eq. (2.104). The interpretation of Eq. (2.104) becomes simpler in the $T = 0$ case and sufficiently small voltage ($V \ll \Gamma$). In such a situation $\tau(\omega)$ can be considered energy independent and the CGF simplifies as

$$S(\chi, t) = \frac{Vt}{2\pi} \text{Log} \{ 1 + \tau(e^{-i\chi} - 1) \}, \quad (2.105)$$

where the prefactor sets the maximum number of electrons that can be transfer in a time interval t . After the projective measurement the maximum of transferred charges

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is an integer N . Applying the Newton's binomial theorem in Eq. (2.105) it is possible to write the GF as

$$Z(\chi, t) = [1 + \tau(e^{i\chi} - 1)]^N = \sum_{n=0}^N \binom{N}{n} \tau^n (1 - \tau)^{N-n} e^{in\chi}, \quad (2.106)$$

which leads to a binomial distribution for the number of tunnel electrons with probabilities $P_n = \binom{N}{n} \tau^n (1 - \tau)^{N-n}$. This distribution tends to the Poisson one in the limit of low transmission coefficient, $\tau \ll 1$. In this limit the probabilities simplify as

$$P_n \approx \frac{(t\Gamma)^n}{n!} e^{-t\Gamma}. \quad (2.107)$$

This same result is obtained when considering the number of particles transfer through a potential barrier, neglecting the effects from the Pauli principle. In this situation the CGF gets a simple expression $S(\chi, t) = t\Gamma(e^{i\chi} - 1)$, which predicts that all the current cumulants are identical and equal to $\langle I^n \rangle = \Gamma$.

In a more general case, the expressions for the cumulants can be obtained differentiating Eq. (2.104). For instance, the current can be written as

$$\langle I(t) \rangle = \int \frac{d\omega}{2\pi} \tau(\omega) [f_R(\omega) - f_L(\omega)], \quad (2.108)$$

which is the Meir-Wingreen expression (2.35), obtained for the expectation value for the current operator. The noise can be obtained deriving twice with respect to the counting field, finding

$$\begin{aligned} \langle I^2(t) \rangle = \int \frac{d\omega}{2\pi} \tau(\omega) \{ & f_L(\omega) [1 - f_L(\omega)] + f_R(\omega) [1 - f_R(\omega)] + \\ & [1 - \tau(\omega)] [f_L(\omega) - f_R(\omega)]^2 \}. \end{aligned} \quad (2.109)$$

This expression has two different contributions. The first one, written in the top part of Eq. (2.109), describes the thermal fluctuations of the electrons at the interface, also known as Johnson-Nyquist noise. Its expression at zero voltage is given by $\langle I^2(t) \rangle = 4TG$ (being G the conductance), consistent with the fluctuation-dissipation theorem. The second term written in the lower part of Eq. (2.109) describes a rather different situation. It is originated by the discreteness of the charge carriers and becomes non-zero when a bias voltage is applied. This source of noise is known as shot noise. In the regime where $T \ll V \ll \Gamma$ the expression can be considered energy-independent, finding $\langle I^2(t) \rangle = V\tau(1 - \tau)/2\pi$. It becomes useful to introduce the Fano factor as

$$F = \langle I^2(t) \rangle / \langle I(t) \rangle, \quad (2.110)$$

which provides a measurement of the effective charge transfer. In the Poissonian limit described above where the probability distribution is described by Eq. (2.107), $F = 1$. In the absence of interactions, this is the maximum value the Fano factor can take. In the low bias limit, the Fano factor is given by $F = \tau(1 - \tau)/\tau$, which describes a reduction of the shot noise in $1 - \tau$ due to the Pauli exclusion principle [64]. Note that the Fano factor tends to zero for a perfect transmitted junction while it maximizes for $\tau \rightarrow 0$.

2.6.2 Interaction effects

In general the calculation of the GF in interacting devices is a formidable task, which has not been solved in general. It is important to note that the counting field not only appears in the tunneling self-energy, but it generally appears also in the interaction self-energy (2.42), since it depends on the system NGFs. Even though if the counting field dependent self-energy is known, a naive inclusion in Eq. (2.101) leads to an overcounting of diagrams, leading to a wrong description of the cumulants. As discussed in Ref. [51], it is more convenient to use the counting field dependent current, deriving it to compute the higher order current cumulants. The counting field dependent current can be written as

$$\langle I(t, \chi) \rangle = \frac{\partial Z(t, \chi)}{\partial(i\chi)} = \left\langle T_c \frac{\partial H_{T,\chi}}{\partial(i\chi)} \exp \left\{ -i \int_c dt_1 H_{T,\chi}(t_1) \right\} \right\rangle. \quad (2.111)$$

In terms of the Green functions, it can be simplified as

$$\langle I(t, \chi) \rangle = \int \frac{d\omega}{2\pi} \tau_\chi(\omega) \left[e^{-i\chi} \Sigma_{T,L}^{-+}(\omega) \Sigma_{T,R}^{+-}(\omega) - e^{i\chi} \Sigma_{T,R}^{-+}(\omega) \Sigma_{T,L}^{+-}(\omega) \right], \quad (2.112)$$

being $\tau_\chi(\omega)$ the determinant of the counting field dependent system Green function, written as

$$\tau_\chi(\omega) = \det \left[\hat{g}_S^{-1} - \hat{\Sigma}_{T,\chi} - \hat{\Sigma}_{int,\chi} \right]^{-1}. \quad (2.113)$$

In the zero counting field limit, Eq. (2.112) recovers the Meir-Wingreen expression (2.35) as expected. The CGF could be in general obtained by integrating Eq. (2.112), finding

$$S(t, \chi) = \int d\chi \langle I(t, \chi) \rangle. \quad (2.114)$$

This integral is non-analytic in a general situation, existing only few examples of approximations where the GF can be integrated in the presence of interactions [29, 35, 51]. From the expression (2.112) it becomes easier to determine the higher order cumulants

2. Theoretical framework in the stationary regime

by successive derivatives with respect to the counting field. For instance, the shot noise can be written as

$$\begin{aligned} \langle I^2 \rangle = & \int \frac{d\omega}{2\pi} \tau(\omega) \left\{ f_L(\omega) [1 - f_L(\omega)] + f_R(\omega) [1 - f_R(\omega)] + [1 - \tau(\omega)] [f_L(\omega) - f_R(\omega)]^2 \right. \\ & \left. + \tau(\omega) [f_L(\omega) - f_R(\omega)]^2 + \frac{\partial \tau_\chi}{\partial(i\chi)} \Big|_{\chi=0} [f_L(\omega) - f_R(\omega)] \right\}. \end{aligned} \quad (2.115)$$

The two terms appearing in the top part of the previous equation are similar to the ones in Eq. (2.109). Note that the first terms, written in the upper part, describe the thermal noise and have a similar functional form as in the non-interacting situation, given by Eq. (2.109), with a renormalized transmission factor due to interaction effects. For this reason, it becomes difficult from an experimental point of view to detect interactions through the equilibrium noise. The terms written in the lower part of Eq. (2.115) describe the shot noise, being the first term the non-interacting contribution. The interaction contribution to the shot noise is given by the last term of Eq. (2.115), where the many-body effects are introduced through the counting field derivative of the τ_χ function.

2.6.3 Factorial cumulants

The higher order cumulants contain a more complete information about the charge transport characteristics. However, its interpretation becomes more challenging since these quantities exhibit strong oscillations in any system parameter. These oscillations turn out to be universal in the absence of interactions [57, 65–67], uncovering all the important information about the electron dynamics. For this reason, the factorial and generalized factorial cumulants have been introduced. They are generalizations of the cumulants measured at finite counting field [68, 69] in the complex $z = e^{i\chi}$ plane

$$\langle c_F^n(z, t) \rangle = \frac{\partial^n}{\partial(i\chi)^n} S(\chi, t) \Big|_z, \quad (2.116)$$

and the factorial current cumulants are defined as $\langle I_F^n(z, t) \rangle = \partial_t \langle c_F^n(z, t) \rangle$. Note that for $z = 1$ the regular cumulants are recovered. It is worth mentioning that a slightly different definition of the factorial cumulants have been given in Ref. [69], which leads to an additional $1/z^n$ prefactor for $c_n^F(z, t)$. It can be shown that the main properties of the factorial cumulants discussed in those references are preserved under the somewhat simpler expression given in Eq. (2.116).

It becomes possible to define also a factorial generating function (FGF) as [70, 71]

$$S_F(z, t) = \frac{\sum_n P_n(t)(z + s)^n}{\sum_n P_N(t)s^n}, \quad (2.117)$$

where s is a biasing field used to move the measurement point from $z = 1$ to $z = 1 + s$. The biasing field can be in general complex, leading to non-Hermitian Hamiltonians. The most analyzed quantities are the factorial cumulants, corresponding to measure at the $z = 0$ point. At this point, all the cumulants of a Poisson probability distribution (2.107) are equal to zero. Thus, $C_F^n(z = 0, t)$ measures deviations from the Poissonian distribution, at variance to the regular cumulants which measures deviations from the Gaussian one. It has been shown that those quantities do not exhibit the universal oscillations mentioned before. Moreover, in the absence of interactions (described by the generalized binomial distribution) their sign is fixed, independently from the parameters [69]. For this reason, they have been suggested for detecting and characterizing electron interactions and correlations at the nanoscale. To understand the generalized factorial cumulants it becomes more convenient to analyze the singular points of the CGF and to discuss their relation to the phase transition phenomena. Finally I would like to comment that, in the following, both the factorial cumulants (measured at $z = 0$) and the generalized factorial cumulants ($z \neq 0, 1$) will be referred as factorial cumulants for simplicity, specifying in each case the measurement point.

2.6.4 Dynamical Yang-Lee zeros

An alternative point of view is provided by the zeros of the GF (2.96), which are logarithmic singularities of the CGF (2.98). It can be demonstrated that those singularities contain a complete information about the transport properties, while providing further insight about the electron interaction effects. At a given time, the maximum number of electrons transfer in each of the directions is bounded. The bound has been commented in the context of a non-interacting device in Eq. (2.105), but similar expressions could be derived for interacting interactions. Thus, the sum of the GF can be truncated between n_{min} and n_{max} and written by performing an index change as

$$Z(z, t) = \left(\frac{1}{z}\right)^{n_{min}} \sum_{n=0}^{n_T} P_n(t) z^n, \quad (2.118)$$

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being $n_T = n_{max} - n_{min}$ the degree of the polynomial determined by the GF. Equivalently, the previous equation can be written as a function of its roots, $z_j(t)$, as

$$Z(z, t) = \left(\frac{1}{z}\right)^{n_{min}} \prod_{j=1}^{n_T} [z - z_j(t)] , \quad (2.119)$$

and the CGF can be expressed as the sum of the binomials appearing in the previous equation. As it will be shown in the next chapter, the prefactor to the product does not contribute to the current cumulants. Thus, the transport properties can be written as a function of $\{z_j\}$. An approximate relation between the high order cumulants ($\langle c^n \rangle$ with $n \gtrsim 6$) and the zeros of the GF has been given in Ref. [69]. That expression has been used to determine experimentally the zeros of the Andreev tunneling events in a non-coherent Josephson junction [72]. When approaching the stationary regime ($t \rightarrow \infty$), the singularities of the CGF arrange forming branches in the complex counting field plane. In that situation the roots of the GF are in principle not isolated and the sum of Eq. (2.118) cannot be truncated.

The importance of understanding the position of the roots of the GF was pointed out some time ago. In the first works analyzing these singular points [73, 74], the authors showed that they lie in the negative real axis for a generalized binomial distribution, i.e. in the absence of interactions. However, when electron correlations take place, the image becomes richer and deviations from this behavior can be found. Some works reporting the appearance of zeros in the complex counting field plane are given in Refs. [29, 69].

Importantly, the FCS has been related to the statistical mechanics theory in the equilibrium situation. For instance, the dynamical trajectories can be related to the microstates in the equilibrium situation. These trajectories are characterized by the activity parameter n , which is related to the number of events during the time span $[0, t]$. Interestingly this analogy is rather general and has been applied to describe a wide range of problems such as the Ising model [75], the glass transition [76] or the quantum jumps between different states [77, 78]. In the set of examples that will be considered along the thesis involving quantum transport through nanoscale systems, the activity is related to the number of tunneling events. The GF can be related to the partition function (with the CGF as the free energy) and the time plays the role of the system volume in equilibrium statistical mechanics. To complete the analogy, the complex exponential of the counting field can be related to the fugacity, which depends on the temperature. By changing the counting field, the system is biased from the common ($z = 1$) to the uncommon ($z \neq 1$)

trajectories, driving eventually the system to a non-equilibrium phase transition. The non-equilibrium phase transition phenomena manifests itself as a discontinuity in the factorial cumulants, measured at finite counting field [78].

The link between the two ingredients, the roots of the GF and the non-equilibrium phase transitions, can be done by extending the works of Refs. [79, 80] developed in equilibrium statistical mechanics to the trajectories ensemble. In these original works, the authors demonstrated that two different phases are separated from each other by a line of zeros of the partition function (known as Yang-Lee zeros). Moreover, at the phase coexistence point, the density of each phase is determined by the density of zeros close to the real fugacity axis in the thermodynamical limit. The order of the phase transition can be determined by the angle at which the zeros intercept the real axis [81, 82]. Similarly, it has been shown that two non-equilibrium phases are separated by a line of zeros of the GF, named as Dynamical Yang-Lee zeros (DYLZs) by analogy. Their position has been also related to the intermittency and phase coexistence phenomena, as described in Refs. [78, 83].

Bibliography

- [1] D. C. Langreth. Friedel sum rule for Anderson's model of localized impurity states. *Phys. Rev.* **150**, 516 (1966).
- [2] L. V. Keldysh. Diagram technique for nonequilibrium processes. *Zh. Eksp. Teor. Fiz.* **47**, 1515 (1964). [*Sov. Phys. JETP*20,1018(1965)].
- [3] Y. Meir and N. S. Wingreen. Landauer formula for the current through an interacting electron region. *Phys. Rev. Lett.* **68**, 2512 (1992).
- [4] R. Bulla, T. A. Costi, and T. Pruschke. Numerical renormalization group method for quantum impurity systems. *Rev. Mod. Phys.* **80**, 395 (2008).
- [5] E. Gull, A. J. Millis, A. I. Lichtenstein, A. N. Rubtsov, M. Troyer, and P. Werner. Continuous-time monte Carlo methods for quantum impurity models. *Rev. Mod. Phys.* **83**, 349 (2011).
- [6] R. Haussmann. *Self-consistent Quantum-Field Theory and Bosonization for Strongly Correlated Electron Systems*. Lecture Notes in Physics Monographs (Springer, 1999).
- [7] E. Kozik, M. Ferrero, and A. Georges. Nonexistence of the Luttinger-Ward functional

BIBLIOGRAPHY

- and misleading convergence of skeleton diagrammatic series for Hubbard-like models. Phys. Rev. Lett. **114**, 156402 (2015).
- [8] P. W. Anderson. Localized magnetic states in metals. Phys. Rev. **124**, 41 (1961).
- [9] A. Hewson. *The Kondo Problem to Heavy Fermions*. Cambridge Studies in Magnetism (Cambridge University Press, 1997).
- [10] P. B. Wiegmann. Towards an exact solution of the Anderson model. Phys. Lett. **80A**, 163 (1980).
- [11] N. Kawakami and A. Okiji. Exact expression of the ground-state energy for the symmetric Anderson model. Physics Letters A **86**, 483 (1981).
- [12] N. Andrei, K. Furuya, and J. H. Lowenstein. Solution of the Kondo problem. Rev. Mod. Phys. **55**, 331 (1983).
- [13] A. Martín-Rodero, F. Flores, M. Baldo, and R. Pucci. A new solution to the Anderson-Newns hamiltonian of chemisorption. Solid State Communications **44**, 911 (1982).
- [14] J. A. White. Self-consistent Green functions for the Anderson impurity model. Phys. Rev. B **45**, 1100 (1992).
- [15] A. Martín-Rodero, E. Louis, F. Flores, and C. Tejedor. Interpolative solution for the periodic Anderson model of mixed-valence compounds. Phys. Rev. B **33**, 1814 (1986).
- [16] A. Martín-Rodero, F. Flores, M. Baldo, and R. Pucci. A new solution to the Anderson-Newns hamiltonian of chemisorption. Solid State Communications **44**, 911 (1982).
- [17] A. L. Yeyati, A. Martín-Rodero, and F. Flores. Electron correlation resonances in the transport through a single quantum level. Phys. Rev. Lett. **71**, 2991 (1993).
- [18] H. Kajueter and G. Kotliar. New iterative perturbation scheme for lattice models with arbitrary filling. Phys. Rev. Lett. **77**, 131 (1996).
- [19] F. B. Anders. A numerical renormalization group approach to non-equilibrium Green functions for quantum impurity models. Journal of Physics: Condensed Matter **20**, 195216 (2008).
- [20] T. Fujii and K. Ueda. Perturbative approach to the nonequilibrium Kondo effect in a

- quantum dot. *Phys. Rev. B* **68**, 155310 (2003).
- [21] N. Agraït, C. Untiedt, G. Rubio-Bollinger, and S. Vieira. Onset of energy dissipation in ballistic atomic wires. *Phys. Rev. Lett.* **88**, 216803 (2002).
 - [22] L. de la Vega, A. Martín-Rodero, N. Agraït, and A. L. Yeyati. Universal features of electron-phonon interactions in atomic wires. *Phys. Rev. B* **73**, 075428 (2006).
 - [23] R. Leturcq, C. Stampfer, K. Inderbitzin, L. Durrer, C. Hierold, E. Mariani, M. G. Schultz, F. von Oppen, and K. Ensslin. Franck-Condon blockade in suspended carbon nanotube quantum dots. *Nature Physics* **5**, 327 EP (2009).
 - [24] H. Park, J. Park, A. K. L. Lim, E. H. Anderson, A. P. Alivisatos, and P. L. McEuen. Nanomechanical oscillations in a single-C60 transistor. *Nature* **407**, 57 EP (2000).
 - [25] T. Holstein. Studies of polaron motion: Part i. the molecular-crystal model. *Annals of Physics* **8**, 325 (1959).
 - [26] A. Mitra, I. Aleiner, and A. J. Millis. Phonon effects in molecular transistors: Quantal and classical treatment. *Phys. Rev. B* **69**, 245302 (2004).
 - [27] R.-P. Riwar and T. L. Schmidt. Transient dynamics of a molecular quantum dot with a vibrational degree of freedom. *Phys. Rev. B* **80**, 125109 (2009).
 - [28] D. F. Urban, R. Avriller, and A. Levy Yeyati. Nonlinear effects of phonon fluctuations on transport through nanoscale junctions. *Phys. Rev. B* **82**, 121414 (2010).
 - [29] Y. Utsumi, O. Entin-Wohlman, A. Ueda, and A. Aharony. Full-counting statistics for molecular junctions: Fluctuation theorem and singularities. *Phys. Rev. B* **87**, 115407 (2013).
 - [30] Y. Murakami, P. Werner, N. Tsuji, and H. Aoki. Interaction quench in the Holstein model: Thermalization crossover from electron- to phonon-dominated relaxation. *Phys. Rev. B* **91**, 045128 (2015).
 - [31] I. G. Lang and Y. A. Firsov. Kinetic theory of semiconductors with low mobility. *Sov. Phys. JETP* **16**, 1301 (1962).
 - [32] S. Maier, T. L. Schmidt, and A. Komnik. Charge transfer statistics of a molecular quantum dot with strong electron-phonon interaction. *Phys. Rev. B* **83**, 085401 (2011).
 - [33] K. Flensberg. Tunneling broadening of vibrational sidebands in molecular transistors.

BIBLIOGRAPHY

- Phys. Rev. B **68**, 205323 (2003).
- [34] R. Seoane Souto, A. L. Yeyati, A. Martín-Rodero, and R. C. Monreal. Dressed tunneling approximation for electronic transport through molecular transistors. Phys. Rev. B **89**, 085412 (2014).
- [35] B. Dong, G. H. Ding, and X. L. Lei. Full counting statistics of a single-molecule quantum dot. Phys. Rev. B **88**, 075414 (2013).
- [36] R. C. Monreal and A. Martín-Rodero. Equation of motion approach to the Anderson-Holstein hamiltonian. Phys. Rev. B **79**, 115140 (2009).
- [37] G. E. Blonder, M. Tinkham, and T. M. Klapwijk. Transition from metallic to tunneling regimes in superconducting microconstrictions: excess current, charge imbalance, and supercurrent conversion. Phys. Rev. B **25**, 4515 (1982).
- [38] C. W. J. Beenakker and H. van Houten. Josephson current through a superconducting quantum point contact shorter than the coherence length. Phys. Rev. Lett. **66**, 3056 (1991).
- [39] M. Hurd, S. Datta, and P. F. Bagwell. Current-voltage relation for asymmetric ballistic superconducting junctions. Phys. Rev. B **54**, 6557 (1996).
- [40] J. C. Cuevas, A. Martín-Rodero, and A. L. Yeyati. Hamiltonian approach to the transport properties of superconducting quantum point contacts. Phys. Rev. B **54**, 7366 (1996).
- [41] E. Scheer, P. Joyez, D. Esteve, C. Urbina, and M. H. Devoret. Conduction channel transmissions of atomic-size aluminum contacts. Phys. Rev. Lett. **78**, 3535 (1997).
- [42] E. Scheer, W. Belzig, Y. Naveh, M. H. Devoret, D. Esteve, and C. Urbina. Proximity effect and multiple Andreev reflections in gold atomic contacts. Phys. Rev. Lett. **86**, 284 (2001).
- [43] A. L. Yeyati, J. C. Cuevas, A. López-Dávalos, and A. Martín-Rodero. Resonant tunneling through a small quantum dot coupled to superconducting leads. Phys. Rev. B **55**, R6137 (1997).
- [44] S. De Franceschi, L. Kouwenhoven, C. Schönenberger, and W. Wernsdorfer. Hybrid superconductor-quantum dot devices. Nature Nanotechnology **5**, 703 EP (2010). Review Article.

- [45] A. Martín-Rodero and A. L. Yeyati. Josephson and Andreev transport through quantum dots. *Advances in Physics* **60**, 899 (2011).
- [46] J. C. Cuevas. Electronic transport in normal and superconducting nanocontacts. Doctoral thesis (1999).
- [47] L. S. Levitov and G. B. Lesovik. Charge distribution in quantum shot noise. *JETP Lett.* **58**, 230 (1993).
- [48] L. S. Levitov, H. Lee, and G. B. Lesovik. Electron counting statistics and coherent states of electric current. *Journal of Mathematical Physics* **37**, 4845 (1996).
- [49] Y. V. Nazarov. Universalities of weak localization. *Ann. Phys. (Leipzig)* **8**, 507 (1999).
- [50] D. A. Bagrets and Y. V. Nazarov. Full counting statistics of charge transfer in Coulomb blockade systems. *Phys. Rev. B* **67**, 085316 (2003).
- [51] A. O. Gogolin and A. Komnik. Towards full counting statistics for the Anderson impurity model. *Phys. Rev. B* **73**, 195301 (2006).
- [52] T. Fujisawa, T. Hayashi, R. Tomita, and Y. Hirayama. Bidirectional counting of single electrons. *Science* **312**, 1634 (2006).
- [53] S. Gustavsson, R. Leturcq, B. Simović, R. Schleser, T. Ihn, P. Studerus, K. Ensslin, D. C. Driscoll, and A. C. Gossard. Counting statistics of single electron transport in a quantum dot. *Phys. Rev. Lett.* **96**, 076605 (2006).
- [54] S. Gustavsson, R. Leturcq, B. Simović, R. Schleser, P. Studerus, T. Ihn, K. Ensslin, D. C. Driscoll, and A. C. Gossard. Counting statistics and super-Poissonian noise in a quantum dot: Time-resolved measurements of electron transport. *Phys. Rev. B* **74**, 195305 (2006).
- [55] E. V. Sukhorukov, A. N. Jordan, S. Gustavsson, R. Leturcq, T. Ihn, and K. Ensslin. Conditional statistics of electron transport in interacting nanoscale conductors. *Nature Physics* **3**, 243 EP (2007).
- [56] C. Fricke, F. Hohls, W. Wegscheider, and R. J. Haug. Bimodal counting statistics in single-electron tunneling through a quantum dot. *Phys. Rev. B* **76**, 155307 (2007).
- [57] C. Flindt, C. Fricke, F. Hohls, T. Novotný, K. Netočný, T. Brandes, and R. J. Haug. Universal oscillations in counting statistics. *Proceedings of the National Academy of Sciences* **106**, 10116 (2009).

BIBLIOGRAPHY

- [58] T. Wagner, P. Strasberg, J. C. Bayer, E. P. Rugeramigabo, T. Brandes, and R. J. Haug. Strong suppression of shot noise in a feedback-controlled single-electron transistor. *Nature Nanotechnology* **12**, 218 EP (2016).
- [59] B. K. Agarwalla, J.-H. Jiang, and D. Segal. Full counting statistics of vibrationally assisted electronic conduction: transport and fluctuations of thermoelectric efficiency. *Phys. Rev. B* **92**, 245418 (2015).
- [60] Z. Yu, G.-M. Tang, and J. Wang. Full-counting statistics of transient energy current in mesoscopic systems. *Phys. Rev. B* **93**, 195419 (2016).
- [61] G. Tang, Z. Yu, and J. Wang. Full-counting statistics of energy transport of molecular junctions in the polaronic regime. *New Journal of Physics* **19**, 083007 (2017).
- [62] L. S. Levitov and M. Reznikov. Counting statistics of tunneling current. *Phys. Rev. B* **70**, 115305 (2004).
- [63] A. Kamenev. *Field Theory of Non-Equilibrium Systems* (Cambridge University Press, 2011).
- [64] C. W. J. Beenakker and C. Schonenberger. Quantum shot noise. *Physics today* **56**, 37 (2003).
- [65] C. Flindt, T. Novotný, A. Braggio, M. Sassetti, and A.-P. Jauho. Counting statistics of non-Markovian quantum stochastic processes. *Phys. Rev. Lett.* **100**, 150601 (2008).
- [66] D. S. Golubev, M. Marthaler, Y. Utsumi, and G. Schön. Statistics of voltage fluctuations in resistively shunted Josephson junctions. *Phys. Rev. B* **81**, 184516 (2010).
- [67] C. Fricke, F. Hohls, N. Sethubalasubramanian, L. Fricke, and R. J. Haug. High-order cumulants in the counting statistics of asymmetric quantum dots. *Applied Physics Letters* **96**, 202103 (2010).
- [68] C. W. J. Beenakker and H. Schomerus. Antibunched photons emitted by a quantum point contact out of equilibrium. *Phys. Rev. Lett.* **93**, 096801 (2004).
- [69] D. Kambly, C. Flindt, and M. Büttiker. Factorial cumulants reveal interactions in counting statistics. *Phys. Rev. B* **83**, 075432 (2011).
- [70] P. Stegmann, B. Sothmann, A. Hucht, and J. König. Detection of interactions via generalized factorial cumulants in systems in and out of equilibrium. *Phys. Rev. B* **92**, 155413 (2015).

- [71] P. Stegmann and J. König. Inverse counting statistics based on generalized factorial cumulants. *New Journal of Physics* **19**, 023018 (2017).
- [72] K. Brandner, V. F. Maisi, J. P. Pekola, J. P. Garrahan, and C. Flindt. Experimental determination of dynamical Lee-Yang zeros. *Phys. Rev. Lett.* **118**, 180601 (2017).
- [73] A. G. Abanov and D. A. Ivanov. Allowed charge transfers between coherent conductors driven by a time-dependent scatterer. *Phys. Rev. Lett.* **100**, 086602 (2008).
- [74] A. G. Abanov and D. A. Ivanov. Factorization of quantum charge transport for noninteracting fermions. *Phys. Rev. B* **79**, 205315 (2009).
- [75] J. M. Hickey, C. Flindt, and J. P. Garrahan. Trajectory phase transitions and dynamical Lee-Yang zeros of the Glauber-Ising chain. *Phys. Rev. E* **88**, 012119 (2013).
- [76] L. O. Hedges, R. L. Jack, J. P. Garrahan, and D. Chandler. Dynamic order-disorder in atomistic models of structural glass formers. *Science* **323**, 1309 (2009).
- [77] J. P. Garrahan, A. D. Armour, and I. Lesanovsky. Quantum trajectory phase transitions in the micromaser. *Phys. Rev. E* **84**, 021115 (2011).
- [78] J. M. Hickey, C. Flindt, and J. P. Garrahan. Intermittency and dynamical Lee-Yang zeros of open quantum systems. *Phys. Rev. E* **90**, 062128 (2014).
- [79] C. N. Yang and T. D. Lee. Statistical theory of equations of state and phase transitions. i. theory of condensation. *Phys. Rev.* **87**, 404 (1952).
- [80] T. D. Lee and C. N. Yang. Statistical theory of equations of state and phase transitions. ii. lattice gas and ising model. *Phys. Rev.* **87**, 410 (1952).
- [81] R. A. Blythe and M. R. Evans. Lee-Yang zeros and phase transitions in nonequilibrium steady states. *Phys. Rev. Lett.* **89**, 080601 (2002).
- [82] R. A. Blythe. An introduction to phase transitions in stochastic dynamical systems. *Journal of Physics: Conference Series* **40**, 1 (2006).
- [83] C. Flindt and J. P. Garrahan. Trajectory phase transitions, Lee-Yang zeros, and high-order cumulants in full counting statistics. *Phys. Rev. Lett.* **110**, 050601 (2013).

Part I.

Transient dynamics in normal nanojunctions

3 Transient dynamics in non-interacting junctions

3.1 Introduction

The analysis of the charge transferred statistics can provide a more detailed information about electron correlation and interaction effects than the mean current itself [1, 2]. It involves the measurement of the probability of detecting a given number of charges in a time interval. While most of the studies have been restricted to the stationary regime, where the probability distribution is determined by measuring over long time periods, the time-dependent situation has been much less analyzed. The determination of the time resolved statistics requires to perform several independent measurements at times shorter than the characteristic system relaxation time.

Recently, some works have analyzed the time resolved transport statistics, focusing on the charge cumulants [3–5] and the factorial cumulants [6] in the incoherent regime. However, the coherent regime has been much less investigated [7–9]. The existence of an universal short time behavior for the higher order cumulants has been reported in both incoherent [4] and coherent [10] regimes.

The goal of this chapter is to introduce the basic ideas about the time-dependent quantum transport through a mesoscopic system. By considering the simplest non-interacting situation, the analytic and the numerical methods are introduced. I consider that the system is suddenly coupled to electronic reservoirs (quench of the tunneling amplitudes). This problem has already been analyzed for the single particle observables (current and

3. Transient dynamics in non-interacting junctions

system population) in Refs. [11, 12] and for the charge statistics in [8, 9]. In the first part of the chapter, the methods introduced in those references are reviewed.

In section 3.4 the exact relation between the zeros of the GF and the charge and current cumulants is presented, derived in Ref. [13], which constitutes the most important result of this chapter. Firstly, an exact expression will be used to study the short time transport, involving a single tunneling event. In this regime, the charge and current cumulants exhibit universal oscillations of increasing amplitude with increasing order. These oscillations are found to be in agreement with the experimental measurements from Ref. [4]. As it will be commented below, the oscillations in the many electron regime tend to be damped due to interference effects, leading to the system relaxation into the steady state.

Moreover, the role of coherence is analyzed. I find that, in the unidirectional coherent regime a second set of the cumulant oscillations become visible due to an increase in the delay of the tunneling events. Finally, the short time bidirectional transport will be analyzed, finding that the amplitude of the universal oscillations is renormalized due to interference effects of the contributions to the cumulants of the electrons transmitted in opposite directions.

3.2 Mean properties

I begin the chapter by studying the time evolution of a system described by Eq. (2.1). This simple example will provide information about the typical relaxation times of the system, allowing to introduce the analytic approximations that will be used to study the dynamics of interacting nanojunctions under simple approaches such as a mean-field approximation or the DTA. I consider the situation in which the electrodes are suddenly coupled to the system (quench), where the tunneling Hamiltonian is described by

$$\hat{H}_T(t) = \theta(t) \sum_{k\sigma,\nu} \left(t_{k\sigma,\nu} \hat{c}_{k\sigma,\nu}^\dagger \hat{d}_\sigma + t_{k\sigma,\nu}^* \hat{d}_\sigma^\dagger \hat{c}_{k\sigma,\nu} \right). \quad (3.1)$$

For simplicity the spin degree of freedom will be omitted in the following of this chapter. This problem has already been analyzed some time ago in the Refs. [11, 12], where situations involving single particle potentials are also studied. In this chapter I will restrict the discussion to the case without any external potential. The solution to the problem is provided by the time-dependent Dyson equation which can be transformed to the triangular representation [14]. Thus, the advanced and retarded components of

the central system Green function are given by

$$G_S^{a,r}(t, t') = g_S^{a,r}(t - t') + \int dt_1 \int dt_2 g_S^{a,r}(t - t_1) \Sigma_T^{a,r}(t_1 - t_2) G_S^{a,r}(t_2, t'). \quad (3.2)$$

The solution to this integral equation is in general non-analytic in the presence of interactions and a numerical treatment becomes necessary. The way of numerically solving this problem by means of a discretization procedure will be discussed in the following section. In the case of a non-interacting nanojunction and a quench of the tunneling amplitudes, Eq. (3.2) can be written as

$$G_S^{a,r}(t, t') = g_S^{a,r}(t, t') + \int_0^t dt_2 K^{a,r}(t - t_2) G_S^{a,r}(t_2, t'), \quad (3.3)$$

where the kernel $K^{a,r}(t - t_2) = \int dt_1 g_S^{a,r}(t - t_1) \Sigma_T^{a,r}(t_1 - t_2)$ becomes time translational invariant, i.e. depending only on the time difference. This property is of fundamental interest since it allows to use the Laplace transform methods for solving the integral equation. Eq. (3.3) can be written in Laplace space as

$$G_S^{a,r}(s) = g_S^{a,r}(s) + K^{a,r}(s) G_S^{a,r}(s), \quad (3.4)$$

with

$$K^{a,r}(s) = \frac{-\Gamma}{s \pm i\epsilon_0}. \quad (3.5)$$

Thus, the solution to the Dyson equation in the Laplace space (3.4) has a compact expression given by

$$G_S^{a,r}(s) = \frac{g_S^{a,r}(s)}{1 - K^{a,r}(s)}, \quad (3.6)$$

and the Green function in time domain can be obtained by taking the inverse Laplace transform of the previous expression. The inverse Laplace transform involves the integration of Eq. (3.6) along the Bromwich contour, containing all the poles of the function. Thus the advanced and retarded Green functions are given by

$$G_S^{a,r}(t - t') = \theta(t - t') \sum_j \text{Res}(\xi_j^{a,r}) e^{\xi_j^{a,r}(t - t')}, \quad (3.7)$$

$\xi_j^{a,r}$ being the poles of Eq. (3.6) and $\text{Res}(\xi_j^{a,r})$ their residues. This expression may become more difficult in the case where Eq. (3.6) presents other kind of singularities such as branchpoints or branchcuts, situations not considered in this thesis. It is worth mentioning that the retarded Green function is expressed as a sum of contributions of each of its poles, without any mixing term. In the case of interacting nanojunctions Eq. (3.7)

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becomes approximately valid and will be denoted as the *single pole approximation*. In the non-interacting situation analyzed along this chapter, Eq. (3.6) has a single pole $\xi^{a,r} = -\Gamma \pm i\epsilon_0$ with a residue equal to the unit. Then, the Keldysh components of the NGF can be obtained through the equation of motion

$$G_S^{+-} = (1 + G_S^r \Sigma_T^r) g_S^{+-} (1 + \Sigma_T^a G_S^a) + G_S^r \Sigma_T^{+-} G_S^a, \quad (3.8)$$

where integration over intermediate times is considered implicitly. Integrating G_S^{+-} it can be found

$$\begin{aligned} G_S^{+-}(t, t') &= i e^{\epsilon_0(t-t')} e^{\Gamma(t+t')} \left\{ n(0) + \frac{\Gamma}{2\pi} \right. \\ &\times \int d\omega [f_L(\omega) + f_R(\omega)] \frac{[e^{(\Gamma+i\epsilon_0-i\omega)t} - 1][e^{(\Gamma-i\epsilon_0+i\omega)t} - 1]}{\Gamma^2 + (\omega - \epsilon_0)^2} \Big\}. \end{aligned} \quad (3.9)$$

A similar expression can be obtained for the $G_S^{-+}(t, t')$ component. The central level charge can be obtained from the previous component as $n(t) = i G_S^{+-}(t, t)$, which has a very compact expression

$$n(t) = n(0) e^{-\Gamma t} + \frac{\Gamma}{2\pi} \int d\omega [f_L(\omega) + f_R(\omega)] \frac{1 + e^{-2\Gamma t} - 2e^{\Gamma t} \cos(\omega - \epsilon_0)t}{\Gamma^2 + (\omega - \epsilon_0)^2}, \quad (3.10)$$

and predicts that the initial configuration $n(0)$ relaxes in times of the order of $t \sim 1/\Gamma$. Analytic integration can be obtained if a finite bandwidth (3.17) or temperature (3.18) is considered. The current could be obtained from Eq. (2.33) in the time domain, which leads to the expression

$$I_\nu(t) = \int_{-\infty}^{\infty} dt_1 \left\{ G_S^r(t-t_1) \Sigma_{T,\nu}^{+-}(t_1-t) - \Sigma_{T,\nu}^{+-}(t_1-t) G_S^a(t_1-t) \right\}. \quad (3.11)$$

In the situation where the system is symmetrically coupled to both electrodes ($\Gamma_L = \Gamma_R = \Gamma/2$), the symmetrized current gets a very simple expression

$$\langle I(t) \rangle = 2\Gamma \theta(t) \text{Re} \int_0^\infty dt_1 \left\{ G_S^r(t-t_1) [f_L(t_1-t) - f_R(t_1-t)] \right\}, \quad (3.12)$$

where $f_\nu(t_1-t)$ is the Fourier transform of the electrodes DOS, described by the Fermi distribution function. This expression constitutes a generalization of the Meir-Wingreen expression given in Eq. (2.35) to the time domain. Note that Eq. (3.12) is also valid in the interacting situation, with the difference that, in that case, the retarded and advanced Green functions may depend on both time arguments in a non-trivial way (not only on the time difference).

3.3 Full counting statistics

The problem of the charge and current cumulants dynamics is more involved since it requires to solve in the time domain the statistics of the charges transfer. The solution of this problem could be obtained by inverting the counting field dependent Dyson equation in the time domain

$$\hat{G}_{S,\chi}(t, t') = [\hat{g}_S^{-1} - \hat{\Sigma}_{T,\chi}]^{-1}, \quad (3.13)$$

where the time dependence of $\hat{g}_S^{-1}$ and $\hat{\Sigma}_{T,\chi}$ is not included explicitly in the preceding expression, since $\hat{G}_{S,\chi}$ may depend on all the previous times. Moreover, the tunneling self-energy contains the information of the counting field as described in Eq. (2.103). At variance with the previously discussed situation with no counting field, it is not possible to convert the matrix equation to a triangular one by performing the Keldysh rotation [14] and the method discussed in the previous section. In other words, the counting field dependent retarded and advanced components may depend on the two time arguments in a non-trivial way. Thus, an analytic expression for the counting field dependent system NGFs in the time domain remains unknown, even in the non-interacting case.

3.3.1 Discretized Dyson equation and the determinant formula

Instead of trying to obtain an analytic approximation to the problem, I am going to use a numerical approach to address the time evolution of the charge and current cumulants. For this purpose, it becomes necessary to discretize the Keldysh contour as depicted in the Fig. 3.1. Thus, the Dyson equation can be written as

$$\hat{\mathbf{G}}_{S,\chi} = [\hat{\mathbf{g}}_S^{-1} - (\Delta t)^2 \hat{\mathbf{\Sigma}}_{T,\chi}]^{-1}, \quad (3.14)$$

where the discretized matrix propagators and self-energies will be denoted in boldtype from now on. The inverse bare system Green function discretized on the contour becomes a $2N \times 2N$ matrix (N being the number of times in the mesh and the factor 2 comes from the Keldysh structure, see Fig. 3.1). The expression of the inverse system NGF is

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Figure 3.1: Discretized Keldysh contour where χ is the counting field, changing sign in each of the branches and Δt is the time step.

derived in detail in Ref. [14], finding

$$i\mathbf{g}_S^{-1} = \left(\begin{array}{ccc|ccc} -1 & & & & & -\rho \\ h_- & -1 & & & & \\ & h_- & -1 & & & \\ & & \ddots & \ddots & & \\ & & & 1 & -1 & \\ \hline & & & & h_+ & -1 \\ & & & & & \ddots \\ & & & & & h_+ & -1 \end{array} \right)_{2N \times 2N}. \quad (3.15)$$

where $h_{\pm} = 1 \mp i\epsilon_0\Delta t$ and $\Delta t = t/N$ is the time step in the discretization. It is worth noticing that this expression corresponds to a discretization of the $i\partial_t - \epsilon_0$ operator in the time contour, evolving from the initial to the final time through the positive Keldysh branch and returning back through the negative one, as described in Fig. (3.1). The lines in the matrix of Eq. (3.15) are used to separate the four different Keldysh components $(i\mathbf{g}_S^{-1})^{\alpha\beta}$ ($\alpha, \beta = \pm$), where the constant element in the $-+$ component (bottom left box) corresponds to the closing of the Keldysh contour. In Eq. (3.15) the initial level charge is determined by $n(0) = \rho/(1 + \rho)$. For an initial equilibrium situation $\rho = e^{-\epsilon_0/T}$.

The discretized tunneling self-energy can be evaluated straightforwardly through the Fourier transform of the tunneling self-energy

$$\begin{aligned} \Sigma_{T,\chi}^{+-}(t-t') &= 2i \sum_{\nu=L,R} \Gamma_{\nu} e^{i\chi_{\nu}} f_{\nu}(t, t'), \\ \Sigma_{T,\chi}^{-+}(t-t') &= 2i \sum_{\nu=L,R} \Gamma_{\nu} e^{-i\chi_{\nu}} [f_{\nu}(t, t') - \delta(t-t')], \end{aligned} \quad (3.16)$$

where χ_{ν} is the counting field in each of the electrodes. In the following I will consider $\chi_R = 0$ and $\chi_L = \chi$ unless stated differently, thus the transport properties at the left

interface will be analyzed. The function $f_\nu(t, t')$ is the Fourier transform of the Fermi function, which has a very simple expression in the zero temperature limit if a finite bandwidth D is considered

$$f_\nu(t, t') = \frac{i}{2\pi} \frac{e^{-i\mu_\nu(t-t')} - e^{iD(t-t')}}{t - t'}. \quad (3.17)$$

In the infinite bandwidth limit the Fourier transform of the Fermi function can be also obtained analytically by considering a small temperature and expanding it in Matsubara frequencies

$$f_\nu(t, t') = i \sum_{n=1}^{\infty} R_n e^{-i\mu_n u(t-t')} e^{-\omega_n |t-t'|} + \frac{\delta(t-t')}{2}, \quad (3.18)$$

where $R_n = T$ are the residues and $\omega_n = (2n+1)\pi T$ the Matsubara frequencies. The convergence of the sum can be significantly improved by using the approximate residues and frequencies proposed in Ref. [15]. The Dirac delta functional appearing in Eqs. (3.16) and (3.18) is converted to a Kronecker delta, including a $1/\Delta t$ prefactor when discretizing in the time mesh. For the diagonal ones the general Keldysh relations can be used

$$\begin{aligned} \Sigma_{T,\chi}^{++}(t, t') &= -\theta(t-t') \Sigma_{T,\chi=0}^{+-}(t, t') - \theta(t'-t) \Sigma_{T,\chi=0}^{+-}(t, t') \\ \Sigma_{T,\chi}^{--}(t, t') &= -\theta(t-t') \Sigma_{T,\chi=0}^{+-}(t, t') - \theta(t'-t) \Sigma_{T,\chi=0}^{+-}(t, t'). \end{aligned} \quad (3.19)$$

Notice that there is an ambiguity in the determination of these self-energies at equal times, where the Heaviside function is not well defined. The stability and the convergence to the correct time evolution depends strongly on the choice of the equal times criteria for $\Sigma_{T,\chi}^{++}(t, t)$ and $\Sigma_{T,\chi}^{--}(t, t)$. The most stable criteria found corresponds to the choice

$$\Sigma_{T,\chi}^{++}(t, t) = \Sigma_{T,\chi}^{--}(t, t) = -\frac{\Sigma_{T,\chi}^{+-}(t, t) + \Sigma_{T,\chi}^{+-}(t, t)}{2}, \quad (3.20)$$

corresponding to define $\theta(0) = 1/2$, at variance to the one for the bare system Green function (3.15).

Interestingly, in this simple example, the GF can be integrated using Grassmann variables [16] or diagrammatic arguments. As a function of the discretized Green functions and self-energies it can be written as

$$Z(\chi, t) = \det [\hat{\mathbf{G}}_{S,\chi}^{-1} \hat{\mathbf{G}}_{S,\chi=0}] = \frac{\det [\hat{\mathbf{g}}_S^{-1} - (\Delta t)^2 \hat{\Sigma}_{T,\chi}]}{\det [\hat{\mathbf{g}}_S^{-1} - (\Delta t)^2 \hat{\Sigma}_{T,\chi=0}]}, \quad (3.21)$$

where the element in the denominator in the right hand term is used to ensure the GF normalization at zero counting field. This expression converges with good accuracy when

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Δt is the smallest system timescale (typically $\Delta t \lesssim 1/D$). In the wideband limit where the discretized self-energy is obtained from Eq. (3.18) we found out that the convergence is usually achieved for $\Delta t \lesssim 1/(10\Gamma)$.

Finally I would like to comment that there could be a way of obtaining a semi-analytic expression for Eq. (3.21) by means of the Toeplitz theory applied to blocked matrices. In appendix A.2 I introduce an approximation based on the single box Toeplitz theory, which provides the appropriate long time limit and the transient in the uni-directional regime $V \gg \Gamma$. Finally, it is worth mentioning that a numerical approximation based on the work of Ref. [17] could be used to improve the performance of the algorithm.

3.4 Universal relation between cumulants and zeros

In this section I derive an exact relation between the zeros of the GF and the charge and current cumulants. As mentioned at the end of the previous chapter, these zeros contain a complete information about the transport properties. Moreover, their position can be related to the non-equilibrium phase transition phenomenon through an analogy to the equilibrium statistical mechanics and the works in Refs. [18, 19] (see subsection 1.5.2 for a more detailed discussion). The importance of these zeros has already been pointed out in the literature in Refs. [20–22]. From Eq. (2.119) the CGF can be written as

$$S(\chi, t) = -n_{min} \log(z) + \sum_{n=1}^{n_T} \log(z - z_n), \quad (3.22)$$

where $z = e^{i\chi}$ and the cumulants can be obtained by successive derivatives of this expression (2.97). In this chapter I will concentrate on the non-interacting case, where the z_n are real non-positive numbers [23, 24]. Some situations deviating from this behavior have been found for interacting nanojunctions in Refs. [6, 20, 25–28]. In that situation the probability of n charges flowing in both directions are related through the fluctuation dissipation theorem as $P_{-n}(t) = P_n(t)e^{nV/T}$, due to thermal agitation. Thus, $z = -e^{V/T}$ separates processes in the positive and negative direction [26]. From the expression (3.22) it becomes clear that the contribution from each zero of the GF is independent from the position of the remaining ones. Taking derivatives on Eq. (3.22), I find that each of the terms contribute to the j charge cumulant as

$$\left. \frac{\partial^j}{\partial \chi^j} \log(z - z_n) \right|_{z=1} = - \sum_{k=1}^{\infty} \frac{k^{j-1}}{z_n^k}. \quad (3.23)$$

This leads to a very compact expression for the charge cumulants [13]

$$c_L^j(t) = - \sum_{n=1}^{n_T} \text{Li}_{1-j} \left[\frac{1}{z_n(t)} \right], \quad (3.24)$$

where Li_{1-j} denotes the polylogarithm function of order $1-j$ [29]. Similar expressions for the right or the symmetrized cumulants can be obtained by taking a different choice of the counting field (see discussion of section 2.6). The expression (3.24) constitutes the central result of this chapter which is valid also for interacting nanojunctions, where the zeros appear as complex conjugated pairs. It is worth noticing that the contributions of the zeros at the coordinates origin ($z = 0$), and written in the first term of Eq. (3.22), vanish for all charge and current cumulants, except for the mean charge. The first polylogarithms have a very simple expression as a function of the zero distribution and the first charge cumulants can be written as

$$c_L(t) = - \sum_n \frac{1}{1 - z_n} - n_{\min}, c_L^2(t) = - \sum_n \frac{z_n}{(1 - z_n)^2}, \quad (3.25)$$

which are related to the mean charge and the charge noise. From the first equality in Eq. (3.25) it becomes clear that the zero closest to the measurement point ($z = 1$) provides the greatest contribution to the charge transferred. For this reason, this zero (z_1) will be denoted in the following as the *dominant* zero. This singular point will be of importance for understanding the short time dynamics as it will be commented in the following section of this chapter. The first equality in Eq. (3.25) also describes that a charge is fully transferred in the positive direction when a zero moves from $z \rightarrow -\infty$ to the $z = 0$ point and in the negative direction when moving from $z = 0$ to $z \rightarrow \infty$. At finite temperature the $\{z_n\}$ describing processes in both directions are separated by a gap in their distribution at $z = -e^{\beta V}$ due to the fluctuation theorem [26].

