Gerhard Dorn, BSc BSc

A Born-Markov master equation approach to correlated quantum systems in non-equilibrium

MASTER'S THESIS

to achieve the university degree of
Diplom-Ingenieur

Master's degree programme: Technische Physik

Technische Universität Graz

Supervisor:

Univ.-Prof. Dipl.-Phys. Dr.rer.nat. Wolfgang von der Linden

Institute of Theoretical and Computational Physics

Graz, May 2015

Deutsche Fassung:
Beschluss der Curricula-Kommission für Bachelor-, Master- und Diplomstudien vom 10.11.2008
Genehmigung des Senates am 1.12.2008

EIDESSTATTLICHE ERKLÄRUNG

Ich erkläre an Eides statt, dass ich die vorliegende Arbeit selbstständig verfasst, andere als die angegebenen Quellen/Hilfsmittel nicht benutzt, und die den benutzten Quellen wörtlich und inhaltlich entnommenen Stellen als solche kenntlich gemacht habe.

Graz, am …… ……..
(Unterschrift)

Englische Fassung:

STATUTORY DECLARATION

I declare that I have authored this thesis independently, that I have not used other than the declared sources / resources, and that I have explicitly marked all material which has been quoted either literally or by content from the used sources.

…… ……..
date (signature)

To my family

Abstract

The derivation of the Born-Markov master equation apt to describe open dissipative quantum systems in non-equilibrium is depicted and compared with the Lindblad formalism. Further it's extensions to quasi-degenerate states and it' evaluation in the system eigenbasis is given for the steady state in the wide band limit.

In the second part the derivation of the self-consistent Born approach an improvement of the before discussed master equation is presented and explicit formulas for the calculation of the steady state and steady state current in a single quantum dot (Anderson Impurity) with spin dependent coupling is given.

In the third part these two methods are numerically evaluated for the single quantum dot and the influence of temperature and coupling strength on the steady state and current characteristics is discussed. Further the quantum dot with spin-dependent coupling and a cyclic triple quantum dot are investigated to show current blocking effects and negative conductance due to Coulomb repulsion respectively quantum interference. The issue of quasi-degenerate states is discussed on the triple quantum dot by variation of gate voltages.

Kurzfassung

Die Born-Markov Mastergleichung und deren Herleitung zur Beschreibung offener dissipativer Quantensysteme im Nicht-Gleichgewicht wird in Kapitel 1 vorgestellt und mit dem Lindblad Formalismus verglichen, welcher eine gültige Lösung für den stationären Zustand garantiert. Anschließend wird auf die Erweiterung auf quasi-entartete Zustände eingegangen und die resultierenden Gleichungen in der System Eigenbasis für den stationären Zustand im Weitbandlimit dargestellt.

Im zweiten Kapitel wird die Herleitung des selbst-konsistenten Born Ansatzes präsentiert, der eine Verbesserung der eingangs erwähnten Born-Markov Mastergleichung darstellt. Weiters werden explizite Formeln für die Berechnung des stationären Zustandes und des stationären Stromes für einen einzelnen Quantenpunkt (Anderson Störstelle) mit Spin-abhängiger Badkoppelung hergeleitet.

Im dritten Kapitel werden diese beiden Methoden schließlich numerisch für den Quantenpunkt ausgewertet, wobei explizit auf den Einfluss von Temperatur und Badkoppelungsstärke auf den stationären Zustand und auf die Spannungs-Strom-Kennlinie eingegangen wird. Anschließend wird bei dem Quantenpunkt mit Spin-abhängiger Badkoppelung und bei einem zyklischen Quantenring, das Verschwinden der elektrischen Leitfähigkeit (Coulomb-Blockade) bzw. eine negative Leitfähigkeit aufgrund von Coulomb Abstoßung beziehungsweise aufgrund von Quanteninterferenz beobachtet. Auf die Behandlung von quasi-entarteten Zuständen ohne sekulare Approximation wird anhand des zyklischen 3-Platz Systems unter Variation der Gatespannung eingegangen.

Contents

Introduction/Motivation

Advances in strongly correlated electron systems Condensed matter physics is a branch in physics that tries to explain physical properties in fluids, solids or more exotic condensed phases of matter [1, 2]. Since the 1930s the Schrödinger equation serves as the basis of condensed matter physics and it is in combination with the Pauli-Dirac equation (to account for relativistic effects) still assumed to describe the physical behaviour of matter correctly. By providing a time evolution of probabilities it incorporates the quantum nature of the relevant elementary particles. Though the Schrödinger equation is not apt to derive directly macroscopic properties because the dimensions of the configuration space go beyond any computable scale. Since it's formulation 1926 a significant part of condensed matter physics is occupied with finding the best approximations to successfully solve the Schrödinger equation. Most of those approaches like mean field theory, Bloch theory or density functional theory [3, 4] assume the interactions of electrons to be small or even neglect them. Still this branch of theories is well apt to describe the metallic, insulating or semiconducting behaviour and many more effects of many materials where the correlation of electrons is not dominant.

Other interesting effects like Mott insulator transition [5] in nickel(II) oxide, the Kondo effect [6] in iron doped gold or high temperature superconductivity [7] in cuprates cannot be addressed by these approaches and need indeed other methods that account for this correlation since in these materials the physics is determined by the strong interaction of electrons. A very prominent model to describe these phenomena is the Hubbard model [8] which itself can be solved exactly only in some special cases. For the general case one faces the problem that the Hilbert space increases exponentially with the number of orbitals/sites N. The corresponding Hamilton matrix is not very decomposable (in contrast to the tight binding model), so the resulting eigenvalue problem is hard to address with increasing N by brute force methods like full diagonalisation, but also more sophisticated approaches like Krylov space methods (Lanczos algorithm) already reach their limits for $N \approx 20$. A more detailed discussion about the limits of the Lanczos method when solving the Hubbard model is given in [9, Chapter 8].

In the last three decades the dynamical mean field theory (DMFT) [10, 11] developed to describe the electronic structure of strongly correlated materials. It maps the in general intractable many-body lattice problem on a single site impurity problem which is also known as the Anderson impurity model [12]. It describes a single interacting quantum dot connected to a non-interacting environment and can be regarded as a combination of a tight binding model and a single site Hubbard model.

Quantum microelectronics Whereas theoretical techniques and understanding of solid states has increased over the past decades also enormous progress has been achieved on the manufacturing side. Since electronic devices have already reached the molecular limit it has become crucial to understand the electronic properties of such nano-structured devices like single-molecule transistors [13]. Quantum phenomena, like quantum blockade, Kondo effect or quantum interference [14, 15], have a great influence on the transport properties in single electron devices and present a rich pool of possible new quantum technologies. At this point it is important to emphasize not to investigate the quantum system as an isolated system because the influence of the contacts cannot be neglected. To include also dissipation we model a so called open quantum system, which is realised by attaching non-interacting thermalized metallic leads to the strongly correlated quantum device.

To describe such an open quantum systems the before mentioned Anderson impurity model serves as a good starting point. Since solving this model is also a crucial step in the before mentioned DMFT several different methods have been developed like numerical renormalization group (nRG) techniques [16, 17], non-equilibrium Green's function technique with non-crossing approximation (nGF+NCA) [18], continuous time Quantum Monte Carlo algorithms (ctQMC) [19], density matrix renormalization group methods (DMRG) [20, 21] or cluster approaches like cluster perturbation theory (CPT) and it's improvement, the variational cluster approach (VCA) [22, 23, 24].

Approach to open quantum systems In this thesis I address two methods to describe the dynamics of such open quantum systems, the Born-Markov master equation approach (BM) [25] and it's extension the self-consistent Born master equation approach (SCB) [26]. These methods shall help to describe quantum microelectronic properties of mesoscopic quantum devices that in near future may be or already are realised [27, 28, 29]. An example of the full application of this method combining ab-initio calculations to determine parameters needed in the model is given in [30] for a benzene ring attached to gold contacts. The master equation approach has also be used in combination with other methods. For example the group around my supervisor Prof. von der Linden has proposed in a recent work [31] the so-called auxiliary master equation approach to act as an impurity solver for single-site DMFT. In a more recent work the group presented a CPT method that is based on the steady state solution of the Born-Markov master equation approach [32].

My thesis is structured the following way: In the first part I give a detailed derivation of the BM method and I explain and motivate the performed approximations. I explain the relation to the Lindblad formalism and discuss the approach for quasi-degenerate states and it's influence on the stability of the solution.

In the second part I explain the SCB approach and discuss it's improvements compared to the first method. With this more sophisticated approach I derive an analytic expression for the steady state of a single quantum dot in the wide band limit that inherits spin dependent coupling and also present the current formula in this SCB approach.

In the third part I give numerical results for different quantum systems, discuss temperature and coupling dependence of both introduced methods and analyse their numerical

instabilities for certain parameter ranges. Further I compare the current-voltage characteristics of the single Anderson impurity model obtained by different methods and reveal the quantum phenomenon of Coulomb blockade respectively negative conductance in two different setups, in one case for spin dependent hopping and in the other case in a cyclic quantum system, hosting interference effects.

1 Born-Markov master equation approach

In this chapter I give the derivation of the Born-Markov master equation approach roughly following [33, 25, 34].

The aim of this approach is to predict the non-equilibrium state of an open quantum system [35], that is characterized by a central system of interest exchanging information with an environment. This environment is realized by thermally stabilized baths b, also referred to as reservoirs or leads which are assumed to be infinitely long and inherit a dissipative character thus information propagated from the system to the baths gets lost (see section 1.4.1). The baths shall not inherit electronic correlation features thus will be modelled with a non-interacting tight binding Hamiltonian (1.4).

The central system s of interest instead is modelled by a Hubbard Hamiltonian (1.5) to include strongly correlated electrons that for instance arise from on-site interactions due to Coulomb repulsion. For this Born-Markov master equation approach basically any system Hamiltonian can be applied that can be fully solved since we need the system eigenstates as eigenbasis to evaluate the final equations. Depending on the strength of the realized non-equilibrium we also have to consider eigenstates corresponding to higher eigenenergies, but for small deviations from equilibrium also Hamiltonians can be treated for which only the energetically lowest eigenspace is tractable. For the calculations described in this work we restrict the number of sites in the central system to $N \leq 6$.

The non-equilibrium situation of the system is realised by coupling it to the baths which are associated with different potentials like an external electric field inducing an electric current. The coupling of the system to the non-interacting leads will be realised by an interaction Hamiltonian (1.2) also used in the Anderson impurity model (AIM). With the special choice of choosing a Hubbard Hamiltonian for the central region this model indeed corresponds to the AIM but as mentioned before any quantum system described by it's eigenspectrum and it's corresponding eigenbasis can be treated with this approach.

We are interested in the the dynamics of a quantum system that is described by the probabilistic mixture of pure states: the density operator. Since we are not interested in the detailed properties of the baths we will average over their degrees of freedom by taking the partial trace and receive the central object of interest used to describe the quantum system: the reduced density operator (see section 1.1.3).

A first order differential equation that describes the time evolution of such probabilities (given by the reduced density operator) is called a **Master Equation** [33]. The starting point to derive such a description of quantum dynamics is - as mentioned in the introduction

- the Schrödinger equation (see section 1.1.4).

The Born and Markov approximations are in detail discussed in section 1.4 and are essential to get a closed equation for the steady state 1.5.

In section 1.6 we will see how the secular approximation guarantees the steady state to represent indeed a valid solution (density matrix) by applying the Lindblad formalism.

1.1 Setup

1.1.1 Techniques

Basic concepts of different quantum mechanical pictures (Schrödinger, Heisenberg, Interaction) presumed the reader will be introduced to the superoperator notation. Several times the practical features of traces and the basic commutator / anticommutator relations are needed. A repetition of complex Laplace and Fourier transforms and their properties and of some functional analytic integration theorems is given in the appendix 4.2.

The following features of the examined system Hamiltonian (Hubbard) will be used during the derivation: The Hubbard Hamiltonian is time invariant and conserves particle number and spin.

Here is a list of approximations respectively assumptions used throughout the work to make a calculation feasible:

- Perturbation theory to second order in the Liouville-von Neumann equation
- Born approximation
- Markov approximation
- Secular approximation
- Adiabatic switch-on of the interaction between bath and system at $t = -\infty$
- Steady state limit $t \to \infty$
- Weak coupling limit (as justification for Born/Markov approximation)

Later on we use the terminology of non-equilibrium Green's functions [36] to describe the current formula in a more common way.

1.1.2 Model system

The examined problem shall consist of a strong interacting fermionic system S coupling weakly to a fermionic reservoir or bath b.

Let $a^s_{i\tau} = a^s_\mu$ be an annihilator ($s = -$) or a creator ($s = +$) of an electron of a spin τ at a system site i (combined index μ represents state $|\mu\rangle$). For the leads let $d^s_{\alpha k\tau}$ be analogously an creator/annihilator of an electron with a spin τ and momentum k, where

the index α indicates on which bath (lead) the operator acts. To refer to the spin of a^s_μ we use τ_μ.

The total Hamiltonian H can be separated according to the affective domain of their operators, so we identify the interaction Hamiltonian H_1, that operates on both, system and bath and is switched on adiabatically at time $t = -\infty$

$$H = H_0 + H_1, \qquad H_0 := H_b + H_s \tag{1.1}$$

The general form of the interaction Hamiltonian is

$$H_1 := \lim_{\varepsilon \to 0^+} e^{-\varepsilon|t|} \sum_{\alpha\mu k\tau} t_{\alpha\mu k\tau} a^\dagger_{\mu(\tau_\mu)} d_{\alpha k\tau} \delta_{\tau_\mu,\tau} + h.c. = e^{-0^+|t|} \sum_{\mu s} s a^s_\mu D^{\bar{s}}_\mu \tag{1.2}$$

$$D^s_\mu := \sum_{\alpha k\tau} t^s_{\alpha\mu k\tau} d^s_{\alpha k\tau} \delta_{\tau_\mu,\tau} \tag{1.3}$$

with the coupling constants ($t^+_{\alpha\mu k\tau} = t^*_{\alpha\mu k\tau}$). So for the bath operators D the bath index α is additionally included in the combined index μ, we may use later $D^s_\mu = \sum_\alpha D^s_{\alpha\mu}$. The spin isn't changed by the interaction Hamiltonian and it's index τ may be used later to discuss spin dependent coupling. We will assume later, that each lead α couples just to one system site μ_α.

The Hamiltonian of the non interacting leads (reservoir) is

$$H_b = \sum_{\alpha k\tau} \epsilon_{\mathbf{k}} d^\dagger_{\alpha k\tau} d_{\alpha k\tau} \tag{1.4}$$

The system Hamiltonian don't has to be specified in the following derivation but for later calculations we will refer to the following Hubbard model:

$$\begin{aligned} H_s = &\sum_{i\tau} \xi_i a^\dagger_{i\tau} a_{i\tau} + b \sum_{i\tau} (a^\dagger_{i\tau} a_{i+1\tau} + a^\dagger_{i+1\tau} a_{i\tau}) \\ &+ U \sum_i (n_{i\uparrow} n_{i\downarrow} - \frac{1}{2}(n_{i\uparrow} + n_{i\downarrow})) \\ &+ V \sum_i (n_{i\uparrow} + n_{i\downarrow} - 1)(n_{i+1\uparrow} + n_{i+1\downarrow} - 1) \end{aligned} \tag{1.5}$$

1.1.3 Modified density operator

To describe the behaviour of the system S under influence of the leads we will start deriving an equation for the full density operator of the total setup, ρ_{bs}, including system and bath. Subsequently we will trace out the bath to get a reduced density operator σ_s of the system S. Summarized in this paragraph some of the main important properties of the density operator are given:

Properties of the density operator $\rho_{sb}(t)$. The density operator is used to describe a quantum system in a mixed state [37, 38, 34]. Main motivation for this is, that it's possible to perform a 'complete' experiment on all quantum systems, for instance it's not possible to clarify in which quantum state a quantum system definitely is. But statements of the probabilistic mixture of pure states (mixed quantum state) are feasible and can be used to describe the quantum system in a statistical way.

The density matrix can be constructed in an eigenstate basis $(|\varphi_i\rangle)_{i\in\mathbb{N}}$ of H

$$\rho(t) = \sum_i p_i |\varphi_i(t)\rangle\langle\varphi_i(t)| \tag{1.6}$$

with $0 \leq p_i \leq 1$ and $\sum_i p_i = 1$. If the basis is orthonormal then the coefficients p_i indicate the probability to find the system in the state $|\varphi_i(t)\rangle$. Arising from the probabilistic construction the density operator is **positive**, which means that for any state $|a\rangle$ in the Hilbert space $\mathcal{H}$ the expression $\langle a|\rho|a\rangle$ gives a positive result, indicating the probability to find a system in that state. The property of positivity is equivalent to having all eigenvalues of ρ greater or equal to zero.

The **trace** of the operator is **equal to one**. The density operator is describing a pure state if and only if the trace of ρ^2 is equal to one. Since it's symmetric construction the density operator is **self-adjoint** (hermitian density matrix) $\rho^\dagger = \rho$.

Modified density operator $\rho_{sb}(t)$. For later use we extend this derivation to a more general operator, in particular we want to regard a modified density operator beside the normal density operator:

$$\rho = \begin{cases} \rho_{bs}, & \eta = +1 \\ [a^s_\kappa \rho_{bs}], & \eta = -1 \end{cases} \tag{1.7}$$
$$\sigma = \mathrm{Tr}_b\Big\{\rho\Big\}$$

The index η is used to distinguish between the two cases. For $\eta = +1$ we refer to the **bosonic density operator** ρ_{bs} whereas we use $\eta = -1$ to label the **fermionic modified density operator** $\tilde{\rho}$.

As the modified density operator is not self-adjoint we use a comma or parenthesis in the scalar product (matrix representation) to indicate on which side of the scalar product the operator acts first:

$$\langle a|(\sigma|b\rangle = \langle a|\sigma^\dagger)|b\rangle \tag{1.8}$$

In general this will be the right side using the short notation for matrix elements $A_{ab} := \langle a|(A|b\rangle$. As we will identify some Hermitian conjugate parts later on, we need a special dagger operator ‡ that works like the usual dagger (adjoint) operator † but acts like a unity operator on the modified (not self-adjoint) density operator $\tilde{\sigma}$, pretending it to be

self-adjoint in some sense:

$$\sigma^{\ddagger} = \sigma \tag{1.9}$$

$$\rho^{\ddagger} = \rho \tag{1.10}$$

$$\langle a|(\,[A^{\dagger}\sigma]^{\ddagger}|b\rangle = \langle a|(\sigma A|b\rangle \tag{1.11}$$

We will need this special dagger notation in section 1.4.3 where we will use the abbreviation $h.c.(\ddagger)$ to indicate that the Hermitian conjugate is performed according to that special dagger operator.

Notation

Operators with the subscripts $_s$ of $_b$ are always referring to density operators, either to reduced system, reduced bath or total density operators. The operators ρ and σ without subscript will be of general type as defined in equation (1.7) and used in the following sections to get the derivation for both cases. Superscripts ^{s} are referring to the dagger operations $(\dagger, \ddagger)$. If we refer explicitly to the modified density operators we will use a tilde to make the setting clear

$$\tilde{\sigma}^s_\kappa = \mathrm{Tr}_b\Big\{\tilde{\rho}^s_\kappa\Big\} = \begin{cases} \mathrm{Tr}_b\Big\{a_\kappa \rho_{bs}\Big\} & s = - \\ \mathrm{Tr}_b\Big\{a^\dagger_\kappa \rho_{bs}\Big\} & s = + \end{cases} \tag{1.12}$$

Note: $(\tilde{\sigma}^s_\kappa)^\dagger \neq \tilde{\sigma}^{\bar{s}}_\kappa$.

We will refer to the definition of the modified density operator in section 1.4.3 where it is of relevance especially when treating the self-consistent Born approach in chapter 2. The time dependence and relationship of both operators is of crucial importance and will be discussed in detail in section 2.1.

1.1.4 Time dependent Schrödinger equation

The time dependent Schrödinger equation is the fundamental starting point to describe the time evolution of a state $|\Psi(t)\rangle$ in the given quantum system described by the (not necessarily time-independent) Hamilton operator H:

$$i\hbar\frac{d}{dt}|\Psi(t)\rangle = H|\Psi(t)\rangle$$

The time evolution of the system described by the (modified) density operator $\rho(t)$ is given by the Liouville-von Neumann equation:

$$i\hbar\frac{d}{dt}\rho(t) = [H, \rho(t)]_- \tag{1.13}$$

where $[\,\cdot\,,\cdot\,]_-$ defines the commutator $[A, B] = AB - BA$. Later we use $[\,\cdot\,,\cdot\,]_+$ for the anticommutator.

1.2 Superoperators

In this section the superoperator notation is introduced to have a more convenient way to write the equations.

Definition: A superoperator is a mapping of an operator $A \in \mathcal{O}$, ($\mathcal{O}$ vector space of operators) onto an operator. We use the word operator to refer to mappings that operate on the Hilbert space of quantum states of the examined quantum system. In quantum mechanics we define the Liouville superoperator or Liouvillian $\mathcal{L}_H$ with respect to the Hamiltonian H as

$$\mathcal{L}_H : \mathcal{O} \to \mathcal{O}, \qquad \mathcal{L}_H(A) := [H, A] = HA - AH \tag{1.14}$$

1.2.1 Properties of superoperators

Theorem 1.1 (Exponentials of the Liouville operator)
We want to prove the exponential application of the Liouville operator:

$$e^{i\mathcal{L}_H t}A = e^{iHt}Ae^{-iHt} \tag{1.15}$$

Proof 1.1
We start with the series representation of the exponential superoperator:

$$e^{i\mathcal{L}_H t}A = \sum_{n=0}^{\infty} \frac{(it)^n}{n!}\mathcal{L}_H^n A \tag{1.16}$$

Now we show by induction that

$$\mathcal{L}_H^n A = \sum_{k=0}^{n}(-1)^k \binom{n}{k} H^{n-k}AH^k \qquad \text{(hypothesis)}$$

$$\mathcal{L}_H^1 A = HA - AH = (-1)^0 H^1 A H^0 + (-1)^1 H^0 A H^1 \qquad \text{(basis)}$$

$$\mathcal{L}_H^{n+1} A = \mathcal{L}_H \mathcal{L}_H^n A = \sum_{k=0}^{n}(-1)^k \binom{n}{k} H^{n+1-k}AH^k - \sum_{k=0}^{n}(-1)^k \binom{n}{k} H^{n-k}AH^{k+1} \qquad \text{(step)}$$

We shift the index of the second sum $k+1 \to k$ and extend the sums as $\binom{n}{n+1} = \binom{n}{-1} = 0$.

$$= \sum_{k=0}^{n+1}(-1)^k \binom{n}{k} H^{n+1-k}AH^k + \sum_{k=1}^{n+1}(-1)^k \binom{n}{k-1} H^{n-k+1}AH^k$$

With $\binom{n}{k} + \binom{n}{k-1} = \binom{n+1}{k}$ we have proven this representation.
Now we have a look at the right side and compare the coefficients of t^n, with $r = n - s$:

$$e^{iHt}Ae^{-iHt} = \sum_{r=0}^{\infty}\frac{(it)^r}{r!}H^r \cdot A \cdot \sum_{s=0}^{\infty}\frac{(-it)^s}{s!}H^s = \sum_{n=0}^{\infty}\sum_{s=0}^{n}\frac{n!}{n!}\cdot\frac{(it)^n(-1)^s}{(n-s)!s!}H^{n-s}AH^s$$

$$\overset{s\equiv k}{=} \sum_{n=0}^{\infty}\frac{(it)^n}{n!}\mathcal{L}_H^n A = e^{i\mathcal{L}_H t}A$$

□

Note: The exponential form of the superoperator is self-adjoint:

$$\left(e^{-i\mathcal{L}_H t}A\right)^\dagger = \left(e^{-iHt}Ae^{iHt}\right)^\dagger = e^{-iHt}A^\dagger e^{iHt} = e^{-i\mathcal{L}_H t}A^\dagger \tag{1.17}$$

Theorem 1.2 (Matrix representation of functions and adjoint functions of Liouville superoperators in the eigenbasis of the system)
Let $\mathcal{L}$ be the Liouvillian superoperator according to the system Hamiltonian acting on the not necessarily self-adjoint operator A and let $|a\rangle, |b\rangle$ be system eigenvectors with eigenenergies E_a and E_b. The function $f : \mathbb{C} \to \mathbb{C}$ shall be an analytic function around zero.

Then the following identities for the matrix representation and the adjoint of functions of the superoperator holds:

$$\left(f(\mathcal{L})A\right)_{ab} = f(E_a - E_b)\left(A\right)_{ab} \tag{1.18}$$

$$\left[f(\mathcal{L})A\right]^\dagger = f(-\mathcal{L})^* A^\dagger \tag{1.19}$$

Note: As the operator in the scalar product $\langle a|\ldots|b\rangle$ is not necessarily self-adjoint we use a comma or a single parenthesis to indicate to which side of the scalar product the operator belongs. Throughout all sections this will be the right side $A_{ab} := \langle a|, A|b\rangle$.

Proof 1.2
We start looking at the Taylor series of $f(\mathcal{L})A = \sum_{n\geq 0} a_n \mathcal{L}^n A$. We show by induction that

$$(\mathcal{L}^n A)_{ab} = (E_a - E_b)^n A_{ab} \qquad A_{ab} := \langle a|(A|b\rangle \qquad \text{(hypothesis)}$$

$$(HA - AH)_{ab} = E_a A_{ab} - A_{ab}E_b = (E_a - E_b)A_{ab} \qquad \text{(basis)}$$

$$(H\mathcal{L}^n A - \mathcal{L}^n AH)_{ab} = E_a(E_a - E_b)^n A_{ab} - (E_a - E_b)^n A_{ab}E_b = (E_a - E_b)^{n+1}A_{ab} \qquad \text{(step)}$$

So we get in the series representation

$$\left(f(\mathcal{L})A\right)_{ab} = \sum_{n\geq 0} a_n \left(\mathcal{L}^n A\right)_{ab} = \sum_{n_\geq 0} a_n (E_a - E_b)^n A_{ab} = f(E_a - E_b)A_{ab}$$

Some examples:

$$\mathrm{Tr}\{f(\mathcal{L})A\} = f(0)\mathrm{Tr}\{A\} \tag{1.20}$$

$$\left(f(\omega - \mathcal{L})A\right)_{ab} = f(\omega - (E_a - E_b))A_{ab} \tag{1.21}$$

For the adjoint operator $\dagger$ acting on the function of a Liouville superoperator we have

$$\begin{aligned}
\left(\left[f(\mathcal{L})A\right]^\dagger\right)_{ab} &= \langle a|, \left[f(\mathcal{L})A\right]^\dagger|b\rangle \\
&= \overline{\langle b|, f(\mathcal{L})A|a\rangle} \\
&= \overline{f(E_b - E_a)\langle b|, A|a\rangle} \\
&= f(-(E_a - E_b))^* \langle a|, A^\dagger|b\rangle \\
&= \left(f(-\mathcal{L})^* A^\dagger\right)_{ab}
\end{aligned}$$

□

Time evolution

The Liouville-von Neumann equation can thus be expressed via superoperators:

$$\dot{\rho}(t) = -\frac{i}{\hbar}[H, \rho(t)] = -\frac{i}{\hbar}\mathcal{L}_H \rho(t) \tag{1.22}$$

From now on $\hbar$ shall be set to 1.

For the time evolution of a not-necessarily self-adjoint (modified) density operator in Schrödinger picture ρ we can use the superpropagator (operator that evolves an operator in time)

$$\rho(t) = e^{-i\mathcal{L}_H(t-t_0)}\rho(t_0) \tag{1.23}$$

Note that the time evolution of ordinary operators in Heisenberg (or interaction picture) picture have the opposite sign in the propagators:

$$A(t) = e^{iHt}Ae^{-iHt} = e^{i\mathcal{L}_H t}A \tag{1.24}$$

Notation of time propagators and superpropagators

As all occurring Hamiltonians are not time dependent (time translation invariant) we can write all time propagators and superpropagators $\mathcal{T}$ as exponentials of the corresponding Hamiltonian respectively Liouvillian and just need one argument (time difference) instead of two for initial and final time $\mathcal{T}(t_f, t_i) = \mathcal{T}(t_f - t_i)$. In order to indicate time intervals from a fixed initial time t_0 we use underlined time arguments as short notation $\underline{t} = (t - t_0)$. If $t_0 = -\infty$, we write $\mathcal{T}(t, -\infty)$. We will use the letter G for time propagators and the calligraphic letter $\mathcal{G}$ for time superoperators with a subscript indicating according to which Hamiltonian / Liouvillian the propagation is carried out $G_x(t) = e^{iH_x t}$. A missing subscript indicates time evolution with respect to the full Hamiltonian H. Some relations with the corresponding Hamiltonians are given below:

$$\mathcal{G}(t)A \;=\; e^{-i\mathcal{L}t}A \;=\; e^{-iHt}Ae^{iHt} \;=\; G(t)AG^\dagger(t) \tag{1.25}$$

Below is a table with abbreviations for propagators, superpropagators and Liouvillians according to the Hamiltonians defined in section 1.1.2:

$G(t) = e^{iHt}$	$\mathcal{G}(t) = e^{i\mathcal{L}t}$	$\mathcal{L}A = [H, A]_-$	$H = H_0 + H_1$
G_0	$\mathcal{G}_0$	$\mathcal{L}_0$	$H_0 = H_b + H_s$
G_s	$\mathcal{G}_s$	$\mathcal{L}_s$	H_s
G_b	$\mathcal{G}_b$	$\mathcal{L}_b$	H_b
G_I	$\mathcal{G}_I$	$\mathcal{L}_1$	H_1

When in the Born approximation scheme we introduce a time superpropagator $\mathcal{U}$ acting on the system space that propagates according to the full Hamiltonian

$$\mathcal{U} : S \to S, \qquad \mathcal{U}(t-t')\sigma_s(t') := \mathrm{Tr}_b\Big\{\mathcal{G}(t-t')\rho_{bs}(t')\Big\} = \sigma_s(t) \tag{1.26}$$
$$\rho_{bs}(t') = \rho_b(t') \otimes \sigma_s(t'), \quad \text{with } \rho_b(t') = \rho_b, \ \forall t' \text{ in Born approx.}$$

The Born approximation with the time invariant bath density matrix is essential as otherwise the operator wouldn't be well defined. Note that the operator is additive in the time argument $\mathcal{U}(t)\mathcal{U}(t') = \mathcal{U}(t+t')$ and that the trace over the bath and the time propagator according to the full Hamiltonian do not commute since the involved interaction Hamiltonian.