The charge noise is described by the second expression in Eq. (3.25). The contribution to the charge noise of each zero vanishes for a complete transmitted electron ($z \rightarrow 0, -\infty$), becoming maximum for the non-interacting situation at $z = -1$ when half an electron is transferred. Similar expressions can be obtained for the higher order cumulants by using the general properties of the polylogarithmic functions $\text{Li}_{j-1}(z) = z \partial_z \text{Li}_j(z)$, although they become less transparent. For the higher order cumulants, the exact expression given in Eq. (3.24) is well approximated by [20, 30]

$$c_L^j(t) \approx (-1)^{j-1} (j-1)! \sum_n \frac{2 \cos \{j \arg [z_n(t) - 1]\}}{|z_n(t) - 1|^j}, \quad (3.26)$$

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valid for $j \gtrsim 6$. This expression predicts the appearance of oscillations in the non-interacting situation with a maximum roughly located at $z = -1$ and decaying for $z \rightarrow 0, -\infty$. The maximum amplitude of the oscillation can be obtained from the previous expression showing that it scales with the cumulant order as

$$\max[c_L^j(t)] \sim (j-1)! \pi^{(-j+1/2)}. \quad (3.27)$$

From Eq. (3.24) it becomes also possible to obtain compact expressions for the current cumulants using the general properties of the polylogarithms as

$$I_L^j(t) = - \sum_{n=1}^{n_T} \frac{z'_n(t)}{z_n(t)} \text{Li}_{-j} \left[\frac{1}{z_n(t)} \right], \quad (3.28)$$

which depends on the position of the zero and its time derivative, $z'_n(t) = \partial_t z_n(t)$.

Finally, compact expressions can be also obtained for the factorial cumulants [6, 20, 31] by simply shifting the measurement point by a quantity s . The FGF can be written as

$$Z_F(z, t; s) = \frac{\sum_n P_n (z+s)^n}{\sum_n s^n P_n}, \quad (3.29)$$

where s is the biasing field and the denominator is a normalization factor, giving no contribution to the transport properties. One can also define the zeros of the FGF, which are just the original $z_j(t)$ shifted by s . Thus, the factorial cumulants are given by

$$c_{F,L}^j(t; s) = - \sum_j \text{Li}_{1-n} \left[\frac{1}{z_j(t) + s} \right]. \quad (3.30)$$

The factorial current cumulants (I_F^n) can be then computed by deriving Eq. (3.30) with respect to the time, finding a similar expression to Eq. (3.28) but replacing the zeros by the displaced zeros.

3.5 Analysis of the short time universality

In this section I study the short time transient transport properties, comparing numerical and analytic results. I start from the simplest situation of a system suddenly coupled to a single electrode, analyzing afterwards the role of coherence and directionality in open quantum systems.

Figure 3.2: Transfer charge cumulants comparing numerical results obtained by evaluating the GF in the discretized Keldysh contour through Eq. (3.21) (full lines) with the analytic expression of Eq. (3.24) (dotted lines) for the system suddenly connected to a single electrode. The position of the zero is given by Eq. (3.33), with $z_{long-t} = 0$. The model parameters are $\Gamma_L = 0.5$, $\Gamma_R = 0$, $\mu_L = 3$, $\epsilon = 0$, $T = 0.1$ and the system is initially empty.

3.5.1 Single electrode junction

I first discuss the short time transport between a mesoscopic system and a single electrode. Although all the time averaged current cumulants vanish at long times in this simple example, it is illustrative for understanding the short time system dynamics. I consider that the system is prepared with a given initial configuration, $n(0) = 0$ or $n(0) = 1$ (note that the spin degree of freedom has been omitted for simplicity), and suddenly connected to the electronic reservoir. Then a current can flow between the system and the electrode, which tends to relax the initial condition. The GF gets a simple expression since only 2 processes are possible

$$Z(\chi, t) \simeq P_0(t) + P_{\pm 1}(t)e^{\pm i\chi}. \quad (3.31)$$

The sign $\pm$ denote the direction of the transport determined by the initial population in the system: positive for initially empty (transport from the electrode to the system), and negative in the opposite case. Then, the charge cumulants can be obtained as derivatives

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Figure 3.3: Current cumulants for the same case as in Fig. 3.2, comparing the analytic results (dotted line) given in Eq. (3.34) with the numerical calculations (full lines).

of the CGF as

$$c_L^j(t) = \left(\frac{\partial}{\partial i\chi} \right)^j \log \left[1 + \frac{1}{z_1} e^{\pm i\chi} \right] \Big|_{\chi=0}, \quad (3.32)$$

where $z_1 = [-P_0(t)/P_{\pm 1}(t)]^{\pm 1}$ is the only root of Eq. (3.31). I have found that the zero is well approximated by

$$z_1 \approx -\frac{1}{(1 - e^{-\Gamma_L t})e^{2\Gamma_L t}} + z_{long-t}, \quad (3.33)$$

where z_{long-t} is the long time position of the zero, related to the asymptotic system charge. In Fig. 3.2 the charge cumulants for increasing orders (from $j = 4$ to 12) are represented comparing the analytic result from Eq. (3.24) with the numerical simulations performed using Eq. (3.21). A remarkable agreement is found using a single fitting parameter (z_{long-t}) for all order cumulants. This constitutes a proof of the validity of Eq. (3.24). As it is shown, the charge cumulants of increasing orders exhibit stronger oscillations, originated by the first electron transfer. The same behavior has been observed experimentally in Ref. [4] in the sequential regime ($V \gg T \gg \Gamma$). Although those measurements were performed in the stationary regime (leads connected in the remote past), the oscillations in that experiment are signatures of the first electron crossing after

the events recording started. At longer times, the oscillations in Fig. 3.2 decay due to the relaxation of the initial condition and the approach to the equilibrium situation.

In this example it is also possible to deduce an universal expression for the current cumulants, normalized with the mean current as

$$\frac{I_L^j(t)}{I_L(t)} = \frac{(z_1 - 1)^2}{z_1} \text{Li}_{-j}(1/z_1), \quad (3.34)$$

where the dependence on the derivative of the zero position appearing in Eq. (3.28) simplifies. In Fig. 3.3 the comparison between the numerical calculations for the normalized current cumulants and Eq. (3.34) is shown, with the single zero (z_1) approximated by (3.33), finding a perfect agreement at short times.

3.5.2 Two electrodes junction: Coherent effects

In the previous subsection I have addressed the unidirectional transport between a mesoscopic system and a single electrode, showing that the short time universal oscillations are due to the first electron passage. However, there are a few remaining questions to be answered still in the non-interacting situation. The first one is about the long time relaxation of oscillations, since each electron transmitted through the junction provides an oscillating contribution, according to Eq. (3.26). The second question arising is about the role of coherence on the system.

For addressing those questions I consider that the system is connected to two electrodes at time ($t = 0$). For simplicity I will discuss first the unidirectional situation ($V \gg \Gamma, T$), analyzing two different regimes: the coherent ($\Gamma \gg T$) and the sequential ($T \gg \Gamma$) regime. In the top panel of Fig. 3.4 some of the high order cumulants in the sequential regime are shown, which exhibit the universal oscillations at short times (shadowed region). The dotted line shows the analytic result where the dominant zero is approximated by Eq. (3.33), in good agreement with the numerical calculations. The oscillation in the charge cumulants progressively deviates from the universality at longer times, relaxing finally for $t \gg 1/\Gamma$.

In the left bottom panel of Fig. 3.4 the position of the zeros of the GF in the negative real axis is represented as a function of the time. The dominant zero is shown in black, corresponding the dotted line to the approximate expression given by Eq. (3.33). As can

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Figure 3.4: In the upper panel I represent the high order charge cumulants, c_L^{10} , c_L^{11} and c_L^{12} in the sequential regime comparing the numerical results (solid lines) and the analytic ones, considering the contribution from a single zero (discontinuous line) approximated by Eq. (3.33) with $z_{long-t} \approx -0.4$. In the lower panel the time evolution of the dominant zeros is presented, together with the polylogarithm $\text{Li}_{11}(1/z)$ (related to the cumulant c_L^{12}). At short time, there is a single dominant zero (gray line) entering the strongly oscillating region, which leads to the short time universal behavior (shadowed region in the upper panel), broken when more zeros approach $z = -1$. The model parameters are $V = 40$, $T = 6$, $\epsilon_0 = 0$, $\Gamma_L = \Gamma_R = 0.5$ and $n(0) = 0$.

be observed, the charge oscillations appear in the regime where the dominant zero (describing the first electron tunneling event) approaches the $z \sim -1$ point, where the mean charge transfer is fractional according to Eq. (3.25). This is illustrated by the shadow in the plot, which qualitatively represents the height of the polylogarithm Li_{-11} , related to the charge cumulant c_L^{12} and represented in the bottom right panel of Fig. 3.4. At longer times more zeros approach the strongly oscillating region ($z \sim -1$), leading to a destructive interference of the oscillations and the relaxation to the steady state. There is also a small probability of detecting a mean charge flowing in the opposite direction to the voltage bias, described by the zero represented in blue in the bottom left panel of Fig. 3.4 due to thermal agitation. Finally, it is worth noticing that the times relaxation for all the cumulants are similar and determined by the zeros dynamics.

Figure 3.5: Upper panel: charge cumulants c_L^{10} , c_L^{11} and c_L^{12} comparing numerical results (solid lines) with the analytic one (dotted lines) from Eq. (3.24). For the analytic result two zeros are considered, being the first one described by Eq. (3.33) with $z_{long-t} = -0.1$, and the second one by (3.35) with $\alpha_{2,long-t} = -0.15$. At very short times, the universal oscillations are seen (black shadowed region), while at intermediate times a second set of oscillations appear (yellow shadowed region). In the lower panels I represent the zeros of the GF (left) determined numerically (full lines) together with approximated ones (dotted lines). The polylogarithm $\text{Li}_{11}(1/z)$ is represented in the bottom right panel. The model parameters are $V = 6$, $T = 0.1$, $\epsilon_0 = 0$, $\Gamma_L = \Gamma_R = 0.5$ and $n(0) = 0$.

The result for the high order charge cumulants in the coherent regime ($T \ll \Gamma$) is represented in Fig. 3.5. Similarly to the previous case, at short times the universal oscillations are observed (black shadowed region). At variance to the sequential regime, another set of extra oscillations appear at longer times ($t \sim \Gamma/\Gamma_L\Gamma_R$). The dotted line represents the analytic result from Eq. (3.24) in good agreement with the numerical results. In this case two zeros have to be included to reproduce the two sets of oscillations: the dominant one given by Eq. (3.33) while the second one is well described as

$$z_2 \approx -\frac{1}{(1 - e^{-\tilde{\Gamma}(t-\Delta\tau)})e^{2\tilde{\Gamma}t}} + z_{2,long-t}, \quad (3.35)$$

with an effective rate $\tilde{\Gamma} = \Gamma_L^2\Gamma_R/\Gamma^2$, which reflects the fact that the second zero is dominated by third order tunneling processes at short times. It is related to the process

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Figure 3.6: Upper panel: high order charge cumulants (full line), showing their deviation with respect to the universal behavior (dotted lines) given by Eq. (3.24) due to the short time bidirectional transport. The corresponding parameters are $V = 0$, $\Gamma_L = \Gamma_R = 0.5$, $\epsilon_0 = 0$, $T = 0.1$ and the system is initially empty. In the lower panel the amplitude of the oscillations are shown for the universal case (red dots), exhibiting an universal scaling given by Eq. (3.27), and for the bidirectional transport (blue squares), where the universal scaling is broken.

where the charge of the system is relaxed through the right electrode and charged again from the left one. A delay time has also been included, given by the characteristic time evolution of the first zero, $\Delta\tau \approx 1/2\Gamma_L$. Differently from the sequential regime studied before, the distance between the first and the second zero has increased (see the lower panel of Fig. 3.5), avoiding them to interfere and producing the second set of oscillations. However, they are not completely independent and their contributions can overlap, leading to a breaking of the universality (change on the oscillations amplitude) of the second set of oscillations.

3.5.3 Bidirectional transport

In this subsection I analyze the situation when the transport at short time is not unidirectional, but electrons are allowed to tunnel in both directions of the junction at short

times. This kind of situation is found for an initially occupied system or for $V \lesssim \Gamma$. In this section I consider the limiting case when $V \approx 0$, where the effects are more pronounced. In the upper panel of Fig. 3.6 I show the higher order cumulants, comparing the numerical results with the analytic ones from Eq. (3.24) considering a single zero described by Eq. (3.33). At very short times ($t \lesssim \Gamma_L^{-1}$) the dominant process corresponds to the central system charging and the universal behavior generated by the first tunneling event is again observed. However, at longer times ($t \gtrsim 1/\Gamma$), the universality is broken and a new kind of oscillations with a renormalized amplitude appear. In the lower panel of Fig. 3.6 we represent the amplitude of the charge transfer cumulants oscillations with respect to the order, for the universal case (red dots) and the bidirectional one (blue squares). In the universal case the amplitude follows the scaling law given in Eq. (3.27), while this law is broken in the bidirectional case. A similar behavior has been observed in Refs. [32, 33] for the short time energy statistics.

The breaking of the universality is due to the interference between two processes involving the short time transport in both directions. It generates two dominant zeros, one starting at $z \rightarrow -\infty$, related to the system charging from the left electrode, and another one at $z \rightarrow 0$, related to the system discharging through the left electrode. As the Fermi edges of the electrodes and the system level are aligned, these two poles are equally dominant, since both processes are equally probable, leading to an interference that produces the universality breaking. The short time universality is recovered in the case when one of the two poles (or, equivalently, the transport in one of the directions) become more favorable, and one can estimate that this happens when the bias voltage becomes bigger than the tunneling rates ($V > \Gamma$).

3.6 Conclusions

In this chapter I have introduced the basic ideas to analyze the problem of the time-dependent transport through mesoscopic nanojunctions. I have discussed the analytic solution for the mean properties (system charge and current), which will be used as an approximate solution to analyze some interacting situations under simple approximations such as the DTA for the electron-phonon interaction. I have also introduced the discretization procedure which can be used for a numerical evaluation of the higher order cumulants. Moreover, I have deduced an analytic expression relating the cumulants and the zeros of the GF. This analytic expression has been used to analyze the uni-

versal oscillatory behavior originated by the first electron transfer in the unidirectional transport. I have also analyzed the universality in the unidirectional transport situation in the sequential ($T \gg \Gamma$) and coherent ($T \ll \Gamma$) regimes. Finally, the bidirectional situation ($V \lesssim \Gamma$) has also been investigated, showing the breaking of the short time universal behavior. In any of the situations mentioned before, the universal behavior of the charge and current cumulants is lost due to interference contributions of the electrons transferred, leading to the system relaxation to the steady state.

Bibliography

- [1] Y. Blanter and M. Büttiker. Shot noise in mesoscopic conductors. *Physics Reports* **336**, 1 (2000).
- [2] Y. Nazarov and Y. Blanter. *Quantum Transport: Introduction to Nanoscience* (Cambridge University Press, 2009).
- [3] C. Fricke, F. Hohls, W. Wegscheider, and R. J. Haug. Bimodal counting statistics in single-electron tunneling through a quantum dot. *Phys. Rev. B* **76**, 155307 (2007).
- [4] C. Flindt, C. Fricke, F. Hohls, T. Novotný, K. Netočný, T. Brandes, and R. J. Haug. Universal oscillations in counting statistics. *Proceedings of the National Academy of Sciences* **106**, 10116 (2009).
- [5] C. Fricke, F. Hohls, C. Flindt, and R. J. Haug. High cumulants in the counting statistics measured for a quantum dot. *Physica E: Low-dimensional Systems and Nanostructures* **42**, 848 (2010). 18th International Conference on Electron Properties of Two-Dimensional Systems.
- [6] P. Stegmann, B. Sothmann, A. Hucht, and J. König. Detection of interactions via generalized factorial cumulants in systems in and out of equilibrium. *Phys. Rev. B* **92**, 155413 (2015).
- [7] D. Kambly and C. Flindt. Time-dependent factorial cumulants in interacting nano-scale systems. *Journal of Computational Electronics* **12**, 331 (2013).
- [8] G.-M. Tang, F. Xu, and J. Wang. Waiting time distribution of quantum electronic transport in the transient regime. *Phys. Rev. B* **89**, 205310 (2014).
- [9] G.-M. Tang and J. Wang. Full-counting statistics of charge and spin transport in the

- transient regime: A nonequilibrium Green's function approach. *Phys. Rev. B* **90**, 195422 (2014).
- [10] R. Seoane Souto, R. Avriller, R. C. Monreal, A. Martín-Rodero, and A. Levy Yeyati. Transient dynamics and waiting time distribution of molecular junctions in the polaronic regime. *Phys. Rev. B* **92**, 125435 (2015).
 - [11] A. Blandin, A. Nourtier, and D. W. Hone. Localized time-dependent perturbations in metals: formalism and simple examples. *J. Phys. France* **37**, 369 (1976).
 - [12] A.-P. Jauho, N. S. Wingreen, and Y. Meir. Time-dependent transport in interacting and noninteracting resonant-tunneling systems. *Phys. Rev. B* **50**, 5528 (1994).
 - [13] R. Seoane Souto, A. Martín-Rodero, and A. Levy Yeyati. Analysis of universality in transient dynamics of coherent electronic transport. *Fortschritte der Physik* **65**, 1600062 (2017).
 - [14] A. Kamenev. *Field Theory of Non-Equilibrium Systems* (Cambridge University Press, 2011).
 - [15] T. Ozaki. Continued fraction representation of the Fermi-Dirac function for large-scale electronic structure calculations. *Phys. Rev. B* **75**, 035123 (2007).
 - [16] M. Esposito, U. Harbola, and S. Mukamel. Nonequilibrium fluctuations, fluctuation theorems, and counting statistics in quantum systems. *Rev. Mod. Phys.* **81**, 1665 (2009).
 - [17] I. D. Abrahams. On the solution of Wiener–Hopf problems involving noncommutative matrix kernel decompositions. *SIAM Journal on Applied Mathematics* **57**, 541 (1997).
 - [18] C. N. Yang and T. D. Lee. Statistical theory of equations of state and phase transitions. i. theory of condensation. *Phys. Rev.* **87**, 404 (1952).
 - [19] T. D. Lee and C. N. Yang. Statistical theory of equations of state and phase transitions. ii. lattice gas and ising model. *Phys. Rev.* **87**, 410 (1952).
 - [20] D. Kambly, C. Flindt, and M. Büttiker. Factorial cumulants reveal interactions in counting statistics. *Phys. Rev. B* **83**, 075432 (2011).
 - [21] C. Flindt and J. P. Garrahan. Trajectory phase transitions, Lee-Yang zeros, and high-order cumulants in full counting statistics. *Phys. Rev. Lett.* **110**, 050601 (2013).
 - [22] J. M. Hickey, C. Flindt, and J. P. Garrahan. Trajectory phase transitions and dynamical

BIBLIOGRAPHY

- Lee-Yang zeros of the Glauber-Ising chain. *Phys. Rev. E* **88**, 012119 (2013).
- [23] A. G. Abanov and D. A. Ivanov. Allowed charge transfers between coherent conductors driven by a time-dependent scatterer. *Phys. Rev. Lett.* **100**, 086602 (2008).
- [24] A. G. Abanov and D. A. Ivanov. Factorization of quantum charge transport for noninteracting fermions. *Phys. Rev. B* **79**, 205315 (2009).
- [25] F. Hassler, M. V. Suslov, G. M. Graf, M. V. Lebedev, G. B. Lesovik, and G. Blatter. Wave-packet formalism of full counting statistics. *Phys. Rev. B* **78**, 165330 (2008).
- [26] Y. Utsumi, O. Entin-Wohlman, A. Ueda, and A. Aharony. Full-counting statistics for molecular junctions: Fluctuation theorem and singularities. *Phys. Rev. B* **87**, 115407 (2013).
- [27] K. Brandner, V. F. Maisi, J. P. Pekola, J. P. Garrahan, and C. Flindt. Experimental determination of dynamical Lee-Yang zeros. *Phys. Rev. Lett.* **118**, 180601 (2017).
- [28] R. S. Souto, A. Martín-Rodero, and A. L. Yeyati. Quench dynamics in superconducting nanojunctions: Metastability and dynamical yang-lee zeros. *Phys. Rev. B* **96**, 165444 (2017).
- [29] M. Abramowitz and I. A. Stegun. *Handbook of Mathematical Functions with Formulas, Graphs and Mathematical Tables* (Dover Publications, Inc., New York, 1965).
- [30] C. Flindt, T. Novotný, A. Braggio, and A.-P. Jauho. Counting statistics of transport through Coulomb blockade nanostructures: high-order cumulants and non-Markovian effects. *Phys. Rev. B* **82**, 155407 (2010).
- [31] P. Stegmann and J. König. Short-time counting statistics of charge transfer in Coulomb-blockade systems. *Phys. Rev. B* **94**, 125433 (2016).
- [32] Z. Yu, G.-M. Tang, and J. Wang. Full-counting statistics of transient energy current in mesoscopic systems. *Phys. Rev. B* **93**, 195419 (2016).
- [33] G. Tang, Z. Yu, and J. Wang. Full-counting statistics of energy transport of molecular junctions in the polaronic regime. *New Journal of Physics* **19**, 083007 (2017).

4 Polaron effects in quench dynamics

4.1 Introduction

As already mentioned in the previous chapter, the study of current fluctuations provide a complete information about the transport phenomenon through the system, such as electron interactions and correlation effects [1, 2]. On the other hand, the time evolution of the physical observables contain information about the characteristic system time scales, which can be important to fabricate devices operating at high frequency [3, 4] or for developing single electron sources [5–8] and single electron detectors [9].

In this context, new methods have to be developed to characterize the charge fluctuations in the time domain. The concept of Waiting Time Distributions (WTD), developed originally in the field of quantum optics and stochastic processes [10], has been more recently introduced to characterize the electron quantum transport [11]. Although initially the works were focused on the incoherent regime [12, 13], a more recent extension to the coherent regime has been done for non-interacting devices [14, 15]. Another approach to the problem, discussed in Ref. [16], is provided by the NGFs. This method has been used to analyze the WTD and transient fluctuations in coherent non-interacting junctions in Refs. [17, 18].

This chapter is devoted to the study of the transient transport properties of a molecular junction, analyzing the effect of an internal vibrational degree of freedom. The simplest model describing this situation is the so-called spinless Anderson-Holstein model

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given in Eq. (2.52) [19]. This simple model provides the basis to understand complex non-equilibrium phenomena occurring in actual systems such as the phonon-assisted tunneling [20, 21] and the Franck-Condon blockade [22]. While this model has extensively been analyzed in the stationary regime [23–30], the time-dependent situation has been much less studied [31]. Some exact calculations using the diagrammatic MC technique have shown that for strong electron-phonon coupling (polaronic regime) the current can evolve to two different values at short times, depending on the system initialization [32]. This behavior was interpreted as a signature of bistability in the system. Later works have shown that the apparent bistability corresponds actually to a long transient dynamics due to polaronic effects [33–37], whose results are in qualitative agreement with the DTA ones.

There is however a lack of works studying the fluctuations dynamics in the time domain. In this context the work of this chapter was done, whose main results have been presented in Ref. [35]. This chapter is focused on the coherent dynamics of the charge fluctuations in the polaronic regime by means of the DTA, described in subsection 2.4.3. This approximation has been shown to provide accurate results for the current and the spectral density in the stationary regime [38]. Moreover, it preserves the appropriate limits such as the FSR at equilibrium situation and zero temperature, and the convergence to the SPA [23] result for large voltage values. Importantly, after publishing this work some more articles appeared studying the dynamics of molecular junctions using the DTA, investigating the current evolution [37, 39] and the energy transfer statistics [40]. More recently, some other works appeared analyzing the WTD in the Born-Markov approximation [41, 42].

In this chapter, I firstly analyze the current and system population evolution. The results are in good agreement with numerically exact results from Ref. [32]. The system relaxation is also analyzed, finding that the interaction of electrons with the localized mode enlarge exponentially the convergence time to the steady state in the polaronic regime. This convergence time is significantly reduced when a bias voltage is applied to the junction. The charge cumulants are also analyzed, finding the short time universal behavior commented in the previous chapter for a non-interacting system which indicates that the DTA is an effective non-interacting scheme with renormalized parameters. However, signatures of the polaron dynamics can be found in the current cumulants and the WTD, which exhibit features at times $\sim 2\pi/\omega_0$ (ω_0 being the phonon frequency) due to interference effects of electrons transferred through different channels. These effects

become more evident in the conductance and the differential Fano factor, which are analyzed at the end of the chapter. A peculiar time evolution is found in the inelastic ($V = \omega_0$) and the elastic threshold ($V = 2\omega_0$).

4.2 Basic theoretical formulation

The objective of this section is to introduce the basic formalism for understanding the dynamics of a molecular junction. I consider the simplest situation described by the spinless Anderson-Holstein model, given by the Hamiltonian of Eq. (2.52), where the system is described by a spinless level coupled to a localized vibrational mode. In this chapter I focus on the polaronic regime where electrons and phonons are strongly coupled ($\lambda^2/\omega_0\Gamma \gg 1$) using the DTA [29, 38] introduced in section 2.4 of the second chapter. In this section I reformulate this approximation to include the time dependence and to study the corresponding counting statistics. Specific tools for the DTA are also introduced in this chapter.

The starting point of this section is the spinless Anderson-Holstein Hamiltonian after the Lang-Firsov transformation given in Eq. (2.64). I consider that the molecule is suddenly coupled to two electrodes at a given time (taken as $t = 0$). Thus, the tunneling Hamiltonian acquire a dependence on time as

$$\tilde{H}_T(t) = \theta(t) \sum_{k,\nu} \left(t_\nu e^{i\chi_\nu/2} c_{k,\nu}^\dagger X d + t_\nu^* e^{-i\chi_\nu/2} d^\dagger X c_{k,\nu} \right), \quad (4.1)$$

where $X = \exp[\lambda/\omega_0(a + a^\dagger)]$ is the polaron cloud operator and χ_ν is the counting field. For simplicity the lead subindex (ν) in the counting field will be dropped along the derivation, being the following derivation valid for any choice (see discussion in section 2.6). The GF is given by the expected value of the time evolution operator [16], which in this problem is given by

$$Z(\chi, t) = \left\langle T_c \exp \left\{ -i \int dt_1 \tilde{H}_{T,\chi}(t_1) \right\} \right\rangle. \quad (4.2)$$

It is possible to obtain the GF in the DTA following the reasoning from Ref. [43] which uses Gaussian integration and Grassmann algebra. However, it becomes more instructive to proceed as discussed in Ref. [44] where the authors suggested to compute the GF integrating the counting field-dependent current (see subsection 2.6.2 for further details).

4. Polaron effects in quench dynamics

In the stationary regime (where the leads are considered to be connected in the remote past), the solution to the problem is described by the NGFs in the frequency domain given in Eq. (2.73). The same Green functions can be obtained in the time domain, including the counting field variable, finding

$$\hat{\tilde{G}}_{DTA,\chi}(t, t') = \hat{\Lambda}(t, t') \hat{G}_{1,\chi}(t, t'), \quad (4.3)$$

where $\hat{\Lambda}$ is the polaron propagator, whose off-diagonal components are given in Eq. (2.66). The function $\hat{G}_1$ is an intermediate Green function used for clarity. This Green function contains information about the coupling to the electrodes and the polaron cloud through dressed tunneling self-energies (2.72) as

$$\hat{G}_{1,\chi}(t, t') = \left[\hat{g}_S^{-1} - \hat{\tilde{\Sigma}}_{T,\chi} \right], \quad (4.4)$$

where

$$\tilde{\Sigma}_{T,\chi}^{\alpha\beta}(t, t') = \Lambda^{\alpha\beta}(t, t') \Sigma_{T,\chi}^{\alpha\beta}(t, t'), \quad (4.5)$$

with the non-interacting tunneling self-energy is given by Eq. (3.16) and the bare polaron propagator in Eq. (2.66). Thus, the counting field-dependent current through any of the electrodes can be obtained making use of Eq. (2.112), finding

$$I_{\nu,\chi}(t) = \int_0^t dt_1 \left[\tilde{G}_{DTA,\chi}^{-+}(t, t_1) \Sigma_{T\nu,\chi}^{+-}(t_1, t) - \tilde{G}_{DTA,\chi}^{+-}(t, t_1) \Sigma_{T\nu,\chi}^{-+}(t_1, t) \right]. \quad (4.6)$$

A further simplification can be done noticing that the off-diagonal components of the bare polaron propagator are related by $\Lambda^{+-}(t, t_1) = \Lambda^{-+}(t_1, t)$. Thus, Eq. (4.6) can then be written as

$$I_{\nu,\chi}(t) = \int_0^t dt_1 \left[G_{1,\chi}^{-+}(t, t_1) \tilde{\Sigma}_{T\nu,\chi}^{+-}(t_1, t) - G_{1,\chi}^{+-}(t, t_1) \tilde{\Sigma}_{T\nu,\chi}^{-+}(t_1, t) \right]. \quad (4.7)$$

Notice that Eqs. (4.4) and (4.7) describe the situation of an effective non-interacting system with renormalized tunneling self-energies ($\tilde{\Sigma}_T$) due to the dressing with the polaronic cloud. The simplification done in Eq. (4.7) cannot be performed in a general situation where the polaronic cloud is dressed with the electron degrees of freedom. The effects of the non-equilibrium phonon population will be analyzed in chapter 5 by means of a perturbative approximation. In this situation the problem becomes more demanding and it is out of the scope of this chapter. The GF can be finally obtained by integrating Eq. (4.7) with respect to the counting field (4.7), following the non-interacting procedure described in Ref. [16] and finding

$$Z(\chi, t) = \det \left[g_S^{-1} - \hat{\Sigma}_{T,\chi} \right] / \det \left[g_S^{-1} - \hat{\Sigma}_{T,\chi=0} \right], \quad (4.8)$$

which is analogous to the non-interacting expression (3.13) with renormalized tunneling amplitudes. A similar result has been obtained in Ref. [29], where the authors demonstrated that the GF for the DTA in the stationary regime is precisely given by the Levitov-Lesovik with the self-energies described in Eq. (2.72). This fact has several implications. The first one is that the DTA does not introduce electron-electron correlation effects mediated by the phonon degree of freedom. In order to take into account of such effects one should include vertex corrections [45] or the phonon non-equilibrium dynamics [46–48] in the DTA scheme. Therefore, the zeros of the GF written in Eq. (4.8) appear in the real negative axis. Despite of the absence of correlation effects, the DTA provides accurate results in the polaronic regime, as described in Ref. [38]. Due to the analogous expression to the non-interacting case (4.8), all the theoretical methods introduced in the previous chapter can be applied to this model. For instance, the GF could be obtained via the discretization procedure mentioned in the previous chapter as

$$Z(\chi, t) = \det \left[\hat{\mathbf{g}}_S^{-1} - (\Delta t)^2 \hat{\Sigma}_{T,\chi} \right] / \det \left[\hat{\mathbf{g}}_S^{-1} - (\Delta t)^2 \hat{\Sigma}_{T,\chi=0} \right]. \quad (4.9)$$

where the discretized version of the inverse bare system Green function is given by Eq. (3.15). The self-energies in the discretized time mesh can be obtained straightforwardly from Eq. (4.5).

4.2.1 Single pole approximation

For the time dependent single particle properties, such as the system population and the mean current, the single pole approximation can be used. This approximation is based on the non-interacting solution described in the section 3.2 of the previous chapter. As in the non-interacting case it becomes possible to write the Dyson equation for the $\hat{G}_1$ Green function given in Eq. (4.4) in the triangular representation in the absence of counting field [43, 49], finding

$$G_1^{a,r}(t, t') = g_0^{a,r}(t, t') + \int_0^t dt_2 K_{DTA}^{a,r}(t, t_2) G_1^{a,r}(t_2, t'), \quad (4.10)$$

where the kernel of the integral equation is given by $K_{DTA}^{a,r}(t-t_2) = \int dt_1 g_S^{a,r}(t-t_1) \tilde{\Sigma}_T(t_1-t_2)$. Similarly to the non-interacting situation, this kernel depends only on the time difference, preserving the time translation invariance of the bare Green functions. Then, using the Laplace formalism, the retarded and advanced Green functions are given by

$$G_1^{a,r}(s) = \frac{g_S^{a,r}(s)}{1 - K_{DTA}^{a,r}(s)}. \quad (4.11)$$

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As in the non-interacting case, the solution in the time domain is given by the inverse Laplace transform, which involves the integration along the Bromwich contour containing the full collection of poles of the function. This is in general a difficult task, since the DTA can exhibit many singular points. However, in the $\Gamma \ll \omega_0$ regime (where the approximation is expected to provide accurate results) the function is well approximated by a single dominant pole, $\xi^{a,r}$, which has to be determined numerically. The retarded and advanced components are then given by

$$G_1^{a,r}(t-t') = \theta(t-t') \text{Res}(\xi^{a,r}) e^{\xi^{a,r}(t-t')}. \quad (4.12)$$

It is important to note that for obtaining the DTA Green function a final polaron dressing is needed, as described by Eq. (4.3).

The Keldysh components can be determined through the kinetic equation. For simplicity I will show the derivation for the charge occupation for an initially empty system level, although a similar expression can be derived for the initially occupied case. In this case, the G_1^{+-} component is given by

$$G_1^{+-}(t, t') = \int_0^t dt_1 dt_2 G_1^r(t, t_1) \tilde{\Sigma}_T^{+-}(t_1 - t_2) G_1^a(t_2, t'), \quad (4.13)$$

and the system population is given by the imaginary part of this Green function, finding

$$n(t) = n(t \rightarrow \infty) \left[1 + 2e^{2\text{Re}(\xi)t} \right] - 2e^{2\text{Re}(\xi)t} n_x(t), \quad (4.14)$$

where

$$n_x(t) = \sum_{\nu=L,R} \frac{\Gamma_\nu}{\pi} \sum_{k=-\infty}^{\infty} \alpha_k \int d\omega f_\nu(\omega + k\omega_0) \frac{\cos[(\omega + \text{Im}(\xi))t]}{\text{Re}(\xi)^2 + [\omega + \text{Im}(\xi)]^2}, \quad (4.15)$$

and the stationary population is given by $n(t \rightarrow \infty) = n_x(0)$. Similarly to the non-interacting situation, the real part of the pole provides an estimation of the system relaxation time after the connection to the electrodes. Finally, the symmetrized current evolution can be obtained through Eq. (3.8). Considering a symmetric coupling to the electrodes for simplicity, I find

$$\langle I(t) \rangle = \frac{\theta(t)}{2} \text{Re} \left\{ \int_0^t dt_1 \tilde{G}_{DTA}^r(t, t_1) [\Gamma_L f_L(t - t_1) - \Gamma_R f_R(t - t_1)] \right\}, \quad (4.16)$$

where the retarded component is given by

$$\tilde{G}_{DTA}^r(t, t_1) = \Lambda^{+-}(t, t_1) G_1^R(t, t_1) + [\Lambda^{+-}(t, t_1) - \Lambda^{-+}(t, t_1)] G_{DTA}^{+-}(t, t_1). \quad (4.17)$$

4.2.2 Short time tunnel limit

Further insight can be obtained by studying the short time evolution, performing a perturbation to the lowest order in the tunneling amplitude at the ν interface. Through this expansion, the GF of the DTA, given in Eq. (4.8) gets a simple expression as

$$Z(\chi, t) \simeq 1 + \int_0^t dt_1 \int_0^t dt_2 \text{Tr}_K \{ [\tilde{\Sigma}_{T\nu, \chi}(t_1, t_2) - \tilde{\Sigma}_{T\nu, \chi=0}(t_1, t_2)] \hat{g}_S(t_2, t_1) \}, \quad (4.18)$$

which can be expanded as

$$Z(\chi_\nu, t) \simeq 1 + \int_0^t dt_1 \int_0^t dt_2 [\tilde{\Sigma}_{T\nu}^{+-}(t_1, t_2) g_S^{-+}(t_2, t_1) (e^{i\chi_\nu} - 1) + \tilde{\Sigma}_{T\nu}^{-+}(t_1, t_2) g_S^{+-}(t_2, t_1) (e^{-i\chi_\nu} - 1)]. \quad (4.19)$$

It is worth noticing that the short time limit and the atomic limits coincide meaning that both are described by the expansion to the lowest order in Γt . The previous equation can be written as

$$Z(\chi_\nu, t) \simeq 1 + \left\{ (e^{i\chi} - 1) A_{\nu,01}(t) [1 - n(0)] + (e^{-i\chi} - 1) A_{\nu,10}(t) n(0) \right\}, \quad (4.20)$$

with

$$A_{\nu,01}(t) = \frac{2\Gamma_\nu}{\pi} \sum_{k=0}^{\infty} \alpha_k \int_{-D}^D d\omega \frac{1 - \cos[(\omega - \tilde{\epsilon} - k\omega_0)t]}{(\omega - \tilde{\epsilon} - k\omega_0)^2} f_\nu(\omega), \quad (4.21)$$

$$A_{\nu,10}(t) = \frac{2\Gamma_\nu}{\pi} \sum_{k=0}^{\infty} \alpha_k \int_{-D}^D d\omega \frac{1 - \cos[(\omega - \tilde{\epsilon} + k\omega_0)t]}{(\omega - \tilde{\epsilon} + k\omega_0)^2} [f_\nu(\omega) - 1], \quad (4.22)$$

where D is the bandwidth of the electrode. In the $t \rightarrow \infty$ limit, $A_{\nu,01}$ and $A_{\nu,10}$ tend to the Fermi Golden rule rates derived for the sequential tunneling regime (see for example Ref. [50]). As expected in the short time limit $\Gamma t \ll 1$, the GF in Eq. (4.8) involves the charge transfer of a single electron at the studied interface. In other words, it only describes the time evolution of the dominant zero of the GF at short times (see discussion of chapter 3). In this approximation, only $P_0(t)$, $P_{-1}(t)$ and $P_1(t)$ contribute, being given by

$$P_q(t) = A_{\nu,01}(t) [1 - n(0)] \delta_{q,1} - A_{\nu,10}(t) n(0) \delta_{q,-1}, \quad (4.23)$$

and the idle time probability is $P_0(t) = 1 - P_1(t) - P_{-1}(t)$. In this simple situation the WTD is proportional to the left current $I_L(t)$ as

$$W(t) = -\frac{d}{dt} P_0(t) = |I_L(t)|. \quad (4.24)$$

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At zero temperature and neglecting contributions from the band edges (which is justified in the wide band approximation) the left current has a very compact expression as

$$I_\nu = \Gamma_\nu(1 - 2n_d) + \frac{2\Gamma_\nu}{\pi} \sum_k \alpha_k \{ \text{Si}[(\mu_\nu + k\omega_0 - \tilde{\epsilon})t] (1 - n_d) - \text{Si}[(\mu_\nu - k\omega_0 - \tilde{\epsilon})t] n_d \} , \quad (4.25)$$

where Si denotes the sine integral function. This expression describes a fast raise of the current at short times. At longer times it predicts a coherent superposition of oscillations, generated by the high order polaronic satellites in the DOS (see for instance Fig. 2.8 of the second chapter).

Note that the initial current is non-zero and equal to $I_\nu = \pm\Gamma_\nu$, with the sign depending on the initial charge ($n(0) = 0$ or $n(0) = 1$). This property fixes also the initial value of the WTD as $W(t) \equiv |I_\nu(0)|$. It should be noticed that the system evolution on time scales smaller than the inverse of the leads bandwidth is neglected, which explains why the initial current can be non-zero [51]. However, for the symmetrized current $\langle I(t) \rangle = [\Gamma_R I_L(t) - \Gamma_L I_R(t)]/\Gamma$ the initial value is zero and the first charge transfer cumulant is a continuous function starting from zero regardless of the initial condition.

4.3 Evolution of system population and current

In this section the results for the time evolution of the system population and current after the connection to the electrodes are presented, focusing on the typical relaxation times. In Ref. [32] the authors showed that, in the polaronic regime, the system current becomes sensitive to the initial occupation in time scales larger than the typical relaxation time for a non-interacting nanojunction ($\tau_{relax} \approx 1/\Gamma$). This feature was interpreted as a signature of the system bistability. Later analysis on the same parameters [33–37] revealed that the apparent bistability is in reality a long time relaxation time due to polaronic effects.

In Fig. 4.1 the system population evolution is represented, comparing the results of the single pole approximation for the DTA given by Eq. (4.14) with the diagrammatic MC simulations from Ref. [32]. Results for the two different initial configurations $n(0) = 0$ and $n(0) = 1$ are shown. As can be observed, the agreement between the DTA (solid

Figure 4.1: Time dependent system population comparing the DTA (full lines) and the diagrammatic MC [32] (symbols) for initially empty (red) and initially full occupied (blue) system level. The model parameters are $\tilde{\epsilon} = -10\Gamma$, $\lambda/\omega_0 = 2$, $\omega_0 = 8\Gamma$ and $V = 26\Gamma$.

lines) and the exact calculations (symbols) is very good for the initially fully occupied situation (blue) which is close to the long time expected value, represented for the DTA by the arrow. For the initially empty situation (red curve) the agreement becomes somewhat worse, as the DTA leads to an overestimation of the system relaxation rate.

In Fig. 4.2 the results for the current evolution are shown for the same parameters as in Fig. 4.1 and two different bias voltages. As can be observed, the agreement between the DTA and the exact calculations is remarkable for the initially fully occupied situation (blue), while it is somewhat poorer for the initially empty case (red). Moreover the agreement becomes better in the low voltage situation (upper panel) than in the higher one (lower panel). This reflects that the DTA describes accurately the system properties at $V < \omega_0$ around the Fermi edge, while this description becomes poorer at higher energies. In both cases the DTA is able to reproduce the features in the current evolution at $t \sim 2\pi/\omega_0$ due to the coherent superposition of oscillations described at short times by Eq. (4.25), although overestimating the system relaxation rate. The origin of this discrepancy could be due to the non-equilibrium polaron dynamics, not included in the approximation [38] and which is expected to be important for $V \gtrsim \omega_0$. The inset of the

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Figure 4.2: Symmetrized transient current for the same parameters as in the Fig. 4.1 with $V = 5\Gamma$ (upper panel) and $V = 26\Gamma$ (lower panel). The inset in the top panel shows the evolution at longer times for the $V = 5\Gamma$ case.

upper panel shows the long time current evolution, which converges to the same stationary value independently from the initial condition. Moreover, for increasing times the polaronic features are gradually damped, leading to a simple exponential convergence to the steady state.

As mentioned before, the interactions drastically increase the system relaxation time. In the low bias voltage situation and for electron-hole symmetry ($\tilde{\epsilon} = 0$), the relaxation rate is given by the width of the peak at Fermi edge. At zero temperature, the relaxation time is given by $\Gamma^{-1} \exp[(\lambda/\omega_0)^2]$, which predicts an exponential increase with the square of the coupling strength. In the voltage biased situation or for $\tilde{\epsilon} \neq 0$ the situation becomes more complex. In the left panel of Fig. 4.3 the relaxation times are represented for increasing values of the interaction strength and different bias voltages, estimated from the inverse of the real part of the pole of the DTA (i.e. $\tau_r \approx -1/\text{Re}(\xi)$). As shown,

Figure 4.3: Left panel: characteristic relaxation time as a function of the electron-phonon coupling strength with $V = 5\Gamma$ (blue curve), 26Γ (green curve) and 50Γ (red curve), for $\tilde{\epsilon} = -10\Gamma$. The black line shows the relaxation rate of the system at the equilibrium situation and for $\tilde{\epsilon} = 0$. The right panel shows the relaxation times as a function of the bias voltage for $\tilde{\epsilon} = 0$ comparing the numerical results (black solid line) with the analytic expression (dashed yellow line), given by Eq. (4.26). The remaining parameters are the same as in Fig. 4.1.

the relaxation behaves exponentially in the polaronic regime. The black curve represents the electron-hole symmetric condition at the equilibrium situation which sets the upper bound to the current convergence time.

In the right panel of Fig. 4.3 the relaxation time is shown as a function of voltage for a perfect transmitting situation and $T = 0$ for simplicity. At low voltage bias ($V < \omega_0$) the transport through the system takes place mainly through the resonance at the Fermi edge, whose width determines the inverse of the system relaxation time. This time is significantly reduced at $V = 2n\omega_0$ which corresponds to the opening of new channels, related to processes mediated by the level in which electrons exchange an integer number of phonons. These processes are schematically represented in the right panel of Fig. 4.3. For $V > 2\omega_0$ (left inset of the right panel) an electron at ω_0 energy can exchange two phonons with another electron from energy $-\omega_0$ (represented as dashed) and mediated

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by the level. For $V > 4\omega_0$ higher order processes are allowed, where an electron can emit 4 phonons, which can be adsorbed by one or two electrons. Similar processes occur at higher voltages, reducing monotonously the relaxation time. An analytic expression of the characteristic relaxation times can be obtained by taking into consideration that all the sidebands inside the voltage window contribute to the relaxation of the system. Thus, it can be written as

$$\tau_r = \sum_{k \in [-\frac{V}{2}, \frac{V}{2}]} \frac{1}{\alpha_k} = \frac{e^{\lambda^2/\omega_0^2}}{\sum_{k \in [-\frac{V}{2}, \frac{V}{2}]} g^{2k}/k!}, \quad (4.26)$$

where the sum is performed over the allowed processes involving the exchange of $2n$ phonons. The result is shown in the right panel as a discontinuous line, exhibiting a quantitative agreement, except for $V \sim 2n\omega_0$, where the relaxation time changes abruptly. Eq. (4.26) also describes the convergence to the non-interacting result ($\tau_r \rightarrow 1/\Gamma$) in the regime where $V \gg \omega_0$, which is consistent with the fact that the DTA tends to the SPA [23] result at high energies (see for instance Fig. 2.8). Interestingly, a similar qualitative behavior has been observed in the WTD in the Born-Markov approximation in Ref. [42]. Finally, it is worth mentioning that this picture might become more complex if the phonon non-equilibrium dynamics is included, since more processes have to be taken into consideration. This situation will be analyzed in the next chapter by means of perturbative approximations for a weak electron-phonon coupling strength.

4.4 Transient statistics and waiting time distribution

In this section I present results for the high order cumulants and the WTD in order to obtain further insight about the system dynamics. In the top panel of Fig. (4.4) the higher order charge cumulants are shown, comparing the DTA numerical results with the exact expression for the charge cumulants (3.24), finding a remarkable agreement at short times. The dominant zero of the GF is well approximated by a simple charging law $z_1 \approx A(1 - e^{-\Gamma_L t})$, where A is its asymptotic value taken as a fitting parameter. In the lower panel of Fig. 4.4 the amplitude of the oscillations are plotted for three different values of the coupling strength, following in any case the universal law from Eq. (3.27), described by the solid line.

Figure 4.4: Upper panel: short time charge cumulants of increasing orders for the same parameters as in Fig. 4.1, comparing the numerical results (solid lines) with the analytic expression from Eq. (3.24). In the lower panel the amplitudes of the oscillations are represented for different values of electron-phonon interaction, $\lambda/\omega_0 = 0, 1$ and 1.5 , following the universal scaling law (black line) from Eq. (3.27).

The results mentioned in the above paragraph suggest that the DTA is an effectively non-interacting approximation where correlation effects are neglected. However, the current cumulants exhibit features of the coupling to the polaronic cloud, shown in the top panel of Fig. 4.5. As can be observed, they exhibit oscillations with greater amplitudes with increasing order, consistent with the short time universality. These oscillations are well described by Eq. (3.28), originated by the dynamics of the dominant zero of the GF. On top of that, additional features are observed at $\sim 2\pi n/\omega_0$ associated to the polaron dynamics. At those times a new polaron mediated conduction channel is opened, producing an abrupt change of the dominant zero velocity.

In the lower panel of Fig. 4.5 the WTD distribution is presented for the two possible

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Figure 4.5: Upper panel: the lower panel shows the transient dynamics of current cumulants $I_L^k(t)$ with $k = 1, 2, 3, 4, 5, 6$ (from top to bottom at short times) for the initially empty case. Bottom panel: waiting time distribution for the initially empty (red) and initially occupied (blue) cases and the same parameters of the lower panel of Fig. 4.2 (lower panel). Inset: probabilities P_0 (full line) and P_1 (dashed line) for the initially empty state.

initial configurations. At $T = 0$, the WTD is fixed to $1/2$ due to the infinite bandwidth limit considered. This value reflects the finite probability of transferring an electron from very low energies, whose dynamics are of the order of the inverse of their energy. For the initially empty situation, the WTD decreases monotonously for increasing times, describing a high probability to transfer an electron at short times. At variance, the initially fully occupied case describes a fast decay and a finite and almost constant value at long times. The slow evolution in this case is related to the blocking of the charge evolution shown in Fig. 4.1. In both cases, the WTD exhibits a series of features at $\sim 2\pi n/\omega_0$ associated with the polaron dynamics. Those features are well described by Eq. (4.25), since at this short times scales only two probabilities (P_0 and P_1) contribute to the GF (see inset in the lower panel of Fig. 4.5), associated to a single electron transfer. This suggests that the typical relaxation time is significantly enlarged by the interactions.

Figure 4.6: Waiting time distribution for $\lambda/\omega_0 = 0$ (green), 1 (blue) and 1.5 (red) for initially empty (upper panel) and initially full occupied (lower panel) with $\tilde{\epsilon} = -\Gamma$, $\omega_0 = 2\Gamma$ and $V = 5\Gamma$. The insets show the corresponding probabilities $P_1(t)$ and $P_2(t)$ (upper panel) and $P_1(t)$ and $P_{-1}(t)$ (lower panel).

In order to analyze the system relaxation to its steady state for the current cumulants and the WTD distribution less extreme parameters will be chosen in the following. In Fig. 4.6 the WTD is represented for increasing values of the electron-phonon coupling parameter and the two initial configurations. For the initially empty configuration (top panel) and for weak coupling between the system and the electrodes, the WTD is well approximated by Eq. (4.25). At the shortest times ($t \lesssim \Gamma^{-1}$), it exhibits an initial linear behavior given by $W(t) \approx \Gamma_L/2 + 2\Gamma_L/\pi \sum_n \alpha_n (\mu_L - \tilde{\epsilon} - n\omega_0)t$, followed by an extreme at $t \approx \pi \{ |\sum_n \alpha_n (\mu_L - \tilde{\epsilon} - n\omega_0) / \sum_n \alpha_n (\mu_L - \tilde{\epsilon} - n\omega_0)^3| \}^{1/2}$. In the non-interacting case (green curve) only the $n = 0$ term contributes and the WTD exhibits an initial positive slope for the parameters in Fig. 4.6, reaching a maximum peak located at $t \approx \pi/|V/2 - \tilde{\epsilon}|$. For increasing coupling strength, the initial slope decreases, becoming eventually negative at large values. For $g \approx 1.3$, the peak converts into a dip, indicating the transition to the strong coupling regime. The probabilities $P_1(t)$ and $P_2(t)$ shown in the insets allow to

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Figure 4.7: Higher order cumulants (normalized with the mean current) for increasing interaction parameter values (same color code as in Fig. 4.6) for the initially empty (left panels) and initially full cases (right panels). Notice that for the occupied case the time axis starts at times slightly larger than 0 in order to avoid the divergent behavior of the normalized cumulants in the $t \rightarrow 0$ limit.

visualize the transference of the first and second electron at the left interface, showing a significant slowing down with increasing interaction.

On the other hand, the initially occupied case (lower panel of Fig. 4.6) exhibits always a short time negative slope, described by $W(t) \approx \Gamma_L/2 - 2\Gamma_L/\pi \sum_n \alpha_n (V/2 - \tilde{\epsilon} + n\omega_0)t$, followed by a dip at very short times $t \approx \pi \{ |\sum_n \alpha_n (\mu_L - \tilde{\epsilon} + n\omega_0) / \sum_n \alpha_n (\mu_L - \tilde{\epsilon} + n\omega_0)^3| \}^{1/2}$ which is associated to the blocking effect due to the Pauli exclusion principle. At variance to the empty case, the dip's evolution is monotonous with the interaction strength, increasing its depth and shifting its position to smaller times. At longer times, the WTD of the weaker interacting case exhibits a maximum related to the higher order process consisting on relaxing the initial condition through the right interface and to charge the system again. For increasing interaction strength, the maximum is shifted to longer times, decreasing also its amplitude due to the Frank-Condon blockade. As shown in the inset, the backward probability (P_{-1}) dominates the short time dynamics, while the revival of the WTD at longer times is controlled by the P_1 . The dip in the WTD is generated by a short time cancellation of these two processes.

Similarly, the increase of the interaction strength has an impact in the evolution and the stationary limit of the higher order cumulants I_L^k . The time evolution of these quantities are shown in Fig. 4.7 for $k = 2, 3$ and 4. The current cumulants are normalized with the time dependent current, which allows one to appreciate more clearly differences with increasing λ . In both situations, the interacting and non-interacting cases, exhibit an increase in the transient amplitude with the cumulant order, consistent with the short time universality (commented in connection to Fig. 4.5). The effect of increasing interaction strength is not only to slow down the dynamics, as commented above, but also to enlarge the relative asymptotic values with respect to the non-interacting situation. The same effects can be observed for the initially occupied case (right panels in Fig. 4.7), where the divergent relative amplitudes at very short times are due to the change of sign of the current.

4.5 Conductance and Fano factor dynamics at $V = n\omega_0$

In this section I present results for the conductance and the differential Fano factor evolution, which contain more clear signatures about the influence of the polaron dynamics in the transient transport properties. This section is focused on the $V \sim n\omega_0$ case where the polaron features are monotonous and easier to understand. The behavior of the conductance at the $V = \omega_0$ threshold has been analyzed in the weakly interacting situation, showing that, the interplay between the elastic and inelastic processes can lead to an increase or decrease of the conductance [52]. More recently a similar qualitative behavior has been found in the polaronic regime in Ref. [38] by means of the DTA. The shot noise has been also analyzed in several works in the limit of weak interaction [53–56], showing that it can either exhibit an increase or a decrease due to the opening of the inelastic channel. The situation at $V = 2\omega_0$ has been analyzed in Ref. [38] for the polaronic regime, indicating that the conductance exhibits an increase and the shot noise a suppression associated with the opening of a sideband.

The transient conductance for $V \sim \omega_0$ and $V \sim 2\omega_0$ are shown in Figs. 4.8 and 4.9 respectively (with $\tilde{\epsilon} = 0$, $\lambda/\omega_0 = 1.5$ and different values of Γ). For comparison the prediction for the tunnel limit, given by Eq. (4.25), is shown as dashed lines. To illustrate the behavior of the noise the differential Fano factor $(\partial F(t)/\partial V$ with $F(t) = \langle I^2 \rangle / (2\langle I \rangle)$) is represented instead of the Fano factor, which allows to see more clearly the differences

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Figure 4.8: Conductance (upper panel) and differential Fano factor (lower panel) for $V = \omega_0$, $g = 1.5$, $\tilde{\epsilon} = 0$ and increasing values of the coupling to the electrodes, $\Gamma/\omega_0 = 0.05$ (blue), 0.25 (green), 1 (yellow) and 2 (red). The dashed line in the upper panel corresponds to the tunnel limit analytic result given by the Eq. (4.25).

in the $\Gamma \rightarrow 0$ limit. As can be observed in these figures, the behavior of the transient conductance in the atomic limit is rather different in the two cases. For the $V = \omega_0$ case the conductance exhibits a sequence of up and down steps at $t \sim 2n\pi/\omega_0$, showing that the polaron dynamics alternatively blocks and deblocks the charge transference through the system. This can lead either to a positive or a negative asymptotic contribution depending on the transmission factor as discussed in Ref. [38]. For the $V = 2\omega_0$ situation the sequence corresponds to steps up showing that the opening of the phonon sideband always tends to increase the system current. The qualitative behavior is well captured by the analytic expression of Eq. (4.25) shown as dashed lines in Figs. 4.8 and 4.9. For increasing Γ the step structure in the conductance is progressively damped. A similar behavior has been observed in Ref. [39] in the stationary limit and after projective measurement, indicating that this peculiar dynamics is not only due to the abrupt connection to the electrodes, but it is a rather general feature of the polaronic regime.

Figure 4.9: Conductance and differential Fano factor for the same as in Fig. 4.8 with $V = 2\omega_0$.

On the other hand, the noise exhibits an interesting different evolution with increasing Γ . While for $\Gamma \rightarrow 0$ the differential noise and the conductance are similar as predicted by the short time limit (4.25), for increasing values of Γ the differential Fano factor can converge either to positive or negative values for $V = \omega_0$. For $V = 2\omega_0$, however, it always converge to negative values, indicating a Fano factor reduction at this point. The behaviors at $V = \omega_0$ and $2\omega_0$ are consistent with the predictions of Ref. [38] for the stationary regime. It is worth noticing that this image might change in the Frank-Condon regime if the non-equilibrium polaron dynamics is included. For instance, in Ref. [50] the authors showed that it can induce a giant Fano factor.

Finally, for values of V that do not commensurate with ω_0 , a less regular time evolution is observed (see for instance the supplemental material of Ref. [35]) and the steps can be either up or down with a criteria given by Eq. (4.25).

4.6 Conclusions

In this chapter the time-dependent counting statistics of the transferred electrons through a resonant level coupled to a localized vibrational mode has been analyzed. By connecting abruptly the electrodes to the molecule, I have investigated the transient evolution of the system, discussing the relaxation time scales and the role of the possible initial conditions. This study is focused on the polaronic regime, where the coupling between the molecule and the electrodes is the smallest energy scale (with $T \ll \Gamma$ to preserve electron coherent effects), by means of the DTA [35] (presented in section 2.4.3 of the second chapter). This approximation has shown a good agreement with the exact MC simulations [32], showing that the transient regime is exponentially enlarged by the interaction. Moreover, the nature of the short time features have been inferred in the tunneling limit, finding that they are due to interference effects generated by the polaron dynamics. Moreover, the typical relaxation times have been analyzed, which are abruptly reduced when opening new elastic channels ($V \sim 2n\omega_0$). An analytic expression is also provided for the electron-hole symmetric point.

In the second part of this chapter I have shown that the high order charge cumulants exhibit oscillations, whose amplitude follow the universal behavior described in the preceding chapter. It indicates that the DTA is an effective non-interacting approximation where correlation effects are neglected. However, features of the polaron dynamics can be observed in the current cumulants and the WTD as jumps. Finally, I have shown that a peculiar evolution of the system is observed when the voltage commensurates with the phonon frequency. While for $V \sim \omega_0$ a series of up and down steps alternate in time for the conductance and the differential Fano factor, for $V \sim 2\omega_0$ these steps show a monotonous behavior, leading to an increase of the conductance and reducing the Fano factor.

There are some remaining questions about the system dynamics that have not been addressed in this chapter. The first one is about the electron correlation effects. The second one refers to the non-equilibrium phonon dynamics. These questions will be partially addressed in the next chapter in the $\lambda < \Gamma$ regime and the for the dynamics of the single particle observables.

Bibliography

- [1] Y. Nazarov and Y. Blanter. *Quantum Transport: Introduction to Nanoscience* (Cambridge University Press, 2009).
- [2] Y. Blanter and M. Büttiker. Shot noise in mesoscopic conductors. *Physics Reports* **336**, 1 (2000).
- [3] L. C. Castro, D. L. John, D. L. Pulfrey, M. Pourfath, A. Gehring, and H. Kosina. Method for predicting fT for carbon nanotube FETs. *IEEE Transactions on Nanotechnology* **4**, 699 (2005).
- [4] Y.-C. Liao, Leiand Lin, M. Bao, R. Cheng, Y. Q. Y. Bai, Jingweiand Liu, K. L. Wang, Y. Huang, and X. Duan. High-speed graphene transistors with a self-aligned nanowire gate. *Nature* **467**, 305 EP (2010).
- [5] G. Fève, A. Mahé, J.-M. Berroir, T. Kontos, B. Plaçais, D. C. Glattli, A. Cavanna, B. Etienne, and Y. Jin. An on-demand coherent single-electron source. *Science* **316**, 1169 (2007).
- [6] E. Bocquillon, V. Freulon, J.-M. Berroir, P. Degiovanni, B. Plaçais, A. Cavanna, Y. Jin, and G. Fève. Coherence and indistinguishability of single electrons emitted by independent sources. *Science* **339**, 1054 (2013).
- [7] J. Dubois, T. Jullien, F. Portier, P. Roche, A. Cavanna, Y. Jin, W. Wegscheider, P. Roulleau, and D. C. Glattli. Minimal-excitation states for electron quantum optics using levitons. *Nature* **502**, 659 EP (2013).
- [8] K. Thibault, J. Gabelli, C. Lupien, and B. Reulet. Pauli-Heisenberg oscillations in electron quantum transport. *Phys. Rev. Lett.* **114**, 236604 (2015).
- [9] I. Neder and F. Marquardt. Coherence oscillations in dephasing by non-Gaussian shot noise. *New Journal of Physics* **9**, 112 (2007).
- [10] N. Van Kampen. *Stochastic Processes in Physics and Chemistry*. North-Holland Personal Library (Elsevier Science, 2011).
- [11] T. Brandes. Waiting times and noise in single particle transport. *Annalen der Physik* **17**, 477 (2008).
- [12] M. Albert, C. Flindt, and M. Büttiker. Distributions of waiting times of dynamic

BIBLIOGRAPHY

- single-electron emitters. *Phys. Rev. Lett.* **107**, 086805 (2011).
- [13] L. Rajabi, C. Pörtl, and M. Governale. Waiting time distributions for the transport through a quantum-dot tunnel coupled to one normal and one superconducting lead. *Phys. Rev. Lett.* **111**, 067002 (2013).
- [14] M. Albert, G. Haack, C. Flindt, and M. Büttiker. Electron waiting times in mesoscopic conductors. *Phys. Rev. Lett.* **108**, 186806 (2012).
- [15] K. H. Thomas and C. Flindt. Waiting time distributions of noninteracting fermions on a tight-binding chain. *Phys. Rev. B* **89**, 245420 (2014).
- [16] M. Esposito, U. Harbola, and S. Mukamel. Nonequilibrium fluctuations, fluctuation theorems, and counting statistics in quantum systems. *Rev. Mod. Phys.* **81**, 1665 (2009).
- [17] G.-M. Tang, F. Xu, and J. Wang. Waiting time distribution of quantum electronic transport in the transient regime. *Phys. Rev. B* **89**, 205310 (2014).
- [18] G.-M. Tang and J. Wang. Full-counting statistics of charge and spin transport in the transient regime: A nonequilibrium Green's function approach. *Phys. Rev. B* **90**, 195422 (2014).
- [19] T. Holstein. Studies of polaron motion: Part i. the molecular-crystal model. *Annals of Physics* **8**, 325 (1959).
- [20] B. J. LeRoy, S. G. Lemay, J. Kong, and C. Dekker. Electrical generation and absorption of phonons in carbon nanotubes. *Nature* **432**, 371 EP (2004).
- [21] S. Sapmaz, P. Jarillo-Herrero, Y. M. Blanter, C. Dekker, and H. S. J. van der Zant. Tunneling in suspended carbon nanotubes assisted by longitudinal phonons. *Phys. Rev. Lett.* **96**, 026801 (2006).
- [22] R. Leturcq, C. Stampfer, K. Inderbitzin, L. Durrer, C. Hierold, E. Mariani, M. G. Schultz, F. von Oppen, and K. Ensslin. Franck-Condon blockade in suspended carbon nanotube quantum dots. *Nature Physics* **5**, 327 EP (2009).
- [23] K. Flensberg. Tunneling broadening of vibrational sidebands in molecular transistors. *Phys. Rev. B* **68**, 205323 (2003).
- [24] M. Galperin, A. Nitzan, and M. A. Ratner. Inelastic tunneling effects on noise properties of molecular junctions. *Phys. Rev. B* **74**, 075326 (2006).

- [25] M. Galperin, M. A. Ratner, and A. Nitzan. Molecular transport junctions: vibrational effects. *Journal of Physics: Condensed Matter* **19**, 103201 (2007).
- [26] L. Mühlbacher and E. Rabani. Real-time path integral approach to nonequilibrium many-body quantum systems. *Phys. Rev. Lett.* **100**, 176403 (2008).
- [27] R. C. Monreal, F. Flores, and A. Martín-Rodero. Nonequilibrium transport in molecular junctions with strong electron-phonon interactions. *Phys. Rev. B* **82**, 235412 (2010).
- [28] S. Maier, T. L. Schmidt, and A. Komnik. Charge transfer statistics of a molecular quantum dot with strong electron-phonon interaction. *Phys. Rev. B* **83**, 085401 (2011).
- [29] B. Dong, G. H. Ding, and X. L. Lei. Full counting statistics of a single-molecule quantum dot. *Phys. Rev. B* **88**, 075414 (2013).
- [30] A. Jovchev and F. B. Anders. Influence of vibrational modes on quantum transport through a nanodevice. *Phys. Rev. B* **87**, 195112 (2013).
- [31] A.-P. Jauho, N. S. Wingreen, and Y. Meir. Time-dependent transport in interacting and noninteracting resonant-tunneling systems. *Phys. Rev. B* **50**, 5528 (1994).
- [32] K. F. Albrecht, H. Wang, L. Mühlbacher, M. Thoss, and A. Komnik. Bistability signatures in nonequilibrium charge transport through molecular quantum dots. *Phys. Rev. B* **86**, 081412 (2012).
- [33] K. F. Albrecht, A. Martín-Rodero, R. C. Monreal, L. Mühlbacher, and A. Levy Yeyati. Long transient dynamics in the Anderson-Holstein model out of equilibrium. *Phys. Rev. B* **87**, 085127 (2013).
- [34] E. Perfetto and G. Stefanucci. Image charge effects in the nonequilibrium Anderson-Holstein model. *Phys. Rev. B* **88**, 245437 (2013).
- [35] R. Seoane Souto, R. Avriller, R. C. Monreal, A. Martín-Rodero, and A. Levy Yeyati. Transient dynamics and waiting time distribution of molecular junctions in the polaronic regime. *Phys. Rev. B* **92**, 125435 (2015).
- [36] E. Perfetto and G. Stefanucci. Transient dynamics in the Anderson-Holstein model with interfacial screening. *Journal of Computational Electronics* **14**, 352 (2015).
- [37] G.-H. Ding, B. Xiong, and B. Dong. Transient currents of a single molecular junction with a vibrational mode. *Journal of Physics: Condensed Matter* **28**, 065301 (2016).

BIBLIOGRAPHY

- [38] R. Seoane Souto, A. L. Yeyati, A. Martín-Rodero, and R. C. Monreal. Dressed tunneling approximation for electronic transport through molecular transistors. *Phys. Rev. B* **89**, 085412 (2014).
- [39] G. Tang, Y. Xing, and J. Wang. Short-time dynamics of molecular junctions after projective measurement. *Phys. Rev. B* **96**, 075417 (2017).
- [40] G. Tang, Z. Yu, and J. Wang. Full-counting statistics of energy transport of molecular junctions in the polaronic regime. *New Journal of Physics* **19**, 083007 (2017).
- [41] D. S. Kosov. Non-renewal statistics for electron transport in a molecular junction with electron-vibration interaction. *The Journal of Chemical Physics* **147**, 104109 (2017).
- [42] D. S. Kosov. Waiting time distribution for electron transport in a molecular junction with electron-vibration interaction. *The Journal of Chemical Physics* **146**, 074102 (2017).
- [43] A. Kamenev. *Field Theory of Non-Equilibrium Systems* (Cambridge University Press, 2011).
- [44] A. O. Gogolin and A. Komnik. Towards full counting statistics for the Anderson impurity model. *Phys. Rev. B* **73**, 195301 (2006).
- [45] L. K. Dash, H. Ness, and R. W. Godby. Nonequilibrium inelastic electronic transport: polarization effects and vertex corrections to the self-consistent Born approximation. *Phys. Rev. B* **84**, 085433 (2011).
- [46] O. Entin-Wohlman, Y. Imry, and A. Aharony. Transport through molecular junctions with a nonequilibrium phonon population. *Phys. Rev. B* **81**, 113408 (2010).
- [47] D. F. Urban, R. Avriller, and A. Levy Yeyati. Nonlinear effects of phonon fluctuations on transport through nanoscale junctions. *Phys. Rev. B* **82**, 121414 (2010).
- [48] Y. Utsumi, O. Entin-Wohlman, A. Ueda, and A. Aharony. Full-counting statistics for molecular junctions: Fluctuation theorem and singularities. *Phys. Rev. B* **87**, 115407 (2013).
- [49] L. V. Keldysh. Diagram technique for nonequilibrium processes. *Zh. Eksp. Teor. Fiz.* **47**, 1515 (1964). [*Sov. Phys. JETP*20,1018(1965)].
- [50] J. Koch and F. von Oppen. Franck-Condon blockade and giant Fano factors in transport through single molecules. *Phys. Rev. Lett.* **94**, 206804 (2005).

- [51] F. Covito, F. G. Eich, R. Tuovinen, M. A. Sentef, and A. Rubio. Transient charge and energy flow in the wide-band limit. arXiv: 1801.08440 (2018).
- [52] L. de la Vega, A. Martín-Rodero, N. Agraït, and A. L. Yeyati. Universal features of electron-phonon interactions in atomic wires. *Phys. Rev. B* **73**, 075428 (2006).
- [53] R. Avriller and A. Levy Yeyati. Electron-phonon interaction and full counting statistics in molecular junctions. *Phys. Rev. B* **80**, 041309 (2009).
- [54] T. L. Schmidt and A. Komnik. Charge transfer statistics of a molecular quantum dot with a vibrational degree of freedom. *Phys. Rev. B* **80**, 041307 (2009).
- [55] F. Haupt, T. Novotný, and W. Belzig. Phonon-assisted current noise in molecular junctions. *Phys. Rev. Lett.* **103**, 136601 (2009).
- [56] M. Kumar, R. Avriller, A. L. Yeyati, and J. M. van Ruitenbeek. Detection of vibration-mode scattering in electronic shot noise. *Phys. Rev. Lett.* **108**, 146602 (2012).

5 Self-consistent approximations

5.1 Introduction

As it has been discussed in the previous chapters, the analysis of the system dynamics provides information about the transport properties such as relaxation times or time delay between tunneling events. In chapter 3 the study of the non-interacting situation has been presented in both the coherent and the sequential regimes. However, the inclusion of interactions becomes essential to understand transport through many mesoscopic systems, such as molecular junctions, quantum dots or nanowires. For this reason, in chapter 4 has been devoted to the study of the transient transport properties through a molecular junction in the presence of strong electron-phonon interaction, by means of the DTA. In this approach most of the correlation effects are neglected, becoming an effective non-interacting approximation with renormalized parameters, although exhibiting a good agreement with numerically exact methods. Thus, the aim of this chapter is to discuss the effect of many-body interactions in the transient transport properties.

The problem of quantum transport through interacting mesoscopic systems in the time domain has been widely discussed in the literature. There is a set of works studying the problem in the sequential regime [1, 2]. However, this chapter is focused on the coherent regime, where the electron quantum nature manifests itself more clearly. This regime has been treated by means of numerical calculations like time-dependent NRG [3–8], MC [9–14] or density functional theory [15–19], among others [20, 21]. These calculations are usually computationally expensive and also limited to some parameter

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range due technical limitations. For this reason, approximations, like the ones based on diagrammatic techniques [22–29] become important to understand the leading processes influencing the system dynamics.

In this chapter a novel diagrammatic self-consistent method is presented for describing the system transport properties in the time domain. Although the algorithm is presented for studying the quench dynamics after a sudden connection to the electrodes, it could also be used to study other problems involving the time evolution of any model parameter, like the case of voltage profiles. The method is based on an iterative inversion of the Dyson equation on the discretized Keldysh contour presented in chapter 3, saving information from the previous time steps to compute the transport properties. The algorithm has several advantages. The first one is related to the information that it provides: one can obtain not only information about the steady state properties, but also on the system time response. Secondly, there is no need to approximate the initial Green functions or self-energies for the self-consistent method, since they are given by the uncoupled ones. Finally, it is also worth remarking that the perturbation theory is developed in the time domain, where the diagrams have usually simpler expressions with respect to the frequency representation.

The accuracy of the method is illustrated by analyzing the transport through systems with localized electron-electron and electron-phonon interactions, using a perturbative diagrammatic expansion of the system self-energies at different levels of approximation. As a test of these approximations, the convergence of the system properties like the mean charge, current and spectral density to their stationary values is analyzed. Also comparisons with numerically exact methods are presented. The method is finally used to discuss the open issue of possible bistable signatures in the system population and current in a molecular junction [30–32], exhibiting localized electron-phonon interactions. It is shown that the inclusions of correlation effects beyond the mean-field approximation tend to eliminate the bistable behavior for a wide range of parameters. In the final part of the chapter an extension of the algorithm for exploring the stationary limit is introduced. The accuracy of this extension is tested by calculating the stationary conductance of a system exhibiting localized electron-electron interaction.

5.2 Self-consistent procedure

In this section the self-consistent procedure in the time domain is presented. The starting point is the Dyson equation for the system NGFs

$$\hat{G}_{S,\sigma}(t, t') = [\hat{g}_{S,\sigma} - \hat{\Sigma}_{T,\sigma} - \hat{\Sigma}_{int,\sigma}]^{-1}, \quad (5.1)$$

where $\hat{\Sigma}_{int,\sigma}$ is the interaction self-energy, that will be discussed for different situations of interest. In general, this self-energy depends on the system Green functions, leading to coupled integral equations that have to be solved numerically (see section 2.2 for a more detailed discussion). In this section the solution to the coupled equations is obtained numerically by discretizing the time Keldysh contour (see chapter 3 for further details about the discretization procedure). In the discrete time mesh, the Dyson equation can be written as

$$\hat{\mathbf{G}}_{S,\sigma}(t, t') = [\hat{\mathbf{g}}_{S,\sigma} - (\Delta t)^2 (\hat{\mathbf{\Sigma}}_{T,\sigma} + \hat{\mathbf{\Sigma}}_{int,\sigma})]^{-1}. \quad (5.2)$$

The self-consistent method that is going to be introduced in this chapter is based on an iterative inversion of the discretized Dyson equation (5.2). If the time step is sufficiently small ($1/\Delta t$ much larger than any other energy parameter in the model), the Green functions in two consecutive time steps are approximately equal. Thus, the interaction self-energy can be computed accurately from the NGFs obtained in the previous time step. Note that this method does not require a self-consistent loop for each time since the convergence of the loop is achieved by a single step for sufficiently small Δt . Since the dimensions of the matrices in equation (5.2) are increased from N to $N + 2$ each time step, there is a set of undefined components in the interaction self-energy. Those components can be extrapolated with a good accuracy by copying the components a time step before as depicted in Fig. 5.1.

The discretization procedure, as well as the expressions for the inverse system Green function [33] and the tunneling self-energy are described in chapter 3. Along this chapter the interaction self-energies considered do not include any spin mixing term. The method can be, however, generalized to include that situation, which will be commented in the following chapters for superconducting nanojunctions. The most stable way of determining the $\hat{\Sigma}_{int}$ is to compute first the off-diagonal Keldysh components, using the Keldysh relations for the diagonal ones, i.e.

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Figure 5.1: Schematic illustration of the self-consistent procedure for the interaction self-energy. Left panel: $\hat{\Sigma}_{int,\sigma}$ at a given time where the shadowed region is used to highlight the final times. In the right panel the same self-energy is represented at the next time step, where the red shadowed region represents the undefined components which are extrapolated by copying the components at the previous time (in blue), preserving the time difference.

$$\begin{aligned}\Sigma_{int,\sigma}^{++}(t,t') &= -[\theta(t-t')\Sigma_{int,\sigma}^{+-}(t,t') + \theta(t'-t)\Sigma_{int,\sigma}^{--}(t,t')] , \\ \Sigma_{int,\sigma}^{--}(t,t') &= -[\theta(t-t')\Sigma_{int,\sigma}^{+-}(t,t') + \theta(t'-t)\Sigma_{int,\sigma}^{++}(t,t')] .\end{aligned}\quad (5.3)$$

Similarly to the tunneling self-energy case discussed in section , the Keldysh diagonal self-energies are not well defined for equal times. It is found that the most stable criteria is given by the condition

$$\Sigma_{int,\sigma}^{++}(t,t) = \Sigma_{int,\sigma}^{--}(t,t) = -\frac{\Sigma_{int,\sigma}^{+-}(t,t) + \Sigma_{int,\sigma}^{--}(t,t)}{2} .\quad (5.4)$$

The physical observables can be determined from the system NFGs, obtained by inverting numerically Eq. (5.2). For instance, the system population per spin is given by $n_\sigma(t) = i\mathbf{G}_{S,\sigma}^{+-}(t,t)$ and the current (2.34) in the discretized mesh is given by

$$I_\nu(t) = \sum_{\sigma=\pm} \sum_{n=1}^N [\mathbf{G}_{S,\sigma}^{+-}(t,t_n)\Sigma_{T,\sigma}^{+-}(t_n,t) - \mathbf{G}_{S,\sigma}^{--}(t,t_n)\Sigma_{T,\sigma}^{--}(t_n,t)] ,\quad (5.5)$$

where the final time is denoted as $t_N = t$. Finally, it becomes possible to determine the auxiliary spectral density resolved in time by calculating the current through weakly coupled probes to the system, following the procedure used in Refs. [13, 29]. In particular, in the limit where the coupling to this probe becomes negligible, the system NGFs are not perturbed by the presence of the probes and the occupied (empty) part of the spectral density can be written as

$$A_\sigma^{o(e)}(\omega, t) = \text{Im} \sum_{n=1}^N \frac{\pm e^{-i\omega(t-t_n)} G_{S,\sigma}^{+-(-+)}(t_n, t)}{2\pi} ,\quad (5.6)$$

The figure shows two Feynman diagrams representing self-energy corrections. The left panel shows the first-order self-energy $\hat{\Sigma}_{HF}(t, t) = \text{tadpole diagram}$, which consists of a vertical wavy line labeled U at time t , connected to a dashed circle with a spin label $\bar{\sigma}$. The right panel shows the second-order self-energy $\hat{\Sigma}^{(2)}(t, t') = \text{bubble diagram}$, which consists of two vertical wavy lines labeled U at times t and t' , connected by two horizontal dashed lines with spin labels $\bar{\sigma}$ and σ .

Figure 5.2: First (left panel) and second order (right panel) self-energy diagrams for the Coulomb interaction. The continuous line denotes the electron propagator in the system with a given spin, dashed line is used to denote the electron propagator with different spin and the wavy line represents the interaction. The single line is used for free propagators, while double line is used to denote propagators dressed with the interaction.

where the total auxiliary DOS is $A_{\sigma}(\omega, t) = A_{\sigma}^o(\omega, t) + A_{\sigma}^e(\omega, t)$. Note that with this definition $A_{\sigma}^{o(e)}$ tends to the expected long time limit. In the following of this chapter, the method described above will be applied to several systems of interest.

5.3 Electron-electron interaction: The Anderson model

The first case considered is the so-called Anderson model [34], which considers a single spin degenerate level, exhibiting local electrostatic repulsion, coupled to metallic electrodes. The model is described by the Hamiltonian in Eq. (2.44).

5.3.1 Hartree-Fock approximation

A simple way of dealing with the interaction term is at the mean-field level (also known in the literature as HF approximation), whose self-energy is schematically represented by the tadpole diagram in the left part of Fig. 5.2. This approximation is exactly solvable in the time domain and constitutes an ideal test for the accuracy of the method described in the previous section. In this approach the problem becomes equivalent to a level under a time dependent external potential

$$\epsilon_{\sigma}(t) = \epsilon_0 + U n_{\bar{\sigma}}(t), \quad (5.7)$$

where $n_{\sigma}(t)$ is the central level occupation per spin. As commented in chapter 3, the problem of an impurity level coupled to metallic leads and under a time-dependent

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potential is exactly solvable using the Keldysh method [35, 36]. For the HF case addressed in this subsection, the Keldysh Green function can be written in a very compact way as

$$G_{HF,\sigma}^{+-}(t, t') = \theta(t)\theta(t')ie^{-i[t\check{\epsilon}_\sigma(t)-t'\check{\epsilon}_\sigma(t')]}e^{-\Gamma(t+t')} \\ \times \left\{ n_\sigma(0) + \frac{1}{\pi} \int d\omega \left[\sum_{\nu=L,R} \Gamma_\nu f_\nu(\omega) \right] g_\sigma(\omega, t) g_\sigma^*(\omega, t') \right\}, \quad (5.8)$$

where

$$\check{\epsilon}_\sigma(t) = \frac{1}{t} \int_0^t d\tau \epsilon_\sigma(\tau), \quad g_\sigma(\omega, t) = \int_0^t d\tau e^{-i[\omega + i\Gamma - \check{\epsilon}_\sigma(\tau)]\tau}, \quad (5.9)$$

which are the terms containing the memory effects from the system history. The time evolution of the central level occupation is then obtained as $n_\sigma(t) = iG_{\sigma, HF}^{+-}(t, t)$ and has the form

$$n_\sigma(t) = e^{-2\Gamma t} \left\{ n_\sigma(0) + \int \frac{d\omega}{\pi} \left[\sum_{\nu=L,R} \Gamma_\nu f_\nu(\omega) \right] |g_\sigma(\omega, t)|^2 \right\}. \quad (5.10)$$

In the stationary regime the population per spin can be obtained by integrating the spectral density up to the Fermi edge, leading to

$$n_\sigma(t \rightarrow \infty) = \frac{1}{\pi} \int_{-\infty}^0 d\omega \frac{4\Gamma_L \Gamma_R}{(\omega - \epsilon_\sigma)^2 + \Gamma^2} = \frac{4\Gamma_L \Gamma_R}{\pi \Gamma^2} \left[\frac{\pi}{2} + \text{atan} \left(\frac{\epsilon_\sigma}{\Gamma} \right) \right], \quad (5.11)$$

where ϵ_σ is given by the Eq. (5.7). This expression leads to two non-linear coupled equations for the spin occupation which can have either one or three solutions [34]. In the case where the equations have only one solution, the system always relaxes to the same long time steady state independently from the initial conditions. However, in the situation where they have three different solutions, the system can converge either to a state with spin degeneracy (singlet state) or with a broken spin symmetry (magnetic solution), depending on the system initial configuration (charge bistable behavior). In the simplest situation of electron-hole symmetry ($\epsilon_0 = -U/2$), $\Gamma_L = \Gamma_R$ and zero temperature, the stationary system population can be written as $n_\sigma = 1/2 \pm \delta$ with $\delta = \arctan(U\delta/\Gamma)/\pi$, and the bistable region is found for $U/\pi\Gamma > 1$. It is worth mentioning that the bistable behavior in the mean-field approximation is known to be a pathological result, which is absent in the exact solution of the Anderson model obtained in terms of the Bethe ansatz [37–39]. It is thus expected that higher order diagrams beyond HF approximation will make the system relax, converging to the expected steady state.

One can compare the result of the numerical method proposed in section 5.2 with Eq. (5.10). In the HF approximation the self-energy is given by the tadpole diagram,

depicted in the left part of Fig. 5.2, whose expression is

$$\Sigma_{HF,\sigma}^{\alpha\beta}(t, t') = \alpha U n_{\bar{\sigma}}(t) \delta(t - t') \delta_{\alpha\beta}, \quad (5.12)$$

where $\alpha, \beta = \pm$ are the Keldysh branch indexes. In the time mesh, the Dirac delta appearing in Eq. (5.12) is converted to a Kronecker δ functional, with an additional factor $1/\Delta t$ due to the discretization. The propagators in the HF approximation can be obtained by inverting

$$\hat{\mathbf{G}}_{HF,\sigma}^{-1} = \hat{\mathbf{g}}_{S,\sigma}^{-1} - (\Delta t)^2 (\hat{\Sigma}_{T,\sigma} + \hat{\Sigma}_{HF,\sigma}), \quad (5.13)$$

and following the numerical procedure presented in the previous section. The dynamical properties of the system can be determined from $\hat{\mathbf{G}}_{HF,\sigma}$. In this mean-field approximation, the self-consistent condition is achieved by storing the charge values at the previous times for each spin and inverting Eq. (5.13). The undefined components at the final times can be extrapolated from by their values a time step before, i.e. $\Sigma_{\sigma,HF}^{\alpha\alpha}(t_{N+1}, t_{N+1}) \approx \Sigma_{\sigma,HF}^{\alpha\alpha}(t_N, t_N)$, leading to accurate results for a sufficiently small Δt .

In Fig. 5.3 the system population for each spin is represented for the electron-hole symmetric situation ($\epsilon_0 = -U/2$), where the effects of the interaction are more pronounced. Results for $U/\pi\Gamma > 1$ are shown, where the mean-field approximation predicts a long time magnetic state [34] when starting from an initially trapped spin in the system, $(n_{\uparrow}(0), n_{\downarrow}(0)) = (1, 0)$. The long time system population is given by the solution of the Eqs. (5.7) and (5.11), and represented by the arrows in the upper panel of Fig. 5.3. As shown, the numerical solution and the exact expression of Eq. (5.10) represented by symbols, are in very good agreement, constituting a proof for the methods accuracy.

Finally, it is worth mentioning that in Ref. [40] the first order perturbation expansion in U/Γ has been treated in a not fully self-consistent way in time domain, comparing the system occupation with numerical results obtained from MC simulations. The authors considered the initial condition to be spin degenerate $(0, 0)$, for avoiding the magnetic long time solution of the HF approximation. They found a good agreement for small to moderate U/Γ values, while the HF solution deviates from the exact calculations when increasing the interaction strength (with an average charge $n(t)$ overpassing the electron-hole symmetric stationary value at large U/Γ). This pathological behavior is due to the approximate character of their HF calculation and is corrected within a fully self-consistent HF treatment (5.10), where the average charge $n(t) = 1/2 \sum_{\sigma} n_{\sigma}(t)$ tends

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Figure 5.3: Time evolution of the central level charge for the Anderson model for the up (solid lines) and down (dashed lines) spins starting from an initial magnetic configuration, $(n_{\uparrow}(0), n_{\downarrow}(0)) = (1, 0)$. In the top panel the results for the HF approximation (blue lines) are compared to the analytic expression (black points), given by Eq. (5.10) whose stationary value is represented by the arrows for $U/\Gamma = 4$, $\epsilon_0 = -U/2$ and in the infinite bandwidth limit. The red lines correspond to the second-order self-energy case. In the bottom panel the n_{σ} is represented calculated using the second order self-energy for increasing values of the interaction, $U/\Gamma = 4$ (red), 6 (green) and 8 (black).

to the correct electron-hole symmetric limit for all U/Γ values.

The long time magnetic moment solution at zero temperature is known to be unphysical, according to the exact model solution [37–39], and expected to be relaxed when including electronic correlations in an appropriate way. In the next section the electron correlation effects are analyzed beyond the HF approximation in the transient regime.

Figure 5.4: Short time level population compared to the MC simulations in Ref. [40] (symbols) for $U/\Gamma = 8, 4, 2, 1$ and 0 , from top to bottom. The remaining parameters are $\epsilon_0 = -U/2$, $T = 0.2\Gamma$, $V = 0$, $D = 10\Gamma$ and the initial occupation $(0, 0)$.

5.3.2 Effects of correlation beyond mean-field

As commented in the previous subsection, the mean-field approximation (self-consistent first order expansion in U/Γ for the electron self-energy) only predicts a system energy shift due to the interaction. In order to include electron correlation effects higher order terms have to be included. In this chapter I will restrict the calculations to the second order term of the interaction self-energy in U/Γ , represented in the right panel of Fig. 5.2, although more complex self-energies can be implemented within this algorithm. As commented in section 2.3, the second order term in the perturbative expansion of the self-energy, which describes the electron hole pair creation, includes correlation effects in a rather precise way [41]. In fact, it can be shown that the exact self-energy in the atomic limit and the $\hat{\Sigma}_X$ have the same functional dependence on the U parameter and on energy. This fact has been used to develop different interpolative approaches, providing reasonable results for a wide range in the electron-electron coupling strength [42–45].

Along this section I will focus on the $\epsilon_0 = -U/2$ case, where correlations effects are more pronounced. As it has been shown in section 2.3, a proper inclusion of the second

Figure 5.5: long time DOS for $U/\Gamma = 4$ (red curves) and $U/\Gamma = 8$ (blue curves). Top panel: comparison between the numerical calculations (solid lines) and the second order perturbation expansion (dotted lines). Bottom panel: comparison between the numerical calculations (solid lines) and NRG calculations from Ref. [46] (dashed lines). The insets show the low frequency convergence of the Kondo resonance, fulfilling the FSR for $\omega = 0$.

order self-energy yields a spectral density in a good agreement with exact calculations. Actually, in the equilibrium situation it is able to describe accurately the position and the height of the charge peaks. It also describes the appearance of the Kondo resonance at the Fermi edge, although its width is somewhat overestimated for $U/\Gamma \gg 1$. Finally, it is worth commenting that the second order self-energy diagrams are calculated using the HF propagators, given in the discretized mesh by inverting Eq. (5.13). In contrast, when using the full dressed propagators, which in principle include information about higher order diagrams, the good analytic properties of the second order self-energy commented above are lost, leading to a poorer description of the spectral density [47].

The off-diagonal Keldysh components of the second order self-energy in the time domain are given by expressions

$$\begin{aligned}\Sigma_{\sigma}^{(2)+-}(t, t') &= -U^2 \mathbf{G}_{HF, \sigma}^{+-}(t, t') \mathbf{G}_{HF, \bar{\sigma}}^{+-}(t, t') \mathbf{G}_{HF, \bar{\sigma}}^{-+}(t', t), \\ \Sigma_{\sigma}^{(2)-+}(t, t') &= -U^2 \mathbf{G}_{HF, \sigma}^{-+}(t, t') \mathbf{G}_{HF, \bar{\sigma}}^{-+}(t, t') \mathbf{G}_{HF, \bar{\sigma}}^{+-}(t', t),\end{aligned}\quad (5.14)$$

where the HF propagators are calculated by inverting Eq. (5.13). As described in the section 5.2, the remaining self-energy components can be determined through the usual Keldysh relations (5.3), with the criteria for equal times given in Eq. (5.4). The propagators in Keldysh space can now be evaluated by inverting Eq. (5.2) with $\hat{\Sigma}_{int, \sigma} = \hat{\Sigma}_{HF, \sigma} + \hat{\Sigma}_{\sigma}^{(2)}$.

The effect of correlations on the electronic and transport properties of the system will be analyzed in the following of this section. In the top panel of Fig. 5.3 the evolution of $n_{\sigma}(t)$ is shown for the case discussed in the previous section, with an initially trapped spin, where the HF approximation predicts a magnetic solution. As shown, when including correlation effects (electron-hole pair creation) the system evolves to a non-magnetic solution corresponding to a singlet state in the stationary limit.

In the bottom panel of Fig. 5.3 the evolution of $n_{\sigma}(t)$ is represented for the same initial magnetic configuration and increasing U/Γ values. It is found that for $U/\Gamma \gtrsim 8$ the initial localized spin is no longer screened by the electrodes, tending to a magnetic solution. The frontier between the monostable and the bistable regimes cannot be set precisely due to that, close to the transition point the method becomes very sensitive to the time step and the electrodes bandwidth. The long time magnetic state indicates a shortcoming of the approximated self-energy for sufficiently large interaction strength, which is expected to be corrected when including higher order terms in the perturbation expansion. The singlet stationary state is, however, always reached when starting from a spin degenerate initial state, i.e. (0,0) or (1,1).

The self-consistent evolution is compared with available calculations for the system population using exact MC simulations [40]. In Fig. 5.4 it is shown the short time evolution of $n_{\sigma}(t)$ for increasing U/Γ and an initial empty level. The results of MC simulations are represented by symbols. As can be observed, there is a good agreement even in the strongly interacting case.

Figure 5.6: Time evolution of the spectral density for $U/\Gamma = 4$ (top panel) and 8 (middle panel). In the bottom panel the height of the central peak is shown for these two cases.

The long time DOS is represented in Fig. 5.5, comparing the numerical results with the expected stationary limit in the upper panel, finding a remarkable agreement. The initial system occupation is chosen to be $(0,0)$, to avoid the system to get trapped in a long time magnetic state. The inset shows the convergence of the Kondo resonance, fulfilling the FSR $A(\omega = 0) = 1/\pi\Gamma$. In the bottom panel of Fig. 5.5 the long time results are compared with the exact NRG results of Ref. [46], showing an overall agreement, except for the Kondo peak whose width is slightly overestimated for $U/\Gamma \gg 1$. To describe the exponential decrease of the Kondo resonance with increasing U/Γ , in principle all order diagrams should be included.

On the other hand, the formation in time of the charge peaks and the Kondo resonance is analyzed in Fig. 5.6. This issue has been investigated in some previous works [22, 49]. One could expect a convergence time of the order of T_K^{-1} , where T_K is the Kondo temperature given in Eq. (2.45). In Fig. 5.6 the spectral density is plotted in the time domain for two different electron-electron interaction strengths, $U/\Gamma = 4$ (top panel) and 8 (middle panel). As shown, the Kondo resonance takes longer to develop for increasing U/Γ values.

Figure 5.7: Short time current comparing the numerical results (solid lines) with the ones obtained using MC technique from Ref. [40] (symbols). Results for $U/\Gamma = 0, 4$ and 8 (from top to bottom) are shown, with $V = 10\Gamma$. The remaining parameters are the same as in Fig. 5.4.

This is analyzed in the bottom panel of Fig. 5.6 where the height of the Kondo resonance, $A(\omega = 0, t)$, is plotted. For these two cases, it is found that the ratio between the exact Kondo temperatures (2.45) is $T_K(U/\Gamma = 4)/T_K(U/\Gamma = 8) \simeq 3.4$, which means that the resonance formation for $U/\Gamma = 8$ would take about 3.4 times longer than for the $U/\Gamma = 4$ case. Remarkably, the ratio of the Kondo peak formation for the calculations in Fig. 5.6 is in good agreement with the expected value, although slightly underestimated due to the overestimation of the central resonance (see the inset in the bottom panel of Fig. 5.5).

The non-equilibrium situation is analyzed in Fig. 5.7, comparing the results from the method introduced in this chapter with the ones from the MC simulations from Ref. [40], finding a quantitative agreement for moderate coupling strengths. For very large interaction strengths (green line) the second order self-energy tends to underestimate the current value, although capturing the general trend.

Finally in Fig. 5.8 the asymptotic $I(V)$ characteristic is shown for increasing U/Γ values compared to the MC results of Ref. [48]. As can be observed, there is a good overall agreement specially for $V > \Gamma$. However, a discrepancy is found at low voltages

Figure 5.8: Asymptotic current value compared to exact MC results from Ref. [48]. I show results for increasing values of the electron-electron interaction, $U/\Gamma = 0, 4, 6, 8, 10, 12$ and 14 from top to bottom. The remaining parameters are $\epsilon_0 = -U/2$, $T = 0.2\Gamma$ and $D = 10\Gamma$.

specially at large U/Γ values. The discrepancy for small voltage values is due to two main reasons. The first one is related to the fact that in Ref. [48] the authors have considered a rather short time ($t \sim 1/\Gamma$) to compute the stationary properties, much shorter than the convergence time in the equilibrium situation (see for instance Fig. 5.6). On the other hand, the second order self-energy tends to overestimate the Kondo resonance, overestimating also the current at low voltages. This shortcoming would be corrected in this electron-hole symmetric case by including higher order diagrams, as shown in Ref. [50]. In this reference the authors included the fourth order diagrams in the stationary limit, which describe the splitting of the Kondo resonance at finite bias voltage.

5.4 Electron-phonon interaction: Spinless Anderson-Holstein model

Another situation of interest that it is going to be analyzed is the one involving transport through a resonant level coupled to a localized phonon mode. This model is described by

Figure 5.9: Second order self-energy diagrams for the spinless Anderson-Holstein interaction. a. Hartree (left panel) and exchange (right panel) diagrams for the Born approximation, using the bare phonon propagator (wavy line). In b. similar approximations are shown with two different phonon self-energies depicted below: RPA [51], where the electronic propagators are considered to be undressed, and the self-consistent MIGDAL [52], where the electronic propagators are fully dressed.

the so-called spinless Anderson-Holstein model whose Hamiltonian is given by Eq. (2.52). In the previous chapter the transient transport properties of this model have been analyzed in the polaronic regime by means of the DTA, which is an effective non-interacting approximation with renormalized tunneling amplitudes [28]. However, in this section the transient dynamics of the spinless Anderson-Holstein model will be analyzed using second order self-consistent approximations, which provide accurate results for weak to moderate electron-phonon coupling strengths. Moreover, these approximations contain information of the leading electron-electron correlation effects mediated by the phonon mode.