1.3 Interaction picture

We want to go now to the interaction picture (Dirac picture, denoted by I) where we treat the coupling Hamiltonian H_1 as a weak perturbation switched on adiabatically at time t_0. Before this starting point, the interaction Hamiltonian is zero and thus the interaction picture is equal to the Schrödinger picture $|\Psi^I(t_0)\rangle = |\Psi^S(t_0)\rangle$. Time independent operators A in the interaction picture have an underlined time dependent argument that also includes implicitly the starting time t_0:

$$A^{(I)}(\underline{t}) = e^{iH_0(t-t_0)} A e^{-iH_0(t-t_0)} = e^{i\mathcal{L}_0 \underline{t}} A, \tag{1.27}$$

The density matrices are denoted explicitly with an $\rho^S(t)$ respectively $\rho^I(\underline{t})$ for distinguishing between Schrödinger and interaction picture.[1]

$$\rho^I(\underline{t}) = G^\dagger(t-t_0)\,\rho^S(t)\,G(t-t_0) = e^{i\mathcal{L}_0(t-t_0)}\rho^S(t), \tag{1.28}$$
$$\rho^I(t_0) = \rho^S(t_0).$$

Liouville-von Neumann equation in the interaction picture

$$\dot{\rho}^I(\underline{t}) \stackrel{!}{=} -i[H_1^I(\underline{t}), \rho^I(\underline{t})] = -i\mathcal{L}_1^I(\underline{t})\rho^I(\underline{t}), \tag{1.29}$$
$$\mathcal{L}_1^I(\underline{t}) := e^{i\mathcal{L}_0(t-t_0)}\mathcal{L}_1. \tag{1.30}$$

Proof 1.3

$$\dot{\rho}^I(\underline{t}) = \frac{d}{dt}\mathrm{e}^{iH_0(t-t_0)}\rho(t)e^{-iH_0(t-t_0)} = \mathrm{e}^{iH_0(t-t_0)}(i[H_0,\rho(t)] + \dot{\rho}(t))\mathrm{e}^{-iH_0(t-t_0)} \tag{1.31}$$

[1] As the Hamilton has no time dependency the stationary solution and thus ρ must be time independent, but as we don't know this solution, we have to start from an arbitrary point (solution of the isolated system) and evolve in time to reach the total stationary solution.

Using (1.22) we get

$$\ldots = \mathrm{e}^{iH_0\underline{t}}(i[H_0,\rho(t)] - i[H_T,\rho(t)])\mathrm{e}^{-iH_0\underline{t}} = -i\mathrm{e}^{i\mathcal{L}_0\underline{t}}[H_1,\rho(t)] = -ie^{i\mathcal{L}_0\underline{t}}\mathcal{L}_1\rho(t)$$

On the other hand we can insert the unity operator $\mathbb{1} = e^{-iH_0\underline{t}}e^{iH_0\underline{t}}$ *between the Hamilton and density operator in the second term of the previous equation and get:*

$$\cdots = -i[e^{i\mathcal{L}_0\underline{t}}H_1, e^{i\mathcal{L}_0\underline{t}}\rho(t)] = -i\mathcal{L}_1^I(\underline{t})\rho^I(\underline{t}), \tag{1.32}$$

regaining the Liouville equation in the interaction picture and extending the interaction notation to superoperators.

$$\mathcal{L}_1^I(\underline{t})A := (e^{i\mathcal{L}_0\underline{t}}\mathcal{L}_1)A \tag{1.33}$$

The brackets indicate, that the exponential superoperator just works on the Hamiltonian in the commutator of the second superoperator We have also received a convenient rule for the composition of superoperators:

$$e^{i\mathcal{L}_0 t}(\mathcal{L}_1 A) = (e^{i\mathcal{L}_0 t}\mathcal{L}_1)(e^{i\mathcal{L}_0 t}A) \tag{1.34}$$

□

1.3.1 Interaction picture for tensor product operators

Next we want to derive the interaction picture of tensor-product operators. Let $A_{sb} = A_s \otimes A_b$ be such an tensor-product operator with $[A_s, A_b] = 0$, then we also get in the interaction picture with $H_0 = H_b + H_s, [H_b, H_s] = 0$ a tensor structure:

$$\begin{aligned} A_{sb}^I(\underline{t}) &= e^{i(H_b+H_s)(t-t_0)}A_{sb}e^{-i(H_b+H_s)(t-t_0)} \\ &= e^{iH_s(t-t_0)}A_s e^{-iH_s(t-t_0)}\, e^{iH_b(t-t_0)}A_b e^{-iH_b(t-t_0)} = A_s^I(\underline{t}) \otimes A_b^I(\underline{t}) \end{aligned} \tag{1.35}$$

1.3.2 Series expansion in the interaction picture

The advantage now is to have only the small perturbation Hamiltonian in the differential equation. We proceed by integration (one Picard iteration though $\rho^I(t)$ in the integral is still unknown) starting from the known state $\rho(t_0)$ of the non interacting system:

$$\rho^I(\underline{t}) = \rho(t_0) - i\int_{t_0}^{t} dt'\mathcal{L}_1(\underline{t}')\rho^I(\underline{t}'). \tag{1.36}$$

Reinserting this expression for $\rho^I(\underline{t})$ into the right part of the Liouville-von Neumann equation (1.22) results in:

Integro-differential form of the Liouville-von Neumann equation
$$\dot{\rho}^I(\underline{t}) = -i\mathcal{L}_1(\underline{t})\rho(t_0) - \int_{t_0}^{t} dt' \mathcal{L}_1(\underline{t})\mathcal{L}_1(\underline{t}')\rho^I(\underline{t}'). \quad (1.37)$$

I want to remark, that this equation in it's present form evaluated in the system eigenbasis is similar to the Redfield-Bloch equation [34], which assumes $t_0 = 0$ and the density operator to be only dependent on it's present state $\rho^S(t') = \rho^S(t)$ (Markovian property).

The drawback of having a more complex structure in the equation above with having an additional integral now is compensated by the fact, that errors in the unknown density operator in the integral are alleviated by the square of the perturbation Hamiltonian that is supposed to be small.

In the spirit of perturbation theory a treatment in higher orders is possible by further expansion of equation 1.36 through recursively inserting the equation of motion of the density matrix.

$$\rho^I(\underline{t}) = \rho(t_0) - i\int_{t_0}^{t} \mathcal{L}_1(\underline{t}')\rho(t_0)dt' - \int_{t_0}^{t}\int_{t_0}^{t'} \mathcal{L}_1(\underline{t}')\mathcal{L}_1(\underline{t}'')\rho(\underline{t}'')dt''dt' \quad (1.38)$$

At this point another method shall be mentioned that is to a certain extent similar to the later discussed self-consistent Born approximation. Main idea is to decompose the Liouville-von Neumann equation for the reduced density operator in an unperturbed part and one corresponding to a self-energy term, which inherits an operator for whom a new equation of motion is defined. This results in an infinite hierarchy of equations that leads to the so-called hierarchical master equation approach [39, 40, 41].

1.3.3 Transformation back to Schrödinger picture

Now that we have expanded the integral of ρ to second order in H_1 we go back to the Schrödinger picture:

$$\begin{aligned}
\rho^S(t) &= e^{-i\mathcal{L}_0(t-t_0)}\rho^I(\underline{t}),\\
\dot{\rho}^S(t) &= -i\mathcal{L}_0\rho^S(t) + e^{-i\mathcal{L}_0(t-t_0)}\dot{\rho}^I(\underline{t}),\\
\dot{\rho}^S(t) &= -i\mathcal{L}_0\rho^S(t) - ie^{-i\mathcal{L}_0(t-t_0)}\mathcal{L}_1(\underline{t})\rho(t_0) - \int_{t_0}^{t} dt' e^{-i\mathcal{L}_0(t-t_0)}\mathcal{L}_1(\underline{t})\mathcal{L}_1(\underline{t}')\rho^I(\underline{t}'). \quad (1.39)
\end{aligned}$$

1.3.4 Trace over bath

Next we want to trace out the bath because we are interested in the system. So we get the reduced (modified) density operator

$$\sigma(t) := \mathrm{Tr}_b\Big\{\rho(t)\Big\} = \begin{cases}\sigma_s(t) & \eta = +1\\ a_j^s\sigma_s(t) & \eta = -1\end{cases} \tag{1.40}$$

and the equation (1.39) turns into:

$$\dot\sigma(t) = -i\mathrm{Tr}_b\Big\{\mathcal{L}_0\rho^S(t)\Big\} - i\mathrm{Tr}_b\Big\{e^{-i\mathcal{L}_0(t-t_0)}\mathcal{L}_1(\underline{t})\rho(t_0)\Big\} - \int_{t_0}^{t} dt'\mathrm{Tr}_b\Big\{e^{-i\mathcal{L}_0(t-t_0)}\mathcal{L}_1(\underline{t})\mathcal{L}_1(\underline{t}')\rho^I(\underline{t}')\Big\}. \tag{1.41}$$

1.4 Born and Markov approximation

1.4.1 Born approximation

To evaluate the trace we assume that the density operator is separable at all times t, thus it can be written as a tensor product of a system and a bath part $\rho(t) = \mathrm{Tr}_b\Big\{\rho(t)\Big\}\otimes\rho_b(t) = \sigma(t)\otimes\rho_b(t)$ (Born approximation). That means, we assume, that there are no entangled states of bath and system for any time, which is reasonable because due to the thermalization of the bath, dissipation occurs and any entanglement would break down instantly due to the very short decoherence times. The interesting fact in this approximation is that the feature of dissipation or open quantum system is introduced just by claiming the density operator to be separable.

This thermalization implies that the reduced density operator of the bath is constant over time $\rho_b(t) = \rho_b = \frac{1}{Z}e^{\beta(H_b-\mu_\alpha)}$. Here we also have introduced the bath potential μ_α that isn't per-se defined in the non-equilibrium situation.

The separability of the density operator holds also in the interaction picture (1.35):

$$\rho^I(\underline{t}) = \big(e^{iH_s(t-t_0)}\sigma^S(t)e^{-iH_s(t-t_0)}\big)\otimes\underbrace{\big(e^{iH_b(t-t_0)}\rho_b^S(0)e^{-iH_b(t-t_0)}\big)}_{=\rho_b} = \sigma^I(\underline{t})\otimes\rho_b \tag{1.42}$$

Features of the reduced density operator concerning entanglement[42]

- The reduced density operator for an entangled pure ensemble is a mixed ensemble.
- A bipartite pure state ρ is entangled if and only if its reduced states are mixed rather than pure.
- There is the Positive Partial Transpose (PPT) condition (also known as Peres–Horodecki criterion) to find out whether a mixed state is entangled or not in the 2×2 and 2×3 dimensional case.

For non-Markovian systems that do not inherit such rapidly decaying correlations in the bath as assumed in this work, their is a generalization of the Born approximation. Instead of restricting the setup to product states and excluding entanglement one can define a more general projection superoperator $\mathcal{P}$ that includes correlations between system and bath by projecting onto the relevant part of the full density operator. This approach can be cast in a more general Lindblad form that allows to treat also highly non-Markovian quantum processes in structured environments [43, 34, 44].

1.4.2 Master Equation for the reduced density operator in the Schrödinger picture

For the first term of the equation (1.41) we decompose the non interacting Hamiltonian and use that system and bath Hamiltonian act on only one factor of the total density operator

$$\mathrm{Tr}_b\Big\{\mathcal{L}_0\rho(t)\Big\} = \mathrm{Tr}_b\Big\{[H_s,\sigma(t)]\rho_b\Big\} + \mathrm{Tr}_b\Big\{\sigma(t)[H_b,\rho_b]\Big\} \tag{1.43}$$

$$= [H_s,\sigma(t)]\underbrace{\mathrm{Tr}_b\Big\{\rho_b\Big\}}_{=1} + \sigma(t)\mathrm{Tr}_b\Big\{\underbrace{[H_b,\rho_b]}_{=0}\Big\}. \tag{1.44}$$

and get

$$\mathrm{Tr}_b\Big\{\mathcal{L}_0\rho(t)\Big\} = [H_s,\sigma(t)]. \tag{1.45}$$

For the second term we first proof the following identity

$$\mathrm{Tr}_b\Big\{e^{-i\mathcal{L}_0(t-t_0)}A\Big\} = \mathrm{Tr}_b\Big\{e^{-i\mathcal{L}_s(t-t_0)}e^{-i\mathcal{L}_b(t-t_0)}A\Big\} = e^{-i\mathcal{L}_s(t-t_0)}\mathrm{Tr}_b\Big\{A\Big\} \tag{1.46}$$

We use that the commutator $[H_s,H_b]$ is zero and let the bath superoperator vanish due to the cyclic invariance ot the bath trace for bath operators. So now we get for the second term using the separability of ρ at time t_0:

$$-i\mathrm{Tr}_b\Big\{e^{-i\mathcal{L}_0(t-t_0)}\mathcal{L}_1(\underline{t})\rho(t_0)\Big\} = -ie^{-i\mathcal{L}_s(t-t_0)}\mathrm{Tr}_b\Big\{[H_1^I(\underline{t}),\sigma(t_0)\rho_b]\Big\} \tag{1.47}$$

Now we look at the representation of the interaction Hamiltonian H_1 (1.2) in the interaction picture (1.35)

$$H_1^b(\underline{t}) = e^{i\mathcal{L}_b(t-t_0)}e^{-0^+|t|}\sum_{\mu s} sa_\mu^s D_\mu^{\overline{s}} = e^{-0^+|t|}\sum_{\mu s} sa_\mu^s D_\mu^{\overline{s}}(\underline{t}) \tag{1.48}$$

$$D_\mu^s(\underline{t}) := \sum_{\alpha k\tau} t_{\alpha\mu k\tau}^s d_{\alpha k\tau}^s(\underline{t})\delta_{\tau_\mu,\tau}$$

and use it to evaluate the first summand of the commutator

$$\mathrm{Tr}_b\Big\{H_1^I(\underline{t})\sigma(t_0)\rho_b\Big\} = e^{-0^+|t|}\sum_{\alpha\mu k\tau s} st^{\bar{s}}_{\alpha\mu k\tau}a^s_{\mu(\tau_\mu)}\sigma(t_0)\underbrace{\mathrm{Tr}_b\Big\{d^{\bar{s}}_{\alpha k\tau}(\underline{t})\rho_b\Big\}}_{=\langle d^{\bar{s}}_{\alpha k\tau}\rangle=0} = 0 \tag{1.49}$$

We get the same result for the other summand of the commutator. So the second term vanishes.

We remain with the third term of equation (1.41), an integral, where we also apply the just noted identity and use that the time evolution for the density matrix just happens in the system space (see equation 1.42).

$$\mathrm{Tr}_b\Big\{e^{-i\mathcal{L}_0(t-t_0)}\mathcal{L}_1(\underline{t})\mathcal{L}_1(\underline{t}')\rho^I(\underline{t}')\Big\} = \mathrm{Tr}_b\Big\{e^{-i\mathcal{L}_s(t-t_0)}\mathcal{L}_1(\underline{t})\mathcal{L}_1(\underline{t}')e^{i\mathcal{L}_s(t'-t_0)}\rho^S(t')\Big\} \tag{1.50}$$

To deal with the bath trace we go from the interaction picture with the Hamiltonian H_0 to a bath interaction picture with the Hamiltonian H_b:

$$\mathcal{L}_1^b(\underline{t}) := e^{i\mathcal{L}_b(t-t_0)}\mathcal{L}_1 = e^{-i\mathcal{L}_s(t-t_0)}\mathcal{L}_1(\underline{t}) \tag{1.51}$$

$$H_1^b(\underline{t}) := e^{i\mathcal{L}_b(t-t_0)}H_1 \tag{1.52}$$

By applying several times equation 1.34 we get

$$e^{-i\mathcal{L}_s(t-t_0)}\mathcal{L}_1(\underline{t})\mathcal{L}_1(\underline{t}')e^{i\mathcal{L}_s(t'-t_0)]}\rho^S(t') \tag{1.53}$$

$$= \mathcal{L}_1^b(\underline{t})e^{-i\mathcal{L}_s(t-t'+t'-t_0)}\mathcal{L}_1(\underline{t}')e^{i\mathcal{L}_s(t'-t_0)}\rho^S(t') = \mathcal{L}_1^b(\underline{t})e^{-i\mathcal{L}_s(t-t')}\mathcal{L}_1^b(\underline{t}')\rho^S(t') \tag{1.54}$$

Thus having

Master equation for the reduced density operator in the Schrödinger picture

$$\dot{\sigma}(t) = -i[H_s,\sigma(t)] - \int_{t_0}^{t} dt' \mathrm{Tr}_b\Big\{\mathcal{L}_1^b(\underline{t})\mathcal{G}_s(t-t')\mathcal{L}_1^b(\underline{t}')\rho^S(t')\Big\} \tag{1.55}$$

$$\mathcal{G}_s(t-t') := e^{-i\mathcal{L}_s(t-t')} \tag{1.56}$$

$$G_s(t-t') := e^{-iH_s(t-t')} \tag{1.57}$$

1.4.3 Evaluation of the commutators

Next we evaluate the superoperator induced commutators in the trace

$$\{\dots\} = [H_1^b(\underline{t}), G_s(t-t')[H_1^b(\underline{t}'), \rho(t')]G_s^\dagger(t-t')] \tag{1.58}$$

$$= \underbrace{H_1^b(\underline{t})G_s(t-t')H_1^b(\underline{t}')\rho(t')G_s^\dagger(t-t')}_{=:T_1} - \underbrace{H_1^b(\underline{t})G_s(t-t')\rho(t')H_1^b(\underline{t}')G_s^\dagger(t-t')}_{=:T_2} \tag{1.59}$$

$$- \underbrace{G_s(t-t')H_1^b(\underline{t}')\rho(t')G_s^\dagger(t-t')H_1^b(\underline{t})}_{=:T_3} + \underbrace{G_s(t-t')\rho(t')H_1^b(\underline{t}')G_s^\dagger(t-t')H_1^b(\underline{t})}_{=:T_4} \tag{1.60}$$

$$= T_1 - T_3 + (T_1 - T_3)^\ddagger = T_1 - T_3 + h.c.(\ddagger) \tag{1.61}$$

Here it is of importance that the Hermitian conjugate is performed due to the previously introduced (see section 1.1.3) modified dagger operator ‡, that doesn't change the modified density operator $\tilde{\sigma}$.

To evaluate T_1 we use again the representation of H_1^b (see equation (1.48) and insert it in T_1.

$$\mathrm{Tr}_b\Big\{T_1\Big\} = \mathrm{Tr}_b\Big\{H_1^b(\underline{t})G_s(t-t')H_1^b(\underline{t}')\rho(t')G_s^\dagger(t-t')\Big\} \tag{1.62}$$

$$= e^{-0^+(|t|+|t'|)}\sum_{\mu\kappa rs} rs\mathrm{Tr}_b\Big\{a_\mu^r D_\mu^{\bar{r}}(\underline{t})G_s(t-t')a_\kappa^s D_\kappa^{\bar{s}}(\underline{t}')\rho(t')G_s^\dagger(t-t')\Big\} \tag{1.63}$$

Now we get a minus sign from the anti-commuting fermionic operators when separating system and bath operators. As we are interested in the situation when the interaction is switched on ($|t| \approx 0$) we can simplify the time argument of the adiabatic convergence term to $(|t| + |t'|) \to |t - t'|$.

$$\mathrm{Tr}_b\Big\{T_1\Big\} = -e^{-0^+|t-t'|}\sum_{\mu\kappa rs} rs\mathrm{Tr}_b\Big\{D_\mu^{\bar{r}}(\underline{t})D_\kappa^{\bar{s}}(\underline{t}')\rho_b\Big\}a_\mu^r G_s(t-t')a_\kappa^s\sigma(t')G_s^\dagger(t-t') \tag{1.64}$$

$$= -e^{-0^+|t-t'|}\sum_{\mu\kappa rs} rs\Big\langle D_\mu^{\bar{r}}(\underline{t})D_\kappa^{\bar{s}}(\underline{t}')\Big\rangle a_\mu^r G_s(t-t')a_\kappa^s\sigma(t')G_s^\dagger(t-t') \tag{1.65}$$

The time dependence of the operators in the correlation function can be transferred to one operator since the bath Hamiltonian is time independent and commutes with ρ_b. Again we use the invariance of the partial trace under cyclic permutation of bath operators. The resulting bath correlation function is now only dependent on the time difference since the starting point of the Heisenberg evolution t_0 has dropped out.

$$\Big\langle D_\mu^{\bar{r}}(\underline{t})D_\kappa^{\bar{s}}(\underline{t}')\Big\rangle = \mathrm{Tr}_b\Big\{e^{iH_b(t-t_0)}D_\mu^{\bar{r}}e^{-iH_b(t-t_0-(t'-t_0))}D_\kappa^{\bar{s}}\underbrace{e^{-iH_b(t-t_0)}\rho_b}_{[H_b,\rho_b]=0]}\Big\} \tag{1.66}$$

$$= \Big\langle D_\mu^{\bar{r}}(t-t')D_\kappa^{\bar{s}}\Big\rangle \tag{1.67}$$

We now evaluate this bath correlation function - see equation (1.3) for the definition of D^s_μ. As particle number, momentum and spin are conserved in the bath Hamiltonian (no superconducting), we have $\delta_{s,\overline{r}}, \delta_{k,k'}$ respectively $\delta_{\tau,\tau'} \Rightarrow \delta_{\mu_\tau,\kappa_\tau}$. As different baths are not correlated we also have $\delta_{\alpha,\alpha'}$ The primes refer to the second operator D_κ.

$$\Big\langle D^{\overline{r}}_\mu(t-t')D^{\overline{s}}_\kappa \Big\rangle = \sum_{\substack{\alpha k\tau \\ \alpha' k'\tau'}} t^s_{\alpha\mu k\tau} t^{\overline{s}}_{\alpha'\kappa k'\tau'} \Big\langle d^s_{\alpha k\tau}(t-t')d^{\overline{s}}_{\alpha' k'\tau'} \Big\rangle \delta_{\alpha,\alpha'}\delta_{k,k'}\delta_{\tau,\tau'}\delta_{\mu_\tau,\kappa_\tau}\delta_{\tau,\mu_\tau} \tag{1.68}$$

$$C^s_{\mu\kappa}(t-t') := e^{-0^+|t-t'|}\Big\langle D^s_\mu(t-t')D^{\overline{s}}_\kappa \Big\rangle \delta_{\alpha,\alpha'}\delta_{k,k'}\delta_{\tau,\tau'} \tag{1.69}$$

With a minus sign resulting from the term rs we get:

$$\mathrm{Tr}_b\Big\{T_1\Big\} = \sum_{\mu\kappa s} C^s_{\mu\kappa}(t-t')a^{\overline{s}}_\mu \cdot \Big(\mathcal{G}_s(t-t')a^s_\kappa \sigma(t')\Big) \tag{1.70}$$

For T_3 we proceed analogously,

$$\mathrm{Tr}_b\Big\{T_3\Big\} := \mathrm{Tr}_b\Big\{G_s(t-t')H^b_1(\underline{t}')\rho(t')G^\dagger_s(t-t')H^b_1(\underline{t})\Big\} \tag{1.71}$$

$$= e^{-0^+(|t|+|t'|)} \sum_{\mu\kappa rs} rs G_s(t-t')\mathrm{Tr}_b\Big\{a^s_\kappa D^{\overline{s}}_\kappa(\underline{t}')\rho_b\sigma(t')G^\dagger_s(t-t')a^r_\mu D^{\overline{r}}_\mu(\underline{t})\Big\} \tag{1.72}$$

We use the same argument for the adiabatic convergence term and the conservation of particle number $\delta_{s,\overline{r}}$. But in contrast to T_1 the system operator σ has to interchange position with a bath operator thus it's commutation relation, which isn't specified yet, is of importance. As mentioned in section (1.1.3), we assume here the operator σ to be either of fermionic $\sigma := a^s_j\sigma_s, (\eta = -1)$ or of bosonic $\sigma := \sigma_s, (\eta = +1)$ type, with σ_s denoting the actual reduced density matrix of the system. Depending on this statistical behaviour we get the sign factor η. Using the cyclic permutation invariance in the bath trace and equation (1.69) we end up with

$$\ldots = -\eta e^{-0^+|t-t'|} \sum_{\mu\kappa rs} rs \underbrace{\mathrm{Tr}_b\Big\{D^{\overline{s}}_\kappa(\underline{t}')\rho_b D^{\overline{r}}_\mu(\underline{t})\Big\}}_{=\langle D^{\overline{r}}_\mu(\underline{t})D^{\overline{s}}_\kappa(\underline{t}')\rangle} G_s(t-t')a^s_\kappa\sigma(t')G^\dagger_s(t-t')a^r_\mu \tag{1.73}$$

$$\mathrm{Tr}_b\Big\{T_3\Big\} = \eta\sum_{\mu\kappa s} C^s_{\mu\kappa}(t-t')\Big(\mathcal{G}_s(t-t')a^s_\kappa\sigma(t')\Big)\cdot a^{\overline{s}}_\mu \tag{1.74}$$

Now with both terms T_1 (1.70) and T_3 the integral in equation (1.55) can be summarized by a commutator $[\,\cdot\,,\cdot\,]_-$ respectively an anticommutator $[\,\cdot\,,\cdot\,]_+$

$$\int_{t_0}^t dt' \mathrm{Tr}_b\Big\{T_1 - T_3\Big\} = \sum_{\mu\kappa s}\int_{t_0}^t dt' C^s_{\mu\kappa}(t-t')\Big[a^{\overline{s}}_\mu, \mathcal{G}_s(t-t')a^s_\kappa\sigma(t')\Big]_{-\eta}. \tag{1.75}$$

Now we move the integral and the summation over κ into the right side of the commutator and define this as the **correlation operator** $A^s_{\mu\sigma(t')}(t)$ which will make notation more compact as our next approximations will all act on this operator. The resulting equations are presented in the following box and will serve as starting point for the next approximations:

BORN MASTER EQUATION

$$\dot{\sigma}(t) = -i\mathcal{L}_s\sigma(t) - \sum_{\mu s}\left[a^{\bar{s}}_{\mu}, A^{s}_{\mu\sigma(t')}(t)\right]_{-\eta} + h.c.(\ddagger) \tag{1.76}$$

$$A^{s}_{\mu\sigma(t')}(t) = \int_{t_0}^{t} dt' \sum_{\kappa} C^{s}_{\mu\kappa}(t-t')\mathcal{G}_s(t-t')a^{s}_{\kappa}\sigma(t') \tag{1.77}$$

$$C^{s}_{\mu\kappa}(t-t') = e^{-0^+|t-t'|}\sum_{\alpha k\tau} t^{s}_{\alpha\mu k\tau}t^{\bar{s}}_{\alpha\kappa k\tau}\langle d^{s}_{\alpha k\tau}(t-t')d^{\bar{s}}_{\alpha k\tau}\rangle$$

1.4.4 Bath correlation function for non-interacting thermally equilibrated leads

Now we want to have a closer look at the previously introduced bath correlation function

$$C^{s}_{\mu\kappa}(t) = \sum_{\alpha} C^{s}_{\alpha\mu\kappa}(t) \tag{1.78}$$

$$C^{s}_{\alpha\mu\kappa}(t) = e^{-0^+|t|}\sum_{k\tau} t^{s}_{\alpha\mu k\tau}t^{\bar{s}}_{\alpha\kappa k\tau}\langle d^{s}_{\alpha k\tau}(t)d^{\bar{s}}_{\alpha k\tau}\rangle \tag{1.79}$$

We can assume the baths to stay in thermal equilibrium since the coupling via the system to the other bath is assumed to be small. Thus we can assign them a temperature and a potential μ_α and describe the reduced bath density operator

$$\rho_b = \frac{e^{-\beta(H_b-\mu_\alpha\hat{N})}}{Z_{\text{leads}}} \tag{1.80}$$

in the grand-canonical ensemble with the grand-canonical partition function Z_{leads} as normalization factor and the particle number operator $\hat{N} = \sum_{\alpha k\tau} d^{\dagger}_{\alpha k\tau}d_{\alpha k\tau} = \sum_{\alpha k\tau}\hat{n}_{\alpha k\tau}$.

For the correlation function of the bath operators we can now evaluate the expectation value in the eigenbasis of the bath Hamiltonian $H_b|n\rangle = \epsilon_n|n\rangle$. (Note, that the creation/annihilation bath operators $d^{s}_{\alpha k\tau}$ in the interaction picture just depend on the time difference t since t_0 dropped out.)

$$\langle d^{s}_{\alpha k\tau}(t)d^{\bar{s}}_{\alpha k\tau}\rangle = \text{Tr}_b\Big\{d^{s}_{\alpha\kappa\tau}(t)d^{\bar{s}}_{\alpha\kappa\tau}\rho_b\Big\} = \sum_n\langle n|\frac{e^{-\beta(H_b-\mu_\alpha\hat{N})}}{Z_{\text{leads}}}e^{iH_bt}d^{s}_{\alpha k\tau}e^{-iH_bt}d^{\bar{s}}_{\alpha k\tau}|n\rangle \tag{1.81}$$

Now we use the following commutation relation of the fermionic bath operators with functions of the bath Hamiltonian H_b:

$$[d^{s}_{\alpha k\tau}, H_b] = \sum_{k'\alpha'\tau'}[d^{s}_{\alpha k\tau}, \epsilon_{k'}\hat{n}_{\alpha' k'\tau'}] = -s\epsilon_k d^{s}_{\alpha k\tau}\cdot\delta_{\alpha\alpha'}\delta_{kk'}\delta_{\tau\tau'}$$

$$d^{s}_{\alpha k\tau}\cdot f(H_b) = f(H_b - s\epsilon_k)\cdot d^{s}_{\alpha k\tau} \tag{1.82}$$

So we get for the correlation function

$$\langle d^s_{\alpha k\tau}(t) d^{\bar{s}}_{\alpha k\tau}\rangle = e^{is\epsilon_k t}\sum_n \langle n|\frac{e^{-\beta(H_b-\mu_\alpha \hat{N})}}{Z_{\text{leads}}}\underbrace{d^s_{\alpha k\tau} d^{\bar{s}}_{\alpha k\tau}}_{\hat{n}^s_{\alpha k\tau}}|n\rangle = e^{is\epsilon_k t}\langle \hat{n}^s_{\alpha k\tau}\rangle \tag{1.83}$$

Using the invariance under cyclic permutation of the trace and equation (1.82) we get

$$\langle \hat{n}^s_{\alpha k\tau}\rangle = \frac{1}{Z_{\text{leads}}}\sum_n \langle n| d^{\bar{s}}_{\alpha k\tau} e^{-\beta\mathcal{H}} d^s_{\alpha k\tau}|n\rangle \tag{1.84}$$

$$= \frac{1}{Z_{\text{leads}}}\sum_n \langle n| e^{-\beta(\mathcal{H}-\bar{s}(\epsilon_k-\mu_\alpha))}\underbrace{d^{\bar{s}}_{\alpha k\tau} d^s_{\alpha k\tau}}_{=1-d^s_{\alpha k\tau}d^{\bar{s}}_{\alpha k\tau}}|n\rangle \tag{1.85}$$

$$\langle \hat{n}^s_{\alpha k\tau}\rangle = e^{-s\beta(\epsilon_k-\mu_\alpha)}(1-\langle \hat{n}^s_{\alpha k\tau}\rangle) \tag{1.86}$$

So we have derived the generalized Fermi function

$$n^s_\alpha(\epsilon_k) := \langle \hat{n}^s_{\alpha k\tau}\rangle = \frac{1}{1+e^{s\beta(\epsilon_k-\mu_\alpha)}} \tag{1.87}$$

Now with inserting a delta distribution to transform the sum into an integral over the real axis $\sum_k \Rightarrow \int d\omega\delta(\epsilon_k-\omega)$, we introduce the convenient spectral density function $S^s_{\alpha\mu\kappa}(\omega)$ that we call in combination with the generalized Fermi function $n^s_\alpha(\omega)$ the bath density function $\Gamma^s_{\alpha\mu\kappa}(\omega)$ and can express the bath correlation function for a bath α as follows:

Bath Correlation Function
for a non-interacting thermally equilibrated lead

$$C^s_{\alpha\mu\kappa}(t) = e^{-0^+|t|}\sum_{k\tau} t^s_{\alpha\mu k\tau} t^{\bar{s}}_{\alpha\kappa k\tau} e^{is\epsilon_k t} n^s_\alpha(\epsilon_k) \tag{1.88}$$

$$= e^{-0^+|t|}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\Gamma^s_{\alpha\mu\kappa}(\omega)e^{is\omega t} \tag{1.89}$$

$$\Gamma^+_{\alpha\mu\kappa}(\omega) = n^+_\alpha(\omega)\Gamma_{\alpha\kappa\mu}(\omega), \qquad \Gamma^-_{\alpha\mu\kappa}(\omega) = n^-_\alpha(\omega)\Gamma_{\alpha\mu\kappa}(\omega) \tag{1.90}$$

$$\Gamma_{\alpha\mu\kappa}(\omega) = 2\pi\sum_{k\tau} t_{\alpha\mu k\tau} t^*_{\alpha\kappa k\tau}\delta(\epsilon_k-\omega) \tag{1.91}$$

$$\mathcal{F}[C^s_{\alpha\mu\kappa}(t)] = 2\sum_{k\tau} t^s_{\alpha\mu k\tau} t^{\bar{s}}_{\alpha\kappa k\tau}\underbrace{\frac{0^+}{(s\omega-s\epsilon_k)^2+(0^+)^2}}_{\xrightarrow{0^+\to 0}\pi\delta(\omega-\epsilon_k)} n^s_\alpha(\omega) \tag{1.92}$$

$$\left(\Gamma^s_{\alpha\mu\kappa}(\omega)\right)^\dagger = \Gamma^s_{\alpha\kappa\mu}(\omega), \qquad \forall\omega\in\mathbb{R} \tag{1.93}$$

The bath density function $\Gamma^s_{\alpha\mu\kappa}$ can be interpreted as the Fourier transform (s dependent integral kernel $e^{is\omega t}$) of the bath correlation function without the adiabatic switch-on factor.