5.4.1 Hartree approximation

As in the previously discussed Anderson model, the analysis is begun with the self-consistent mean-field approximation. This approach is given by the tadpole diagram depicted in the left part of Fig. 5.9a. Its Keldysh components are given by

$$\Sigma_H^{\alpha\beta}(t, t') = \alpha \delta(t - t') \delta_{\alpha\beta} \lambda^2 \int d\tau [d^{++}(t, \tau) - d^{+-}(t, \tau)] n(\tau), \quad (5.15)$$

where $n(t)$ is the self-consistent system charge and $\hat{d}$ is the free phonon propagator, given in Keldysh space by Eq. (2.55). In the following I will consider, the phonon mode to be in thermal equilibrium when using its bare propagator, with a population described by the Bose-Einstein distribution function $n_p = (e^{\omega_0/T} - 1)^{-1}$. Since most of the calculations are performed at zero or very small temperature, $n_p = 0$. Using the Keldysh relations, Eq. (5.15) can then be written as

$$\Sigma_H^{\alpha\beta}(t, t') = \alpha \lambda^2 \delta(t - t') \delta_{\alpha\beta} \int_0^t d\tau d^R(t, \tau) n(\tau), \quad (5.16)$$

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where $d^R(t, t')$ is the retarded free phonon propagator, whose expression is given by

$$d^R(t, t') = -2\theta(t - t') \sin[\omega_0(t - t')] . \quad (5.17)$$

It is worth noticing that the electron-phonon interaction introduces retardation effects even in the Hartree approximation, differently from the electron-electron interaction situation discussed in the previous section. These effects are important in the transient regime, mostly in the situation when the electron and the phonon dynamics are equally fast ($\omega_0 \sim \Gamma$), decreasing when increasing ω_0 . For $\omega_0 \gg \Gamma$ the phonon dynamics is much faster than the electronic one, and it can be considered that the phonon effects are instantaneous. In this limit ($\omega_0 \gg \epsilon_0, \Gamma$), the Hartree term can be written as

$$\Sigma_H^{\alpha\beta}(t, t') \approx -\alpha\delta(t - t')\delta_{\alpha\beta}\frac{2\lambda^2}{\omega_0}n(t) , \quad (5.18)$$

which induces a renormalization of the system energy, depending on the system instantaneous population as $\epsilon(t) = \epsilon_0 + (2\lambda^2/\omega_0)n(t)$. This limit has been analyzed in Ref. [31]. Similarly to the Anderson model discussed in the previous section, the time-dependent population in the phonon adiabatic regime can be exactly obtained as

$$n(t) = e^{-2\Gamma t} \left\{ n(0) + \int \frac{d\omega}{\pi} \left[\sum_{\nu=L,R} \Gamma_\nu f_\nu(\omega) \right] |g(\omega, t)|^2 \right\} , \quad (5.19)$$

where

$$\check{\epsilon}(t) = \frac{1}{t} \int_0^t d\tau \left[\epsilon_0 + \frac{2\lambda^2}{\omega_0} n(\tau) \right], \quad g(\omega, t) = \int_0^t d\tau e^{-i[\omega + i\Gamma - \check{\epsilon}(\tau)]\tau} . \quad (5.20)$$

In the stationary regime, the population can be written as

$$n(t \rightarrow \infty) = \frac{4\Gamma_L\Gamma_R}{\pi\Gamma^2} \left\{ \frac{\pi}{2} + \text{atan} \left[\epsilon_0 - \frac{2\lambda^2}{\omega_0} n(t \rightarrow \infty) \right] \right\} . \quad (5.21)$$

This equation has either one or three solutions, depending on the model parameters, reflecting the existence of two different regimes: one where only a long-time state is possible (monostable regime) and the other one where the system can converge to three different long time solutions, depending on the initial configuration (bistable regime). For the electron-hole symmetric point ($\epsilon_0 = \lambda^2/\omega_0$) and for a symmetrically coupled system ($\Gamma_L = \Gamma_R$), the charge bistable regime is found for $\lambda^2/\pi\omega_0\Gamma > 1$.

Both regimes are illustrated in Fig. 5.10, where the system population has been computed using the algorithm described in section 5.2. As in the Anderson model, the charge evolves to the stationary regime given by the solution of Eq. (5.21) and denoted by the

Figure 5.10: Evolution of the central level charge as a function of time for an initially full (top panel) and empty level (bottom panel). The dashed line represents the evolution using an instantaneous Hartree term (5.18), while the solid one corresponds to the full Hartree calculation (5.16). The dependence on phonon frequency is also illustrated for the values: $\omega_0 = 8\Gamma$ (red), 2Γ (green) and Γ (blue). The rest of parameters are $\lambda = 1.5\Gamma$, $V = 0$ and the central level is set to $\epsilon_0 = \lambda^2/\omega_0$, thus preserving electron-hole symmetry.

arrows in the figure. Fig. 5.10 also illustrates the effect of retardation effects on the Hartree self-energy, showing that the evolution progressively deviates from the adiabatic approximation given in Eq. (5.18) when reducing the phonon frequency. Moreover, the system population exhibits oscillations due to phonon retardation effects, which induce a superposition of oscillations with a period $2\pi/\omega_0$ but with amplitudes proportional to the instantaneous system population. The charge oscillations are damped at times $t \gg 1/\Gamma, 2\pi/\omega_0$, indicating the losing of coherence. However, in the adiabatic regime ($\omega_0 \gg \Gamma$), where the phonon and electron dynamics decouple, the oscillations persist on time. They are mostly visible for the $n = 1$ situation (black curve of the top panel of Fig. 5.10). It is worth commenting that an additional damping could be introduced by dressing the phonon propagator in the Hartree term, given in Eq. (5.16).

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Finally, as it can be seen in Fig. 5.10, for the smallest value of ω_0 the system can reach two different charge values, depending on the initial configuration. This charge bistable behavior has been predicted before in Refs. [30, 31] by means of the self-consistent Hartree approximation. The possibility of a bistable behavior was suggested some time ago [53–55] and has experienced a recent revival by studying the mechanical properties [12, 56–58], which bears clear signatures of a transition to a bistable regime. It is important to mention that this phonon displacement bistable behavior does not imply necessarily a bistable behavior for the charge or the current, as predicted by the mean field approximation. As even this simple spinless Anderson-Holstein model is not exactly solvable, this issue is still under debate [10, 32, 59]. It seems plausible that, at least for equilibrium conditions and $T = 0$ correlation effects destroys the charge and current bistability predicted by the mean field solution [12]. This issue is addressed in the following section.

5.4.2 Effects of correlation beyond Hartree approximation

Differently from the Anderson model previously discussed, the exact solution of the spinless Anderson-Holstein model is still unknown. Thus, the possibility of finding a charge bistable regime as predicted by the mean-field approximation and the role of electron correlations are still open questions. These issues are partially addressed in this section using three different self-consistent approximations, differing on the way the phonon propagator is treated in the exchange term.

Firstly, I consider the simplest situation where the phonon propagator is kept undressed, known as the self-consistent Born approximation and given by the diagrams depicted in the left panel of Fig. 5.9. The electron propagators are, however, fully dressed with the interaction. Then, the electron self-energies can be written as

$$\begin{aligned}\Sigma_H^{\alpha\beta}(t, t') &= -2\alpha\lambda^2\delta_{\alpha\beta}\delta(t-t')\int_0^t d\tau \sin[\omega_0(t-\tau)]n(\tau), \\ \Sigma_X^{\alpha\beta}(t, t') &= i\alpha\beta\lambda^2 G^{\alpha\beta}(t, t')d^{\alpha\beta}(t, t'),\end{aligned}\tag{5.22}$$

where $G^{\alpha\beta}$ denotes the Keldysh components of the fully dressed electron propagators and $n(t)$ is the final self-consistent charge. In this case, the interaction self-energy entering Eq. (5.2) is given by $\hat{\Sigma}_{int} = \hat{\Sigma}_H + \hat{\Sigma}_X$.

This fully self-consistent approximation can be straightforwardly implemented within the numerical procedure of section 5.2, where the self-energies of Eqs. (5.22) are com-

Figure 5.11: Evolution of the central level charge as a function of the time for an initially empty (dashed line) and initially full level (solid line). The blue and red lines correspond to the Hartree and self-consistent Born approximation, respectively. The parameters are the same as in Fig. 5.10 with $\omega_0 = \Gamma$.

puted from the final Green functions and then stored for each time step. In Fig. 5.11 the system population is shown for a case where the Hartree approximation predicts a charge bistable behavior. As can be observed, the inclusion of correlations eliminates the bistable behavior predicted by the mean-field approximation, leading to the convergence of the system population to the same long time solution independently from the initial configuration. This behavior is maintained up to quite large values of $\lambda^2/\omega_0\Gamma$. At very large values of $\lambda^2/\omega_0\Gamma$, when approaching the polaronic regime, the Born approximation breaks down, indicating the need to include higher order diagrams. For addressing this regime another kind of approximations has to be used, like for instance in the lines of the ones discussed in Refs. [32, 60–63]. These results suggest that the population bistable behavior of the central level charge predicted in Refs. [30, 31] is a spurious feature of the mean field approximation which disappears when correlation effects are included. This is in agreement with the predictions of exact numerical calculations of Ref. [12], at least for the equilibrium case and at sufficiently low temperatures.

In order to improve the range of validity of the Born approximation, the phonon

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renormalization due to the coupling to the electrons should be included. This coupling induces a phonon frequency renormalization and the appearance of a natural phonon lifetime as described in Ref. [64]. One of the simplest ways of including the electron backaction on the phonon degree of freedom is through a RPA-like [51] approach. In this approximation the phonon propagator is dressed using the corresponding Dyson equation as

$$\hat{D} = (\hat{d}^{-1} - \hat{\Pi})^{-1}, \quad (5.23)$$

where $\hat{D}(t, t') = -i \langle T_c[\varphi(t)\varphi^\dagger(t')] \rangle$, with $\hat{\varphi} = a + a^\dagger$, $\hat{d}^{-1}$ is the inverse free-phonon propagator and $\hat{\Pi}$ is the phonon self-energy given by

$$\Pi^{\alpha\beta}(t, t') = -i\alpha\beta\lambda^2 G^{\alpha\beta}(t, t') G^{\beta\alpha}(t', t). \quad (5.24)$$

The phonon self-energy is depicted in the bottom part of the right panel of Fig. 5.9 for two different schemes: the RPA, where the electron lines are kept undressed, and the Migdal approximation, where the electron propagators entering in Eq. (5.24) are fully dressed with the interaction.

As for the electronic propagator, Eq. (5.23) is going to be discretized in the Keldysh contour. It requires to find a stable expression for the inverse phonon propagator as the one found for the electronic case (3.15). To the best of my knowledge, this issue has not been previously addressed in the literature. The difficult of this task lies on the fact that the discretized free phonon propagator given in Eq. (2.55) is singular and not invertible. Thus, the method requires a regularization of the singularity, which is discussed in appendix B, finding

$$\mathbf{d}^{-1} = \frac{1}{2\delta} \left(\begin{array}{ccc|ccc} h_0^+ & -1 & & & & \\ -1 & h & -1 & & & \\ & \ddots & \ddots & \ddots & & \\ & & -1 & h & -1 & \\ & & & -1 & h_N^+ & \\ & & & & c & \\ \hline & & & & c & \\ & & & & h_N^- & 1 \\ & & & & 1 & -h & 1 \\ & & & & & \ddots & \ddots & \ddots \\ & & & & & & 1 & -h & 1 \\ & & & & & & & 1 & h_0^- \end{array} \right)_{2N \times 2N}, \quad (5.25)$$

where $\delta = \Delta t \omega_0$ and $h = 2(1 - \delta^2/2)$. The information about the initial phonon state is encoded in the components $h_0^\pm = \pm h/2 + i\delta(1 + \rho_0^2)/(1 - \rho_0^2)$ and $h_{0N} = -2i\delta\rho_0/(1 - \rho_0^2)$,

Figure 5.12: Long time spectral density for the self-consistent MIGDAL (solid line), RPA (dashed line) and Born (dotted line) approximations, compared to NRG calculations (yellow dots). In the inset we show the convergence of the central resonance to the FSR. The parameters are: $\lambda = 1.5$, $\omega_0 = 2$, $V = 0$, $\epsilon_0 = \lambda^2/\omega_0$, $\Gamma = 1$ and $D = 30$.

where $\rho_0 = n_p(0)/[n_p(0) + 1]$ and $n_p(0)$ is the initial phonon population. In the case of an initially equilibrated phonon population, $\rho_0 = e^{-\omega_0/k_B T}$. The regularization procedure requires introducing an infinitesimal quantity η which enters in the matrix elements connecting both Keldysh branches: $c = -2i\delta/\eta$ and $h_N^\pm = \pm h/2 - c$. The parameter, η , can be interpreted as a small phonon relaxation rate which has to be taken such as $1/\eta \gg t, 1/\omega_0$ for a good convergence to the expected free propagator when inverting Eq. (5.25).

It should be noticed that this problem with the inversion of the free phonon propagator has been avoided in the literature by neglecting fast oscillating terms of the type $\langle \hat{T}_c[a(t)a(t')] \rangle$ and $\langle \hat{T}_c[a^\dagger(t)a^\dagger(t')] \rangle$ in the diagrammatic expansion of $\hat{D}$ (for example see the inverse expression provided in Ref. [33]). This approximation corresponds to the so-called rotating wave approximation, which describes the regime where the phonon timescale is much faster than the electron dynamics ($\omega_0 \gg \Gamma, \lambda$) [65, 66].

In Fig. 5.12 the long-time spectral function is shown for the three approximations considered in this section using the same parameters as in Fig. 5.11 with $\omega_0 = 2\Gamma$. It

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Figure 5.13: Short time current, comparing the three approximations with the exact MC calculations from Ref. [9]. Results for the three approximations described in the text: self-consistent Born (dotted line), RPA (dashed line) and MIGDAL (solid line) approximations are shown. Parameters: $\lambda = 8$, $\omega_0 = 10$, $D = 20$, $T = 0.2$, $\Gamma = 1$ and $V = 0$ (left panels) and 4 (right panels)

corresponds to a case with a rather strong electron-phonon interaction ($\lambda^2/\omega_0\Gamma \sim 1$), although still far from the strong polaronic regime. The results of the three approximations are compared with a NRG calculation (see appendix A for numerical details) showing quantitative agreement. Remarkably, the RPA and the MIGDAL approximations eliminate the pathological feature of the second order self-energy at $\omega = \omega_0$ [60, 67]. Moreover, they are able to describe the phonon renormalization, observed in the first sideband, to higher energies. It is worth mentioning that these two approximations are, however, not able to reproduce the energy shift to smaller energies in the regime $\omega_0 \ll \Gamma$. For describing that regime, approximations beyond the second order for the phonon renormalization have to be developed. Additionally, an extra feature can be observed at $\sim 2\omega_0$, associated to the appearance of a second phonon sideband. The inset illustrates the convergence at low frequencies to the expected FSR for equilibrium and electron-hole symmetric situation [68], showing that the MIGDAL approximation tends to underestimate the central peak width.

In Fig. 5.13 results for the current evolution are shown in the regime $V < \omega_0$ and

towards the polaronic regime ($\lambda^2/\omega_0\Gamma = 6.4$). Results for the left, right and symmetrized currents are shown, comparing them with the ones from MC simulations presented in Ref. [9]. In the equilibrium situation where the non-equilibrium effects of phonons are expected to be smaller (left panels), the three approximations provide accurate results for the current evolution, although the RPA and the MIGDAL results seem to exhibit a better agreement with the MC results. In the $\omega_0 > V \neq 0$ regime, (right panels of Fig. 5.13) where the non-equilibrium phonon effects are expected to be still small, the RPA describes accurately the current dynamics while the other two approaches deviate from the numerical results. In the case of the MIGDAL approximation, the origin of the disagreement could be due to the poorer description of the spectral properties at low frequencies as shown in the inset of Fig. 5.12. For the self-consistent Born approximation, the disagreement is due to the absence of the non-equilibrium phonon effects.

Finally in Fig. 5.14 the same results are represented in the regime $V > \omega_0$, where the non-equilibrium phonon dynamics is expected to become important. As shown, the MIGDAL approximation describes rather accurately the current evolution. It means that this approximation is able to describe accurately the average transport properties in the voltage window. The RPA calculations show a deviation with respect to the exact calculations for the electron-hole symmetric situation (left panels) becoming better at low transmission factors. Finally, the self-consistent Born approximation leads to results deviating, in any of the cases represented in Fig. 5.14, from the exact calculations. This is indicative of the importance of a good description of the phonon dynamics, mainly for large electron-phonon coupling strengths and for $V > \omega_0$.

5.5 Electron-electron and electron-phonon interactions

The final situation that is analyzed in this chapter is the spin-degenerate Anderson-Holstein model, where both electron-electron and electron-phonon interactions are present in the system. The model is described by the Hamiltonian

$$H = \sum_{\sigma=\uparrow,\downarrow} H_{0,\sigma} + V_{e-e} + H_{ph} + V_{e-ph} \quad (5.26)$$

where $H_{0,\sigma}$ is the non-interacting part given by Eq. (2.1), $V_{e-e} = U n_{\uparrow} n_{\downarrow}$, $H_{ph} = \omega_0 a^{\dagger} a$ and $V_{e-ph} = \lambda(a + a^{\dagger}) \sum_{\sigma} n_{\sigma}$.

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Figure 5.14: Same quantities as in Fig. 5.13 for $V = 32\Gamma$ and $\epsilon_0 = \lambda^2/\omega_0$ (left panels) and $\epsilon_0 = \lambda^2/\omega_0 - 16$ (right panels).

Similarly to the previously discussed situations, the mean-field approximation induces a renormalization of the system energy, which in the adiabatic phonon regime can be written as $\epsilon_\sigma(t) = \epsilon_0 + Un_\sigma(t) - 2\lambda^2/\omega_0 n_\sigma(t)$. In this regime the system population evolution can be solved exactly finding similar expressions to Eqs. (5.10) and (5.19). In the stationary situation two different regimes can be found: a regime with only a long time charge state (monostable regime) and the other one with three possible steady state populations (bistable regime). Similarly to the previous examples analyzed, the inclusion of correlation effects tend to make the system relax to the same steady state independently from the initial configuration. In this case the interaction self-energy is the sum of the self-energies of the two kinds of interactions considered, $\hat{\Sigma}_{int} = \hat{\Sigma}_{e-e} + \hat{\Sigma}_{e-ph}$, depicted in Figs. 5.2 and 5.9, respectively.

In the panels of Fig. 5.15 the long time spectral density is compared to the exact NRG results from Ref. [69] for the MIGDAL approximation (top panel) and the RPA (bottom one) for the phonon self-energies. For the smallest electron-phonon coupling strength (in red) both approximations exhibit an overall agreement, although they do not describe the broadening of the central resonance due to the phonon interaction, as given by exact calculations of Refs. [60, 69, 70]. The agreement becomes poorer when increasing λ (blue curves). In fact, the second order approximations are not able to describe properly the

Figure 5.15: Long time DOS (lines) compared with equilibrium NRG results (symbols) from Ref. [69], for two different electron-phonon coupling parameters: $\lambda = 1.89$ (red) and $\lambda = 3.14$ (blue). I represent results for the MIGDAL in top panel and for the RPA in the bottom one. The remaining parameters are $U = 6.3$, $\omega_0 = 3.14$, $\epsilon_0 = \lambda^2/\omega_0 - U/2$, $\Gamma_R = \Gamma_L = 0.5$, $V = 0$ and an initial condition $(0, 0)$.

transition to the insulating phase, appearing for $\lambda^2/\omega_0 \gtrsim U/2$. To explore this parameter regime, one would need to develop an approximation correctly interpolating between the perturbative and the strong polaronic regimes. The appearance of the insulating phase in the time domain has already been discussed in Ref. [29] by means of the non-crossing approximation, although this approximation does not describe the convergence to the FSR at the equilibrium situation.

Finally, the transient spectral function is represented in Fig. 5.16 for $U/\Gamma = 8$ and $\lambda/\Gamma = 0$ (top panel) and $\lambda/\Gamma = 2$ (middle panel) for the RPA. Similar results are obtain for the MIGDAL case. As shown, even far from the insulating phase, in the Kondo

Figure 5.16: Time evolution of the equilibrium DOS for $U = 8$ and $\lambda = 0$ (top panel) and $\lambda = 2$ (middle panel). The central peak height evolution is shown in the bottom panel for the $\lambda = 0$ case in red and the $\lambda = 2$ case in blue. The remaining parameters are $\omega_0 = 2$, $\epsilon_0 = \lambda^2/\omega_0 - U/2$ and $\Gamma = 1$

dominates the electron-phonon interaction modifies significantly the system dynamics. Firstly, it leads to significantly longer convergence times, as illustrated in the bottom panel where the height of the central resonance, $A(\omega = 0, t)$, is represented. It also exhibits additional oscillations with a period $\sim 2\pi/\omega_0$, indicating that, even in the Kondo dominated regime, the resonance at the Fermi edge is significantly affected by the phonon dynamics. Moreover the decaying time of this oscillations has been enlarged due to the electron-electron interaction (not shown), indicating that the electron-electron interaction increases phonon retardation effects.

5.6 Calculation of the steady state properties

Finally, I would like to close this chapter discussing a generalization of the method introduced in this chapter to the stationary regime, that will be subject of future work. Firstly note that the numerical method described in section 5.2 can be computationally heavy

Figure 5.17: Scheme of the extension to the method to calculate steady state properties where the left panel illustrate the convergence to the steady state and the right one the calculations of the stationary properties for decreasing voltage values, starting from the NGFs and the self-energies computed at the end of the first step.

and time consuming when computing stationary properties: it may require in principle a full time calculation for any set of parameters. Moreover, a lot of unneeded information is obtained about the system dynamics. A more efficient way to proceed is to make the system evolve to its steady state, where the stationary Green functions and self-energies are computed. Then, a similar self-consistent method can be applied by changing the desired model parameter. This method converges and provides accurate results if the parameter is changed adiabatically. So, the self-energy is well approximated by the one with the previous model parameters and the convergence of the self-consistent method is reached by a single iteration, as in the case of the time dependent algorithm discussed in the section 5.2. The extension of the method is schematically explained in Fig. 5.17 for a I-V curve, showing the two steps of the numerical calculations. Furthermore, the numerical calculations can be significantly shorter and their precision improved if the part of the transient part of the charge transfer is subtracted.

The power of the method is illustrated by calculating the conductance as a function of the voltage for the Anderson model, shown in the top panel of Fig. 5.18. In the equilibrium situation, all the curves converge to the unit of conductance, consistent with the fulfilling of the FSR (see inset) as discussed in section 5.3. At low voltages a linear decay is observed with an slope increasing with the electron-electron interaction strength, which is a signature of the width decreasing of the Kondo peak and consistent with the exact calculations from [41]. However, the increasing of the conductance at $V \sim \Gamma$ for large U/Γ is missing in the second order approach of the self-energy. This is due to the

Figure 5.18: Top panel: conductance for $U/\Gamma = 0$ (black), 4 (red), 6 (yellow), 8 (green) and 10 (blue) for the electron-hole symmetric situation $\epsilon_0 = -U/2$. The inset shows the convergence at low bias voltage. Lower panel: asymptotic spectral density for increasing voltage values and $U/\Gamma = 8$.

splitting of the Kondo resonance, which is a higher order process [50]. At voltages $V \sim U$ an increase of the conductance appears when the charge peak enter the voltage window. In the bottom panel of the figure 5.18 the long time spectral density is represented for increasing voltage values, showing the disappearance of Kondo resonance with increasing voltage.

5.7 Conclusions

In this chapter I have presented an efficient way to calculate the transient transport properties of interacting nanojunctions. To illustrate the accuracy of the method, different self-consistent diagrammatic approximations have been implemented to study the transient and the steady state regimes. The method can be generalized to more com-

plex self-energies. Along the chapter three paradigmatic problems have been considered: the Anderson model for the electron-electron interaction, the spinless Anderson-Holstein model for electron-phonon interaction and the spin-degenerate Anderson-Holstein model, where both interactions are present. For the Anderson model I have shown that the long-time magnetic solution tends to relax when including correlation effects through a second order perturbation expansion. Moreover, the second order self-energy has been shown to provide accurate results in both the transient and the stationary regimes. In the case of the spinless Anderson-Holstein model I have shown that the electron correlations tend to destroy the charge and current bistable behavior for a wide range of parameters. Although this is indicative of the absence of a charge bistable behavior for a wide range of parameters, it is not inconsistent with another kinds of bistable behaviors found in the mechanical properties such as in the displacement spectrum [12, 58]. Moreover, two different schemes for dressing the phonon propagator (denoted as RPA and MIGDAL) have been introduced, illustrating the importance of describing the non-equilibrium population of phonons in the strong coupled regime and for voltages bigger than the phonon frequency. For the spin-degenerate Anderson-Holstein model, the results from the second order approximations describe in a rather satisfactory way the exact numerical results for the Kondo dominated regime. However, these approximations are not able to describe some features when increasing the electron-phonon coupling strength, such as the transition to the insulating state. This may require the implementation of approximations interpolating from the weak coupling to the polaronic regime. Finally, a possible extension of the method for the calculation of the steady state properties has been discussed, based on the combination of two self-consistent loops, one in time domain until reaching the steady state, and the other one in the stationary regime by changing one of the model parameters. Some preliminary results have been shown for the Anderson model, showing the accuracy of this extension.

Bibliography

- [1] D. Kambly, C. Flindt, and M. Büttiker. Factorial cumulants reveal interactions in counting statistics. *Phys. Rev. B* **83**, 075432 (2011).
- [2] P. Stegmann and J. König. Violation of detailed balance for charge-transfer statistics in Coulomb-blockade systems. *physica status solidi (b)* **254**, 1600507 (2017). 1600507.
- [3] F. B. Anders and A. Schiller. Real-time dynamics in quantum-impurity systems: a

BIBLIOGRAPHY

- time-dependent numerical renormalization-group approach. *Phys. Rev. Lett.* **95**, 196801 (2005).
- [4] F. Heidrich-Meisner, A. E. Feiguin, and E. Dagotto. Real-time simulations of nonequilibrium transport in the single-impurity Anderson model. *Phys. Rev. B* **79**, 235336 (2009).
- [5] J. Eckel, F. Heidrich-Meisner, S. G. Jakobs, M. Thorwart, M. Pletyukhov, and R. Egger. Comparative study of theoretical methods for non-equilibrium quantum transport. *New Journal of Physics* **12**, 043042 (2010).
- [6] E. Eidelstein, A. Schiller, F. Güttge, and F. B. Anders. Coherent control of correlated nanodevices: A hybrid time-dependent numerical renormalization-group approach to periodic switching. *Phys. Rev. B* **85**, 075118 (2012).
- [7] F. Güttge, F. B. Anders, U. Schollwöck, E. Eidelstein, and A. Schiller. Hybrid NRG-DMRG approach to real-time dynamics of quantum impurity systems. *Phys. Rev. B* **87**, 115115 (2013).
- [8] H. T. M. Nghiem and T. A. Costi. Generalization of the time-dependent numerical renormalization group method to finite temperatures and general pulses. *Phys. Rev. B* **89**, 075118 (2014).
- [9] L. Mühlbacher and E. Rabani. Real-time path integral approach to nonequilibrium many-body quantum systems. *Phys. Rev. Lett.* **100**, 176403 (2008).
- [10] K. F. Albrecht, H. Wang, L. Mühlbacher, M. Thoss, and A. Komnik. Bistability signatures in nonequilibrium charge transport through molecular quantum dots. *Phys. Rev. B* **86**, 081412 (2012).
- [11] G. Cohen, E. Gull, D. R. Reichman, and A. J. Millis. Green’s functions from real-time bold-line monte Carlo calculations: Spectral properties of the nonequilibrium Anderson impurity model. *Phys. Rev. Lett.* **112**, 146802 (2014).
- [12] J. Klatt, L. Mühlbacher, and A. Komnik. Kondo effect and the fate of bistability in molecular quantum dots with strong electron-phonon coupling. *Phys. Rev. B* **91**, 155306 (2015).
- [13] K. F. Albrecht, A. Martín-Rodero, J. Schachenmayer, and L. Mühlbacher. Local density of states on a vibrational quantum dot out of equilibrium. *Phys. Rev. B* **91**, 064305 (2015).

- [14] M. Ridley, V. N. Singh, E. Gull, and G. Cohen. Numerically exact full counting statistics of the nonequilibrium Anderson impurity model. *Phys. Rev. B* **97**, 115109 (2018).
- [15] X. Zheng, F. Wang, C. Y. Yam, Y. Mo, and G. Chen. Time-dependent density-functional theory for open systems. *Phys. Rev. B* **75**, 195127 (2007).
- [16] S. Kurth, G. Stefanucci, E. Khosravi, C. Verdozzi, and E. K. U. Gross. Dynamical Coulomb blockade and the derivative discontinuity of time-dependent density functional theory. *Phys. Rev. Lett.* **104**, 236801 (2010).
- [17] A.-M. Uimonen, E. Khosravi, A. Stan, G. Stefanucci, S. Kurth, R. van Leeuwen, and E. K. U. Gross. Comparative study of many-body perturbation theory and time-dependent density functional theory in the out-of-equilibrium Anderson model. *Phys. Rev. B* **84**, 115103 (2011).
- [18] E. Khosravi, A.-M. Uimonen, A. Stan, G. Stefanucci, S. Kurth, R. van Leeuwen, and E. K. U. Gross. Correlation effects in bistability at the nanoscale: Steady state and beyond. *Phys. Rev. B* **85**, 075103 (2012).
- [19] Y. Kwok, Y. Zhang, and G. Chen. Time-dependent density functional theory for quantum transport. *Frontiers of Physics* **9**, 698 (2014).
- [20] H. Wang and M. Thoss. Multilayer formulation of the multiconfiguration time-dependent Hartree theory. *The Journal of Chemical Physics* **119**, 1289 (2003).
- [21] S. Weiss, J. Eckel, M. Thorwart, and R. Egger. Iterative real-time path integral approach to nonequilibrium quantum transport. *Phys. Rev. B* **77**, 195316 (2008).
- [22] P. Nordlander, M. Pustilnik, Y. Meir, N. S. Wingreen, and D. C. Langreth. How long does it take for the Kondo effect to develop? *Phys. Rev. Lett.* **83**, 808 (1999).
- [23] M. Plihal, D. C. Langreth, and P. Nordlander. Transient currents and universal time scales for a fully time-dependent quantum dot in the Kondo regime. *Phys. Rev. B* **71**, 165321 (2005).
- [24] A. Komnik. Transient dynamics of the nonequilibrium Majorana resonant level model. *Phys. Rev. B* **79**, 245102 (2009).
- [25] P. Myöhänen, A. Stan, G. Stefanucci, and R. van Leeuwen. Kadanoff-Baym approach to quantum transport through interacting nanoscale systems: From the transient to the steady-state regime. *Phys. Rev. B* **80**, 115107 (2009).

BIBLIOGRAPHY

- [26] E. Perfetto and G. Stefanucci. Image charge effects in the nonequilibrium Anderson-Holstein model. *Phys. Rev. B* **88**, 245437 (2013).
- [27] S. Latini, E. Perfetto, A.-M. Uimonen, R. van Leeuwen, and G. Stefanucci. Charge dynamics in molecular junctions: Nonequilibrium Green's function approach made fast. *Phys. Rev. B* **89**, 075306 (2014).
- [28] R. Seoane Souto, R. Avriller, R. C. Monreal, A. Martín-Rodero, and A. Levy Yeyati. Transient dynamics and waiting time distribution of molecular junctions in the polaronic regime. *Phys. Rev. B* **92**, 125435 (2015).
- [29] H.-T. Chen, G. Cohen, A. J. Millis, and D. R. Reichman. Anderson-Holstein model in two flavors of the noncrossing approximation. *Phys. Rev. B* **93**, 174309 (2016).
- [30] P. D'Amico, D. A. Ryndyk, G. Cuniberti, and K. Richter. Charge-memory effect in a polaron model: equation-of-motion method for Green functions. *New Journal of Physics* **10**, 085002 (2008).
- [31] R.-P. Riwar and T. L. Schmidt. Transient dynamics of a molecular quantum dot with a vibrational degree of freedom. *Phys. Rev. B* **80**, 125109 (2009).
- [32] K. F. Albrecht, A. Martín-Rodero, R. C. Monreal, L. Mühlbacher, and A. Levy Yeyati. Long transient dynamics in the Anderson-Holstein model out of equilibrium. *Phys. Rev. B* **87**, 085127 (2013).
- [33] A. Kamenev. *Field Theory of Non-Equilibrium Systems* (Cambridge University Press, 2011).
- [34] P. W. Anderson. Localized magnetic states in metals. *Phys. Rev.* **124**, 41 (1961).
- [35] A.-P. Jauho, N. S. Wingreen, and Y. Meir. Time-dependent transport in interacting and noninteracting resonant-tunneling systems. *Phys. Rev. B* **50**, 5528 (1994).
- [36] A. Blandin, A. Nourtier, and D. W. Hone. Localized time-dependent perturbations in metals: formalism and simple examples. *J. Phys. France* **37**, 369 (1976).
- [37] P. B. Wiegmann. Towards an exact solution of the Anderson model. *Phys. Lett.* **80A**, 163 (1980).
- [38] N. Kawakami and A. Okiji. Exact expression of the ground-state energy for the symmetric Anderson model. *Physics Letters A* **86**, 483 (1981).

- [39] N. Andrei, K. Furuya, and J. H. Lowenstein. Solution of the Kondo problem. *Rev. Mod. Phys.* **55**, 331 (1983).
- [40] T. L. Schmidt, P. Werner, L. Mühlbacher, and A. Komnik. Transient dynamics of the Anderson impurity model out of equilibrium. *Phys. Rev. B* **78**, 235110 (2008).
- [41] J. E. Han and R. J. Heary. Imaginary-time formulation of steady-state nonequilibrium: Application to strongly correlated transport. *Phys. Rev. Lett.* **99**, 236808 (2007).
- [42] A. Martín-Rodero, F. Flores, M. Baldo, and R. Pucci. A new solution to the Anderson-Newns hamiltonian of chemisorption. *Solid State Communications* **44**, 911 (1982).
- [43] A. Martín-Rodero, E. Louis, F. Flores, and C. Tejedor. Interpolative solution for the periodic Anderson model of mixed-valence compounds. *Phys. Rev. B* **33**, 1814 (1986).
- [44] A. L. Yeyati, A. Martín-Rodero, and F. Flores. Electron correlation resonances in the transport through a single quantum level. *Phys. Rev. Lett.* **71**, 2991 (1993).
- [45] H. Kajueter and G. Kotliar. New iterative perturbation scheme for lattice models with arbitrary filling. *Phys. Rev. Lett.* **77**, 131 (1996).
- [46] F. B. Anders. A numerical renormalization group approach to non-equilibrium Green functions for quantum impurity models. *Journal of Physics: Condensed Matter* **20**, 195216 (2008).
- [47] J. A. White. Self-consistent Green functions for the Anderson impurity model. *Phys. Rev. B* **45**, 1100 (1992).
- [48] P. Werner, T. Oka, and A. J. Millis. Diagrammatic monte Carlo simulation of nonequilibrium systems. *Phys. Rev. B* **79**, 035320 (2009).
- [49] H. T. M. Nghiem and T. A. Costi. Time evolution of the Kondo resonance in response to a quench. *Phys. Rev. Lett.* **119**, 156601 (2017).
- [50] T. Fujii and K. Ueda. Perturbative approach to the nonequilibrium Kondo effect in a quantum dot. *Phys. Rev. B* **68**, 155310 (2003).
- [51] Y. Utsumi, O. Entin-Wohlman, A. Ueda, and A. Aharony. Full-counting statistics for molecular junctions: Fluctuation theorem and singularities. *Phys. Rev. B* **87**, 115407 (2013).

BIBLIOGRAPHY

- [52] Y. Murakami, P. Werner, N. Tsuji, and H. Aoki. Interaction quench in the Holstein model: Thermalization crossover from electron- to phonon-dominated relaxation. *Phys. Rev. B* **91**, 045128 (2015).
- [53] A. Gogolin and A. Komnik. Multistable transport regimes and conformational changes in molecular quantum dots. *arXiv:cond-mat/0207513* (2002).
- [54] A. S. Alexandrov, A. M. Bratkovsky, and R. S. Williams. Bistable tunneling current through a molecular quantum dot. *Phys. Rev. B* **67**, 075301 (2003).
- [55] M. Galperin, M. A. Ratner, and A. Nitzan. Hysteresis, switching, and negative differential resistance in molecular junctions: a polaron model. *Nano Letters* **5**, 125 (2005). PMID: 15792425.
- [56] G. Micchi, R. Avriller, and F. Pistolesi. Mechanical signatures of the current blockade instability in suspended carbon nanotubes. *Phys. Rev. Lett.* **115**, 206802 (2015).
- [57] G. Micchi, R. Avriller, and F. Pistolesi. Electromechanical transition in quantum dots. *Phys. Rev. B* **94**, 125417 (2016).
- [58] R. Avriller, B. Murr, and F. Pistolesi. Bistability and displacement fluctuations in a quantum nanomechanical oscillator. *Phys. Rev. B* **97**, 155414 (2018).
- [59] E. Y. Wilner, H. Wang, G. Cohen, M. Thoss, and E. Rabani. Bistability in a nonequilibrium quantum system with electron-phonon interactions. *Phys. Rev. B* **88**, 045137 (2013).
- [60] A. Martín-Rodero, A. Levy Yeyati, F. Flores, and R. C. Monreal. Interpolative approach for electron-electron and electron-phonon interactions: From the kondo to the polaronic regime. *Phys. Rev. B* **78**, 235112 (2008).
- [61] S. Maier, T. L. Schmidt, and A. Komnik. Charge transfer statistics of a molecular quantum dot with strong electron-phonon interaction. *Phys. Rev. B* **83**, 085401 (2011).
- [62] B. Dong, G. H. Ding, and X. L. Lei. Full counting statistics of a single-molecule quantum dot. *Phys. Rev. B* **88**, 075414 (2013).
- [63] R. Seoane Souto, A. L. Yeyati, A. Martín-Rodero, and R. C. Monreal. Dressed tunneling approximation for electronic transport through molecular transistors. *Phys. Rev. B* **89**, 085412 (2014).
- [64] D. F. Urban, R. Avriller, and A. Levy Yeyati. Nonlinear effects of phonon fluctuations on

- transport through nanoscale junctions. *Phys. Rev. B* **82**, 121414 (2010).
- [65] D. Meyer, A. C. Hewson, and R. Bulla. Gap formation and soft phonon mode in the Holstein model. *Phys. Rev. Lett.* **89**, 196401 (2002).
 - [66] B. K. Agarwalla, M. Kulkarni, S. Mukamel, and D. Segal. Tunable photonic cavity coupled to a voltage-biased double quantum dot system: diagrammatic nonequilibrium green's function approach. *Phys. Rev. B* **94**, 035434 (2016).
 - [67] M. A. Laakso, D. M. Kennes, S. G. Jakobs, and V. Meden. Functional renormalization group study of the Anderson–Holstein model. *New Journal of Physics* **16**, 023007 (2014).
 - [68] A. Jovchev and F. B. Anders. Influence of vibrational modes on quantum transport through a nanodevice. *Phys. Rev. B* **87**, 195112 (2013).
 - [69] G. S. Jeon, T.-H. Park, and H.-Y. Choi. Numerical renormalization-group study of the symmetric Anderson-Holstein model: Phonon and electron spectral functions. *Phys. Rev. B* **68**, 045106 (2003).
 - [70] A. C. Hewson and D. Meyer. Numerical renormalization group study of the Anderson-Holstein impurity model. *Journal of Physics: Condensed Matter* **14**, 427 (2002).

Part II.

Transient dynamics in superconducting nanojunctions

6 Quench dynamics in superconducting nanojunctions

6.1 Introduction

In the previous chapters the time dynamics of nanodevices has been analyzed in detail for several situations of interest. While chapter 3 was devoted to the non-interacting situation, in chapters 4 and 5 some paradigmatic interacting situations were analyzed, showing that the electron correlations can lead to a peculiar dynamics and approach to the steady state. This chapter is, however, devoted to the study of the superconducting nanojunctions, situation that has been much less analyzed in the literature [1–5].

In recent years, some proposals have suggested to use superconducting devices as a building blocks for quantum information purposes [6]. While the first works were based on the use of macroscopic Josephson junctions, nanoscaled systems like QPCs or QDs have recently been explored for quantum information applications, such as for the generation of electron entanglement through a Cooper pair splitting [7, 8] or the so-called Andreev qubits [9–11]. Moreover, the discovery of topological states of the matter has generated a huge activity in the field. It is particularly interesting the one dimensional topological superconductor case, which is supposed to host Majorana-like excitations at its ends [12] and can be used for (topologically) protected quantum computation (for recent reviews see [13, 14]).

However, in these devices an unexpectedly large quasiparticle population is found, which undermines the system coherence [15–22]. In superconducting nanodevices, these

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quasiparticles excited above the superconducting gap can eventually decay to the ABSs leading to a non-equilibrium population, what is called quasiparticle “poisoning”. Although it can be an obstacle for the implementation of the Majorana qubits [23–26], the long living excitations in the ABSs [27, 28] have been suggested as possible realizations of a spin qubit [29, 30].

While most of the studies have been focused on the stationary regime [31–36], in this chapter I present the analysis of the time dependent dynamics of mesoscopic superconductors. This is of essential interest for understanding the way the quasiparticles get trapped in the system and to develop new strategies for coherently controlling the system state. I shall analyze the quench dynamics of a superconducting atomic contact, focusing on the evolution of the system population, the current and the occupied spectral function. I pay special attention to the formation of the ABSs and also to their long time population after the connection. I consider the model describing a QD coupled to superconducting electrodes, which can also describe the situation of a superconducting QPC in the limit where the coupling to the electrodes is much larger than the superconducting gap.

Both phase and voltage driven situations are analyzed. In the first case the system gets trapped into a metastable state at long times for generic parameters, characterized by a lower supercurrent than the expected stationary value. It is also shown that an extra relaxation mechanism is necessary to relax the generated quasiparticles and to make the system reach its expected steady state. The role of the initial conditions is also analyzed, showing that the system dynamics depends strongly in the system preparation before the connection in the QD regime, while the dynamics progressively becomes independent from the initial conditions when moving to the QPC regime. This independence, however, does not imply the system relaxation to its stationary ground state and signatures of the quasiparticle trapping are found in the current evolution. Differently, when a bias voltage is applied to the junction, the system relaxes to the steady state due to the ABSs dynamics. This fact will be used to propose a new way of coherently control the system state by means of voltage pulses with different amplitudes.

Figure 6.1: Left panel: schematic representation of the nanojunction formed on a superconducting loop immersed in a magnetic field which allows to fix the phase difference ϕ between the two parts of the superconductor. Along this chapter two different ways of initializing the system are going to be considered. The first one described in a) consists on a quench of the tunneling amplitudes. The second one depicted in b) consists on a voltage drop at $t = 0$.

6.2 Model and formalism

In this section I present the model and the basic theoretical formalism to address the system dynamical properties. Along this chapter the Nambu representation is used to simplify the notation (see section 2.5 for further details). For simplicity, I will concentrate first on the quench of the tunneling amplitudes, analyzing at the end of the chapter the situation of a voltage step to control the system initialization. The Hamiltonian of the system is given in Eq. (2.78) where the tunneling part gets a dependence on time to describe the sudden connection between the system and the electrodes as

$$\bar{H}_T(t) = \theta(t) \sum_{k\nu} t_\nu \bar{\Psi}_{k\nu}^\dagger \bar{h}_t \bar{\Psi}_0. \quad (6.1)$$

In order to describe the superconducting electrodes, the Green function formalism from Refs. [33, 37] is used. In the equilibrium stationary regime, the system properties are well described by the retarded or advanced Green function, as discussed in section 2.5 of the second chapter. In a time-dependent non-equilibrium situation, however, the full Keldysh and Nambu structure has to be considered. Thus, the system Green function is given by

$$\hat{\hat{G}}_S = \left[\hat{\hat{g}}_S^{-1} - \hat{\hat{\Sigma}}_T \right]^{-1}. \quad (6.2)$$

where the $\hat{\hat{}}$ and the $\bar{}$ are used to denote the Keldysh and Nambu structure, respectively. Similarly to the previous chapters, the problem will be addressed by discretizing the

6. Quench dynamics in superconducting nanojunctions

Keldysh contour (see Fig. 3.1). Then, the discretized system Green function can be computed as

$$\hat{\mathbf{G}}_S = \left[\hat{\mathbf{g}}_S^{-1} - (\Delta t)^2 \hat{\mathbf{\Sigma}}_T \right]^{-1}, \quad (6.3)$$

which involves a matrix inversion. The inverse Green function of the uncoupled system can be written in Nambu space as

$$\hat{\mathbf{g}}_S^{-1} = \begin{pmatrix} \hat{\mathbf{g}}_{11}^{-1} & 0 \\ 0 & \hat{\mathbf{g}}_{22}^{-1} \end{pmatrix}, \quad (6.4)$$

where the subindex 1 and 2 are used to denote the spin up and spin down part, respectively. The component $\hat{\mathbf{g}}_{11}^{-1}$ is the inverse system Green function for the spin up electrons, described by Eq. (3.15). The spin down hole part is given by $\hat{\mathbf{g}}_{22}^{-1} = -(\hat{\mathbf{g}}_{11}^{-1})^*$, where the initial density matrix in this case is defined as $\rho_2 = [1 - n_\downarrow(0)]/n_\downarrow(0)$. Note that the occupation per spin can be calculated from Eq. (6.3), and given by $n_\uparrow(t) = -i\mathbf{G}_{11}^{+-}(t, t)$ and $n_\downarrow(t) = -i\mathbf{G}_{22}^{+-}(t, t)$.

On the other hand, the tunneling self-energies for each of the electrodes are given by

$$\hat{\mathbf{\Sigma}}_{T,\nu} = \begin{pmatrix} \hat{\Sigma}_{T,11} & \hat{\Sigma}_{T,12} \\ \hat{\Sigma}_{T,21} & \hat{\Sigma}_{T,22} \end{pmatrix}, \quad (6.5)$$

which contain spin mixing terms given by the off-diagonal terms ($\hat{\Sigma}_{21}$ and $\hat{\Sigma}_{12}$). In the time domain the tunneling self-energy components can be written as

$$\Sigma_{\nu,jk}^{\alpha\beta}(t, t') = \theta(t)\theta(t') s_\alpha s_\beta s_j s_k (V_\nu^0)^2 e^{i(s_j - s_k)\phi_\nu/2} g_{jk}^{\alpha\beta}(t - t'), \quad (6.6)$$

where $\alpha = \pm$ denotes the Keldysh branch, $j, k = 1, 2$ the Nambu component and $\nu = L, R$ the electrode. The global sign and the dependence on the superconducting phase is determined through $s_\pm = \pm 1$ and $s_j = (-1)^{(j+1)}$. The Fourier transform of the uncoupled lead Green functions can be analytically obtained for the finite bandwidth situation by Fourier transforming the BCS Green functions in frequency representation [37], finding

$$g_{11}^{+-}(t, t') = g_{22}^{+-}(t, t') = -\frac{i\Delta}{\pi D} H_1^{(1)}[\Delta(t - t')] + 2\frac{e^{iD(t-t')}}{D\pi^2(t - t')}, \quad (6.7)$$

while $g_{11}^{-+}(t, t') = g_{21}^{-+} = [g_{11}^{+-}]^*$, and for the anomalous components

$$g_{12}^{+-}(t, t') = g_{21}^{+-}(t, t') = -\frac{\Delta}{\pi D} H_0^{(1)}[\Delta(t - t')] - 2\frac{i\Delta}{\pi^2 D} E_1(-i\Delta|t - t'|), \quad (6.8)$$

while $g_{12}^{-+}(t, t') = g_{21}^{-+} = -[g_{12}^{+-}]^*$. In the above expressions $H_{0,1}^{(1)}$ denote the first kind Hankel functions and E_1 is the exponential integral with imaginary argument. The remaining components can be determined using the usual Keldysh relations

$$\begin{aligned}\Sigma_{T,jk}^{++}(t, t') &= -[\theta(t-t')\Sigma_{T,jk}^{-+}(t, t') + \theta(t'-t)\Sigma_{T,jk}^{+-}(t, t')] \\ \Sigma_{T,jk}^{--}(t, t') &= -[\theta(t-t')\Sigma_{T,jk}^{+-}(t, t') + \theta(t'-t)\Sigma_{T,jk}^{-+}(t, t')],\end{aligned}\quad (6.9)$$

with $j, k = 1, 2$. As mentioned in the previous chapters, there is an ambiguity on the definition of the Heaviside functions at $t = t'$. The most stable choice for the self-energies consist on taking

$$\Sigma_{T,jk}^{++}(t, t) = \Sigma_{T,jk}^{--}(t, t) = -\frac{\Sigma_{T,jk}^{-+}(t, t) + \Sigma_{T,jk}^{+-}(t, t)}{2}. \quad (6.10)$$

The discretized expression of the self-energies can be obtained through a direct evaluation of equations (6.7), (6.8), (6.9) and (6.10) in the discrete time mesh. For a good convergence in the calculation of the system Green function (6.3) it is necessary to take the inverse of the time step to be larger than the any energy parameter in the model. As I will consider in the following the bandwidth to be the largest energy scale, it may require $1/\Delta t \gtrsim D$.