A Fourier transform including the adiabatic switch-on term would anyway result in a Lorentz peak converging to a delta distribution.

As we will see later from the Markov approximation we will consider times $t > 0$, thus we can combine the adiabatic switch on factor with omega: $\omega^+ = \omega + i0^+$.

1.4.5 Decay behaviour of the bath correlation function in the wide band limit

We want to get an analytic estimation for the time-dependent bath correlation function in the wide band limit ($\Gamma^s_{\alpha\mu\kappa}(\omega) = \Gamma^s_{\alpha\mu\kappa} n^s_\alpha(\omega)$) assuming the spectral density function $S^s_{\alpha\mu\kappa}$ for each bath α to be constant. Since the integral of the generalized Fermi function diverges in one direction we introduce the convergence factor $e^{-\eta|\omega|}$, with taking the limit $\eta \to 0^+$ in the end.

$$C^s_{\mu\kappa\alpha}(t) = \Gamma^s_{\alpha\mu\kappa} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{e^{is\omega t}}{1 + e^{s\beta(\omega - \mu_\alpha)}} = \Gamma^s_{\alpha\mu\kappa} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{e^{i\omega t - \eta|\omega|}}{1 + e^{\beta(\omega - s\mu_\alpha)}} \tag{1.94}$$

This integral of the form

$$\int_{-\infty}^{\infty} d\omega \frac{e^{i\omega t - \eta|\omega|}}{1 + e^{\beta\omega}} = 2\pi i \sum_{\omega_0} \mathrm{Res}(f, \omega_0) \tag{1.95}$$

can be solved using the residue theorem, assuming that the integral in the upper complex plain $\omega = Re^{i\varphi}, \varphi \in [0, \pi]$ vanishes for large R. The complex roots of the denominator in the upper complex plane are the Matsubara frequencies $\omega_0 = i\frac{2\pi}{\beta}(n + \frac{1}{2}), n \in \mathbb{N}_0$. A Taylor expansion of the denominator around ω_0 leads to:

$$1 + e^{\beta\omega} = \underbrace{1 + e^{\beta\omega_0}}_{=0} + \beta \underbrace{e^{\beta\omega_0}}_{=-1}(\omega - \omega_0) + \frac{\beta^2}{2!}(-1)(\omega - \omega_0)^2 + \dots = (\omega - \omega_0)(-\beta \dots) \tag{1.96}$$

So the residues of the integrand f can be calculated

$$\mathrm{Res}(f, \omega_0) = \lim_{\omega \to \omega_0} (\omega - \omega_0) \frac{e^{i\omega t - \eta|\omega|}}{1 + e^{\beta\omega}} \tag{1.97}$$

$$= \lim_{\omega \to \omega_0} \frac{e^{i\omega t - \eta|\omega|}}{-\beta - \frac{\beta^2}{2!}(\omega - \omega_0) - \dots} \tag{1.98}$$

$$= -\frac{e^{i\omega_0 t - \eta|\omega_0|}}{\beta} = -\frac{e^{-\frac{2\pi}{\beta}(n + \frac{1}{2})t - \eta|\omega_0|}}{\beta} \tag{1.99}$$

Thus the solution yields

$$\int_{-\infty}^{\infty} f(\omega) d\omega = -2\pi i \sum_{n \geq 0} \frac{e^{-\frac{2\pi}{\beta}(n + \frac{1}{2})t}}{\beta} = -\frac{2\pi i}{\beta} e^{-\frac{\pi t}{\beta}} \frac{1}{1 - e^{-\frac{2\pi t}{\beta}}} = -\frac{\pi i}{\beta} \frac{1}{\sinh(\frac{\pi t}{\beta})} \tag{1.100}$$

So we have as a result for the bath correlation function in the wide band limit

Decaying behaviour of the bath correlation function in the wide band limit

$$C^s_{\mu\kappa\alpha}(t) \approx \frac{1}{\sinh(\frac{\pi t}{\beta})} \tag{1.101}$$

So we indeed see, that the bath correlation function is strongly peaked around $t = 0$ and decays with approximately $\frac{\Gamma}{t}$. Figure 1.4.5 illustrates the relaxation of a random initial state calculated in the Born Markov master equation approach in contrast to the corresponding bath correlation function. The fact that this function is peaked around zero will allow to regard the system as Markovian and perform further approximations.

Laplace transform of the bath correlation function in the wide band limit

For the Laplace transform of the bath correlation function we can use the dispersive relation. We only have to take care of the sign factor s since it was used to define the Fourier transform of $C^s_{\alpha\mu\kappa}(t)$

$$\begin{aligned} C^s_{\alpha\mu\kappa}(\omega) &= \int_0^\infty dt e^{i\omega t} C_{\alpha\mu\kappa}(t) = \int_{-\infty}^\infty \frac{d\omega'}{2\pi} \Gamma^s_{\alpha\mu\kappa}(\omega') \int_0^\infty dt e^{i\omega t} e^{i(s\omega' + i0^+)t} \\ &= \int_{-\infty}^\infty \frac{d\omega'}{2\pi} \Gamma^s_{\alpha\mu\kappa}(\omega') \frac{i}{\omega + s\omega' + i0^+} \end{aligned}$$

Applying again the wide band limit, assuming the spectral distribution to be flat and constant reveals with some calculations (see also 4.2)

Laplace transform of the Bath correlation function in the wide band limit

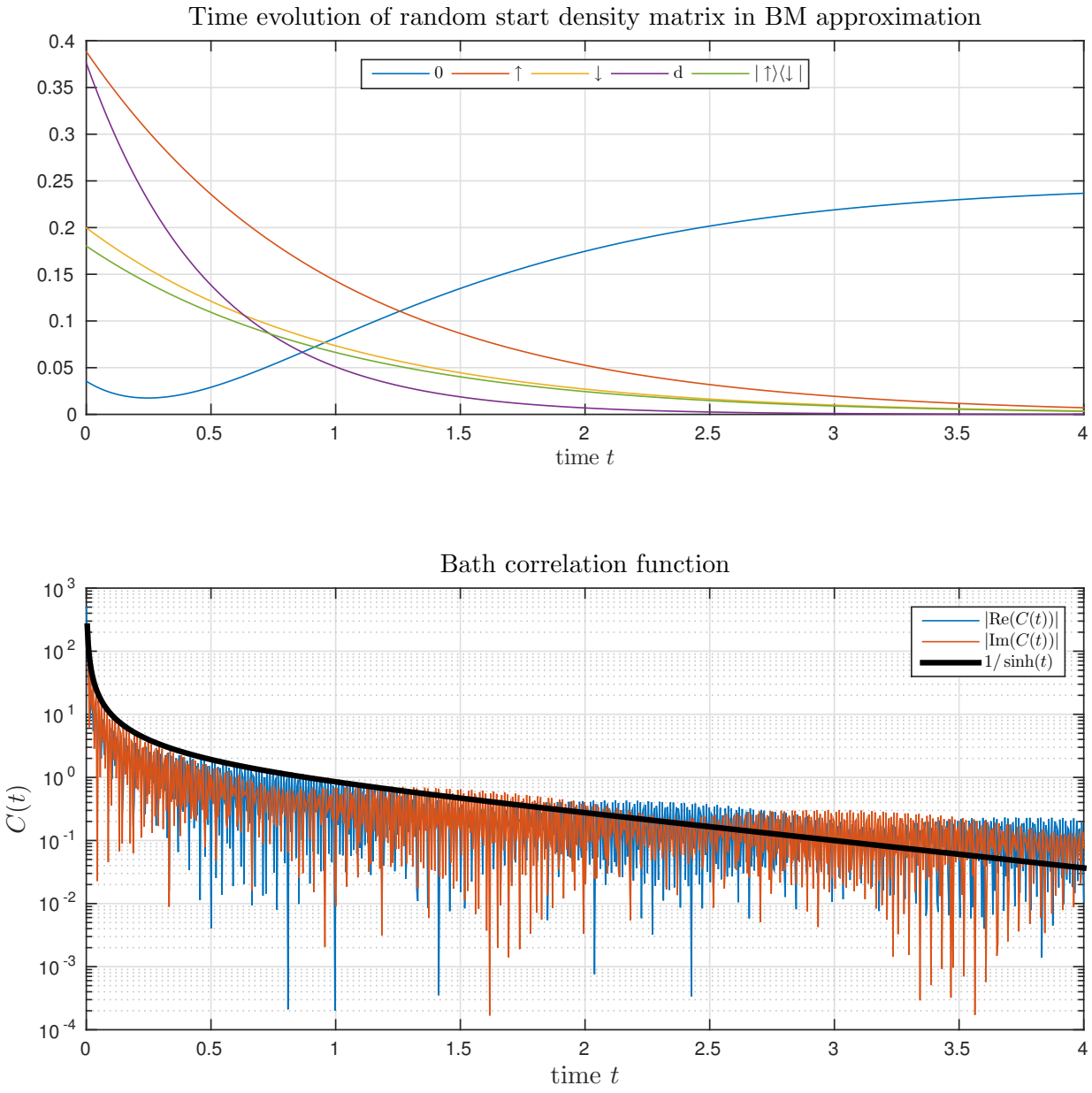
$$C^s_{\alpha\mu\kappa}(\omega) = \Gamma^{(s)}_{\alpha\mu\kappa} \left(\frac{1}{2} n^s_\alpha(-s\omega) + \frac{i}{2\pi} \mathrm{Re}\left[\Psi\left(\frac{1}{2} + i \frac{\beta(\omega + s\mu_\alpha)}{2\pi} \right) \right] \right) \tag{1.102}$$

with $\Psi(z)$ the digamma function.

1.4.6 Markov approximation

If we look at the correlation operator in the Born master equation (1.77) in it's present form, we see that the (modified) density operators is involved at different times (t') (convolution), thus making it's evaluation quite complicated.

$$A^s_{\mu\sigma(t')}(t) = \sum_\alpha \int_{t_0}^t dt' C^s_{\alpha\mu\kappa}(t - t') e^{-iH_s(t-t')} a^s_\kappa \sigma(t') e^{iH_s(t-t')}$$



quantum dot: $U = 10$, $\beta = 10$, $\Gamma = 0.5$, $\xi_0 = 12$, $V_b = 4$

Figure 1.1: In the upper panel there is given the calculated time evolution of the occupation densities of a strong interacting quantum dot in the Born-Markov approach. In the second panel the calculated correlation time $C_(t)$ and is it's envelope an inverse sinus hyperbolicus is plotted.

Our aim is to simplify this time dependency and get a correlation operator that contains the (modified) density operator just at one time.

$$C^s_{\alpha\mu\kappa}(t) = e^{-0^+|t|} \sum_{k\tau} t^s_{\alpha\mu k\tau} t^{\bar{s}}_{\alpha\kappa k\tau} e^{is\epsilon_k t} n^s_\alpha(\epsilon_k) \tag{1.103}$$

$$= e^{-0^+|t|} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma^s_{\alpha\mu\kappa}(\omega) e^{is\omega^+ t} \tag{1.104}$$

Thus we perform a variable transformation as most of the factors just depend on the time difference $t - t'$

$$\tau := t - t', \qquad \tau \in (t - t_0, 0), \qquad dt' = -d\tau \tag{1.105}$$

and we set $t_0 = -\infty$ as we have switched on the interaction adiabatically at that time. After switching the integration limits we have for the correlation operator:

$$A^s_{\mu\sigma(t-\tau)}(t) = \sum_{\alpha\kappa} \int_0^\infty d\tau C^s_{\alpha\mu\kappa}(\tau) \mathcal{G}_s(\tau) a^s_\kappa \sigma(t - \tau) \tag{1.106}$$

This equation will be the starting point for two slightly different approximations, the first one the so called **Markov approximation**, whereas later we will develop a self-consistent approach from this equation (see section 2.1). Main idea of the Markov approximation is to claim that the dynamics $\dot{\sigma}(t)$ of the (modified) density operator σ are just depending on it's present state $\dot{\sigma}(t) \propto \sigma(t)$ with no time propagator $\mathcal{G}_s$ acting on it anymore. To see the details we write out the propagators and insert a unity operator $\mathbb{1} = e^{iH_s\tau} e^{-iH_s\tau}$ at the left side of σ:

$$A^s_{\mu\sigma(t-\tau)}(t) = \int_0^\infty d\tau C^s_{\alpha\mu\kappa}(\tau) e^{-iH_s\tau} a^s_\kappa e^{iH_s\tau} \underbrace{e^{-iH_s\tau} \sigma(t - \tau) e^{iH_s\tau}}_{=\mathcal{G}_s(\tau)\mathcal{U}(t-\tau,-\infty)\rho(-\infty)} \tag{1.107}$$

The Markov approximation states that

$$\mathcal{G}_s(\tau)\sigma(t - \tau) \approx \sigma(t) \qquad \big(= \underbrace{\mathcal{U}(\tau)\mathcal{U}(t - \tau, -\infty)}_{\mathcal{U}(t,-\infty)} \sigma(-\infty) \big) \tag{1.108}$$

Note that due to the Born approximation we can always factorize our full density operator ρ into a system and a constant bath part, but the time evolution of the whole operator cannot be described by a restriction just to the system. The crucial difference of full evolution and system evolution is the interplay with the bath that for instance allows to change the occupation densities in the system which in an isolated environment would remain constant. As this coupling to the bath is assumed to be weak (weak coupling limit [35]) the interaction with the bath happens in such a slow way, that for small values of τ we can neglect it.

Secondly that approximation is justified for bigger values of τ since the bath correlation term in the integral in equation (1.77) is sharply peaked at $\tau = 0$ (see section 1.4.5), thus cancelling eventual errors.

The Markovian property of the system can be explained physically by the weak coupling and the thermal stabilization of the baths. Any excitation that enters a bath dissipates on a much shorter time-scale than the interaction time scale. So the baths cannot give any feedback.

Here we summarize the formulas for the Born-Markov master equation:

BORN-MARKOV MASTER EQUATION

$$\dot{\sigma}(t) = -i\mathcal{L}_s\sigma(t) - \sum_{\mu s}\left[a_\mu^{\bar{s}}, A^s_{\mu\sigma(t)}(t)\right]_{-\eta} + h.c.(\ddagger) \tag{1.109}$$

$$A^s_{\mu\sigma(t)}(t) = \int_0^\infty d\tau \sum_{\alpha\kappa} C^s_{\alpha\mu\kappa}(\tau) e^{-iH_s\tau} a^s_\kappa e^{iH_s\tau}\sigma(t)$$

$$C^s_{\alpha\mu\kappa}(\tau) = \int_{-\infty}^\infty \frac{d\omega}{2\pi}\Gamma^s_{\alpha\mu\kappa}(\omega) e^{is\omega^+\tau}$$

Note that this derivation is done also for the modified density operator $\tilde{\sigma}$ (dependent on η and $\ddagger$). The adiabatic switch-on parameter is hidden in $\omega^+ = \omega + i0^+$.

1.5 Evaluation of the Born-Markov master equation

1.5.1 Steady state

The Born-Markov master equation gives us a formula for the time derivative of the reduced system matrix σ_s ($\eta = +$). Our main goal is to calculate the steady state $\overline{\sigma}$ of the quantum system. As we assume our setup to be ergodic we can say that we are in steady state at $t \to \infty$. The property of a steady state $\dot{\overline{\sigma}} = 0$ will help us to develop a closed set of equations to solve it.

The following box contains the formulas to which we will refer later when calculating the steady state and the steady state current.

STEADY STATE IN THE BORN-MARKOV MASTER EQUATION FOR THE REDUCED SYSTEM DENSITY OPERATOR σ_s

$$0 = -i\mathcal{L}_s\overline{\sigma} - \sum_{\mu s}\left[a^{\bar{s}}_{\mu}, A^{s}_{\mu\overline{\sigma}}\right]_- + h.c.(\dagger) \tag{1.110}$$

$$A^{s}_{\mu\overline{\sigma}} = \int_0^\infty d\tau \sum_{\alpha\kappa} C^{s}_{\alpha\mu\kappa}(\tau) e^{-iH_s\tau} a^{s}_{\kappa} e^{iH_s\tau}\overline{\sigma} \tag{1.111}$$

$$A^{s}_{\mu\overline{\sigma}} = \int_{-\infty}^\infty \frac{d\omega}{2\pi} \sum_{\alpha\kappa} \Gamma^{s}_{\alpha\mu\kappa}(\omega)\left(\mathcal{G}_s(s\omega)a^{s}_{\kappa}\right)\overline{\sigma} \tag{1.112}$$

There is also an alternative representation of the correlation operator $A^{s}_{\mu\overline{\sigma}}$ using the Fourier representation of the bath correlation function and containing the Laplace transform of the time propagator $\mathcal{G}_s(s\omega)$. Note that the sign factor s is present now in the argument of the superpropagator.

$$\int_0^\infty d\tau C^{s}_{\mu\kappa}(\tau)\mathcal{G}_s(\tau) = \int_0^\infty d\tau \int_{-\infty}^\infty \frac{d\omega}{2\pi}\Gamma^{s}_{\mu\kappa}(\omega)e^{is\omega^+\tau}\mathcal{G}_s(\tau) \tag{1.113}$$

$$= \int_{-\infty}^\infty \frac{d\omega}{2\pi}\Gamma^{s}_{\mu\kappa}(\omega)\underbrace{\int_0^\infty d\tau e^{is\omega^+\tau}\mathcal{G}_s(\tau)}_{\mathcal{G}_s(s\omega)} \tag{1.114}$$

1.5.2 Evaluation in system eigenbasis

Now we want to evaluate this Born-Markov master equation for the reduced system density operator σ_s in the system eigenbasis consisting of eigenstates $H_s|a\rangle = E_a|a\rangle$ using the notation: $\sigma_{ab} := \langle a|\sigma_s|b\rangle$. We start by rewriting the steady state solution:

$$0 = -i\mathcal{L}_s\overline{\sigma} - \sum_{\mu\kappa s}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\Gamma^s_{\mu\kappa}(\omega)\left[a^{\overline{s}}_\mu, \int_0^\infty d\tau e^{is\omega^+\tau}e^{-iH_s\tau}a^s_\kappa e^{iH_s\tau}\overline{\sigma}\right] + h.c.(\dagger) \tag{1.115}$$

To derive an equation of the form $\dot{\sigma} = K\sigma$ we insert unity operators $\mathbb{1} = \sum_a |a\rangle\langle a|$ on the left and right side of the density operator σ_s.

The commutator then becomes:

$$\sum_{ab}\left[a^{\overline{s}}_\mu, \int_0^\infty d\tau e^{i(s\omega - H_s + i0^+)\tau}a^s_\kappa e^{iE_a\tau}|a\rangle\sigma_{ab}\langle b|\right] \tag{1.116}$$

$$= \sum_{ab}\left[a^{\overline{s}}_\mu, \frac{i}{s\omega - H_s + E_a + i0^+}a^s_\kappa|a\rangle\sigma_{ab}\langle b|\right] \tag{1.117}$$

$$= i\sum_{abcd}|c\rangle\underbrace{\langle c|a^{\overline{s}}_\mu|d\rangle}_{=:Q^{\overline{s}}_{\mu,cd}}(s\omega - \underbrace{(E_d - E_a)}_{E_{da}} + i0^+)^{-1}\underbrace{\langle d|a^s_\kappa|a\rangle}_{Q^s_{\kappa,da}}\sigma_{ab}\langle b| \tag{1.118}$$

$$-|d\rangle(s\omega - E_{da} + i0^+)^{-1}Q^s_{\kappa,da}\sigma_{ab}Q^{\overline{s}}_{\mu,bc}\langle c| \tag{1.119}$$

Here we have introduced the handy Q-matrices Q^s_κ which represent the eigenbasis representation of the system annihilators/creators $Q^s_{\kappa,da} := \langle d|a^s_\kappa|a\rangle$. We still have to evaluate the frequency integral in equation (1.115), that will be a function of the system energy gap E_{da}. We label this function $F^s_{\mu\kappa}$ and note that all information about the bath and applied potentials are encoded in this expression via the spectral density function $S^s_{\alpha\mu\kappa}$ and the generalized Fermi functions n^s_α both combined in the bath density function $\Gamma^s_{\alpha\mu\kappa}(\omega) = S^s_{\alpha\mu\kappa}(\omega)n^s_\alpha(\omega)$.

Born-Markov master equation in the system eigenbasis

$$0 = -i\mathcal{L}_s\overline{\sigma} - \sum_{\substack{\mu\kappa s\\ abcd}}(F^s_{\mu\kappa})_{da}\left(Q^{\overline{s}}_{\mu,cd}Q^s_{\kappa,da}\sigma_{ab}|c\rangle\langle b| - Q^s_{\kappa,da}\sigma_{ab}Q^{\overline{s}}_{\mu,bc}|d\rangle\langle c|\right) + h.c. \tag{1.120}$$

$$(F^s_{\mu\kappa})_{da} = \sum_\alpha F^s_{\alpha\mu\kappa}(E_{da}), \qquad F^s_{\alpha\mu\kappa}(E_{da}) = i\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\frac{S^s_{\alpha\mu\kappa}(\omega)n^s_\alpha(\omega)}{s\omega - E_{da} + i0^+} \tag{1.121}$$

1.5.3 Effective bath coupling in the wide band limit

We call this function $F^s_{\alpha\mu\kappa}$ the effective bath coupling function as it inherits all information about the bath and is just dependent on the systems energy differences ΔE. Throughout this work we assumed the spectral density function $S(\omega)$ occurring in the integral to be flat respectively constant. We label this coupling constants with $\Gamma^{(s)}_{\alpha\mu\kappa}$ without argument. In this so-called wide band limit the integral has an analytic solution derived in the appendix 4.2.

$$F^s_\alpha(E) = i\Gamma_{\alpha\mu\kappa}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\frac{n^s_\alpha(\omega)}{s\omega - E + i0^+} = \frac{\Gamma_{\alpha\mu\kappa}}{2}\left(n^s_\alpha(sE) + \frac{i}{\pi}\mathrm{Re}\left[\Psi\left(\frac{1}{2} + i\frac{\beta(E - s\mu_\alpha)}{2\pi}\right)\right]\right) \tag{1.122}$$

with $\Psi(z)$ the digamma function, see figure 4.1.

1.6 Secular Approximation and Lindblad formalism

In this section we want to discuss the issue how it is guaranteed that we have indeed a the steady state solution, which represents a valid reduced density matrix of our system. The demanded properties imply certain restrictions on our Born-Markov master equation which represents the evolution of some starting state. If this state at t_0 was valid then it's sufficient to demand that this evolution preserves these features. So if the Born-Markov master equations conserves trace, hermiticity and positivity of any given initial state, then the steady state, which is reached for $t \to \infty$ will be valid.

A closer look at the the given equations let us recognize that hermiticity and trace preservation are already guaranteed, compare [33]. The challenging part is to check for positivity. This will lead us to the now discussed secular approximation that is the restriction necessary to transform the Born-Markov master equation into the Lindblad formalism, which itself embodies the generalization such valid dynamical maps [45].

1.6.1 Lindblad formalism

Here I give the definition of a Lindblad form following [33]:

Definition 1.1

(Lindblad Form) A master equation of Lindblad form to describe the time evolution of a quantum system represented by the density operator ρ has the following structure:

$$\dot\rho(t) = -i[H, \rho(t)] + \sum_{\alpha\beta=1}^{N^2-1}\gamma_{\alpha\beta}\Big(L_\alpha\rho(t)L^\dagger_\beta - \frac{1}{2}\Big\{L^\dagger_\beta L_\alpha, \rho(t)\Big\}\Big) \tag{1.123}$$

where the hermitian operator $H = H^\dagger$ can be interpreted as an effective Hamiltonian and $\gamma_{\alpha\beta} = \gamma^*_{\beta\alpha}$ is a positive semi-definite matrix, that means it fulfils $\sum_{\alpha\beta} x^*_\alpha\gamma_{\alpha\beta}x_\beta \geq 0$ for

all elements x of the according Hilbert space (or, equivalently that the spectrum of $(\gamma_{\alpha\beta})$ is non-negative.

As mentioned in the introduction of the section the Lindblad form is the generalized form of a Markovian master equation that guarantees the conservation of trace, hermiticity and positivity of the evolved density operator. Our aim will it now be to match our Born-Markov master equation into a Lindblad form to gain these desired features.

1.6.2 Secular Approximation

In order to transform equation (1.120) to Lindblad form we write out the hermitian conjugate, where we can relabel the eigenbases to match the first expression[2]:

$$\dot{\sigma}_s(t) = -i\mathcal{L}_s\sigma_s(t) - \sum_{\substack{\mu\kappa s\\ abcd}} F^s_{\mu\kappa}(E_{da})\left(Q^{\bar{s}}_{\mu,cd}Q^s_{\kappa,da}\sigma_{ab} - Q^s_{\kappa,da}\sigma_{ab}Q^{\bar{s}}_{\mu,bc}\right) \tag{1.124}$$

$$+ \overline{F^s_{\mu\kappa}(E_{bc})}\left(\sigma_{ab}Q^{\bar{s}}_{\kappa,bc}Q^s_{\mu,cd} - Q^s_{\mu,da}\sigma_{ab}Q^{\bar{s}}_{\kappa,bc}\right) \tag{1.125}$$

From this we recognize that if the energy differences E_{da} and E_{bc} are the same we can condense the bath coupling functions F and get

$$\dot{\sigma}_s(t) = -i\mathcal{L}_s\sigma_s(t) + \sum_{\substack{\mu\kappa s\\ abcd}} \left(F^s_{\mu\kappa}(E_{da}) + \overline{F^s_{\mu\kappa}(E_{bc})}\right)Q^s_{\kappa,da}\sigma_{ab}Q^{\bar{s}}_{\mu,bc}\delta_{E_{da}E_{bc}} \tag{1.126}$$

$$- \left(F^s_{\mu\kappa}(E_{da})Q^{\bar{s}}_{\mu,cd}Q^s_{\kappa,da}\sigma_{ab} + \overline{F^s_{\mu\kappa}(E_{bc})}\sigma_{ab}Q^{\bar{s}}_{\kappa,bc}Q^s_{\mu,cd}\right)\delta_{E_{da}E_{bc}} \tag{1.127}$$

Now we decompose the function $F^s_{\mu\kappa}(\omega) = N^s_{\mu\kappa}(\omega) + i\Lambda^s_{\mu\kappa}(\omega)$ in it's real and imaginary part. The first expression in the last equation can be expressed by the real part, whereas for the second expression we get an anticommutator for the real and a commutator for the imaginary part, which is added as so-called Lamb-Shift Hamiltonian to the Liouvillian before the sum.

$$\dot{\sigma}_s(t) = -i[H_s + H_{\mathrm{LS}}, \sigma_s(t)] \tag{1.128}$$

$$+ 2\sum_{\substack{\mu\kappa s\\ abcd}} N^s_{\mu\kappa}(E_{da})\left(Q^s_{\kappa,da}\sigma_{ab}Q^{\bar{s}}_{\mu,bc}\delta_{E_{da}E_{bc}} - \left\{Q^{\bar{s}}_{\mu,cd}Q^s_{\kappa,da}|c\rangle\langle a|, \sigma_s(t)\right\}\right) \tag{1.129}$$

$$H_{\mathrm{LS}} = \sum_{\substack{\mu\kappa s\\ acd}} \Lambda^s_{\mu\kappa}(E_{da})Q^{\bar{s}}_{\mu,cd}Q^s_{\kappa,da}|c\rangle\langle a| \tag{1.130}$$

[2]Here we have marked the dependence of the F functions from the Q-matrices in blue.

Now we can relabel the black Q-matrix in the commutator to match the black Q-matrix before to be able to extract them:

$$\dot{\sigma}_s(t) = -i[H_s + H_{\mathrm{LS}}, \sigma_s(t)] \tag{1.131}$$

$$+ \sum_{\substack{\mu\kappa s \\ abcd}} \underbrace{= 2N^s_{\mu\kappa}(E_{da}) Q^s_{\kappa,da} Q^{\overline{s}}_{\mu,bc} \delta_{E_{da}E_{bc}}}_{:=\gamma_{da,bc}} \left(|d\rangle\langle a|\sigma_s(t)|b\rangle\langle c| - \frac{1}{2}\Big\{ |b\rangle\langle c||d\rangle\langle a|, \sigma_s(t) \Big\} \right) \tag{1.132}$$

With identifying $|d\rangle\langle a| = L_\alpha$, $|b\rangle\langle c| = L^\dagger_\beta$ we have successfully transformed the master equation to Lindblad form.

The approximation that made this possible concerned the energy differences: $E_{da} = E_{bc}$. In connection with the first term in the Lindblad form this restricts the density operator to states with the same energy. This approximation is also called random phase approximation and can be motivated in the interaction picture by neglecting fast oscillating terms that average to zero. The frequency of those terms is determined by the energy difference of used eigenstates. For big energy gaps this approximation is justified, since the frequency is higher but for quasi-degenerate states this approach fails.

For practical application the secular approximation can be easily implemented by just regarding those entries in the density matrix that belong to eigenstates with the same energy, so it will be block-diagonal in all degenerate states. From here the step to a pure rate equation representing the diagonal of the density matrix doesn't seem far but as we will see with numerical examples in last chapter these effects of degenerate states play an important role to resolve for example interference effects.

1.6.3 Quasi-degenerate states - non secular approximation

As mentioned in the treatment of the secular approximation the occurrence of quasi-degenerate states that for instance may arise from slightly deviated on site-energies due to symmetry breaking lead contacts may be a problem for the correct description of the dynamics of such a system. In section 3.4.2 I numerically examine such a case realised by a triple quantum dot where interference effects of degenerate states are fundamental for the current characteristics.