6.2.1 Single pole approximation

Similarly to the non-interacting case for normal junctions discussed in the section 3.2, it becomes possible to obtain analytic expressions for the single particle observables evolution after the sudden connection between the electrodes and the system by means of the single pole approximation. In the absence of counting field, the Keldysh [38] rotation can be performed leading to the Dyson equation for the retarded and advanced components that can be written as

$$\bar{G}_S^{a,r}(t, t') = \bar{g}_S^{a,r}(t - t') + \int dt_1 \int dt_2 \bar{g}_S^{a,r}(t - t_1) \bar{\Sigma}_T^{a,r}(t_1 - t_2) \bar{G}_S^{a,r}(t_2, t'), \quad (6.11)$$

where in this case the Nambu structure is implicitly considered. As in the case of a nanodevice coupled to normal electrodes discussed in section 3.2, it can be shown that the kernel of the equation depends only on the time difference, preserving the time translation symmetry of the uncoupled Green functions. Thus, the time dependent retarded and advanced Green functions only depend on the time difference for $t, t' > 0$ and they can be written as

$$\bar{G}_S^{a,r}(t, t') = \theta(t)\theta(t')\bar{G}_{stat}^{a,r}(t - t'). \quad (6.12)$$

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Due to the time translation symmetry, the Laplace methods can be applied, finding the analytic solution to the integral equation (6.11)

$$\bar{G}_{stat}^{a,r}(s) = \begin{pmatrix} i s - \epsilon_0 - \Gamma \bar{G}_{11}^{a,r}(s) & (e^{i\phi_L} \Gamma_L + e^{i\phi_R} \Gamma_R) g_{12}^{a,r}(s) \\ (e^{-i\phi_L} \Gamma_L + e^{-i\phi_R} \Gamma_R) g_{12}^{a,r}(s) & i s + \epsilon_0 - \Gamma \bar{G}_{11}^{a,r}(s) \end{pmatrix}, \quad (6.13)$$

which is precisely the stationary Green function given in Eq. (2.87) with $\omega = i s$. This Green function has two kind of singularities. Firstly, it has two poles located at $\pm \epsilon_A$, with $\epsilon_A < \Delta$ which corresponds to the the position of the two ABSs. On the other hand, it has a branchcut which describes the continuum of states above and below the superconducting gap. In the limit $\Gamma/\Delta \ll 1$ the contribution from the continuum spectrum becomes negligible and the time dependent Green function is well approximated as

$$\bar{G}_S^{a,r} \approx \theta(t)\theta(t') \sum_{\pm} \begin{pmatrix} p_{\pm} & \pm p_{12} \\ \pm p_{12}^* & p_{\pm} \end{pmatrix} e^{\pm i \epsilon_A (t-t')}, \quad (6.14)$$

where $|p_{12}| = \sqrt{p_+ p_-}$. Simple expressions can be obtained in the regime $\epsilon_0 < \Gamma$, finding

$$\epsilon_A = \sqrt{\epsilon_0^2 + \Gamma^2 \cos^2(\phi/2) + (\Gamma_L + \Gamma_R)^2 \sin^2(\phi/2)}, \quad (6.15)$$

and

$$p_{\pm} = \frac{\Delta}{2\sqrt{\Delta^2 + \epsilon_A^2}} \left(1 - \frac{\Gamma}{\Delta} \mp \frac{\epsilon}{\epsilon_A} \right). \quad (6.16)$$

The off-diagonal Keldysh components of the system Green function can be obtained through the equation of motion

$$\bar{G}_S^{+-} = \bar{G}^r \bar{\Sigma}_T^{+-} \bar{G}^a + (1 + \bar{G}^r \bar{\Sigma}_T^r) \bar{g}_S^{+-} (1 + \bar{G}^a \bar{\Sigma}_T^a), \quad (6.17)$$

where integration over intermediate times is considered implicitly and the bare system Green function is given by

$$\bar{g}_S^{+-} = i e^{-i \epsilon_0 (t-t')} \begin{pmatrix} n_{\uparrow}(0) & 0 \\ 0 & 1 - n_{\downarrow}(0) \end{pmatrix}. \quad (6.18)$$

In the simplest situation of an initial occupation $(n_{\uparrow}(0), n_{\downarrow}(0)) = (0, 1)$, only the first term in Eq. (6.17) contributes and the time dependent spin up population can be written as

$$n_{\uparrow}(t) = \sum_{\pm} n_{\pm}(t) + n_{+-}(t). \quad (6.19)$$

Note that the spin down population for this initial configuration is simply given by $n_{\downarrow}(t) = 1 - n_{\uparrow}(t)$. The first term in the previous expression can be written as

$$n_{\pm}(t) = -\frac{2p_{\pm}}{\pi} \int_{-\infty}^{-\Delta} \frac{\omega \Gamma \mp 4p_+ p_- \Delta \epsilon_A}{\omega_{\mp} \sqrt{\omega^2 - \Delta^2}} [1 - \cos(\omega_{\mp} t)] d\omega, \quad (6.20)$$

where $\omega_{\pm} = \omega \pm \epsilon_A$. This term describes the population of each ABS, denoted by the $\pm$ subindex for the upper and lower state, respectively. It is well approximated for $t > 1/\Gamma$ as

$$n_{\pm}(t) = n_{\pm}(t \rightarrow \infty) + \frac{\Gamma}{\Delta} \left(1 \pm \frac{\epsilon_A}{\Gamma}\right) \frac{\sin[(\Delta \pm \epsilon_A)t - \pi/4]}{\sqrt{2\pi\dot{\Delta}t}}, \quad (6.21)$$

$n_{\pm}(t \rightarrow \infty) = (\Gamma/2 \pm \epsilon_A/\pi)/\Delta$ being the long time population of the two ABSs after the quench. The second term in Eq. (6.19) is the interference term given by

$$n_{+-} = \frac{2(p_+ - p_-)}{\pi} \int_{-\infty}^{-\Delta} \frac{4p_+p_- \Delta \epsilon_A}{\omega_+ \omega_- \sqrt{\omega^2 - \Delta^2}} [1 + \cos(2\epsilon_A t) - \cos(\omega_+ t) - \cos(\omega_- t)] d\omega, \quad (6.22)$$

which predicts a population oscillation due to the entanglement between the ABSs, generated by the sudden connection. At time $t > 1/\Delta$ the interference term (6.22) is well approximated by

$$n_{+-}(t) \approx -\frac{\epsilon_0}{\pi\Delta} \left(1 - \frac{\epsilon_0^2}{\epsilon_A^2}\right) \left[1 + \cos(2\epsilon_A t) + \sqrt{\frac{2\pi}{\Delta t}} \cos(\epsilon_A t) \sin\left(\Delta t - \frac{\pi}{4}\right)\right]. \quad (6.23)$$

Finally, when the system is prepared in a non-magnetic initial configuration, i.e. $(n_{\uparrow}(0), n_{\downarrow}(0)) = (0, 0)$ or $(1, 1)$, there is a contribution from the second term of Eq. (6.17). For instance, for the initially empty case the population can be written as

$$n_{\sigma}(t) = \sum_{\pm} n_{\pm}(t) + n_{+-}(t) + n_{00}(t), \quad (6.24)$$

with the extra contribution given by

$$n_{00}(t) = 2|p_{12}|^2 [1 - \cos(2\epsilon_A t)], \quad (6.25)$$

which describes undamped oscillations with a period $2\pi/\epsilon_A$ and an amplitude given by

$$|p_{12}|^2 = \frac{(1 - \Gamma/\Delta)^2 - \epsilon_0^2/\epsilon_A^2}{4(1 + \epsilon_A^2/\Delta^2)}. \quad (6.26)$$

6.3 Quench dynamics

In this section I present the main results for the time evolution of the single particle observables (system population, DOS and current) after a sudden connection to the electrodes and for a phase driven nanojunction.

Firstly, I analyze the system population in the regime $\Gamma \ll \Delta$, where the ABSs contribution dominates and the analytic expressions found in the previous section by means

Figure 6.2: Time evolution of the spin up component of the system population after the connection to the electrodes for an initial condition $(n_{\uparrow}(0), n_{\downarrow}(0)) = (0, 1)$. The red and blue curves of the mean panel show the population of the lower and upper ABS respectively, obtained from Eq. (6.21), while the green curve is the sum of both contributions. The parameters are $\Gamma = 0.05\Delta$, $\phi = 2$ and $\epsilon_0 = 0$. The dotted line corresponds to the analytic solution given by Eq. (6.19). In the inset longer times spin up population are represented the same situation as in the main panel (green curve) and for $\epsilon_0 = 0.025\Gamma$ (Yellow curve), which has been shifted down for clarity.

of the single pole approximation are valid. In Fig. 6.2 the time dependent spin up contribution to the system population is represented for an initial magnetic configuration $(n_{\uparrow}(0), n_{\downarrow}(0)) = (0, 1)$ and the electron-hole symmetric situation. Notice that the spin down contribution is simply given by $n_{\downarrow}(t) = 1 - n_{\uparrow}(t)$. As illustrated by the green curve of the main panel of Fig. 6.2, the population exhibits a fast linear increasing at short times, with a slope set by Γ , followed by an oscillatory behavior around the asymptotic value at longer times. The dotted line in Fig. 6.2 represents the analytic expression given in Eq. (6.19), in good agreement with the numerical results. In this electron-hole symmetric situation the contributions from each ABS are well separated due to a vanishing of the

Figure 6.3: Spin up component of the system population for increasing coupling to the electrodes, $\Gamma/\Delta = 0.05$ (top panel), 0.5 (middle panel) and 2 (lower panel). Results for the four possible initial configurations are shown: the red curve is used for the $(0,0)$ case, the blue one for the $(1,1)$ case and the solid and dotted green lines for the initially spin up and spin down polarized initial state, respectively. The black dashed lines in the top panel show the comparison to the results with the analytic population given in Eq. (6.24) for the initially empty case.

interference term of Eq.(6.22). The contributions for both of the states are represented in the main panel by the red curve for the lower ABS and the blue curve for the upper one. As it is shown, the population of the upper ABS is higher than the lower one due to the abrupt connection. An inversion of this tendency is observed and described by the analytic expression of Eq. (6.19) at $\Gamma/\Delta \sim 1$, where the population of the lower ABS becomes higher than the upper one.

Figure 6.4: Top panel: spin up population for the same parameters as in the middle panel of Fig. 6.3. The middle and the bottom panel show the time evolution occupied density of states for the $(0,0)$ and the $(0,1)$ cases respectively, and the same parameters as the top panel.

In the inset longer times are represented for the electron-hole symmetric case (green curve) and the $\epsilon_0 = 0.025\Gamma$ case (yellow curve), using the analytic expression given in Eq. (6.19). For the electron-hole symmetric situation, a beat of frequencies can be observed due to the superposition of the two different frequencies of the two states, whose periods are given by $2\pi/(\Delta \pm \epsilon_A)$. As described by Eq. (6.23), the oscillations decay with a rather weak power law (as $t^{-1/2}$). When the electron-hole symmetry is broken, the sudden connection between the system and the electrodes couples the two ABSs generating an additional oscillations with a period $2\pi/\epsilon_A$, which do not decay on time, as described by

Eq. (6.23). Remarkably, these oscillations are signatures of the entanglement between the ABSs generated by the initial conditions.

In Fig. 6.3 the spin up population time evolution is represented for three different couplings to the electrodes, $\Gamma/\Delta = 0.05$ (top panel), 0.5 (middle panel) and 2 (lower panel). The results correspond to the electron-hole symmetric situation and for the possible initial configurations. In the $\Gamma \lesssim \Delta$ regime, for initially trapped quasi-particles (i.e. configurations (0,0) and (1,1), represented in red and blue, respectively), the population exhibits large oscillations. These oscillations are well described in the $\Gamma/\Delta \ll 1$ regime by Eq. (6.25), represented as dashed lines in the top panel. In contrast, for the initially trapped spins (solid and dotted green curves), the system converges to a long time magnetic state. Remarkably, the long time population for this initial configuration is a bound for the oscillations observed for the initially trapped quasiparticle situation. This suggests that the relaxation of the initial conditions occurs at a very short time, becoming blocked afterwards. In the lower panels of Fig. 6.4 results for increasing values of Γ/Δ are shown, illustrating that the system population becomes gradually independent from the initial condition. In the QPC limit ($\Gamma \gg \Delta$) the expected stationary value $n_\sigma \sim 0.5$ for both spins is reached. However, as it will be shown in the following, this relaxation does not imply necessarily a relaxation to the steady state regime.

Further insight can be obtained from the occupied part of the DOS, represented in the middle and the bottom panels of Fig. 6.4 for the initial occupations (0,0) and (0,1), respectively. In the top panel the population evolution is shown for the different initial configurations. At the shortest times the population tends to relax to the stationary result given by $n_\sigma = 1/2$ for the electron-hole symmetric situation. However, a change in this tendency is observed at times of the order $t \sim 2/\Delta$ coinciding with the incipient formation of the ABSs inside the superconducting gap which blocks the charge relaxation. The blocking of the population relaxation produce the convergence of the system to a long time magnetic state for the (0,1) and (1,0) initial occupations. This behavior corresponds to a high probability of trapping quasiparticles in the ABSs, as illustrated in the bottom panel of Fig. 6.4, where the populations of the upper and the lower states are similar. On the other hand, for the initially empty or full level the system remains non-magnetic, although exhibiting undamped population oscillations with a period $\sim \pi/|\epsilon_A|$, as commented above. As shown in the middle panel of Fig. 6.4, the charge oscillations are due to coherent population and depopulation of both ABSs, simultaneously.

Figure 6.5: Left current evolution for the three different initial configurations: dashed red curve for (0,0) case, dotted blue line for the (1,1) situation and the solid green line represents the (0,1) case. Inset: symmetrized current. The parameters are the same as in Fig. 6.4

In the main panel of Fig. 6.5 the current at the left interface is represented for the same situations as in Fig. 6.4. Results for the initially empty (dashed red curve), initially fully occupied (dotted blue curve) and for the (0,1) initial configuration (solid green line) are represented. For the situations of initially trapped quasiparticle an oscillatory behavior is found for the left current with the same period than the one observed for the population evolution in Fig. 6.4. The (0,1) case exhibits oscillations with smaller amplitude and which decay as a weak power law ($t^{-1/2}$), as discussed above for the system population. In the inset of Fig. 6.5 the symmetrized current ($\langle I \rangle = (I_L - I_R)/2$) is represented, which is independent from the initial conditions. This demonstrates that the current and charge oscillations are due to a symmetric flow of quasiparticles between the system and the electrodes, which tend to populate the two ABSs simultaneously and are canceled out when the current is symmetrized. Thus, in the following I will concentrate on the symmetrized quantities.

In the top panel of Fig. 6.6 the symmetrized current is represented for the perfect trans-

Figure 6.6: Top panel: symmetrized current evolution for three different tunneling amplitudes $\Gamma/\Delta = 10$ (red), 5 (green) and 1 (blue), at the perfect transmission situation and at fixed phase difference $\phi = 2$. The full lines correspond to the case of no external relaxation mechanism while the dashed ones represent the case when a phenomenological broadening is added the spectral densities of size $1/\tau_{in} = 0.02\Delta$. The arrows indicate the stationary values for thermal equilibrium. The lower panels show the corresponding time-dependent occupied spectral densities without (bottom left panel) and including the external relaxation mechanism for the $\Gamma = 10$ case.

mitting situation and increasing tunneling rate values, from bottom to top. The solid line represents the quench result while the arrows on the right part of the panel represents the expected stationary value [37]. As shown, the long time current differs from the expected stationary result. Moreover, for $\Gamma/\Delta \lesssim 1$ the current changes sign with respect to the stationary result, a signature of the ABS population inversion mentioned above. This behavior is due to the quasiparticle poisoning (trapping of quasiparticles within the ABSs), which is a rather general behavior, independently from the system parameters.

Figure 6.7: Transient current as a function of the switching rate, $f(t) = \theta(t)[1 - \exp(-\alpha t)]$, with stationary tunneling amplitudes $\Gamma_L = \Gamma_R = 5$. Results for $\alpha = 5, 1$ and 0.25 are shown, from top to bottom. The dashed line represents the quench result. The remaining parameters are the same as in Fig. 6.4.

This is illustrated in the bottom left panel of Fig. 6.6, where the occupied DOS is represented in the QPC regime, where the population converges to the expected stationary result ($n_\sigma = 0.5$). As shown, the relaxation of the current happens at short times, until the ABSs are created at times $\sim 2/\epsilon_A$. After the ABSs appear in the superconducting gap, the system dynamics is frozen, leading to a lower asymptotic current.

In order to evacuate the quasiparticles trapped in the system two different, but equivalent, mechanisms can be used. The first one consists on adding a finite broadening to the branchpoints in the bare lead Green functions given in Eqs. (2.86). This broadening could be induced for example by the interactions, as discussed in Ref. [39]. The other way of making the quasiparticles relax is by weakly connecting the system to a normal electrode, enabling the quasiparticles to tunnel to the normal region. Similar mechanisms have been used to evacuate quasiparticles in transmon qubits [40]. For any of the two situations mentioned before, the current relaxes to the steady state with a rate of the

Figure 6.8: Long time population of the upper (red lines) and the lower (blue lines) ABS, computed by using the Eqs. (6.27) and (6.28). The solid line represents the quench result for different tunneling amplitudes while the dotted one represents the result for a smooth system connection with an effective tunneling rate given by $\Gamma_{eff} = \Gamma[1 - (1 - \exp(-\alpha t_A))]$ with $t_A = \sqrt{(2)}/\Delta$. The dashed lines represent the universal QPC behavior, described in the appendix C.

order of the broadening of the spectral function or the coupling to the normal region, respectively. This behavior is illustrated by the dashed lines in the top panel of Fig. 6.6, where a finite broadening of the gap edges of the order of $1/\tau_{in} = 0.02\Delta$ is considered. This value is extremely large compared to the experimental values. For instance in Ref. [27] relaxation times of the order of $1 - 100\mu s$ have been reported, corresponding to inelastic rates $1/\tau_{in} \sim 10^{-5} - 10^{-7}\Delta$. The occupied spectral function shown in the bottom right panel of Fig. 6.6 for the QPC regime illustrates the decay on time of the population of the upper ABS. In this case the charge is transferred to the lower ABS, which gets fully populated and significantly wider with respect to the case where no extra relaxation mechanism is included.

In order to show that the trapping of the system in a metastable state is not due to the abrupt connection to the electrodes, in Fig. 6.7 the current is represented for the

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same case as in Fig. 6.6 with $\Gamma = 10$ and for a smooth connection to the electrodes described by the function $f(t) = \theta(t)[1 - \exp(-\alpha t)]$, α being the connection rate. Results for increasing connection rates are shown from bottom to top. For a sufficiently fast connection rate the quench result is recovered, represented by the dashed line in the Fig. 6.7. For smoother connection rates the system exhibits a lower long time supercurrent, changing sign for the smaller connection rates, which is a signature of the system getting trapped in a more excited long time state.

This behavior is better understood by analyzing the ABS population. From the condition that both ABSs carry current in opposite directions, the long time current is given by

$$\langle I \rangle = (n_- - n_+) I_A + I_c, \quad (6.27)$$

where I_A is equilibrium supercurrent and I_c the contribution from the continuum spectrum [37]. In the electron-hole symmetric situation, the asymptotic population of the ABSs is bounded by

$$n_- + n_+ = 1. \quad (6.28)$$

Eqs. (6.27) and (6.28) constitute a system of linear coupled equations which is used to determine the population of the two ABSs, represented in Fig. 6.8 for increasing values of the tunneling amplitude. As shown, for $\Gamma \ll \Delta$ the upper ABS gets more populated than the lower one which leads to a supercurrent flowing in the opposite direction with respect to the expected stationary value. This behavior is inverted at $\Gamma \sim 1$ as predicted by the approximate expression given in Eq. (6.19). At large Γ values, the population tends to the universal QPC result represented by the discontinuous horizontal lines (see appendix C for more details).

Finally, the dotted lines in Fig. 6.8 represent the Andreev states population for the smooth connection case, with an effective tunneling rate, Γ_{eff} , given by the instantaneous tunneling rate at the characteristic formation of the ABS, i.e. $\Gamma_{eff} = \Gamma[1 - (1 - \exp(-\alpha t_A))]$ with $t_A \approx \sqrt{2}/\Delta$. A good agreement is found between the small Γ case and the smooth connection rate cases, finding also a population inversion at $\Gamma_{eff} \sim 1$. This indicates that for the smooth connection case the relaxation of the initial conditions happens at very short times, until the ABSs are formed. Afterwards they move adiabatically to their long time stationary value without exchanging charge.

Figure 6.9: Left panel: transient evolution of the current of a biased voltage nanojunction for increasing voltage values $V/\Delta = 4, 2, 1$ and 0.25 from bottom to top. In the right panel the stationary current value is shown. The parameters are $\epsilon = 0$ and $\Gamma_L = \Gamma_R = 5\Delta$.

6.4 AC-Josephson effect

In this section I summarize the main results for the transport properties of a voltage driven superconducting nanojunction. The voltage is supposed to be applied symmetrically, i.e. $\mu_L = -\mu_R = V/2$, which can be incorporated to the superconducting phase of the corresponding electrode through a gauge transformation, leading to $\phi_\nu(t) = \phi_\nu(0) + \mu_\nu t$ (with $\nu = L, R$).

The current evolution after the quench to the electrodes is represented in Fig. 6.9 for different bias voltages, for a perfect transmitting junction and in the QPC regime ($\Gamma \gg \Delta$). At short times a linear increase of the current is observed due to the abrupt connection to the electrodes. At times of the order $t \sim \pi/V$ the current converges to the expected stationary value. After converging, the current exhibits oscillations with a period π/V corresponding to the ac-Josephson effect. The amplitude of the oscillations decays with the increasing voltage, disappearing in the $V \gg \Delta$ limit. The long time

Figure 6.10: Long time averaged current in the QPC regime ($\Gamma = 60\Delta$) for different transmission factors ($\tau = 1, 0.9, 0.7, 0.5, 0.3$ and 0.1 , from top to bottom), compared to the dc stationary values (dashed lines) [33, 41]. The inset shows the convergence for the subgap voltages.

averaged stationary value is shown in the right panel of Fig. 6.9.

In Fig. 6.10 the long time averaged dc component of the current is represented for different transmission values. As shown, the numerical calculations (solid lines) are in very good agreement with the previous results obtained by the standard stationary methods given in Refs. [33, 41, 42] and shown as dotted lines in the figure. The agreement is somewhat poorer in the regime of small bias voltage ($V/\Delta \lesssim 1/10$) and the perfect transmission situations, due to an enlargement of the transient times making the calculation more demanding. The transient times in particular diverge in the equilibrium situation, where the system gets trapped in a metastable state as mentioned in the previous section. In the inset of Fig. 6.10 the dc currents for the subgap voltages are represented, which exhibit the features from the MAR processes (see section 2.5.1 for more details).

In Fig. 6.11 the dc current is represented for a voltage biased junction in the QD regime. The agreement between the numerical calculations and the stationary ones from

Figure 6.11: Long time averaged current in the QD regime ($\Gamma/\Delta = 1$) for different system energies, $\epsilon/\Delta = 0$ (red), 0.5 (green) and 1 (blue), which corresponds to transmissions values $\tau = 1, 0.8$ and 0.5 , respectively. The numerical calculations (solid lines) are compared to the stationary results (dashed lines) from Ref. [34]. Inset: zoom on the low bias limit where the curves are shifted up for clarity.

Refs. [34, 43, 44] is remarkable, as in the QPC regime. In the inset I show results for voltages smaller than the superconducting gap, exhibiting the expected subgap structure due to multiple Andreev reflections, even for the perfect transmission situation. It is worth mentioning that the transient dynamics in the QD is enlarged with respect to the QPC regime. For this reason it becomes more demanding to obtain accurate results for the stationary current, and small differences can be seen for voltages $V \lesssim \Delta/4$.

In Fig. 6.12 the occupied density of states is represented for the perfect transmission situation and increasing voltage values. In the top panel the occupied DOS is represented for a subgap voltage ($V = \Delta/4$), where the time evolution of the ABSs is adiabatic [44]. At very short times the population of the ABSs can differ from the equilibrium one, as mentioned in the previous section. However, when the states approach the gap edges the non-equilibrium population relaxes by transferring them to the continuum of states, and

Figure 6.12: Time evolution of the occupied spectral function for a bias voltage nanojunction in the QPC regime. Results for the perfect transmitting situation are shown, for a subgap voltage ($V = 0.25\Delta$) in the top panel and for a voltage bigger than the gap ($V = 2\Delta$) in the lower one. For the smaller voltage situation the expected adiabatic evolution of the ABSs is represented by the dashed red curves.

the system reaches the stationary condition. In the stationary regime the ABSs motion (represented by the dashed red line) leads to a charge pumping between the lower and the upper continuum of states [44]. This leads to a small density of quasiparticles excited over the superconducting gap, visible at the times where the upper state approaches the gap edges, which is able to relax in a time period. Moreover, the occupied ABS exhibits oscillations which can be due to coherent effects whose origin is the coupling to the probes used to measure the spectral density of the coupling between the two ABSs. Additionally to the main features related to the ABS, some replicas of the ABSs appear due to their non-adiabatic evolution.

In the regime $V > \Delta$, the ABS evolution becomes strongly non-adiabatic and it becomes progressively difficult to resolve them in the DOS. This case is represented in the

Figure 6.13: Time evolution of the occupied spectral density in the QPC regime and for different transmission factors $\tau = 0.96$ (top panel) and $\tau = 0.8$ (bottom panel). The dashed red curves is used to illustrate the adiabatic evolution of the ABSs. The remaining parameters are the same as in the top panel of Fig. 6.12.

bottom panel of Fig. 6.12 for $V = 2\Delta$. In this case the ABSs movement is completely non-adiabatic although some periodic features can be observed due to their movement. In this situation, the excited quasiparticles above the superconducting gap are unable to relax in a time period, leading to an almost homogeneous occupied DOS in the voltage window. However, a small change in the intensity is observed at the Fermi edge. At this point the two states approach each other, leading to the Landau-Zener phenomenon. The change on the spectral intensity is due to a non-perfect population transfer between the subgap states due to their fast movement.

Finally, in the non-perfect transmitting situation a gap between the two states opens, with increasing amplitude for decreasing transmission values as $\Delta_A = 2\Delta\sqrt{1-\tau}$. This situation has already been discussed in the stationary regime in Refs. [41, 44], finding that for subgap voltages the states evolve adiabatically, except when the states approach each

Figure 6.14: Main panel: Current evolution after a bias voltage switch off at $t = 0$ at a final phase difference $\phi = 2$. Results for three different pulse amplitudes are shown, $V/\Delta = 2, 16$ and 100 from top to bottom and for the perfect transmission situation. The black arrow denotes the expected stationary value. In the inset the current evolution during and just after the dc pulse is shown. The red dotted line represents the quench result.

other. At those times, the Landau-Zener transitions between the states have a transfer probability given by $P = \exp[-\pi\Delta(1 - \tau)/V]$. In Fig. 6.13 the non-perfect transmission situation is represented for a subgap voltage ($V = 0.25\Delta$). I show results for $\tau = 0.96$ (top panel), which corresponds to a transmission probability between states ~ 0.5 , and $\tau = 0.8$, where the probability of transition is negligible. While in the first case the upper ABS gets some population, in the lower one it remains always empty. In both cases quasiparticles initially trapped in the ABSs relax by transferring them to the continuum of states, similarly to the perfect transmission situation previously discussed.

6.5 Voltage pulse initialization

In this last section of the chapter I discuss the possibility of coherently control the ABSs population by means of voltage pulses. In the main panel of Fig. 6.14 the current evolu-

tion is shown after a switch off of the voltage at $t = 0$ for increasing V/Δ values from top to bottom. I focus on the QPC regime and consider a fixed final phase difference taken as $\phi = 2$. For $V \sim \Delta$ (blue curve), the long time system current evolves to a state close to the stationary result, represented by the arrow in Fig. 6.14. In this case, the voltage pulse is a way to relax the initially trapped quasiparticles to the continuum spectrum. This mechanism resembles the *antidote* protocols proposed in Ref. [27] to overcome quasiparticle poisoning, which consists on approaching the ABSs to the gap edges where the quasiparticles can be transferred to the continuum spectrum, from where they can move away from the junction. For increasing voltage values, the current progressively deviates from the stationary result, converging to the quench result (dotted line) for $V/\Delta \gg 1$, where the system losses completely the memory of the previous history. Remarkably, this is a way to address experimentally the quench dynamics of the system without manipulating the tunneling amplitudes. In the inset of Fig. 6.14 the current evolution is shown during and just after the voltage pulse.

Finally, in Fig. 6.15 the occupied DOS is represented after a voltage pulse of amplitude $V = 2\Delta$. As it is shown in the top, the lower ABS gets populated, while the upper one occupation is almost negligible, illustrating the convergence to the system ground state. It is worth mentioning that the final state of the system may depend strongly on the final phase. For instance, for a nearly perfect transmission situation and a final phase difference $\pi < \phi < 2\pi$ the upper ABS will become almost completely populated, as shown in the lower panel of Fig. 6.15. The situation with finite transmission coefficient is slightly more complex. For voltages much smaller than the Andreev gap, the lower ABS is always populated (see the lower panel of Fig. 6.13). Thus, after the voltage switch off the system will converge to the ground state independently from the final phase difference. In the case where the voltage is comparable to the gap between the ABSs (whose ac-Josephson spectral density is represented in the top panel of the Fig. 6.13) the system can either converge to the ground state for a phase difference in the range $0 < \phi < \pi$ or it can lead to a mixed state with trapped quasiparticles for $\pi \lesssim \phi < 2\pi$, where the population of both states depend on the Landau-Zener transition probability.

6.6 Conclusions

In this chapter I have presented an analysis of the transient dynamics of a nanoscale device coupled to two superconducting electrodes. Both phase and voltage driven situa-

Figure 6.15: Occupied spectral density after a voltage pulse with an amplitude $V/\Delta = 2$ and a final phase difference $\phi = 2$ (top panel) and -2 (bottom panel). The arrows show the asymptotic position of the two ABSs. The remaining parameters are the same as in Fig. 6.14.

tions have been analyzed. In the phase driven situation I have shown that, under general conditions, the system gets trapped in a metastable state characterized by a lower steady state supercurrent and the trapping of quasiparticles within the ABSs (quasiparticle poisoning). Moreover, in the QD regime the long time state becomes sensitive to its initial preparation and can lead to a long time magnetic state for an initially trapped spins in the system. For the initially empty or occupied case, however, the system population is always non-magnetic but exhibiting strong oscillations due to a symmetric flux of quasiparticles between the electrodes and the system. The dependence on the system initialization is lost for the symmetrized current. I have also shown that the system relaxation is blocked by the appearance of the ABSs in the gap, created at times $\sim 2/\epsilon_A$, whose population becomes frozen. To enforce the system to relax, an extra mechanism has to be considered such as a phenomenological broadening of the electrodes spectral density at the gaps edge or the coupling to a normal metal electrode.

In the voltage driven situation the system converges to the steady state, independently from the initial conditions, exhibiting the ac behavior expected for the current. The long time current has been also analyzed, in very good agreement with the conventional stationary methods for different transmissions. The relaxation of the system can be related to the ABS movement. For subgap voltages I have shown that the initially trapped quasiparticles can relax through the continuum spectrum when the ABSs approach the gap edges. In the last part of the chapter, I have analyzed the possibility of coherently control the ABS population by the use of voltage pulses. For voltages $V \sim \Delta$ the quasiparticles initially trapped in the system relax and the system remains in the final state just before the voltage drop. This means that the voltage switch off does not induce extra quasiparticles in the system for small voltages. This method can be used to initialize the system in the ground, the excited state or a mixture between them, depending on the final phase difference, the system transmission and the voltage. However, for large voltages ($V \gg \Delta$) the quench result is recovered providing a feasible experimental way to access the quench dynamics. Finally, it is worth mentioning that the method could be in principle generalized to treat more complex interactions [45], multiterminal problems [46–48] or topological superconductivity [49, 50].

Bibliography

- [1] E. Perfetto, G. Stefanucci, and M. Cini. Equilibrium and time-dependent Josephson current in one-dimensional superconducting junctions. *Phys. Rev. B* **80**, 205408 (2009).
- [2] G. Stefanucci, E. Perfetto, and M. Cini. Time-dependent quantum transport with superconducting leads: A discrete-basis Kohn-Sham formulation and propagation scheme. *Phys. Rev. B* **81**, 115446 (2010).
- [3] B. Tarasinski, D. Chevallier, J. A. Hutasoit, B. Baxevanis, and C. W. J. Beenakker. Quench dynamics of fermion-parity switches in a Josephson junction. *Phys. Rev. B* **92**, 144306 (2015).
- [4] J. Weston and X. Waintal. Linear-scaling source-sink algorithm for simulating time resolved quantum transport and superconductivity. *Phys. Rev. B* **93**, 134506 (2016).
- [5] R. Taranko and T. Domanski. How long does it take to form the Andreev quasiparticles? *arXiv:1705.08755* .
- [6] M. H. Devoret and R. J. Schoelkopf. Superconducting circuits for quantum information:

BIBLIOGRAPHY

- an outlook. *Science* **339**, 1169 (2013).
- [7] L. Hofstetter, S. Csonka, J. Nygård, and C. Schönenberger. Cooper pair splitter realized in a two-quantum-dot y-junction. *Nature* **461**, 960 EP (2009).
 - [8] L. G. Herrmann, F. Portier, P. Roche, A. L. Yeyati, T. Kontos, and C. Strunk. Carbon nanotubes as Cooper-pair beam splitters. *Phys. Rev. Lett.* **104**, 026801 (2010).
 - [9] A. Zazunov, V. S. Shumeiko, E. N. Bratus, J. Lantz, and G. Wendin. Andreev level qubit. *Phys. Rev. Lett.* **90**, 087003 (2003).
 - [10] N. M. Chtchelkatchev and Y. V. Nazarov. Andreev quantum dots for spin manipulation. *Phys. Rev. Lett.* **90**, 226806 (2003).
 - [11] C. Janvier, L. Tosi, L. Bretheau, Ç. Ö. Girit, M. Stern, P. Bertet, P. Joyez, D. Vion, D. Esteve, M. F. Goffman, H. Pothier, and C. Urbina. Coherent manipulation of Andreev states in superconducting atomic contacts. *Science* **349**, 1199 (2015).
 - [12] A. Y. Kitaev. Unpaired Majorana fermions in quantum wires. *Physics-Uspekhi* **44**, 131 (2001).
 - [13] V. Lahtinen and J. K. Pachos. A short introduction to topological quantum computation. *SciPost Phys.* **3**, 021 (2017).
 - [14] R. Aguado. Majorana quasiparticles in condensed matter. *Riv. Nuovo Cimento* **40**, 523 (2017).
 - [15] J. M. Martinis, M. Ansmann, and J. Aumentado. Energy decay in superconducting Josephson-junction qubits from nonequilibrium quasiparticle excitations. *Phys. Rev. Lett.* **103**, 097002 (2009).
 - [16] M. Lenander, H. Wang, R. C. Bialczak, E. Lucero, M. Mariani, M. Neeley, A. D. O'Connell, D. Sank, M. Weides, J. Wenner, T. Yamamoto, Y. Yin, J. Zhao, A. N. Cleland, and J. M. Martinis. Measurement of energy decay in superconducting qubits from nonequilibrium quasiparticles. *Phys. Rev. B* **84**, 024501 (2011).
 - [17] G. Catelani, R. J. Schoelkopf, M. H. Devoret, and L. I. Glazman. Relaxation and frequency shifts induced by quasiparticles in superconducting qubits. *Phys. Rev. B* **84**, 064517 (2011).
 - [18] D. Ristè, C. C. Bultink, M. J. Tiggelman, R. N. Schouten, K. W. Lehnert, and L. DiCarlo. Millisecond charge-parity fluctuations and induced decoherence in a

- superconducting transmon qubit. *Nature Communications* **4**, 1913 EP (2013). Article.
- [19] L. Bretheau, Ç. Ö. Girit, C. Urbina, D. Esteve, and H. Pothier. Supercurrent spectroscopy of Andreev states. *Phys. Rev. X* **3**, 041034 (2013).
 - [20] R. Avriller and F. Pistolesi. Andreev bound-state dynamics in quantum-dot Josephson junctions: A washing out of the $0-\pi$ transition. *Phys. Rev. Lett.* **114**, 037003 (2015).
 - [21] E. M. Levenson-Falk, F. Kos, R. Vijay, L. Glazman, and I. Siddiqi. Single-quasiparticle trapping in aluminum nanobridge Josephson junctions. *Phys. Rev. Lett.* **112**, 047002 (2014).
 - [22] A. Beshpalov, M. Houzet, J. S. Meyer, and Y. V. Nazarov. Theoretical model to explain excess of quasiparticles in superconductors. *Phys. Rev. Lett.* **117**, 117002 (2016).
 - [23] J. C. Budich, S. Walter, and B. Trauzettel. Failure of protection of Majorana based qubits against decoherence. *Phys. Rev. B* **85**, 121405 (2012).
 - [24] D. Rainis and D. Loss. Majorana qubit decoherence by quasiparticle poisoning. *Phys. Rev. B* **85**, 174533 (2012).
 - [25] J. R. Colbert and P. A. Lee. Proposal to measure the quasiparticle poisoning time of Majorana bound states. *Phys. Rev. B* **89**, 140505 (2014).
 - [26] S. M. Albrecht, E. B. Hansen, A. P. Higginbotham, F. Kuemmeth, T. S. Jespersen, J. Nygård, P. Krogstrup, J. Danon, K. Flensberg, and C. M. Marcus. Transport signatures of quasiparticle poisoning in a Majorana island. *Phys. Rev. Lett.* **118**, 137701 (2017).
 - [27] M. Zgirski, L. Bretheau, Q. Le Masne, H. Pothier, D. Esteve, and C. Urbina. Evidence for long-lived quasiparticles trapped in superconducting point contacts. *Phys. Rev. Lett.* **106**, 257003 (2011).
 - [28] D. G. Olivares, A. L. Yeyati, L. Bretheau, Ç. Ö. Girit, H. Pothier, and C. Urbina. Dynamics of quasiparticle trapping in Andreev levels. *Phys. Rev. B* **89**, 104504 (2014).
 - [29] C. Padurariu and Y. V. Nazarov. Spin blockade qubit in a superconducting junction. *EPL (Europhysics Letters)* **100**, 57006 (2012).
 - [30] S. Park and A. L. Yeyati. Andreev spin qubits in multichannel Rashba nanowires. *Phys. Rev. B* **96**, 125416 (2017).

BIBLIOGRAPHY

- [31] C. W. J. Beenakker and H. van Houten. Josephson current through a superconducting quantum point contact shorter than the coherence length. *Phys. Rev. Lett.* **66**, 3056 (1991).
- [32] M. Hurd, S. Datta, and P. F. Bagwell. Current-voltage relation for asymmetric ballistic superconducting junctions. *Phys. Rev. B* **54**, 6557 (1996).
- [33] J. C. Cuevas, A. Martín-Rodero, and A. L. Yeyati. Hamiltonian approach to the transport properties of superconducting quantum point contacts. *Phys. Rev. B* **54**, 7366 (1996).
- [34] A. L. Yeyati, J. C. Cuevas, A. López-Dávalos, and A. Martín-Rodero. Resonant tunneling through a small quantum dot coupled to superconducting leads. *Phys. Rev. B* **55**, R6137 (1997).
- [35] E. Scheer, P. Joyez, D. Esteve, C. Urbina, and M. H. Devoret. Conduction channel transmissions of atomic-size aluminum contacts. *Phys. Rev. Lett.* **78**, 3535 (1997).
- [36] E. Scheer, W. Belzig, Y. Naveh, M. H. Devoret, D. Esteve, and C. Urbina. Proximity effect and multiple Andreev reflections in gold atomic contacts. *Phys. Rev. Lett.* **86**, 284 (2001).
- [37] A. Martín-Rodero and A. L. Yeyati. Josephson and Andreev transport through quantum dots. *Advances in Physics* **60**, 899 (2011).
- [38] L. V. Keldysh. Diagram technique for nonequilibrium processes. *Zh. Eksp. Teor. Fiz.* **47**, 1515 (1964). [*Sov. Phys. JETP*20,1018(1965)].
- [39] R. C. Dynes, V. Narayanamurti, and J. P. Garno. Direct measurement of quasiparticle-lifetime broadening in a strong-coupled superconductor. *Phys. Rev. Lett.* **41**, 1509 (1978).
- [40] R.-P. Riwar, A. Hosseinkhani, L. D. Burkhardt, Y. Y. Gao, R. J. Schoelkopf, L. I. Glazman, and G. Catelani. Normal-metal quasiparticle traps for superconducting qubits. *Phys. Rev. B* **94**, 104516 (2016).
- [41] D. Averin and A. Bardas. ac Josephson effect in a single quantum channel. *Phys. Rev. Lett.* **75**, 1831 (1995).
- [42] E. N. Bratus, V. S. Shumeiko, and G. Wendin. Theory of subharmonic gap structure in superconducting mesoscopic tunnel contacts. *Phys. Rev. Lett.* **74**, 2110 (1995).

- [43] G. Johansson, E. N. Bratus, V. S. Shumeiko, and G. Wendin. Resonant multiple Andreev reflections in mesoscopic superconducting junctions. *Phys. Rev. B* **60**, 1382 (1999).
- [44] A. L. Yeyati, A. Martín-Rodero, and E. Vecino. Nonequilibrium dynamics of Andreev states in the Kondo regime. *Phys. Rev. Lett.* **91**, 266802 (2003).
- [45] R. Maurand, T. Meng, E. Bonet, S. Florens, L. Marty, and W. Wernsdorfer. First-order $0-\pi$ quantum phase transition in the Kondo regime of a superconducting carbon-nanotube quantum dot. *Phys. Rev. X* **2**, 011009 (2012).
- [46] A. Freyn, B. Douçot, D. Feinberg, and R. Mélin. Production of nonlocal quartets and phase-sensitive entanglement in a superconducting beam splitter. *Phys. Rev. Lett.* **106**, 257005 (2011).
- [47] T. Jonckheere, J. Rech, T. Martin, B. Douçot, D. Feinberg, and R. Mélin. Multipair dc Josephson resonances in a biased all-superconducting bijunction. *Phys. Rev. B* **87**, 214501 (2013).
- [48] A. H. Pfeffer, J. E. Duvauchelle, H. Courtois, R. Mélin, D. Feinberg, and F. Lefloch. Subgap structure in the conductance of a three-terminal Josephson junction. *Phys. Rev. B* **90**, 075401 (2014).
- [49] J. Alicea. New directions in the pursuit of Majorana fermions in solid state systems. *Reports on Progress in Physics* **75**, 076501 (2012).
- [50] C. W. J. Beenakker. Random-matrix theory of Majorana fermions and topological superconductors. *Rev. Mod. Phys.* **87**, 1037 (2015).

7 Counting statistics in superconducting nanojunctions

7.1 Introduction

In the previous chapter, the transient dynamics of superconducting nanojunctions after a sudden connection to the electrodes has been analyzed. As discussed, the system relaxation is blocked due to the subgap states formation, leading to a non-equilibrium population. The fact of quasiparticle trapping in the ABSs, also known as quasiparticle poisoning [1–4], has been shown to be a completely general feature and independent from the initial conditions or the system connection procedure. The relaxation to the steady state requires extra relaxation mechanisms and takes place in much longer time scales than the characteristic ABS formation time, as discussed in Ref. [1]. However, what was missing in the analysis of the previous chapter is a systematic characterization of the system state. As it will be shown in this chapter, the system state cannot be identified through the single particle properties only (current, population and spectral density), but a FCS analysis is needed [5]. The current statistics of superconducting nanojunctions have been discussed in the literature [6–11] in the stationary regime. In this chapter, however, the analysis is performed in the time-domain after the system connection, where as shown previously, the system gets trapped in a metastable state.

In the stationary regime, the subgap spectrum of a nanodevice involving a single transmission channel, like a superconducting atomic contact [7, 12], or a superconducting quantum dot [13], is characterized by the population of its ABSs. Four different possibilities can be found, which are depicted in the right panel of Fig. 7.1. In the ground

Figure 7.1: Left panel: Andreev bound states spectrum for a typical single channel nanojunction as a function of the superconducting phase difference ϕ . In the inset a scheme of a phase biased superconducting nanojunction is shown. Right panel: schematic representation of the four many body states which can be generated by changing the population of the ABSs (see text).

state ($|-\rangle$) only the lower ABS is occupied. There is a possible excitation with the same parity, consisting on exciting the electron from the lower to the upper ABS. This state is equivalent to add two quasiparticle with energy $|\epsilon_A|$ ($|\epsilon_A|$ being the energy separation between the ABSs and the Fermi level) and will be referred in the following simply as the excited state ($|+\rangle$). Finally, there are two more possible excitations, which are degenerate, consisting on completely populating or depopulating both ABSs simultaneously. These states will be referred to as odd states ($|odd_{1,2}\rangle$), since they have opposite parity with respect to the ground state. This picture is in principle only valid in the stationary regime. The state of the system has been measured in experiments [3, 14], driven by the proposals to develop the so-called Andreev qubits [15, 16]. The validity of this simplified image in the time-dependent regime is however not so clear. Moreover, it is unclear what is the role of the continuum of states, absent in this simplified picture.

While the subgap spectrum has been analyzed by coupling the system to a microwave resonator [3, 14], the possible determination of the system state through transport measurements only has remained unexplored. These measurements would constitute a less invasive method, since they do not require from external sources of energy which may drive the system out of equilibrium. In this chapter this possibility is analyzed by studying the FCS of the superconducting nanojunctions. Moreover, the question about the

minimal amount of information needed to fully characterize the system state is addressed in this chapter.

On the other hand, the roots of the system GF have been proposed as a tool to characterize the system state, since they contain a complete information about the charge transfer cumulants through Eq. (3.24). They also provide a way to characterize the possible non-equilibrium phases [17]. In connection to the theory of phase transitions in the equilibrium statistical mechanics (see section 1.5.1 for more details), phenomena like the intermittency [18, 19] or non-equilibrium phase transitions [20] have been analyzed. Recently, the Yang-Lee zeros have been experimentally measured in a superconducting device in the incoherent regime [21]. By means of the high order cumulants of the transitions between many body states, the authors of Ref. [21] have determined the position of the dominant zeros (the ones with larger real part). In this chapter I will address the complementary question about the Yang-Lee zeros of the charge statistics in the coherent regime at shorter times than the typical Markovian time [1]. Moreover, by using a simplified description of the GF at times much longer than the characteristic ABS formation, the asymptotic distribution of the zeros is analyzed.

Recently, it has been shown that the factorial cumulants contain valuable information about the system properties. These quantities are cumulants measured at finite counting field values, which is equivalent to add an external field which drives the system dynamics to uncommon trajectories (see section 3.4 for more details). These quantities have been demonstrated to be useful for characterizing interactions [22–25] or non-equilibrium phases [26–28]. However, it is still unclear how to determine them directly without addressing the full charge probability distribution, which may require from challenging measurement. In particular, in superconducting devices the quantum transport involve a bidirectional transference of charges due to Andreev reflections.

Finally, in the last part of the chapter I briefly discuss the situation of a superconducting nanojunction coupled to a bosonic mode. This model can be used to understand the problem of the coupling to a localized vibrational mode (for example a molecule) or to a photonic cavity mode. This last situation is of interest in connection to recent experiments [3, 14]. I show that, even in the situation where the frequency of the mode is much smaller than the superconducting gap and weakly coupled to the electrons, by driving it out of equilibrium a non-thermal population of the ABSs is observed, in agreement with the experimental observations.

7.2 Formalism

The time-dependent transport properties of the system are fully characterized by the GF defined on the Keldysh contour as [29, 30]

$$Z(\chi, t) = \left\langle T_K \exp \left[-i \int_C dt' \bar{H}_{T,\chi}(t') \right] \right\rangle_0, \quad (7.1)$$

where the expression for the tunneling Hamiltonian is given in Eq. (6.1). Note that the counting field, χ , is introduced in the Hamiltonian, appearing as a phase factor which modulates the tunneling amplitudes as $\bar{t}_{\nu,\chi} = \bar{t}_{\nu,\chi=0} \bar{\sigma}_z e^{i\bar{\sigma}_z \chi_\nu}$. For simplicity, the symmetrized cumulants will be analyzed along this chapter, unless stated differently. As in the case of normal junctions discussed in chapter 3, it can be shown that the GF can be written as a Fredholm determinant

$$Z(\chi, t) = \frac{\det \left[\hat{g}_S^{-1} - \hat{\Sigma}_{T,\chi} \right]}{\det \left[\hat{g}_S^{-1} - \hat{\Sigma}_{T,\chi=0} \right]}, \quad (7.2)$$

where the expression for the uncoupled system Green function is given in Eq. (6.4). The counting field-dependent self-energy is given by

$$\left[\hat{\Sigma}_{T\nu,\chi}(t, t') \right]_{jk}^{\alpha\beta} = e^{i(s_\alpha s_j - s_\beta s_k) \chi_\nu / 2} \left[\hat{\Sigma}_{T\nu}(t, t') \right]_{jk}^{\alpha\beta}, \quad (7.3)$$

where $\hat{\Sigma}_{T\nu}$ is given in Eq. (6.6). The discretized expression for the tunneling self-energy can be straightforwardly obtained by evaluating Eq. (7.3). Then, the transport properties can be calculated from the GF given by Eq. (7.2), which can be decomposed as $Z(\chi, t) = \sum_n P_n(t) \exp(i n \chi)$, $P_n(t)$ being the probabilities of detecting n electrons transferred between the connection time and t . In the superconducting case these probabilities can take eventually negative values, and for this reason in the following they will be denoted as quasi-probabilities in the following. Two possible explanations have been given in the literature to explain this behavior. The first one is related to the fact that the number of electrons in the leads (and thus the initial system state) is not well defined for a system with a well defined phase difference (note they are conjugated variables) [8, 31]. The second one, given in Ref. [32], relates it to interference processes between different trajectories in the Keldysh contour. For the situation analyzed along this chapter, both explanations seem to be equally valid, and there is in principle no way to discern the reason why the $P_n(t)$ can get small negative values. However, as it will be commented below, positive asymptotic probabilities can be defined by averaging P_n over a certain n interval.

7.2.1 Coarse grained statistics

In this subsection I introduce a simplified description of the GF to understand the numerical results, valid for $t \gg 1/\Delta$. As shown in the previous chapter, for sufficiently long times the subgap states are formed, getting a non-equilibrium population. Moreover, as described in Refs. [1, 3], the system relaxation and transition occur at much longer time scales. For times much shorter than those scales, where transitions between states are unlikely, the GF is described by three time-independent probabilities

$$Z(\chi, t) \approx P_- e^{i\chi_-(t)} + P_+ e^{i\chi_+(t)} + P_{odd} e^{i\chi_{odd}(t)}, \quad (7.4)$$

where $P_{\pm, odd}$ are the probabilities of populating the ground, the excited and the odd states, respectively, and $\chi_{\pm, odd} = \chi I_{\pm, odd} t$ describe the position of the three probability maxima. This position is determined by the current the state carry, which is given by

$$I_{\pm} = I_c \pm |I_A|, \quad I_{odd} = I_c, \quad (7.5)$$

where $|I_A|$ is the supercurrent carried by the ABSs and I_c the contribution of the continuum spectrum, which vanishes both for $\Gamma \gg \Delta$ and $\Gamma \ll \Delta$, becoming maximal when $\Gamma \sim \Delta$. Note that both odd states carry the same current and cannot be resolved by means of electron transport measurement. To distinguish between them another kind of measurements has to be performed. From Eq. (7.4) the approximate expressions for the first two current cumulants at long times are simple and given by

$$\begin{aligned} \langle I \rangle &\simeq P_- I_- + P_+ I_+ + P_{odd} I_{odd}, \\ \langle I^2 \rangle &\simeq t \left[(P_- I_-^2 + P_+ I_+^2 + P_{odd} I_{odd}^2) - (P_- I_- + P_+ I_+ + P_{odd} I_{odd})^2 \right]. \end{aligned} \quad (7.6)$$

These two expressions, together with the normalization condition ($P_+ + P_- + P_{odd} = 1$) constitute a complete set of linear equations that can be solved, leading to

$$P_{\pm} = \frac{\langle I^2 \rangle / t + 2(\langle I \rangle^2 - I_c^2)}{4I_A^2} - \frac{\langle I \rangle - I_c}{2I_A} \left(\frac{2I_c}{I_A} \pm 1 \right), \quad (7.7)$$

and $P_{odd} = 1 - (P_- + P_+)$. Thus, the system state can be fully identify from the current and noise measurements, in the simple example of a single channel superconducting nanojunction. In general, if the nanojunction exhibits more conduction channels, higher order cumulants are required to be measured to address the system state.

On the other hand, from Eq. (7.4), the position of the zeros of the GF is given by

$$z_{\pm}^{I_A t} \approx \frac{-P_{odd}^2 \pm \sqrt{P_{odd}^2 - 4P_- P_+}}{2P_-}, \quad (7.8)$$

which for $t \gg 1/\Delta$ corresponds to two branches, converging to the unitary circle centered in the coordinates origin. An analogous behavior has been found in Ref. [33] for the two dimensional Ising model in the equilibrium situation. The current cumulants can be computed through the exact expression given in Eq. (3.24), finding

$$\langle I^n \rangle \approx n I_A^n t^{n-1} \sum_{\pm} \text{Li}_{1-n} \left(\frac{1}{z_{\pm}} \right), \quad (7.9)$$

which describe the way the cumulants diverge with time due to the coexistence of the three many-body states, which carry different current. Finally, the factorial cumulants can be determined similarly as discussed in section 3.4 by including a biasing field.

7.3 Quench dynamics

In this section the main results for the time evolution of a phase biased superconducting nanojunctions after a sudden connection to the electrodes is presented. The upper panel of Fig. 7.2 shows the short time evolution of the quasiprobabilities, evolving from an uni-modal distribution to a tri-modal one. The three probability maxima are related to the three states (ground, excited and odd states) described in the previous section. In the lower panel of Fig. 7.2 cuts before (blue) and just after this transition (red) are shown. At very short times, the probability distribution exhibits only a single maximum. The behavior at these times is well described by a bidirectional Poisson distribution (see appendix D), represented by the discontinuous line in the lower panel. The green line shows the probability distribution at the typical ABSs formation time, which becomes asymmetric exhibiting a net charge transfer through the system. It deviates from the bidirectional Poisson distribution (dotted line). Remarkably, the fit provides an estimation of the number of electrons crossing the junction in each direction to create the ABSs of the order of ~ 3 . At longer times (red curve), the distribution exhibits three maxima, indicating the coexistence between the three different many body states.

The three maxima become more clearly visible at longer times, as shown in the main panel of Fig. 7.3. The character of each maximum can be inferred from their motion in time, which is related to the state current as $n_{\pm, \text{odd}} = I_{\pm, \text{odd}} t$, where the stationary state current is given by Eq. (7.5). The slope of the probability maximum moving towards a positive number of electrons transferred is given by $|I_A| + I_c$ (represented as the dashed line), thus related to the ground state. Similarly, the other two probability maxima evolve with slopes given by I_c (middle maximum) and $|I_A| + I_c$ (bottom maximum),

Figure 7.2: Top panel: time evolution of the quasiprobabilities. Lower panel: quasiprobabilities at three different times $t = 1$ (blue), 3 (green) and 6 (red). The dashed and dotted black lines are fits using the bidirectional Poisson distribution (also known as birth-death distribution).

which can be related to the odd and excited states, respectively. Thus, a FCS analysis provide information about the system state [1, 34], which is not accessible from the single particle observables analyzed in the previous chapter. In the right panel of Fig. 7.2, the quasiprobabilities at $t \gg 1/\Delta$ are represented, where the many-body probabilities ($P_{\pm}, P_{odd}$) can be extracted by integrating around the three maxima of the probability distribution.

In Fig. 7.4 the asymptotic many-body probabilities are shown, as a function of the stationary ABS energy, $|\epsilon_A|$. Results for the QPC (left panel) and for the QD regime (right panel) are represented. In the QPC regime, the odd state exhibits an increasing value for

Figure 7.3: Main panel: $P_n(t)$ represented in the (t, n) plane for a perfectly transmitted junction. The dashed lines represent the corresponding current for each of the states given in Eq. (7.5). In the right panel the quasiprobabilities are represented at $t = 50/\Delta$. The remaining parameters are $\Gamma/\Delta = 10$ and $\phi = 2$.

decreasing ϵ_A , reaching $P_{\text{odd}} = 0.5$ for $\epsilon_A \rightarrow 0$, while $P_- = P_+ = 0.25$. In this case, the result is independent from the initial conditions (as discussed in the previous chapter for the current and the system population) or the system transmission (results for $\tau = 0.95$ and 0.9 are shown as squares and circles, respectively). Thus, the QPC results are universal in the sense that they only depend on the value of ϵ_A . The dotted line represents the solution from the approximate rate equation, described in appendix C and schematically shown in the inset of Fig. 7.4, where the rates are inversely proportional to the energy distance to the lower gaps edge, i.e. $\Gamma_{\pm} \propto 1/(\Delta \pm \epsilon_A)$ and $\Gamma_{\text{odd}} \propto 1/\Delta$. This simplified description provides results in good agreement with the numerical ones for small ϵ_A values. It also provides the qualitative behavior when the ABSs approach the gaps edges, although overestimating the ground state weight, while P_{odd} is underestimated. On the other hand, in the QD regime the result becomes dependent on the system initial configuration, as described by the solid lines for the $(0, 0)$ initialization and the dashed ones for the $(0, 1)$. In this regime, the system keeps memory of the initial configuration. In particular, when reducing Γ the final state tends to preserve the electron parity number in the system from the initial one, converging predominantly to the even states for $(0, 1)$

Figure 7.4: Asymptotic probabilities, representing P_- in red, P_+ in blue and P_{odd} in green color. Results for the QPC regime ($\Gamma/\Delta = 10$) are shown in the left panel for $\tau = 1$ (solid line), 0.95 (squares) and 0.9 (dots). In the right one results in the QD regime are shown for the initially empty situation (dashed lines) and for an initially trapped spin (solid lines) are shown.

and $(1, 0)$ initializations, while the odd states dominate for the fully empty or occupied ones. Moreover, this behavior is rather independent from the asymptotic ABSs position, as shown in the right panel of Fig. 7.4.

Finally, in Fig. 7.5 the symmetrized shot noise is represented for different tunneling rates and initial configurations. As can be observed, the symmetrized shot noise becomes sensitive to the initial configurations in the QD regime, where different long time states are reached as a function of the system initialization. This result is at variance with the symmetrized current (see for instance the inset of Fig. 6.5 of the previous chapter), indicating that current fluctuations contain information about the system state, not included in the mean current. This also illustrates the fact that the quantum state of the system cannot be completely inferred from the single particle observables, but it requires the knowledge of the current fluctuations. In all the situations described in Fig. 7.5 the shot noise grows linearly in time, indicating the coexistence between the three many-

Figure 7.5: Symmetrized shot noise of the transferred charges for different initial conditions $(n_{\uparrow}(0), n_{\downarrow}(0)) = (0, 0)$ or $(1, 1)$ (continuous lines) and $(1, 0)$ (dashed lines) for increasing values of the tunneling amplitudes, $\Gamma/\Delta = 0.05$ (red), 0.5 (blue) and 2 (green).

body states after the system connection. The long time behavior is determined by the lower equality in Eq. (7.6). Moreover, the noise is larger for the case of initially trapped quasi-particles. It becomes gradually independent from the initial configurations when Γ is increased, until reaching the QPC regime, where the long time evolution of the system does not depend on its initialization.

7.3.1 Yang-Lee zeros and phase coexistence

Complementary information can be obtained from the position of the roots of the GF (DYLZs) which, according to Eq. (3.24) contain a complete information about the system transport properties. In Fig. 7.6 I represent $1/|Z(z, t)|$ for increasing time values, where the bright spots correspond to the position of the DYLZs. At the shortest time (top panel), all the roots of the GF appear in the real negative axis, signature of uncorrelated tunneling events [22, 35–37]. For times $\sim 1/\Delta$ (middle panel of Fig. 7.6) superconducting correlations start to be important in the system, and the zeros appear in the complex

Figure 7.6: Short time behavior of $1/|Z(z, t)|$ as a function of $z = e^{i\chi}$ complex variable. Results for times $\Delta \cdot t = 0.5, 1$ and 7 (panels from top to bottom) are shown. The red squares and yellow diamonds are used to mark the dominant DYLZs evolution. The parameter values are $\Gamma/\Delta = 2$, $\epsilon = 0$ and $\phi = 2$

z -plane as complex conjugate pairs marked with the red squares. For times $t \gg 1/\Delta$ (lower panel of Fig. 7.6), the DYLZs marked with the symbols approach the measurement point $z = 1$. These roots will be referred to as *dominant zeros*, since they provide the main contribution to the charge transfer, according to Eq. (3.24). In particular, all the cumulants diverge for a zero located at $z = 1$.

The long time distribution of DYLZs is represented in Fig. 7.7, showing their tendency to accumulate in the complex z -plane forming two branches. The position of the dominant zeros is well described by Eq. (7.8). At long times it predicts that the roots arrange in the complex plane forming two concentric circles centered at the coordinates origin (red lines in Fig. 7.7). Although the description of the roots farther from the measurement point is poorer due to details absent in the coarse grained description of subsection 7.2.1 (such as the peak's width), the agreement improves with increasing

Figure 7.7: Long time ($t \sim 25/\Delta$) behavior of $1/|Z|$ illustrating the accumulation of DYLZs around two circles which can be described by the coarse grained expression of Eq. (7.8), represented as red lines. Results for $\Gamma = 2$ (top panel) and 0.5 (bottom panel) are shown. The remaining parameters are the same as in Fig. 7.6.

times. In analogy to the equilibrium statistical mechanics (see section 1.5.1 for more details), the two circles can be understood as phase coexistence lines [33, 38], which separate three different phases. The nature of each of the phases can be inferred from their transport properties. In the limit $t \rightarrow \infty$, the two circles tend to converge to the unitary one ($|z| = 1$), leading to the coexistence of three phases at the measurement point, $z = 1$, which thus becomes a *triple point*. This image is consistent with the one provided before by the quasi-probabilities in Fig. 7.2. It is worth mentioning that if an additional relaxation mechanism is included in the model, as the ones discussed in the previous chapter, the asymptotic radius of the circles is reduced moving the coexistence point to smaller $|z|$ values, becoming the phase coexistence not visible at the measurement point.

In Fig. 7.8 the shot noise is shown for the same parameters as in Fig. 7.6, where the dotted line represents the numerical result. The green line corresponds to the result

Figure 7.8: Shot noise evolution for the same parameters as in Fig. 7.6. The dotted line is the numerical result, while the green, blue and black curves correspond to including an increasing number of DYLZs in the exact expression of Eq. (3.24). While for the green curve only the two dominant DYLZs (denoted by the red squares in Fig. 7.6) are used in the calculation, for the blue one also the subdominant ones (yellow diamonds in Fig. 7.6) are also included. Finally, the black curve represents the convergence to the numerical result when including a higher number of singularities (around 10 in this case).

given by Eq. (3.24) where only two dominant zeros have been considered (represented as red squares in Fig. 7.6). As shown, by including only information about the dominant zeros, the long time behavior of the shot noise is not well described. For obtaining the way the shot noise grows in time, also the subdominant zeros (yellow diamonds in Fig. 7.6) have to be included, leading to an accurate description of the slope (blue line in the figure). This result is at variance with the case analyzed in Ref. [33], where only two phases coexist and thus only the information about the two dominant zeros were needed. In this case, however, two branches of DYLZs have to be described since $z = 1$ becomes a triple point in the long time limit. For obtaining a quantitative agreement with the numerical result, a higher number of zeros have to be included, as shown by the black curve where ~ 10 DYLZs have been considered.

Figure 7.9: Factorial current (top) and noise (bottom panel) as a function of the bias field s . Results for three different times, $t = 10$ (blue), 25 (green) and 50 (red) are shown. The arrows in the bottom panel denote the noise maxima, determined through the simplified description given by the coarse grained FCS of the section 7.2.1. The parameters are the same as in Fig. 7.6.

Finally, in Fig. 7.9 the first two factorial cumulants (factorial current and noise) are shown, which corresponds to displace the measurement point on the positive real z -axis a quantity s from $z = 1$. Results for increasing times and the same parameters as in the lower panel of Fig. 7.7 are represented. The factorial current tends to exhibit a jump at the measurement point ($s = 0$), indicated by the dashed vertical line. Moreover, the current goes from negative values for $s < 0$ (inside the two circles depicted in Fig. 7.7) to positive ones for $s > 0$ (outside the circles). This indicates that inside the circles the even excited state is dominant, at variance with the points outside the circles where the ground state dominates. For $s \simeq 0$ (between the circles of Fig. 7.7) the current almost vanishes, indicating a dominance of the odd state. On the other hand, the factorial noise tends to develop two peaks approaching the $s = 0$ point for increasing times. The arrows in the figure are used to denote the maxima position using the coarse grained descrip-

Figure 7.10: FCS of a voltage biased junction. In the top panel I represent the time evolution of the probability after the contact formation. In the lower panel the evolution of the shot noise is shown, illustrating the convergence to the steady state. The parameters are $\Gamma/\Delta = 10$, $\tau = 1$ and $V/\Delta = 0.25$

tion, which corresponds to the intersection of the circles with the real z axis in the lower panel of Fig. 7.7. The increasing of the noise value at $s = 0$ with time is again a signature of phase coexistence. I would like to emphasize that, in the analogy with statistical mechanics, the $z = 1$ point would be a first order phase transition point where the first derivative of the GF (which plays the role of the partition function in the analogy) tends to exhibit a step and the second derivative tends to diverge.

7.4 Finite bias voltage dynamics

In this section the main results for the transport properties through a voltage biased junction are presented. While the single particle properties were presented in section 6.4, in this section the FCS results are analyzed. The upper panel of Fig. 7.10 shows results

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for the time evolution of the quasi-probabilities for $V < \Delta$ and in the QPC regime. At very short times after the junction formation, the probability distribution exhibits three maxima, related to the phase coexistence between the three many body states described above. At times longer than the expected quasiparticle relaxation times ($t > \pi/V$), the slope of the three peaks become equal, reflecting the convergence to the stationary regime characterized by the presence of a single quantum phase. Although the three maxima are visible in the probability distribution for large times, they are no longer reflecting a coexistence between three phases since their time evolution is similar (thus carrying the same current). For $V > \Delta$ the initially trapped quasiparticles are able to relax at short times, avoiding the short time phase coexistence and exhibiting a single quasiprobability maximum.

In the lower panel of Fig. 7.10 the shot noise evolution is shown for the same bias voltage as in the upper panel. A linear increase of the noise is observed at short times, related to the phase coexistence. At longer times, when the trapped quasiparticles in the ABS relax, the noise relaxes to the stationary situation, characterized by an oscillatory behavior consistent to the ac-Josephson effect. The maximum of the shot noise correspond to the subgap states approaching each other, where they can exchange population (see for instance the upper panel of Fig. 6.12). While the minimum is found when the ABSs reach the gap's edge.

The long time averaged shot noise behavior in the QPC regime is shown in Fig. 7.11, comparing the numerical results (solid lines) with the expected stationary value (dashed lines) from Refs. [6, 9, 10], for different transmission coefficients. In the upper panel the high transmission situations are presented, while the low transmission ones are shown in the lower panel, where the structure due to the MAR becomes more evident. The inset of the upper panel shows the shot noise in an enlarged scale for the subgap voltages, exhibiting a remarkable agreement with the stationary results, except for extremely small voltages ($V/\Delta \lesssim 1/10$) where the relaxation time becomes longer and the computation more demanding. Additionally, the finite Γ value induces a small deviation at the lower voltages, mainly visible for the perfect transmitting situation. As already discussed in Refs. [6, 9, 10], the Fano factor $\langle I^2 \rangle / \langle I \rangle$ diverges in the $V \rightarrow 0$ limit, reflecting the increase in the effective transmitted charge due to the multiple Andreev reflection processes of increasing order.

In Fig. 7.12 results for the long time averaged shot noise are represented for the QD

Figure 7.11: Long time averaged noise for different transmissions values ($\tau = 1, 0.99, 0.98, 0.96$ and 0.9 , from top to bottom in the upper panel and $0.7, 0.5, 0.3$ and 0.1 in the bottom one) in the QPC regime ($\Gamma = 60\Delta$), compared to the dc stationary values (dashed lines) [6, 9, 10]. Inset of the top panel: zoom on the low bias limit.

regime (I consider $\Gamma/\Delta = 1$) and different level positions. Note that, at variance with the QPC regime, signatures of the MAR can be observed even in the perfectly transmitted situation (represented by the red curve). The inset shows the results for subgap voltages in an enlarged scale. To the best of my knowledge, these results have not been reported before in the literature and can be relevant to describe some recent experiments [39]. Moreover, it is worth remarking that the time-resolved technique can be used to obtain steady state results in a relatively simple way, compared to the standard stationary methods.

Figure 7.12: Long time averaged noise in the QD regime ($\Gamma = \Delta$) for different positions, $\epsilon_0/\Delta = 0$ (red curve), 0.5 (green curve) and 1 (blue curve), corresponding to transmissions $\tau = 1$, 0.8 and 0.5 , respectively. Inset: zoom on the low biased junction where the curves are shifted up by ϵ_0/Δ for clarity.

7.4.1 Dynamical Yang-Lee zeros

In this subsection the main results for the DYLZs of a voltage biased nanojunction are presented. In Fig. 7.13 I represent $1/|Z(z, t)|$ in the complex z -plane for increasing times, from top to bottom and for $V/\Delta > 1$. In the top panel, the result at $t = 1/\Delta$ is represented, when the superconducting correlations start to become important in the system. As in the phase biased case (represented in the middle panel of Fig. 7.6 for the same parameters but with $V = 0$), the dominant zeros, marked with the red squares and yellow diamonds, appear as conjugate pairs in the complex plane. However, an extra zero appear in the negative real axis, related to the quasiparticle relaxation (black circle). At intermediate times (middle panel of Fig. 7.13), when this point becomes dominant, the rest of the DYLZs approaches the negative real axis. At longer times (bottom panel), all the GF roots converge to the negative real axis, exhibiting a small splitting close to $z = 0$

Figure 7.13: DYLZs in the voltage biased situation for a perfect transmitting junction for increasing time values, $\Delta \cdot t = 1, 7$ and 12 , from top to bottom. The red squares, yellow diamonds and black circle are used to denote the dominant, subdominant and the quasiparticle relaxation zeros, respectively. The inset in the lower panel zooms over the negative real axis. The parameters are the same as in Fig. 7.10 with $V/\Delta = 2$.

which is reduced when increasing Γ . The green dots in Fig. 7.13 represent the steady state DYLZs, obtained from the result described in Refs. [9, 10], in qualitative agreement with the numerical calculations. The inset of the lower panel shows the behavior close to the z -real axis in an enlarged scale.

In Fig. 7.14 the $1/|Z(\chi, t)|$ function is represented in the complex z -plane for a subgap voltage and for increasing times from top to bottom. At short times (upper and middle panels) the DYLZs tend to converge to the unitary circle, signature of phase coexistence. However, the dominant zeros do not approach the $z = 1$ with increasing time (see the bottom panel of Fig. 7.14), meaning that the density of zeros close to this point at $t \gg 1/\Delta$ is zero and there is no coexistence between phases. Thus, the cumulants converge to the steady state result (as commented in connection to Figs. 7.11 and 7.12), although

Figure 7.14: DYLZs for the $V/\Delta = 0.25$ situation. The remaining parameters are the same as in Fig. 7.13.

the charge statistics keeps some memory about the initial phase coexistence (see for instance the top panel of Fig. 7.10). Similarly to the case of voltages bigger than the superconducting gap, the DYLZs converge to their steady state, represented by the green lines in the lower panel of Fig. 7.14 and computed as above from the steady state results from Refs. [9, 10]. Note that the number of stationary branches is related to the number of different multiple Andreev processes contributing to the charge transport through the system at the corresponding bias, roughly given by $2\Delta/V$.

Figure 7.15: Long time populations of the many body states after the voltage switch off for increasing voltage values, $V/\Delta = 2, 16$ and 100 from left to right. The red, blue and green curves represent the probability of finding the system in the ground (P_-), excited (P_+) and odd states (P_{odd}) after the voltage pulse, respectively. The parameters are $\Gamma/\Delta = 10$, $\tau = 1$ and the final phase $\phi = 2$.

7.5 Voltage pulse initialization

In this section I analyze the system state after a voltage step. In section 6.5 of the previous chapter, the single particle properties have been analyzed after a voltage drop, as a function of the value of the voltage. It was shown that the current can converge either to the expected stationary value for small voltages or to the quench result for $V/\Delta \gg 1$. In Fig. 7.15 the asymptotic probabilities are represented in the QPC regime, for increasing initial voltage values from left to right. For $V/\Delta \sim 1$, the probability for the system to converge to the ground state after the voltage drop is very high (~ 0.95), while there is a small probability for the system to become trapped in the odd state. This is related to the processes where the quasiparticles excited above the superconducting gap decay to the upper ABSs. Remarkably, the probability of the excited state vanishes. For increasing bias voltage, the many-body population converges gradually to the quench result, reached for $V/\Delta \gg 1$ (right panel of Fig. 7.15). Remarkably, this provides a feasible way of testing the quench dynamics of a superconducting device without manipulating the

Figure 7.16: DYLZs after a voltage pulse of amplitude $V = 2\Delta$. Results for times $t/\Delta = 1, 6$ and 14 are shown from top to bottom. The remaining parameters are the same as in Fig. 7.15).

tunneling amplitudes.

Finally, in Fig. 7.16 The DYLZs are presented for increasing times after a voltage pulse of amplitude $V/\Delta \sim 1$ (corresponding to the left panel of Fig. 7.15). Just after the voltage drop (top panel) the zeros of the system are mostly localize in the real negative axis, as described in the lower panel and the inset of Fig. 7.13. For increasing times (middle and bottom panels of Fig. 7.16) they tend to converge to a single unit circle, signature of the phase coexistence between two states. This image is compatible with the two phases coexistence, as described in the left panel of Fig. 7.15. As commented above, the character of each of the phases can be inferred from the factorial cumulants (not shown for this case), finding that outside the circle ($s > 0$) the ground state is dominant,

while inside ($s < 0$) the odd state dominates.

7.6 Coupling to a bosonic mode

In the final part of this chapter the situation of a superconducting nanojunction coupled to a bosonic mode is studied. It can be used to analyze rather different experimental situations such as the superconducting junction coupled to a localized phonon mode [40–42] or to a microwave resonator. This last case is of special interest since recent experiments have demonstrate that microwave radiation can be used to measure and control the system state [3, 14].

7.6.1 Model and formalism

In this section I introduce the minimal model to describe the superconducting system coupled to a bosonic mode. Two equivalent situations under the DTA approximation can be studied. The first one consists on a mode coupled to the local charge in the system, whose Hamiltonian is given by

$$\bar{H} = \bar{H}_S + \bar{H}_{leads}^{BCS} + \bar{H}_T(t) + \bar{H}_{e-p} + H_p \quad (7.10)$$

where the non-interacting part of the Hamiltonian is described by Eqs. (2.80) and (2.79) in Nambu space. As in the previously studied situations, I also consider the tunneling term to be connected at the initial time, whose Hamiltonian is given in Eq. (6.1). The phonon part of the Hamiltonian is given by $H_p = \omega_0 a^\dagger a$, ω_0 being the frequency mode. The interaction part describing the coupling to the bosonic reservoir is given by

$$\bar{H}_{e-p} = \sum_{\sigma} \lambda (a + a^\dagger) \bar{\Psi}^\dagger \bar{\sigma}_0 \bar{\Psi}, \quad (7.11)$$

where λ is the electron-phonon coupling strength. By performing the Lang-Firsov transformation (see section 2.4 for a more detailed discussion) the Hamiltonian can be rewritten as

$$\tilde{\bar{H}} = \tilde{\bar{H}}_0 + \bar{H}_{leads} + \tilde{\bar{H}}_T, \quad (7.12)$$

where $\tilde{\bar{H}}_0 = \bar{\Psi}_0^\dagger \bar{h}_{vib} \bar{\Psi}_0$ with $\bar{h}_{vib} = (\epsilon_0 - \lambda^2/\omega_0) \bar{\sigma}_z$. The many body interactions are included in the tunneling Hamiltonian part as

$$\tilde{\bar{H}}_T = \theta(t) \sum_{k\nu} t_\nu X \bar{\Psi}_{k\nu}^\dagger \bar{h}_t \bar{\Psi}_0. \quad (7.13)$$

7. Counting statistics in superconducting nanojunctions

where X is the polaron cloud operator in the Nambu representation. As described in chapter 4, the GF of the DTA has a non-interacting form. In the superconducting situation is given by

$$Z(\chi, t) = \frac{\det \left[\hat{g}_0^{-1} - \hat{\Sigma}_{DTA, \chi} \right]}{\det \left[\hat{g}_0^{-1} - \hat{\Sigma}_{DTA, \chi=0} \right]}, \quad (7.14)$$

with the dressed self-energy given by

$$[\Sigma_{DTA, \chi}(t, t')]_{jk}^{\alpha\beta} = \left[\hat{\Sigma}_{T\nu, \chi}(t, t') \right]_{jk}^{\alpha\beta} \left[\hat{\Lambda}(t, t') \right]_{jk}^{\alpha\beta}. \quad (7.15)$$

The bare tunneling self energies are described in Eq. (7.3) and $\hat{\Lambda}$ is the polaron propagator. In the Nambu representation the polaron propagator is given by $\hat{\Lambda}(t, t') = \langle \{ \bar{\Phi}(t) \bar{\Phi}(t') \} \rangle$ with the spinor described by $\bar{\Phi} = (X, X^\dagger)^T$. In the Nambu Keldysh space the functions are given by [42]

$$\begin{aligned} \Lambda_{11}^{\mp, \pm}(t - t') &= \Lambda_{22}^{\mp, \pm}(t - t') = \exp \left\{ -g^2 \left[n_p (1 - e^{\mp i \omega_0 (t - t')}) + (n_p + 1) (1 - e^{\pm i \omega_0 (t - t')}) \right] \right\}, \\ \Lambda_{12}^{\mp, \pm}(t - t') &= \Lambda_{21}^{\mp, \pm}(t - t') = \exp \left\{ -g^2 \left[n_p (1 + e^{\mp i \omega_0 (t - t')}) + (n_p + 1) (1 + e^{\pm i \omega_0 (t - t')}) \right] \right\}, \end{aligned} \quad (7.16)$$

where n_p is the boson number, considered to be constant for a driven system and left as a free parameter. The Keldysh diagonal components can be determined through the usual Keldysh relations given in Eqs. (3.19) and (3.20). Although the DTA does not describe the adsorption or emission of real bosons, it contains valuable information about the inelastic processes. It can be checked that, under the DTA, similar expressions are obtained for the GF and the tunneling self-energy if the bosonic mode is coupled to the current operator, whose Hamiltonian is given by

$$\bar{H} = \bar{H}_S + \bar{H}_{leads}^{BCS} + \tilde{H}_T(t), \quad (7.17)$$

where the modified tunneling term is

$$\tilde{H}_T(t) = \theta(t) \sum_{\sigma} \left[t_{Lk} c_{Lk, \sigma}^\dagger e^{i\lambda/\omega_0(a+a^\dagger)} + t_{Rk} c_{Rk, \sigma}^\dagger e^{-i\lambda/\omega_0(a+a^\dagger)} + \text{H.C.} \right]. \quad (7.18)$$

Expanding the previous equation with respect to the coupling to the phonon mode, it is found

$$\tilde{H}_T(t) \approx H_T(t) + \theta(t) \frac{\lambda}{\omega_0} \left[\sum_{\sigma} i I_k (a + a^\dagger) + \text{H.C.} \right] + \dots, \quad (7.19)$$

being $I_k = t_{Lk} c_{Lk, \sigma}^\dagger + t_{Rk} c_{Rk, \sigma}^\dagger$. From the previous expression it becomes more evident that the Hamiltonian of Eq. (7.17) describes the coupling between the boson mode and the system current.

Figure 7.17: Stationary occupied spectral density around $\phi = \pi$. The parameters are $\Gamma/\Delta = 10$, $\omega_0/\Delta = 0.1$, $\lambda/\omega_0 = 0.05$ and $n_p = 10$. The upper and the lower panels represents $\tau = 1$ and 0.99 , respectively.

7.6.2 Single particle properties

I begin first by analyzing the single particle properties of the system. In Fig. 7.17 the stationary occupied spectral density in the QPC regime is represented for the perfect transmission situation (top panel) and for $\tau = 0.99$ (bottom panel). I show results for the subgap spectrum, around the $\phi = \pi$ point. Although the coupling to the phonon mode is rather small ($\lambda^2/\omega_0\Gamma \sim 10^{-3}$), the upper state gets a finite population due to the coupling to the driven mode. This population increases with increasing coupling parameter λ . Similarly, it is also increased when raising n_p , becoming almost zero for $n_p = 0$ and the same parameters of Fig. 7.17. This is indicative that the driven mode induces a coupling between the two ABSs, leading to a ground state characterized by a finite population in both subgap states. It is worth mentioning that the DTA does not describe the level anti-crossing around $\epsilon_A = \omega_0$ [43, 44] due its approximate character. This is, however, described by other approximations like the PTA [45], although not shown in this section.

Figure 7.18: Current evolution after a voltage pulse initialization with amplitude $V/\Delta = 2$ and final phase difference $\phi = 2$. Situations for $n_p = 0, 20, 50$ and 100 are shown, from top to bottom. The arrows denote the expected current stationary value. The remaining parameters are the same as in Fig. 7.17.

In Fig. 7.18 the current evolution is shown after a voltage pulse of amplitude $V/\Delta \sim 1$, for increasing n_p from top to bottom. The arrows denote the expected stationary value in the equilibrium situation, whose subgap contribution has already been analyzed in Ref. [42]. As shown, the current after the pulse evolves to its stationary value at long time for all possible values of n_p . This is indicative that the voltage pulse initialization is an efficient way of relaxing the initially trapped quasiparticles generated during the system connection, similarly to the non-interacting situation discussed in the previous chapter. However, the value reached is slightly smaller, due to some residual quasiparticles trapped in the system, as it will be shown in the next section.

Figure 7.19: Asymptotic probability to find the system in the ground (red line), excited (blue line) and odd states (green curve), for increasing values of the coupling to the boson mode. The parameters are the same as in Fig. 7.17 with $n_p = 0$ and $\phi = 3$.

7.6.3 Counting statistics

In this section I present the FCS analysis for a boson mode coupled to a superconducting system by means of the DTA approximation. In Fig. 7.19 the asymptotic many-body probabilities are represented for increasing electron-phonon coupling for $n_p = 0$. As shown, for small couplings, the system converges to the ground state with a high probability (~ 0.9) as in the non-interacting case (see the left panel of Fig. 7.15). However, for increasing interaction strengths the population of the odd and even excited states are also increased. The asymptotic value is reached for $\lambda/\omega_0 \simeq 0.7$ where $P_{odd} \sim 0.5$ and $P_{\pm} \sim 0.25$. It is worth remarking that the results shown correspond to the perfect transmission situation, although a similar qualitative behavior and the same asymptotic value is obtained when decreasing the transmission coefficient.

Finally, in Fig. 7.20 I represent the asymptotic many-body probabilities after the voltage drop for increasing phonon number. I show results in the weakly coupled regime,

Figure 7.20: The same probabilities as in Fig. 7.19 are represented for increasing number of n_p and for $\lambda/\omega_0 = 0.05$. The remaining parameters are the same as in Fig. 7.17.

where the system relaxes to the steady state for $n_p = 0$. This regime is relevant since it is the one studied in the experiments [3]. As shown in the figure, by increasing n_p , the population of the odd state is increased monotonously, reaching its asymptotic value $P_{odd} \sim 0.5$ for $n_p \sim 50$. Moreover, even in the regime where the phonon frequency is larger than the energy distance between the ABSs, but much smaller than the superconducting gap ($\epsilon_A \ll \omega_0 \ll \Delta$), the mode can induce quasiparticles trapped in the subgap states.

7.7 Conclusions

In this chapter I have presented a FCS analysis of superconducting nanojunctions. Both the phase biased and voltage biased situations have been analyzed. The first part of the chapter has been devoted to the study of the phase biased situation, showing that the system converges to a metastable state after the formation of the junction. This state is characterized by the many-body probabilities, which are only accessible through the charge or current fluctuations. Moreover, I have demonstrated that the system state in

this case can be obtained through the long time behavior of the first two current cumulants (current and shot noise). Both, the QPC and the QD regimes have been analyzed. In the QPC regime, the asymptotic populations of the many-body states exhibit an universal behavior, dependent only on the stationary ABS energy. In the QD regime, however, the asymptotic populations preserves memory of the symmetry of the initial condition. The zeros of the GF have been also analyzed for the phase biased situation, showing that they can be used to determine the phase transition points in the complex counting field plane. This can be related to the phase transition theory in the equilibrium statistical mechanics described in Refs. [33, 38]. Moreover, I have shown that the factorial cumulants become of importance to characterize each of the phases.

For the voltage biased junction I have shown that the system converges to the expected steady state due to quasiparticle relaxation. I have shown that the time-dependent algorithm becomes useful to analyze steady state properties which are rather difficult to address using conventional techniques. This is illustrated by calculating the stationary shot noise of a superconducting QD, which has not been addressed before in the literature. In this case, I have also shown that the zeros of the GF converge to their expected steady state result, where the number of branches is determined by the possible number of MAR processes. Also the state after a voltage pulse has been analyzed, showing that depending on the amplitude of the pulse the system can converge to the ground state ($V \lesssim \Delta$) or to the quench result ($V \gg \Delta$).

Finally, the last part of the chapter has been devoted to the problem of a boson mode coupled to a superconducting nanodevice. This analysis can be relevant to understand recent experiments [3, 14] where a high probability of system trapping in an odd state has been measured. By using the DTA, a non-equilibrium population of the ABSs have been observed for increasing number of bosons. Moreover, by performing a FCS analysis the many-body probabilities can be obtained, in a similar way as for the non-interacting superconducting nanojunctions. I have found that the population of the odd state increases with increasing electron-boson coupling or the number of bosons in the junction. Moreover, for large values of the boson population, the probability to find the odd state after a quantum measurement is ~ 0.5 , in agreement with recent experiments [3, 14].

Bibliography

- [1] M. Zgirski, L. Bretheau, Q. Le Masne, H. Pothier, D. Esteve, and C. Urbina. Evidence for long-lived quasiparticles trapped in superconducting point contacts. *Phys. Rev. Lett.* **106**, 257003 (2011).
- [2] L. Bretheau, Ç. Ö. Girit, C. Urbina, D. Esteve, and H. Pothier. Supercurrent spectroscopy of Andreev states. *Phys. Rev. X* **3**, 041034 (2013).
- [3] C. Janvier, L. Tosi, L. Bretheau, Ç. Ö. Girit, M. Stern, P. Bertet, P. Joyez, D. Vion, D. Esteve, M. F. Goffman, H. Pothier, and C. Urbina. Coherent manipulation of Andreev states in superconducting atomic contacts. *Science* **349**, 1199 (2015).
- [4] S. M. Albrecht, E. B. Hansen, A. P. Higginbotham, F. Kuemmeth, T. S. Jespersen, J. Nygård, P. Krogstrup, J. Danon, K. Flensberg, and C. M. Marcus. Transport signatures of quasiparticle poisoning in a Majorana island. *Phys. Rev. Lett.* **118**, 137701 (2017).
- [5] R. S. Souto, A. Martín-Rodero, and A. L. Yeyati. Andreev bound states formation and quasiparticle trapping in quench dynamics revealed by time-dependent counting statistics. *Phys. Rev. Lett.* **117**, 267701 (2016).
- [6] J. C. Cuevas, A. Martín-Rodero, and A. L. Yeyati. Shot noise and coherent multiple charge transfer in superconducting quantum point contacts. *Phys. Rev. Lett.* **82**, 4086 (1999).
- [7] R. Cron, M. F. Goffman, D. Esteve, and C. Urbina. Multiple-charge-quanta shot noise in superconducting atomic contacts. *Phys. Rev. Lett.* **86**, 4104 (2001).
- [8] W. Belzig and Y. V. Nazarov. Full counting statistics of electron transfer between superconductors. *Phys. Rev. Lett.* **87**, 197006 (2001).
- [9] J. C. Cuevas and W. Belzig. Full counting statistics of multiple Andreev reflections. *Phys. Rev. Lett.* **91**, 187001 (2003).
- [10] J. C. Cuevas and W. Belzig. dc transport in superconducting point contacts: a full-counting-statistics view. *Phys. Rev. B* **70**, 214512 (2004).
- [11] S. Pilgram and P. Samuelsson. Noise and full counting statistics of incoherent multiple Andreev reflection. *Phys. Rev. Lett.* **94**, 086806 (2005).

- [12] E. Scheer, P. Joyez, D. Esteve, C. Urbina, and M. H. Devoret. Conduction channel transmissions of atomic-size aluminum contacts. *Phys. Rev. Lett.* **78**, 3535 (1997).
- [13] A. Martín-Rodero and A. L. Yeyati. Josephson and Andreev transport through quantum dots. *Advances in Physics* **60**, 899 (2011).
- [14] M. Hays, G. de Lange, K. Serniak, D. J. van Woerkom, D. Bouman, P. Krogstrup, J. Nygård, A. Geresdi, and M. H. Devoret. Direct microwave measurement of Andreev-bound-state dynamics in a proximitized semiconducting nanowire. *arXiv:1711.01645* (2017).
- [15] A. Zazunov, V. S. Shumeiko, E. N. Bratus, J. Lantz, and G. Wendin. Andreev level qubit. *Phys. Rev. Lett.* **90**, 087003 (2003).
- [16] N. M. Chtchelkatchev and Y. V. Nazarov. Andreev quantum dots for spin manipulation. *Phys. Rev. Lett.* **90**, 226806 (2003).
- [17] I. Bena, M. Droz, and A. Lipowski. Statistical mechanics of equilibrium and nonequilibrium phase transitions: The Yang-Lee formalism. *International Journal of Modern Physics B* **19**, 4269 (2005).
- [18] J. M. Hickey, C. Flindt, and J. P. Garrahan. Trajectory phase transitions and dynamical Lee-Yang zeros of the Glauber-Ising chain. *Phys. Rev. E* **88**, 012119 (2013).
- [19] J. M. Hickey, C. Flindt, and J. P. Garrahan. Intermittency and dynamical Lee-Yang zeros of open quantum systems. *Phys. Rev. E* **90**, 062128 (2014).
- [20] C. Flindt and J. P. Garrahan. Trajectory phase transitions, Lee-Yang zeros, and high-order cumulants in full counting statistics. *Phys. Rev. Lett.* **110**, 050601 (2013).
- [21] K. Brandner, V. F. Maisi, J. P. Pekola, J. P. Garrahan, and C. Flindt. Experimental determination of dynamical Lee-Yang zeros. *Phys. Rev. Lett.* **118**, 180601 (2017).
- [22] D. Kambly, C. Flindt, and M. Büttiker. Factorial cumulants reveal interactions in counting statistics. *Phys. Rev. B* **83**, 075432 (2011).
- [23] P. Stegmann, B. Sothmann, A. Hucht, and J. König. Detection of interactions via generalized factorial cumulants in systems in and out of equilibrium. *Phys. Rev. B* **92**, 155413 (2015).
- [24] P. Stegmann and J. König. Short-time counting statistics of charge transfer in Coulomb-blockade systems. *Phys. Rev. B* **94**, 125433 (2016).

BIBLIOGRAPHY

- [25] P. Stegmann, J. König, and S. Weiss. Coherent dynamics in stochastic systems revealed by full counting statistics. *arXiv:1803:02175* (2018).
- [26] J. M. Hickey, S. Genway, I. Lesanovsky, and J. P. Garrahan. Time-integrated observables as order parameters for full counting statistics transitions in closed quantum systems. *Phys. Rev. B* **87**, 184303 (2013).
- [27] S. Genway, J. M. Hickey, J. P. Garrahan, and A. D. Armour. Trajectory phases of a quantum dot model. *Journal of Physics A: Mathematical and Theoretical* **47**, 505001 (2014).
- [28] R. S. Souto, A. Martín-Rodero, and A. L. Yeyati. Quench dynamics in superconducting nanojunctions: Metastability and dynamical yang-lee zeros. *Phys. Rev. B* **96**, 165444 (2017).
- [29] L. S. Levitov, H. Lee, and G. B. Lesovik. Electron counting statistics and coherent states of electric current. *Journal of Mathematical Physics* **37**, 4845 (1996).
- [30] M. Esposito, U. Harbola, and S. Mukamel. Nonequilibrium fluctuations, fluctuation theorems, and counting statistics in quantum systems. *Rev. Mod. Phys.* **81**, 1665 (2009).
- [31] A. Shelankov and J. Rammer. Charge transfer counting statistics revisited. *EPL (Europhysics Letters)* **63**, 485 (2003).
- [32] P. P. Hofer and A. A. Clerk. Negative full counting statistics arise from interference effects. *Phys. Rev. Lett.* **116**, 013603 (2016).
- [33] T. D. Lee and C. N. Yang. Statistical theory of equations of state and phase transitions. ii. lattice gas and ising model. *Phys. Rev.* **87**, 410 (1952).
- [34] A. Zazunov, A. Brunetti, A. L. Yeyati, and R. Egger. Quasiparticle trapping, Andreev level population dynamics, and charge imbalance in superconducting weak links. *Phys. Rev. B* **90**, 104508 (2014).
- [35] A. G. Abanov and D. A. Ivanov. Allowed charge transfers between coherent conductors driven by a time-dependent scatterer. *Phys. Rev. Lett.* **100**, 086602 (2008).
- [36] D. A. Ivanov and A. G. Abanov. Phase transitions in full counting statistics for periodic pumping. *EPL (Europhysics Letters)* **92**, 37008 (2010).
- [37] Y. Utsumi, O. Entin-Wohlman, A. Ueda, and A. Aharony. Full-counting statistics for molecular junctions: Fluctuation theorem and singularities. *Phys. Rev. B* **87**, 115407

- (2013).
- [38] C. N. Yang and T. D. Lee. Statistical theory of equations of state and phase transitions. i. theory of condensation. *Phys. Rev.* **87**, 404 (1952).
 - [39] C. Schönenberger and M. Ferrier (private communications) .
 - [40] P. Stadler, W. Belzig, and G. Rastelli. Ground-state cooling of a mechanical oscillator by interference in Andreev reflection. *Phys. Rev. Lett.* **117**, 197202 (2016).
 - [41] Z. Cao, T.-F. Fang, Q.-F. Sun, and H.-G. Luo. Inelastic Kondo-Andreev tunneling in a vibrating quantum dot. *Phys. Rev. B* **95**, 121110 (2017).
 - [42] B. Dong, G. H. Ding, and X. L. Lei. Full counting statistics of phonon-assisted Andreev tunneling through a quantum dot coupled to normal and superconducting leads. *Phys. Rev. B* **95**, 035409 (2017).
 - [43] J. Sköldberg, T. Löfwander, V. S. Shumeiko, and M. Fogelström. Spectrum of Andreev bound states in a molecule embedded inside a microwave-excited superconducting junction. *Phys. Rev. Lett.* **101**, 087002 (2008).
 - [44] L. Bretheau, Ç. Ö. Girit, M. Houzet, H. Pothier, D. Esteve, and C. Urbina. Theory of microwave spectroscopy of Andreev bound states with a Josephson junction. *Phys. Rev. B* **90**, 134506 (2014).
 - [45] S. Maier, T. L. Schmidt, and A. Komnik. Charge transfer statistics of a molecular quantum dot with strong electron-phonon interaction. *Phys. Rev. B* **83**, 085401 (2011).

8 General conclusions and outlook

8.1 English

The research carried out during this thesis covers several areas in the field of electron quantum transport, focusing on the transient regime. On the one hand, chapters 3-5 are devoted to the analysis of quantum transport through nanojunctions coupled to normal metal electrodes. While chapter 3 is focused on the non-interacting situation, chapters 4 and 5 analyze the role of interaction effects in both transient and stationary regimes. On the other hand, chapters 6 and 7 are devoted to the superconducting situation, investigating both the single particle observables and the system fluctuations. In this final chapter the general conclusions of the thesis are presented, discussing also some open questions and the future possible research lines.