To circumvent the possibly wrong results accomplished with the before introduced approximation one can simply not use it and solve the problem in the full configuration space (1.120), so we also consider density matrix elements of different energies though the block diagonal structure with respect to particle number and total spin will be still maintained due to the symmetry properties of the system Hamiltonian. The improvements and drawback of a non-positive density matrix are also discussed in 3.4.2.

2 Self-consistent Born approximation

In this section I give the derivation of a more sophisticated method compared to the Born-Markov master equation approach. The aim is to include the features of the level broadening in our current characteristics as they are not sufficient treated in the before introduced method. A more detailed discussion concerning level broadening is given in section 3.1.

In this derivation I follow the papers of Jin, Li, et. al. [26], [46] and present the current formula for an interacting quantum dot with spin dependent coupling, which has been also discussed with a similar method of Born-Markov master equation enhanced cluster perturbation theory (meCPT) in [32].

2.1 Self-consistent Born-Markov master equation

The main idea of this approach is to derive a self-consistent way to include the quite original problem in the solution of the problem. This recursive approach effectively re-includes higher-order diagrams that manifest for example in higher order tunnelling contributions which in turn result in the broadening effect, energy shift and the interplay between the coherent multiple tunnelling and the on-site strong Coulomb interaction that are essential to the Kondo effect[26] .

Starting point is the Born master equation (1.76) for the reduced system density operator σ_s where we already have set $t_0 = -\infty$, corresponding to adiabatically switching on the interaction. For the correlation operator $A^s_{\mu\sigma}$ we just have obtained right before applying the Markov approximation:

$$A^s_{\mu\sigma(t')}(t) = \int_{-\infty}^{t} dt' \sum_{\kappa} C^s_{\mu\kappa}(t-t')\mathcal{G}_s(t-t')a^s_\kappa\sigma(t') \tag{2.1}$$

The central object of that equation is

$$\mathcal{G}_s(t-t')[a^s_\kappa\sigma_s(t')] = \mathrm{Tr}_b\Big\{\mathcal{G}_0(t-t')[a^s_\kappa\rho_{bs}(t')]\Big\} \tag{2.2}$$

For each time t' we identify the term in the square brackets with the (reduced) modified density operator $\tilde{\rho}^s_\kappa$ as we have introduced them in section 1.1.3 at time zero.

$$\tilde{\rho}^s_\kappa(0) = [a^s_\kappa\rho_{bs}(t')] \tag{2.3}$$

$$\tilde{\sigma}^s_\kappa(0) = \mathrm{Tr}_b\Big\{a^s_\kappa\rho_{bs}(t')\Big\} = a^s_\kappa\sigma_s(t') = a^s_\kappa\mathcal{U}(t',-\infty)\sigma_s(-\infty) \tag{2.4}$$

Self-consistent approach

Now we need the time evolution of this object $\tilde{\sigma}$ which is currently done just with respect to the system. The key of the self-consistent approach is now to regard the object $\tilde{\rho}$ as time evolved by the full Hamiltonian H through exchanging the superoperator $\mathcal{G}_s(\tau) \to \mathcal{U}(\tau)$.

$$\tilde{\sigma}^s_\kappa(t) = \mathrm{Tr}_b\Big\{\tilde{\rho}^s_\kappa(t)\Big\} = \mathcal{U}(t)\tilde{\sigma}^s_\kappa(0) \tag{2.5}$$

$$\tilde{\rho}^s_\kappa(t) = \mathcal{G}(t)\tilde{\rho}^s_\kappa(0) \tag{2.6}$$

$$\tilde{\sigma}^s_\kappa(t) = \mathcal{U}(t)a^s_\kappa\mathcal{U}(t',-\infty)\sigma_s(-\infty) \tag{2.7}$$

To indicate this generalization to the full Hamiltonian in the correlation operator A we use the calligraphic $\mathcal{A}$:

$$\mathcal{A}^s_\mu(t) = \int_{-\infty}^{t} dt' \sum_\kappa C^s_{\mu\kappa}(t-t')\mathcal{U}(t-t')\tilde{\sigma}^s_\kappa(0), \qquad \tilde{\sigma}^s_\kappa(0) = a^s_\kappa\sigma_s(t') \tag{2.8}$$

In contrast to the Markov approximation done in section 1.4.6 at the same point, we gain here an equation that still depends on different times t'. We still need to know the reduced density matrix at all times $t' < t$ in order to get the system dynamics at time t. As the bath correlation function is strongly peaked at $t' \approx t$, we postulate (similar to the Markov approximation) that $\sigma_s(t')$ is not depending on the integration variable t' but on a fixed time $\bar{t}$ close to t.

We now perform a variable transformation $\tau = t - t'$ and show that for $t \to \infty$ also the density operator at time $\bar{t}$ converges into steady state $\sigma_s(\bar{t}) \to \overline{\sigma}$. For $t \to \infty$ we have

$$\sigma_s(\bar{t}) = \underbrace{\sigma_s(t)\Big|_{t\to\infty}}_{=\overline{\sigma}} + \underbrace{\frac{d}{dt}\sigma_s(t)\Big|_{t\to\infty}}_{=0}\underbrace{(t-\bar{t})}_{\approx\text{const.}} + \ldots = \overline{\sigma} \tag{2.9}$$

$$\mathcal{A}^s_{\mu\overline{\sigma}}(t\to\infty) = \int_0^\infty d\tau \sum_\kappa C^s_{\mu\kappa}(\tau)\mathcal{U}(\tau)a^s_\kappa\overline{\sigma} \tag{2.10}$$

Also in steady state we cannot solve the stationary solution $\overline{\sigma}$ directly since we still have the time evolution operator $\mathcal{U}$ to consider that acts on the modified density operator $a^s_\kappa\overline{\sigma} =: X^s_\kappa$. We now introduce this new notation for the time evolution of the modified density operator starting in steady state[1] $X^s_\kappa(\tau) = \mathcal{U}(\tau)a^s_\kappa\overline{\sigma}$ to emphasize the difference in the time evolution of density and the modified density operator in steady state.

Although this time evolution $X^s_\kappa(\tau) = \mathcal{U}(\tau)X^s_\kappa(0)$ is similar to the problem we wanted to solve originally for the reduced **system** density operator.

$$\sigma_s(t) = \mathcal{U}(t-t_0)\sigma_s(t_0) \tag{$\star$}$$

[1] Remember that we have defined the modified density operator $\tilde{\sigma}^s_\kappa(t)$ for an arbitrary starting time t', so $\tilde{\sigma}^s_\kappa(t) = \mathcal{U}(t)a^s_\kappa\sigma_s(t')$. Note that also for $t' = 0$, $\tilde{\sigma}^s_\kappa(t) \neq a^s_\kappa\sigma_s(t)$, as the time evolution of this new operator behaves differently.

We call this a **self-consistent** approach since in the treatment of the problem ($\star$) we need the solution of the almost same problem.

One could also regard this problem as a fix point equation ($x = f(\tilde{x})$), with x the solution of the problem ($\star$) depending on the solution of the almost same problem $\tilde{x}$ and solve this recursively under the presumption $x \approx \tilde{x}$ and the involved function f to be a contraction (Banach fixed-point theorem).

To have this equation in frequency space we use the Fourier representation of the bath correlation function $C^s_{\mu\kappa}$, swap integrals and Laplace transform the superpropagator:

$$\mathcal{A}^s_{\mu\tilde{\sigma}}(t \to \infty) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \sum_{\kappa} \Gamma^s_{\mu\kappa}(\omega) \underbrace{\mathcal{U}(s\omega) X^s_\kappa(0)}_{=X^s_\kappa(s\omega)}, \qquad X^s_\kappa(0) = a^s_\kappa \overline{\sigma} \tag{2.11}$$

The Laplace transform of the superpropagator $\mathcal{U}(\omega)$ is defined by it's effect on the operator space:

$$\mathcal{U}(\omega)A = \int_0^{\infty} dt e^{i\omega^+ t} \mathcal{U}(t)A \tag{2.12}$$

In the next section we will concentrate on the time evolution of X^s_κ and it's evolution in frequency space which we will need to calculate the steady state self-consistently.

The following box sums up all equations in the self-consistent Born approach. The equations in the second half for the modified density operator in frequency space are derived in the following section.

Self-consistent Born-Markov master equation

$$0 = \dot{\overline{\sigma}}(t) = -i\mathcal{L}_s \overline{\sigma} - \sum_{\mu s} \left[a^{\overline{s}}_\mu, \mathcal{A}^s_{\mu\overline{\sigma}}(t \to \infty) \right]_- + h.c. \tag{2.13}$$

$$\mathcal{A}^s_{\mu\overline{\sigma}}(t \to \infty) = \sum_{\kappa} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma^s_{\mu\kappa}(\omega) \underbrace{\mathcal{U}(s\omega) X^s_\kappa(0)}_{X^s_\kappa(s\omega)}, \qquad X^s_\kappa(0) = a^s_\kappa \overline{\sigma} \tag{2.14}$$

$$X^s_\kappa(\omega) = \Big(i(\mathcal{L}_s - \omega) + \Sigma(\omega) \Big)^{-1} X^s_\kappa(0) \tag{2.15}$$

$$\Sigma(\omega)(X^s_\kappa(\omega)) := \sum_{\nu\tau} \Big(\{ a^{\overline{\tau}}_\nu, A^\tau_{\nu X}(\omega) \} + \{ (A^\tau_{\nu X}(-\omega))^\ddagger, a^\tau_\nu \} \Big) \tag{2.16}$$

$$A^\tau_{\nu X}(\omega) = \sum_j C^\tau_{\nu j}(\omega - \mathcal{L}_s) a^\tau_j X^s_\kappa(\omega) \tag{2.17}$$

$$(A^\tau_{\nu X}(-\omega))^\ddagger = \sum_j C^\tau_{\nu j}(\mathcal{L}_s - \omega)^* X^s_\kappa(\omega) a^{\overline{\tau}}_j \tag{2.18}$$

2.2 Laplace transform of the Born-Markov equation

We consider now the modified density operator as we want to get it's evolution in ω space. In section 1.4.3 we have already shown that the modified density operator $\tilde{\sigma}^s_\kappa(t)$[2] underlies the same equation of motion as the system density matrix σ_s with the small difference, that in equation 1.76 there is an anticommutator instead of a commutator and a modified dagger operator $\ddagger$ that doesn't change the modified density operator $\tilde{\sigma}$.

BORN-MARKOV EQUATION FOR THE MODIFIED DENSITY OPERATOR

$$\dot{\tilde{\sigma}}^s_\kappa(t) = -i\mathcal{L}_s\tilde{\sigma}^s_\kappa(t) - \sum_{\nu\tau}\left(\left\{a^{\overline{\tau}}_\nu, A^\tau_{\nu\tilde{\sigma}(t')}(t)\right\} + \left\{\left(A^\tau_{\nu\tilde{\sigma}(t')}(t)\right)^\ddagger, a^\tau_\nu\right\}\right) \tag{2.19}$$

$$A^\tau_{\nu\tilde{\sigma}(t')}(t) = \int_0^t dt' \sum_j C^\tau_{\nu j}(t-t')\mathcal{G}(t-t')a^\tau_j\tilde{\sigma}^s_\kappa(t') \tag{2.20}$$

$$\mathcal{G}(t-t') = e^{-i\mathcal{L}_{0s}(t-t')} \tag{2.21}$$

$$C^\tau_{\nu j}(t) = \int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\Gamma^\tau_{\nu j}(\omega)e^{i\tau\omega^+ t} \tag{2.22}$$

We want to have a look at the complex Laplace transformed Born-Markov equation. The main transformation rules are summarized in the appendix 4.1 and their application gives:

$$-i\omega\tilde{\sigma}^s_\kappa(\omega) - \tilde{\sigma}^s_\kappa(0) = -i\mathcal{L}_s\tilde{\sigma}^s_\kappa(\omega) - \sum_{\nu\tau}\left(\{a^{\overline{\tau}}_\nu, A^\tau_{\nu\tilde{\sigma}(\omega)}(\omega)\} + \{\left(A^\tau_{\nu\tilde{\sigma}(\omega)}(-\omega)\right)^\ddagger, a^\tau_\nu\}\right) \tag{2.23}$$

First we evaluate $(A^\tau_\nu)(\omega)$. Starting time of the evolution is $t_0 = 0$ when we can express the modified density operator directly by the reduced system density operator $\tilde{\sigma}^s_\kappa(0) = [a^s_\kappa\sigma_s(t')]$. We apply the convolution theorem (see section 4.1.2) for the Laplace transform of the appearing convolution.

$$\begin{aligned}(A^\tau_{\nu\tilde{\sigma}(\omega)})(\omega) &= \sum_j\int_0^\infty dt e^{i(\omega+i0^+)t}\int_0^t dt' C^\tau_{\nu j}(t-t')\mathcal{G}(t-t')a^\tau_j\tilde{\sigma}^s_\kappa(t') \\ &= \sum_j \mathfrak{L}\left[(C^\tau_{\nu j}\circ\mathcal{G}) * (a^\tau_j\tilde{\sigma}^s_\kappa)\right] \\ &= \sum_j \mathfrak{L}\left[C^\tau_{\nu j}(t)e^{-i\mathcal{L}_s t}\right]\cdot a^\tau_j\mathfrak{L}\left[\tilde{\sigma}^s_\kappa(t)\right] \\ &= \sum_j C^\tau_{\nu j}(\omega-\mathcal{L}_s)\left(a^\tau_j\tilde{\sigma}^s_\kappa(\omega)\right)\end{aligned} \tag{2.24}$$

As the superoperator is of exponential form we have applied the shifting theorem. The resulting function of a superoperator still acts on the operator to it's right side.

[2] In this section we are not necessarily in steady state thus not using X^s_κ for the modified density operator.

Now we have to evaluate the Laplace transform of the Hermitian conjugated operator and we use the identity $\mathfrak{L}\left[(A_\nu^\tau(t))^\ddagger\right] := \left((A_\nu^\tau)^\ddagger\right)(\omega) = (A_\nu^\tau(-\omega))^\ddagger$. We start by switching the sign of the argument:

$$A_\nu^\tau(-\omega) = \sum_j C_{\nu j}^\tau(-\omega - \mathcal{L}_s) a_j^\tau \tilde{\sigma}_\kappa^s(-\omega) \tag{2.25}$$

Note that there is a superoperator in the argument of the function $C_{\nu j}^\tau$ thus the application of the adjoint operator has to be treated with care as done in theorem 1.2, stating $(C_{\nu j}^\tau(\mathcal{L}_s))^\dagger = C_{\nu j}^\tau(-\mathcal{L}_s)^*$:

$$(A_\nu^\tau(-\omega))^\ddagger = \sum_j \left(C_{\nu j}^\tau(-\omega - \mathcal{L}_s)\right)^\dagger (\tilde{\sigma}_\kappa^s(-\omega))^\ddagger a_j^{\overline{\tau}} \tag{2.26}$$

$$= \sum_j \left(C_{\nu j}^\tau(-\omega + \mathcal{L}_s)\right)^* \underbrace{\mathfrak{L}\left[\tilde{\sigma}_\kappa^s(t)^\ddagger\right]}_{=\mathfrak{L}[\tilde{\sigma}_\kappa^s(t)]} a_j^{\overline{\tau}} \tag{2.27}$$

$$= \sum_j \left(C_{\nu j}^\tau(\mathcal{L}_s - \omega)\right)^* \tilde{\sigma}_\kappa^s(\omega) a_j^{\overline{\tau}} \tag{2.28}$$

Laplace transform of the Born-Markov master equation for the modified density operator

$$-i\omega\tilde{\sigma}_\kappa^s(\omega) - \tilde{\sigma}_\kappa^s(0) = -i\mathcal{L}_s\tilde{\sigma}_\kappa^s(\omega) - \Sigma(\omega)\tilde{\sigma}_\kappa^s(\omega) \tag{2.29}$$

$$\Sigma(\omega)(\tilde{\sigma}_\kappa^s(\omega)) := \sum_{\nu\tau}\left(\{a_\nu^{\overline{\tau}}, A_{\nu\tilde{\sigma}}^\tau(\omega)\} + \{(A_{\nu\tilde{\sigma}}^\tau(-\omega))^\dagger, a_\nu^\tau\}\right) \tag{2.30}$$

$$A_{\nu\tilde{\sigma}}^\tau(\omega) = \sum_j C_{\nu j}^\tau(\omega - \mathcal{L}_s) a_j^\tau \tilde{\sigma}_\kappa^s(\omega) \tag{2.31}$$

$$(A_{\nu\tilde{\sigma}}^\tau(-\omega))^\dagger = \sum_j C_{\nu j}^\tau(\mathcal{L}_s - \omega)^* \tilde{\sigma}_\kappa^s(\omega) a_j^{\overline{\tau}} \tag{2.32}$$

From this equation we receive an approximation of the Laplace transformed propagator $\mathcal{U}(\omega)$ that we need in the self-consistent current formula (2.50) in order to evaluate it.

Laplace transformed propagator $\mathcal{U}(\omega)$

$$\mathcal{U}(\omega)\tilde{\sigma}_\kappa^s(0) = \tilde{\sigma}_\kappa^s(\omega) = \frac{1}{i(\mathcal{L}_s - \omega) + \Sigma(\omega)}\tilde{\sigma}_\kappa^s(0) \tag{2.33}$$

2.3 Current from self-consistent Born-Markov master equation in steady state

In this section we develop formulas for the current using the two presented approximations and compare them to the Meir-Wingreen current formula [47], which describes the current through an interacting electron system using the non-equilibrium Keldysh formalism. The cited paper gives also a good overview of different types of current formulas in the different regimes, like the Landauer-Büttiker formula in the non-interacting case.

2.3.1 Formula for the current in the Born-Markov approximation

In general the total current in the system is defined by the time derivative of charge ($e = 1, \hbar = 1$):

$$\dot{Q} = \frac{d}{dt}\langle N_s \rangle = \frac{d}{dt}\mathrm{Tr}_s\Big\{N_s \rho_{bs}\Big\} = \mathrm{Tr}_s\Big\{N_s \dot{\sigma}_s\Big\} = 0 \tag{2.34}$$

and according to the continuity equation it is equal to zero since ingoing current is equal to the negative outgoing current. For $\dot{\sigma}_s$ in the above equation we apply the Born-Markov master equation (1.109) for the reduced system density operator σ_s

$$\begin{aligned} J(t) = \underbrace{\frac{e}{h}}_{\text{set to 1}} \dot{Q}(t) &= -i\mathrm{Tr}_s\Big\{N_s[H_s, \sigma_s(t)]_-\Big\} - \sum_{\mu s}\mathrm{Tr}_s\Big\{N_s[a_\mu^{\bar{s}}, A^s_{\mu\sigma_s}(t)]_- + h.c.(\dagger)\Big\} \\ &= -i\mathrm{Tr}_s\Big\{\underbrace{[N_s, H_s]_-}_{=0}\sigma_s\Big\} - 2\,\mathrm{Re}\Big(\sum_{\mu s}\mathrm{Tr}_s\Big\{\underbrace{[N_s, a_\mu^{\bar{s}}]_-}_{-s a_\mu^{\bar{s}}} A^s_{\mu\sigma_s}(t)\Big\}\Big) \\ &= 2\sum_\mu \mathrm{Re}\left(\mathrm{Tr}_s\Big\{a_\mu A^+_{\mu\sigma_s}(t) - a_\mu^\dagger A^-_{\mu\sigma_s}(t)\Big\}\right) = 0 \end{aligned} \tag{2.35}$$

If we just look at the terms of the sum according to one bath index α we get the contribution of just that bath and thus an effective current. We consider the stationary current in steady state, thus $t \to \infty$, $\sigma_s(t) \to \overline{\sigma}$ (1.112).

$$J_\alpha = 2\sum_\mu \mathrm{Re}\left(\mathrm{Tr}_s\Big\{a_\mu A^+_{\alpha\mu\overline{\sigma}} - a_\mu^\dagger A^-_{\alpha\mu\overline{\sigma}}\Big\}\right) \tag{2.36}$$

$$A^s_{\alpha\mu\overline{\sigma}}(t\to\infty) = \sum_\kappa \int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\Gamma^s_{\alpha\mu\kappa}(\omega)\mathcal{G}_s(s\omega)[a^s_\kappa\overline{\sigma}]$$

So the current formula can be written as:

$$J_\alpha = 2\sum_{\mu\kappa}\mathrm{Re}\left(\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\Gamma^{(+)}_{\alpha\mu\kappa}(\omega)\mathrm{Tr}_s\Big\{a_\mu\mathcal{G}_s(\omega)[a_\kappa^\dagger\overline{\sigma}]\Big\} - \Gamma^{(-)}_{\alpha\mu\kappa}(\omega)\mathrm{Tr}_s\Big\{a_\mu^\dagger\mathcal{G}_s(-\omega)[a_\kappa\overline{\sigma}]\Big\}\right) \tag{2.37}$$

Complex conjugated of the trace

For the negative s we now want to get rid of the negative frequency argument in the superpropagator. As we just need the real part in the formula we can also work with the adjoint. Thus we remember $(\Gamma^s_{\alpha\mu\kappa})^\dagger(\omega) = \Gamma^s_{\alpha\kappa\mu}(\omega)$ and use that $\mathrm{Re}(A) = \mathrm{Re}(A^\dagger)$

$$\begin{aligned}
&\mathrm{Tr}_s\Big\{a^\dagger_\mu \mathcal{G}_s(-\omega)[a_\kappa\overline{\sigma}]\Big\}^\dagger = \int_0^\infty dt \mathrm{Tr}_s\Big\{a^\dagger_\mu e^{i(-\omega)t}\mathcal{G}_s(t)[a_\kappa\overline{\sigma}]\Big\}^\dagger \qquad (2.38)\\
&= \int_0^\infty dt \mathrm{Tr}_s\Big\{a^\dagger_\mu e^{-i\omega t}e^{-iH_s t}[a_\kappa\overline{\sigma}]e^{iH_s t}\Big\}^\dagger = \int_0^\infty dt \mathrm{Tr}_s\Big\{e^{i\omega t}e^{-iH_s t}[\overline{\sigma}a^\dagger_\kappa]e^{iH_s t}a_\mu\Big\}\\
&= \mathrm{Tr}_s\Big\{a_\mu \int_0^\infty dt e^{i\omega t}\mathcal{G}_s(t)[\overline{\sigma}a^\dagger_\kappa]\Big\} = \mathrm{Tr}_s\Big\{a_\mu \mathcal{G}_s(\omega)[\overline{\sigma}a^\dagger_\kappa]\Big\}
\end{aligned}$$

Markov approximation

Applying the Markov approximation has in fact excluded the steady state from being time propagated further so in fact we can write using the complex conjugated representation of the second trace

$$\begin{aligned}
J_\alpha = 2\sum_{\mu\kappa}\mathrm{Re}\Big(&\int_{-\infty}^\infty \frac{d\omega}{2\pi}\int_0^\infty dt e^{i\omega^+ t}\Gamma^{(+)}_{\alpha\mu\kappa}(\omega)\mathrm{Tr}_s\Big\{a_\mu e^{-iH_0 t}a^\dagger_\kappa e^{iH_0 t}\overline{\sigma}\Big\} \qquad (2.39)\\
&- \Gamma^{(-)}_{\alpha\kappa\mu}(\omega)\mathrm{Tr}_s\Big\{e^{-iH_0 t}a^\dagger_\kappa e^{iH_0 t}a_\mu\overline{\sigma}\Big\}\Big)
\end{aligned}$$

Here we cannot identify greater and lesser Green's function as we will do later in the self-consistent Born approach but we show the evaluation in the system eigenbasis

$$\begin{aligned}
J_\alpha = 2\sum_{\mu\kappa}\sum_{abcd}\mathrm{Re}\Big(&\int_{-\infty}^\infty \frac{d\omega}{2\pi}\Gamma^{(+)}_{\alpha\mu\kappa}(\omega)\int_0^\infty dt \mathrm{Tr}_s\Big\{e^{i(\omega^+ - E_b + E_c)t}|a\rangle\langle a|a_\mu|b\rangle\langle b|a^\dagger_\kappa|c\rangle\langle c|\overline{\sigma}|a\rangle\langle a|\Big\} \qquad (2.40)\\
&- \Gamma^{(-)}_{\alpha\kappa\mu}(\omega)\mathrm{Tr}_s\Big\{e^{i(\omega^+ - E_a + E_b)t}|a\rangle\langle a|a^\dagger_\kappa|b\rangle\langle b|a_\mu|c\rangle\langle c|\overline{\sigma}|a\rangle\langle a|\Big\}\Big)\\
= 2\sum_{\mu\kappa}\sum_{abcd}\mathrm{Re}\Big(&\int_{-\infty}^\infty \frac{d\omega}{2\pi}\Gamma^{(+)}_{\alpha\mu\kappa}(\omega)\int_0^\infty dt \mathrm{Tr}_s\Big\{e^{i(\omega^+ - E_b + E_c)t}|a\rangle\langle a|a_\mu|b\rangle\langle b|a^\dagger_\kappa|c\rangle\langle c|\overline{\sigma}|a\rangle\langle a|\Big\} \qquad (2.41)\\
&- \Gamma^{(-)}_{\alpha\kappa\mu}(\omega)\mathrm{Tr}_s\Big\{e^{i(\omega^+ - E_a + E_b)t}|a\rangle\langle a|a^\dagger_\kappa|b\rangle\langle b|a_\mu|c\rangle\langle c|\overline{\sigma}|a\rangle\langle a|\Big\}\Big)
\end{aligned}$$

The integrals is similar to the one already solved in (1.122)

$$\begin{aligned}
F^s_{\alpha\mu\kappa}(E_{bc}) &= \int_{-\infty}^\infty \frac{d\omega}{2\pi}\Gamma^{(\pm)}_{\alpha\mu\kappa}(\omega)\int_0^\infty dt e^{i(\omega^+ - E_{bc})t} = \int_{-\infty}^\infty \frac{d\omega}{2\pi}\Gamma^{(\pm)}_{\alpha\mu\kappa}\frac{in^s_\alpha(\omega)}{\omega - E_{bc} + i0^+}\\
&= \frac{\Gamma_{\alpha\mu\kappa}}{2}\left(n^s_\alpha(E_{bc}) + \frac{i}{\pi}\mathrm{Re}\Big[\Psi\Big(\frac{1}{2} + i\frac{\beta(E_{bc}-\mu_\alpha)}{2\pi}\Big)\Big]\right) \qquad (2.42)
\end{aligned}$$

The full formula with Q-matrices is given in the box below:

CURRENT FORMULA FOR THE BORN-MARKOV MASTER EQUATION

$$J_\alpha = 2\sum_{\mu\kappa} \mathrm{Re}\Big(\mathrm{Tr}_s\Big\{Q_\mu \widetilde{Q}^{\dagger,+}_\kappa \overline{\sigma}\Big\} - \mathrm{Tr}_s\Big\{\widetilde{Q}^{\dagger,-}_\kappa Q_\mu \overline{\sigma}\Big\}\Big) \tag{2.43}$$

$$\widetilde{Q}^{\dagger,s}_\kappa = \sum_{ab} F^s_{\alpha\mu\kappa}(E_{ab})\langle a|a^\dagger_\kappa|b\rangle |a\rangle\langle b| \tag{2.44}$$

Self-consistent approach

We identify the expressions in the square brackets with the reduced modified density operators as we have introduced them in the sections 1.1.3 and 2.1, $X^s_\kappa(0) = [a^s_\kappa \overline{\sigma}]$.

We can also make the self-consistent Born approximation here, exchanging the time evolution operator $\mathcal{G}_s$ by $\mathcal{U}$ which propagates the modified density operator according to the full Hamiltonian H. The equation is given below (2.50).

Green's function description

Our next aim is to receive a formula in terms of Green's function. For the first trace we define

$$\phi^+_{\mu\kappa}(\omega) := \mathrm{Tr}_s\Big\{a_\mu \tilde{\sigma}^\dagger_\kappa(\omega)\Big\} = \mathrm{Tr}_s\Big\{a_\mu \mathcal{U}(\omega) a^\dagger_\kappa \overline{\sigma}\Big\} \tag{2.45}$$

Since we just need the real part of the second trace we define for it's adjoint (2.38):

$$\phi^-_{\mu\kappa}(\omega) := \mathrm{Tr}_s\Big\{a_\mu \mathcal{U}(\omega) \overline{\sigma} a^\dagger_\kappa\Big\} \tag{2.46}$$

For the two traces we have the the initial condition $\phi^s_{\mu\kappa}(0) = \mathrm{Tr}_s\Big\{a_\mu (a^s_\kappa \overline{\sigma})^{\overline{s}}\Big\}$. Remembering the Born approximation we can insert the trace over bath and have in time space:

$$\phi^+_{\mu\kappa}(t) = \mathrm{Tr}\{a_\mu e^{-i\mathcal{L}_s t} a^\dagger_\kappa \overline{\rho}\} = \mathrm{Tr}\{a_\mu e^{-iH_s t} a^\dagger_\kappa \overline{\rho} e^{iH_s t}\} = \mathrm{Tr}\{(e^{iH_s t} a_\mu e^{-iH_s t}) a^\dagger_\kappa \overline{\rho}\} = \langle a_\mu(t) a^\dagger_\kappa\rangle$$
$$\phi^-_{\mu\kappa}(t) = \mathrm{Tr}\{a_\mu e^{-i\mathcal{L}_s t} \overline{\rho} a^\dagger_\kappa\} = \mathrm{Tr}\{a_\mu e^{-iH_s t} \overline{\rho} a^\dagger_\kappa e^{iH_s t}\} = \mathrm{Tr}\{a^\dagger_\kappa (e^{iH_s t} a_\mu e^{-iH_s t}) \overline{\rho}\} = \langle a^\dagger_\kappa a_\mu(t)\rangle$$

Here we have used the cyclic property of the trace and note that for time evolution of annihilation or creation operators in Heisenberg picture we have in contrast to density operators the opposite sign in the exponential terms. We now identify these expressions with the greater respectively lesser Green's functions:

$$G^>_{\mu\kappa}(t) = -i\phi^+_{\mu\kappa}(t), \qquad G^<_{\mu\kappa}(t) = i\phi^-_{\mu\kappa}(t) \tag{2.47}$$

Now we have to go to frequency space. Since $\phi^s_{\mu\kappa}(\omega)$ is the Laplace transform of $isG^s_{\mu\kappa}(t)$ we use the dispersive relation for the Fourier transform 4.1.3

$$G^{<,>}_{\mu\kappa}(\omega) = \int_{-\infty}^{\infty} dt e^{-i\omega t} G^{<,>}_{\mu\kappa}(t)$$
$$G^{<,>}_{\mu\kappa}(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{i\omega t} G^{<,>}_{\mu\kappa}(\omega)$$
$$\phi^s_{\mu\kappa}(\omega) = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{-sG^s_{\mu\kappa}(\omega')}{\omega + \omega' + i0^+}$$

with the same mapping of s for the Green's functions as given in equation (2.47): $(+ \equiv >)$. Since $\phi_{\mu\kappa}(t) := \phi^+_{\mu\kappa}(t) + \phi^-_{\mu\kappa}(t) = \langle\{a_\mu(t), a^\dagger_\kappa\}\rangle$ combined with $-i\Theta(t)$ gives the retarded Green's function we get for the Laplace transform which implicates the theta function:

$$\phi_{\mu\kappa}(\omega) = iG^r_{\mu\kappa}(\omega) \tag{2.48}$$

Formulas for the current

$$J_\alpha = 2\sum_{\mu\kappa} \mathrm{Re}\left(\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma^{(+)}_{\alpha\mu\kappa}(\omega)\phi^+_{\mu\kappa}(\omega) - \Gamma^{(-)}_{\alpha\kappa\mu}(\omega)\phi^-_{\mu\kappa}(\omega)\right) \tag{2.49}$$
$$= 2\sum_{\mu\kappa} \mathrm{Re}\left(\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma^{(+)}_{\alpha\mu\kappa}(\omega)\mathrm{Tr}_s\Big\{a_\mu \tilde{\sigma}^+_\kappa(\omega)\Big\} - \Gamma^{(-)}_{\alpha\mu\kappa}(\omega)\mathrm{Tr}_s\Big\{a^\dagger_\mu \tilde{\sigma}^-_\kappa(-\omega)\Big\}\right) \tag{2.50}$$
$$\tag{2.51}$$

We now decompose the bath density function into spectral and Fermi function

$$\Gamma^s_{\alpha\mu\kappa}(\omega) = S_{\alpha(\mu\kappa)_{\Pi(s)}}(\omega) n^s_\alpha(\omega),$$

with $\Pi(s)$ denoting a transposition of the indices for negative s. Note that for both values of s, we get the same sequence of indices $\mu\kappa$ in the spectral density function S since in the above formula the indices are already twisted for negative s. So we can combine both ϕ's by using $n^-_\alpha(\omega) = 1 - n^{(+)}_\alpha(\omega)$ to regain the retarded Green's function. By taking the difference of the current formulas according to right and left bath we additionally get a formula of

$$J_\alpha = 2\sum_{\mu\kappa} \mathrm{Re}\Big[\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} S_{\alpha\mu\kappa}(\omega) n_\alpha(\omega) \underbrace{\big(\phi^+_{\mu\kappa}(\omega) + \phi^-_{\mu\kappa}(\omega)\big)}_{=iG^r_{\mu\kappa}(\omega)} - S_{\alpha\mu\kappa}(\omega)\phi^-_{\mu\kappa}(\omega)\Big] \tag{2.52}$$

$$J = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \mathrm{Tr}_{\mu\kappa}\Big\{\mathrm{Re}\Big(\Big[\boldsymbol{S}_R(\omega) n_R(\omega) - \boldsymbol{S}_L(\omega) n_L(\omega)\Big] i\boldsymbol{G}^R(\omega) - \Big[\boldsymbol{S}_R(\omega) - \boldsymbol{S}_L(\omega)\Big]\boldsymbol{\phi}^-(\omega)\Big)\Big\} \tag{2.53}$$

In the second equation we introduced a matrix notation $\boldsymbol{G}$ for the representation of $G_{\mu\kappa}$ and interpreted the sums over both variables as trace of a matrix product.[3] We assume here the spectral function $\boldsymbol{S}$ to be adjoint according to μ and κ and can interpret the real part of $\mathrm{Tr}\{i\boldsymbol{S}\boldsymbol{G}^r\}$ as $i\mathrm{Tr}\{\boldsymbol{S}(\boldsymbol{G}^r - \boldsymbol{G}^a)/2\}$ since the retarded Green's function is the adjoint of the advanced Green's function in frequency space.