8.1.1 Normal nanojunctions

As already discussed in the introductory chapter, the system fluctuations contain a more complete information about the system dynamics than the mean current itself. For this reason, chapter 3 has been devoted to the analysis of the charge statistics in the non-interacting situation. An exact relation between the zeros of the generating function of the transferred electrons and the charge and current cumulants has been derived, valid also in the interacting situation. This relation reproduces the short time universal oscillatory behavior in the high order charge cumulants measured in Ref. [1] in the sequential

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regime. These oscillations are due to the first electron passage and this is the true reason why they are observed either if the measurement is performed in the transient or the steady state regime. The role of coherence in the system is analyzed, observing the appearance of a new set of oscillations at longer times with renormalized amplitudes, in addition to the universal ones. Furthermore, the universality is broken for the small bias voltage situation where the short time transport is bidirectional, due to interference effects of electrons flowing in both of the directions. In any of the situations mentioned above, the oscillatory behavior of the high order cumulants is damped at long times due to an interference effect between the contributions from the different charge transfer processes.

Chapter 4 is devoted to the analysis of the transient dynamics of a molecular junction in the presence of electron-phonon interaction. In that chapter the polaronic regime is explored, characterized by a strong coupling to the vibrational mode and achieved in some experiments [2]. To explore this regime, the DTA is used, which has been developed during the thesis and correctly describes the polaronic limit in both the steady state [3] and the transient regimes [4]. It is found that the interaction tends to enlarge exponentially the relaxation time, although no signatures of a bistable behavior are observed in the charge or the current cumulants. The short time universal oscillatory behavior in the high order cumulants mentioned above is also observed, indicating that the DTA does not include electron correlation effects. Nevertheless, the peculiar system dynamics is correctly described, exhibiting abrupt jumps in the current and noise due to the opening of the phonon sidebands. A particular behavior is found when the bias voltage commensurates with the phonon frequency. When the voltage is an even multiple of the phonon frequency, a series of steps up in the conductance and steps down in the differential Fano factor appear. This corresponds to the elastic threshold where the stationary current is increased [5]. On the other hand, for a bias voltage equal to an odd multiple of the phonon frequency the conductance evolution is oscillatory, converging either to a positive or negative value depending on the system transmission [6]. Interestingly, a similar qualitative behavior has been found in the stationary regime after a projective measurement protocol in Ref. [7].

Finally, in chapter 5 a novel self-consistent algorithm is introduced to treat interactions in the transient regime. The accuracy of the algorithm is discussed by means of perturbative approximations for the electron-electron and the electron-phonon interactions, finding a remarkable agreement with exact numerical results in both the transient

regime and the steady state limit. For the electron-electron interaction case, it is found that the self-consistent mean-field approximation predicts a long time magnetic solution when the system is initialized with a well defined spin. This long time magnetic moment, known to be unphysical [8–10], relaxes when correlation effects are included, converging to the expected singlet solution. The correlation effects, included through a second order expansion in the interaction Hamiltonian, correctly describes the Kondo peak formation at times of the order of the inverse of the Kondo temperature [11]. On the other hand, the mean-field solution for the electron-phonon interaction case predicts a bistable behavior. This bistability tends to disappear for a wide range of parameters when electron correlation effects mediated by phonons are included. This is indicative of the absence of a charge bistable behavior in the polaronic regime at zero temperature, although a rigorous proof is still lacking. Note that it does not necessarily implies that another kind of bistability can be found in the system like, for instance, the one predicted in the phonon displacement in Refs. [12, 13]. The non-equilibrium phonon dynamics is also analyzed, finding that their proper description becomes essential to obtain accurate results in the moderate and the polaronic regimes. Finally, I also analyzed the situation when both electron-electron and electron-phonon interactions are present. It is found that, even in the Kondo dominated regime, the transient spectral function carry important information about the hybrid character of the Kondo peak, which is somehow hidden in the stationary regime.

After the study presented in chapters 3-5 there are several question that remain to be answered. Some of them are summarized here. The first one is related to the role of electron correlation effects in the charge and current cumulants. Since the zeros of the GF of an interacting system appear as complex conjugate pairs [14, 15], a breaking of the short time universality is expected to happen, according to the exact expression derived in this thesis [16]. It would be interesting to characterize the short time dynamics of interacting systems, proposing a way of detecting electron correlation effects. In particular, the previous study would be of fundamental interest for the electron-phonon interaction situation, where signatures of a bistable behavior were found in the phonon displacement [12, 13], but absent in the system population and mean current. Since the current fluctuations contain a more complete information about the system state than the mean charge and current, it would interesting to analyze whether signatures of this possible bistable behavior can be found in the shot noise or in the higher order cumulants.

There are several ways of addressing the questions mentioned above in different regimes.

Firstly, it is possible to include the counting field variable in a rather straightforward way in the algorithm introduced in chapter 5. It would allow to determine the shot noise and the higher order cumulants by successive derivatives of the counting field dependent current [17]. This would be of great interest for addressing the question of the possible bistable behavior for the electron-phonon interaction situation. Another possibility for addressing this question in the polaronic regime is by including electron correlation effects within the DTA scheme. In relation to this issue, it would be interesting to compute the roots of the generating function in combination to the factorial cumulants [15], which allows to identify the different non-equilibrium phases as described in Ref. [18]. Moreover, a possible interesting extension for the algorithm of chapter 5 consists in including non-perturbative approximations, interpolating between the weak and the strong coupling regimes [19–21]. In this sense, it would be relevant to investigate the transition to the insulating phase in a system involving electron-electron and electron-phonon interactions [22, 23], analyzing if the transition happens suddenly in time or, in contrast, if the system exhibits dynamical phase transitions in the sense of the ones proposed in Ref. [24]. Finally, recent experiments have shown that the dynamics of an interacting nanojunction can be analyzed by coupling electromagnetic modes to the system [25]. It would be also of relevance to study the effect of these modes in the system evolution.

8.1.2 Superconducting nanojunctions

The second part of the thesis, composed by chapters 6 and 7, is devoted to the study of the transient dynamics of superconducting nanojunctions. The situation of a spin degenerate level coupled to BCS electrodes is analyzed, which describes the point contact case in the strongly coupled regime. Chapter 6 is focused on the single electron properties: system population, mean current and spectral density. Firstly, it is found that the system population does not relax to its expected steady state value for weak to moderate coupling strengths to the electrodes. Instead, the population exhibits either a long time magnetic moment for an initially trapped spin, or an undamped oscillation for the empty and occupied initializations. Although the population tends to its expected value in the point contact regime, the system exhibits a lower supercurrent with respect to the expected stationary value. This is indicative of an asymptotic non-equilibrium occupation of the ABSs, known as quasiparticle poisoning. It is worth mentioning that the quasiparticle poisoning happens under rather general conditions, irrespectively of the system parameters or the connection procedure to the BCS electrodes. In order to make the system converge to the expected equilibrium situation an extra relaxation mecha-

nism has to be included. The voltage biased situation has been also analyzed, finding that the system converges to the steady state, being the initially trapped quasiparticles relaxed to the continuum spectrum when subgap ABSs approach the gap edges. At long times, the time averaged stationary value of the current is recovered, leading to results in quantitative agreement with the calculations done with standard stationary methods [26].

In chapter 7 the study of chapter 6 is completed by analyzing the charge transferred counting statistics of a superconducting nanojunction in the time domain. It is shown that the charge statistics exhibits three maxima, related to the possible ways of populating the ABSs (named as many body states). The asymptotic occupation of these many body states can be obtained using two equivalent methods: integrating the charge transfer probabilities around the distribution maxima or through the long time current and the shot noise. While the system state exhibits an universal behavior in the point contact regime converging to an asymptotic state which only depends on the position of the ABSs, in the intermediate regime the long time many body populations depend on the system initialization. In particular, in the limit of weakly coupled electrodes, the system initial state remains unperturbed after the connection to the electrodes. By analyzing the short time statistics the development of the Andreev bound states has been addressed, finding that they are formed at times of the order of the inverse of the superconducting gap. This corresponds to a surprisingly small number of electrons crossing the junction in each direction (of the order of 3). As mentioned above, the system relaxes to the expected steady state for the voltage biased situation. The algorithm developed during the thesis reproduces with a good accuracy the stationary shot noise in the point contact regime [27], providing also results for the quantum dot regime which, to the best of my knowledge, has not been addressed in the literature before. Additional insight is provided by the analysis of zeros of the generating function. It has been shown that, in the phase biased situation, they arrange forming two branches in the complex counting field plane which converge to the unit circle at long times. It has been demonstrated that there is a close analogy between phase transition theory in equilibrium statistical mechanics [28] and the charge counting statistics [29]. In this relation, the factorial cumulants can be used to characterize different non-equilibrium phases. The possibility of manipulating the system state by the use of voltage pulses has been also explored. It has been found that, for voltages of the order of the superconducting gap, the system can be prepared either in the even ground or even excited state, depending on the final phase difference. When the pulse amplitude is increased, the quench result is gradually

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recovered. Finally, the situation of a superconducting nanojunction coupled to a bosonic mode has been analyzed by means of the DTA. Even in the weakly interacting regime, when the mode is driven out of equilibrium quasiparticles get trapped within the ABSs. It has been shown that, for a strongly driven mode the occupation of the odd states reaches ~ 0.5 for values of $\epsilon_A \rightarrow 0$, consistent with the recent experimental observations [30, 31].

There are several open questions that remain to be answered after the analysis presented in chapters 6 and 7. The first one is related to the role of interactions in the system thermalization, which would lead to the quasiparticle relaxation due to the inelastic effects. According to the experimental results of Ref. [32], the quasiparticles relaxation in superconducting atomic contacts occurs on time scales larger than $1\mu s$. In particular, it could be interesting to analyze the situation of a local electrostatic repulsion, which can lead to the appearance of the π phase, where the current suddenly changes sign in the stationary situation [33]. It would be worth analyzing the electron correlation effects near the phase transition point and to investigate if the insulating phase appears suddenly in time or, in contrast, if dynamical phase transitions occur in the time evolution [24]. In this situation, it becomes also important to address the shot noise in the voltage biased situation, which has been recently measured [34]. Another open question is related to the situation of a boson mode coupled to the superconducting nanojunction, briefly discussed in the final part of chapter 7. In this direction it can be interesting to investigate the role of electron correlation effects through the approximations discussed in chapter 5, analyzing also the differences between the driven and the undriven mode situations.

The questions mentioned above can be addressed in several ways. For instance, a straightforward extension of the algorithm proposed in section 5 to the superconducting situation can be used to address the single particle properties using self-consistent perturbative approximations. For the electron-electron interacting situation the second order approximation in the coupling parameter has shown to properly describe the transition to the π phase in Ref. [35]. On the other hand, the self-consistent perturbative approximations for the electron-phonon interaction presented in chapter 5 are also expected to provide accurate results in the superconducting situation for weak to moderate coupling strengths. Both the equilibrated and unequilibrated phonon regimes can be analyzed using those approximations. Moreover, the algorithm can be generalized to include the counting field variable, as mentioned above. It will allow to evaluate also the current cumulants, important to describe some recent experiments like for instance the ones an-

alyzing the shot noise for a system exhibiting local electrostatic repulsion [34].

It is also worth mentioning that a possible extension of the work done in the chapters 6 and 7 consists on analyzing the situation involving topological materials. For instance, it would be interesting to analyze the situation of a one dimensional topological superconductor, which exhibits Majorana-like states at its ends. By analyzing the dynamics, singular signatures of the topological situation could be found, which may lead to a Majorana detection proposal. It is also of essential importance to investigate the robustness of the topological superconductor against the quasiparticle poisoning. In this sense, the system state could be achieved in the equilibrium and non-equilibrium situation, addressing the internal system state through a counting statistics analysis as done for the conventional BCS electrodes in chapter 7. The interacting situation including Majorana bound states is also worth to be studied.

To summarize, this thesis proposes novel methodologies to explore the transient dynamics and the steady properties of mesoscopic systems. It can be used for a numerical evaluation of the charge and current statistics of interacting and superconducting nanoscopic junctions. The research performed in this thesis addresses, not only some recent hot topics in the field, but it also revisits some previously studied problems, providing an interesting complementary point of view by analyzing the system evolution. Although a quite complete overview of the transport paradigmatic problems have been provided, there are many promising lines which can be followed in the future to better understand the transport phenomena at the nanoscale. For instance, it is interesting the connection to the recent experimental advances to measure the transport in time domain [30, 31, 36], which surely will renovate the current interest in the field. It is then justified to assert that the nanoelectronics field has a bright future ahead.

8.2 Castellano

La investigación llevada desarrollada a lo largo de la tesis cubre diferentes áreas del campo del transporte cuántico, centrándose en el régimen transitorio. Por un lado, los capítulos 3-5 están dedicados al análisis del transporte electrónico a través de uniones nanoscópicas acopladas a electrodos metálicos. Mientras el capítulo 3 analiza la situación no interactuante, en los capítulos 4 y 5 se discuten los efectos de las interacciones electrónicas en el régimen transitorio y estacionario. Por otro lado, los capítulos 6 y 7 es-

tán dedicados al estudio del caso superconductor, investigando los observables de una partícula y las fluctuaciones de carga y corriente del sistema. En este capítulo final se presentan las conclusiones generales de la tesis, discutiendo algunas cuestiones abiertas así como posibles futuras líneas de investigación.

8.2.1 Uniones normales

Como se menciona en el capítulo introductorio, las fluctuaciones del sistema contienen información completa a cerca de la dinámica del sistema. Por esta razón, el capítulo 3 está dedicado al análisis de la estadística de electrones transferidos a través de una unión no interactuante. En ese capítulo se deriva una relación exacta entre los cumulantes de la carga y la corriente transferida y los ceros de la función generatriz, también válida para situaciones con interacción. Dicha relación reproduce las oscilaciones observadas en los cumulantes de orden alto [1] en el régimen secuencial. Estas oscilaciones a tiempos cortos son generadas por el paso del primer electrón, razón por la cual son observadas tanto en el régimen estacionario como en el transitorio. Los efectos de la coherencia electrónica son analizados, observando la aparición de una nueva secuencia de oscilaciones con amplitudes renormalizadas a tiempos más largos, aparte de las universales anteriormente mencionadas. Además, la universalidad es destruida cuando el transporte a tiempos cortos es bidireccional, debido a la interferencia de las contribuciones de las cargas transferidas en sentidos opuestos. En cualquier caso, las oscilaciones son atenuadas a tiempos largos debido a la interferencia de los procesos de transferencia electrónica.

El capítulo 4 está dedicado al análisis de la dinámica transitoria de uniones moleculares en presencia de la interacción electrón-fonón. El capítulo se centra en el régimen polarónico caracterizado por un acoplo fuerte al modo de vibración, medido en algunos experimentos [2]. Este régimen es explorado usando la aproximación DTA, desarrollada a lo largo de la tesis y que describe correctamente los regímenes estacionario [3] y transitorio [4]. Debido a la interacción, el tiempo de relajación del sistema es alargado exponencialmente, aunque no hay evidencias de un comportamiento biestable en las propiedades electrónicas. Al igual que en el caso no interactuante, los cumulantes de orden alto presentan oscilaciones universales a tiempos cortos, indicando que la DTA no incluye efectos de correlación electrónica. Sin embargo, dicha aproximación describe correctamente la dinámica del sistema, que manifiesta saltos en la corriente y en el ruido debido a la apertura en tiempo de procesos elásticos e inelásticos. Un comportamiento peculiar es observado para voltajes proporcionales a la frecuencia del fonón. Para voltajes múltiples

pares de la frecuencia del fonón una serie de escalones hacia arriba aparecen en la conductancia (y hacia abajo en el factor de Fano diferencial). Esta situación se corresponde con el umbral elástico, donde la corriente presenta un incremento en el régimen estacionario [5]. Por otro lado, para voltajes iguales a múltiplos impares de la frecuencia del fonón, la conductancia presenta un comportamiento oscilatorio, convergiendo a tiempos largos a valores positivos o negativos dependiendo de la transmisión [6]. Un comportamiento similar ha sido observado después de una medida proyectiva en el régimen estacionario en la referencia [7].

Finalmente, en el capítulo 5 se introduce un nuevo algoritmo para tratar interacciones de una manera autoconsistente en el régimen transitorio. La precisión del algoritmo es discutido usando aproximaciones perturbativas para las interacciones electrón-electrón y electrón-fonón, encontrando un excelente acuerdo con los cálculos numéricos exactos en los regímenes transitorios y estacionarios. En el caso de la interacción electrón-electrón, la aproximación autoconsistente de campo medio describe la convergencia a tiempo largo a una solución magnética cuando el sistema es inicializado con un determinado spin. Dicha solución magnética se sabe que no física [8–10], y es relajada cuando los efectos de correlación del sistema son incluidos, convergiendo al estado singlete. A través de una aproximación perturbativa de segundo orden en el término de interacción del Hamiltoniano los efectos de correlación son incluidos, describiendo correctamente la formación del pico Kondo a tiempos del orden del inverso de la temperatura Kondo [11]. Por otro lado, la aproximación de campo medio para la interacción electrón-fonón predice la aparición de un comportamiento biestable para interacciones fuertes. Esta biestabilidad tiende a desaparecer para un amplio rango de parámetros cuando los efectos de correlación electrónica mediados por los fonones son incluidos. Esto es indicativo de una ausencia de biestabilidad en las propiedades electrónicas del sistema en el régimen polarónico y a temperatura cero, aunque hasta el momento no existe una demostración rigurosa. Es importante mencionar que esto no implica necesariamente que otro tipo de comportamiento biestable, como por ejemplo en el desplazamiento del fonón [12, 13], no pueda existir. Los efectos de la dinámica de no equilibrio de fonones es también analizada, demostrando que su correcta descripción es esencial para obtener resultados precisos en el régimen de acoplo intermedio y polarónico. Finalmente, la situación que involucra interacciones electrón-electrón y electrón-fonón simultáneamente es estudiada. Incluso en el régimen en el que el efecto Kondo domina, la evolución de la función espectral contiene información importante a cerca del carácter híbrido del pico Kondo que, de alguna manera, está oculto en el régimen estacionario.

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Después del estudio presentado en los capítulos 3-5 existen varias cuestiones que faltan por ser respondidas, algunas de las cuales son resumidas a continuación. La primera de ellas está relacionada con los efectos de correlación electrónica en los cumulantes de la corriente y la carga. Dado que en dicho caso los ceros de la función generatriz aparece como pares complejos conjugados [14, 15], una ruptura de la universalidad a tiempos cortos de los cumulantes de orden alto se espera que ocurra, de acuerdo a la relación exacta derivada en la tesis [16]. En esta dirección sería interesante el caracterizar la dinámica a tiempos cortos, proponiendo maneras de detectar los efectos de correlación electrónica. En particular, el estudio mencionado previamente sería de fundamental interés en el caso de la interacción electrón-fonón, donde evidencias de un comportamiento biestable han sido encontradas en el desplazamiento del fonón [12, 13], aunque no en la población del sistema y en la corriente. Dado que las fluctuaciones de corriente contienen una información más completa que las propiedades de una sola partícula, sería importante analizar evidencias del posible comportamiento biestable en el ruido o en los cumulantes de orden más alto.

Hay diferentes maneras de abordar las cuestiones mencionadas anteriormente. En primer lugar sería posible incluir la variable del campo de recuento de una manera relativamente directa en el algoritmo introducido en el capítulo 5, que permitiría acceder al ruido y a los cumulantes de orden elevado a través de derivadas sucesivas [17]. Esta extensión del algoritmo sería importante para analizar el posible comportamiento biestable en el caso de interacción electrón-fonón. Otra posibilidad para analizar esta cuestión en el régimen polarónico sería a través de la inclusión de correlaciones electrónicas en el esquema de la DTA. En relación a esta pregunta sería interesante determinar los ceros de la función generatriz así como el cálculo de cumulantes factoriales [15], que permitirían identificar y caracterizar las diferentes fases de no equilibrio tal y como se describe en la referencia [18]. Además, una posible extensión del algoritmo presentado en el capítulo 5 podría consistir en incluir aproximaciones no perturbativas que interpolen bien entre los regímenes de acoplo débil y fuerte [19–21]. En concreto sería relevante analizar la transición aislante en un sistema que involucra interacción electrón-electrón y electrón-fonón [22, 23], estudiando si ésta sucede abruptamente en tiempo o si, por el contrario, el sistema presenta transiciones de fase dinámicas como las propuestas en la referencia [24]. Finalmente, algunos experimentos recientes han demostrado que la dinámica de sistemas interactuantes puede ser analizada acoplando modos electromagnéticos a la unión [25]. Sería interesante estudiar el efecto de estos modos en la dinámica

electrónica.

8.2.2 Uniones superconductoras

La segunda parte de la tesis, compuesta por los capítulos 6 y 7, está dedicada al estudio de la dinámica transitoria de uniones superconductoras. Para estudiar este problema se ha tomado el caso de un nivel con degeneración de spin acoplado a electrodos BCS, que describe la situación de un contacto atómico en el régimen de acoplo fuerte a los electrodos. El capítulo 6 se centra en el estudio de las propiedades de electrones individuales: población del sistema, corrientes y densidades espectrales. En primer lugar, se ha encontrado que la población del sistema no se relaja al valor estacionario esperado para acoplos débiles o moderados a los electrodos. Esto induce, o bien una solución magnética a tiempo largo para un sistema inicializado con un determinado spin, o bien una oscilación en el tiempo que no se amortigua para un sistema inicialmente vacío o lleno. Por otro lado, a pesar de que la población del sistema en el régimen de contacto tiende al valor esperado a tiempo largo, la super-corriente presenta un valor menor con respecto al estacionario. Esto es indicativo de que los estados de Andreev presentan una población de no equilibrio, también llamado *quasiparticle poisoning* (envenenamiento por cuasipartículas sería su traducción literal). Es importante mencionar que esto ocurre para condiciones bastante generales, independientemente de los parámetros del sistema o del proceso de conexión. Para hacer que el sistema relaje a la situación de equilibrio, mecanismos adicionales tienen que ser incluidos. Además, la situación de no equilibrio con voltaje es analizada, mostrando que las cuasipartículas son relajadas cuando los estados de Andreev se acercan al borde del gap superconductor. A tiempos largos, el valor de la corriente promediada en tiempo recupera el límite estacionario [26].

En el capítulo 7 se completa el estudio del capítulo 6 incluyendo el análisis de la estadística de cargas transferidas a través de la unión superconductora en el tiempo. Se muestra que la estadística muestra tres máximos relacionados con las tres posibles ocupaciones de los estados Andreev (estados de muchos cuerpos). La ocupación asintótica de los estados de muchos cuerpos puede ser obtenida de dos maneras equivalentes: integrando la estadística de cargas transferidas alrededor de sus máximos o a través de la evolución de la corriente y el ruido. Mientras que el sistema presenta un comportamiento universal dependiente solo de la energía de los estados Andreev en el régimen de contacto, para acoplos intermedios a los electrodos la población depende de la inicialización del sistema. En particular, en el límite de acoplo débil a los electrodos, el estado del sistema

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permanece inalterado después de la conexión. Analizando la estadística a tiempos cortos, la formación de los estados de Andreev puede ser estudiada, encontrando que son formados a tiempos del orden del inverso del gap superconductor. Esto se corresponde con la transferencia de un número sorprendentemente pequeño de electrones cruzando la unión en ambos sentidos (de en torno a 3 electrones). Como se menciona anteriormente, la corriente del sistema se relaja en el caso de un voltaje aplicado a la unión. El algoritmo desarrollado a lo largo de la tesis es también capaz de reproducir los resultados para el ruido en el régimen de contacto con buena precisión, así como de proporcionar resultados en el régimen de punto cuántico que, hasta donde sé, no han sido reportados anteriormente en la literatura. Un punto de vista alternativo es proporcionado por los ceros de la función generatriz. En la tesis se muestra que, en la situación de equilibrio, los ceros se agrupan en el plano complejo de la variable de recuento formando dos ramas que convergen al círculo unidad a tiempos largos. Se demuestra, además, que existe una relación estrecha entre la teoría de transiciones de fase en la mecánica estadística de equilibrio y la estadística de recuento de cargas transferidas [29]. En esta relación, los cumulantes factoriales pueden ser utilizados para caracterizar las diferentes fases de no equilibrio. La posibilidad de manipular del estado del sistema a través de pulsos de voltaje es también explorada. Se encuentra que, para voltajes del orden del gap superconductor el sistema es inicializado en el estado fundamental o excitado, dependiendo de la fase final. Sin embargo, si la amplitud del pulso se incrementa, el resultado tiende a recuperar gradualmente el caso de la conexión abrupta a los electrodos. Finalmente, la situación de una unión superconductora acoplada a un modo bosónico es analizada a través de la aproximación DTA. Incluso en el régimen de interacción débil, cuando el modo bosónico es excitado fuera del equilibrio, existe una posibilidad de excitar cuasipartículas que, eventualmente, quedan atrapadas en los estados de Andreev. Se demuestra además que, para un modo muy excitado, la población del estado de paridad opuesta al estado fundamental es ~ 0.5 para $\epsilon_A \rightarrow 0$, consistente con las recientes observaciones experimentales [30, 31].

Existen numerosas cuestiones abiertas después del trabajo presentado en los capítulos 6 y 7. La primera está relacionada con el efecto de las interacciones en la termalización del sistema, que podría producir la relajación de las cuasipartículas atrapadas al incluir efectos inelásticos. De acuerdo con los resultados experimentales de la referencia [32], el tiempo de relajación de las cuasipartículas en un contacto superconductor es más largo que $1\mu s$. En particular sería interesante analizar la situación de repulsión electrostática local, que produce la aparición de la fase π , donde la corriente estacionaria presenta un

cambio de signo [33]. También sería interesante analizar los efectos de la correlación electrónica cerca de la transición, investigando si ésta sucede de forma abrupta en tiempos o si, por el contrario, transiciones dinámicas aparecen [24]. En esta situación es importante estudiar el ruido de no equilibrio que ha sido recientemente medido experimentalmente [34]. Otra cuestión relevante es la que involucra un modo bosónico acoplado a la unión, discutida brevemente al final del capítulo 7. En este respecto, sería importante investigar el papel de las correlaciones electrónicas a través de las aproximaciones usadas en el capítulo 5. También sería de utilidad el discutir las diferencias entre las situaciones de un modo excitado o no excitado por fuentes externas.

Las cuestiones planteadas anteriormente pueden ser respondidas de diferentes maneras. En primer lugar, una extensión del algoritmo propuesto en el capítulo 5 para el caso superconductor podría servir para analizar las propiedades de electrones individuales a través de aproximaciones autoconsistentes. Por ejemplo, la interacción electrón-electrón podría ser analizado a través de una aproximación de segundo orden que describe adecuadamente la transición a la fase π , tal y como se discute en la referencia [35]. Por otro lado, es de esperar que las aproximaciones autoconsistentes del capítulo 5 para la interacción electrón-fonón también proporcionen buenos resultados en la situación superconductora para acoplos moderados o intermedios. Las situaciones de equilibrio y no equilibrio de fonones pueden ser analizadas a través de esas aproximaciones. Además, el algoritmo podría ser generalizado para incluir el campo de recuento, tal y como se ha mencionado anteriormente. Esto permitiría la evaluación de los cumulantes de la corriente, importantes para describir algunos experimentos recientes que involucran repulsión electrostática local [34].

Es importante mencionar también que una posible extensión de estos trabajos podría consistir en analizar situaciones que involucran materiales topológicos. En concreto, sería relevante analizar la el caso de un superconductor topológico en cuyos extremos aparecen estados de Majorana. Analizando su dinámica, evidencias del carácter topológico del material podrían emerger, lo que podría ser utilizado para proponer un método para detectar Fermiones de Majorana. También sería de esencial importancia investigar la robustez del superconductor topológico frente a las posibles cuasipartículas atrapadas en los estados de Andreev. En este sentido, el estado del sistema podría ser determinado en las situaciones de equilibrio y de no equilibrio a través de la estadística de cargas transferidas, al igual que se hizo en el capítulo 7 para superconductores BCS. Finalmente, el caso interactuante también es relevante de ser analizado en presencia de estados de los

Majorana.

Resumiendo, en la tesis se proponen una serie de metodologías para acceder a la dinámica transitoria y propiedades estacionarias de sistemas mesoscópicos. Estos métodos se podrían usar para calcular numéricamente la corriente promedio y su estadística en sistemas nanoscópicos interactuantes. La investigación desarrollada en esta tesis cubre, no solo algunos temas candentes en el campo, sino que también proporciona una visión alternativa de algunos problemas ya estudiados en el campo a través del análisis de la dinámica en tiempo real. Aunque la tesis presenta una visión bastante amplia del campo, analizando muchos de los ejemplos paradigmáticos, hay varias líneas de investigación prometedoras con las que se podría profundizar en el entendimiento del transporte electrónico en la nanoescala. En concreto, es interesante la conexión con los recientes avances experimentales en la medida del transporte en el dominio temporal, que seguramente renovarán el interés en el campo [30, 31, 36]. Por tanto, está justificado afirmar que el campo de la nanoelectrónica tiene un brillante futuro por delante.

Bibliography

- [1] C. Flindt, C. Fricke, F. Hohls, T. Novotný, K. Netočný, T. Brandes, and R. J. Haug. Universal oscillations in counting statistics. *Proceedings of the National Academy of Sciences* **106**, 10116 (2009).
- [2] H. Park, J. Park, A. K. L. Lim, E. H. Anderson, A. P. Alivisatos, and P. L. McEuen. Nanomechanical oscillations in a single-C60 transistor. *Nature* **407**, 57 EP (2000).
- [3] R. Seoane Souto, A. L. Yeyati, A. Martín-Rodero, and R. C. Monreal. Dressed tunneling approximation for electronic transport through molecular transistors. *Phys. Rev. B* **89**, 085412 (2014).
- [4] R. Seoane Souto, R. Avriller, R. C. Monreal, A. Martín-Rodero, and A. Levy Yeyati. Transient dynamics and waiting time distribution of molecular junctions in the polaronic regime. *Phys. Rev. B* **92**, 125435 (2015).
- [5] L. Mühlbacher and E. Rabani. Real-time path integral approach to nonequilibrium many-body quantum systems. *Phys. Rev. Lett.* **100**, 176403 (2008).
- [6] L. de la Vega, A. Martín-Rodero, N. Agraït, and A. L. Yeyati. Universal features of electron-phonon interactions in atomic wires. *Phys. Rev. B* **73**, 075428 (2006).

- [7] G. Tang, Y. Xing, and J. Wang. Short-time dynamics of molecular junctions after projective measurement. *Phys. Rev. B* **96**, 075417 (2017).
- [8] P. B. Wiegmann. Towards an exact solution of the Anderson model. *Phys. Lett.* **80A**, 163 (1980).
- [9] N. Kawakami and A. Okiji. Exact expression of the ground-state energy for the symmetric Anderson model. *Physics Letters A* **86**, 483 (1981).
- [10] N. Andrei, K. Furuya, and J. H. Lowenstein. Solution of the Kondo problem. *Rev. Mod. Phys.* **55**, 331 (1983).
- [11] H. T. M. Nghiem and T. A. Costi. Time evolution of the Kondo resonance in response to a quench. *Phys. Rev. Lett.* **119**, 156601 (2017).
- [12] J. Klatt, L. Mühlbacher, and A. Komnik. Kondo effect and the fate of bistability in molecular quantum dots with strong electron-phonon coupling. *Phys. Rev. B* **91**, 155306 (2015).
- [13] R. Avriller, B. Murr, and F. Pistolesi. Bistability and displacement fluctuations in a quantum nanomechanical oscillator. *Phys. Rev. B* **97**, 155414 (2018).
- [14] A. G. Abanov and D. A. Ivanov. Allowed charge transfers between coherent conductors driven by a time-dependent scatterer. *Phys. Rev. Lett.* **100**, 086602 (2008).
- [15] D. Kambly, C. Flindt, and M. Büttiker. Factorial cumulants reveal interactions in counting statistics. *Phys. Rev. B* **83**, 075432 (2011).
- [16] R. Seoane Souto, A. Martín-Rodero, and A. Levy Yeyati. Analysis of universality in transient dynamics of coherent electronic transport. *Fortschritte der Physik* **65**, 1600062 (2017).
- [17] A. O. Gogolin and A. Komnik. Towards full counting statistics for the Anderson impurity model. *Phys. Rev. B* **73**, 195301 (2006).
- [18] R. S. Souto, A. Martín-Rodero, and A. L. Yeyati. Quench dynamics in superconducting nanojunctions: Metastability and dynamical yang-lee zeros. *Phys. Rev. B* **96**, 165444 (2017).
- [19] A. Martín-Rodero, E. Louis, F. Flores, and C. Tejedor. Interpolative solution for the periodic Anderson model of mixed-valence compounds. *Phys. Rev. B* **33**, 1814 (1986).

BIBLIOGRAPHY

- [20] A. L. Yeyati, A. Martín-Rodero, and F. Flores. Electron correlation resonances in the transport through a single quantum level. *Phys. Rev. Lett.* **71**, 2991 (1993).
- [21] H. Kajueter and G. Kotliar. New iterative perturbation scheme for lattice models with arbitrary filling. *Phys. Rev. Lett.* **77**, 131 (1996).
- [22] A. C. Hewson and D. Meyer. Numerical renormalization group study of the Anderson-Holstein impurity model. *Journal of Physics: Condensed Matter* **14**, 427 (2002).
- [23] G. S. Jeon, T.-H. Park, and H.-Y. Choi. Numerical renormalization-group study of the symmetric Anderson-Holstein model: Phonon and electron spectral functions. *Phys. Rev. B* **68**, 045106 (2003).
- [24] M. Heyl, A. Polkovnikov, and S. Kehrein. Dynamical quantum phase transitions in the transverse-field Ising model. *Phys. Rev. Lett.* **110**, 135704 (2013).
- [25] D. Shaoqing, K. Yoshida, H. Ikutaro, and K. Hirakawa. Terahertz dynamics of electron-vibron coupling in single molecules with tunable electrostatic potential. *arXiv:1712.07339* (2017).
- [26] J. C. Cuevas, A. Martín-Rodero, and A. L. Yeyati. Hamiltonian approach to the transport properties of superconducting quantum point contacts. *Phys. Rev. B* **54**, 7366 (1996).
- [27] J. C. Cuevas, A. Martín-Rodero, and A. L. Yeyati. Shot noise and coherent multiple charge transfer in superconducting quantum point contacts. *Phys. Rev. Lett.* **82**, 4086 (1999).
- [28] C. N. Yang and T. D. Lee. Statistical theory of equations of state and phase transitions. i. theory of condensation. *Phys. Rev.* **87**, 404 (1952).
- [29] C. Flindt and J. P. Garrahan. Trajectory phase transitions, Lee-Yang zeros, and high-order cumulants in full counting statistics. *Phys. Rev. Lett.* **110**, 050601 (2013).
- [30] C. Janvier, L. Tosi, L. Bretheau, Ç. Ö. Girit, M. Stern, P. Bertet, P. Joyez, D. Vion, D. Esteve, M. F. Goffman, H. Pothier, and C. Urbina. Coherent manipulation of Andreev states in superconducting atomic contacts. *Science* **349**, 1199 (2015).
- [31] M. Hays, G. de Lange, K. Serniak, D. J. van Woerkom, D. Bouman, P. Krogstrup, J. Nygård, A. Geresdi, and M. H. Devoret. Direct microwave measurement of Andreev-bound-state dynamics in a proximitized semiconducting nanowire.

- arXiv:1711.01645 (2017).
- [32] M. Zgirski, L. Bretheau, Q. Le Masne, H. Pothier, D. Esteve, and C. Urbina. Evidence for long-lived quasiparticles trapped in superconducting point contacts. *Phys. Rev. Lett.* **106**, 257003 (2011).
 - [33] A. Martín-Rodero and A. L. Yeyati. Josephson and Andreev transport through quantum dots. *Advances in Physics* **60**, 899 (2011).
 - [34] C. Schönenberger and M. Ferrier (private communications) .
 - [35] M. Žonda, V. Pokorný, V. Janiš, and T. Novotný. Perturbation theory for an Anderson quantum dot asymmetrically attached to two superconducting leads. *Phys. Rev. B* **93**, 024523 (2016).
 - [36] J. Dubois, T. Jullien, F. Portier, P. Roche, A. Cavanna, Y. Jin, W. Wegscheider, P. Roulleau, and D. C. Glattli. Minimal-excitation states for electron quantum optics using levitons. *Nature* **502**, 659 EP (2013).

A Numerical renormalization group

In this appendix the NRG algorithm is introduced, which has been used along the thesis to obtain equilibrium transport properties. This algorithm allows for a non-perturbative treatment of impurity problems coupled to electronic and/or bosonic baths. The method was originally developed by Kenneth G. Wilson in the middle of the 1970s decade and initially used to solve the Kondo model in the equilibrium situation [1]. After the original works, the method has been extended to large variety of problems involving localized electrostatic repulsion [2] or electron-phonon interactions [3, 4]. More recently, the algorithm has been extended to the non-equilibrium and the non-stationary situations [5–7].

The essential idea of the method is the same in all the cases. The method is based on a logarithmic discretization of the conductance band, which leads to an exponentially increasing precision for the energies close to the Fermi edge. This problem can be mapped into an impurity coupled to a semi-infinite chain of sites with decreasing tunneling amplitudes between the nearest neighbors. The algorithm treats the problem in an interacting way, discarding the higher energy states, which have exponentially small contributions to the system ground state (and thus, to the low temperature system properties). In this chapter the method is briefly explained, discussing the solution for the spinless Anderson-Holstein model.

A.1 Algorithm

In this first section, the algorithm is going to be formulated for the non-interacting situation. Although this has been done a long time ago [1, 2], more recent improvements have extended the method to compute the high energy equilibrium properties [5, 6]. In order to introduce the general algorithm, the simplest model of an spinless impurity

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coupled to a single electrode is considered

$$H = H_{lead} + H_S + H_T, \quad (\text{A.1})$$

where the Hamiltonian of the electrode and the system are given by

$$H_{leads} = \sum_k \epsilon_k c_k^\dagger c_k, \quad H_S = \epsilon_0 d^\dagger d, \quad (\text{A.2})$$

where c and d are the annihilation operators in the electrode and the system respectively. The tunneling term is given by

$$H_T = \sum_k t_k (c_k^\dagger d + d^\dagger c_k). \quad (\text{A.3})$$

A continuous representation is usually more convenient (see for example Ref. [8] for derivation details), finding that the transformed lead and tunneling Hamiltonians are written as

$$H_{lead} = \int_{-D}^D d\epsilon \epsilon b_k^\dagger(\epsilon) b_k(\epsilon), \quad H_T = \int_{-D}^D d\epsilon \rho^{-1/2}(\epsilon) t(\epsilon) [b^\dagger(\epsilon) d + d^\dagger b(\epsilon)], \quad (\text{A.4})$$

where D is the bandwidth and b the electrode annihilation operator in the continuous notation. A further simplification follows by considering the density of states at the Fermi edge and the tunneling amplitude to be energy independent (wideband approximation), i.e. $\rho(\epsilon) = \rho_0$ and $t(\epsilon) = t$. Thus, the tunneling rate can be written as $\Gamma = \pi t^2 \rho$. Then, the Hamiltonian is given by

$$\frac{H_0}{D} = \int_{-1}^1 d\epsilon \epsilon b_k^\dagger(\epsilon) b_k(\epsilon) + \frac{\epsilon_0}{D} d^\dagger d + \left[\frac{\Gamma}{\pi D} \right]^{1/2} \int_{-1}^1 d\epsilon [b^\dagger(\epsilon) d + d^\dagger b(\epsilon)], \quad (\text{A.5})$$

where all the energy parameters are normalized with respect to the electrode's bandwidth.

A.1.1 Logarithmic discretization

The accuracy of the method is related to the logarithmic discretization of the electrode's band. For instance, for a parameter $\Lambda > 1$ the band can be discretized in subintervals separated by Λ^{-n} , with $n \in \mathbb{N}$, as depicted in the top panel of Fig. A.1. In this way, the energies around the Fermi edge can be determined with arbitrary precision since the sampling around this point can be taken as smooth as desired by increasing the number of intervals. The precision over the higher energies can be improved by making Λ smaller, $\Lambda \rightarrow 1$ corresponding to the continuum limit. In each subinterval it is possible to define a complete set of orthonormal functions

Figure A.1: Top panel: logarithmic discretization of the conduction band using the Λ parameter from the NRG algorithm. In the bottom panel the mapping to the chain is shown, where the blue site on the left represents the impurity and the higher order sites are used to resolve progressively small energies.

$$\Psi_{nk}^{\pm}(\epsilon) = \begin{cases} A_{nk} e^{\pm i\omega_n} & \text{if } \Lambda^{-(n+1)} \leq |\epsilon| \leq \Lambda^{-n} \\ 0 & \text{Otherwise,} \end{cases} \quad k \in \mathbb{Z}, \quad (\text{A.6})$$

where the constant prefactor can be obtained by imposing the normalization condition to the previously written wavefunctions, finding $A_{nk} = \Lambda^{n/2}/\sqrt{1 - 1/\Lambda}$. By imposing orthogonality between the functions it is possible to find that $\omega_n = 2\pi\Lambda^n/(1 - 1/\Lambda)$. The discretized wavefunctions allow to write the continuous operator from Eqs. (A.4) in a discretized way as

$$b(\epsilon) = \sum_{nk} [a_{nk}^+ \Psi_{nk}^+(\epsilon) + b_{nk}^- \Psi_{nk}^-(\epsilon)], \quad (\text{A.7})$$

which can be considered as a discrete Fourier transform of the continuous operator

$$a_{nk} = \int_{-1}^1 d\epsilon b(\epsilon) \Psi_{nk}^+, \quad b_{nk} = \int_{-1}^1 d\epsilon b(\epsilon) \Psi_{nk}^-, \quad (\text{A.8})$$

where the operators a_{nk} and b_{nk} satisfy the usual anticommutation relations. The tunneling Hamiltonian from Eq. (A.4) can be written as

$$\int_{-1}^1 d\epsilon a(\epsilon) = \sqrt{1 - \Lambda^{-1}} \sum_n \Lambda^{-n/2} (a_{n0} + b_{n0}), \quad (\text{A.9})$$

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which illustrates that the impurity only couples to the $k = 0$ operators. Thus, the tunneling Hamiltonian in the discretized form is given by

$$H_T = \sqrt{\frac{\Gamma}{\pi D(1 - \Lambda^{-1})}} \left\{ \sum_n \lambda^{-n/2} [(a_{n0}^\dagger + b_{n0}^\dagger) d + d^\dagger (a_{n0} + b_{n0})] \right\}. \quad (\text{A.10})$$

It becomes useful to define a new fermionic operator as

$$f_0 = \sqrt{\frac{1 - \Lambda^{-1}}{2}} \sum_{n=0}^{\infty} (a_{n0} + b_{n0}) \Lambda^{-n/2} = \frac{1}{\sqrt{2}} \int_{-1}^1 b(\epsilon), \quad (\text{A.11})$$

which satisfy the usual anticommutation relation $[f_0, f_0^\dagger] = 1$. Then, the tunneling Hamiltonian can be simplified, finding

$$H_T = \sqrt{\frac{2\Gamma}{\pi D}} (f_0^\dagger d + d^\dagger f_0). \quad (\text{A.12})$$

On the other hand the kinetic energy term is given in the discrete mesh by

$$\begin{aligned} \int_{-1}^1 d\epsilon \epsilon a^\dagger(\epsilon) a(\epsilon) &= \frac{1 + \Lambda^{-1}}{2} \sum_{nk} (a_{nk}^\dagger a_{nk} - b_{nk}^\dagger b_{nk}) + \\ \frac{1 - \Lambda^{-1}}{2\pi i} \sum_{n, k \neq k'} \frac{\Lambda^{-n}}{k - k'} (a_{nk}^\dagger a_{nk'} - b_{nk}^\dagger b_{nk'}) \exp \left[\frac{2\pi i(k - k')}{1 - \Lambda^{-1}} \right]. \end{aligned} \quad (\text{A.13})$$

The second term of the previous equation, written in the lower part, is usually neglected for several reasons. The first reason is related to the fact that it tends to zero in the continuous limit ($\Lambda \rightarrow 1$). Moreover, it is strongly oscillating. Furthermore, note that the system only couples to the $k = 0$ terms, as mentioned above. For these reasons, the Hamiltonian of the electrode can be approximated with good precision as

$$H_{lead} \approx \frac{1 + \Lambda^{-1}}{2} \sum_{n=0}^{\infty} (a_n^\dagger a_n - b_n^\dagger b_n), \quad (\text{A.14})$$

where the k subindex term has been dropped for simplicity. Differently from the previously discussed tunneling term (A.12), any unitary transformation to a new set of operators will produce non-diagonal coupling terms, i.e. $f_n^\dagger f_{n'}$ with $n \neq n'$. The best choice is an unitary transformation to a tridiagonal representation which can be mapped into a linear tight-binding chain with only first neighbor hopping terms (as depicted in the lower panel of Fig. A.1), finding

$$\lambda^{-n} (a_n^\dagger a_n - b_n^\dagger b_n) \rightarrow \lambda^{-n/2} \xi_n (f_n^\dagger f_{n+1} + f_{n+1}^\dagger f_n), \quad (\text{A.15})$$

where

$$\xi_n = [1 - \lambda^{(n+1)}] \sqrt{1 - \Lambda^{-(2n+1)}} \sqrt{1 - \Lambda^{-(2n+3)}}, \quad (\text{A.16})$$

which exponentially converges to the unit when $n \rightarrow \infty$. Then, summarizing, the Hamiltonian can be written as

$$\frac{H}{D} = \frac{1 + \Lambda^{-1}}{2} \sum_{n=0}^{\infty} \Lambda^{-n/2} \xi_n [f_n^\dagger f_{n+1} + f_{n+1}^\dagger f_n] + \left[\frac{2\Gamma}{\pi D} \right]^{1/2} (f_0^\dagger d + d^\dagger f_0) + \frac{\epsilon_0}{D} d^\dagger d. \quad (\text{A.17})$$

Note that the spin degree of freedom can be straightforwardly included, as well as interaction terms like the local Coulomb repulsion in the Hamiltonian (A.1).

A.1.2 Recursive diagonalization

In order to find a numerical solution to the problem, it becomes convenient to write the Hamiltonians as a sequence

$$H_N = \Lambda^{(N-1)/2} \left\{ \sum_{n=1}^{N-1} \Lambda^{-n/2} \xi_n [f_n^\dagger f_{n+1} + f_{n+1}^\dagger f_n] + \tilde{\Gamma} (f_0^\dagger d + d^\dagger f_0) + \tilde{\epsilon}_0 d^\dagger d \right\} \quad (\text{A.18})$$

where $\tilde{\epsilon}_0 = 2/(1 + \Lambda^{-1})$ and $\tilde{\Gamma} = 2\Gamma[2/(1 + \Lambda^{-1})]^2/\pi D$. Note that the original Hamiltonian given in Eq. (A.17) is recovered by taking the limit

$$\frac{H}{D} = \lim_{N \rightarrow \infty} \frac{1 + \Lambda^{-1}}{2} \Lambda^{-(N-1)/2} H_N. \quad (\text{A.19})$$

The sequence of Hamiltonians can be solved in an iterative way by using the recursive relation

$$H_{N+1} = \sqrt{\Lambda} H_N + \xi_N (f_N^\dagger f_{N+1} + f_{N+1}^\dagger f_N), \quad (\text{A.20})$$

which relates the Hamiltonian H_{N+1} with the eigenvalues of the Hamiltonian in a previous step. By increasing the number of sites, the dimension of the Hamiltonian is also increased exponentially. Due to the computers memory limitation, the maximum number of sites that can be diagonalized exactly is rather small (~ 20 for the spinless model). In order to overcome this limitation, a truncation can be done over the states higher in energy, as depicted in the top right panel of Fig. A.2, since they are expected to provide smaller contributions to the ground state. After the truncation procedure two different kind of states appear: the kept states ($|k\rangle$), which are used in the next iteration, and the discarded ones ($|d\rangle$). Although the discarded states are not used in the subsequent NRG iterations, they contain valuable information about the high energy system properties, as it will be discussed below.