For the right hand side we can apply a similar trick since adding the adjoint resembles two times the real part. We now show that the adjoint of $\boldsymbol{\phi}^-$ in fact embodies the continuation to the full Fourier transform of the lesser Green's function $\boldsymbol{\phi}^-(\omega) + (\boldsymbol{\phi}^-(\omega))^\dagger = -i\boldsymbol{G}^<(\omega)$. Written out with indices we show that $\phi^-_{\mu\kappa}(\omega) + \phi^-_{\kappa\mu}(\omega)^\dagger = -iG^<_{\mu\kappa}(\omega)$. Looking at the second addend we have

$$\begin{aligned}(\phi^-_{\kappa\mu}(\omega))^\dagger &= \left(\int_0^\infty d\omega e^{i\omega t}\phi_{\kappa\mu}(t)\right)^\dagger = \int_0^\infty dt e^{-i\omega t}\langle a^\dagger_\mu a_\kappa(t)\rangle^\dagger \\ &\overset{t\to -t}{=} -\int_0^{-\infty} dt e^{i\omega t}\langle a^\dagger_\kappa(-t)a_\mu\rangle = \int_{-\infty}^0 dt e^{i\omega t}\underbrace{\langle a^\dagger_\kappa a_\mu(t)\rangle}_{=\phi^-_{\mu\kappa}(t)}\end{aligned}$$

which comes out to be in fact the continuation of the Laplace to the full Fourier transform.

Finally we have received the Meir-Wingreen formula [47]:

Current formula in the self-consistent Born approach in the Meir-Wingreen representation
$$J = \frac{ie}{2\hbar}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\mathrm{Tr}\left\{(\boldsymbol{S}_R n_R - \boldsymbol{S}_L n_L)(\boldsymbol{G}^r - \boldsymbol{G}^a) + (\boldsymbol{S}_R - \boldsymbol{S}_L)\boldsymbol{G}^<\right\} \quad (2.54)$$

For assuming the spectral density of both functions to be linear dependent $\boldsymbol{S}_L = \lambda \boldsymbol{S}_R$ one can recast the Landauer-Büttiker type of current formula, see [26].

[3] $\boldsymbol{S}$ represents switched indices $\mu\kappa$ in $S_{\alpha\kappa\mu}$ to match the matrix multiplication.

2.4 Evaluation of the self-consistent Born-Markov master equation for a quantum dot with spin dependent coupling

2.4.1 Model

In this section we want to analytically derive an equation for the current and the reduced density matrix of a single interacting quantum dot weakly coupled to two non-interacting baths, referred to as left and right bath ($\alpha \in \{L, R\}$) (Anderson impurity). We use the same Hamiltonians for the baths and the coupling as defined in section 1.1.2.

As each bath just couples to the one system site we have $\delta_{\mu\kappa}$ in the interaction Hamiltonian and the bath correlation function so there is one degree of freedom in the spin left $\kappa = \tau \in \{\uparrow, \downarrow\}$. For the baths we assume to have a flat spectral density distribution (wide band limit). Thus the spectral density function of the baths looks like[4]

$$\Gamma^s_{\alpha\kappa\kappa}(\omega) = n^s_\alpha(\omega) 2\pi |t_{\alpha\kappa}|^2 \underbrace{\sum_k \delta(\omega - \epsilon_k)}_{=\text{constant}} = n^s_\alpha(\omega)\Gamma_{\alpha\kappa} \tag{2.55}$$

Additionally we assume that the coupling from the baths to the quantum dot is spin dependent, namely blocked for one spin to one bath:

$$\Gamma_{L\uparrow} = \Gamma_{L\downarrow} = \Gamma_{R\uparrow} = \Gamma, \qquad \Gamma_{R\downarrow} = 0 \tag{2.56}$$

2.4.2 Formulas of the self-consistent problem

Starting point is the self-consistent Born-Markov master equation for the reduced system density operator σ_s in steady state, see also equation (2.13).

[4] We have condensed the summation over spin τ in the summation over system site κ.

SELF-CONSISTENT BORN-MARKOV MASTER EQUATION FOR THE QUANTUM DOT FOR SPIN DEPENDENT COUPLING IN STEADY STATE

$$0 = \dot{\overline{\sigma}}_s = -i\mathcal{L}_s\overline{\sigma} - \sum_{\kappa s}\left[a^{\overline{s}}_\kappa, \mathcal{A}^s_{\kappa\tilde{\sigma}}\right]_- + h.c. \tag{2.57}$$

$$\mathcal{A}^s_{\kappa\tilde{\sigma}} = \sum_\alpha \int_{-\infty}^{\infty} \frac{d\omega}{2\pi}\Gamma_{\alpha\kappa} n^s_\alpha(\omega)\underbrace{\mathcal{U}(s\omega)\tilde{\sigma}^s_\kappa(0)}_{\tilde{\sigma}^s_\kappa(s\omega)}, \qquad \tilde{\sigma}^s_\kappa(0) := [a^s_\kappa\overline{\sigma}] \tag{2.58}$$

$$\Gamma_{L\kappa} = \Gamma_{R\uparrow} = \Gamma, \quad \Gamma_{R\downarrow} = 0 \tag{2.59}$$

$$\tilde{\sigma}^s_\kappa(\omega) = \Big(i(\mathcal{L}_s - \omega) + \Sigma(\omega)\Big)^{-1}\tilde{\sigma}^s_\kappa(0) \tag{2.60}$$

$$\Sigma(\omega)(\tilde{\sigma}^s_\kappa(\omega)) := \sum_{\nu\tau}\Big(\{a^{\overline{\tau}}_\nu, A^\tau_{\nu\tilde{\sigma}}(\omega)\} + \{\big(A^\tau_{\mu\tilde{\sigma}}(-\omega)\big)^\ddagger, a^\tau_\nu\}\Big) \tag{2.61}$$

$$A^\tau_{\nu\tilde{\sigma}}(\omega) = C^\tau_\nu(\omega - \mathcal{L}_s)a^\tau_\nu\tilde{\sigma}^s_\kappa(\omega) \tag{2.62}$$

$$(A^\tau_{\nu\tilde{\sigma}}(-\omega))^\ddagger = C^\tau_\nu(\mathcal{L}_s - \omega)^*\tilde{\sigma}^s_\kappa(\omega)a^{\overline{\tau}}_\nu \tag{2.63}$$

The aim is to solve this bunch of equations to get the stationary solution of the system $\overline{\sigma}$. Thus we have assumed the matrix $\langle a|\dot{\overline{\sigma}}_s|b\rangle = \dot{\sigma}_{ab}$ in the first equation to be zero and endeavour to gain an equation of the form $0 = K\overline{\sigma}$ where K is a rank four tensor and often referred to as Lindblad operator [33].

First step will be to solve the second half of equations to get an expression for the modified density operator $\tilde{\sigma}$. We use $X^s_\kappa := \tilde{\sigma}^s_\kappa(\omega)$ to refer to this operator throughout this derivation and don't write explicitly the dependence of ω since it's not relevant until the evaluation of the integral in the end.

As basis set we use the system eigenbasis consisting of $|0\rangle, |\uparrow\rangle, |\downarrow\rangle, |d\rangle = |\uparrow\downarrow\rangle$. We denote the half occupied state with $|\kappa\rangle$, address the opposite spin direction with $|\overline{\kappa}\rangle$ and use κ also for the sign in the basis representation $|d\rangle = a^\dagger_\uparrow a^\dagger_\downarrow|0\rangle$ when creators might have to switch position. So $a^\dagger_\kappa|\overline{\kappa}\rangle = \kappa|d\rangle$ depending on the spin ($\uparrow$: $+$, $\downarrow$: $-$).

We start with the matrix representation of the first equation. Since the left hand side is self-adjoint this holds also for the right hand side where the imaginary part cancels:

$$0 = \dot{\sigma}_{ab} = -i(E_a - E_b)\sigma_{ab} - 2\mathrm{Re}\left(\sum_{\kappa s}\langle a|a^{\overline{s}}_\kappa\mathcal{A}^s_{\kappa\tilde{\sigma}}|b\rangle - \langle a|\mathcal{A}^s_{\kappa\tilde{\sigma}}a^{\overline{s}}_\kappa|b\rangle\right) \tag{2.64}$$

Selection rules

Since the time evolution $\mathcal{U}$ of the modified density operator according to the full Hamiltonian preserves the particle number in each spin sector, $\mathcal{A}^s_\kappa$ and X^s_κ have non zero matrix

elements for the same indices as a^s_κ, whose are given below :

$$\begin{aligned}\langle d|a^\dagger_\kappa|\overline{\kappa}\rangle &= \kappa \\ \langle \kappa|a^\dagger_\kappa|0\rangle &= 1 \\ \langle \overline{\kappa}|a_\kappa|d\rangle &= \kappa \\ \langle 0|a_\kappa|\kappa\rangle &= 1\end{aligned} \tag{2.65}$$

Therefore equation (2.64) has non-zero matrix elements just on the diagonal, so the resulting density operator will will be diagonal in any case.

The same selection rules will be applicable to the self energy operator (ΣX^s_κ) (2.61), when determining non-zero matrix elements, since in all terms occur a pairing of $a^\dagger_\nu$ and a_ν.

Evaluation in the eigenbasis

We continue with equation (2.64) with inserting a unity operator $\mathbb{1} = \sum_c |c\rangle\langle c|$ that leads to

$$0 = \mathrm{Re}\left(\sum_{\kappa s}\sum_c \langle a|a^{\overline{s}}_\kappa|c\rangle\langle c|\mathcal{A}^s_{\kappa\tilde{\sigma}}(\omega)|a\rangle - \langle a|\mathcal{A}^s_{\kappa\tilde{\sigma}}(\omega)|c\rangle\langle c|a^{\overline{s}}_\kappa|a\rangle\right) \tag{2.66}$$

With the selection rules $\langle a|a^{\overline{s}}_\kappa|c\rangle \in \{0, \pm 1\}$ we get for the different eigenstates:

$$\begin{aligned}
a = 0: \qquad & 0 = \sum_\kappa \mathrm{Re}\left(\langle 0|a_\kappa|\kappa\rangle\langle\kappa|\mathcal{A}^+_\kappa|0\rangle - \underbrace{\langle 0|\mathcal{A}^-_\kappa|\kappa\rangle}_{=:\mathcal{A}^-_{\kappa,\kappa}}\langle\kappa|a^\dagger_\kappa|0\rangle\right) \\
a = \kappa \in \{\uparrow,\downarrow\}: \qquad & 0 = \mathrm{Re}\Big(\langle\kappa|a^\dagger_\kappa|0\rangle\langle 0|\mathcal{A}^-_\kappa|\kappa\rangle + \underbrace{\langle\kappa|a_{\overline{\kappa}}|d\rangle}_{=\overline{\kappa}}\langle d|\mathcal{A}^+_{\overline{\kappa}}|\kappa\rangle \\
& \quad - \langle\kappa|\mathcal{A}^+_\kappa|0\rangle\langle 0|a_\kappa|\kappa\rangle - \langle\kappa|\mathcal{A}^-_{\overline{\kappa}}|d\rangle\underbrace{\langle d|a^\dagger_{\overline{\kappa}}|\kappa\rangle}_{=\overline{\kappa}}\Big) \\
a = d: \qquad & 0 = \sum_\kappa \mathrm{Re}\left(\underbrace{\langle d|a^\dagger_\kappa|\overline{\kappa}\rangle}_{=\kappa}\langle\overline{\kappa}|\mathcal{A}^-_\kappa|d\rangle - \langle d|\mathcal{A}^+_\kappa|\overline{\kappa}\rangle\underbrace{\langle\overline{\kappa}|a_\kappa|d\rangle}_{=\kappa}\right) \\
& = \sum_\kappa \mathrm{Re}\left(\underbrace{\langle d|a^\dagger_{\overline{\kappa}}|\kappa\rangle}_{=\overline{\kappa}}\langle\kappa|\mathcal{A}^-_{\overline{\kappa}}|d\rangle - \langle d|\mathcal{A}^+_{\overline{\kappa}}|\kappa\rangle\underbrace{\langle\kappa|a_{\overline{\kappa}}|d\rangle}_{=\overline{\kappa}}\right)
\end{aligned}$$

with the now introduced short notation for the matrix elements $\mathcal{A}^s_\kappa$. As we gain minus signs from some matrix elements we include them in the notation with the tilde. We will see that these factors will cancel out since the final result shouldn't depend on the basis

representation in Fock space.

$$\begin{aligned} \mathcal{A}^{+}_{\kappa,0} &= \langle\kappa|\mathcal{A}^{+}_{\kappa}|0\rangle & \mathcal{A}^{-}_{\kappa,\kappa} &= \langle 0|\mathcal{A}^{-}_{\kappa}|\kappa\rangle \\ \tilde{\mathcal{A}}^{+}_{\overline{\kappa},\kappa} &= \overline{\kappa}\langle d|\mathcal{A}^{+}_{\overline{\kappa}}|\kappa\rangle & \tilde{\mathcal{A}}^{-}_{\overline{\kappa},d} &= \overline{\kappa}\langle\kappa|\mathcal{A}^{-}_{\overline{\kappa}}|d\rangle \end{aligned} \tag{2.67}$$

So we get

$$\begin{aligned} a = 0: \quad & 0 = \sum_{\kappa} \mathrm{Re}\left(\mathcal{A}^{+}_{\kappa,0} - \mathcal{A}^{-}_{\kappa,\kappa}\right) \\ a = \kappa: \quad & 0 = \mathrm{Re}\left(\mathcal{A}^{-}_{\kappa,\kappa} + \tilde{\mathcal{A}}^{+}_{\overline{\kappa},\kappa} - \mathcal{A}^{+}_{\kappa,0} - \tilde{\mathcal{A}}^{-}_{\overline{\kappa},d}\right) \\ a = d: \quad & 0 = \sum_{\kappa} \mathrm{Re}\left(\tilde{\mathcal{A}}^{-}_{\overline{\kappa},d} - \tilde{\mathcal{A}}^{+}_{\overline{\kappa},\kappa}\right) \end{aligned} \tag{2.68}$$

We can use the same notation for the matrix elements of $X^s_k := \tilde{\sigma}^s_k(\omega)$ appearing in the formula of the operators $\mathcal{A}$ because the other terms are just scalar factors.

We remain with evaluating the real part of:

$$\begin{aligned} \mathrm{Re}\left(\mathcal{A}^{s}_{\kappa,E}\right) &= \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \sum_{\alpha} \Gamma_{\alpha\kappa} n^{s}_{\alpha}(\omega) \mathrm{Re}\left(X^{s}_{\kappa,E}(s\omega)\right) \\ &\overset{s\omega\to z}{=} \int_{-\infty}^{\infty} \frac{dz}{2\pi} \sum_{\alpha} \Gamma_{\alpha\kappa} n^{s}_{\alpha}(sz) \mathrm{Re}\left(X^{s}_{\kappa,E}(z)\right) \end{aligned} \tag{2.69}$$

This equation inherits a big numerical cost, as one has to calculate these matrix elements $X^s_{\kappa,E}$ for every frequency ω before integrating over the real axis.

We proceed by simplifying the generalized Fermi function using the symmetry in the bath potentials $\mu_\alpha = -\mu_{s\alpha}$:

$$n^{s}_{\alpha}(s\omega) = \begin{cases} (1 + e^{\beta(\omega-\mu_\alpha)})^{-1}, & s = + \\ (1 + e^{-\beta(-\omega-\mu_\alpha)})^{-1} = n^{+}_{s\alpha}(\omega), & s = - \end{cases} \tag{2.70}$$

With the special choice of spin dependent coupling constants (2.56) and $n(\omega) = \sum_\alpha n_\alpha(\omega)$ we get:

$$\begin{aligned} g^{s}_{\kappa}(\omega) &:= \sum_{\alpha} \Gamma_{\alpha\kappa} n^{+}_{s\alpha}(\omega) \\ g^{+}_{\uparrow}(\omega) &= \Gamma n(\omega) \\ g^{-}_{\uparrow}(\omega) &= \Gamma n(\omega) \\ g^{+}_{\downarrow}(\omega) &= \Gamma n_L(\omega) \\ g^{-}_{\downarrow}(\omega) &= \Gamma n_R(\omega) \end{aligned} \tag{2.71}$$

So we have

$$
\begin{aligned}
a = 0: \qquad & 0 = \int_{-\infty}^{\infty} d\omega \mathrm{Re}\left(n(\omega)X^{+}_{\uparrow,0} - n(\omega)X^{-}_{\uparrow,\uparrow} + n_L(\omega)X^{+}_{\downarrow,0} - n_R(\omega)X^{-}_{\downarrow,\downarrow}\right) && (2.72)\\
a = \uparrow: \qquad & 0 = \int_{-\infty}^{\infty} d\omega \mathrm{Re}\left(n(\omega)X^{-}_{\uparrow,\uparrow} + n_L(\omega)\tilde{X}^{+}_{\downarrow,\uparrow} - n(\omega)X^{+}_{\uparrow,0} - n_R(\omega)\tilde{X}^{-}_{\downarrow,d}\right)\\
a = \downarrow: \qquad & 0 = \int_{-\infty}^{\infty} d\omega \mathrm{Re}\left(n_R(\omega)X^{-}_{\downarrow,\downarrow} + n(\omega)\tilde{X}^{+}_{\uparrow,\downarrow} - n_L(\omega)X^{+}_{\downarrow,0} - n(\omega)\tilde{X}^{-}_{\uparrow,d}\right)\\
a = d: \qquad & 0 = \int_{-\infty}^{\infty} d\omega \mathrm{Re}\left(n_R(\omega)\tilde{X}^{-}_{\downarrow,d} - n_L(\omega)\tilde{X}^{+}_{\downarrow,\uparrow} + n(\omega)\tilde{X}^{-}_{\uparrow,d} - n(\omega)\tilde{X}^{+}_{\uparrow,\downarrow}\right)
\end{aligned}
$$

Here the constant coupling factor Γ drops out. The same happens in the derivation of the normal Born-Markov master equation, thus the current curvature is independent of the coupling strength. In the self-consistent Born-Markov master equation this factor will come in again through the self-consistent determined operator X - see section 2.4.3.

Below we summarize this first evaluation with the general functions $g^s_\kappa(\omega) = \sum_\alpha \Gamma_{\alpha\kappa} n^s_\alpha(\omega)$:

Evaluation of the stationary solution of the interacting quantum dot in it's eigenbasis

$$0 = K \cdot \overline{\sigma}_{aa} \qquad (2.73)$$

$$
\begin{aligned}
a = 0: \qquad & 0 = +A_1 + A_2, \qquad & A_1 &:= \int_{-\infty}^{\infty} d\omega \mathrm{Re}\left(g^{+}_{\uparrow}(\omega)X^{+}_{\uparrow,0} - g^{-}_{\uparrow}(\omega)X^{-}_{\uparrow,\uparrow}\right) && (2.74)\\
a = \uparrow: \qquad & 0 = -A_1 + A_3, \qquad & A_2 &:= \int_{-\infty}^{\infty} d\omega \mathrm{Re}\left(g^{+}_{\downarrow}(\omega)X^{+}_{\downarrow,0} - g^{-}_{\downarrow}(\omega)X^{-}_{\downarrow,\downarrow}\right)\\
a = \downarrow: \qquad & 0 = -A_2 + A_4, \qquad & A_3 &:= \int_{-\infty}^{\infty} d\omega \mathrm{Re}\left(g^{+}_{\downarrow}(\omega)\tilde{X}^{+}_{\downarrow,\uparrow} - g^{-}_{\downarrow}(\omega)\tilde{X}^{-}_{\downarrow,d}\right)\\
a = d: \qquad & 0 = -A_3 - A_4, \qquad & A_4 &:= \int_{-\infty}^{\infty} d\omega \mathrm{Re}\left(g^{+}_{\uparrow}(\omega)\tilde{X}^{+}_{\uparrow,\downarrow} - g^{-}_{\uparrow}(\omega)\tilde{X}^{-}_{\uparrow,d}\right)
\end{aligned}
$$

From this structure it's obvious that the column sum is always zero, thus the resulting matrix K will be singular revealing the stationary solution.

2.4.3 Evaluation of X^s_κ in the system eigenbasis

As we need the matrix elements $X^s_{\kappa,E}$ we look at the matrix elements E, E' of equation (2.60)

$$\begin{aligned}[a^s_\kappa \overline{\sigma}] &= (i(\mathcal{L}_s - \omega) + \Sigma(\omega))\,(X^s_\kappa(\omega)) \\ [a^s_\kappa \overline{\sigma}]_{EE'} &= i(E - E' - \omega)\,(X^s_\kappa(\omega))_{EE'} + \Big(\Sigma(\omega) X^s_\kappa(\omega)\Big)_{EE'}\end{aligned} \tag{2.75}$$

As $\overline{\sigma}$ has to be diagonal in the system eigenbasis (conservation of particle number in spin sectors) we get for the possible non zero matrix elements the same selection rules as defined in equation (2.65).

The first term on the right side with the Liouville superoperator is evaluated easily (see theorem 1.2), whereas the self-energy operator Σ will be more involved to get an expression in the desired matrix elements.

The general definition of the self-energy operator (see equations (2.61) - (2.18)) contains the bath correlation function $C^s_{\mu\nu}$ for which $\delta_{\mu,\nu}$ applies, as there is just one system site to connect the baths to (compare also section 2.4.1). We use the index ν to describe the remaining spin degree of freedom (similar to κ). A more detailed look at the bath correlation function and it's representation according to equation (4.19) reveals:

$$C^\tau_\nu(\omega) = \sum_\alpha C^\tau_{\alpha\nu\nu} = \sum_\alpha \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{i}{\omega + \tau\omega' + i0^+} \Gamma^\tau_{\alpha\nu\nu}(\omega') \tag{2.76}$$

with $\nu \in \{\uparrow, \downarrow\}$. Here also the wide band approximation is applied with spin dependent coupling:

$$\Gamma^\tau_{\alpha\nu\nu}(\omega') = n^\tau_\alpha(\omega')\Gamma_{\alpha\nu}, \qquad \Gamma_{L\nu} = \Gamma_{R,\uparrow} = \Gamma, \quad \Gamma_{R,\downarrow} = 0 \tag{2.77}$$

At this point of the self-consistent Born-Markov approximation the constant coupling factor comes in again.

So we get similar to $g^s_\kappa(\omega)$:

$$C^\tau_\nu(\omega) = \sum_\alpha \Gamma_{\alpha\nu} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{i \cdot n^\tau_\alpha(\omega')}{\omega + \tau\omega' + i0^+} \tag{2.78}$$

$$\overset{\tau\omega' \to z}{=} \sum_\alpha \Gamma_{\alpha\nu} \underbrace{\int_{-\infty}^{\infty} \frac{dz}{2\pi} \frac{i \cdot n^+_{\tau\alpha}(z)}{\omega + z + i0^+}}_{=: u_{\tau\alpha}(\omega)} \tag{2.79}$$

$$C^+_\uparrow(\omega) = \Gamma u(\omega), \qquad u(\omega) = u_L(\omega) + u_R(\omega) \tag{2.80}$$

$$C^-_\uparrow(\omega) = \Gamma u(\omega) \tag{2.81}$$

$$C^+_\downarrow(\omega) = \Gamma u_L(\omega) \tag{2.82}$$

$$C^-_\downarrow(\omega) = \Gamma u_R(\omega) \tag{2.83}$$

The derivation of the analytic expression of the function u is given in the Appendix 4.2.

We remain with the missing matrix elements of equation (2.75):

$$\begin{aligned}\Sigma(\omega)X^s_\kappa(\omega) = \sum_{\nu\tau}\Big(&a^{\overline{\tau}}_\nu C^\tau_\nu(\omega-\mathcal{L}_s)a^\tau_\nu X^s_\kappa(\omega)\\ &+ C^\tau_\nu(\omega-\mathcal{L}_s)a^\tau_\nu X^s_\kappa(\omega)a^{\overline{\tau}}_\nu\\ &+ C^\tau_\nu(\mathcal{L}_s-\omega)^* X^s_\kappa(\omega)a^{\overline{\tau}}_\nu a^\tau_\nu\\ &+ a^\tau_\nu C^\tau_\nu(\mathcal{L}_s-\omega)^* X^s_\kappa(\omega)a^{\overline{\tau}}_\nu\Big)\end{aligned}$$

$$\begin{aligned}\Big(\Sigma(\omega)X^s_\kappa\Big)_{EE'} = \sum_{F\nu\tau}\Big(&\langle E|a^{\overline{\tau}}_\nu|F\rangle C^\tau_\nu(\omega-(F-E'))\langle F|a^\tau_\nu X^s_\kappa|E'\rangle\\ &+ C^\tau_\nu(\omega-(E-E'))\langle E|a^\tau_\nu X^s_\kappa a^{\overline{\tau}}_\nu|E'\rangle\\ &+ C^\tau_\nu(E-E'-\omega)^*\langle E|X^s_\kappa a^{\overline{\tau}}_\nu a^\tau_\nu|E'\rangle\\ &+ \langle E|a^\tau_\nu|F\rangle C^\tau_\nu(F-E'-\omega)^*\langle F|X^s_\kappa a^{\overline{\tau}}_\nu|E'\rangle\Big)\end{aligned}$$

We start with $s=+$:

$$\begin{aligned}\Big(\Sigma(\omega)X^+_\kappa\Big)_{d\overline{\kappa}} = \sum_{F\nu\tau}\Big(&\underbrace{\langle d|a^{\overline{\tau}}_\nu|F\rangle}_{\delta_{\tau,-}\delta_{F,\overline{\nu}}\langle d|a^\dagger_\nu|\overline{\nu}\rangle} C^\tau_\nu(\omega-(F-E_{\overline{\kappa}}))\underbrace{\langle F|a^\tau_\nu X^+_\kappa|\overline{\kappa}\rangle}_{\langle\overline{\nu}|a_\nu|d\rangle X^+_{\kappa,\overline{\kappa}}}\\ &+ C^\tau_\nu(\omega-(E_d-E_{\overline{\kappa}}))\quad\underbrace{\langle d|a^\tau_\nu X^+_\kappa a^{\overline{\tau}}_\nu|\overline{\kappa}\rangle}_{\delta_{\tau,+}\delta_{\nu,\overline{\kappa}}\langle d|a^\dagger_{\overline{\kappa}}|\kappa\rangle X^+_{\kappa,0}\langle 0|a_{\overline{\kappa}}|\overline{\kappa}\rangle}\\ &+ C^\tau_\nu(E_d-E_{\overline{\kappa}}-\omega)^*\quad\underbrace{\langle d|X^+_\kappa a^{\overline{\tau}}_\nu a^\tau_\nu|\overline{\kappa}\rangle}_{X^+_{\kappa,\overline{\kappa}}(\delta_{\tau,+}\delta_{\nu,\kappa}\kappa^2+\delta_{\tau,-}\delta_{\nu,\overline{\kappa}})}\\ &+ \underbrace{\langle d|a^\tau_\nu|F\rangle}_{\delta_{\tau,+}\delta_{F,\overline{\nu}}\langle d|a^\dagger_{\overline{\kappa}}|\kappa\rangle} C^\tau_\nu(F-E_{\overline{\kappa}}-\omega)^*\underbrace{\langle F|X^+_\kappa a^{\overline{\tau}}_\nu|\overline{\kappa}\rangle}_{\delta_{\nu,\overline{\kappa}}X^+_{\kappa,0}\langle 0|a_{\overline{\kappa}}|\overline{\kappa}\rangle}\Big)\end{aligned}$$

$$\begin{aligned}\Big(\Sigma(\omega)X^+_\kappa\Big)_{d\overline{\kappa}} = &[C^-_\kappa(\omega-(E_{\overline{\kappa}}-E_{\overline{\kappa}}))+C^-_{\overline{\kappa}}(\omega-(E_\kappa-E_{\overline{\kappa}}))]X^+_{\kappa,\overline{\kappa}}\\ &+\overline{\kappa}C^+_{\overline{\kappa}}(\omega-(E_d-E_{\overline{\kappa}}))X^+_{\kappa,0}\\ &+[C^+_\kappa(E_d-E_{\overline{\kappa}}-\omega)^*+C^-_{\overline{\kappa}}(E_d-E_{\overline{\kappa}}-\omega)^*]X^+_{\kappa,\overline{\kappa}}\\ &+\overline{\kappa}C^+_{\overline{\kappa}}(E_\kappa-E_{\overline{\kappa}}-\omega)^*X^+_{\kappa,0}\end{aligned}$$

We proceed with the second matrix element for $s = +$:

$$\begin{aligned}\left(\Sigma(\omega)X^{+}_{\kappa}\right)_{\kappa 0} = \sum_{F\nu\tau}\Big(&\underbrace{\langle\kappa|a^{\overline{\tau}}_{\nu}|F\rangle C^{\tau}_{\nu}(\omega-(F-E_0))\langle F|a^{\tau}_{\nu}X^{+}_{\kappa}|0\rangle}_{(\delta_{\tau,+}\delta_{\nu,\overline{\kappa}}\delta_{F,d}\overline{\kappa}^2+\delta_{\tau,-}\delta_{\nu,\kappa}\delta_{F,0})C^{\tau}_{\nu}(\omega-(F-E_0))X^{+}_{\kappa,0}}\\ &+C^{\tau}_{\nu}(\omega-(E_\kappa-E_0))\quad\underbrace{\langle\kappa|a^{\tau}_{\nu}X^{+}_{\kappa}a^{\overline{\tau}}_{\nu}|0\rangle}_{\delta_{\tau,-}\delta_{\nu,\overline{\kappa}}\langle\kappa|a_{\overline{\kappa}}|d\rangle X^{+}_{\kappa,\overline{\kappa}}\langle\overline{\kappa}|a^{\dagger}_{\overline{\kappa}}|0\rangle}\\ &+C^{\tau}_{\nu}(E_\kappa-E_0-\omega)^{*}\ \underbrace{\langle\kappa|X^{+}_{\kappa}a^{\overline{\tau}}_{\nu}a^{\tau}_{\nu}|0\rangle}_{X^{+}_{\kappa,0}\delta_{\tau,+}(\delta_{\nu,\kappa}+\delta_{\nu,\overline{\kappa}})}\\ &+\underbrace{\langle\kappa|a^{\tau}_{\nu}|F\rangle}_{\langle\kappa|a_{\overline{\kappa}}|d\rangle}C^{\tau}_{\nu}(F-E_{\overline{\kappa}}-\omega)^{*}\quad\underbrace{\langle F|X^{+}_{\kappa}a^{\overline{\tau}}_{\nu}|0\rangle}_{\delta_{\tau,-}\delta_{\nu,\overline{\kappa}}\delta_{F,d}X^{+}_{\kappa,\overline{\kappa}}\langle\overline{\kappa}|a^{\dagger}_{\overline{\kappa}}|0\rangle}}\Big)\end{aligned}$$

$$\begin{aligned}\left(\Sigma(\omega)X^{+}_{\kappa}\right)_{\kappa 0} &= [C^{+}_{\overline{\kappa}}(\omega-(E_d-E_0))+C^{-}_{\kappa}(\omega-(E_0-E_0))]X^{+}_{\kappa,0}\\ &+\overline{\kappa}C^{-}_{\overline{\kappa}}(\omega-(E_\kappa-E_0))X^{+}_{\kappa,\overline{\kappa}}\\ &+[C^{+}_{\kappa}(E_\kappa-E_0-\omega)^{*}+C^{+}_{\overline{\kappa}}(E_\kappa-E_0-\omega)^{*}]X^{+}_{\kappa,0}\\ &+\overline{\kappa}C^{-}_{\overline{\kappa}}(E_d-E_0-\omega)^{*}X^{+}_{\kappa,\overline{\kappa}}\end{aligned}$$

We remain with $s = -$:

$$\begin{aligned}\left(\Sigma(\omega)X^{-}_{\kappa}\right)_{\overline{\kappa}d} = \sum_{F\nu\tau}\Big(&\underbrace{\langle\overline{\kappa}|a^{\overline{\tau}}_{\nu}|F\rangle C^{\tau}_{\nu}(\omega-(F-E_d))\langle F|a^{\tau}_{\nu}X^{-}_{\kappa}|d\rangle}_{(\delta_{\tau,+}\delta_{\nu,\kappa}\delta_{F,d}\kappa^2+\delta_{\tau,-}\delta_{\nu,\overline{\kappa}}\delta_{F,0})C^{\tau}_{\nu}(\omega-(F-E_d))X^{-}_{\kappa,d}}\\ &+C^{\tau}_{\nu}(\omega-(E_{\overline{\kappa}}-E_d))\quad\underbrace{\langle\overline{\kappa}|a^{\tau}_{\nu}X^{-}_{\kappa}a^{\overline{\tau}}_{\nu}|d\rangle}_{\delta_{\tau,+}\delta_{\nu,\overline{\kappa}}\langle\overline{\kappa}|a^{\dagger}_{\overline{\kappa}}|0\rangle X^{-}_{\kappa,\kappa}\langle\kappa|a_{\overline{\kappa}}|d\rangle}\\ &+C^{\tau}_{\nu}(E_{\overline{\kappa}}-E_d-\omega)^{*}\quad\underbrace{\langle\overline{\kappa}|X^{-}_{\kappa}a^{\overline{\tau}}_{\nu}a^{\tau}_{\nu}|d\rangle}_{X^{-}_{\kappa,d}\delta_{\tau,-}(\delta_{\nu,\kappa}\kappa^2+\delta_{\nu,\overline{\kappa}}\overline{\kappa}^2)}\\ &+\underbrace{\langle\overline{\kappa}|a^{\tau}_{\nu}|F\rangle}_{\langle\overline{\kappa}|a^{\dagger}_{\overline{\kappa}}|0\rangle}C^{\tau}_{\nu}(F-E_d-\omega)^{*}\quad\underbrace{\langle F|X^{-}_{\kappa}a^{\overline{\tau}}_{\nu}|d\rangle}_{\delta_{\tau,+}\delta_{\nu,\overline{\kappa}}\delta_{F,0}X^{-}_{\kappa,\kappa}\langle\kappa|a_{\overline{\kappa}}|d\rangle}}\Big)\end{aligned}$$

$$\begin{aligned}\left(\Sigma(\omega)X^{-}_{\kappa}\right)_{\overline{\kappa}d} &= [C^{+}_{\kappa}(\omega-(E_d-E_d))+C^{-}_{\overline{\kappa}}(\omega-(E_0-E_d))]X^{-}_{\kappa,d}\\ &+\overline{\kappa}C^{+}_{\overline{\kappa}}(\omega-(E_{\overline{\kappa}}-E_d))X^{-}_{\kappa,\kappa}\\ &+[C^{-}_{\kappa}(E_{\overline{\kappa}}-E_d-\omega)^{*}+C^{-}_{\overline{\kappa}}(E_{\overline{\kappa}}-E_d-\omega)^{*}]X^{-}_{\kappa,d}\\ &+\overline{\kappa}C^{+}_{\overline{\kappa}}(E_0-E_d-\omega)^{*}X^{-}_{\kappa,\kappa}\end{aligned}$$

And finally:

$$\begin{aligned}\left(\Sigma(\omega)X^-_\kappa\right)_{0\kappa} = \sum_{F\nu\tau}\Big(&\underbrace{\langle 0|a^{\overline{\tau}}_\nu|F\rangle C^\tau_\nu(\omega-(F-E_\kappa))\langle F|a^\tau_\nu X^-_\kappa|\kappa\rangle}_{\delta_{\tau,-}(\delta_{\nu,\kappa}\delta_{F,\kappa}+\delta_{\nu,\overline{\kappa}}\delta_{F,\overline{\kappa}})C^\tau_\nu(\omega-(F-E_\kappa))X^-_{\kappa,\kappa}}\\ &+C^\tau_\nu(\omega-(E_0-E_\kappa))\quad\underbrace{\langle 0|a^\tau_\nu X^-_\kappa a^{\overline{\tau}}_\nu|\kappa\rangle}_{\delta_{\tau,-}\delta_{\nu,\overline{\kappa}}\langle 0|a_{\overline{\kappa}}|\overline{\kappa}\rangle X^-_{\kappa,d}\langle d|a^\dagger_{\overline{\kappa}}|\kappa\rangle}\\ &+C^\tau_\nu(E_0-E_\kappa-\omega)^*\quad\underbrace{\langle 0|X^-_\kappa a^{\overline{\tau}}_\nu a^\tau_\nu|\kappa\rangle}_{X^-_{\kappa,\kappa}(\delta_{\tau,-}\delta_{\nu,\kappa}+\delta_{\tau,+}\delta_{\nu,\overline{\kappa}}\overline{\kappa}^2)}\\ &+\underbrace{\langle 0|a^\tau_\nu|F\rangle}_{\delta_{\tau,-}\langle 0|a_{\overline{\kappa}}|\overline{\kappa}\rangle}C^\tau_\nu(F-E_\kappa-\omega)^*\quad\underbrace{\langle F|X^-_\kappa a^{\overline{\tau}}_\nu|\kappa\rangle}_{\delta_{\nu,\overline{\kappa}}\delta_{F,\overline{\kappa}}X^-_{\kappa,d}\langle d|a^\dagger_{\overline{\kappa}}|\kappa\rangle}\Big)\end{aligned}$$

$$\begin{aligned}\left(\Sigma(\omega)X^-_\kappa\right)_{0\kappa} &= [C^+_\kappa(\omega-(E_\kappa-E_\kappa))+C^+_{\overline{\kappa}}(\omega-(E_{\overline{\kappa}}-E_\kappa))]X^-_{\kappa,\kappa}\\ &+\overline{\kappa}C^-_{\overline{\kappa}}(\omega-(E_0-E_\kappa))X^-_{\kappa,d}\\ &+[C^-_\kappa(E_0-E_\kappa-\omega)^*+C^+_{\overline{\kappa}}(E_0-E_\kappa-\omega)^*]X^-_{\kappa,\kappa}\\ &+\overline{\kappa}C^-_{\overline{\kappa}}(E_{\overline{\kappa}}-E_\kappa-\omega)^*X^-_{\kappa,d}\end{aligned}$$

For every κ and s value there are two non zero matrix elements that couple. We label the matrix element with the lower particle number in the ket vector $n=1$ and the other $n=2$.

The following table shows the index mapping for the different operators X^s_κ:

m	0	κ	$\overline{\kappa}$	d
$X^+_{\kappa,m}$	1		2	
$X^-_{\kappa,m}$		1		2

Now we introduce the abbreviation $C^s_{\kappa,nm}$ with $n,m\in\{1,2\}$ in order to label those coupling functions in ω. A tilde is used to indicate that the expression contains the factor $\overline{\kappa}$. A summary of all those derived functions is given in (2.87).

$$\begin{pmatrix}(\sum(\omega)X^s_\kappa)_1\\(\sum(\omega)X^s_\kappa)_2\end{pmatrix}=\begin{pmatrix}C^s_{\kappa,11}&\tilde{C}^s_{\kappa,12}\\\tilde{C}^s_{\kappa,21}&C^s_{\kappa,22}\end{pmatrix}\begin{pmatrix}X^s_{\kappa,1}\\X^s_{\kappa,2}\end{pmatrix}\tag{2.84}$$

We look now back at equation (2.75) and have the linear equation system for a given s and κ. Note that the matrix elements of the modified density operator with index two $[a^s_\kappa\overline{\sigma}]_2$ we get a factor of κ similar to (2.65):

$$\begin{pmatrix}\sigma_1\\\kappa\sigma_2\end{pmatrix}=\underbrace{\begin{pmatrix}i(E_1-\omega)+C^s_{\kappa,11}&\tilde{C}^s_{\kappa,12}\\\tilde{C}^s_{\kappa,21}&i(E_2-\omega)+C^s_{\kappa,22}\end{pmatrix}}_{=:D^s_\kappa}\cdot\begin{pmatrix}X^s_{\kappa,1}\\X^s_{\kappa,2}\end{pmatrix}\tag{2.85}$$

With the inversion of this matrix D^s_κ we get a closed form for the time evolution of the modified density operators:

$$\begin{pmatrix} X^s_{\kappa,1} \\ X^s_{\kappa,2} \end{pmatrix} = \frac{1}{\det(D^s_\kappa)} \begin{pmatrix} D^s_{\kappa,22} & -\overline{\kappa} C^s_{\kappa,12} \\ -\overline{\kappa} C^s_{\kappa,21} & D^s_{\kappa,11} \end{pmatrix} \cdot \begin{pmatrix} \sigma_1 \\ \kappa \sigma_2 \end{pmatrix} \tag{2.86}$$

with $D^s_{\kappa,ii}(\omega) := i(E_i - \omega) + C^s_{\kappa,ii}(\omega)$. In the first line the sign factor $-\overline{\kappa} = \kappa$ drops out whereas in the second line the factor κ remains but since we need this factor for the searched tilde expressions $\tilde{X}^s_{\kappa,2} = \kappa X^s_{\kappa,2}$, as used in equation (2.74), it drops out.

The analytical expressions for all functions $C^s_{\kappa,ij}$ are given below.

SUMMARIZED FORMULAS FOR SPIN-DEPENDENT SELF-ENERGY EQUATION

$$
\begin{aligned}
C^{+}_{\uparrow,11} &= \Gamma\left(u_L(\omega - E_{d0}) + u(\omega) + u(E_{\uparrow 0} - \omega)^* + u_L(E_{\uparrow 0} - \omega)^*\right) \\
C^{+}_{\uparrow,12} &= \Gamma\left(u_R(\omega - E_{\uparrow 0}) + u_R(E_{d0} - \omega)^*\right) \\
C^{+}_{\uparrow,21} &= \Gamma\left(u_L(\omega - E_{d\downarrow}) + u_L(E_{\uparrow\downarrow} - \omega)^*\right) \\
C^{+}_{\uparrow,22} &= \Gamma\left(u(\omega) + u_R(\omega - E_{\uparrow\downarrow}) + u(E_{d\downarrow} - \omega)^* + u_R(E_{d\downarrow} - \omega)^*\right) \\
C^{-}_{\uparrow,11} &= \Gamma\left(u(\omega) + u_L(\omega - E_{\downarrow\uparrow}) + u(E_{0\uparrow} - \omega)^* + u_L(E_{0\uparrow} - \omega)^*\right) \\
C^{-}_{\uparrow,12} &= \Gamma\left(u_R(\omega - E_{0\uparrow}) + u_R(E_{\downarrow\uparrow} - \omega)^*\right) \\
C^{-}_{\uparrow,21} &= \Gamma\left(u_L(\omega - E_{\downarrow d}) + u_L(E_{0d} - \omega)^*\right) \\
C^{-}_{\uparrow,22} &= \Gamma\left(u(\omega) + u_R(\omega - E_{0d}) + u(E_{\downarrow d} - \omega)^* + u_R(E_{\downarrow d} - \omega)^*\right) \\
C^{+}_{\downarrow,11} &= \Gamma\left(u(\omega - E_{d0}) + u_R(\omega) + u_L(E_{\downarrow 0} - \omega)^* + u(E_{\downarrow 0} - \omega)^*\right) \\
C^{+}_{\downarrow,12} &= \Gamma\left(u(\omega - E_{\downarrow 0}) + u(E_{d0} - \omega)^*\right) \\
C^{+}_{\downarrow,21} &= \Gamma\left(u(\omega - E_{d\uparrow}) + u(E_{\downarrow\uparrow} - \omega)^*\right) \\
C^{+}_{\downarrow,22} &= \Gamma\left(u_R(\omega) + u(\omega - E_{\downarrow\uparrow}) + u_L(E_{d\uparrow} - \omega)^* + u(E_{d\uparrow} - \omega)^*\right) \\
C^{-}_{\downarrow,11} &= \Gamma\left(u_L(\omega) + u(\omega - E_{\uparrow\downarrow}) + u_R(E_{0\downarrow} - \omega)^* + u(E_{0\downarrow} - \omega)^*\right) \\
C^{-}_{\downarrow,12} &= \Gamma\left(u(\omega - E_{0\downarrow}) + u(E_{\uparrow\downarrow} - \omega)^*\right) \\
C^{-}_{\downarrow,21} &= \Gamma\left(u(\omega - E_{\uparrow d}) + u(E_{0d} - \omega)^*\right) \\
C^{-}_{\downarrow,22} &= \Gamma\left(u_L(\omega) + u(\omega - E_{0d}) + u_R(E_{\uparrow d} - \omega)^* + u(E_{\uparrow d} - \omega)^*\right)
\end{aligned}
\tag{2.87}
$$

$$
\begin{aligned}
C^{+}_{\kappa,11} &= [C^{+}_{\overline{\kappa}}(\omega - (E_d - E_0)) + C^{-}_{\kappa}(\omega) + C^{+}_{\kappa}(E_\kappa - E_0 - \omega)^* + C^{+}_{\overline{\kappa}}(E_\kappa - E_0 - \omega)^*] \\
\tilde{C}^{+}_{\kappa,12} &= \overline{\kappa}[C^{-}_{\overline{\kappa}}(\omega - (E_\kappa - E_0)) + C^{-}_{\overline{\kappa}}(E_d - E_0 - \omega)^*] \\
\tilde{C}^{+}_{\kappa,21} &= \overline{\kappa}[C^{+}_{\overline{\kappa}}(\omega - (E_d - E_{\overline{\kappa}})) + C^{+}_{\overline{\kappa}}(E_\kappa - E_{\overline{\kappa}} - \omega)^*] \\
C^{+}_{\kappa,22} &= [C^{-}_{\kappa}(\omega) + C^{-}_{\overline{\kappa}}(\omega - (E_\kappa - E_{\overline{\kappa}})) + C^{+}_{\kappa}(E_d - E_{\overline{\kappa}} - \omega)^* + C^{-}_{\overline{\kappa}}(E_d - E_{\overline{\kappa}} - \omega)^*] \\
C^{-}_{\kappa,11} &= [C^{+}_{\kappa}(\omega) + C^{+}_{\overline{\kappa}}(\omega - (E_{\overline{\kappa}} - E_\kappa)) + C^{-}_{\kappa}(E_0 - E_\kappa - \omega)^* + C^{+}_{\overline{\kappa}}(E_0 - E_\kappa - \omega)^*] \\
\tilde{C}^{-}_{\kappa,12} &= \overline{\kappa}[C^{-}_{\overline{\kappa}}(\omega - (E_0 - E_\kappa)) + C^{-}_{\overline{\kappa}}(E_{\overline{\kappa}} - E_\kappa - \omega)^*] \\
\tilde{C}^{-}_{\kappa,21} &= \overline{\kappa}[C^{+}_{\overline{\kappa}}(\omega - (E_{\overline{\kappa}} - E_d)) + C^{+}_{\overline{\kappa}}(E_0 - E_d - \omega)^*] \\
C^{-}_{\kappa,22} &= [C^{+}_{\kappa}(\omega) + C^{-}_{\overline{\kappa}}(\omega - (E_0 - E_d)) + C^{-}_{\kappa}(E_{\overline{\kappa}} - E_d - \omega)^* + C^{-}_{\overline{\kappa}}(E_{\overline{\kappa}} - E_d - \omega)^*]
\end{aligned}
$$

Explicit current formula

Here we also derive an explicit current formula for the single quantum dot with spin dependent coupling. Starting point is equation (2.37)

$$J_\alpha = 2\sum_\kappa \mathrm{Re}\left(\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\Gamma_{\alpha\kappa}n_\alpha^{(+)}(\omega)\mathrm{Tr}_s\Big\{a_\kappa X_\kappa^+(\omega)\Big\} - \Gamma_{\alpha\kappa}n_\alpha^{(-)}(\omega)\mathrm{Tr}_s\Big\{a_\kappa^\dagger X_\kappa^-(-\omega)\Big\}\right)$$

With assuming the spin down hopping to and from the right lead to be zero we just have to consider the spin up contributions when looking at the current to the right lead.

$$J_R = 2\mathrm{Re}\left(\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\Gamma_{R\uparrow}n_R^{(+)}(\omega)\mathrm{Tr}_s\Big\{a_\uparrow X_\uparrow^+(\omega)\Big\} - \Gamma_{R\uparrow}n_R^{(-)}(\omega)\mathrm{Tr}_s\Big\{a_\uparrow^\dagger X_\uparrow^-(-\omega)\Big\}\right)$$

Evaluation of the trace

$$\mathrm{Tr}_s\Big\{a_\kappa^{\bar{s}}X_\kappa^s\Big\} = \begin{cases}\langle 0|a_\kappa|\kappa\rangle\langle\kappa|X_\kappa^+|0\rangle + \langle\overline{\kappa}|a_\kappa|d\rangle\langle d|X_\kappa^+|\overline{\kappa}\rangle, & s=+\\ \langle\kappa|a_\kappa^\dagger|0\rangle\langle 0|X_\kappa^-|\kappa\rangle + \langle d|a_\kappa^\dagger|\overline{\kappa}\rangle\langle\overline{\kappa}|X_\kappa^-|d\rangle, & s=-\end{cases}$$

$$\phi_\kappa^+(\omega) := \mathrm{Tr}_s\Big\{a_\kappa X_\kappa^+(\omega)\Big\} = \frac{1}{\det(D_\kappa^+)}\Big(D_{\kappa,22}^+\overline{\sigma}_{00} + C_{\kappa,12}^+\overline{\sigma}_{\overline{\kappa}\overline{\kappa}} + \underbrace{\kappa\kappa}_{=1}\big(C_{\kappa,21}^+\overline{\sigma}_{00} + D_{\kappa,11}^+\overline{\sigma}_{\overline{\kappa}\overline{\kappa}}\big)\Big)$$

$$\phi_\kappa^-(-\omega) := \mathrm{Tr}_s\Big\{a_\kappa X_\kappa^+(-\omega)\Big\} = \frac{1}{\det(D_\kappa^-)}\Big(D_{\kappa,22}^-\overline{\sigma}_{\kappa\kappa} + C_{\kappa,12}^-\overline{\sigma}_{dd} + \kappa\kappa\big(C_{\kappa,21}^-\overline{\sigma}_{\kappa\kappa} + D_{\kappa,11}^-\overline{\sigma}_{dd}\big)\Big)$$

The ϕs correspond with those introduced in the section about the current 2.3.1. So we have here the following relations

Green's functions

$$\phi_\kappa^+(\omega) = \frac{1}{\det(D_\kappa^+)}\Big(\big(D_{\kappa,22}^+(\omega) + C_{\kappa,21}^+(\omega)\big)\overline{\sigma}_{00} + \big(C_{\kappa,12}^+(\omega) + D_{\kappa,11}^+(\omega)\big)\overline{\sigma}_{\overline{\kappa}\overline{\kappa}}\Big) \tag{2.88}$$

$$\phi_\kappa^-(-\omega) = \frac{1}{\det(D_\kappa^-)}\Big(\big(D_{\kappa,22}^-(-\omega) + C_{\kappa,21}^-(-\omega)\big)\overline{\sigma}_{\kappa\kappa} + \big(C_{\kappa,12}^-(-\omega) + D_{\kappa,11}^-(-\omega)\big)\overline{\sigma}_{dd}\Big) \tag{2.89}$$

$$iG_\kappa^r(\omega) = \phi_\kappa^+(\omega) + \phi_\kappa^-(\omega) \tag{2.90}$$

3 Numerical results

In this part I present numerical results obtained by a Matlab implementation of the Born-Markov master equation and the self-consistent Born approach as described in section 1.5 respectively in section 2.4. I will examine the influence of the applied potential, temperature in the bath and the influence of the coupling strength on the steady state of the single quantum dot for the two methods.

Next I will present the calculated current characteristics, compare them with other methods and show the Coulomb blockade due to spin dependent coupling in a quantum dot respectively due to interference phenomena in a cyclic triple quantum dot depending on an applied gate voltage. In the end I show results for the Born-Markov approach with and without the secular approximation in order to deal with quasi-degenerate states.

As explained in the introduction of chapter 1 the method of Born-Markov master equation is apt for all kind of system Hamiltonians that can be exact solved or at least solved for lowest energy levels. For my calculations I restricted all central quantum systems to the Hubbard model[1] since the main focus was to solve strongly correlated electron systems under the influence of the Markovian baths. The corresponding Hamiltonian H_s used to describe the systems is defined in (1.5). Note that the system is defined to be particle hole symmetric for zero on-site energy ξ_0. The coupling from the baths to the quantum system is assumed to be weak (weak coupling limit) and the attached baths are described by a flat density distribution (wide band limit), which allow us to use the analytic expression for the bath correlation function (1.102).

3.1 Single quantum dot

Temperature and potential dependence for Born-Markov approximation - instabilities

We start our discussion of the features of the just introduced methods with the most simple model of a single quantum dot as here some analytical solutions are available. The Fock basis represents the eigenbasis. For zero on-site energy $\xi = 0$ and switched on interaction $U > 0$ the one particle sector spans the eigenspace of the groundstate. If the system is isolated from the environment ($H_1 = 0$), we will find only those degenerate groundstates to be populated. Any prepared magnetization will be conserved. Now we attach one bath, characterized by it's potential $\mu = 0$ with respect to the system and it's inverse temperature β. The coupling strength to the system is given by $\Gamma = 1$. The solution of the

[1] When taking into account the non-interacting bath we gain the Anderson impurity model.

Born-Markov master equation shows that the occupation probability changes according to the temperature as depicted in figure 3.1. The energy gap to the first excited level for that system is one energy unit which thermally can be overcome with increasing temperature. For low temperatures the Boltzmann factor gets so small that the steady state solution is not unique any more. I implemented a procedure that projects a randomly chosen initial state on that zero eigenspace which corresponds to the time evolution for infinite time. The observed magnetization depends on the initial state. This strategy also breaks down for higher values of β due to division by zero in the Gram-Schmidt orthonormalization.

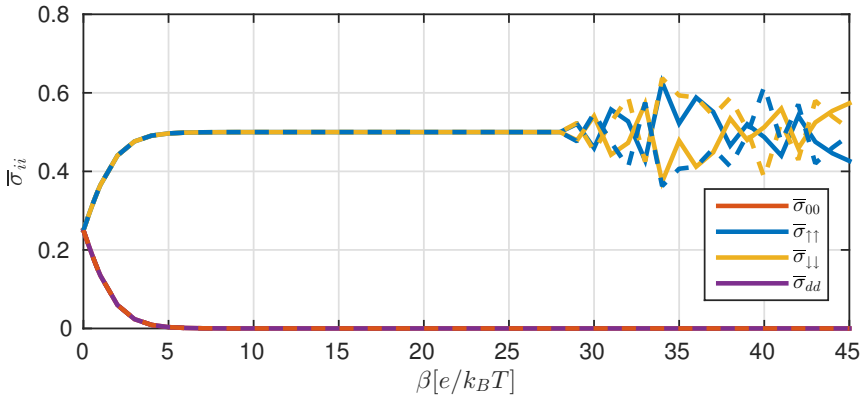


Figure 3.1: Densities of the system eigenstates as a function of inverse temperature for the single quantum dot attached to a bath with zero potential: $U = 2|b|$, $\Gamma = |b|$. The variation of spin states for high values of β origin from the randomly chosen initial state which were used to reach a steady state, since in that region no unique solution could be found.

In a next step we examine the influence of an applied bath potential V_b on the occupation density for different inverse temperatures (see figure 3.2). The particle occupation changes when the applied potential is in the range of the energy gap of the system. The shape of this transition strongly depends on the temperature. We can see also how the Born-Markov master equation method fails for low temperatures (high β) and low applied potential as the steady state is not unique.

We also want to discuss the influence of the coupling strength on the occupation density and see an important difference of the two methods, namely the Born-Markov master equation approach and it's self-consistent enhancement. Figure 3.3 shows the occupation densities of a quantum dot for a small applied potential $V_b = 0.2$ (to seperate zero and double occupied states) as a function of coupling strength calculated with both methods. The coupling strength in the Born-Markov master equation approach has no influence on the occupation density whereas the self-consistent Born master equation accounts for the level broadening that results from coupling to the bath.

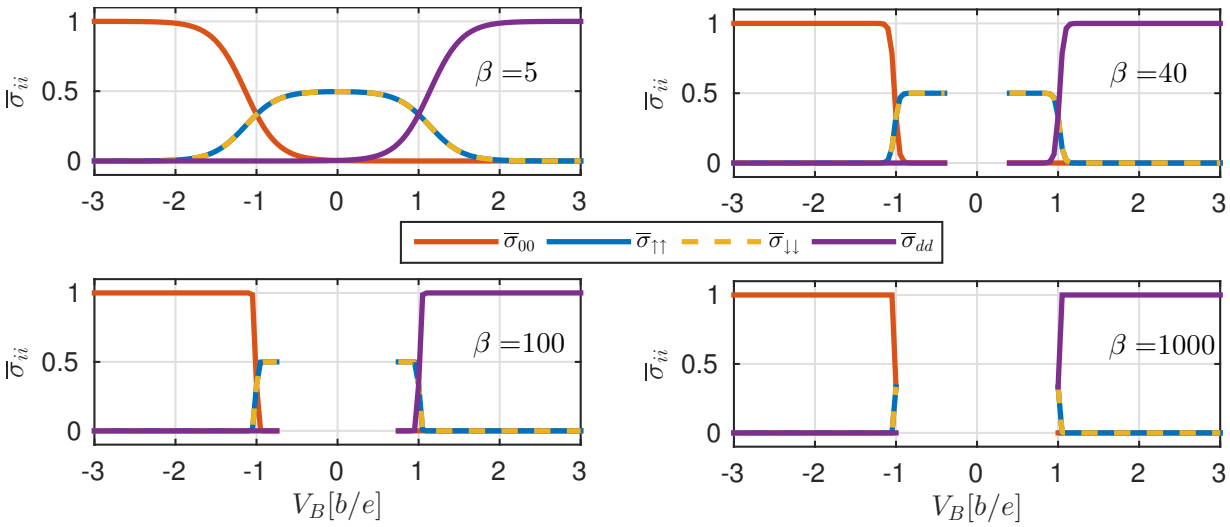


Figure 3.2: Densities of the system eigenstates of the single quantum dot attached to a bath as a function of bias voltage V_b: $U = 2|b|$, $\Gamma = |b|$.

3.1.1 Non-interacting quantum dot - current voltage characteristics

Now we want to attach two leads $\alpha \in \{L, R\}$ to the quantum dot and calculate the current. The potential difference of the central system to the leads shall be symmetric $\mu_L = -\mu_R$, so we have for the applied bias voltage $V_B = \mu_L - \mu_R = 2\mu_L$.