It is worth mentioning that by using the system symmetries the Hamiltonian can be divided into different subspaces. This is important, not only because it improves drastically the algorithm performance, but it also avoids possible artificial perturbations in

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Figure A.2: Scheme of the NRG iterative procedure. In the second panel on the top part the eigenenergies from the previous calculation are scaled by $\sqrt{\Lambda}$. The third panel describes the energy spectrum after adding a new site to the chain (as described in the lower part of the figure), where the number states is doubled. Finally, the eigenspace is truncated discarding the states with higher energies (dashed lines) and keeping the lower ones.

the NRG truncation procedure. For instance, in the spin degenerate Anderson model the truncation over the complete set of eigenstates of the Hamiltonian can lead to the appearance of an artificial magnetic field (for further details see the discussion in Ref. [9]). In the model of Eq. (A.1) the number of electrons is well defined, becoming a good quantum number. Thus, in the following the states are going to be labeled as $|Q, l\rangle_N$, where Q represents the number of electrons and N the number of sites. The states just before the diagonalization are going to be labeled as $|Q, m_0\rangle_N = |Q, m\rangle_{N-1} \otimes |0\rangle_N$ and $|Q, m_1\rangle_N = |Q-1, m\rangle_{N-1} \otimes |1\rangle_N$, where the subindex in the definition is used to denote the occupation of the last site. These states are related to the ones after the diagonalization through

$$|Q, l\rangle_N = \sum_{m_i} U(Q, l; Q-i, m_i) |Q-i, m_i\rangle_N, \quad (\text{A.21})$$

where U is the eigenvectors matrix. The eigenenergies of the state $|Q, l\rangle_N$ will be denoted in the following as $\epsilon_N^{Q, l}$. The final part of this section is devoted to explain the way the physical observables are calculated, focusing on the spectral function. Also, an

extension based on the use of the discarded states is introduced, to address the finite energy properties.

A.1.3 Spectral properties

Although the NRG algorithm has been shown to be powerful to obtain the position of the localized states in the system [10], it can also be used to compute the physical observables around the Fermi edge. It becomes specially precise close to the Fermi level, due to the logarithmic discretization. In this section I will concentrate on the determination of the equilibrium spectral density, although a more complete information can be obtained about the system by means of the NRG (some examples are described in Ref. [11]).

The calculation of the spectral functions involve the calculation of the retarded Green function of the product of two operators. For instance, considering two fermionic operators, A and B , the retarded Green function is defined as

$$G_{A,B}^r(t, t') = -i\theta(t - t') \sum_n \langle [A(t), B(t')]_+ \rangle, \quad (\text{A.22})$$

where the bracket represents the usual anti-commutation operation. For the localized spectral density, $A = d$ and $B = d^\dagger$, the spectral properties can be obtained through the Lehman representation (see the subsection 2.2.2 for further details) as

$$A(\epsilon) = \sum_{Q,l,l'} \text{Im} \left\{ \left[\frac{|{}_N \langle Q, l | d | Q+1, l' \rangle_N|^2}{\epsilon - (\epsilon_N^{Q+1,l'} - \epsilon_N^{Q,l}) + i\eta} + \frac{|{}_N \langle Q, l | d^\dagger | Q-1, l' \rangle_N|^2}{\epsilon + (\epsilon_N^{Q+1,l'} - \epsilon_N^{Q,l}) + i\eta} \right] \rho_l \right\}, \quad (\text{A.23})$$

being η an infinitesimium and the density matrix $\rho_l = \exp(-\epsilon_N^{Q,l}/T)/Z$, where Z is the normalization factor, $Z = \sum_l \exp(-\epsilon_N^{Q,l}/T)$. Note that the NRG method provides discrete states that have to be correctly broaden. It has been found that one of the most precise way of broaden the resonances is to use a combination of the Gaussian function

$$\delta(\epsilon - \epsilon_N) \rightarrow \frac{1}{2\pi} \frac{b}{(\epsilon - \epsilon_N)^2 + b^2}, \quad (\text{A.24})$$

close to the Fermi edge and a log-Gaussian for larger energy scales [12]

$$\delta(\epsilon - \epsilon_N) \rightarrow \frac{e^{-b^2/4}}{b \epsilon_N \sqrt{\pi}} \exp \left[-\frac{(\log \epsilon - \log \epsilon_N)^2}{b^2} \right]. \quad (\text{A.25})$$

The parameter b is an artificial broadening that has to be included, whose value is usually taken around $b \sim 0.5$.

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Similarly to the Hamiltonian sequence (A.18), the matrix elements from Eq. (A.23) can be obtained iteratively, similarly to the Hamiltonian described above, as

$${}_N \langle Q, l | d^\dagger | Q-1, l' \rangle_N = \sum_{m_i, m'_i} U_N^*(Q, l; Q-i, m_i) U_N(Q-1, l; Q-1-i, m'_i) \times {}_N \langle Q-i, m_i | d^\dagger | Q-1-i, m'_i \rangle_N. \quad (\text{A.26})$$

Similar expressions can be obtained for the other matrix element of Eq. (A.23).

A.1.4 Discarded states

The method described in the previous subsection is powerful to determine the equilibrium system properties around the Fermi edge. However, the higher energy properties away the Fermi level are determined with increasing error, since to keep them in the truncation procedure not many chain sites have to be considered (and thus the system ground state could be determined in a rather imprecise way). This limitation is due to the truncation method described above. The algorithm can be improved by keeping information about the discarded states as described in Refs. [5, 6]. In those references, the authors proposed a rather different way of understanding the NRG algorithm. Instead of adding a site to the chain at each step, they considered the complete chain from the beginning connecting one more site each step. Moreover, they showed that the discarded states, together with the states at the final iteration constitute a complete set of eigenstates of the system. Thus, the sum over the states of Eq. (A.23) has to be performed over the final states and the discarded ones in the previous iterations.

In this subsection I will consider the temperature to be sufficiently small so the density matrix from Eq. (A.23) is well approximated at the final iteration. It is worth mentioning that if this is not the case, the discarded states have to be considered in the density matrix as described in Ref. [13]. Thus, in this case two different kind of terms contribute to the spectral properties. The first one involves the calculation of the correlation functions at the final iterations $({}_N \langle Q, l | d^\dagger | Q-1, l' \rangle_N)$, which can be determined iteratively as described by Eq. (A.27). The second term involves the calculation of the correlation matrix between a final state and a discarded one in the previous iterations, which can be obtained as

$${}_N \langle Q, l | d^\dagger | Q', l'_{dis} \rangle_{N'} = \sum U_N^*(Q+1, l; Q+1-i, m_i^N) \cdots U_{N'+1}^*(Q'+i, m^{N'+1}; Q'+1, n_i) \times {}_N \langle Q'+1, n | d^\dagger | Q', l'_{dis} \rangle_{N'}. \quad (\text{A.27})$$

Figure A.3: Schematic representation of the calculation of the $_N \langle Q, l | d^\dagger | Q - 2, l'_{dis} \rangle_{N-3}$ term, where three different terms contributions to the sum of Eq. (A.27) are given by the different paths connecting the states and represented by the different colors. The horizontal line in each of the charge subspaces is used to visually divided the discarded states (upper part) and the kept ones (lower part)

Note that this calculation involves the product of $N - N'$ matrices where the sum is performed over all the possible paths connecting the final and the discarded states. This is illustrated for the $_N \langle Q + 1, l | d^\dagger | Q - 2, l'_{dis} \rangle_{N-3}$ term in Fig. A.3 where three different contributions are found (the three paths are represented with different colors). This extension of the algorithm requires to save information of all the possible correlation matrices involving kept and discarded states, and the kept part of the eigenvectors. Finally, it is worth mentioning that the energy of the discarded states is well approximated by their value just before the truncation procedure.

A.1.5 The z -average

In the final part of the section I will introduce the z -averaging, discussed originally in Ref. [14]. The idea consists on averaging over independent NRG calculations, whose discretization is shifted in energy by a quantity Λ^{-z} , as depicted in Fig. A.4. By changing the z value in the interval $[0, 1)$ (Note that $z = 0$ and $z = 1$ correspond to the same discretization of the band), the discrete states move in energy leading to result closer to the continuum limit when $z \rightarrow \infty$. Then, the physical properties can be obtained by

Figure A.4: Energy scheme for the shifted logarithmic discretization of the conduction band (red lines) proposed in Ref. [14].

averaging over the different discretizations. This idea has several advantages. The first one is that it provides results closer to the continuum limit. In this sense, the results are less dependent on the broadening kernel and the broadening parameter (if taken small enough) considered to compute the spectral properties. Moreover, by averaging over the N_z results obtained for the different choices of z , the unphysical oscillations appearing in the low temperature observables are damped and greater values of Λ can be taken, speeding up the calculation significantly [14].

In the shifted discretization of the band the wavefunctions are given by

$$\Psi_{nk}^{\pm}(\epsilon) = \begin{cases} A_{nk} e^{\pm i\omega_n} & \text{if } \Lambda^{-(n+z+1)} \leq |\epsilon| \leq \Lambda^{-(n+z)} \quad k \in \mathbb{Z}, \\ 0 & \text{Otherwise,} \end{cases} \quad (\text{A.28})$$

where by imposing the normalization and the orthogonality conditions it is possible to find the amplitude and the frequencies of the wavefunctions, given by

$$A_{nk} = \Lambda^{n/2} \sqrt{\frac{\Lambda^{z-1}}{1 - \Lambda^{-1}}}, \quad \omega_n = 2\pi \frac{\Lambda^{n+z-1}}{1 - \Lambda^{-1}}. \quad (\text{A.29})$$

These new wavefunctions induce a change in the prefactor term of the kinetic energy term, which can be written as

$$\frac{H_T}{D} = \frac{1 + \Lambda^{-1}}{2\Lambda^{z-1}} \sum_{n=0}^{\infty} \Lambda^{-n/2} \xi_n (f_n^{\dagger} f_{n+1} + f_{n+1}^{\dagger} f_n), \quad (\text{A.30})$$

where ξ_n is given by Eq. (A.16). With these definitions, the iterative procedure and the expressions introduced before can be also be applied to this situation.

A.2 Holstein model

In this section I discuss the NRG results applied to the spinless Anderson-Holstein situation. This problem has been discussed in the literature in the equilibrium [3, 4, 16]

Figure A.5: Spectral density of the NRG method. Results for the numerical developed during the thesis (solid lines) are compared to the ones from Ref. [15] in the equilibrium situation. Results for $\lambda/\omega_0 = 1.5$ (red line) and 2 (blue line) are shown. The inset illustrates the convergence to the FSR. The remaining are $\Gamma = \omega_0$, $\epsilon_0 = \lambda^2/\omega_0$, $\Lambda = 1.5$, $n_p^{max} = 200$, $N_z = 128$ and the maximum dimension per subspace is taken to be 10^3 .

and the non-equilibrium situation [15]. The Hamiltonian is given by

$$H = H_0 + H_p + H_{e-p}, \quad (\text{A.31})$$

where the non-interacting Hamiltonian is given by Eq. A.1, the Hamiltonian of the phonon is $H_p = \omega_0 a + a^\dagger$, ω_0 being the phonon frequency, and the electron-phonon coupling term $H_{e-p} = \lambda d^\dagger d(a + a^\dagger)$, with λ being the coupling strength. After discretizing the electronic reservoir, the Hamiltonian can be written as

$$\frac{H}{D} = H_0 + \frac{H_p}{D} + \frac{H_{e-p}}{D}, \quad (\text{A.32})$$

where the phonon terms are simply scaled with the bandwidth. Note that the electron-phonon coupling makes the Hilbert space at the first iteration infinite, since the vibrational mode can be excited in any state. However, for moderate coupling strengths, the

Figure A.6: Spectral density for the NRG calculation (solid lines) compared to the approximated DTA from Eq. (2.75). Results for $\lambda/\omega_0 = 0.5, 1$ and 1.5 are shown in the panels, from left to right. The remaining parameters are the same as in Fig. A.5 with $\Gamma = 0.2\omega_0$

space can be truncated to a finite n_p^{max} , since higher order excitations are highly energetic and unlikely. This makes the initial Hilbert space to be of dimension $2 \times (n_p^{max} + 1)$. For converging the algorithm, the maximum number of phonons needs to be much larger than the average number, since the problem requires from an accurate description of excitations involving a large number of phonons. I make the estimation that the system convergence is reached for $n_p^{max} \gtrsim 4n_p$ (being n_p the stationary phonon number). Moreover, the number of states per subspace has to be such that the high order phonon excitations are not discarded in the earliest iterations. A typical choice is to take the dimension of the kept subspace to be $\gtrsim 4n_p^{max}$. Finally, the energy cutoff in the band has to be taken as the largest energy scale and chosen to be $D = 30\omega_0$ in the following calculations.

In Fig. A.5 the spectral density is represented for $\lambda = 1.5$ (in red) and 2 (blue). The results from the algorithm developed during the thesis (solid lines) are compared to the ones from Ref. [15] (represented as symbols), finding a remarkable agreement. This constitutes a test for the method accuracy. I would like to comment that, although the method is exact, the smooth details may depend on the details of the broadening kernel,

Figure A.7: Position of the first phonon sideband for increasing λ and the situation represented in Figs. A.5 (red squares) and A.6 (blue circles).

which can lead to small quantitative differences as observed in the dips, mainly in the $\lambda = 2$ case. The inset of Fig. A.5 illustrates the convergence of the DOS to the FSR in the equilibrium electron-hole symmetric case. As shown, the agreement between both calculations at low frequencies is perfect, describing the exponential decrease of the central resonance as $\Gamma \rightarrow \Gamma e^{-\lambda^2/\omega_0^2}$ for strong electron-phonon interaction.

In Fig. A.6 the polaronic regime ($\Gamma \ll \omega_0, \lambda$) is analyzed. Results for $\lambda/\omega_0 = 0.5, 1$ and 1.5 are represented from left to right, showing the appearance of the higher order phonon sidebands. To the best of my knowledge, this regime has not been explored before in the literature by means of the NRG method. In this regime, the z -average [14] introduced in the section A.1.5 is essential to obtain reliable results, since the broadening kernels with the usual broadening parameter lead to an artificial smoothing of the spectral function. The numerical results (solid lines) are compared with the approximated DTA (see section 2.4.3 for more details), represented as dashed lines, showing a better agreement in the strong electron-phonon coupling regime. Importantly, the DTA describes accurately

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the exponential decrease of the resonance at the Fermi edge, fulfilling the situations the FSR in the equilibrium electron-hole symmetric situation.

As observed in Fig. A.6, the energy shift of the phonon sidebands is always to higher frequencies and maximal in the regime where $\lambda \sim \omega_0$. The position of the first resonance is represented in Fig. A.7 for the parameters of Fig. A.5 in red color and for the $\Gamma \ll \omega_0$ in blue color. The maximum of the curves are roughly located at $\lambda^2/2\omega_0\Gamma \sim 1$, which leads to $\lambda_{max} \sim \sqrt{2\Gamma\omega_0}$. Moreover, the displacement becomes maximal for the $\Gamma = \omega_0$ case where the electron and the phonon timescales are of the same order. In both situations represented in Fig. A.6, the shift of the first sideband tends to zero in the strong electron-phonon coupling regime.

Finally, it is worth mentioning that the regime $\omega_0 \ll \Gamma$, λ is challenging to be achieved through this NRG procedure described above. In this regime the low energy states become strongly degenerated, which may require the use of huge matrices to not discard important information in the NRG iterations. This problem could be overcome with the use of the coherent states of the harmonic oscillator. To the best of my knowledge this possibility remains unexplored in the case of the NRG method applied to the spinless Anderson-Holstein model, and remains as a possible future research line.

Bibliography

- [1] K. G. Wilson. The renormalization group: Critical phenomena and the Kondo problem. *Rev. Mod. Phys.* **47**, 773 (1975).
- [2] H. R. Krishna-murthy, J. W. Wilkins, and K. G. Wilson. Renormalization-group approach to the Anderson model of dilute magnetic alloys. i. static properties for the symmetric case. *Phys. Rev. B* **21**, 1003 (1980).
- [3] D. Meyer, A. C. Hewson, and R. Bulla. Gap formation and soft phonon mode in the Holstein model. *Phys. Rev. Lett.* **89**, 196401 (2002).
- [4] A. C. Hewson and D. Meyer. Numerical renormalization group study of the Anderson-Holstein impurity model. *Journal of Physics: Condensed Matter* **14**, 427 (2002).
- [5] F. B. Anders and A. Schiller. Real-time dynamics in quantum-impurity systems: a

- time-dependent numerical renormalization-group approach. *Phys. Rev. Lett.* **95**, 196801 (2005).
- [6] F. B. Anders and A. Schiller. Spin precession and real-time dynamics in the Kondo model: time-dependent numerical renormalization-group study. *Phys. Rev. B* **74**, 245113 (2006).
 - [7] M. Pletyukhov, D. Schuricht, and H. Schoeller. Relaxation versus decoherence: Spin and current dynamics in the anisotropic Kondo model at finite bias and magnetic field. *Phys. Rev. Lett.* **104**, 106801 (2010).
 - [8] R. Bulla, T. Pruschke, and A. C. Hewson. Anderson impurity in pseudo-gap Fermi systems. *Journal of Physics: Condensed Matter* **9**, 10463 (1997).
 - [9] M. Sindel. Numerical renormalization group studies of quantum impurity models in the strong coupling limit. Doctoral thesis (2004).
 - [10] A. Martín-Rodero and A. L. Yeyati. The Andreev states of a superconducting quantum dot: mean field versus exact numerical results. *Journal of Physics: Condensed Matter* **24**, 385303 (2012).
 - [11] R. Bulla, T. A. Costi, and T. Pruschke. Numerical renormalization group method for quantum impurity systems. *Rev. Mod. Phys.* **80**, 395 (2008).
 - [12] R. Bulla, T. A. Costi, and D. Vollhardt. Finite-temperature numerical renormalization group study of the Mott transition. *Phys. Rev. B* **64**, 045103 (2001).
 - [13] A. Weichselbaum and J. von Delft. Sum-rule conserving spectral functions from the numerical renormalization group. *Phys. Rev. Lett.* **99**, 076402 (2007).
 - [14] W. C. Oliveira and L. N. Oliveira. Generalized numerical renormalization-group method to calculate the thermodynamical properties of impurities in metals. *Phys. Rev. B* **49**, 11986 (1994).
 - [15] A. Jovchev and F. B. Anders. Influence of vibrational modes on quantum transport through a nanodevice. *Phys. Rev. B* **87**, 195112 (2013).
 - [16] G. S. Jeon, T.-H. Park, and H.-Y. Choi. Numerical renormalization-group study of the symmetric Anderson-Holstein model: Phonon and electron spectral functions. *Phys. Rev. B* **68**, 045106 (2003).

In this appendix the theory of Toeplitz matrices is introduced (for a review see [1]). This theory allows for an analytic treatment of matrices whose entries depend only on the difference between the row and column indices. The Toeplitz matrices appear when studying a large number of physical systems such as electromagnetic waves [2] or spins in a lattice through the Ising model [3, 4]. Analytic expressions are developed to describe the asymptotic regime, where the matrix dimension tends to infinite. This regime is important from the physical point of view since it corresponds to the continuous time limit.

On the other hand, the block Toeplitz matrices are much less understood since its general asymptotic expressions are generally unknown. These matrix are found for example when studying waves at the interfaces of two media. In this appendix I will relate the time resolved transport statistics to the calculation of the determinant of a block Toeplitz matrix, whose leading contribution at large times recovers the Levitov result [5]. The transient regime is also explored by means of an ansatz based on the known Toeplitz results.

A.3 Basic mathematical theory

In this first section of the chapter I make a brief review of the mathematical formalism for treating the Toeplitz matrices (for a review see Ref. [1]). In the next sections the connection between the Toeplitz theory and the expressions for computing the transport properties is done. A Toeplitz matrix is a matrix whose entries only depend on the difference between the row and column indices, i.e.

$$T_n = \begin{bmatrix} t_0 & t_{-1} & t_{-2} & \cdots & t_{1-n} \\ t_1 & t_0 & t_{-1} & \cdots & t_{2-n} \\ t_2 & t_1 & t_0 & \cdots & \vdots \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ t_{n-1} & t_{n-2} & t_{n-3} & \cdots & t_0 \end{bmatrix}_{n \times n}, \quad (\text{A.33})$$

where $\{t_k\}$ are complex numbers in general. I am interested on the asymptotic properties of the matrices which is equivalent to take an infinite number of discretization intervals. When the sequence of matrix entries is absolutely summable ($\sum_{k=-\infty}^{\infty} |t_k| < \infty$), i.e. it is in the Wiener class, the matrix entries can be understood as the coefficients of the discrete Fourier transform of a continuous variable f , also known as symbol of the matrix

$$f(\phi) = \sum_{k=-\infty}^{\infty} t_k e^{ik\phi}, \quad (\text{A.34})$$

where f is defined in the $\phi \in [0, 2\pi]$ interval. Note that the function is real if and only if the matrix is Hermitian ($t_k^* = t_{-k}$). A usual notation for the matrix from Eq. (A.33) is $T_n(f)$, which can be understood as a sequence of matrices of dimensions $n \times n$ and indicates that its entries are obtained through the discrete Fourier transform of the function f .

These matrices have very interesting asymptotic properties ($n \rightarrow \infty$) that will be reviewed in the following. The asymptotic inverse of a Toeplitz matrix can be obtained as

$$T_n(f) \sim T_n(1/f). \quad (\text{A.35})$$

Moreover, the sequence of products of j Toeplitz matrices is asymptotically equivalent to the Toeplitz matrix of the symbol product, i.e

$$\prod_{i=1}^j T_n(f_i) \sim T_n\left(\prod_{i=1}^j f_i\right). \quad (\text{A.36})$$

Note that this property is equivalent to the convolution theorem for the symbols. Probably the most important result in relation to this thesis topic are the so-called Szegő theorems, which determines the asymptotic of the matrix determinant. In order to simplify the formulation of these theorems, it becomes useful to introduce the notation

$$[\log(f)]_k \equiv \frac{1}{2\pi} \int_0^{2\pi} d\phi \log[f(\phi)] e^{-ik\phi}. \quad (\text{A.37})$$

The weak Szegő theorem [6] describes the asymptotic value of the determinant of a Toeplitz matrix which, under some conditions on the smoothness of the function, is described by

$$\lim_{n \rightarrow \infty} \frac{\det[T_n(f)]}{\det[T_{n-1}(f)]} = \exp\{[f(\phi)]_0\}. \quad (\text{A.38})$$

This equation implies that the asymptotic limit of the determinant is given by

$$\lim_{n \rightarrow \infty} \det[T_n(f)] = \exp\{n[f(\phi)]_0\}. \quad (\text{A.39})$$

A more complete result was derived in Ref. [7] where Szegő analyzed the subleading term of the determinant, finding

$$\lim_{n \rightarrow \infty} \frac{\det[T_n(f)]}{\exp\{n[f(\phi)]_0\}} = E(f), \quad (\text{A.40})$$

where

$$E(f) = \sum_{k=1}^{\infty} k [f(\phi)]_k [f(\phi)]_{-k}. \quad (\text{A.41})$$

Eq. (A.40) is also known as the strong Szegő theorem, and provides the subdominant contribution to the main one, given in Eq. (A.38). Thus, the complete asymptotic expression for the determinant of a Toeplitz matrix is

$$\lim_{n \rightarrow \infty} [T_n(f)] = E(f) \exp \{n [f(\phi)]_0\} . \quad (\text{A.42})$$

This expression is only valid if the symbol f is smooth enough (see Ref. [1] for more precise mathematical conditions). For studying singular symbols, exhibiting for instance a discontinuous behavior, the Fisher-Hartwig conjecture [8] could be used. This result has been used in Ref. [9] to analyze the time evolution of Luttinger liquids after rectangular pulses.

A.3.1 Block Toeplitz matrices

After introducing the basic results for the Toeplitz matrices, it is interesting to generalize the results to the block situation. A block Toeplitz matrix can be written as

$$D_c(\{f_{i,j}\}) = \begin{bmatrix} T_n(f_{11}) & T_n(f_{12}) & \cdots & T_n(f_{1c}) \\ T_n(f_{21}) & T_n(f_{22}) & \cdots & T_n(f_{2c}) \\ \vdots & \vdots & \ddots & \vdots \\ T_n(f_{c1}) & T_n(f_{c2}) & \cdots & T_n(f_{cc}) \end{bmatrix}_{n c \times n c} , \quad (\text{A.43})$$

where each block has the form of Eq. (A.33). The determinant of the block Toeplitz matrices is in general unknown and the most complete result was provided by H. Widom in Ref. [10]. In that reference the author stated that the determinant of a block Toeplitz matrix can be written in a rather similar way as for the scalar case

$$\lim_{n \rightarrow \infty} [D_c(\hat{f})] \sim G^{n+1}[\hat{f}] E[\hat{f}] , \quad (\text{A.44})$$

where the $\hat{}$ symbol is used to indicate the matrix internal structure. Similarly to the scalar case, G is the dominant term given by

$$G^{n+1}[\hat{f}] = \exp \{ (n+1) \log [\det(\hat{f})] \} , \quad (\text{A.45})$$

where the determinant is taken on the matrix of the symbols. The dominant term can be simplified when considering the set of eigenvalues $\{\lambda_j(\phi)\}$ of the $\hat{f}$ matrix in the continuous notation, finding

$$G[\hat{f}] = \exp \left\{ \sum_{j=1}^c \log [\lambda_j(\phi)]_0 \right\} . \quad (\text{A.46})$$

The second term of Eq. (A.44) describes the subleading term, given by

$$E[\hat{f}] = \det \left[D_c(\hat{f}) D_c(\hat{f}^{-1}) \right]. \quad (\text{A.47})$$

Although the previous equation is elegant, it becomes challenging to be evaluated since it involves the product of two huge matrices (operation whose computational cost is similar to a direct calculation of the determinant itself). There exist only few block Toeplitz matrices whose subleading term (E) is analytic [11]. For instance, the Ising model with first neighbors interactions can be reduced to the scalar situation via a Wiener-Hopf factorization [3, 4]. There are only two kind of block Toeplitz matrices, not reducible to the scalar case whose expression for the determinant is known: the one described in Ref. [12] and used to study the dimer model, and the one for the von-Neumann entropy given in Ref. [13]. Another alternative to compute the determinant of the block Toeplitz matrix consists on the use of approximations. For instance, in Ref. [2] the author uses Padé approximants to obtain the matrix determinant with a good accuracy.

In order to obtain an approximation for the subleading term of Eq. (A.44), I reformulate the scalar solution in terms of the matrix eigenvalues ($\lambda_j(\phi)$) as

$$E[\hat{f}] \approx \sum_{j=1}^c \sum_{k=1}^{\infty} k [\log \lambda_j(\phi)]_k [\log \lambda_j(\phi)]_{-k}. \quad (\text{A.48})$$

Although this naive approximation is not expected to be valid in general, its result goes in the correct direction, as it will be commented below.

A.4 Transport properties

In this section of the chapter, the connection between the Toeplitz theory and the FCS analysis is established. I will focus on the non-interacting situation, whose analytic solution is still unknown. The starting point of the section is the GF of a system coupled to metallic electrodes at the initial time, which is given by the Fredholm determinant as [14]

$$Z(\chi, t) = \det \left[\hat{\mathbf{G}}_{S,\chi}^{-1} \hat{\mathbf{G}}_{S,\chi=0} \right] = \frac{\det \left[\hat{\mathbf{g}}_S^{-1} - \hat{\Sigma}_{T,\chi} \right]}{\det \left[\hat{\mathbf{g}}_S^{-1} - \hat{\Sigma}_{T,\chi=0} \right]}, \quad (\text{A.49})$$

where the expressions for the system Green function and self-energy are described in the chapter 3, as well as the numerical solution to the problem. The inverse system Green

Figure A.8: Evolution of the probability distribution of a non-interacting nanojunction coupled to normal metal electrodes. The result from the Szegő theorem (red solid lines) are compared to the numerical evaluation of Z_T (blue dashed lines). In the top panel Only the main contribution from the Szegő theorem for the block Toeplitz matrices (A.45) is considered, while in the lower one the approximation of Eq. (A.47) for the subdominant part is also included. The parameters are $\epsilon_0 = 0$ and $V = 2\Gamma$.

function can be factorized into a block Toeplitz and a non-Toeplitz contributions as

$$i\mathbf{g}_{S,T}^{-1} = \left[\begin{array}{ccc|ccc} 1 & & & & & \\ h^- & 1 & & & & \\ & h^- & 1 & & & \\ \hline & & 0 & 1 & & \\ & & & h^+ & 1 & \\ & & & & h^+ & 1 \end{array} \right], \quad i\delta\mathbf{g}_S^{-1} = \left[\begin{array}{c|c} & \rho \\ \hline & i \end{array} \right], \quad (\text{A.50})$$

being $h_{\pm} = 1 \mp i\epsilon_0\Delta t$ and the inverse system Green function is given by the sum of these two contributions. Then, the GF can be factorized as

$$Z(\chi, t) = \frac{\det[\hat{\mathbf{g}}_{S,T}^{-1} - \hat{\Sigma}_{T,\chi}]}{\det[\hat{\mathbf{g}}_{S,T}^{-1} - \Sigma_{T,\chi=0}]} \frac{\det[\hat{\mathbb{I}} + \delta\hat{\mathbf{g}}_S^{-1}\hat{\mathbf{G}}_{S,\chi}]}{\det[\hat{\mathbb{I}} + \delta\hat{\mathbf{g}}_S^{-1}\hat{\mathbf{G}}_{S,\chi=0}]} \equiv Z_T(\chi, t)Z_{NT}(\chi, t), \quad (\text{A.51})$$

where $\hat{\mathbf{G}}_{S,\chi} = [\hat{\mathbf{g}}_{S,T}^{-1} - \hat{\Sigma}_{T,\chi}]^{-1}$. The first term, (Z_T) , describes the Toeplitz contribution, while Z_{NT} contains information about the connection and the closing of the Keldysh contour (see Fig. 3.1). Note that the no Toeplitz part only contain terms, which depend on the inverse of the Toeplitz part and is subdominant at long times. In this section, I will focus only on the Toeplitz part which constitutes the leading term of the determinant. It is worth remarking that the dominant contribution of the Toeplitz part, described by Eq. (A.45), correctly describes (after some manipulation) the asymptotic limit given by the Levitov formula (2.104) [5]. The $(n+1)$ prefactor in the exponential is converted to the final time, when transforming the integral from the $\phi \in [0, 2\pi]$ variable to ω .

Although the initial condition and the contour closing terms are not considered in the Toeplitz part, there is a transient related to the system connection. The analytic results using the Szegő theorem for the block Toeplitz matrices and the numerical one are shown in the Fig. A.8. In the top panel only the contribution from the dominant term (A.45) is represented. As observed, there is some disagreement between the results at short times, coinciding for $t \gg 1/\Gamma$ where the Levitov result is recovered and the subleading term becomes negligible. Moreover, at times $t \sim 1/\Gamma$, the probabilities computed using the weak Szegő theorem exhibit small negative parts, which is a pathological feature. This pathology is partially cured through the ansatz given in Eq. (A.48) for the subleading term, plotted in the lower panel of the Fig. A.8. This illustrates the importance of correctly describing the subleading contribution in the transient regime.

As a final comment, I would like to remark that the subleading terms becomes specially important for superconducting nanojunctions. As described in the chapter 7, the system connection can lead to a non-equilibrium population of the subgap states that do not relax on time. This means that the importance of the subleading term in superconducting nanojunctions is not necessarily reduced with increasing time.

Bibliography

- [1] R. M. Gray. Toeplitz and circulant matrices: A review. *Foundations and Trends in Communications and Information Theory* **2**, 155 (2006).
- [2] I. D. Abrahams. On the solution of Wiener–Hopf problems involving noncommutative matrix kernel decompositions. *SIAM Journal on Applied Mathematics* **57**, 541 (1997).
- [3] B. M. McCoy and T. T. Wu. Theory of Toeplitz determinants and the spin correlations of the two-dimensional Ising model. ii. *Phys. Rev.* **155**, 438 (1967).
- [4] K. Tanaka, T. Morita, and K. Hiroike. Spin pair correlation function of the Ising model on the brickwork lattice with second neighbor interactions. *Physica A: Statistical Mechanics and its Applications* **171**, 350 (1991).
- [5] L. S. Levitov and G. B. Lesovik. Charge distribution in quantum shot noise. *JETP Lett.* **58**, 230 (1993).
- [6] G. Szegő. Ein grenzwertsatz über die Toeplitzschen determinanten einer reellen positiven funktion. *Mathematische Annalen* **76**, 490 (1915).
- [7] G. Szegő. On certain Hermitian forms associated with the Fourier series of a positive function. *Comm. Sémin. Math. Univ. Lund* 228 (1952).
- [8] M. E. Fisher and R. E. Hartwig. *Toeplitz Determinants: some Applications, Theorems, and Conjectures*, 333–353 (Wiley-Blackwell, 2007).
- [9] I. V. Protopopov, D. B. Gutman, and A. D. Mirlin. Luttinger liquids with multiple Fermi edges: Generalized Fisher-Hartwig conjecture and numerical analysis of Toeplitz determinants **52**, 165 (2012).
- [10] H. Widom. Asymptotic behavior of block Toeplitz matrices and determinants. ii. *Advances in Mathematics* **21**, 1 (1976).
- [11] P. Deift, A. Its, and I. Krasovsky. Toeplitz matrices and Toeplitz determinants under the impetus of the Ising model: Some history and some recent results. *Communications on Pure and Applied Mathematics* **66**, 1360 (2013).
- [12] E. L. Basor and T. Ehrhardt. Asymptotics of block Toeplitz determinants and the classical dimer model. *Communications in Mathematical Physics* **274**, 427 (2007).
- [13] B.-Q. Jin and V. E. Korepin. Quantum spin chain, Toeplitz determinants and the

- Fisher-Hartwig conjecture. *Journal of Statistical Physics* **116**, 79 (2004).
- [14] M. Esposito, U. Harbola, and S. Mukamel. Nonequilibrium fluctuations, fluctuation theorems, and counting statistics in quantum systems. *Rev. Mod. Phys.* **81**, 1665 (2009).

Inverse free boson propagator

In this appendix the problem of obtaining the inverse of the free phonon propagator along the discretized Keldysh contour is discussed in detail. A similar problem has already been discussed in Ref. [1] where the author considers the problem of bosonic particles on a single level, whose Hamiltonian is given by

$$H_{ph,0} = \omega_0 b^\dagger b, \quad (\text{B.1})$$

being ω_0 the phonon energy. The free phonon propagator of the mode described by the Hamiltonian (B.1) is defined as $\hat{d}_0(t, t') = -i \langle T_c b(t) b^\dagger(t') \rangle$, whose inverse can be obtained in a similar way as the electronic propagator [1], finding

$$i\mathbf{d}_0^{-1} = \left(\begin{array}{ccc|ccc} -1 & & & & & \\ h_- & -1 & & & & \\ & h_- & -1 & & & \\ & & \ddots & \ddots & & \\ & & & 1 & & \\ \hline & & & & -1 & \\ & & & & h_+ & -1 \\ & & & & & \ddots & \ddots \\ & & & & & & h_+ & -1 \end{array} \right)_{2N \times 2N}, \quad (\text{B.2})$$

with $h_\pm = 1 \pm i\Delta t\omega_0$. Note that the expression of Eq. (B.2) is equivalent to discretize the operator $i\partial_t - \omega_0$ on the time contour with an initial condition given by $\rho(\omega_0) = n_p(0)/[1 + n_p(0)]$, which depends on the initial phonon population $n_p(0)$.

To the best of my knowledge, the case of the inverse of the inverse of the propagator $\hat{d}(t, t') = -i \langle \hat{T}_c [\hat{\varphi}(t) \hat{\varphi}^\dagger(t')] \rangle$ (with $\hat{\varphi} = b + b^\dagger$) has not been discussed in the literature. This inverse becomes mathematically more demanding since it requires the discretization of the second order differential operator

$$H_{ph} = \frac{p^2}{2} + \frac{\omega_0^2}{2} x^2, \quad (\text{B.3})$$

B. Inverse free boson propagator

with $p = -i\partial_x$ and $x = \sqrt{1/2\omega_0}\hat{\phi}$. Moreover, it is worth remarking that the discretized version of the free phonon propagator given in Eq. (2.55) is singular and cannot be inverted. In this appendix, the way of regularizing this singular behavior is also discussed. From the definition given in Ref. [1], the system partition function is given by

$$Z = \frac{\text{Tr}[\mathcal{U}_c \rho]}{\text{Tr}[\rho]}, \quad (\text{B.4})$$

where $\mathcal{U}_c = \mathcal{U}^+(t_{2N}, t_{N+1})\mathcal{U}^-(t_N, t_1)$ is the evolution operator on the Keldysh contour and $\rho = e^{-H/T}$ the initial density matrix. Expanding Z in coordinate space and for $N = 3$ it is possible to find

$$\begin{aligned} \text{Tr}[\mathcal{U}_c \rho] = \int dx_1 \cdots dx_6 & \langle x_6 | \mathcal{U}_{-\Delta t} | x_5 \rangle \langle x_5 | \mathcal{U}_{-\Delta t} | x_4 \rangle \langle x_4 | \mathbb{I} | x_3 \rangle \\ & \langle x_3 | \mathcal{U}_{\Delta t} | x_2 \rangle \langle x_2 | \mathcal{U}_{\Delta t} | x_1 \rangle \langle x_1 | \rho | x_6 \rangle \end{aligned} \quad (\text{B.5})$$

being $\mathcal{U}_{\Delta t} = e^{-iH_{ph}\Delta t}$. Note that the last term in the top part of Eq. (B.5) represents the branch changing in the Keldysh contour while the last one in the lower part the contour closing at the final time. The matrix components can be determined by means of the so-called Mehler kernel [2]

$$\langle x | e^{-iH_{ph}t} | y \rangle = \frac{\exp\{i[(x^2 + y^2)\cos(\omega_0 t) - 2xy]/2\sin(\omega_0 t)\}}{\sqrt{2\pi i \sin(\omega_0 t)}}. \quad (\text{B.6})$$

Discretizing the previous expression with a time step, I find

$$\langle x | e^{\mp iH_{ph}\Delta t} | y \rangle = \frac{\exp\{\pm i[(x^2 + y^2)(1 - \delta^2/2) - 2xy]/2\delta\}}{\sqrt{2\pi i \delta}} \quad (\text{B.7})$$

with $\delta = \omega_0 \Delta t$. Similarly, for the Keldysh contour closing term

$$\langle x | \rho | y \rangle = \sqrt{\frac{\rho_0}{\pi(1 - \rho_0^2)}} \exp\left[-\frac{(1 + \rho_0^2)(x^2 + y^2)}{2(1 - \rho_0^2)} + \frac{2xy\rho_0}{1 - \rho_0^2}\right], \quad (\text{B.8})$$

where $\rho_0 = n_p(0)/[n_p(0) + 1]$ contains information about the initial phonon population, $n_p(0)$. The final step for obtaining the inverse is to regularize the delta function, given by the term

$$\langle x | \mathbb{I} | y \rangle \approx \frac{1}{\sqrt{2\pi\eta}} e^{-(x-y)^2/2\eta}, \quad (\text{B.9})$$

where η is an infinitesimium. Finally, the inverse of the free phonon propagator can be obtained identifying the components of

$$Z = \int dx_1 \cdots dx_{2N} e^{ix^T d^{-1}x}, \quad (\text{B.10})$$

finding the expression

$$\mathbf{d}^{-1} = \frac{1}{2\delta} \left(\begin{array}{cccc|cccc} h_0^+ & -1 & & & & & & h_{0N} \\ -1 & h & -1 & & & & & \\ & \ddots & \ddots & \ddots & & & & \\ & & -1 & h & -1 & & & \\ & & & -1 & h_N^+ & c & & \\ & & & & c & h_N^- & 1 & \\ & & & & & 1 & -h & 1 \\ & & & & & & \ddots & \ddots & \ddots \\ & & & & & & & 1 & -h & 1 \\ & & & & & & & & 1 & h_0^- \end{array} \right)_{2N \times 2N}, \quad (\text{B.11})$$

where $h = 2(1 - \delta^2/2)$. The information about the initial phonon number is encoded in the components at the initial time $h_0^\pm = \pm h/2 + i\delta(1 + \rho_0^2)/(1 - \rho_0^2)$ and $h_{0N} = -2i\delta\rho_0/(1 - \rho_0^2)$. Finally, the regularization terms appear in the branch changing point, $c = -2i\delta/\eta$ and $h_N^\pm = \pm h/2 - c$. Note that all the prefactors appearing in the Mehler kernel expression only normalize the partition function, having no influence on the phonon dynamics. The particular case of $N = 2$ can be obtained straightforwardly following the procedure described above, finding

$$i\mathbf{d}_{N=2}^{-1} = \left(\begin{array}{cccc} 1 - \frac{\delta^2}{2} + i\delta\frac{1+\rho_0^2}{1-\rho_0^2} & -1 & 0 & -2i\frac{\delta\rho_0}{1-\rho_0^2} \\ -1 & 1 - \frac{\delta^2}{2} + 2i\frac{\delta}{\eta} & -2i\frac{\delta}{\eta} & 0 \\ 0 & -2i\frac{\delta}{\eta} & -1 + \frac{\delta^2}{2} + 2i\frac{\delta}{\eta} & 1 \\ -2i\frac{\delta\rho_0}{1-\rho_0^2} & 0 & 1 & -1 + \frac{\delta^2}{2} + i\delta\frac{1+\rho_0^2}{1-\rho_0^2} \end{array} \right). \quad (\text{B.12})$$

Bibliography

- [1] A. Kamenev. *Field Theory of Non-Equilibrium Systems* (Cambridge University Press, 2011).
- [2] A. Erdélyi. *Higher Transcendental Functions*. volume 2, Bateman Project, Higher Transcendental Functions (McGraw-Hill, New York, 1953).

Interpretation in terms of rate equations

In this appendix I introduce a simplified rate equation which can be used to compute the many-body probabilities $P_{\pm}(t)$ and $P_{odd}(t) = P_{odd,\uparrow}(t) + P_{odd,\downarrow}(t)$ of a superconducting nanojunction after a quench of the tunneling rates. The rate equation is depicted in the right panel of Fig. C.1 and given by

$$\begin{aligned}\frac{dP_-}{dt} &= -2\Gamma_o(t)P_- + \Gamma_-(t)P_o \\ \frac{dP_o}{dt} &= 2\Gamma_o(t)P_- - [\Gamma_-(t) + \Gamma_+(t)]P_o + 2\Gamma_o(t)P_+ \\ \frac{dP_+}{dt} &= \Gamma_+(t)P_o - 2\Gamma_o(t)P_+, \end{aligned} \tag{C.1}$$

where $\Gamma_{odd}(t)$ and $\Gamma_{\pm}(t)$ are the time-dependent rates for transitions between states and $P_-(t) + P_+(t) + P_{odd}(t) = 1$. In the QPC regime the long time population of the many-body states become universal and only dependent on the ABSs asymptotic population (see the discussion of the section 7.3). In this regime, the rates can be approximated by means of the simple perturbation theory, scaling as their energy distance to the lower gaps edge (see left panel of Fig. C.1). Thus, $\Gamma_{odd} \approx \Gamma^2/\Delta$ and $\Gamma_{\pm} \approx \Gamma^2/(\Delta \pm \epsilon_A)$.

The corresponding results are represented in the left panel of Fig. 7.4 compared to the numerical results. As shown, the rate equations from Eq. (C.1) provide a quantitative description of the $\epsilon_A \rightarrow 0$ limit, where $P_{odd} = 0.5$ and $P_{\pm} = 0.25$. On the other hand, they describe qualitatively the behavior for increasing ϵ_A where the poisoning probability is reduced. The disagreement at finite ϵ_A values may be due to the fact that the ABSs are not only charged from the states close to the gap, but lower energy states may also contribute.

The situation in the QD regime becomes richer since the asymptotic many-body prob-

C. Interpretation in terms of rate equations

Figure C.1: Left panel: stationary energy spectrum. The arrows in the figure indicate the approximated rates explaining the universal many-body populations in the QPC regime (see discussion on the section 7.3). in the right panel the simplified rate equation is represented.

abilities depend on the initial configuration (see the right panel of Fig. 7.4). In this regime, the rates are explicitly time dependent since the system relaxation can occur until the subgap states are developed, when the dynamics become blocked. However, the way the rates decay on time cannot be inferred from the numerical results and has to be left as a free parameter, which may lead to a non representative fit of the results (too many free parameters for fitting few independent variables).

Bidirectional Poisson distribution

In this appendix I show the calculation details for the bidirectional Poisson distribution, which is also known in the literature as birth-death distribution [1]. Starting from an initial population n , there is a birth rate (b) and a death rate (d), connecting the subspaces with different populations (see Fig. D.1). The distribution is considered to be Markovian, thus the probability of a birth or a death is independent from the previous history. In this case, the population describes the number of electrons transferred through the junction (the initial population is take as $n = 0$), where the sing of the population determines the transport direction. The calculation of the population is done in the following way. I begin with an initial probability distribution represented by the vector $P(0) = (\dots, 0, 1, 0, \dots)^T$. The differential system of equations can be solved by discretizing on time and using the recursive relation $P(t + dt) = M \cdot P(t)$, where

$$M = \begin{pmatrix} \ddots & \ddots & & & \\ \ddots & a & b dt & & \\ & d dt & a & \ddots & \\ & & \ddots & \ddots & \end{pmatrix}, \quad (\text{D.1})$$

and $a = 1 - (b + d)dt$ is the probability of staying at the same subspace after dt . The matrix space is truncated to high dimensions to avoid the border effects. In the equilibrium situation and at short times the birth and death rates become equal (there is no preferable direction for the transport), and the dynamics is characterized by a single parameter $n_{\rightarrow}(t) = b dt = d dt$ describing the net charge transfer in one of the directions.

The results for a superconducting nanojunction is represented in the lower panel of Fig. 7.2. By analyzing the time where the bidirectional Poisson distribution deviates from the numerical result, the ABS formation can be determined, finding that they are developed for $n_{\rightarrow}(t) \sim 3$. Thus, the number of electrons crossing the junction needed necessary for

Figure D.1: Scheme of the birth-death process. The birth process (which has a probability b), allows to go from n to $n + 1$ charges transferred through the junction. In the short time limit, we consider $b = d$, due to the absence of bias voltage.

the ABS to be formed is surprisingly small (they are of the order of 3 electrons crossing the junction in both directions of the junction).

Bibliography

- [1] C. Gardiner. *Stochastic Methods: A Handbook for the Natural and Social Sciences*. Springer Series in Synergetics (Springer Berlin Heidelberg, 2009).

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List of publications

Publications related to the content of this Thesis:

1. *Build-up of vibron-mediated electronic correlations in molecular electronics* Rémi Avriller, **Rubén Seoane Souto**, Álvaro Martín Rodero and Alfredo Levy Yeyati. In preparation.
2. *Transient dynamics in interacting nanojunctions within self-consistent perturbation theory* **Rubén Seoane Souto**, Rémi Avriller, Alfredo Levy Yeyati and Álvaro Martín Rodero. New J. Phys. **20**, 083039 (2018).
3. *Quench dynamics in superconducting nanojunctions: metastability and dynamical Yang-Lee zeros* **Rubén Seoane Souto**, Álvaro Martín Rodero and Alfredo Levy Yeyati. Physical Review B, **96** (16), 165444 (2017)
4. *Analysis of universality in transient dynamics of coherent electronic transport* **Rubén Seoane Souto**, Álvaro Martín Rodero and Alfredo Levy Yeyati. Fortschritte der Physik, **65** 1600062 (2017).
5. *Andreev bound states formation and quasiparticle trapping in quench dynamics revealed by time-dependent counting statistics* **Rubén Seoane Souto**, Álvaro Martín Rodero and Alfredo Levy Yeyati. Physical Review Letters, **117** (26), 267701 (2016).
6. *Transient dynamics and waiting time distribution of molecular junctions in the polaronic regime* **Rubén Seoane Souto**, Rémi Avriller, Rosa Carmina Monreal, Álvaro Martín Rodero and Alfredo Levy Yeyati. Physical Review B, **92** (12), 125435 (2015).
7. *Dressed tunneling approximation for electronic transport through molecular transistors* **Rubén Seoane Souto**, Alfredo Levy Yeyati, Álvaro Martín Rodero and Rosa Carmina Monreal. Physical Review B, **88** (8), 085412 (2014).

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This thesis is devoted to the analysis of the electronic transport properties through nanoscopic junctions. It is focused on the time response of the nanojunctions, analyzing not only the mean current but also its fluctuations, which contain valuable information on the underlying microscopic processes. In particular, the thesis analyzes in detail the coherent regime, where the quantum nature of the electrons manifests itself more clearly. This study can be of relevance in connection to the increasing demand of fabricating smaller and faster devices. One of the aims of this thesis is to analyze the effects of electron interactions and superconducting correlations in the time evolution of the system. Special attention is paid to the system relaxation to the expected steady state and the possible metastable and bistable behaviors.