We are going to evaluate the current formulas given in section 2.3.1 and first look at the non-interacting quantum dot with on-site energy $\xi_0 = 0$. All states are degenerate thus the density matrix in steady state comes out to be $\overline{\sigma}_{ab} = \frac{1}{4}\delta_{ab}$. The current operator in the Born-Markov current formula 2.43 is

$$\begin{aligned} I_{00} &= -2\Gamma n_L^+(0) \\ I_{dd} &= 2\Gamma n_L^-(0) = 2\Gamma(1 - n_L^+(0) \\ I_{\kappa\kappa} &= \Gamma(n_L^-(0) - n_L^+(0)) = \Gamma(1 - 2n_L^+(0)) \end{aligned}$$

So evaluating the trace in the current formula yields

$$\langle I_L \rangle = \mathrm{Tr}(I\overline{\sigma}) = \Gamma(1 - 2n_L^+(0)) = \Gamma\left(1 - \frac{2}{e^{-\beta\mu_L} + 1}\right) = \Gamma \tanh(\frac{\beta\mu}{2}) \tag{3.1}$$

Although in the non-interacting case for $T = 0$ the current for such a quantum dot can be calculated analytically for example with non-equilibrium Green's function method [36] and is given by

$$\langle I \rangle = \frac{2\Gamma}{\pi}\mathrm{Arctan}(\frac{\mu}{\Gamma}) \tag{3.2}$$

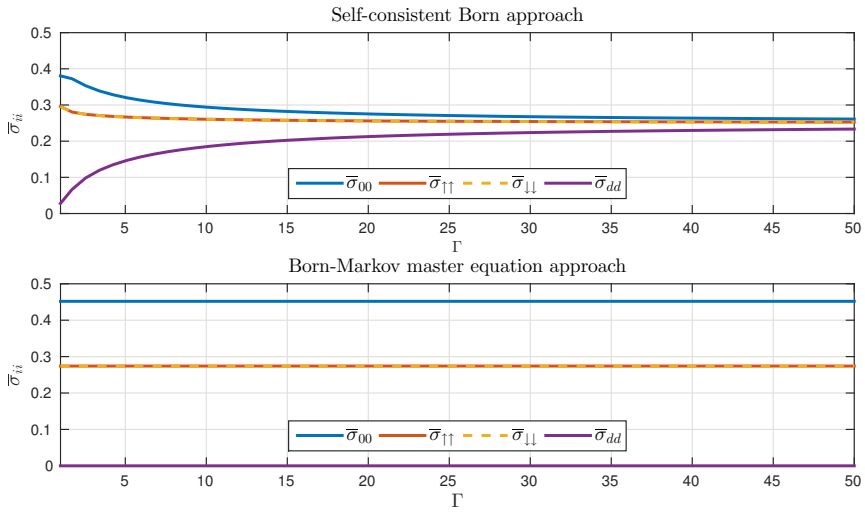


Figure 3.3: Densities of the system eigenstates of the single quantum dot attached to a bath as a function of the coupling strength Γ calculated with the Born Markov and the self-consistent Born approach with parameters $U = 2$, $V_b = 0.1$, $\xi_0 = 1$.

As visualized in figure 3.4 we see clearly that for $U = 0$ the Born-Markov current is wrong and misses the influence of the coupling constant Γ. In contrast the self-consistent Born approach reproduces the level broadening as it recovers the same Green's functions as with the exact non-equilibrium Green's functions approach ([26])

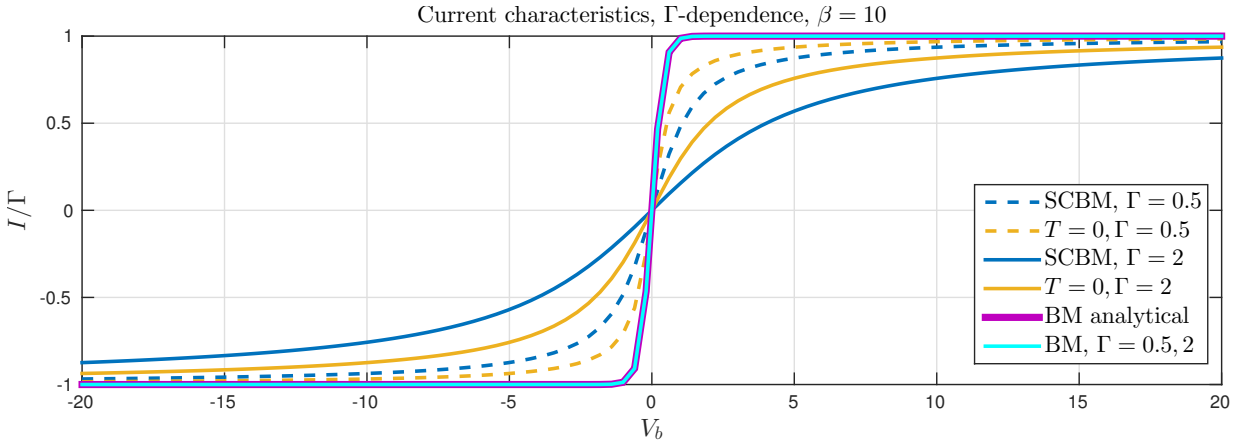


Figure 3.4: Current voltage characteristics of the non-interacting quantum dot for different temperatures and different coupling strengths calculated with different methods and compared to the exact zero temperature result.

3.2 Interacting quantum dot

In this section we approach the interacting quantum dot as treated also in the Anderson impurity model. For on-site energy $\xi_0 \neq 0$ we have three system levels. Here we can see a typical single electron transport effect, the so-called Coulomb blockade, a situation when the current can't rise though voltage increases. Reason for this is the one-electron energy level that becomes populated when current starts to flow. With increasing voltage additional charge carriers would like to participate in transport but the already present electron hinders those other electrons from entering due to Coulomb repulsion. Applying a sufficient high voltage the double occupied energy state gets favourable and the current can increase again to reach it's maximum value that's proportional to the coupling strength Γ. Figure 3.5 shows the result for the two introduced methods. One can clearly see how the first current plateau corresponds with the occupation of the one particle sector.

Comparison of methods

I also want to present a comparison of different methods. In figure 3.6 several different methods approach the same problem.

- Mean field method (Mf)
- Steady state Cluster Perturbation Theory (stsCPT)
- Master equation enhanced cluster perturbation theory (meCPT)
- Born-Markov master equation approach (BM)
- Self-consistent Born-Markov approach (SCB)

As depicted in that plot the mean field method basically averages over the plateau and doesn't resolve the Coulomb blockade for applied voltages in the energy range around the first excited state. Steady state cluster perturbation theory correctly reproduces the first excitation but not the plateau. It's enhancement - a combination of Born-Markov master equation and cluster perturbation theory as described in [32] repairs that effect. A similar situation is present for the master equation approach in connection with the level broadening. Whereas the Born-Markov approach doesn't account for the level broadening connected to the coupling strength, it's enhancement, the self-consistent Born approach indeed incorporates the influence of the bath coupling.

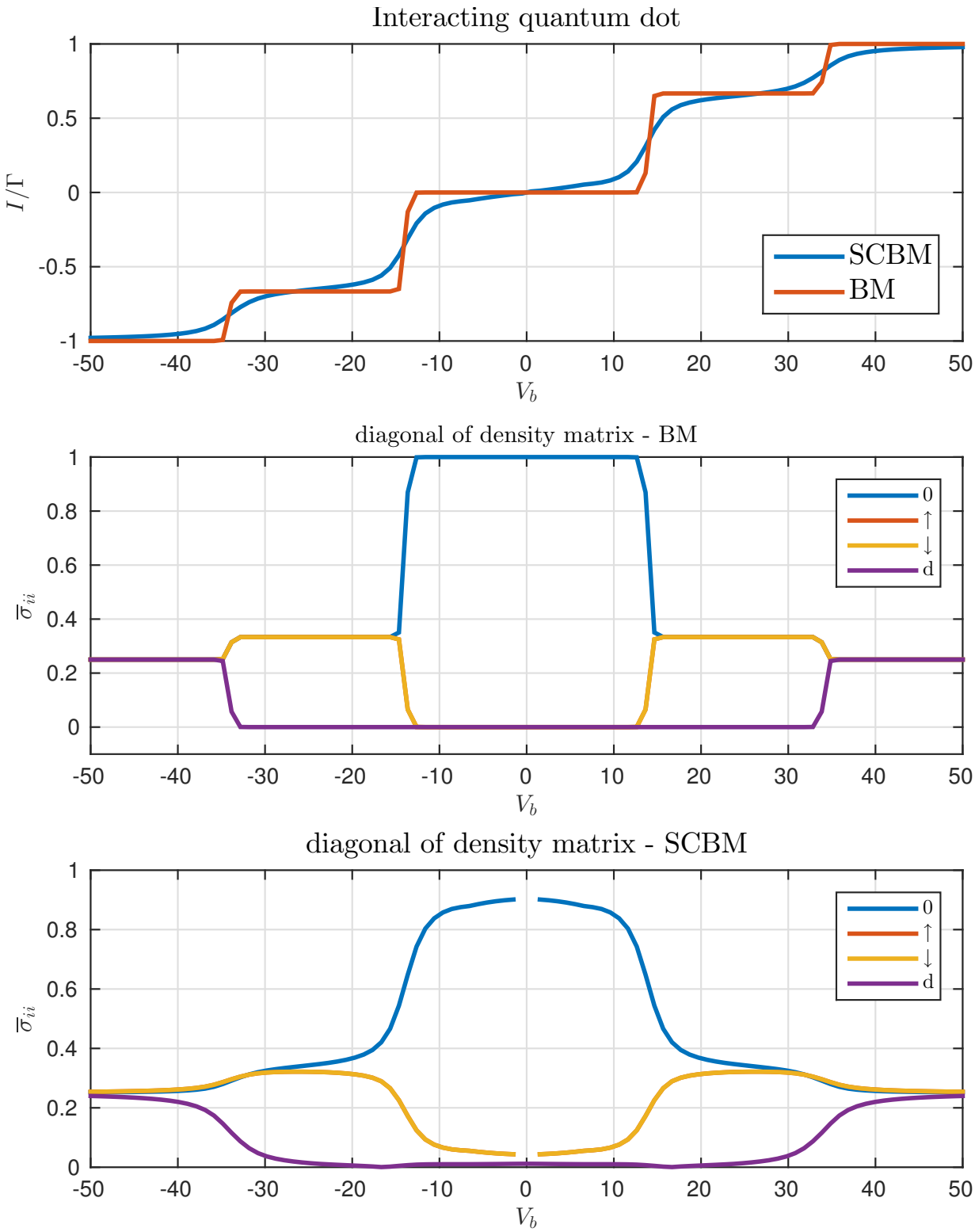


Figure 3.5: Current voltage characteristics and occupation densities of the interacting quantum dot for Born-Markov and self-consistent Born approach.

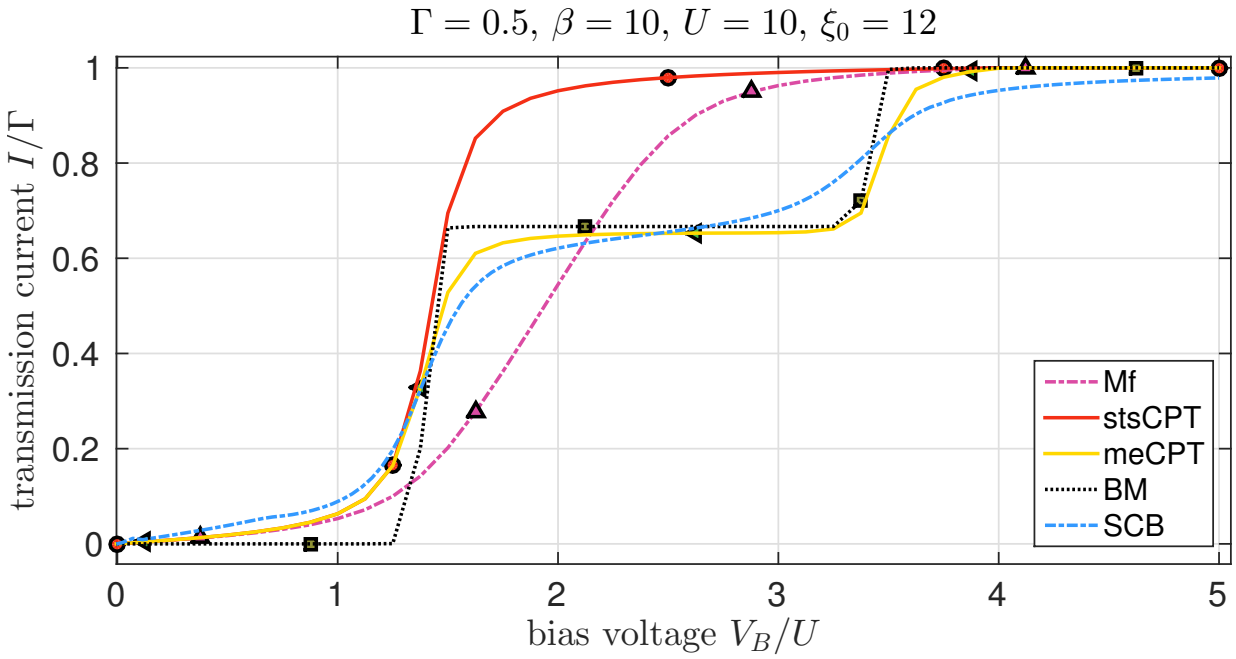
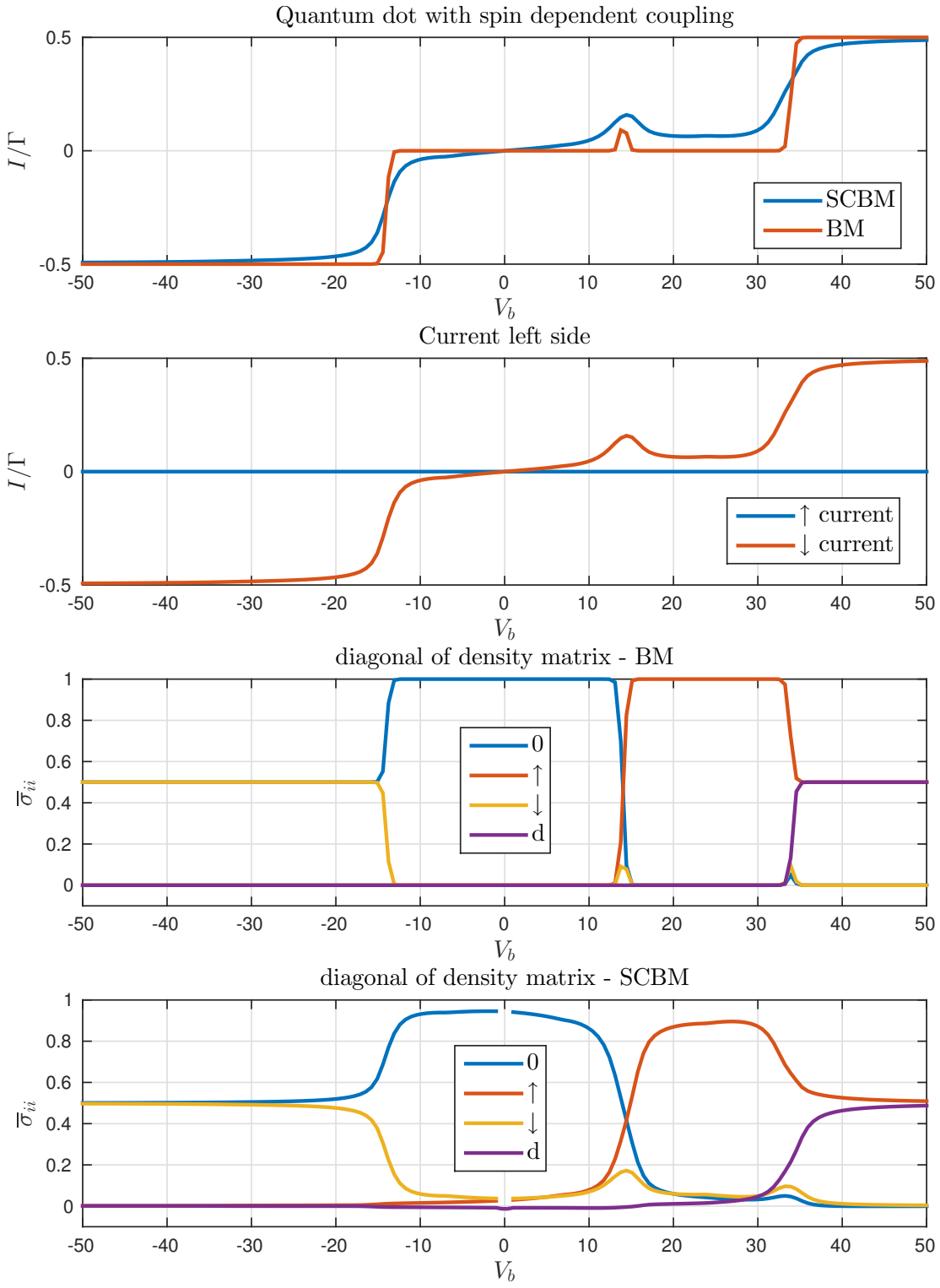


Figure 3.6: Current voltage characteristics of the interacting quantum dot for different methods

3.3 Spin dependent coupling

As mentioned in the introduction we will have a look at the realisation of a negative conductance in form of a quantum dot with spin dependent coupling, where spin up coupling to the right lead is forbidden $\Gamma_{R\uparrow} = 0$. In figure 3.7 we see the current characteristics and the occupation densities for both methods (BM and SCB). Since spin up electrons are not allowed to tunnel to the right lead, the one particle state fills up with those electrons that hinder spin down electrons to pass through. This results in a total current blockade in the BM approach whereas in the SCB approach it finds a minimum. For sufficient high bias voltage the two particle sector gets populated so the spin down electrons can pass through again.

The same results are also reproduced in [32] where the same setup has been examined with the before mentioned master equation enhanced cluster perturbation theory.



$U = 10$, $\xi_0 = 12$, $\Gamma_{L\kappa} = \Gamma_{R\downarrow} = 0.5$, $\Gamma_{R\uparrow} = 0$, $\beta = 10$

Figure 3.7: Current voltage characteristics and occupation densities of the interacting quantum dot with spin dependent coupling.

3.4 Interference in triple quantum dot

Beside spin dependent coupling there is also another mechanism that leads to the previously examined blocking effect respectively the occurrence of negative differential conductance. We can observe this phenomena also in quantum ring system due to destructive interference of degenerate system eigenstates. More discussion about the symmetric properties of cyclic quantum systems and the consequential quantum interference effects are given in the paper of Darau et. al. [15], where also the same method (Born-Markov master equation) is used to calculate the steady state current of a benzene molecule.

I examined a smaller ring system, the triple quantum dot, where I could reproduce the blocking effect and made a parameter study to see the non symmetric behaviour with respect to the gate voltage when compared to the six-site system. This system can be regarded as single electron transistor by tuning the system via the on-site energy ξ_0 which I refer to as gate voltage V_g.

The application of self-consistent Born to that system was not implemented yet so the presented results are calculated using the BM-method with a lacking of level broadening effects. This system was also examined with the before mentioned meCPT approach [32].

3.4.1 Born-Markov master equation results

In figure 3.8 you can see the differential conductance, the current and the average particle number of the triple quantum dot as a function of bias voltage and gate voltage. In figure 3.9 the current characteristics and the relevant occupation densities of the lowest energy states of the two and three particle sector are given.

3.4.2 Quasi-degenerate states

In this section I want shortly discuss the treatment of quasi-degenerate states. In section 1.6.3 we already discussed that a valid solution of the Born-Markov equation in the sense of a valid density matrix is just guaranteed when applying the secular approximation which restricts the treatment of the dynamics to the energy block diagonal subspace of the density matrix. If we consider a perturbation of our system that in a realistic system will always be probable, for example by varying the on-site energy of one system site we split our spectrum and formally degenerate states become quasi-degenerate. Since in the secular approximation we now suddenly neglect interactions between states of the now separated energies we also don't account for their dynamics that may be important for the current characteristics.

In figure 3.10 I have depicted such a situation where the on-site energy ξ_3 of the system site that is not connected to any bath is slightly changed. I performed the calculations with and without the secular approximation (see also [15]). The drawbacks of not staying in Lindblad formalism when not applying secular approximation are not yet visible for that small perturbation but one can observe negative currents for positive applied voltage, non-zero current for zero applied voltage and negative eigenvalues of the steady state density

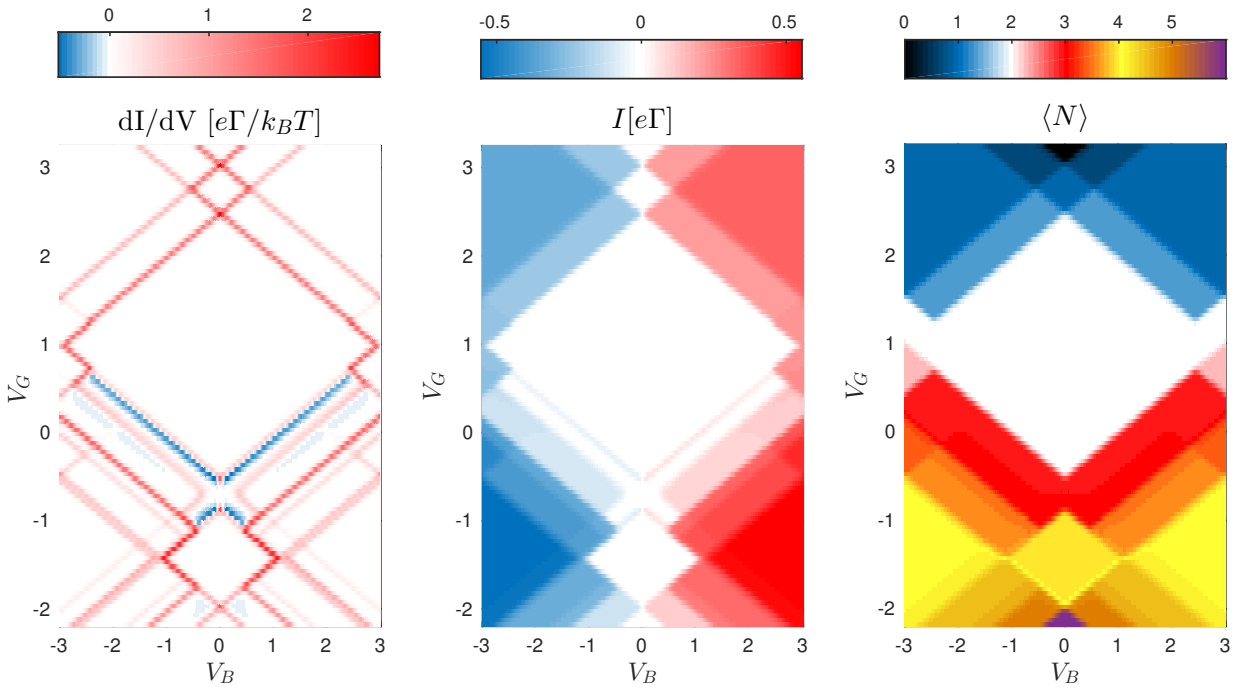


Figure 3.8: Stability diagram for the triple quantum dot obtained with Born-Markov master equation approach. From left to right there are plotted differential conductance dI/dV, current I and average particle number $\langle N \rangle$ as a function of bias voltage V_B and gate voltage V_G. Parameters were chosen to be: $U = 2|b|$, $\Gamma = 0.05|b|$, $\beta = 50$

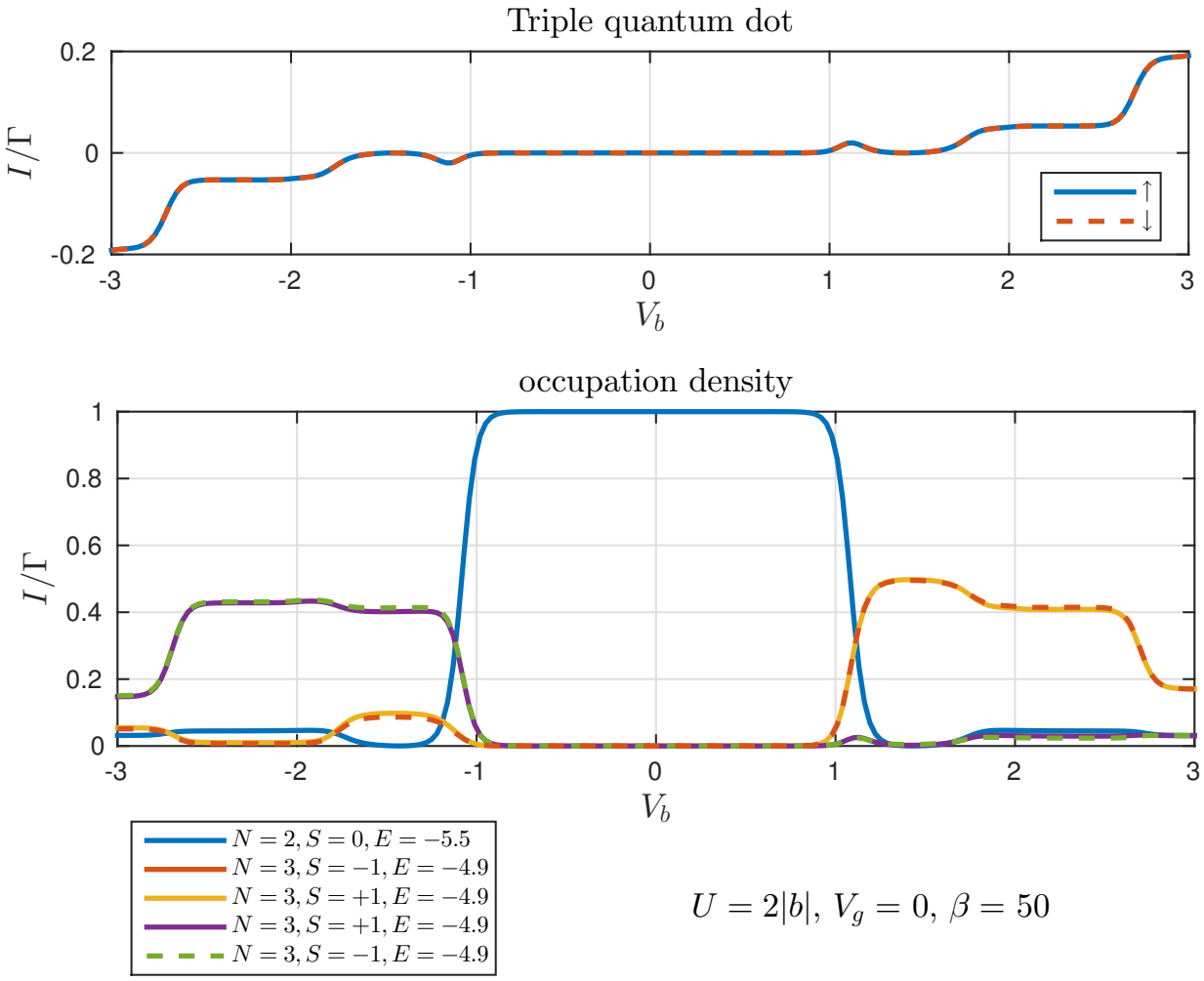


Figure 3.9: Current characteristics for the triple quantum dot obtained with Born-Markov master equation approach in the same setup as in figure 3.8. In the lower panel the relevant density matrix occupation probabilities are given.

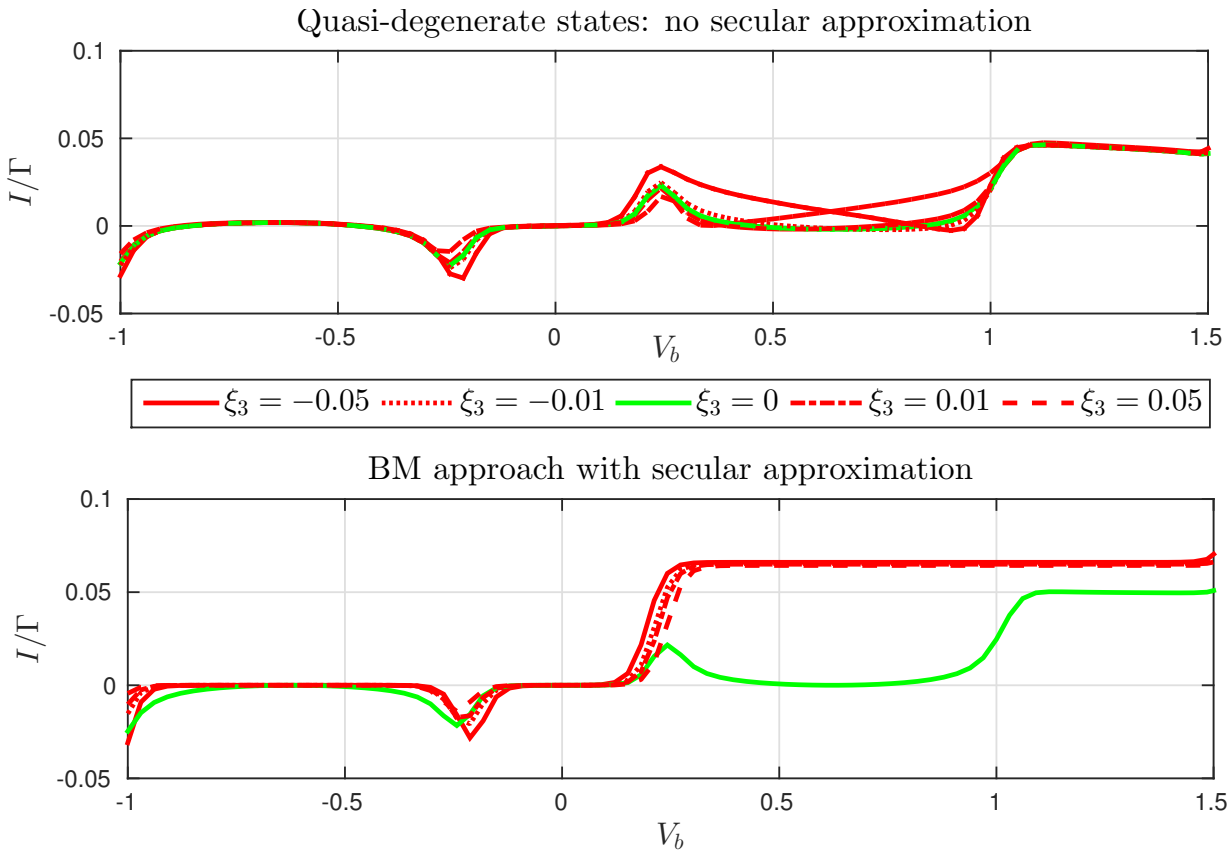


Figure 3.10: Current characteristics for the triple quantum dot obtained with Born-Markov master equation approach in the same setup as in figure. In the lower panel the relevant density matrix occupation probabilities are given.

matrix if the gap of the quasi-degeneracy gets bigger. Nevertheless this method still accounts for the correlations between the quasi-degenerate states and provides a continuation from the degenerate to the non-degenerate case.

Conclusion

In this work the detailed derivation of the Born-Markov master equation (BE) and it's extension the self-consistent Born approach (SCB) were presented. These methods are well apt to describe small open quantum systems in non-equilibrium. The environment of the quantum system was modelled by thermally equilibrated non-interacting baths that were assumed to be in the wide band limit (constant spectral density function), so that an analytic derivation of the bath hybridization function was feasible.

The system was described by the Hubbard model to investigate correlation effects of electrons in the current transport such as Coulomb blocking and negative conductance. For a single site system this open quantum system corresponds with the Anderson impurity model.

I discussed the criteria for a unique and valid solution which is gained with the secular approximation in the Lindblad formalism and examined the continuation to quasi-degenerate states. Since here it's not possible to neglect the interference effects of these hardly separated energy states a non secular solution provides a good extension. The errors of non-zero current at zero bias voltage and invalid steady states can be neglected for small energy gaps of the quasi-degenerate states.

For both approaches the formulas for the steady state and for the current were given and explicitly derived for the quantum dot with special focus at spin-dependent coupling in the SCB approach. I could show the correspondence of the current formula in the SCB approach with the Meir-Wingreen formula.

Both methods were implemented in Matlab and applied to describe the current characteristics of a single quantum dot and a triple quantum dot. I discussed the improvement of the SCB method concerning level broadening effects that were not reproduced in the BM method. In order to compare both methods with analytical current formulas I addressed a non-interacting system for which the SCB method reproduces the exact result.

Further I showed that the Born-Markov approach runs in numerical problems at low temperatures. A coupling strength dependence in terms of the current shape is just present at the SCB method since in the Born-Markov approach the corresponding factor Γ drops out. I also compared different current-voltage characteristics obtained from different numerical methods applied on the single Anderson impurity problem and discussed the benefits of self-consistent Born approach and master equation enhanced cluster perturbation theory ([32]) in terms of the correct reproduction of the current levels and the level broadening. Finally I could reproduce the quantum phenomena of Coulomb blockade and negative conductance in two different setups, in one case for spin dependent hopping and in the other case in a cyclic quantum system, hosting interference effects.

Acknowledgements

First of all, I want to thank my supervisor Prof. Dr. Wolfgang von der Linden for introducing me to the topic of open quantum systems in non-equilibrium. He helped me a lot in working through the relevant papers, which incorporated the pathway to the introduced methods, and gave me the freedom to follow my own ideas. I also liked to approach this topic from different formalisms since they helped me to accomplish my understanding.

I also want to thank my colleagues and friends at university who cheer up my working day during lunch or during the coffee breaks. I especially want to thank the colleagues in the office, Antonius Dorda, Marin Nuss, Christoph Heil, Delia Fugger and Christopher Albert for their help on open question, their cooperation and just for having a sympathetic ear.

Last but not least I want to thank my parents, siblings, my grandma and all other relatives who supported me not only financially but as a family who encourages and motivates me and gave me mentally support when days work not so well. And special thanks to my nieces who teach me explain simple things in a simple way.

4 Appendix

4.1 Fourier transform and incomplete Fourier transform

4.1.1 Fourier transform

The Fourier transform of a function f in time space shall be defined:

$$f(t) := \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} e^{i\omega' t} F(\omega'). \tag{4.1}$$

For the inverse Fourier transform we have:

$$F(\omega') = \int_{-\infty}^{\infty} dt e^{-i\omega' t} f(t). \tag{4.2}$$

Fourier transform of the complex conjugated Be aware that the operations Fourier transform and complex conjugation don't commute though a factor of minus one in the arguments remains preserved. Let $F(\omega')$ be the Fourier transform of $f(t)$, then we have

$$\mathcal{F}[f(-t)] = \int_{-\infty}^{\infty} dt e^{-i\omega' t} f(-t) \overset{t\to -t'}{=} \int_{-\infty}^{\infty} dt' e^{i\omega' t'} f(t') = F(-\omega') \tag{4.3}$$

$$\mathcal{F}[f(t)^*] = \int_{-\infty}^{\infty} dt e^{-i\omega' t} f(t)^* = \left(\int_{-\infty}^{\infty} dt e^{i\omega' t} f(t) \right)^* = (F(-\omega'))^* \tag{4.4}$$

$$= (\mathcal{F}[f(-t)])^* \tag{4.5}$$

So symmetric functions remain symmetric in Fourier space.

4.1.2 Complex Laplace transform

We define the incomplete Fourier transform (Laplace transform):

$$f(\omega) = \mathfrak{L}[f(t)] = \int_0^{\infty} f(t) e^{i(\omega + i0^+)t} dt. \tag{4.6}$$

The $i0^+$ factor is a convergence factor that allows for instance to integrate constant functions. We use the abbreviation ω^+ which will indicate that in this integral there is this convergence factor of $e^{-0^+|t|}$ no matter what the actual sign before the ω is. We use $f(\omega)$ to describe the Laplace transform and $F(\omega)$ to describe the Fourier transform of $f(t)$.

Laplace transform of the complex conjugated In contrast to the Fourier transform a factor of minus one in the argument is not preserved though the relation for the complex conjugation is still valid:

$$\mathfrak{L}\left[f(-t)\right] = \int_0^\infty f(-t)e^{i\omega^+ t}dt \overset{t\to -t'}{=} \int_{-\infty}^0 f(t')e^{-i\omega^+ t'}dt' \neq f(-\omega) \tag{4.7}$$

$$\mathfrak{L}\left[f(t)^*\right] = \int_0^\infty f^*(t)e^{i\omega^+ t}dt = \left(\int_0^\infty f(t)e^{-i\omega^+ t}dt\right)^* = (f(-\omega))^* \tag{4.8}$$

Derivation rule for the complex Laplace transform

$$L[f'(t)] = \left[e^{i\omega^+ t}f(t)\right]_0^\infty - (i\omega - 0^+)\int_0^\infty e^{i\omega^+ t}f(t)dt \tag{4.9}$$

$$- i\omega f(\omega) - f(0) \tag{4.10}$$

The pre-factor $-i\omega$ arises from the complex exponential.

Shift theorem For the Laplace (Fourier) transform of a function $f(t)$ multiplied with an exponential term we can write

$$\mathfrak{L}[f(t)e^{iat}] = \int_0^\infty e^{i\omega t}f(t)e^{iat}dt \tag{4.11}$$

$$= \int_0^\infty e^{i(\omega+a)t}f(t)dt = F(\omega + a) \tag{4.12}$$

with $F(\omega) = \mathfrak{L}[f(t)]$.

Convolution theorem We show that the Laplace (Fourier) transform of a given convolution of the functions $f(t)$ and $g(t)$ can be expressed as the product of the individual Laplace (Fourier) transforms of the two functions:

$$\mathfrak{L}[f * g] = \int_0^\infty dt e^{i\omega^+ t}\int_0^t dt' f(t-t')g(t') \tag{4.13}$$

$$= \int_0^\infty dt e^{i\omega^+ t}\int_0^\infty dt' \mathbb{1}(t-t')f(t-t')g(t') \tag{4.14}$$

$$= \int_0^\infty dt' e^{i\omega^+ t'}\int_0^\infty dt e^{i\omega^+(t-t')}\mathbb{1}(t-t')f(t-t')g(t') \tag{4.15}$$

We swap the integrals allowed due to Fubini's theorem and substitute with respect to t, $\tau := t - t'$, $\tau \in (-t', \infty)$, shorten the integration limits by the Heaviside function $\mathbb{1}$ and

swap integrals again.

$$\mathfrak{L}[f*g] = \int_0^\infty dt' e^{i\omega^+ t'} \int_{-t'}^\infty d\tau e^{i\omega^+ \tau} \mathbb{1}(\tau) f(\tau) g(t') \tag{4.16}$$

$$= \int_0^\infty dt' e^{i\omega^+ t'} \int_0^\infty d\tau e^{i\omega^+ \tau} f(\tau) g(t') \tag{4.17}$$

$$= \underbrace{\int_0^\infty d\tau e^{i\omega^+ \tau} f(\tau)}_{f(\omega)} \underbrace{\int_0^\infty dt' e^{i\omega^+ t'} g(t')}_{=g(\omega)} = \mathfrak{L}[f] \cdot \mathfrak{L}[g] \tag{4.18}$$

The proof for the Fourier transform works analogously.

4.1.3 Connection between both transformations

Let $f(\omega)$ and $F(\omega')$ be the complex Laplace respectively the Fourier transform of $f(t)$. Then we gain the following dispersive relation:

$$f(\omega) = \int_0^\infty f(t) e^{i\omega^+ t} dt = \int_0^\infty \int_{-\infty}^\infty \frac{d\omega'}{2\pi} F(\omega') e^{i\omega' t} e^{i\omega^+ t} dt \tag{4.19}$$

$$= \int_{-\infty}^\infty \frac{d\omega'}{2\pi} \left(\int_0^\infty dt e^{i(\omega^+ + \omega')t} \right) F(\omega') = \int_{-\infty}^\infty \frac{d\omega'}{2\pi} \frac{i}{\omega + \omega' + i0^+} F(\omega')$$

4.2 Evaluation of the Laplace transform of the bath correlation function in the wide band limit

Theorem 4.1 (Sokhotski-Plemelj theorem)
Let $\phi(x)$ be a continuous integrable function on the real axis ($\phi \in C^1(\mathbb{R}) \cap L^1(\mathbb{R})$), then

$$\lim_{\varepsilon \to 0^+} \int_{-\infty}^\infty dx \frac{\phi(x)}{x \pm i\varepsilon} = \left(\mathrm{pv} \frac{1}{x} \mp i\pi\delta(x) \right) \phi(x) \tag{4.20}$$

Proof 4.1
We expand on a real denominator:

$$\lim_{\varepsilon \to 0^+} \int_{-\infty}^\infty dx \frac{\phi(x)}{x \pm i\varepsilon} = \underbrace{\lim_{\varepsilon \to 0^+} \int_{-\infty}^\infty dx \frac{x^2}{x^2 + \varepsilon^2} \frac{\phi(x)}{x}}_{=I_1} \mp i\pi \underbrace{\lim_{\varepsilon \to 0^+} \int_{-\infty}^\infty dx \frac{\varepsilon}{\pi(x^2 + \varepsilon^2)} \phi(x)}_{=I_2} \tag{4.21}$$

We take the half of the first integral I_1 and perform a variable transformation $x \to -x'$ and combine those halves again

$$I_1 = \lim_{\varepsilon \to 0^+} \int_{-\infty}^\infty dx \frac{x^2}{x^2 + \varepsilon^2} \frac{\phi(x) - \phi(-x)}{2x} \tag{4.22}$$

We can express the integral as a limit and swap the two limits

$$I_1 = \lim_{\eta\to 0^+}\lim_{\varepsilon\to 0^+}\int_{|x|>\eta} dx \frac{x^2}{x^2+\varepsilon^2}\frac{\phi(x)-\phi(-x)}{2x} \tag{4.23}$$

Now we want to apply Lebesgue's dominated convergence theorem. We find a majorant of the integrand for $\varepsilon \to 0$ and see that the integral $\int_{|x|>\eta} dx \frac{\phi(x)-\phi(-x)}{2x}$ is finite since $x = 0$ is excluded and $\phi \in L^1(\mathbb{R})$. So we swap the limit and the integral. With the remaining limit we have the definition of the Cauchy principal value:

$$I_1 = \mathcal{P}\int_{-\infty}^{\infty} dx \frac{\phi(x)}{x} \tag{4.24}$$

For the second integral I_2 we add and subtract $\phi(0)$ in the numerator. The limes can be separated since both summands are assumed to converge:

$$I_2 = \lim_{\varepsilon\to 0^+}\varepsilon\int_{-\infty}^{\infty}\frac{x}{\pi(x^2+\varepsilon^2)}\frac{\phi(x)-\phi(0)}{x}dx + \frac{\phi(0)}{\pi}\lim_{\varepsilon\to 0^+}\int_{-\infty}^{\infty} dx\frac{\varepsilon}{x^2+\varepsilon^2} \tag{4.25}$$

We show that the first term goes to zero. We split the integration domain. For $|x| > 1$ we apply the dominated convergence theorem and the integrand becomes zero. For $|x| \leq 1$ we show that the absolute value of the integral is zero. So we apply the mean value theorem, the triangle inequality and perform the remaining integral:

$$\lim_{\varepsilon\to 0^+}\varepsilon\left|\int_{-1}^{1} dx\frac{x}{\pi(x^2+\varepsilon^2)}\frac{\phi(x)-\phi(0)}{x}\right| \leq \lim_{\varepsilon\to 0^+}\sup_{\eta\in(-1,1)}|\phi'(\eta)|\varepsilon\int_{-1}^{1} dx\frac{|x|}{\pi(x^2+\varepsilon^2)}$$

$$= \underbrace{\sup_{\eta\in(-1,1)}|\phi'(\eta)|\frac{1}{\pi}}_{=C}\lim_{\varepsilon\to 0^+}\varepsilon\underbrace{\int_0^1 dx\frac{2x}{(x^2+\varepsilon^2)}}_{=\log(x^2+\varepsilon^2)\big|_0^1} = C\lim_{\varepsilon\to 0^+}\varepsilon\log(1+\frac{1}{\varepsilon^2}) \overset{\text{l'Hosp.}}{=} C\lim_{\varepsilon\to 0^+}\frac{\frac{1}{1+\frac{1}{\varepsilon^2}}\frac{-2}{\varepsilon^3}}{-\frac{1}{\varepsilon^2}} = 0$$

For the last remaining integral we can restrict the integration domain to $|x| \leq 1$, since we know the rest to be zero. Then we substitute $\frac{x}{\varepsilon} = y$, $dx = \varepsilon dy$ and use $\arctan(x) \xrightarrow{x\to\pm\infty} \pm\frac{\pi}{2}$:

$$\lim_{\varepsilon\to 0^+}\int_{-1}^{1} dx\frac{\varepsilon}{(x^2+\varepsilon^2)} = \lim_{\varepsilon\to 0^+}\underbrace{\int_{-1/\varepsilon}^{1/\varepsilon} dy\frac{1}{(y^2+1)}}_{\arctan(y)} = \pi \tag{4.26}$$

So we have

$$I_2 = \phi(0) \tag{4.27}$$

It can be shown in general that the integral kernel in I_2 is a nascent delta distribution, it converges in the weak sense for $\varepsilon \to 0^+$

$$\frac{\varepsilon}{\pi(x^2+\varepsilon^2)} \xrightarrow{\varepsilon\to 0^+} \delta(x) \tag{4.28}$$

□

For the shifted function $x = y + E$ we have

$$\lim_{\varepsilon\to 0^+}\int_{-\infty}^{\infty} dx \frac{\phi(y)}{y+E+i\varepsilon} = \left(\mathrm{pv}\frac{1}{x} - i\pi\delta(x)\right)\phi(x-E) \tag{4.29}$$

Theorem 4.2 (Evaluation of the bath correlation function)
The Laplace transform of the bath correlation function $C^s_{\alpha\mu\kappa}(t)$ using the dispersive relation is given by

$$C^s_{\alpha\mu\kappa}(\omega) = \int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\Gamma^s_{\alpha\mu\kappa}(\omega')\frac{i}{\omega + s\omega' + i0^+} \tag{4.30}$$

In the wide band limit this simplifies to (using the Sokhotski-Plemelj theorem (4.1))

$$C^s_{\alpha\mu\kappa}(\omega) = i\Gamma^{(s)}_{\alpha\mu\kappa}\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\frac{n^s_\alpha(\omega')}{\omega+s\omega'+i0^+} = i\frac{\Gamma^{(s)}_{\alpha\mu\kappa}}{2\pi}\int_{-\infty}^{\infty}dz\frac{n^s_\alpha(s(z-\omega))}{z+i0^+} \tag{4.31}$$

$$= \Gamma^{(s)}_{\alpha\mu\kappa}\left(\frac{1}{2}n^s_\alpha(-s\omega) + \frac{i}{2\pi}\underbrace{\mathrm{pv}\left(\frac{n^s_\alpha(s(z-\omega))}{z}\right)}_{=:\Lambda^s_\alpha(\omega)}\right) = \Gamma^{(s)}_{\alpha\mu\kappa}\cdot F^s_\alpha(\omega) \tag{4.32}$$

with the principal value integral:

$$\Lambda^s_\alpha(\omega) = \mathcal{P}\int_{-\infty}^{\infty}\frac{n^s_\alpha(s(x-\omega))}{x}dx \overset{x=\omega+sy}{=} \mathcal{P}\int_{-\infty}^{\infty}\frac{n^s_\alpha(y)}{\omega+sy}dy = \mathrm{Re}\left[\Psi\left(\frac{1}{2}+i\frac{\beta(\omega+s\mu_\alpha)}{2\pi}\right)\right] \tag{4.33}$$

Proof 4.2
As the generalized Fermi function in the integrand $n^s_\alpha\big(s(x-\omega)\big) = \big(1+e^{\beta((x-\omega)-s\mu_\alpha)}\big)^{-1}$ doesn't fulfil this Schwartz space condition $n^s_\alpha\big(s(x-\omega)\big) \xrightarrow{x\to-\infty} 1$ we change our flat band assumption slightly by adding an auxiliary exponential convergence term $e^{\eta x}$, with $\eta\to 0^+$.

$$\mathcal{P}\int_{-\infty}^{\infty}\frac{dx}{2\pi}\frac{n^s_\alpha\big(s(x-\omega)\big)}{x}$$

$n^s_\alpha(\omega) = \big(1+e^{s\beta(\omega-\mu_\alpha)}\big)^{-1}$

For the principal value integral we have a look at the closed Cauchy integral, where the integration along a semicircle with infinitely large radius and along the semicircle cutting out the singularity at zero vanish due to the exponential terms. So we apply the residual theorem. The roots of the Fermi function are the Matsubara frequencies

$$x_n = \omega + s\mu_\alpha + i\frac{2\pi}{\beta}(n+1/2).$$

We get for the sum of the residuals:

$$\oint_{-\infty}^{\infty} \frac{n_\alpha^s\big(s(x-\omega)\big)}{x}dx = 2\pi i \sum_{n=0}^{\infty} \frac{e^{\eta x_n}}{-\beta}\frac{1}{x_n} = -\frac{2\pi i}{\beta}\sum_{n=0}^{\infty}\frac{e^{\eta(\omega+s\mu_\alpha+i\frac{2\pi}{\beta}(n+1/2))}}{\omega+s\mu_\alpha+i\frac{2\pi}{\beta}(n+1/2)}$$

We extract the factor $i2\pi/\beta$ from the denominator, define a new convergence factor $\tilde{\eta} = \eta \cdot i2\pi/\beta$ and define $z := \frac{1}{2} - i\frac{\beta}{2\pi}(\omega + s\mu_\alpha)$ and find:

$$= -e^{z\tilde{\eta}} \sum_{n=0}^{\infty} \frac{e^{i\tilde{\eta}n}}{z+n}$$

As the exponential term before the sum only depends on η we can already evaluate the limit and sent it to one. For $\eta > 0$, we still have the convergence term in the sum $e^{i\tilde{\eta}n} \propto e^{-2\pi\eta n/\beta} \xrightarrow{n\to\infty} 0$ and so also the sum converges

$$C := \sum_{n=1}^{\infty} \frac{\left(e^{-2\pi\eta/\beta}\right)^n}{n} = -\ln(1-e^{-2\pi\eta/\beta}) < \infty$$

Adding and subtracting that constant results in

$$-\sum_{n=0}^{\infty}\frac{e^{i\tilde{\eta}n}}{z+n} = -\frac{1}{z} + \sum_{n=1}^{\infty}\left(\frac{e^{i\tilde{\eta}n}}{n} - \frac{e^{i\tilde{\eta}n}}{z+n}\right) - C = -\frac{1}{z} + \sum_{n=1}^{\infty}\frac{ze^{i\tilde{\eta}n}}{n(n+z)} - C$$

One can show that the constant C that goes to infinity for $\eta \to 0^+$ cancels out with the complex conjugated. Now we can neglect the convergence factor in the sum which behaves like $\frac{1}{n^2}$. Compared to the series expansion of the digamma function Ψ we find:

$$\Psi(z+1) = \sum_{n=1}^{\infty}\frac{z}{n(n+z)} - \gamma, \quad \Psi(z+1) = \Psi(z) + \frac{1}{z}$$

$$-\frac{1}{z} + \sum_{n=1}^{\infty}\frac{z}{n(n+z)} - C = \Psi(z) - \gamma - C$$

with the Euler-Mascheroni constant $\gamma \approx 0.57721$. Another representation of the complex digamma function is given by the integral expression

$$\Psi(z) = \int_0^{\infty}\frac{e^{-t}}{t} - \frac{e^{-zt}}{1-e^{-t}}dt \tag{4.34}$$

$$\mathrm{Re}\big(\Psi(a+bi)\big) = \int_0^{\infty}\frac{e^{-t}}{t} - \frac{e^{-at}\cos(bt)}{1-e^{-t}}dt \tag{4.35}$$

As we just need the real part we can see from this expression that the sign of the complex argument in z doesn't matter, so we find for

$$\Lambda_\alpha^s(\omega) = \mathrm{Re}(\Psi(z)) - \gamma - C, \quad z = \frac{1}{2} + i\frac{\beta}{2\pi}(\omega + s\mu_\alpha) \tag{4.36}$$

□

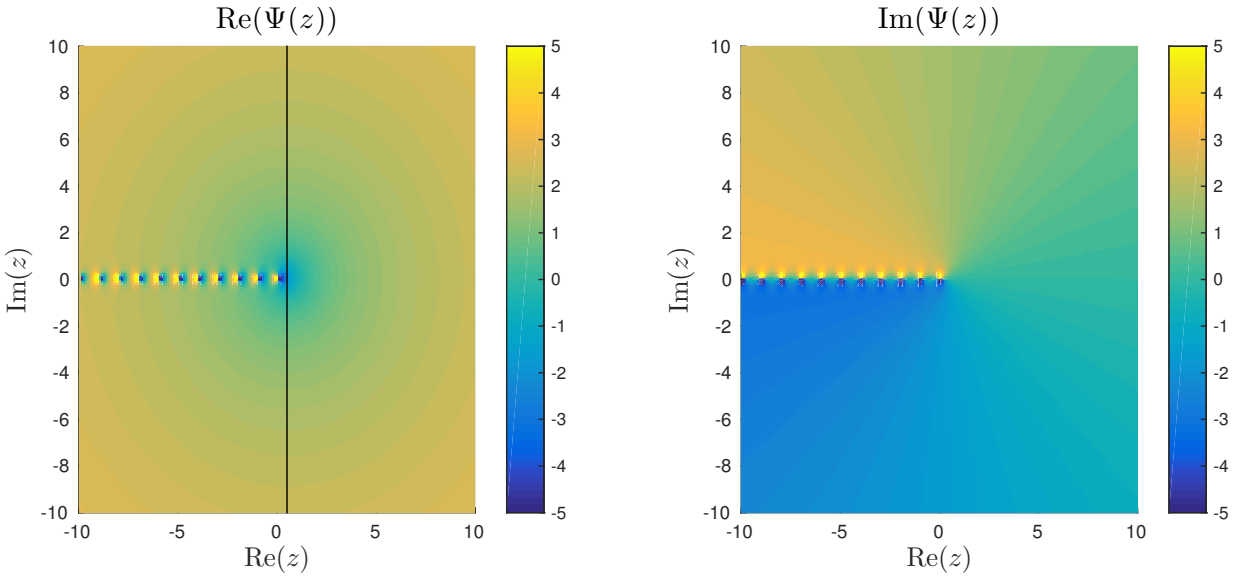


Figure 4.1: Real and imaginary part of the digamma function. The black line marks the relevant parameter space used in the analytic expression for the bath correlation function.

List of Figures

Bibliography

[1] Philip L Taylor and Olle Heinonen. *A quantum approach to condensed matter physics*. Cambridge University Press, 2002.

[2] Walter Kohn. *An essay on condensed matter physics in the twentieth century*. Springer, 1999.

[3] Walter Kohn and Lu Jeu Sham. Self-consistent equations including exchange and correlation effects. *Physical Review*, 140(4A):A1133, 1965.

[4] Reiner M Dreizler and Eberhard Engel. *Density functional theory*. Springer, 2011.

[5] Masatoshi Imada, Atsushi Fujimori, and Yoshinori Tokura. Metal-insulator transitions. *Reviews of Modern Physics*, 70(4):1039, 1998.

[6] David Goldhaber-Gordon, Hadas Shtrikman, D Mahalu, David Abusch-Magder, U Meirav, and MA Kastner. Kondo effect in a single-electron transistor. *Nature*, 391(6663):156–159, 1998.

[7] Elbio Dagotto. Correlated electrons in high-temperature superconductors. *Reviews of Modern Physics*, 66(3):763, 1994.

[8] John Hubbard. Electron correlations in narrow energy bands. In *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, volume 276, pages 238–257. The Royal Society, 1963.

[9] Eva Pavarini, Erik Koch, Dieter Vollhardt, and Alexander Lichtenstein. *The LDA+ DMFT Approach to Strongly Correlated Materials–Lecture Notes of the Autumn School 2011 Hands-on LDA+ DMFT*. 2011.

[10] Antoine Georges and Gabriel Kotliar. Hubbard model in infinite dimensions. *Physical Review B*, 45(12):6479, 1992.

[11] Dieter Vollhardt. Dynamical mean-field theory for correlated electrons. *Annalen der Physik*, 524(1):1–19, 2012.

[12] Philip Warren Anderson. Localized magnetic states in metals. *Physical Review*, 124(1):41, 1961.

[13] Gianaurelio Cuniberti, Giorgos Fagas, and Klaus Richter. *Introducing Molecular Electronics*. Springer, 2010.

[14] Jiwoong Park, Abhay N Pasupathy, Jonas I Goldsmith, Connie Chang, Yuval Yaish, Jason R Petta, Marie Rinkoski, James P Sethna, Héctor D Abruña, Paul L McEuen, et al. Coulomb blockade and the kondo effect in single-atom transistors. *Nature*, 417(6890):722–725, 2002.

[15] D. Darau, G. Begemann, A. Donarini, and M. Grifoni. Interference effects on the transport characteristics of a benzene single-electron transistor. *Phys. Rev. B*, 79:235404, Jun 2009.

[16] Kenneth G Wilson. The renormalization group: Critical phenomena and the kondo problem. *Reviews of Modern Physics*, 47(4):773, 1975.

[17] TA Costi, AC Hewson, and V Zlatic. Transport coefficients of the anderson model via the numerical renormalization group. *Journal of Physics: Condensed Matter*, 6(13):2519, 1994.

[18] Ned S Wingreen and Yigal Meir. Anderson model out of equilibrium: Noncrossing-approximation approach to transport through a quantum dot. *Physical Review B*, 49(16):11040, 1994.

[19] Emanuel Gull, Andrew J Millis, Alexander I Lichtenstein, Alexey N Rubtsov, Matthias Troyer, and Philipp Werner. Continuous-time monte carlo methods for quantum impurity models. *Reviews of Modern Physics*, 83(2):349, 2011.

[20] Martin Nuss, Martin Ganahl, Hans Gerd Evertz, Enrico Arrigoni, and Wolfgang von der Linden. Steady-state and quench-dependent relaxation of a quantum dot coupled to one-dimensional leads. *Phys. Rev. B*, 88:045132, Jul 2013.

[21] Tomonori Shirakawa and Seiji Yunoki. Block lanczos density-matrix renormalization group method for general anderson impurity models: Application to magnetic impurity problems in graphene. *Physical Review B*, 90(19):195109, 2014.

[22] M. Potthoff, M. Aichhorn, and C. Dahnken. Variational cluster approach to correlated electron systems in low dimensions. *Phys. Rev. Lett.*, 91(20):206402, November 2003.

[23] Martin Nuss, Enrico Arrigoni, Markus Aichhorn, and Wolfgang von der Linden. Variational cluster approach to the single-impurity anderson model. *Phys. Rev. B*, 85:235107, Jun 2012.

[24] Martin Nuss, Christoph Heil, Martin Ganahl, Michael Knap, Hans Gerd Evertz, Enrico Arrigoni, and Wolfgang von der Linden. Steady-state spectra, current, and stability diagram of a quantum dot: A nonequilibrium variational cluster approach. *Phys. Rev. B*, 86:245119, Dec 2012.

[25] Howard J. Carmichael. *Statistical Methods in Quantum Optics 1: Master Equations and Fokker-Planck Equations*. Springer, 2010.

[26] Jinshuang Jin, Jun Li, Yu Liu, Xin-Qi Li, and YiJing Yan. Improved master equation approach to quantum transport: From born to self-consistent born approximation. *The Journal of Chemical Physics*, 140(724):–, 2014.

[27] Manabu Kiguchi, Oren Tal, Sören Wohlthat, Fabian Pauly, Michael Krieger, Darko Djukic, Juan Carlos Cuevas, and Jan M van Ruitenbeek. Highly conductive molecular junctions based on direct binding of benzene to platinum electrodes. *Physical review letters*, 101(4):046801, 2008.

[28] Mark A Reed, C Zhou, CJ Muller, TP Burgin, and JM Tour. Conductance of a molecular junction. *Science*, 278(5336):252–254, 1997.

[29] F Molitor, S Dröscher, J Güttinger, A Jacobsen, C Stampfer, T Ihn, and K Ensslin. Transport through graphene double dots. *Applied Physics Letters*, 94(22):222107, 2009.

[30] Dmitry A. Ryndyk, Andrea Donarini, Milena Grifoni, and Klaus Richter. Many-body localized molecular orbital approach to molecular transport. *Phys. Rev. B*, 88:085404, Aug 2013.

[31] Antonius Dorda, Martin Nuss, Wolfgang von der Linden, and Enrico Arrigoni. Auxiliary master equation approach to nonequilibrium correlated impurities. *Physical Review B*, 89(16):165105, 2014.

[32] Martin Nuss, Gerhard Dorn, Antonius Dorda, Wolfgang von der Linden, and Enrico Arrigoni. Master equation based steady-state cluster perturbation theory. *arXiv preprint arXiv:1505.01683*, 2015.

[33] Gernot Schaller. *Open Quantum Systems Far from Equilibrium*. Springer, Cham ; New York, auflage: 2014 edition, January 2014.

[34] Heinz-Peter Breuer and Francesco Petruccione. *The Theory of Open Quantum Systems*. Oxford University Press, 2002.

[35] Ángel Rivas and Susana F Huelga. *Open Quantum Systems*. Springer, 2012.

[36] H. Haug and A.-P. Jauho. *Quantum Kinetics in Transport and Optics of Semiconductors*. Springer-Verlag GmbH, 2., corr. printing edition, December 1996.

[37] Paul Adrien Maurice Dirac. The principles of quantum mechanics 4th ed (the international series of monographs on physics no 27), 1967.

[38] U. Fano. Description of states in quantum mechanics by density matrix and operator techniques. *Rev. Mod. Phys.*, 29:74–93, Jan 1957.

[39] Jinshuang Jin, Xiao Zheng, and YiJing Yan. Exact dynamics of dissipative electronic systems and quantum transport: Hierarchical equations of motion approach. *The Journal of chemical physics*, 128(23):234703, 2008.

[40] R Härtle, G Cohen, DR Reichman, and AJ Millis. Decoherence and lead-induced interdot coupling in nonequilibrium electron transport through interacting quantum dots: A hierarchical quantum master equation approach. *Physical Review B*, 88(23):235426, 2013.

[41] R Härtle, G Cohen, DR Reichman, and AJ Millis. Transport through an anderson impurity: Current ringing, non-linear magnetization and a direct comparison of continuous-time quantum monte carlo and hierarchical quantum master equations. *arXiv preprint arXiv:1505.01283*, 2015.

[42] Ryszard Horodecki, Paweł Horodecki, Michał Horodecki, and Karol Horodecki. Quantum entanglement. *Reviews of Modern Physics*, 81(2):865, 2009.

[43] Heinz-Peter Breuer. Non-markovian generalization of the lindblad theory of open quantum systems. *Physical Review A*, 75(2):022103, 2007.

[44] Hendrik Weimer. *Open Quantum Systems*. 2014.

[45] Goran Lindblad. On the generators of quantum dynamical semigroups. *Communications in Mathematical Physics*, 48(2):119–130, 1976.

[46] Yu Liu, JinShuang Jin, Jun Li, XinQi Li, and YiJing Yan. Number-resolved master equation approach to quantum transport under the self-consistent born approximation. *Science China Physics, Mechanics and Astronomy*, 56(10):1866–1873, 2013.

[47] Yigal Meir and Ned S. Wingreen. Landauer formula for the current through an interacting electron region. *Phys. Rev. Lett.*, 68:2512–2515, Apr 1992.